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THE WASTE ISOLATION PILOT PLANT (WIPP) ACTINIDE SOURCE TERM: TEST PLAN FOR THE CONCEPTUAL MODEL AND THE DISSOLVED CONCENTRATION SUBMODEL

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ABSTRACT

This Test Plan describes the conceptual model for the Waste Isolation Pilot Plant (WIPP) Actinide Source Term, and the approach for developing numerical models to describe the dissolved concentrations of actinides that could occur at the WIPP Site. The conceptual model is the framework for reducing a complex chemical system to a simpler system amenable to mathematical description. The numerical model to describe dissolved actinide concentrations will provide quantitative predictions of dissolved actinide concentrations in WIPP brines, as influenced by coupled chemical processes represented by reactions between the brine and WIPP wastes, including gas generation.

The actinides of interest to the WIPP Performance Assessment are thorium (Th), uranium (U), neptunium (Np), plutonium (Pu), and americium (Am). Dissolved actinide concentration submodels are needed for the +III actinides Pu(III) and Am(III); the +IV actinides Pu(IV), U(IV), Np(IV), and Th(IV); the +V actinides Pu(V), Np(V), and Am(V); and the +VI actinides Pu(VI), U(VI), and Np(VI). The "models" referred to are data bases of chemical parameters to be used in an existing mathematical framework (thermodynamics and the Pitzer activity coefficient model), as implemented by existing computer codes. These data bases make use of a set of thermodynamic parameters from the literature called the Harvie-Moller-Weare (HMW) parameterization of the Pitzer activity coefficient model. The laboratory and modeling effort described in this Test Plan augments the HMW model so that it can be applied to the actinides and oxidation states important to the WIPP Performance Assessment. It appears that one data base each will be needed to describe the +III, +V, and +VI oxidation states, while several data bases will be required to describe the chemical behavior of the +IV actinides. This Test Plan presents the rationale, design, and justification of the work in progress for quantifying dissolved actinide concentrations for the WIPP Performance Assessment.

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ACRONYMS/INITIALISMS

ANL	Argone National Laboratory
ASME	
AST	Actinide Source Term
ASTP	Actinide Source Term Program
BIR	Baseline Inventory Report
DOE/CAO	U.S. Department of Energy Carlsbad Area Office
DOE/WPIO	U.S. Department of Energy WIPP Project Integration Office (the precursor to DOE/CAO)
DQO	Data Quality Objectives
DRP	Disposal Room Program
EDTA	ethylenediaminetetraacetic acid
EPA	U.S. Environmental Protection Agency
ES&H	Environment, Safety, and Health
FSU	Florida State University
GGP	Gas Generation Program
HMW/FW	Harvie-Moller-Weare/Felmy-Weare (Harvie et al., 1984; Felmy and Weare, 1986)
HPLC	high pressure liquid chromatograph
IC	ion chromatograph
ICP-MS	inductively-coupled plasma mass spectrometer
ICP-OES	inductively-coupled plasma optical emissions spectrometer
IR	infrared
LANL	Los Alamos National Laboratory
LBNL	Lawrence Berkeley National Laboratory
LLNL	Lawrence Livermore National Laboratory
LSC	liquid scintillation counter
M&TE	measuring and testing equipment
NBS	National Bureau of Standards
NEA	Nuclear Energy Agency
NIST	National Institute of Standards and Technology
PA	Performance Assessment
PNC	Power Reactor and Nuclear Fuel Development Corporation of Japan
PNL	Pacific Northwest Laboratory
QA	quality assurance
QAP	Quality Assurance Procedure
QAPD	Quality Assurance Program Description
QAPP	Quality Assurance Program Plan
SIT	Specific Interaction Theory
SNL	Sandia National Laboratories
SPC	Salado Primary Constituent
STTP	Actinide Source-Term Waste Test Program
TIC	total inorganic carbon
TRU	transuranic
TTA	1-thenoyl-3,3,3-trifluoroacetone
UV	ultraviolet
WIPP	Waste Isolation Pilot Plant

Chemical Symbols

Am	americium
Np	neptunium
Pu	plutonium
Th	thorium
U	uranium

1.0 INTRODUCTION

The Waste Isolation Pilot Plant (WIPP) is a proposed repository for defense-generated transuranic (TRU) wastes. These wastes are typically a nonradioactive matrix that has been contaminated with actinide elements such as plutonium (Pu), uranium (U), americium (Am), neptunium (Np), and thorium (Th). The waste matrices include cemented sludges, cellulosic materials, iron and/or steel and aluminum metals, resins, and pyrochemical salts.

The Actinide Source Term (AST) is the name given to the concentrations of actinides that could be mobilized by brines and thus could migrate from the WIPP facility. The movement of brines in and above the WIPP Site is considered under the human intrusion scenarios (see for example Lappin et al., 1989) and is outside the scope of this Test Plan. The product of mobile actinide concentrations times brine flow rates yields the flux of actinides that could leave the WIPP disposal horizon. Information on mobile actinide concentrations will be provided to the WIPP Performance Assessment Department to support the WIPP compliance application.

Work on the Actinide Source Term is performed under the Actinide Source Term Program (ASTP). The ASTP contains three principal activities: development of a dissolved actinide concentration submodel, development of a colloid submodel, and tests with actual transuranic (TRU) waste. The solubility submodel will predict dissolved actinide concentrations under WIPP conditions; the colloid submodel will predict colloidal actinide concentrations under WIPP conditions as they prove to be important in concentrated WIPP brines. The tests with actual TRU waste will demonstrate actinide behavior in simulated disposal room environments, provide a check on the important chemical mechanisms for mobilizing actinides, and test the dissolved concentration and colloid submodels.

The development of the AST dissolved actinide concentration submodel is actually the development of a data base for use with existing computer codes. The mathematical equations necessary to describe equilibrium in aqueous/mineral/gaseous systems are fully developed, as are the computer codes to implement these equations. What is lacking is the data base, the set of aqueous and solid chemical species containing actinides, the standard chemical potentials of these species, and the specific ion interaction parameters required to describe the interactions between these species and other constituents of brines in WIPP scenarios.

In summary, the Actinide Source Term Program is composed of three separate yet interdependent areas: a dissolved actinide concentration submodel, a colloidal actinide submodel, and tests with actual TRU waste. The three subprograms all contribute to the process of building and testing an overall numerical model that describes both the concentrations of actinides that could be mobilized from WIPP disposal rooms and the concentrations of actinides that could be subject to chemical retardation in the Culebra.

1.1 Objectives of the Actinide Source Term Program

The objective of this study is a technically credible phenomenological model describing concentrations of mobile dissolved actinides in the brine compositions that occur or are likely to occur at the WIPP Site. A model is a conceptual framework used to describe a system—including assumptions, mathematical equations, parameters necessary for the mathematics, and any computer codes that may be used to solve the equations. A phenomenological model is a mechanistic description of the scientific principles that govern the behavior of a system. A technically credible model is a consistent scientific description of a system supported by measurements of behavior; it is a model that has been reviewed and gained the support of outside experts in relevant scientific disciplines. Chemical logic indicates that an equilibrium thermodynamic model would both meet design objectives and be attainable with several years of effort. Thus, an equilibrium thermodynamic model for dissolved actinide concentrations is the specific deliverable necessary to achieve the study objectives.

The development of the conceptual model, including the logic leading to selection of a thermodynamic model description, is given in Section 2.0.

1.2 Description of the Relevant WIPP Environment

The WIPP is intended to store defense-generated transuranic (TRU) wastes. Transuranic wastes may contain such items as ordinary laboratory trash, protective clothing, glove boxes, column packing materials, and machining equipment contaminated with actinides, as well as process waste streams containing actinide residues such as pyrochemical salts and solidified sludges. Details on the types of wastes are given in the Baseline Inventory Report (BIR) (Department of Energy 1995). The wastes are typically a highly heterogeneous assortment of these and other materials stored in steel drums or standard waste boxes.

The WIPP design basis involves emplacing the waste into the disposal rooms in its existing forms without reprocessing. Several proposed options include backfill of crushed salt or a crushed salt/bentonite mixture. Creep closure of the bedded salt would then seal in the waste and isolate it from the environment.

The WIPP disposal horizon is located within a natural bedded salt formation approximately 655 meters (2150 ft) below ground surface in the Salado Formation (Lappin et al., 1989). The salt is predominantly halite, $NaCl(s)$, but also contains accessory evaporite minerals such as anhydrite, $CaSO_4(s)$, gypsum, $CaSO_4 \bullet 2H_2O(s)$, polyhalite, $K_2MgCa_2(SO_4)_4 \bullet 2H_2O(s)$, and magnesite, $MgCO_3(s)$, (Brush, 1990), as well as fractured anhydrite marker beds and clay seams. There are small quantities of intergranular and intragranular brines associated with the salt at the repository horizon. This brine is a highly concentrated solution (about 10 times that of sea water) of predominantly Na-Mg-K-Cl-SO_4 containing smaller amounts of Ca, CO_3, and B. The brine has been in contact with the evaporite salts for geologic time and is saturated with respect to the evaporate minerals. As such, it holds little potential for dissolving Salado minerals. The compositional variations in Salado brine from point to point and between intra- and intergranular brines are less than about 20%.

Underlying the Salado Formation is the Castile Formation, composed of bedded anhydrite. The Castile Formation in the vicinity of the WIPP Site is known to contain localized pockets of brine under sufficient pressure to force brine to the land surface. Castile brines are predominantly saturated sodium chloride solutions containing calcium and sulfate, and small concentrations of other elements, and are about eight times more concentrated than sea water. Above the Salado in the vicinity of the WIPP Site is the Rustler Formation, a part of which is the Culebra Dolomite Member. The Culebra is a fractured dolomite, $CaMg(CO_3)_2(s)$, layer approximately 8 meters thick, and is significant because it represents the only contiguous ground water in the area. Down gradient from the WIPP Site are wells into the Culebra used for watering cattle. Culebra brines are predominantly sodium chloride with potassium, magnesium, calcium, sulfate, and carbonate; these brines range from about one to four times the concentration of sea water.

1.3 Relationships to Other WIPP Programs

The ASTP is developing information on the chemical behavior of actinides in WIPP brines, both to provide as input to the draft certification compliance application and to continue to confirm knowledge of repository processes. There are several WIPP programs that either impact or are impacted by actinide chemistry, or both. These include the Gas Generation Program (GGP), the Disposal Room Program (DRP), and the NonSalado Chemical Retardation and Transport Program.

The GGP is examining the potential rates of generation of gases caused by interactions among brines, wastes, and microbes. Generation of carbon dioxide gas, $CO_2(g)$, through microbial degradation of cellulosic materials in the waste is of particular importance to the ASTP. Carbon dioxide gas, especially at high pressures, could markedly alter the composition of WIPP brines through gas dissolution. Dissolved $CO_2(g)$ could make the brine acidic, and acidic conditions tend to increase the solubility of actinides. High $CO_2(g)$ pressures in combination with waste/brine interactions that make the system basic could create large concentrations of carbonate, which is a powerful complexing ligand for actinides, and could thus tend to increase solubilities.

The chemical behavior of actinides in the disposal rooms is a coupled process among the GGP, the DRP, and the ASTP. The GGP in combination with the DRP are evaluating potential backfill additives for their ability to remove $CO_2(g)$ from the gas phase. Mechanisms that remove $CO_2(g)$ from the gas phase and incorporate it into the aqueous and/or solid phases necessarily alter the brine compositions. Because of the strong coupling between gas generation, waste form, backfill additives, and actinide solution chemistry, the impact of the GGP and DRP on dissolved actinide concentrations must be assessed within the ASTP.

The information gathered under the Actinide Source Term Program is needed for the WIPP programs investigating physical and chemical retardation. The transport behavior of actinides is strongly dependent on whether the actinides are in colloidal or dissolved form. The movement of dissolved substances in both the Salado and Rustler Formations is quite different from the movement of colloids. Dissolved actinides will diffuse down a concentration gradient within the matrix in the process called physical retardation, while some colloidal actinides will not, depending on the size of the colloid relative to pore throat size. Also, actinides that are already adsorbed to colloids may not interact chemically with porous media and thus may not be subject to chemical retardation phenomena. Thus, the relative amounts in each form are important.

Chemical retardation behavior for actinides is also strongly influenced by actinide speciation. Typically, uncomplexed actinide cations sorb more strongly than anionic actinide complexes. Information about actinide complexes with organic and inorganic ligands in the various WIPP brines is one of the main kinds of information being generated by the ASTP.

1.4 Reference WIPP Brine Compositions

Several synthetic reference brines are being used to examine actinide behavior for the WIPP. The formulations of these brines are based on the measured compositions of brines collected from the Salado and Castile Formations, and the Culebra Dolomite Member of the Rustler Formation. A common synthetic brine used to simulate Salado brine is called Brine A. Another brine commonly used is called SPC (Salado Primary Constituent) Brine, a truncated version of Brine A with the trace elements removed (see Novak, 1993). Synthetic ERDA-6 Brine represents Castile brine, and AISinR and H17 Brines mimic compositions measured at the Air Intake Shaft (AIS) and well H-17 in the Culebra Dolomite. The compositions of these brines are given in Table 1 (Brush, 1990; Novak, 1992; Robinson, 1991).

Table 1. Compositions of synthetic brines used for the WIPP.

Constituent	AISinR (Culebra) molal	H-17 (Culebra) molal	SPC (Salado) molal	ERDA-6 (Castile) molal

Sodium	0.639	2.48	2.00	5.35
Potassium	0.00845	0.0324	0.840	0.106
Magnesium	0.0214	0.0783	1.56	0.0208
Calcium	0.0170	0.0422	0.0164	0.0123
Chlorine	0.564	2.62	5.83	5.27
Sulfur	0.0791	0.0792	0.0436	0.187
Carbon	0.00177	0.000866	0.000176	0.0166
Boron	0.00277	-	0.0218	0.0692
Bromine	-	-	0.0109	0.0121

2.0 DEVELOPMENT OF CONCEPTUAL MODEL FOR DISSOLVED ACTINIDE CONCENTRATIONS

WIPP disposal scenarios assume that brine from the Salado Formation could seep into disposal rooms and contact TRU wastes when waste containers have been crushed because of creep closure. Should this occur, actinides could dissolve or form colloids in these brines. This in itself does not affect the ability of the WIPP to isolate wastes from the environment. However, this necessitates a thorough understanding of the types and quantities of the mobile actinides possible so that the flux of actinides from the repository can be evaluated.

Federal regulations (40 CFR 191; 40 CFR 194) governing the certification of the WIPP for waste disposal require the WIPP Performance Assessment to consider the possibility that, at some future time, the WIPP could be inadvertently intersected by one or more boreholes during prospecting for natural resources. Such drilling could provide a pathway for actinides mobilized in the brines to migrate away from the repository horizon.

Research supporting the WIPP Performance Assessment suggests that, should human intrusion into the repository occur, actinides mobilized in Salado brines could migrate to the Culebra and then be transported down gradient to the accessible environment. Further, should a human intrusion borehole intersect both the repository and a brine pocket in the Castile, Castile brine could contact waste, mobilize actinides in the Salado, and flow up to the Culebra. Finally, should more than one borehole intersect the repository, and one of these boreholes also intersect a Castile brine pocket, brine could flow into and through a waste panel before it flows up into the Culebra. These possibilities drive the question "What are the concentrations of actinides that could be mobilized from the WIPP repository horizon under human intrusion scenarios?"

The answer to this question is complex, but can be reduced to another question: "What are the brine compositions?" Brine composition is the primary determining factor for the chemical behavior of actinides in the aqueous and solid phases. There are two native, approximately uniform, brine compositions from the Salado and Castile Formations. However, interactions between the brines, the wastes, and backfill materials hold the potential for altering brine compositions significantly. Contact between brines and wastes could lower the pH in areas with high radiolysis; raise the pH in areas with cementitious materials, including backfills; increase the carbonate concentrations through promotion of microbial action that produces $CO_2(g)$; decrease magnesium concentration through brucite ($Mg(OH)_2(s)$) precipitation at moderately basic conditions; and produce many other scenarios. In addition, organic ligands, also called chelating agents, in the waste could dissolve and stabilize actinides in solution, and iron corrosion products could form colloids that sorb actinides. All of these potential brine/waste/backfill interactions can have a significant impact on brine compositions and thus on mobile actinide concentrations.

The mobile actinide concentration needed for evaluation of the WIPP with respect to regulatory requirements is the sum of dissolved and colloidal actinides. Dissolved actinides are in a broad category that includes hydrated ions, hydrolysis species, complexes with inorganic ligands, and complexes with inorganic and organic ligands (chelating agents). Colloids, particles small enough to remain suspended in solution by Brownian motion, are stable if they do not coagulate and/or settle out of solution by gravity. Colloidal actinides occur in two forms: intrinsic colloids, which are polymers of actinide cations bridged with oxygen or hydroxide, also called eigencolloids; and carrier colloids, which are otherwise nonradioactive colloids to which actinides have sorbed.

This Test Plan covers the Dissolved Actinide Concentration Submodel for the Actinide Source Term Program. The Colloidal Actinide Concentration Submodel for the Actinide Source Term Program is presented in Papenguth and Behl (1995). The processes governing the concentrations of mobile actinides include dissolution/precipitation, complexation with organic ligands, formation and persistence of actinide intrinsic colloids, formation and persistence of carrier colloids, and adsorption of actinides onto mobile carrier colloids or immobile substrates. All of these mechanisms have a fundamental dependence on brine compositions. Colloid formation and persistence have in addition a dependence on kinetics and particle size. Adsorption in addition has a dependence on the chemical nature of the substrate, regardless of whether the substrate is mobile (colloidal) or immobile. Because of their inherent differences in nature and properties, dissolved species and colloids have been separated into two separate but interacting study areas in support of the WIPP Performance Assessment.

The chemical situation expected in WIPP disposal rooms is too complex to be described exactly. Thus, several key assumptions are employed to make the problem tractable and to provide a measure of conservatism to the description. For the dissolved concentration submodel, these assumptions are (1) actinides do not sorb on immobile substrates, and (2) the system can be described with equilibrium thermodynamics for complexation and precipitation/dissolution reactions, but not for oxidation-reduction reactions. Related issues for actinide colloids can be found in the Test Plan for colloids (Papenguth and Behl, 1995).

Assumption 1 for the dissolved actinide submodel provides both a great deal of simplification to the problem and a measure of conservatism to the conceptual model. Actinides will sorb to solids. However, the majority of information about actinide sorption has not been focused toward developing quantitative models to understand and predict it. Models for actinide adsorption for even the simplest substrates are in early stages of development. A large effort would be required to characterize even a few of the expected sorption substrates in the WIPP, to measure actinide sorption from brines onto these substrates, and to develop quantitative models to describe the results. In addition, the nature of the substrates in the WIPP may change over the 10,000 year regulatory time frame, and their effectiveness in remaining sorptive for this period cannot be ascertained. Thus, an attempt to quantify adsorption in the WIPP disposal room environment would not appear to have a high potential for success given the current state of knowledge and the complexity of the chemical conditions. Assuming no sorption is conservative in that the mobility of actinides is overestimated.

Assumption 2 for the dissolved actinide submodel also provides a great simplification in the description of the system and an additional measure of conservatism in the conceptual model. Implicit in this assumption is the corollary:

"actinides in the waste dissolve immediately when contacted with brine." Actinides will not dissolve immediately, and the time required for dissolution could be a significant fraction of the regulatory time frame, given the relative insolubility of the expected form, e.g., $PuO_2(s)$. However, determination of the kinetics of actinide dissolution for the actinides in WIPP wastes would require location and characterization of the actinides in the heterogeneous waste mixture, another insurmountable task. The assumption of instantaneous dissolution obviates this problem, and potentially yields higher concentrations of mobile actinides, particularly in the early years of the regulatory time frame, which is conservative with respect to regulatory requirements.

A corollary to assumption 2 is that: "if oversaturated, actinide solid phases precipitate immediately." This corollary appears to be nonconservative, but this concern is mitigated by further considerations. The formation of actinide precipitates in the laboratory is typically examined over a time scale of days to months, a period that can be considered instantaneous compared with 10,000 years. Also, the form of the solid precipitates determined in the laboratory may be metastable. That is, the solids determined in the laboratory could convert to more stable forms as they age (see, for example, Neck et al., 1992). Accompanying this change from metastable to stable is a decrease in solubility. Therefore, using laboratory-determined, possibly metastable, solids for the precipitated actinide phases also adds conservatism. Intraaqueous complexing reactions occur rapidly at WIPP temperatures, so the assumption is acceptable in this regard.

Finally, the dissolved actinide submodel assumes that oxidation-reduction reactions are not considered to be at thermodynamic equilibrium. Information in the literature (see for example the discussions in Brush, 1990) demonstrates that oxidation-reduction equilibrium is often not attained in low temperature geochemical systems like the WIPP, even over geologic time. The kinetics are very slow, and conversion among oxidation states is strongly influenced by other factors. The WIPP disposal room environment will likely contain regions in which radiolysis would produce oxidizing conditions, regions in which large quantities of metallic iron will produce reducing environments, and regions where microbes may catalyze conversion among actinide oxidation states. The oxidation states of the actinides within WIPP disposal rooms cannot be predicted based on oxidation-reduction equilibrium. This feature could be added relatively simply once the dissolved actinide submodel is completed, requiring only information on oxidation potential among different actinide oxidation states already in the literature.

3.0 BRIEF REVIEW OF AQUEOUS THERMODYNAMICS

The principles of aqueous thermodynamics are well established, as are the nonlinear algebraic equations describing aqueous thermodynamics (see, for example, Denbigh, 1981; Garrels and Christ, 1991; Stumm and Morgan, 1981). The additional information needed to apply this model to dissolved actinide concentrations are parameters to use within these equations. These parameters include (1) identities of aqueous and solid phase chemical species, (2) standard chemical potentials for each species, and (3) constants for the activity coefficient submodel for each aqueous species.

Chemical models are built element by element, one oxidation state at a time. Two unique features are required of the actinide dissolved concentration model: a description of the chemical behavior of concentrated electrolytes (brines) in the WIPP system, and a description of the chemical behavior of the actinides themselves, which have not been studied extensively under environmental conditions.

Aqueous thermodynamics is a powerful conceptual and mathematical tool for describing chemical equilibrium in aqueous systems. The methodology is most often applied to reactions among dissolved species (intraaqueous reactions), reactions among dissolved and solid species (solubility, or precipitation/dissolution reactions), and reactions among dissolved species and gases (gas dissolution/exsolution reactions). Thermodynamics provides mathematical relationships among the identities, compositions, and pressures of the solid, aqueous and gas phases, and can predict changes in the compositions of all phases caused by changes in any phase. Of most importance for the WIPP, thermodynamics can predict the equilibrium composition of an aqueous phase in simultaneous contact with a gas phase and multiple solid phases, and the changes in all phases that occur in response to changes in other phases. For example, the change in the composition of a brine, and the equilibrium precipitation or dissolution of solid phases, can be modeled as a function of increasing carbon dioxide pressure over the solutions, or as a function of the addition of solid calcium oxide.

The discipline of thermodynamics is well developed and well documented. Numerous standard reference texts focusing on different applications are available, including Denbigh (1981), Smith and Van Ness (1975), Prausnitz (1969), Stumm and Morgan (1981), and Garrels and Christ (1990). A brief overview of the aspects of thermodynamics of particular importance for the Actinide Source Term dissolved actinide concentration submodel is presented below.

3.1 Standard Thermodynamic Relationships

Thermodynamics describes equilibrium among aqueous, solid, and gas species in the general reaction

$$\alpha A^a + \beta B^b \leftrightarrow A_\alpha B_\beta^{\alpha a+\beta b} \qquad \text{for reaction r} \qquad (1)$$

where A and B represent chemical species with charges a and b, respectively. The charges can be positive, negative, or zero. The symbols α and β represent stoichiometric coefficients for the reaction, and describe the proportions in which the reactants A^a and B^b combine to form the product $A_\alpha B_\beta^{\alpha a+\beta b}$. Thermodynamics provides several important mathematical relationships for describing Reaction 1. The Gibbs free energy for the r^{th} reaction, ΔG_r, is defined as

$$\Delta G_r \equiv \mu_{A_\alpha B_\beta^{\alpha a+\beta b}} - \alpha \mu_{A^a} - \beta \mu_{B^b} \qquad (2)$$

where μ_i is the chemical potential of the i^{th} species. The expression for the chemical potential depends on the phase of the species. For aqueous, solid, and gaseous species, the chemical potentials are given by

$$\mu_i \equiv \mu_i^o + RT \ln a_i = \mu_i^o + RT \ln \gamma_i m_i \qquad \text{i=aqueous} \qquad (3a)$$

$$\mu_i \equiv \mu_i^o + RT \ln a_i \qquad \text{i=solid} \qquad (3b)$$

$$\mu_i \equiv \mu_i^o + RT \ln a_i = \mu_i^o + RT \ln f_i = \mu_i^o + RT \ln \phi_i p_i \qquad \text{i=gas} \qquad (3c)$$

where a_i is the activity, γ_i is the activity coefficient, m_i is the molality (moles per kilogram H_2O), ϕ_i is the partial pressure...

for the activities of adsorbed or solid solution species also exist, but are beyond the scope of this work. Activity coefficients are functions of solution composition. Development of model parameters for activity coefficients is a major focus of ASTP dissolved concentration model development, as discussed below. Development of models for gas phase compositions in the WIPP is the purview of the Gas Generation Program.

The standard free energy of the r^th^ reaction, ΔG_r^o, is given by

$$\Delta G_r^o = \mu^o_{A_\alpha B_\beta^{\alpha a+\beta b}} - \alpha\, \mu^o_{A^a} - \beta\, \mu^o_{B^b} \qquad \text{for reaction r} \qquad (4)$$

where μ_i^o are constants called the standard chemical potentials, or the standard free energies of formation, for the i^th^ species. The superscript "o" is used to indicate the standard state. The standard reference state is typically 25°C and 1 atmosphere pressure, where the vast majority of thermodynamic information has been developed. Although thermodynamics is developed and applicable for other temperatures, the changes over a narrow, e.g., 5°C, temperature range are small.

The r^th^ reaction is at equilibrium when $\Delta G_r = 0$. At equilibrium, species activities satisfy the relationship:

$$\frac{-\left[\mu^o_{A_\alpha B_\beta^{\alpha a+\beta b}} - \alpha\, \mu^o_{A^a} - \beta\, \mu^o_{B^b}\right]}{RT}$$

$$= \ln(a_{A_\alpha B_\beta^{\alpha a+\beta b}}) - \alpha \ln(a_{A^a}) - \beta \ln(a_{B^b}) \qquad (5)$$

(compare with Equations 2 and 3). Thermodynamics can also be cast in the form of equilibrium constants by taking the exponential of the above expressions. The equilibrium constant is defined as

$$K^{eq}_{rxn1} \equiv \exp\left[\frac{-\Delta G^o_{rxn1}}{RT}\right] \qquad (6)$$

Equilibrium is given by

$$K_{rxn1}^{eq} = \frac{\left\{a_{A_\alpha B_\beta^{\alpha a+\beta b}}\right\}}{\left\{a_{A^a}\right\}^\alpha \left\{a_{B^b}\right\}^\beta} \tag{7}$$

Although the description in terms of standard chemical potentials and equilibrium constants are mathematically equivalent, the free energy of formation description is more general because it is independent of chemical reactions.

3.2 The Distinction between Molality and Molarity

Aqueous thermodynamics is defined for the molality scale, moles per kilogram H_2O, symbol m. A common unit used in aqueous chemistry is the molarity scale, moles per liter solution, symbol M, $\underline{M}$, or c. This distinction is not very important in dilute solutions at 25°C, where solution density is ~1.0 kg/liter and the mass of dissolved salts in the solutions is not great. However, in concentrated electrolytes such as the brines important to the WIPP, where solution density is not approximately ~1.0 kg/liter, and where the mass of dissolved salts is a considerable fraction of the solution mass, molality is not approximately equal to molarity. Molarity (volume based units) can be converted to molality (mass based units) if solution composition and density are known. Molality is necessary for thermodynamic modeling, while molarity is a simpler unit to use in the laboratory. Because both are in common use, care must be taken that they are specified and treated appropriately.

3.3 The pH, pcH, and pmH Scales

The hydrogen ion concentration in a solution is one of the most important variables governing equilibrium because H^+ or its "reciprocal" species OH^- participate in many aqueous reactions. Unfortunately, it is also one of the more difficult quantities to determine because H^+ is a component of water. In aqueous solutions, the H^+ concentration can vary over the range from 1 to 10^{-14} molal or wider, while the water concentration is always 55.5 molal. Thus, relative to the hydrogen contained in the water, the free hydrogen ion concentration is usually very small. The "pH" scale was defined to handle the wide possible H^+ concentrations, as the negative base 10 logarithm of the hydrogen ion activity, i.e.,

$$pH \equiv -\log(a_{H^+}) = -\log(\gamma_{H^+}\, m_{H^+}) \tag{8}$$

(Bates, 1973; Bates and Guggenheim, 1960). This definition by the National Bureau of Standards (NBS), now the National Institute of Standards and Technology (NIST), was based on dilute buffer systems. In addition, the definition was based on a choice of a reference state for the activity coefficients. In dilute solutions, the hydrogen ion activity can be measured directly with an ion selective pH electrode calibrated to the NIST scale. Unfortunately, the reference defined for the NIST pH scale is not applicable to concentrated electrolytes, which has led to some complications in terminology and measurements when discussing brine systems (Knauss et al., 1990; 1991; Mesmer, 1991).

From a thermodynamic point of view, Equation 8 is general, with the calculated pH value being dependent on the activity coefficient model. In dilute solutions, a reference activity coefficient model has been defined and accepted. However, for concentrated electrolytes, reference standards for pH have not yet been defined and adopted. Measurement of the hydrogen ion activity in concentrated electrolytes is difficult because of factors related to the performance of ion specific electrodes in concentrated electrochemical cells.

The quantities pcH and pmH were developed to work around the difficulty of specifying the pH in concentrated electrolyte solutions. These quantities are defined as the negative base 10 logarithms of the hydrogen ion molarity and molality, respectively, or

$$pcH \equiv -\log(c_{H^+}) \tag{9}$$

$$pmH \equiv -\log(m_{H^+}) \tag{10}$$

The pcH and pmH values are not dependent on a choice of reference activity coefficients or models. The pcH or pmH values contain all necessary information for interpretation with thermodynamic models, yet do not suffer from the ambiguities for pH in brines associated with a choice of a model for γ_i. An experimental method for determining hydrogen ion concentration is discussed in Rai et al. (1995).

4.0 BRINE MODELS AND ACTIVITY COEFFICIENT MODELS FOR THE WIPP ACTINIDE SOURCE TERM PROGRAM

4.1 Selection of Appropriate Models

The chemical behavior of the concentrated electrolytes that occur in evaporites like the WIPP host geology has been extensively studied and documented. Several thermodynamic models to describe the chemistry of concentrated electrolytes have been proposed, including Harvie et al. (1984), Felmy and Weare (1986), and Wood (1975). Four modeling approaches, including the Pitzer formalism, a "Harned's Rule" formalism, a hybrid of the ion pairing and Pitzer formalisms, and the Specific Interaction Theory (SIT) were examined in detail as candidate models, as discussed in Appendix A. The Harvie-Møller-Weare/Felmy-Weare parameterization of the Pitzer model (Harvie et al., 1984; Felmy and Weare, 1986) called HMW/FW henceforth, was selected as the best formalism and primary concentrated electrolyte data base for the WIPP Actinide Source Term dissolved concentration submodel. HMW/FW provides an established data base for description of solubility behavior in the Na-K-Mg-Ca-H-Cl-SO_4-OH-HCO_3-CO_3-CO_2-B-H_2O system, including the significant inorganic constituents of WIPP brines. Rather than develop a new description of the chemical behavior of the nonactinide chemical constituents in WIPP brines, it was decided to use the best existing mathematical formalism, and concentrate on including the actinides of interest to the WIPP and the organic waste constituents that may impact actinide dissolved concentrations.

A great deal of the information needed for the dissolved actinide model exists in the peer-reviewed literature, and represents experiments that do not need to be repeated. However, additional data for actinides are necessary and must be measured. An important facet of this model development is reinterpretation of published data to be consistent with the HMW/FW framework. If reliable data sets are available for a particular element and oxidation state, these data need only be supplemented by a small number of chemical measurements for the WIPP. If little or no reliable data are available for a particular element and oxidation state, complete suites of experiments will eventually be necessary to determine reliable data sets. Evaluation of literature data guides the experimental work, and reinterpretation of new data in combination with literature data provides the final model. This Test Plan describes this interplay.

4.2 Standing Applications of the HMW/FW Parameterization of the Pitzer Model

The existing HMW/FW data base is already capable of modeling most of the important interactions among WIPP brines—the evaporite salts that occur in and around the WIPP Site, potential waste or backfill additives such as calcium oxide or calcium hydroxide, and the carbon dioxide gas that may be generated within the disposal rooms. An example of this modeling capability applied to carbon dioxide gas dissolving in the standard Salado brine surrogate SPC Brine is given in Novak (1993). The HMW/FW data base has numerous applications for modeling and explaining chemical interactions in brines, for both the WIPP and other systems. Felmy and Weare (1986) included boron species into the HMW formulation, and used the resulting model to help understand the formation of borate mineral deposits in Searles Lake, California. The HMW model has been used to postulate compositions of Permian sea water based in part on fluid inclusion data taken from the Salado Formation at the WIPP site and from the Wellington Formation in Kansas (Horita et al., 1991). Extensions of the HMW model have been applied to systems at temperatures below 25°C (Spencer et al., 1990) and to temperatures in the range from 0°C to 250°C (Greenberg and Møller, 1989). Also, the solubility of methane in brines over a wide range in temperatures and pressures has been modeled (Duan et al., 1992). Other applications are documented in the literature, including McCaffrey et al. (1987), Eugster et al. (1980), and Stein and Krumhansl (1988).

4.3 The Pitzer Formalism for Activity Coefficient in Concentrated Electrolytes

The Pitzer activity coefficient model is a semi-empirical formalism for calculating activity coefficient in aqueous systems. The mathematics are based on a virial expansion of ion interactions in solutions as determined through chemical statistical mechanics theories of electrolytes and incorporate the Debye-Hückel limiting law for dilute solutions (Pitzer, 1991). While the Pitzer formalism is more complex than other activity coefficient models such as the Debye-Hückel, the extended Debye-Hückel, the Davies equation (see, for example, Table 3.3, p. 135, Stumm and Morgan, 1981), the B-dot (Helgeson, 1969), and the SIT (Grenthe and Wanner, 1992) models, it is more powerful and subsumes them. Thus, although most commonly applied to concentrated electrolytes, the Pitzer formalism is also appropriate for use with dilute solutions. Unlike all the other activity coefficient models listed above, the Pitzer model is developed for and has been shown to work for electrolytes as concentrated as those in the WIPP system. To illustrate accepted ranges of applicability, Table 3.3, p. 135 of Stumm and Morgan (1981), lists approximate maximum useful concentrations as 0.005, 0.1, and 0.5 Molar for the Debye-Hückel, the extended Debye-Hückel, and the Davies equation, respectively. Grenthe and Wanner (1992) give 3.5 molal as the approximate maximum ionic strength for SIT applications. In contrast, the Pitzer formalism has been applied successfully to evaporite systems with concentrations greater than 10 molal (Felmy and Weare, 1986). The range of ionic strengths in the WIPP system is approximately 0.8 to 8 molal.

The Pitzer equations for activity coefficients were developed to account for the chemical effects that become important in concentrated salt solutions. Summaries of the Pitzer formalism and various applications are given in Pitzer (1991); the developmental history of the theory can be traced through the articles referenced therein. The parameterization of this formalism for evaporite systems is documented in Harvie and Weare (1980), Harvie et al. (1984), and Felmy and Weare (1986). The following summary of the Pitzer equations is based on the discussion in Harvie et al. (1984).

The most important ion interactions in solution are binary cation-anion interactions. Binary ion-neutral interactions may also be important, but likely less so than cation-anion binary interactions. Three body interactions, e.g., cation-cation-anion or cation-anion-anion, give higher order effects that may not be important for the expected low concentrations of actinides in WIPP brines. Equation A.3b from Harvie et al. (1984), when simplified to include only binary cation-anion and ion-neutral interactions, takes the form

$$\ln \gamma_M = z_M^2 F + \sum_{a=1}^{N_a} m_a (2B_{Ma} + ZC_{Ma}) + |z_M| \sum_{c=1}^{N_c} \sum_{a=1}^{N_a} m_c m_a C_{ca}$$

$$+ \sum_{n=1}^{N_n} m_n(2\lambda_{nM}) \tag{11}$$

for cation M interacting with anionic and neutral species. An analogous form holds for anions, symbol X. The binary cation-anion interactions are represented by B_{MX}, itself a function, and the constants C_{MX}. Binary cation-neutral and anion-neutral interactions are represented by the constants λ_{nM} and λ_{nX}, respectively. The variable F represents the Debye-Hückel term, predominantly a function of ionic strength, z_M is the charge on the ion, and Z is a linear concentrated-weighted sum of solution charge. The summations in Equation 11 extend over all anionic species N_a, all cationic species N_c, or all neutral species N_n. All symbols are defined in Harvie et al. (1984).

B_{MX} is a function describing the chemical interactions between species M and species X. The form of this function depends on the charge of the individual ions.

$$B_{MX} \equiv \beta_{MX}^{(0)} + \beta_{MX}^{(1)} g(-2\sqrt{I}) + \beta_{MX}^{(2)} g(-12\sqrt{I}) \quad \text{1-(n}\geq\text{1) electrolytes} \tag{12a}$$

$$B_{MX} \equiv \beta_{MX}^{(0)} + \beta_{MX}^{(1)} g(-1.2\sqrt{I}) + \beta_{MX}^{(2)} g(-12\sqrt{I}) \quad \text{2-2 electrolytes} \tag{12b}$$

$$B_{MX} \equiv \beta_{MX}^{(0)} + \beta_{MX}^{(1)} g(-2\sqrt{I}) + \beta_{MX}^{(2)} g(-50\sqrt{I}) \quad \text{2-(n>2) electrolytes} \tag{12c}$$

(Pitzer and Silvester, 1978). The function g is given by

$$g(y) \equiv \frac{2(1-(1-y)e^{-y})}{y^2} \tag{13}$$

C_{Ma} is a constant, related to the tabulated quantity C_{MX}^{ϕ}, by the expression

$$C_{MX} \equiv \frac{C_{MX}^{\phi}}{2\sqrt{|z_M z_X|}} \tag{14}$$

The value Z is given by

$$Z \equiv \sum_i |z_i| m_i \tag{15}$$

were z_i is the charge on the ith ion. The Pitzer formalism requires up to four parameters to describe the binary interactions between cations and anions, $\beta_{MX}^{(0)}$, $\beta_{MX}^{(1)}$. $\beta_{MX}^{(2)}$, and C_{MX}^{ϕ}, depending on the charge on the ions and also the concentration ranges the ions have in solution. However, for many systems these parameters can be set to zero. The constants $\beta_{MX}^{(0)}$, $\beta_{MX}^{(1)}$. $\beta_{MX}^{(2)}$, and C_{MX}^{ϕ} are called specific ion interaction parameters or Pitzer parameters. Many more details on the HMW parameterization of the Pitzer model, including the complete equations, are given in Harvie et al. (1984).

The simplest version of the Pitzer activity coefficient model requires a single parameter for cation-anion interactions, $\beta_{MX}^{(0)}$. The $\beta_{MX}^{(0)}$ parameter is most important in the intermediate concentration range, e.g., ~1 to ~6.0 molal. This model is similar in form to the Specific Interaction Theory (SIT) model, which is recommended for solutions with ionic strengths less than 3.5 molal (Grenthe and Wanner, 1992). However, there are important distinctions between the SIT model and the Pitzer model. The SIT model is developed for solutions of one dominant electrolyte, and the only intended purpose of SIT is to extrapolate stability constant data to zero ionic strength. The Pitzer formalism is more powerful, particularly for complex brine systems, because interactions between all cations and anions are included, not just interactions with a single dominant background electrolyte. The SIT model has not been used to predict, let alone successfully predict, solubility behavior in complex concentrated brines like those at the WIPP Site.

The $\beta_{MX}^{(1)}$ parameter in Equation 12 multiplies a decaying exponential function of the ionic strength and thus is only important in the low ionic strength range, e.g., ~0.2 to ~1 molal. The $\beta_{MX}^{(2)}$ parameter is multiplied by a exponential function that decays even faster than that which multiplies $\beta_{MX}^{(1)}$ because of the different constants used in the "g(y)" function. Thus, $\beta_{MX}^{(2)}$ is only important and necessary for highly charged species at very low concentrations; few cation-anion pairs with one ion univalent require a nonzero value for the $\beta_{MX}^{(2)}$ parameter. (The only 2-1 electrolyte in the Harvie and Weare (1980) data base for which a nonzero parameter is needed is the Ca^{++}-OH^{-} pair.) The parameter C_{MX} is multiplied by a quadratic function of concentration, and thus becomes increasingly important at high concentrations, e.g., > ~ 5.0 m or higher.

5.0 Data Quality Objectives

The AST dissolved concentration model is intended to predict the dissolved concentrations of actinides in complex natural brines that have interacted with various waste forms and, potentially, with disposal room backfill additives. These systems are complex and contain many important influences, including brine compositions, the identities and concentrations of inorganic and organic species dissolved in the brines, and the hydrogen ion concentration.

The overall objective is a parameterized numerical model that predicts actinide solubilities within one order of magnitude in these complex systems. The value of one order of magnitude was chosen because it is a reasonable accuracy within the precision of Performance Assessment calculations. This value provides an acceptable degree of uncertainty for dissolved concentration information within the WIPP Performance Assessment.

The order-of-magnitude goal for the overall model does not suggest that order-of-magnitude accuracy and precision is acceptable for laboratory measurements. The acceptable level for precision and accuracy is a function of the particular analytical method used, and is determined by good scientific and laboratory practices. The precision and accuracy for every analysis is recorded in the quality assurance (QA) documentation as required by the QA plan (see section 12.0).

The overall model is intended to include all important interactions in Na-K-Mg-Ca-Cl-SO_4-CO_3-F-PO_4 brines containing organic ligands. Parameters are being determined from the important interactions in simpler chemical systems, e.g., Np(V) solubility in NaCl or Na_2CO_3 media. The dissolved concentration model is then built step-wise, e.g., first including hydrolysis, then chloride interactions, then carbonate interactions, then oxalate complexation, etc. Salts and ligands are examined in order of importance to the overall dissolved concentrations, from most important to lesser importance but still significant. This order is based on existing chemical information and reflects the best technical judgment on the important of individual effects. The prioritization thus represents an optimization of work efforts. As additional ligands, both inorganic and organic, are added to the overall model, the model predictions for actinide solubilities in the complex disposal room brines will converge to the measured values. An order-of-magnitude represents the convergence criterion for the process of building the overall model. In principle, when all effects in the complex systems are included, the model should provide exact predictions. However, the smaller effects are considered minor within the uncertainty needed for the WIPP Performance Assessment.

The overall model is developed from individual models of simple chemical systems. For example, the initial Np(V) solubility model in $NaClO_4/NaCl/Na_2CO_3$ media was developed from three separate systems: (1) NpO_2^+-$NpO_2OH(aq)$-$NpO_2(OH)_2^-$-Na^+-ClO_4^--H^+-OH^--$NpO_2OH(am)$-$NpO_2OH(aged)$-H_2O; (2) NpO_2^+-Na^+-Cl^--H^+-OH^--H_2O; and (3) NpO_2^+-$NpO_2(CO_3)^-$-$NpO_2(CO_3)_2^{3-}$-$NpO_2(CO_3)_3^{5-}$-Na^+-ClO_4^--H^+-OH^--$NaNpO_2CO_3(s)$-H_2O, as documented in Novak and Roberts (1995). The combination of these three systems was used to model the NpO_2^+- $NpO_2OH(aq)$- $NpO_2(OH)_2^-$- $NpO_2(CO_3)^-$- $NpO_2(CO_3)_2^{3-}$- $NpO_2(CO_3)_3^{5-}$- Na^+- Cl^-- H^+- OH^-- $NpO_2OH(am)$- $NpO_2OH(aged)$- $NaNpO_2CO_3(s)$- H_2O system. The combined system model has order-of-magnitude accuracy, which is within design specifications, while the individual models, (1), (2), and (3) above, have about a factor of two accuracy. See Novak and Roberts (1995) for details.

The Data Quality Objective (DQO) process is an EPA-endorsed process for describing a problem, the kinds of data needed to solve the problem, and the decisions that are intended to be made as a result of the data (U.S. Environmental Protection Agency, 1994). The DQO process as applied to the Actinide Source Term dissolved concentration submodel is documented here.

5.1 Step 1: State the Problem

DQO guidelines ask for a statement of the potential problem in a larger context. The following is the problem in a large context: does the WIPP comply with applicable U.S. Federal Regulations, particularly in the context of inadvertent human intrusion into the repository after decommissioning? To address this question, one needs to know how much radioactive material could potentially be released from the WIPP disposal facility horizon; what form this radioactivity would assume, i.e., dissolved or colloidal; what fraction of the radioactivity could reach the

Culebra; and how much of it could be transported to the regulatory boundary. The objective of this study is to determine the quantities of actinides that could be dissolved in WIPP brines and transported from the WIPP facility to the accessible environment (U.S. Department of Energy, 1994b).

5.2 Step 2: Identify the Decision

5.2.1 Questions the study will try to resolve

What are the ranges of dissolved actinide concentrations that could exist in WIPP Salado, Castile, and Culebra brines?

5.2.2 Possible actions that will solve the problem

Note that "the problem" here is in the context of the "big picture." The problem is determining whether the WIPP will comply with applicable U.S. Federal Regulations, as determined with the formal Performance Assessment process. Information on the ranges of dissolved actinide concentrations that could exist in WIPP Salado, Castile, and Culebra brines will be used in the WIPP Performance Assessment calculations to compare predicted WIPP performance with the Federal Regulations.

5.3 Step 3: Identify the Input to the Decision

The input to the decision are results of numerical calculations estimating the dissolved concentrations of Th, U, Np, Pu, and Am as a function of the anticipated variability of WIPP brine compositions. The numerical model requires data from laboratory measurements of the chemical properties of Th, U, Np, Pu, and Am as a function of the dominant composition variables in WIPP brines.

5.4 Step 4: Define the Study Boundaries

Information is needed for the potential range of chemical conditions, including hydrogen ion concentration, concentrations of inorganic and organic species present in WIPP brines, and partial pressures of CO_2(g) that may be in contact with the brines. These include the natural brine compositions and the changes in brine compositions caused by interaction between the brine and the TRU wastes emplaced in the WIPP.

5.5 Step 5: Develop a Decision Rule

The parameters of interest: dissolved actinide concentrations

The scale of decision making: Are measurements at laboratory scale adequate? We believe that laboratory-scale measurements, in conjunction with "pilot plan" scale measurements (Phillips and Molecke, 1993; Villarreal and Phillips, 1993) are sufficient to address this issue.

The action level: Are there minimum concentrations below which dissolved actinide concentrations are unimportant? Yes. Sensitivity studies performed as part of the WIPP Performance Assessment indicated that dissolved actinide concentrations below 10^{-11} Molar will not have a significant impact and are thus unimportant. Because most laboratory measurement techniques have detection limits in the range from 10^{-9} to 10^{-6} Molar, we are unable to measure concentrations with sufficient limits to be measuring in the unimportant range.

The alternative actions: If dissolved actinide concentrations, when characterized, indicate that the WIPP does not comply with U.S. Federal Regulations, redesign of the disposal system may be required.

5.5.1 The Decision Rule

Quantification of the dissolved concentrations of actinides correlated to brine composition is the target input for the WIPP Performance Assessment. Should these concentrations lead to noncompliance with U.S. Federal Regulations, the concentrations can guide the choice of waste type selection or engineering modifications to create conditions in the WIPP with lower dissolved actinide concentrations, and an improved chance for compliance.

5.6 Step 6: Specify Limits on Decision Errors

True values and measured values of the parameters of interest (see Step 5) differ by the total error, which is a combination of sampling design errors, sample collection errors, instrumental errors, and method errors. These errors are described as uncertainties in the measurements. In development of the Actinide Source Term dissolved concentration submodel, the maximum tolerable uncertainty with any measurement is generally 100% (a factor of 2) at most.

5.7 Step 7: Optimize the Design for Obtaining Data

Much information on dissolved actinide concentrations exist in the published literature, as documented in other sections of this Test Plan, especially "Design Process for the Actinide Source Term Dissolved Concentration Submodel" (Section 6.3). The design for experiments and model development was created assuming that all relevant and reliable literature data would be fully incorporated into the dissolved concentration submodel. In addition, the design for obtaining data and model was optimized considering the Decision Rule and constraints imposed by decision errors, schedule, cost, and quality assurance requirements (see Section 12.0).

6.0 EXPERIMENTAL PROGRAM

The objective of this study is a numerical model describing the dissolved concentrations of the actinides Am, Np, Pu, Th, and U as a function of brine composition. These dissolved concentrations will be limited by solubility-controlling solids for each of the actinides in the brines. The actinides Am, Np, Pu, Th, and U were determined to be most important with respect to repository performance in the 1992 Performance Assessment calculations (Helton et al., 1992). The actinide oxidation states of interest are Am(III), Am(V), Pu(III), Pu(IV), Pu(V), Pu(VI), Th(IV), U(IV), U(VI), Np(IV), Np(V), and Np(VI). The important ions in WIPP brines are H^+, Na^+, K^+, Mg^{2+}, OH^-, Cl^-, CO_3^{2-}, SO_4^{2-}, and Ca^{2+}. The organic ligands that are contained in the wastes and are believed to be important are acetate; ascorbate; citrate; ethylenediaminetetraacetic acid (EDTA); α-hydroxyisobutyrate; lactate; oxalate; 8-hydroxyquinoline (oxine); 1,10-phenanthroline; and 1-thenoyl-3,3,3-trifluoroacetone (TTA) (Brush, 1990).

The anions phosphate (PO_4^{3-}) and fluoride (F^-) and the cations Al^{3+} and Fe^{2+}/Fe^{3+}, may be important at influencing dissolved actinide concentrations. (The anions are a component of the waste; the cations will be formed through corrosion of aluminum and iron metals.) Parameters for these species will not be determined, but will be included as they are available in the literature. The assumption that these effects are unimportant will be challenged as part of the experimental program in tests with actual and simulated WIPP brines.

The expected temperature of the WIPP disposal rooms during the regulatory time frame is 28°C (Section 1.10.1, Brush, 1990). The small differences in thermodynamic properties between 25°C and 28°C are well within acceptable uncertainty for the WIPP system. For these reasons, the small differences in properties over this temperature range are not significant for the WIPP, and all information will be developed for 25°C. Literature information taken at ~20°C is acceptable for model parameterization as well. This is a retroactive Data Quality Objective for temperature. All information that will beused to develop the model will either be taken at 25±5°C or adjusted using established thermodynamic temperature relationships to 25°C.

The development of the AST dissolved actinide concentration submodel is actually the development of a data base for use with existing computer codes. The mathematical equations necessary to describe equilibrium in aqueous/mineral/gaseous systems are fully developed, as are computer codes to implement these equations. What is lacking is the data base, the set of aqueous and solid chemical species containing actinides, the standard chemical potentials of these species, and the specific ion interaction parameters required to describe the interactions between these species and other constituents of brines in WIPP scenarios.

6.1 The "Oxidation State Analogy": Similarities in Chemical Properties of f-Elements in the Same Oxidation State

The strategy for developing dissolved concentration models groups the elements by oxidation state because actinide (and lanthanide) elements in the same oxidation state have similar chemical properties. For example, Am(III) and Pu(III), and the lanthanide Nd(III), form the same aqueous species and isostructural solid phases. Therefore, the detailed chemical measurements required to parameterize the model are only necessary for one of these elements, for example Nd(III), but the resulting model is applicable to Am(III) and Pu(III) as well as Nd(III). The oxidation state analogy allows a significant reduction in the number of experimental systems that need to be examined in order to develop model parameters. For example, Nitsche (1991) uses the oxidation state analogy, specifying that the tables "show the stability constants used . . . for the reactions of neptunium, plutonium, and americium with ligands present in J-13 groundwater. Italic numbers indicate that no data were available and that estimates were used instead; the estimates were derived from other actinide systems of the same oxidation state." Nitsche (1991) applies the oxidation state analogy to actinides in the +III, +IV, +V, and +VI oxidation states.

Elements and oxidation states important to the WIPP Performance Assessment and the development of the WIPP dissolved actinide concentration submodel are shown in Table 2. The oxidation state analogy cannot be applied indiscriminately; its applicability must be demonstrated. The program includes provisions to demonstrate that a model developed from Nd(III), for example, is applicable to Am(III) and Pu(III). Unequivocal demonstration is straightforward and requires fewer resources and less time than the model development itself. There is less total data on the chemical analogy for the +V actinides (PuO_2^+, NpO_2^+, and AmO_2^+) and the +VI actinides (PuO_2^{2+}, UO_2^{2+}, and NpO_2^{2+}). However, available evidence suggests that the oxidation state analogy holds for these systems. In contrast to the +III, +IV, and +VI oxidation states, the analogy for the +IV actinides Pu^{4+}, U^{4+}, Np^{4+}, and Th^{4+} is not quantitative, but nevertheless can serve as a good indicator for the chemical behavior of a system, and can result in a conservative approximation for An(IV) behavior.

Table 2. Elements and oxidation states important to the WIPP Performance Assessment and the WIPP solubility submodel development. Elements/oxidation states in the same column exhibit similar chemical behavior.

Element	+III Oxidation State	+IV Oxidation State	+V Oxidation State	+VI Oxidation State
Plutonium (actinide)	Pu(III)	Pu(IV)	Pu(V)	Pu(VI)
Uranium (actinide		U(IV)		U(VI)
Neptunium (actinide)		Np(IV)	Np(V)	Np(VI)
Americium (actinide)	Am(III)		Am(V)	
Thorium (actinide)		Th(IV)		
Neodymium (lanthanide)	Nd(III)			

The primary variables influencing chemical behavior in aqueous systems are ionic radius, r, ionic charge, z, and the volumetric charge density, z/r^3. Actinides and lanthanides, the f-elements, have approximately the same ionic radius when in the same oxidation state because the f electron shell is inside the s electron shell, and it is the s electron shell that determines the ionic radius. There is a small variation in ionic radius among f-elements with the same oxidation state, but this variation typically results in only a small difference in chemical properties. The oxidation state analogy is supported by data in the literature. However, although chemical principles provide convincing arguments for the validity of the analogy, experimental verification is still necessary. Such verification is a part of this program.

Chemical behavior depends on the element, but not significantly on the isotope of the element. Thus, the same solubility model applies to ^{238}Pu and ^{239}Pu, for example. The chemical equilibrium model will thus calculate the total dissolved Pu, for example. The radioactivity of the total dissolved Pu, for example, is calculated from the known abundance of each of the isotopes in the system.

6.2 Experiments for Developing Chemical Data for Thermodynamic Models

The process for developing a thermodynamic model for dissolved actinide concentration is to (1) design experiments to measure the change in a chemical property of the actinide as a function of salt or hydrogen ion concentration; (2) perform the experiments; (3) generate model parameters through data analysis using a thermodynamic formalism and a data base consistent with existing information on the properties of the actinide; and (4) test the model through comparison with additional experiments not used to obtain the model parameters.

Planned and completed experiments for the Actinide Source Term dissolved concentration model are given in detail in Appendix B. This section provides a general discussion of the types of experiments that can be performed to get the information needed to develop thermodynamic models.

Experiments the can generate useful information for parameterizing thermodynamic models include isopiestic measurements, electromotive force (EMF) measurements, calorimetry, osmotic pressure measurements, solubility, and extraction measurements. The choice of an experimental system depends on many factors such as concentration ranges, particular properties of interest, hazards, and contamination risks. Thermodynamics is a powerful analysis tool, and the parameters generated from each experimental type are applicable to the other experimental types. Many different sources and kinds of data are necessary to develop a robust model.

Solubility experiments and extraction experiments are particularly useful for developing actinide solubility models. Potentiometric titration and spectrophotometric measurements may also be useful. The essential features of these experiments are described below.

6.2.1 Solubility Experiments

Solubility experiments measure the solubility of a particular solid phase of an actinide in a particular oxidation state as a function of salt concentration. These experiments are designed to hold constant as many variables as possible, to determine (1) the solubility controlling solid(s); (2) the important aqueous species; and (3) the variation of activity coefficients. Typically these experiments vary either the hydrogen ion concentration, or the concentration of the background electrolyte, or both. The solid phase in the experiments is characterized and identified, and the variation in dissolved concentration is measured as a function of the independent variables. Steady-state conditions may be reached from oversaturation or undersaturation, as described below. Solubility experiments provide information on which chemical species are important, and provide values for μ_f^o for the solids that form, μ_f^o for the important aqueous species, and Pitzer parameters $\beta^{(0)}$, $\beta^{(1)}$, $\beta^{(2)}$, and C^{ϕ}, as needed to describe the system.

6.2.1.1 Solubility Experiments from Oversaturation

Solubility experiments from oversaturation are usually performed when the identity of the solid phase that will form is unknown. The actinide in the oxidation state of interest is added in dissolved form at a concentration expected to be larger than the solubility limit for the experimental conditions. The supersaturated aqueous phase is unstable, causing the actinide to form a solid precipitate. (If no precipitation occurs, the actinide concentration is probably not above the solubility limit for the experimental conditions, so more actinide is added.) The concentration of actinide in the aqueous phase will decrease to a constant value as dynamic steady-state conditions are reached. The actinide precipitates in whatever solid phase is most favored under the particular experimental conditions. The solid phase thus formed is kinetically favored (metastable), and is not necessarily the thermodynamically most stable phase under these conditions. This adds conservatism to the conceptual model because it provides the solid phases that form most rapidly, which may have higher solubilities than the thermodynamically more stable solids. These experiments are particularly useful when the identity of the favored phase is unknown. However, these experiments tend to require more time to reach steady-state than undersaturation experiments.

6.2.1.2 Solubility Experiments from Undersaturation

Solubility experiments from undersaturation are the reverse of experiments from oversaturation. A solid phase is added to the reaction vessels, and the solid dissolves until steady-state conditions are reached. These experiments are particularly useful when one knows the identity, but obviously not the solubility, of the (meta-)stable solid phase for the given conditions. Addition of the (meta-)stable solid phase decreases the time needed to reach steady-state. Undersaturation experiments are usually used after some preliminary characterization of the system, such as that provided by oversaturation experiments.

It is possible that the added solid is not the (meta-)stable phase under the experimental conditions. In this case, the solid will dissolve and oversaturate the aqueous phase with respect to the actual (meta-)stable solid phase, which will then precipitate. Experiments of this type are a hybrid between oversaturation and undersaturation experiments, and will likely require more time than undersaturation experiments. However, because a solid phase is present initially, the (meta-)stable solid may precipitate faster because of the availability of nucleation sites. Thus, these experiments are likely to require less reaction time to reach steady-state than oversaturation experiments. This process is demonstrated for the conversion of $Nd_2(CO_3)_3(s)$ to $NaNd(CO_3)_2 \bullet 6H_2O(s)$ in $NaHCO_3$ and Na_2CO_3 solutions by Rao et al. (1994).

6.2.2 Extraction Experiments

Extraction experiments measure the partitioning of an actinide between an aqueous and an organic phase. There are no solid phases in these experiments, thus no information about solubilities *per se* is obtained. Rather, these experiments focus on the formation of complexes, both inorganic and organic, and the variation of activity coefficients with salt concentration. The information determined in extraction experiments complements data from solubility experiments, confirming and substantiating data from solubility experiments. Extraction experiments can be used to measure chemical conditions not easily examined in solubility experiments. Equilibrium between the immiscible phases is attained much more rapidly than solubility experiments (hours versus days or weeks), and thus more data can be generated. In addition, more salt concentrations can be examined, resulting in better definition of the dependence of actinide species concentrations on salt concentration. Extraction experiments yield information on μ_f^o for the important aqueous species, and Pitzer parameters $\beta^{(0)}$, $\beta^{(1)}$, $\beta^{(2)}$, and C^{ϕ}, as needed to describe the system.

6.2.3 Potentiometric Titration Experiments

Potentiometric titration experiments measure the dissociation of a weak acid or base, the hydrolysis of a metal ion, or the complexation between metal ions and ligands. Potentiometry is the most commonly used method for the determination of dissolution constants, and is complementary to spectroscopic methods such as ultraviolet (UV) absorption spectroscopy, near infrared (near IR) absorption spectroscopy, and Raman spectroscopy. The equilibrium constants determined through potentiometric titration at different background electrolyte concentrations can be analyzed together to yield the standard chemical potentials and ion interaction parameters for the species present.

6.2.4 Spectrophotometric Measurements

Extraction experiments can not be used to quantify solution chemical behavior in systems when the solubility of the organic ligand in the organic phase is high and/or a synergistic effect is present. Absorption spectrophotometry can be applied to these systems as an alternative experimental method, or to complement extraction or potentiometric experiments. Spectroscopy measures the absorbance of individual species in solution as a function of wavelength. Different dissolved species absorb at different wavelengths, and the absorption changes with changes in the chemical environment. Changes in the absorption spectrum of a ligand as a function of hydrogen ion concentration in the aqueous phase can be used to determine the ligand dissociation (protonation) constants. Changes in the absorption spectrum of a complex with a constant concentration of a ligand or metal ion can be used to determine apparent stability constants for the complex formed. The apparent stability constants for several background electrolyte concentrations can, in turn, be used to calculate the standard chemical potentials for the various chemical species involved, as well as ion interaction parameters for these species.

6.3 Design Process for the Actinide Source Term Dissolved Actinide Concentration Submodel

The Actinide Source Term Program solubility submodel was designed through extensive review of (1) the information needed for the Performance Assessment, (2) existing work for the WIPP, and (3) the literature. The primary contributors to this process were Hans W. Papenguth (6748), Craig F. Novak (6748), and E. James Nowak (6719) of Sandia National Laboratories (SNL). The results of this review were incorporated into the Actinide Source Term Model Development Plan, which was reviewed by members of the radiochemistry, geochemistry, colloid chemistry, and geochemical modeling communities, as well as by the Department of Energy WIPP Project Integration Office (DOE/WPIO) and the National Academy of Sciences WIPP Committee. The reviewers included Tex Blomeke (National Academy of Sciences WIPP Committee), Carol Bruton (Lawrence Livermore National Laboratory [LLNL]), Gregory R. Choppin (Florida State University [FSU]), Fred Ernsberger (National Academy of Sciences WIPP Committee), Rodney C. Ewing (National Academy of Sciences WIPP Committee), Andrew R. Felmy (Pacific Northwest Laboratory [PNL]), Bruce Honeyman (Colorado School of Mines), Ken Jackson (LLNL), Bob Mesmer (Oak Ridge National Laboratory), Frank Millero (University of Miami), Heino Nitsche (then at Lawrence Berkeley National Laboratory [LBNL]), Cynthia E.A. Palmer (LLNL), Dhanpat Rai (PNL), Robert J. Silva (LLNL), and Tom Wolery (LLNL). The comments from the reviewers shaped and refined the program into that which is documented here. The Model Development Plan has evolved into this Test Plan and the Test Plan for colloids in the WIPP system (Papenguth and Behl, 1995). The description of the AST dissolved concentration model in this document reflects all the input received in the review process.

6.4 Data Base for NonActinide Model

The Pitzer formalism has been parameterized for the brine system, i.e., concentrated electrolytes without actinides, in Harvie et al. (1984) and Felmy and Weare (1986), referred to in this document as HMW/FW. These articles provide extensive description of the parameter fitting process and the data used to determine these parameters. More importantly, the HMW/FW articles demonstrate the utility of this model for predicting equilibrium solid phases and brine compositions for various systems. The Pitzer formalism is the only existing thermodynamic model that is applicable to the concentrated electrolytes and evaporates. Other models such as the SIT (Grenthe et al., 1992) may be applicable to concentrated electrolytes, but this has not yet been demonstrated, and such demonstration would require considerable time and resources to accomplish.

The HMW/FW data base is the data base for the nonactinide portion of the AST dissolved actinide concentration submodel, and is also being used by the WIPP Gas Generation Program (Brush, 1990). The required species, with standard chemical potentials, and specific ion interaction parameters are listed in the preliminary data base for An(III) and An(V) in Appendix C.

Models for the chemistry of phosphate, though not in HMW/FW, were developed and added to the model in much the same manner as the actinides are being included by the ASTP. Chemical properties of F^- and PO_4^{3-} were measured and modeled using values for free energies of formation and ion interaction parameters from the HMW/FW data base when needed for H^+, OH^-, H_2O, Na^+, K^+, etc. Thus these data are consistent.

The actinide chemical models use the values from HMW/FW as a basis for development of parameters for the actinides. This ensures consistency among all parameters generated, allowing all to be used simultaneously, as required to model the WIPP actinide chemistry system.

6.5 Systems under Study, Including Status of Information Available

The general approach to developing thermodynamic solubility models is the same for all oxidation states and all ligands. The five steps are

(1) evaluate existing literature information for the actinide chemical properties;

(2) design the additional experiments necessary to develop the model for WIPP purposes;

(3) perform the experiments;

(4) analyze the experimental data, together with reliable data from the literature, to generate model parameters; and

(5) challenge the model and model assumptions by comparison with data from independent experiments similar to the systems to be modeled.

Step (5), which includes demonstrating the model developed for +III, or +V, or +VI actinides, applies to all the +III, +V, and +VI actinides relevant to the WIPP, as assumed in the oxidation state analogy (see Section 6.1). Because of the nonquantitative nature of the oxidation state analogy for the solubilities of +IV actinides, the demonstration is that the U(IV) model will predict higher dissolved concentrations for Np(IV) and Pu(IV) than are actually measured, and thus use of the U(IV) model for predicting Np(IV) and Pu(IV) concentrations would be conservative.

Throughout this document, the “chemical symbols” An(III), An(IV), An(V), and An(VI) are used to refer to a general actinide in the +III, +IV, +V, and +VI oxidation states, respectively.

6.5.1 The +III actinides (and lanthanides): Pu(III) and Am(III) (and Nd[III])

The +III actinides of interest to the WIPP are Am(III) and Pu(III). The lanthanide Nd(III) typically also has the same chemical properties. The An(III) model, generated as given in Section 6.4, will be followed by demonstrations where the model accurately describes the chemical behavior of both Am(III) and Pu(III), by a combination of comparisons with existing literature information and comparisons with additional experiments designed specifically to perform this demonstration. For the +III system, working with Nd(III) provides a particular advantage because Nd is not radioactive, which simplifies the laboratory work.

Once developed, the model will be tested against data that was not used to develop the model, including solubility experiments in complex synthetics WIPP brines (see for example, Khalili et al., 1994) and against selected data from the Actinide Source-Term Waste Test Program (STTP) (Phillips and Molecke, 1993; Villarreal and Phillips, 1993). The model will be used to predict the behavior of the actinides in the WIPP system, such as is shown for the simple example of the solubility of Am(III) in a closed system of SPC brine as a function of pmH (Appendix E).

Data for the +III actinides and lanthanides are the most extensive of all the oxidation states of interest. The An(III) model is being developed simultaneously from individual data for the Pu(III), Am(III), and Nd(III) systems. The An(III) model is parameterized in the Pitzer formalism for the (pseudo-)elements Na, Cl, SO_4, CO_3, and PO_4 as of March 1995. The model requires the aqueous species Am^{3+}, $AmCO_3^+$, $Am(CO_3)_2^-$, $Am(CO_3)_3^{3-}$, $Am(OH)_2^+$, and $Am(OH)_3^0$, and the solid species $Am(OH)_3(s)$, $AmOHCO_3(s)$, $NaAm(CO_3)_2(s)$, and $AmPO_4(c)$ to explain existing data. These are represented by the following reactions, as written for Am(III):

$$Am^{3+} + CO_3^{2-} \leftrightarrow AmCO_3^+ \qquad (16)$$

$$Am^{3+} + 2\,CO_3^{2-} \leftrightarrow Am(CO_3)_2^- \qquad (17)$$

$$Am^{3+} + 3\,CO_3^{2-} \leftrightarrow Am(CO_3)_3^{3-} \qquad (18)$$

$$Am^{3+} + 2\,OH^- \leftrightarrow Am(OH)_2^+ \qquad (19)$$

$$Am^{3+} + 3\,OH^- \leftrightarrow Am(OH)_3^0 \qquad (20)$$

$$Am(OH)_3(s) \leftrightarrow Am^{3+} + 3\,OH^- \qquad (21)$$

$$\mathrm{AmOHCO_3(s)} \leftrightarrow \mathrm{Am^{3+}} + \mathrm{OH^-} + \mathrm{CO_3^{2-}} \qquad (22)$$

$$\mathrm{NaAm(CO_3)_2{\bullet}6H_2O(s)} \leftrightarrow \mathrm{Na^+} + \mathrm{Am^{3+}} + 2\ \mathrm{CO_3^{2-}} + 6\ \mathrm{H_2O} \qquad (23)$$

$$\mathrm{AmPO_4(c)} \leftrightarrow \mathrm{Am^{3+}} + \mathrm{PO_4^{3-}} \qquad (24)$$

Equations 16-24 represent the minimum set of orthogonal reactions for americium(III) species; other reactions among the species can be found through linear combination of these reactions. The parameterizations of data are documented in source reports as follows: Pu^{3+}-Na^+-Cl^- in Felmy et al. (1989); Am^{3+}-Na^+-HCO_3^- in Felmy et al. (1990); Am^{3+}-Na^+-SO_4^{2-}-PO_4^{3-} in Rai et al. (1994); $Am(OH)_2^+$ and $Am(OH)_3^0$ in Rai et al., 1992b; and Nd^{3+}-Na^+-CO_3^{2-} in Rao et al. (1994). A summary of these source reports, including standard chemical potentials and ion interaction parameters for this model are given in Novak (1995a; 1995b) in Appendix D.

The Am^{3+} model as described here is fully developed with only a small amount of work remaining to test the model. The first step is showing whether this model adequately represents solubility data in mixed $NaCl/NaHCO_3$ and $NaCl/Na_2CO_3$ media, which is testing the initial assumption that ternary interactions, e.g., Na^+-$Am(CO_3)_3^{3-}$-Cl^-, are unimportant. The second step is testing the predictive ability of the model by comparison with solubility experiments in complex synthetic WIPP brines.

Experiments planned for the remainder of FY95 and the first half of FY96, along with a summary of accomplishments for the Actinide Source Term dissolved concentration submodel development, are documented in Appendix B.

6.5.2 The +IV actinides: Pu(IV), U(IV), Np(IV), and Th(IV)

The +IV actinides of interest to the WIPP are Pu(IV), U(IV), Np(IV), and Th(IV). Models for Th(IV) and U(IV) solubility in WIPP brines are being generated as given in Section 6.4. Actinides in the +IV oxidation data exist as highly charged tetravalent ions Pu^{4+}, U^{4+}, Np^{4+}, and Th^{4+}. The variation in charge to radius ratio for the tetravalent actinides is greater than for other oxidation states, and larger differences in the chemical behavior among the +IV actinides is expected. Therefore, individual models for Th(IV) and U(IV) are being developed, using the oxidation state analogy as a guide to expected properties, rather than relying on it more extensively. These models will be evaluated against data for Pu(IV) and Np(IV) solubility to demonstrate that they can be used conservatively to predict the chemical behavior of these actinides.

Data for the +IV actinides are the fairly extensive, although more information is required because the oxidation state analogy cannot be as extensively applied to this oxidation state. The major inorganic interactions of the +IV actinides are being studied simultaneously, with full advantage being taken of information being generated for the Power Reactor and Nuclear Fuel Development Corporation of Japan (PNC) by Andy R. Felmy and Dhanpat Rai of PNL.

The Th(IV) dissolved concentration model is parameterized in the Pitzer formalism for interactions in the Na^+-K^+-Cl^--SO_4^{2-} system as of March 1995. This model requires the aqueous species Th^{4+}, $Th(SO_4)_2^0$, and $Th(SO_4)_3^{2-}$, and the solid species $Th(OH)_4(am)$ to describe the data pertinent to the WIPP (Felmy et al., 1991; Felmy and Rai, 1992; Roy et al., 1992). These chemical species are represented by the following reactions:

$$Th^{4+} + 2\,SO_4^{2-} \leftrightarrow Th(SO_4)_2^0 \qquad (25)$$

$$Th^{4+} + 3\,SO_4^{2-} \leftrightarrow Th(SO_4)_3^{2-} \qquad (26)$$

$$Th(OH)_4(am) \leftrightarrow Th^{4+} + 4\,OH^- \qquad (27)$$

Equations 25-27 represent the minimum set of orthogonal reactions for thorium(IV) species; other reactions among the species can be found through linear combination of these reactions. The parameterizations of data are documented in source reports as follows: Th^{4+} interactions with Cl^- and the standard chemical potential of

$Th(OH)_4$(am) in Felmy et al. (1991); Th^{4+} interactions with Cl^- extended to 3m in Roy et al. (1992); and Th^{4+} interactions with Na^+, K^+, and SO_4^{2-} in Felmy and Rai (1992). Standard chemical potentials for each species are shown in Table 3; the required Pitzer parameters are shown in Tables 4a and 4b.

Table 3. Standard chemical potentials for the Th(IV) model as of March 1995.

Species	Standard Chemical Potential, μ_f^0/RT	Source Report
Th^{4+}	–284.227	Felmy et al., 1991
$Th(SO_4)_2^0$	–911.69	Felmy and Rai, 1992
$Th(SO_4)_3^{2-}$	–1214.0	Felmy and Rai, 1992
$Th(OH)_4$(am)	–451.4076	Felmy et al., 1991

Table 4a. Pitzer parameters for the Th(IV) model as of March 1995 (Part 1).

Cation-Anion Pair	$\beta^{(0)}$	$\beta^{(1)}$	$\beta^{(2)}$	C^ϕ	Source Report	Status
Na^+-$Th(SO_4)_3^{2-}$	0.12	-	-	-	Felmy and Rai, 1992	
K^+-$Th(SO_4)_3^{2-}$	0.90	-	-	-	Felmy and Rai, 1992	
Th^{4+}-HSO_4^-	1.44	-	-	-	Felmy and Rai, 1992	needs to be tested
H^+-$Th(SO_4)_3^{2-}$	0.84	-	-	-	Felmy and Rai, 1992	needs to be tested
Th^{4+}-Cl^-	1.092	13.7	–160	–0.112	Roy et al., 1992	

Table 4b. Pitzer parameters for the Th(IV) model as of March 1995 (Part 2).

Parameter	Value	Source	Status
$\Theta_{H^+-Th^{4+}}$	0.60	Roy et al., 1992	
$\Psi_{H^+-Th^{4+}-Cl^-}$	0.37	Roy et al., 1992	
$\lambda_{Th(SO_4)_2^0-HSO_4^-}$	0.68	Felmy and Rai, 1992	needs to be tested
$\lambda_{Th(SO_4)_2^0-Cl^-}$	0.29	Felmy and Rai, 1992	needs to be tested

The values for ion interaction parameters in Tables 4a and 4b marked "needs to be tested" were developed using older, less well determined values for ion interactions parameters between Th^{4+} and Cl^- (Felmy et al., 1991). The data used to determine these values must be reexamined with the current best values for Th^{4+}-Cl^- interactions from Roy et al. (1992)

Additional experimental work quantifying dissolved thorium concentrations in concentrated electrolytes has been performed. However, these results have not yet been interpreted with the appropriate thermodynamic formalism needed for the WIPP. This work includes the solubility of $Th(OH)_4$(am) in concentrated NaCl and $MgCl_2$ solutions (performed for SNL/WIPP by Felmy and Rai, PNL); the solubility of $Th(OH)_4$(am) in $NaClO_4/NaHCO_3$ media at constant CO_2(g) pressures (Östhols et al., 1994); and the solubility of $Th(OH)_4$(am) as a function of $NaHCO_3$ and Na_2CO_3 concentrations (Rai et al., 1995). All of these data are in the process of being interpreted with the Pitzer activity coefficient formalism in order to be incorporated into the WIPP Th(IV) dissolved concentration model. The parameters values marked as "needs to be tested" in Table 4 will be either verified or modified as part of this process.

The U(IV) dissolved concentration model is parameterized in the Pitzer formalism for dilute solutions over the pH range 2 to 12 as of March 1995. This model requires the aqueous species U^{4+} and UOH^{3+}, and the solid species $U(OH)_4$(am), which can also be written as $UO_2 \bullet xH_2O$(am), to describe the data pertinent to the WIPP (Rai et al., 1990). These chemical species are represented by the following reactions:

$$U^{4+} + OH^- \leftrightarrow UOH^{3+} \qquad (28)$$

$$U(OH)_4(am) \leftrightarrow U^{4+} + 4\ OH^- \qquad (29)$$

Equations 28-29 represent the minimum set of orthogonal reactions for uranium(IV) species; other reactions among the species can be found through linear combination of these reactions. The parameterizations of data are documented in Rai et al. (1990) and include ion interaction parameters between U^{4+} and Cl^- and between UOH^{3+} and Cl^-, and standard chemical potentials for the chemical species. Note that the ion interaction parameters between U^{4+} and Cl^- were taken directly from the Th^{4+}-Cl^- system by analogy with good success. This suggests that ion interaction parameters developed for Th^{4+} may be applicable to U^{4+} and the other +IV actinides. However, the definitive values for Th^{4+}-Cl^- ion interaction parameters from Roy et al. (1992) were not available when the work of Rai et al. (1990) was prepared. Available standard chemical potentials for U(IV) species are shown in Table 5; the required Pitzer parameters are shown in Table 6.

Table 5. Standard chemical potentials for the U(IV) model as of March 1995.

Species	Standard Chemical Potential, μ_f^0/RT	Source Report	Status
U^{4+}	–214.2	Fuger and Oetting, 1976	
UOH3+	–308.7	Rai et al., 1990	needs to be refit
$U(OH)_4$(am)	–396.4	Rai et al., 1990	needs to be refit

Table 6. Pitzer parameters for the U(IV) model as of March 1995.

Cation-Anion Pair	$\beta^{(0)}$	$\beta^{(1)}$	C^{ϕ}	Source Report	Status
UOH^{3+}-Cl^-	0.17	6.2	0.03	Rai et al., 1990	needs to be refit
U^{4+}-Cl^-	1.014	13.33	–0.1034	Rai et al., 1990; from Pitzer and Mayorga, 1973	needs to be refit

The values for ion interaction parameters in the above tables marked "needs to be refit" were developed using the older, less well determined values for ion interactions parameters between U^{4+} and Cl^- , obtained by analogy with the Th^{4+}-Cl^- system. The data used to determine these values must be refit with the current best values for U^{4+}-Cl^- interactions, taken by analogy from the Th^{4+}-Cl^- interactions in Roy et al. (1992)

Additional experimental work quantifying dissolved uranium concentrations in concentrated electrolytes has been performed. However, these results have not yet been interpreted with the appropriate thermodynamic formalism needed for the

WIPP. This work includes the solubility of $U(OH)_4$(am) in concentrated NaCl and $MgCl_2$ solutions (performed for SNL/WIPP by Felmy and Rai, PNL); the formation of uranium carbonato complexes in sodium perchlorate media using visible and ultraviolet spectroscopy (Bruno et al., 1989); and the solubility of $U(OH)_4$(am) as a function of $NaHCO_3$ and Na_2CO_3 concentrations (Rai et al., 1995). All of these data are in the process of being interpreted with the Pitzer activity coefficient formalism in order to be incorporated into the WIPP U(IV) dissolved concentration model. The parameters values marked as "needs to be refit" in Tables 5 and 6 will be redetermined as part of this process.

Experiments planned for the remainder of FY95 and the first half of FY96, along with a summary of accomplishments for the Actinide Source Term dissolved concentration submodel development, are documented in Appendix B.

6.5.3 The +V actinides: Pu(V), Np(V), and Am(V)

The +V actinides of interest to the WIPP are Pu(V) and Np(V), and perhaps Am(V), which has been identified in concentrated sodium chloride solutions with high alpha activity (Magirius et al., 1985). A model for the +V actinides is being generated as given in Section 6.4. Actinides in the +V oxidation data exist as the oxycations plutonyl, neptunyl, and americyl, PuO_2^+, NpO_2^+, AmO_2^+, respectively. While +V is a dominant oxidation state for plutonium in the environment (Clark et al., 1995), Pu(V) is difficult to study under laboratory conditions because it disproportionates into Pu(IV) and Pu(VI) (see, for example, Nitsche et al., 1992; 1994). However, +V is the dominant neptunium oxidation state under most conditions, which simplifies measurement of Np(V) chemical properties relative to Pu(V). The focus of Pu(V) work will be to demonstrate that the An(V) model, developed primarily from Np(V) measurements, is applicable to Pu(V) and Am(V).

A preliminary model for Np(V) solubility in the Na-CO_3-HCO_3-Cl-ClO_4-H-OH-H_2O system was synthesized from data in the literature augmented with additional experimental data from LBNL (Novak and Roberts, 1995). As discussed in Novak and Roberts (1995), this model represents the best existing tool for estimating Np(V) solubility, and by analogy the solubility of Pu(V) and Am(V), and is successful at predicting the measured solubility of Np(V) in two synthetic WIPP

Culebra brines. The Np(V) model, and by analogy the Pu(V) and Am(V) models, is parameterized for the elements Na, Cl, CO_3, and ClO_4 (the last of which is not important for the WIPP but is invaluable for interpreting data in the literature) as of March 1995. The model requires the aqueous species NpO_2^+, $NpO_2OH(aq)$, $NpO_2(OH)_2^-$, $NpO_2CO_3^-$, $NpO_2(CO_3)_2^{3-}$, and $NpO_2(CO_3)_3^{5-}$, and the solid species $NpO_2OH(am)$, $NpO_2OH(aged)$, and $NaNpO_2CO_3(s)$ to explain the available data. These are represented by the reactions

$$NpO_2^+ + OH^- \leftrightarrow NpO_2OH(aq) \tag{30}$$

$$NpO_2^+ + 2\,OH^- \leftrightarrow NpO_2(OH)_2^- \tag{31}$$

$$NpO_2^+ + CO_3^{2-} \leftrightarrow NpO_2CO_3^- \tag{32}$$

$$NpO_2^+ + 2\,CO_3^{2-} \leftrightarrow NpO_2(CO_3)_2^{3-} \tag{33}$$

$$NpO_2^+ + 3\,CO_3^{2-} \leftrightarrow NpO_2(CO_3)_3^{5-} \tag{34}$$

$$NpO_2OH(am) \leftrightarrow NpO_2^+ + OH^- \tag{35}$$

$$NpO_2OH(aged) \leftrightarrow NpO_2^+ + OH^- \tag{36}$$

$$NaNpO_2CO_3(s) \leftrightarrow Na^+ + NpO_2^+ + CO_3^{2-} \tag{37}$$

Equations 30-37 represent the minimum set of orthogonal reactions for neptunium(V) species; other reactions among the species can be found through linear combination of these reactions. The parameterizations of data are documented in the source report Novak and Roberts (1995). Standard chemical potentials for each species are shown in Table 7; the required Pitzer parameters are shown in Table 8. These numbers are also presented in Novak (1995a; 1995b) in Appendix D.

Experiments planned for the remainder of FY95 and the first half of FY96, along with a summary of accomplishments for the Actinide Source Term dissolved concentration submodel development, are documented in Appendix B.

Table 7. Standard chemical potentials available for Np(V) species.

Species	μ_f^o/RT	Species	μ_f^o/RT
$NpO_2OH(am)$	–452.642	$NaNpO_2CO_3(s)$	–713.707
$NpO_2OH(aged)$	–454.010	$NpO_2CO_3^-$	–594.492
$NpO_2OH(aq)$	–438.518	$NpO_2(CO_3)_2^{3-}$	–808.403
$NpO_2(OH)_2^-$	–505.829	$NpO_2(CO_3)_3^{5-}$	–1019.918

Table 8. Specific ion interaction parameters available for the Np(V) solubility model.

$\beta^{(0)}_{NpO_2^+-ClO_4^-}$	0.312	$\beta^{(0)}_{Na^+-NpO_2CO_3^-}$	0.161	$\beta^{(0)}_{Na^+-NpO_2(CO_3)_3^{5-}}$	1.97
$\beta^{(0)}_{NpO_2^+-Cl^-}$	0.169	$\beta^{(0)}_{Na^+-NpO_2(CO_3)_2^{3-}}$	0.407	$\beta^{(1)}_{Na^+-NpO_2(CO_3)_3^{5-}}$	16

6.5.4 The +VI actinides: Pu(VI), U(VI), and Np(VI)

The +VI actinides of interest to the WIPP are Pu(VI), U(VI), and perhaps Np(VI). Solubility model for +VI actinides are being generated as given in Section 6.4. Actinides in the +VI oxidation data exist as the oxycations plutonyl, uranyl, and neptunyl, PuO_2^+, UO_2^+, NpO_2^+, respectively. The +VI oxidation state is dominant for uranium except under highly reducing conditions, and is one of the dominant oxidation states for plutonium under oxic conditions.

A great deal of information about U(VI) chemistry is available in the literature, as summarized in the Nuclear Energy Agency (NEA) review of uranium chemistry (Grenthe et al., 1992). Grenthe et al. (1992) primarily contains stability (formation) constants for uranyl complexes with hydroxide and carbonate, which are equivalent to standard chemical potentials. However, this information has not yet been interpreted in a manner consistent with the Pitzer formalism, and thus is not yet usable for a WIPP U(VI) solubility model. Specific ion interaction parameters exist for UO_2^{2+} with Cl^-, SO_4^{2-}, ClO_4^-, and NO_3^- (Pitzer, 1991), given in Table 9. The perchlorate and nitrate parameters may not be directly useful for application to the WIPP, but are invaluable for interpreting existing data taken in these media. The source reports for these parameters are being evaluated to assess the interpretation methods for these data; this information may be usable as it stands. The literature information for solubility controlling solids is less complete, and additional experimental work must be performed to identify and characterize these solids in the WIPP system, and generate values for the standard chemical potentials of these solids.

Scoping experiments designed to determine the solids controlling the dissolved concentration of uranyl ion in concentrated NaCl, Na_2SO_4, and Na_2CO_3 electrolytes have been completed at LLNL. This information was used to focus additional experimental work to develop the U(VI) dissolved concentration submodel on uranyl carbonate complexation with carbonate.

Experiments planned for the remainder of FY95 and the first half of FY96, along with a summary of accomplishments for the Actinide Source Term dissolved concentration submodel development, are documented in Appendix B.

Table 9. Pitzer parameters available for the U(VI) model as of March 1995.

Cation-Anion Pair	$\beta^{(0)}$	$\beta^{(1)}$	$\beta^{(2)}$	C^{ϕ}	Source
UO_2^{2+}-Cl^-	0.4274	1.644	-	–0.03686	p. 105, Pitzer, 1991
UO_2^{2+}-SO_4^{2-}	0.322	1.827	not determined	–0.0176	p. 109, Pitzer, 1991
UO_2^{2+}-ClO_4^-	0.6113	2.144	-	0.02169	p. 105, Pitzer, 1991
UO_2^{2+}-NO_3^-	0.4607	1.613	-	–0.03154	p. 105, Pitzer, 1991

6.5.5 Organic Ligand Complexation with Actinides

Organic ligands are treated in the same manner as inorganic ligands within the Actinide Source Term dissolved concentration model. Reactions such as reduction of organic carboxylic acids to ketones or alcohols are not included because they do not have an impact on the actinide dissolved concentrations. Rather, only protonation/deprotonation of the organic ligands (acid/base behavior) is included as needed, as well as ionic interactions with actinides and brine cations such as Na^+ and Mg^{2+}. Reactions converting organic ligands to other forms are catalyzed by processes external to aqueous chemistry, such as microbial action or radiolysis. These effects are being evaluated for importance by the WIPP Gas Generation Program.

The organic ligands that are expected to significantly enhance dissolved actinide concentrations are acetate, citrate, EDTA, lactate, and oxalate. The complexation of these organic ligands with Am^{3+}, Th^{4+}, NpO_2^+, and UO_2^{2+} is being quantified using extraction methods. The actinides are being measured in parallel in the sequence acetate, oxalate, lactate, citrate, and EDTA, or approximately in order of the chemical complexity of each system. This ordering allows experience from measuring the chemically simpler systems to be applied to the more complex chemical systems.

Experiments planned for the remainder of FY95 and the first half of FY96, along with a summary of accomplishments for the Actinide Source Term dissolved concentration submodel development, are documented in Appendix B.

6.6 Determinations of Oxidation States of Actinides that will Exist in the WIPP Environment

A meeting to discuss actinide oxidation states under anticipated WIPP disposal room conditions was held at Sandia on 3 May 1995. Attending the meeting were DOE Carlsbad Area Office (DOE/CAO) personnel, SNL personnel, contractors to the SNL AST work, and other scientists working with actinide elements and nuclear waste disposal issues. One of the conclusions of this meeting was the agreement that David L. Clark (LANL), Andrew R. Felmy (PNL), and Dhanpat Rai (PNL) would prepare a joint proposal for work addressing the WIPP information need for actinide oxidation states. Ruth F. Weiner (6747) was selected as the

Sandian responsible for technical management of this work. A draft proposal has been prepared and is being negotiated among the concerned parties.

6.7 Tests of the Conceptual Model Against Experiments with Actual TRU Waste

The tests with actual TRU wastes (the Source-Term Waste Test Program, see Phillips and Molecke, 1993; Villarreal and Phillips, 1993) are an additional source of information about the oxidation state distributions of actinides in the WIPP system. A conservative model suggests either postulating the most mobile oxidation states or sampling the distribution of actinide oxidation states could be modeled using a probabilistic sampling method. The sum of the mass of each element in each oxidation state always sums to the total mass of the element, making a bounded space.

The final component of the ASTP is a suite of tests with actual transuranic (TRU) waste, called the Actinide Source-Term Waste Test Program (STTP) (Phillips and Molecke, 1993; Villarreal and Phillips, 1993). In these tests, various types of actual TRU wastes from Los Alamos National Laboratory will be submerged in synthetic Salado and Castile brines inoculated with microbial populations from in and near the WIPP Site. The types of wastes include relatively homogeneous pyrochemical salts, sludges, cemented sludges, and highly heterogeneous mixtures of metals and combustible materials. Individual test containers of these waste types will be subjected to a range of conditions including large sorbents/colloid concentrations, high organic ligand concentrations, high CO_2(g) pressures, low metallic iron content, and high radiolysis fields. These experiments are not designed to examine the coupled processes of waste/brine interactions for all waste types and all anticipated chemical conditions. Rather, the experimental conditions were selected as those most likely to promote higher dissolved and/or colloidal actinide concentrations. These tests are too complicated to use for developing models for mobile actinide concentrations, but they approach the set of expected chemical conditions in the repository as closely as can be simulated at a "pilot plant" scale. These test provide a "reality check" of the phenomena that can produce actinides in mobile form.

7.0 COMPUTER CODES FOR THERMODYNAMIC MODEL DEVELOPMENT AND WIPP APPLICATION

7.1 Computer Codes in Use for the WIPP Actinide Source Term

Computer codes are tools for performing mathematical calculations. There are two types of codes required for development of the AST dissolved concentration submodel: a fitting code for calculating values of standard chemical potentials and specific ion interaction parameters, and a code that predicts chemical behavior based on WIPP scenarios and these calculated parameters.

The central capability of both types of codes is the minimization of the total free energy for the chemical system. The total free energy, ΔG, is given by

$$\Delta G = \sum_{i=1}^{N_s} \mu_{f,i}^{o} + RT \ln a_i \qquad (38)$$

where the summation extends over all the N_s chemical species present regardless of phase. A mathematically equivalent statement of this minimum specifies that the free energy of each reaction is zero (compare with Equation 3). In addition, many equilibrium codes contain mass balance constraints, meaning that the mass of an element in all species and phases must sum to a user-specified total amount. These two conditions, equilibrium and mass balance, result in a set of nonlinear algebraic equations that must be solved simultaneously to calculate the equilibrium conditions. There are numerous different algorithms for calculating chemical equilibrium, as summarized by Smith and Missen (1982).

In addition to the ability to solve the basic equations for calculating chemical equilibrium, a computer code must contain instructions for calculating activity coefficients using the Pitzer formalism. The data base accompanying the code, if any, will necessarily be replaced with a data base composed of the parameters in the existing HMW/FW data set (Harvie et al., 1984; Felmy and Weare, 1986), augmented with the parameters for dissolved actinides documented in this report and those in development by the AST dissolved concentration model program.

The computer code NONLIN is available for fitting the thermodynamic parameters required by the AST dissolved actinide concentration submodel program. NONLIN was developed by A.R. Felmy, and uses the MINPACK nonlinear least-squares programs in combination with a chemical equilibrium program based on the Gibbs free-energy minimization procedure of Harvie et al. (1987). NONLIN has been used to interpret data for the majority of actinide systems in concentrated electrolytes, including the articles by Felmy and Rai (1992), Felmy et al. (1989, 1990, 1991), Novak and Roberts (1995), Rai et al. (1990; 1994), Rao et al. (1994), and Roy et al. (1992). Because of its importance in developing parameters, NONLIN is being brought to the Sandia WIPP quality assurance requirements as defined in WIPP Quality Assurance Procedure 19-1, Rev. 2.

There are several existing computer codes capable of calculating chemical equilibrium using standard chemical potentials and Pitzer specific ion interaction parameters. These codes, included here for completeness, are EQ3/6 (Wolery, 1992), FMT (see for example the memoranda in Appendices A and C), PHRQPITZ (Plummer et al., 1988), and REACT (Bethke, 1994). All of these codes are capable of making the calculations required for generating information for the Performance Assessment regarding dissolved actinide concentrations in WIPP scenarios. However, for ease of use and code Quality Assurance considerations, only NONLIN and FMT are being used in Actinide Source Term dissolved concentration model development, and are presently being qualified to Sandia WIPP quality assurance requirements as defined in WIPP Quality Assurance Procedure 19-1, Rev. 2.

The development of parameters for describing the dissolved concentrations of actinides in brines is independent of any computer code. These parameters will produce the same computational results using any computer code that correctly implements aqueous thermodynamic equilibrium calculations using the Pitzer activity coefficient formalism.

7.2 Making Predictions for WIPP Scenarios

The prediction of dissolved actinide concentrations for Am(III) in Salado simulant SPC Brine in a closed system is presented in Appendix A. This illustration shows different solubility-controlling solid phases for Am(III) depending on the pmH: $AmOHCO_3(s)$ and $Am(OH)_3(s)$ for the range $8<pmH<8.6$, and

$Am(OH)_3(s)$ for pmH>8.6. This system has only a small molality of total inorganic carbon (TIC), m_{TIC}, where

$$m_{TIC} = m_{CO_3^{2-}} + m_{HCO_3^-} + m_{CO_2(aq)} + m_{CaCO_3(aq)} + m_{MgCO_3(aq)}$$

and is closed to entry of additional $CO_2(g)$ from the gas phase. The hydroxide solid $Am(OH)_3(s)$ likely is the predicted solubility-controlling solid because of the small amount of TIC. In systems containing larger amount of TIC, or open to a gas phase containing $CO_2(g)$ as expected in some WIPP scenarios, the expected solubility-controlling solid at high pmH values is either $AMOHCO_3(s)$ or $NaAm(CO_3)_2 \bullet 6H_2O(s)$. Simulations for systems that more closely approximate the expected post-closure conditions in the WIPP are in progress. Model calculations such as these will be used to construct look-up tables giving Am(III) dissolved concentrations as a function of the important chemical variables in the WIPP disposal rooms, for use in Performance Assessment calculations.

The following example illustrates the steps needed to create lookup tables of actinide concentrations as a function of brine composition. Assume Salado brine contacts a cellulosic TRU waste form containing organic ligands with a calcium hydroxide backfill. Assume carbon dioxide gas is generated, attaining some relatively high but unspecified partial pressure. Assume further that an E1E2 scenario (WIPP Performance Assessment Department, 1992-1993) brings Castile brine into the repository horizon, where it contacts waste and brine. Finally, assume this brine flows out of the Salado and into the Rustler. This scenario can be simulated by the following steps.

1) Calculate the mass of actinides in the control volume of interest, e.g., a panel, and the mass of organic ligands and $Ca(OH)_2(s)$ present in that volume.

2) Calculate the volume of brine contacting the waste.

3) Calculate the equilibrium aqueous and solid phase compositions based on (1) and (2) (results in one equilibrium state).

4) Calculate changes to the equilibrium state from (3) as a function of $CO_2(g)$ pressure (results in a series, call it S_i, of equilibrium states).

5) Calculate changes to the equilibrium states from (4) as a function of the volume of Castile brine added (results in a series of equilibrium states, S_j, for each of the S_i states; call them Sij).

These calculations can all be performed with the titration options included in FMT. The resulting information can be summarized in a form usable for WIPP Performance Assessment calculations.

These calculations will indicate the relative importance of the CO_2(g) fugacity (pressure) and the Castile to Salado brine volume ratio in determining dissolved actinide concentrations for a given waste type and backfill. Such an assessment for the ranges of dissolved actinide concentrations for a particular scenario can guide phased acceptance of waste types and choices for backfill materials or other engineered alternatives. The information will be passed directly to Performance Assessment in the form of look-up tables.

8.0 DATA ACQUISITION PLAN

The responsibilities for laboratory work, parameter determination, and WIPP modeling predictions for the Actinide Source Term dissolved actinide concentration submodel are divided among FSU, LBNL, LLNL, PNL, and SNL. Sandia National Laboratories is responsible for the overall design and coordination of the technical work, technical management, and model predictions for the various WIPP scenarios. These include the impact of waste form, backfill, and other potential engineering alternatives on brine compositions. Model parameters are based on data sets in the literature and laboratory measurements specifically sponsored by this program. Parameterization of the models for each oxidation state will be performed by the groups that generate the data in collaboration with SNL. The direct participation of SNL in modeling is essential to ensure consistency in data interpretation and to make sure that the information can be assimilated directly with the other portions of the dissolved actinide concentration submodel.

The laboratory work is divided among Angonne National Laboratory (ANL), FSU, LANL, LBNL, LLNL, and PNL based on the chemical properties of actinide elements and the specific capabilities of the Principal Investigators at each location. In addition, work is divided into actinide interactions with inorganic and organic ligands. Experiments measuring the solubilities of the +III and +IV actinides in inorganic salt solutions are being performed at PNL by Andrew R. Felmy, Dhanpat Rai, and coworkers under Sandia contract #AF-3339. Experiments measuring the solubilities of the +V actinides in inorganic salt solutions are being performed at LBNL by Ilham Al Mahamid and coworkers (formerly by David E. Hobart and coworkers) under Sandia contract #AH-5592. Experiments demonstrating the oxidation state analogy for Pu(V), Np(V), and Am(V) are being conducted at LANL by Wolfgang Runde in collaboration with David L. Clark (LANL) under contract #AN-1756. Experiments measuring the solubilities of the +VI actinides in inorganic salt solutions are being performed at LLNL by Cynthia E.A. Palmer and coworkers under Sandia contract #AF-3341. Complexation of An(III), An(IV), An(V), and An(VI) with organic ligands is being measured at FSU by Gregory R. Choppin and coworkers contract #AH-5590. Empirical solubility measurements for actinides in complex synthetic WIPP brines, both with and without organic ligands present, are being conducted at ANL by Donald T. Reed and coworkers under contract #AP-2267. Craig F. Novak (SNL) is the Program Manager for the above contracts and the

Principle Investigator for the Actinide Source Term dissolved actinide concentration submodel. Robert C. Moore and Chris C. Crafts are interpreting the data at SNL in conjunction with Novak.

The Management Plan for this work is Novak et al. (1994).

9.0 COMPLIANCE JUSTIFICATION

The concentrations of dissolved actinides have consistently been shown to be among the most important variables in evaluating the performance of the WIPP for compliance with the U.S. Environmental Protection Agency (EPA) regulation 40 CFR Part 191 (WIPP Performance Assessment Division, 1991; Helton et al., 1992; WIPP Performance Assessment Department, 1992-1993; Sandia WIPP Project, 1995). More recently, the concentrations of actinides in colloidal form have been shown to be among the important variables as well. Therefore, reliable information about dissolved actinide concentrations and colloidal actinide concentrations, collectively called the Actinide Source Term, are required for the WIPP Performance Assessment.

10.0 PROVISIONS FOR SIGNIFICANT EVENTS

Significant events related to preparation and conduct of the modeling plan that require documentation or action are identified in the SNL five year program plan for the WIPP.

11.0 OPERATIONAL REGULATORY REQUIREMENTS

The laboratory work conducted to support this test plan is being done at FSU, LANL, LBNL, LLNL, and PNL. The Department of Energy requires that these contractors to Sandia follow all federal, state, and local regulatory requirements for its technical and operational activities. The Environment, Safety, and Health (ES&H) departments, or their equivalent, at each of the contractor locations have developed procedures and guidelines for various activities to ensure that the contractor site is in compliance with all regulatory requirements. All laboratory work conducted at these sites will comply with these ES&H procedures and guidelines, and Sandia approved quality assurance (QA) programs. All laboratories were audited to NQA standards by qualified Sandia auditors.

12.0 QUALITY ASSURANCE REQUIREMENTS

All activities conducted by Sandia and its contractors in support of this test plan will be conducted in accordance with the Quality Assurance (QA) requirements described in the WIPP Quality Assurance Program Description (QAPD) and the supporting QA Procedures (QAPs). The fundamental elements of the QAPD and the related QAPs are addressed in this section.

12.1 Document Control

This test plan is a controlled document. Therefore, its preparation, issuance, and any major changes to it in the future will be controlled through procedures outlined in QAP 6-1 (rev. 1), Document Control Program. In addition, the guidelines provided by Arthur (1993) have also been used in preparing this test plan. **(Carol Crawford had trouble with this wording: "This is a SAND report. Thus, it will be distributed through NTIS, etc.")**

12.2 Computer Software

Any computer software used in support of the modeling efforts described in this Test Plan will be controlled through documentation requirements specified in Section 19 of the QAPD, and QAP 19-1 (rev. 2, [pending]).

12.3 Quality Assurance Requirements at Contractor Facilities

All laboratory work conducted to support the Actinide Source Term dissolved concentration submodel development effort must be performed under a Quality Assurance program reviewed and approved by Sandia and audited to evaluate the adequacy and effectiveness of the QA Program implementation. Contractors to Sandia for this program were required to prepare and implement a Quality Assurance Project Plan based on DOE Order 5700.6C with implementing criteria from ASME NQA-1 (1989) until 31 July 1995. After July 1995, the QA program basis shifted to include ASME NQA-1 (1989), ASME NQA-2a (1990) addenda (Part 2.7) to ASME NQA-2 (1989), and ASME NQA-3 (1989), excluding Section 2.1 (b).

13.0 HEALTH AND SAFETY

Chemical measurements will be performed in standard analytical chemistry laboratories capable of working with radionuclides. Good laboratory practices are required.

Work at Sandia National Laboratories is controlled by SNL ES&H practices as defined in the SNL ES&H manual. Work by contractors is controlled by company and site-specific ES&H practices.

14.0 DELIVERABLES AND SCHEDULE

Milestones and schedules for the Actinide Source Term dissolved concentration submodel are given in Appendix B. These milestones and schedules were designed to meet the requirements of the WIPP Disposal Decision Plan (Department of Energy, 1994), and were current in terms of budget as of June 1995.

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APPENDIX A: MEMORANDUM DOCUMENTING THE REASONING BEHIND SELECTING THE PITZER ACTIVITY COEFFICIENT MODEL AS THE FORMALISM FOR THE ACTINIDE SOURCE TERM DISSOLVED CONCENTRATION SUBMODEL

Novak, C.F. 1992. "Potential Models for Actindie Solubilities in WIPP Brines." memorandum to E.J. Nowak, 18 November 1992.

Note: handwritten corrections are included in the text

Sandia National Laboratories

Albuquerque, New Mexico 87185

date: 18 November 1992

to: E. Jim Nowak, 6345

Craig F Novak

from: Craig F. Novak, 6119

subject: Potential Models for Actinide Solubilities in WIPP Brines

SUMMARY AND CONCLUSIONS

Three types of models for brines were found in the literature, a Pitzer approach, an ion pairing (also called stability constant or speciation) approach, and a hybrid between these two. Pitzer type models have been applied to various brine and evaporite systems including the WIPP, and considerable efforts have already been taken for incorporating actinides into these models–efforts that need not be repeated by WIPP. Ion pairing models for brine systems have been proposed but have not actually been applied or compared with data, and a hybrid ion pairing and Pitzer model has been proposed without comparison to data. Therefore, it appears that the Pitzer model would be the best choice for use by the WIPP program.

INTRODUCTION

A model is needed for estimating the solubilities of actinides in WIPP brines. Such a model would likely have two parts: one for the nonradioactive brine constituents including Na, K, Mg, Ca, Cl, SO4, B, and CO3/HCO3, and another for the actinides of concern, including Am, Cm, U, Th, Ra, Pu, and Np. In the interest of defensibility and effective use of resources, use of an existing brine model with a good track record in the scientific community is essential. The first section of this memo reviews brine models and systems to which they have been applied. The second section reviews the ways in which actinides have been incorporated into brine models and the ways it has been suggested that actinides be incorporated into brine models.

BRINE MODELS

A literature review has revealed only two chemical models, the Pitzer approach and a Harned's Rule approach, for the major brine constituents that have been applied with some success to brine systems. The Pitzer approach is supported by many more articles and has been applied to many more systems than the Harned's Rule approach, which, though it uses different mathematics, is actually a simpler version of the Pitzer approach.

Significantly, the review turned up no applications of speciation-type models to brine systems. Also, no models using Hydration Theory (see Chapter 8, Brush; 1990), a model proposed for WIPP application, were found. One of the originators of Hydration Theory, Tom Wolery, indicated in a phone conversation that it works only up to about 2 molal, as compared with the greater than 8 molal needed for WIPP brines.

Pitzer Approach for Modeling Brines

The current best model existing for brines appears to be that based on the work of Pitzer (see for example Pitzer, 1991), which has been extensively applied to numerous systems. The data necessary to parameterize the Pitzer model are binary and ternary interactions among ions[1]. An initial parameterization was performed by Harvie and Weare (1980), who demonstrated that parameterization from binary and ternary data allowed prediction of quaternary (4-component) and quinary (5-component) systems. Harvie and Weare (1980) also compare their results with the model of Wood (1975), discussed in more detail below, and show better agreement with experimental data than Wood achieved. Some implications and predictions of the Harvie and Weare (1980) model are examined further by Harvie et al. (1982). Also, the evaporation path of sea water, both when brines can back-react with previously precipitated minerals[2], and when brines cannot back-react with previously precipitated minerals[3], are examined in general by Eugster et al. (1980), and in particular for the formation of the German Zechstein salt deposits by Harvie et al. (1980).

The model of Harvie and Weare (1980) was extended by Harvie et al. (1984) to include carbonate species. Because the authors of this paper are Harvie, Møller, and Weare, this is often referred to as the Harvie-Møller-Weare, or HMW, (parameterization of the Pitzer) model; frequently used a basis for additional parameterization. Additional binary and ternary data were used to parameterize the model, which was then used to predict solubility data in quaternary and quinary systems not considered by Harvie and Weare (1980).

In a further refinement, Felmy and Weare (1986) included boron species into the HMW formulation, and used the resulting model to help understand the formation of borate mineral deposits in Searles Lake, California.

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[1] a binary system has one cation, one anion, and water, such as Na^+-Cl^--H_2O; a ternary system has either two cations, one anion, and water, such as Na^+-K^+-Cl^--H_2O, or one cation, two anions, and water, such as Na^+-$SO_4^=$-Cl^--H_2O

[2] also called equilibrium evaporation path

[3] also called fractionation evaporation path

The HMW model has been used to postulate compositions of Permian seawater based in part on fluid inclusion data taken from the Salado Formation at the WIPP site and from the Wellington Formation in Kansas (Horita et al., 1991). Extensions of the HMW model have been applied to systems at temperatures below 25°C (Spencer et al., 1990) and to temperatures in the range from 0°C to 250°C (Greenberg and Møller, 1989). Also, the solubility of methane in brines over a wide range in temperatures and pressures have been modeled (Duan et al., 1992). Numerous other applications are documented in the literature.

Harned's Rule Approach for Modeling Brines

A different chemical model for brines was proposed by Wood (1975), using a variation of Harned's Rule for determining activity coefficients. The method was fairly successful for the Na-K-Mg-Ca-Cl-H_2O and Na-Mg-Cl-H_2O systems. However, after studying Wood (1975), it became apparent that the model is a three parameter model that is only slightly less complex than that of Pitzer, so it cannot truly be considered a competing model. The following extended quote from Wood (1975) seems germane to the topic of this memo. References to "the brine model presented here" can be considered equally applicable to the Pitzer approach; the ion pairing model is equivalent to a stability constant approach.

> The ion pairing model has proven successful in describing the thermodynamics of seawater...as well as more dilute solutions, but has not yet been applied to more concentrated solutions. It has been suggested that, owing to the markedly different nature of the chemical behavior between brines and dilute waters, different models be developed for each.... But such differences may be more apparent than real, and to treat dilute solutions with one model and brines with another would probably eventually create more problems. The progressive evaporation of natural waters leads to a saturated brine *via* continuous thermodynamic functions. Such processes would be difficult to predict if different chemical models were used for various intervals. The brine model presented here can be applied to solutions compositions ranging from infinitely dilute waters to saturated brines.

Although the article is seventeen years old, it appears that the statement "The ion pairing model...has not yet been applied to more concentrated solutions" remains true. I was unable to locate any models for brine systems that are not valid for dilute waters as well.

MODELING ACTINIDE SOLUBILITIES IN BRINES

Three types of models for actinide solubilities in groundwaters have been located in the literature: a Pitzer approach, a hybrid of the Pitzer and an ion pairing approach, and an exclusively ion pairing approach. The three methods, with brief descriptions and existing applications, are outlined below.

Pitzer Approach for Actinides in Brines

Recently, Pitzer-type models have been used to interpret solubility and other data for actinides in various brine and dilute aqueous media. Rai et al. (1990) studied the solubility of $UO_2 \bullet xH_2O(am)$ in dilute solutions. Felmy et al. (1991) examined the solubility of hydrous thorium(IV) oxide in chloride media, and Felmy and Rai (1992) analyzed thorium(IV) solubility data in the Na^+-K^+-Li^+-NH_4^+-$SO_4^=$-HSO_4^--H_2O system. Felmy et al. (1990) modeled the solubility of $AmOHCO_3(s)$ in the Na-Am-CO_3-H_2O system, and Felmy et al. (1989) investigated $Pu(OH)_3(s)$ solubility in both dilute solutions and high ionic strength chloride brines from the Permian Basis, called PBB1 and PBB3.

The work of Felmy et al. (1989) may have particular significance for the WIPP. The element concentrations of Brine A (Brush, 1990) and of PBB1 (ionic strength=5.7) and PBB3 (ionic strength=10.) from Felmy et al. (1989) are given in Table 2 and Figure 1, which show that Brine A can be considered to be intermediate in concentration between PBB1 and PBB3. In their analysis of the Permian Basis brines PBB1 and PBB3, Felmy et al. (1989) found that $Pu(OH)_3(s)$ solubility as a function of pH could be well represented by considering Pu^{+++} as the sole aqueous form of plutonium. Because Cl^- is the dominant cation, Felmy et al. (1989) considered only the interactions between Pu^{+++} and Cl^- when parameterizing the Pitzer ion interaction model. Values for these interactions were taken from data for the analogous Nd^{+++}-Cl^- system. The model works well for the PBB brines, and may be expected to work well for Pu^{+++} solubility in Brine A, assuming that $Pu(OH)_3(s)$ is the stable solid phase[4]. ~~It is interesting to note that the concentration of total inorganic carbon in PBB1 is approximately four times larger than in Brine A, yet a model that does not include specific interactions between~~ ~~Pu^{+++} and $HCO_3^-/CO_3^=$~~ ~~succeeds at describing the data. This suggests that the model may be successful for~~ ~~Pu^{+++}~~ ~~in WIPP Brine A as well.~~

All carbonate was removed from the PBB brines before the experiment. CFN 28MAR95

Hybrid Pitzer and Ion Pairing Model

A hybrid of the Pitzer and ion pairing model has been proposed by Millero (1992) for rare earth (lanthanide) elements with valence +III, which are frequently employed as model compounds for actinides in the +III oxidation state. Millero (1992) uses the Pitzer expressions for elements for which data exist, and an ion pairing model for lanthanide elements when data are not available to derive Pitzer parameters. The constants needed for

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[4] Experiments designed to measure the solubility of Pu^{+++} in Brine A are in the initial stages at Lawrence Berkeley Laboratory under a contract with Heino Nitsche.

ion pairing model were determined by reinterpreting data from the literature to be consistent with the Pitzer model. This hybrid model is applied by Millero (1992) to lanthanide speciation in seawater, and in WIPP Brine A and Brine B (Brush, 1990). However, the speciation calculations are not compared with data, so it is difficult to judge whether the model of Millero (1992) is adequate to describe such systems.

The Specific Ion Interaction (SIT) Approach

The Nuclear Energy Agency of the Organisation for Economic Cooperation and Development (OECD/NEA) advocates the specific ion interaction theory, or SIT, for solutions with actinides (Grenthe and Wanner, 1992). This is an ion pairing model with extended Debye-Hückel activity coefficients that is generally used to extrapolate stability constant data, usually in $NaClO_4$ or NaCl solutions up to about 4 molal ionic strength, to zero ionic strength. The SIT is based on the assumption that only species with opposite charges interact; neutral species do not interact with any species, and species of like charge do not interact. The theory is equivalent to a truncated version of the Pitzer model. The NEA has published data for uranium that is consistent with this model (Grenthe et al., 1992), but I have not seen any modeling efforts using these data in brines, or any comparison of these data with experimental results. Choppin and Du (SAND92-7068J, in review) measure stability constants by extraction for UO_2Cl^+, $UO_2NO_3^+$, $EuCl^{++}$, and $EuNO_3^{++}$ complexes in 3, 5, 7, and 9 molar (3.5, 6.5, 10, 14.1 molal) $NaClO_4$ solutions, and compare the data with SIT predictions. The SIT predictions are reasonable only through ~1 molal ionic strength for both europium species, through ~ 3.5 molal ionic strength for the UO_2Cl^+ species, but through ~10 molal ionic strength or greater for the $UO_2NO_3^+$ complexes. Because SIT apparently has not been applied to brine systems, and because conceptual difficulties exist for extrapolating data from a simple electrolyte solution to multicomponent brine solutions, the potential for success of SIT at estimating actinide solubilities in WIPP brines may be low.

Existing Computer Codes

Numerous computer codes for performing chemical equilibrium calculations are available. Perhaps the best known are EQ3/6 (Wolery, 1979), developed through Lawrence Livermore National Laboratories, and PHRQPITZ (Plummer et al., 1988), a version of PHREEQE (Parkhurst et al., 1980), developed through the U.S. Geological Survey. These codes are formally documented and have been used worldwide, thus either of these would probably meet quality assurance requirements relatively easily. Both EQ3/6 and PHREEQE were judged to be suitable for performance assessment of nuclear waste storage by INTERA (1983) before the Pitzer activity model was incorporated into either code. However, PHRQPITZ is easier to use because it is more straightforward, and also seems to require significantly less computer time to run. The actual computer code to be used for calculations is not of particular importance at this time because correctly parametrized thermodynamic data could be used with nearly any existing chemical equilibrium code.

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Table 1. Calculated Ionic Strengths* of Salado, Castile, and Culebra Brines.

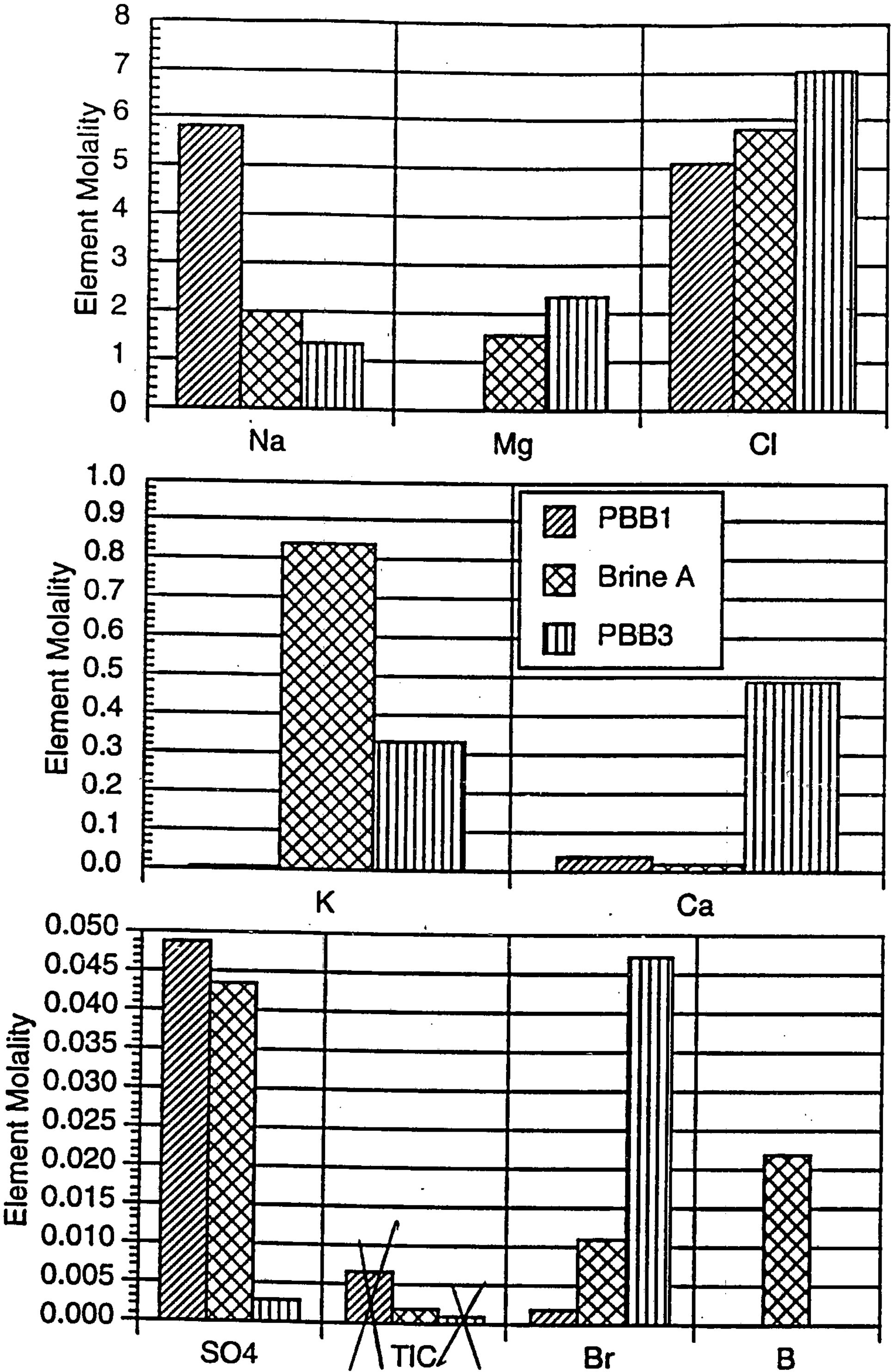
Formation	Brine Name	Ionic Strength	Data Source
Salado	SB1	8.8	Brush (1990)
"Standard" Salado Brine	Brine A	7.8	Brush (1990)
Castile	ERDA6	5.8	Brush (1990)
Culebra	H-17	3.0	Novak (1992)
Culebra	DOE-1	2.5	Novak (1992)
Culebra	H-11	2.2	Novak (1992)
Culebra	H-3	1.1	Novak (1992)
Culebra	AIS	0.85	Novak (1992)

* Ionic strengths were calculated using the Pitzer activity approach with the PHRQPITZ code (Plummer et al., 1988).

Table 2. Element concentrations for brines PBB1 and PBB3, for which solubility data for $Pu(OH)_3(s)$ have been interpreted with the Pitzer model, and Brine A.

Element	PBB1 Molality	Brine A Molality	PBB3 Molality
Na	5.83	2.00	1.34
K	0.00247	0.840	0.330
Mg	0.00555	1.56	2.36
Ca	0.0374	0.0164	0.487
Cl	5.13	5.83	7.03
SO4	0.0490	0.0436	0.00264
TIC ϕ	~~0.00658~~	0.00176	~~0.000837~~
Br	0.00195	0.0109	0.0474
B	-	0.0218	-

ϕ CFN 7SW95



CFN 7JUN95

Figure 1. Element molal concentration for Permain Basis Brines PBB1 and PBB3 (Felmy et al., 1989), and the WIPP Salado Brine Surrogate Brine A (Brush, 1990).

APPENDIX B: DESCRIPTIONS OF THE EXPERIMENTAL PROGRAM FOR THE ACTINIDE SOURCE TERM DISSOLVED CONCENTRATION MODEL

Novak, C.F. 1995. “Experimental and Modeling Program for Determining Mobile Dissolved Actinide Concentrations for Scenarios Relevant to the WIPP, Revision 1.” memorandum to E.J. Nowak. 7 June 1995.

Novak, C.F. 1995. “Experimental and Modeling Program for Determining Mobile Dissolved Actinide Concentrations for Scenarios Relevant to the WIPP: Summary of Program Accomplishments from Inception through 31 January 1995.” memorandum to Butch Stroud and Dick Lark. 6 March 1995.

Sandia National Laboratories

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date: 7 June 1995

to: E. James Nowak, Dept. 6719, MS 1320

Craig F Novak

from: Craig F. Novak, Dept. 6748, MS 1320, 848-0619

subject: Experimental and Modeling Program for Determining Mobile Dissolved Actinide Concentrations for Scenarios Relevant to the WIPP, Revision 1.

WBS 1.1.1.1.4, 1.1.5.1.2, 1.1.5.2.2, 1.1.1.2.6

This memorandum documents the experimental programs involved in developing the model for mobile dissolved actinide concentrations in WIPP brines. This document describes the technical and scheduling aspects of the program, contingent on the Sandia funding request for the Actinide Source Term Dissolved Concentration Model Development effort of $3.6M for FY96.

All the contractors involved in development of the dissolved concentration model contributed to the design of the experimental program. All contractors had the opportunity to review Revision 0 this document to create Revision 1. However, this document had not been through the review process at Sandia.

This document contains three major sections. The first section provides an overview of the Actinide Source Term, what it is, and some general technical details. The second section is a list of the major experimental activities required for developing the actinide dissolved concentration models. The third section contains Gantt charts showing the expected schedule for the experimental activities.

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OVERVIEW: THE ACTINIDE DISSOLVED CONCENTRATION SUBPROGRAM OF THE ACTINIDE SOURCE TERM (AST)

Definition: The Dissolved Concentration Model

The dissolved concentration model is a set of mathematical equations and constants that will be used to predict dissolved actinide concentrations as a function of brine composition and hydrogen ion concentration. The mathematical equations are based on aqueous thermodynamics (see for example, Stumm and Morgan, 1981; Garrels and Christ, 1990). Many of the parameters needed to apply these equations to WIPP brines, at least WIPP brines in the absence of actinides and organic species, are given in the literature (Harvie et al., 1984; Felmy and Weare, 1986, Pitzer, 1991). The development of a dissolved concentration model for actinides involves determining the additional parameters required to apply the existing thermodynamic models to actinide elements of interest to the WIPP Project. These parameters will be the "standard chemical potentials" of actinide-containing species, either aqueous or solid, and the "ion interaction parameters," or "Pitzer parameters," for actinide-bearing solution species. The set of mathematical equations using Pitzer parameters to describe activity coefficients in aqueous solution is commonly called the Pitzer model or the Pitzer activity coefficient formalism.

Only some of the necessary standard chemical potentials or ion interaction parameters for actinide species are available in the literature. Some of the missing but needed parameters can be reliably estimated using the oxidation state analogy for actinides. Because of programmatic constraints, it is not possible to determine all ion interaction parameters and all standard chemical potentials for every possible species. The methodology presented here was developed to provide the maximum amount of information with given resources.

Thermodynamic parameters that cannot be reliably estimated from existing information must be determined from laboratory measurements of chemical properties. The kinds of experiments necessary to determine the parameters are generally well developed, however unanticipated experimental behavior may be encountered for the less well explored actinide systems. The application of existing experimental methods to actinides will provide the parameters needed for the WIPP. The experiments planned through March 1996 in this effort are detailed in this memorandum.

The Actinides of Interest

The WIPP dissolved concentration model program is focusing on the actinide elements thorium (Th), uranium (U), neptunium (Np), plutonium (Pu), and americium (Am), based on sensitivity analyses performed in conjunction with the 1992 WIPP Performance Assessment (WIPP Performance Assessment Department, 1992). Because it is the outer electron configuration that determines chemical properties for the most part, the chemistry of all isotopes of the same element is the same. For example, ^{239}Pu and ^{244}Pu have the same chemical properties, although their nuclear decay properties may be very different.

The most important consideration for the chemistry of the actinide elements is the oxidation state. The oxidation states that the above elements may assume under WIPP conditions are: Pu(III) and Am(III); Th(IV), U(IV), and Np(IV), Pu(IV); Np(V), Pu(V), and Am(V); and U(VI), Np(VI), and Pu(VI).

The Oxidation State Analogy

A chemical principle that can be applied to f-elements like actinides and lanthanides is the oxidation state analogy: f-elements in the same oxidation state exhibit similar chemical behavior. Thus, Pu(III) and Am(III) have quantitatively similar behavior, and this quantitative similarity extends neodymium(III), Nd(III), a lanthanide element. Based on all available evidence, Np(V), Pu(V), and Am(V) have quantitatively similar chemical behavior, and U(VI), Np(VI) and Pu(VI) also have quantitatively similar behavior. However, the oxidation state analogy is not perfect, and for the +IV elements Th(IV), U(IV), Np(IV), and Pu(IV) the analogy is only qualitative. The +IV actinides do show regular trends in behavior, including a systematic decrease in solubility in the series Th(IV), U(IV), Np(IV), Pu(IV) under the same chemical conditions.

The oxidation state analogy reduces the amount of experimental and modeling work required to develop the dissolved concentration model for actinides. That is, a model developed for Nd(III) can be applied quantitatively to Pu(III) and Am(III); a model developed for Np(V) can be applied quantitatively to Pu(V) and Am(V); and a model developed for U(VI) can be applied quantitatively to Np(VI) and Pu(VI). Although the oxidation state analogy must be confirmed experimentally for each system, the process of verifying an existing model is much simpler than developing a new one. Thus, the oxidation state analogy reduces the total effort to obtain dissolved concentration models. Because the oxidation state analogy is only qualitative for the +IV actinides, more work is required to develop models for these elements. However, the qualitative analogy does focus the experimental work required, and reduces total effort, though not as significantly for the +IV actinides as for the other oxidation states.

The current program is: develop a model for Nd(III) behavior and show that it applies to Am(III) and Pu(III); develop a model for Np(V) behavior and show that it applies to Pu(V) and Am(V); develop a model for U(VI) behavior and show that it applies to Pu(VI) and Np(VI); and develop models for U(IV) and Th(IV) behavior, and show that these models overpredict the solubility behavior of Np(IV) and Pu(IV). The following discussion uses Am(III), Th(IV), U(IV), Np(V), and U(VI) as prototypical for each oxidation state. The reader should understand that, through the oxidation state analogy and subject to its limitations, the other actinide elements are included as well.

Aqueous Interactions between Actinides and Brine Constituents

Dissolved actinides in acidic aqueous media generally form the aquo ions: Am^{3+}, Th^{4+}, U^{4+}, NpO_2^{+}, and UO_2^{2+}, called americium(III) ion, thorium(IV) ion, uranium(IV) ion, neptunyl(V) ion, and uranyl(VI) ion, respectively. The total dissolved concentrations of each actinide is the sum of all cationic and anionic complexes and the aquo ion. Anionic brine species are particularly important for complexing the cationic aquo ions. The chemical components of the brine can be divided into two major categories: inorganic ligands such as hydroxide, chloride, sulfate, and carbonate, and organic ligands such as acetate, citrate, and EDTA. In general, the hydrogen ion concentration (generally referred to as pH, although this is only meaningful for low ionic strength systems) is a major influence on dissolved actinide concentrations because the hydrogen ion or the hydroxyl ion reacts chemically with nearly every species in aqueous solution.

Simply put, the dissolved concentration model seeks to describe qualitatively the influence of brine composition on the dissolved actinide concentrations. Not all of the interactions between actinides and aqueous species have the same importance. For example, interactions between cationic actinides and brine cations like sodium ion, Na^+, and potassium ion, K^+, are generally weak because of repulsive forces. However, cationic brine constituents like Na+ or K+ can have strong interactions with anionic actinides complexes like $Am(CO_3)_3^{3-}$. On the other hand, ion interactions between cationic actinides and brine anions such as hydroxide, OH^-, and chloride, Cl^-, are generally stronger because of attractive forces. These principles have been used to prioritize interactions into a list of those likely to be the most important. While it would not be possible to definitely state "this interaction is important and that interaction is unimportant" without some experimental evidence, chemical principles provide the best guidance for this prioritization.

Thermodynamic formalism for aqueous species accounts for interactions between actinides and brine constituents in two categories: specific ion interactions and complex formation. Specific ion interactions, as described by specific ion interaction parameters or "Pitzer parameters," are generally used for weaker cation-anion type interactions between actinides and inorganic ligands. Complex formation, as described by standard chemical potentials (which are related to stability or formation constants) are generally used to describe strong interactions; hydrolysis (reactions with water or hydroxide), carbonate complexation, and complexation with organic ligands fall into this category. There is no sharp demarcation between the two methods; sometimes either description will be adequate. Also, there are systems that require ion interaction parameters with actinide complexes.

Complex formation follows the general chemical reaction

$$An^{n+} + mL^{p-} \leftrightarrow AnL_m^{(n-mp)} \qquad (1)$$

where An^{n+} represents any of the actinides, L^{p-} is an anionic ligand (either organic or inorganic), and $AnL_m^{(n-mp)}$ is an actinide complex resulting from the complexation of An by m ligands. Depending on the charges of the individual reactants, the product species can be cationic, anionic, or neutral. A constant called the "standard chemical potential" of the product species must be determined for each such chemical reaction (by definition the standard chemical potentials of the reactant species are known).

By contrast, specific ion interactions are not associated with chemical reactions, but are related to the concentrations of individual ions and how the concentration of one ion affects concentrations of other ions. The most common types of ion interactions are binary ion interactions, such as cation-anion interactions, neutral-cation or neutral-anion interactions, and cation-cation or anion-anion interactions. Cation-anion interactions tend to be the strongest, although cation-cation or anion-anion interactions can be very strong for highly charged species. The Pitzer activity coefficient formalism also includes ternary interactions such as cation-cation-anion, cation-anion-anion, and cation-anion-neutral interactions, which tend to be important only when some of the species involved are present in high concentration. The symbols $\beta_{mx}^{(0)}$, $\beta_{mx}^{(1)}$, $\beta_{mx}^{(2)}$, and C_{mx}^{ϕ} represent specific ion interaction parameters between cation m and anion x; the β parameters are binary interaction parameters and the C^{ϕ} parameters are technically ternary (three body) interaction parameters but are determined from binary data. The symbols λ_{nm} and λ_{nx} are used for neutral-cation and neutral-anion interaction parameters, respectively. While these symbols are included in the formalism, often it is not required to have values for each parameter, e.g., frequently only the $\beta_{mx}^{(0)}$ parameter is required to describe the data for a particular system.

It is simpler to describe aqueous chemical behavior using only specific ion interaction parameters and no aqueous complexes. However, very strong complexes cannot be described adequately well in this manner, and thus some complexation generally must be included. It should be noted that whenever a complex is added, another species is created that can have specific ion interactions with cationic, anionic, and neutral species, adding to the overall complexity of the chemical description.

Actinide Solid Phases

The aqueous chemical behavior of actinides is part of the problem for determining dissolved actinide concentrations, the other part is identification and characterization of actinide-bearing solid phases. The aqueous chemistry dictates the capacitance of the aqueous phase to hold actinides, while the solid phase actually controls the concentrations of actinides. Solubility studies are designed to provide information both about the aqueous phase and about the solid phase. The identity of the solid phase, including the stoichiometry, is essential for thermodynamic analysis.

THE EXPERIMENTAL PROGRAM

The experimental program for determining dissolved actinide concentrations is divided into five portions, primarily dictated by chemical considerations. One major division is between the effects of inorganic and organic ligands; the other major division is by oxidation state. The +III, +IV, +V, and +VI inorganic models are being developed separately, and each serves as the basis model for that oxidation state. The organic models are being developed independently relying on information from the inorganic models. The experimental work is divided as follows: +III and +IV at PNL (Felmy, Rai), +V at LBL (Al Mahamid) and at LANL (Runde), +VI at LLNL (Palmer), and the organic work at FSU (Choppin). The status of each of these portions, including information available and ongoing and planned experimental work through 31 March 1996, is detailed below.

The +III Actinide Model; Inorganic Interactions (Contract AF-3339; PNL)

Extensive research on the chemistry of +III f-elements in concentrated electrolytes has been reported in the literature independently of the WIPP Project, including the papers Felmy et al. (1989, 1990), and Rai et al. (1983, 1992a, 1992b, 1994). Additional WIPP-supported research is given in Rao et al. (1994). WIPP research for modeling the dissolved concentrations of +III actinides includes integrating existing information into a single data base, demonstrating that the model works in the ternary electrolyte systems Na^+-Cl^--CO_3^{2-} and Na^+-Cl^--HCO_3^-, demonstrating the predictive ability of the model in complex synthetic WIPP brines, and confirming the oxidation state analogy for Nd(III), Am(III), and Pu(III).

+III Experiment: Solubility of Nd(III)$OHCO_3$(c) and NaNd(III)$(CO_3)_2$(c) in Mixed NaCl/Na_2CO_3 and NaCl/$NaHCO_3$ Media

Experiments measuring the solubility of Nd(III)$OHCO_3$(c) and NaNd(III)$(CO_3)_2$(c) in mixed NaCl/Na_2CO_3 and NaCl/$NaHCO_3$ media were initialized in November 1994. These experiments consist of three sets of five samples each: one set with 2m NaCl and Na_2CO_3 concentrations ranging from 0.1 to 2.0m; one set with 4m NaCl and Na_2CO_3 concentrations ranging from 0.1 to 2.0m; and one set with 2m NaCl and $NaHCO_3$ concentrations ranging from 0.1 to 0.5m. These experiments are designed to demonstrate whether anion-anion ion interaction parameters are necessary to describe $Nd(CO_3)_2^-$-Cl^- or $Nd(CO_3)_3^{3-}$-Cl^- interactions, which may be important because of the potentially large concentrations of these anions. The experiments are designed to support the existing model parameters, but could also be used with existing data to determine better parameter values.

+III Experiment: Solubility of Nd(III)$OHCO_3$(c) and NaNd(III)$(CO_3)_2$(c) in Complex Synthetic WIPP Brines

Experiments measuring the solubility of Nd(III)$OHCO_3$(c) and NaNd(III)$(CO_3)_2$(c) in complex synthetic WIPP brines were initiated in January 1995. These experiments are being performed in a truncated Castile brine based on the composition of brine from well ERDA6. These brines are truncated in that the small amounts of calcium and magnesium are not included. The second brine is based on G-Seep from the Salado Formation. The solubility of Nd(III) will be measured as a function of hydrogen ion concentration and CO_2(g) pressure. Brine compositions were developed from those reported by Brush (1990). This will allow all features of the thermodynamic dissolved concentration model for Nd(III) to be tested, and will complete the model development portion for +III actinides

+III Experiment: Confirm the Oxidation State Analogy for Nd(III)-Am(III)-Pu(III)

The oxidation state analogy for similarity of the +III f-elements is firmly established, but no single report collects and documents this work. This activity compiles this information, and will repeat a selection of the experiments with Nd(III) in WIPP brines using Am(III) and Pu(III) instead of Nd(III). These experiments will confirm the oxidation state analogy for Nd(III), Am(III), and Pu(III) for WIPP specific brines.

The +IV Actinide Model; Inorganic Interactions (Contract AF-3339; PNL)

Extensive research on the chemistry of +IV actinides in concentrated electrolytes has been reported in the literature independently of the WIPP Project, although not as much information is available as for the +III f-elements. Articles on the chemical behavior of Th(IV) include Felmy and Rai (1992), Felmy et al. (1991, 1993), and Roy et al. (1992). WIPP research for developing the +IV dissolved concentration model focuses primarily on measuring additional experimental data to get needed ion interaction parameters for the major brine constituents, including chloride, sulfate, and magnesium. Specific ion interactions between the +IV actinides and Mg^{2+} are necessary because of the very high charge on the +IV actinides. The high charge makes cation-cation interactions important enough that they must be included in the dissolved concentration models. An additional complication for the +IV actinides is the oxidation state analogy is not quantitative for Th(IV), U(IV), Pu(IV), and Np(IV), as it is for the other oxidation states. Thus, experimental work for each of the actinide elements is necessary.

Programmatic constraints do not allow the complete development of individual Th(IV), U(IV), Np(IV), and Pu(IV) models. Development of the Th(IV) and U(IV) models is continuing. The dissolved concentrations of Np(IV) and Pu(IV) will be calculated with the U(IV) model, which will predict higher dissolved concentrations than would actually occur, and would thus be conservative. Empirical solubility experiments measuring Np(IV) and Pu(IV))

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solubilities in complex synthetic WIPP brines will show that this is indeed a conservative approach.

+IV Experiment: Solubility of $Th(IV)(OH)_4(s)$ in NaCl and $MgCl_2$ Media

The solubility of hydrous thorium oxide, $Th(IV)(OH)_4(s)$ and associated ion interaction parameters are reported in the literature for up to 3m NaCl (Felmy et al., 1991; Roy et al. 1992). Because WIPP brines contain up to 6m NaCl and up to 8m NaCl/$MgCl_2$, the solubility of $Th(IV)(OH)_4(s)$ in 4 and 6m NaCl, and in 1, 1.8, and 3m $MgCl_2$ were studied as a function of hydrogen ion concentration. The measured aqueous concentrations of Th(IV) in these systems were reported in the December 1994 Monthly Report from PNL; analyses of the solid phases are in progress. Preliminary analysis of the data indicates that specific ion interactions between Th^{4+} and Cl^-, Na^+, and Mg^{2+} are required to interpret the data. Interpretation of these data with the appropriate ion interaction parameters will provide a thermodynamic model capable of predicting the solubility of hydrous thorium oxide in the environmentally important hydrogen ion concentration range in concentrated Na-Mg-Cl brines from the WIPP.

Complementary research on carbonate complexation with Th(IV) is in progress at PNL. This work is funded by the Power Reactor and Nuclear Fuel Development Corporation of Japan (PNC), and thus is neither under the control of Sandia nor DOE. However, SNL is negotiating with PNL to provide funding for interpretation of these data, and thus they may come under the WIPP schedule. SNL intends to rely on this information as part of the model for Th(IV) solubility in WIPP brines, and requires this information for developing Th(IV) solubility functionality tables for WIPP Performance Assessment.

+IV Experiment: Solubility of $UO_2 \cdot xH_2O(am)$ in NaCl and $MgCl_2$ Media

High chloride concentrations can significantly influence the solubility of hydrous uranium(IV) oxides. Most of the available data for complexation of U(IV) with chloride were taken in HCl and $HClO_4$ (Grenthe et al., 1992) at chloride concentrations substantially lower than those observed in WIPP brines. Limited data reporting the solubility of amorphous uranium dioxide are available (Aguilar et al., 1991), but these data are suspected to reflect U(VI) properties, not U(IV) properties, because of inadequate control of the uranium oxidation state. Thus, additional experimental work was performed to measure chloride interactions with U(IV).

U(IV) is readily oxidized to U(VI) by even small amounts of oxygen, so extreme precautions are required to prevent this oxidation. These precautions include: preparing the U(IV) stock solution in 0.5M HCl, where U(IV) is relatively stable; storing the solution under an argon atmosphere and contacting the solution with uranium metal; conducting experiments in low redox potential solutions with a calculated $O_2(g)$ content of less than 10^{-75} atm; and conducting the experiments in the presence of Eu^{2+} (europium) to maintain very low $O_2(g)$ fugacities. The difficulty of maintaining uranium in solution as U(IV) has implications for the uranium oxidation state distribution in WIPP Performance Assessment calculations. Because U(IV) oxidizes readily, it is not possible to assert that uranium will occur in WIPP disposal rooms as U(IV), regardless of overall anoxic conditions in the disposal rooms and regardless of the presence of metallic iron. The oxidation-reduction behavior of uranium and the other redox-sensitive actinides will be quantified under a separate contract with LANL and PNL, as discussed in the last section of this memorandum.

As part of this study, six sets of electrolyte solutions were prepared: 0.2, 1.0, and 6.0m NaCl, and 1.0, 2.0, and 3.0m $MgCl_2$. The solubility of $UO_2 \bullet xH_2O(am)$ was measured as a function of between 5 and 8 hydrogen ion concentrations. The analyses of aqueous phase concentrations have been completed; solid phase characterization is underway. Interpretation of these data with the appropriate ion interaction parameters will provide a thermodynamic model capable of predicting the solubility of uranium hydrous oxide in the environmentally important hydrogen ion concentration range in concentrated Na-Mg-Cl brines from the WIPP.

Complementary research on carbonate complexation with U(IV) is in progress at PNL. This work is funded by the Power Reactor and Nuclear Fuel Development Corporation of Japan (PNC), and thus is neither under the control of Sandia nor DOE. However, SNL is negotiating with PNL to provide funding for interpretation of these data, and thus they may come under the WIPP schedule. SNL intends to rely on this information as part of the model for U(IV) solubility in WIPP brines, and requires this information for developing U(IV) solubility functionality tables for WIPP Performance Assessment.

+IV Experiment: Solubility of $U(IV)(OH)_4(s)$ in Na_2SO_4 Media

The sulfate anion may be an important inorganic ligand influencing the dissolved concentration of U(IV) in WIPP brines. Experiments measuring this effect will be conducted as a function of Na_2SO_4 concentration to determine the magnitude of this effect. The experiments will be designed such that specific ion interaction parameters and the standard chemical potentials of important U(IV)-SO_4^{2-} species can be determined. The equivalent chemical model for Th(IV) behavior in Na_2SO_4 required the aqueous species $Th(SO_4)_2(aq)$ and $Th(SO_4)_3^{2-}$. This model also required the $\beta^{(0)}$ interaction parameter for Na^+-$Th(SO_4)_3^{2-}$ and H^+-$Th(SO_4)_3^{2-}$ (Felmy and Rai, 1992). Therefore, by analogy, these could be important effects for U(IV) in sodium sulfate media.

+IV Experiment: Solubility of Th(IV), U(IV), Np(IV), and Pu(IV) in Complex Synthetic Brines

The goal of the actinide dissolved concentration model development is reliable prediction of the concentrations of the various oxidation states of the actinides in WIPP brines. This reliability must be demonstrated by comparison with data that was not used to develop the model. The most relevant and interpretable of such data would be in WIPP specific brines under different breach and backfill scenarios. Thus, the solubility of the +IV actinides in synthetic WIPP brines must be measured. These experiments will be conducted in several relevant WIPP brines as a function of hydrogen ion concentration and $CO_2(g)$ pressure, to mimic the chemical conditions expected in the WIPP disposal rooms.

The experiments with Th(IV) and U(IV) in complex synthetic WIPP brines will challenge the dissolved concentration models for these elements. The experiments with Np(IV) and Pu(IV) will demonstrate that the solubility of these +IV actinides are lower than that of U(IV), and thus that Np(IV) and Pu(IV) concentrations will be conservatively predicted by the U(IV) model.

The +V Actinide Model; Inorganic Interactions Part 1 (Contract AH-5592; LBL)

Neptunium(V) is the prototype actinide for development of the +V model, which will be applied to Pu(V) and Am(V). A preliminary model for Np(V) solubility as a function of Na^+, Cl^-, OH^-, and CO_3^{2-} concentrations is developed and documented in Novak and Roberts (1994). This serves as the basis for all +V actinide solubility modeling, and will be augmented as new data is available.

Additional experimental work is needed to confirm and refine parameters for neptunyl interactions with chloride, carbonate, and potassium, and to test these parameters in mixed $NaCl/NaHCO_3/Na_2CO_3$ media and complex synthetic brine simulants for the WIPP. The experimental work concludes with a demonstration of the validity of the Np(V)-Pu(V) oxidation state analogy in several WIPP specific brines under expected repository conditions.

+V Experiment: Solubility of Np(V) in K_2CO_3 Media

The solubility of Np(V) is being studied in potassium carbonate media, in experiments initiated in January 1995. These experiments are designed to: determine the standard chemical potential for $KNpO_2CO_3 \bullet xH_2O(s)$; confirm and refine the existing values for the standard chemical potentials of $NpO_2(CO_3)_2^{3-}$ and $NpO_2(CO_3)_3^{5-}$, and establish values for ion interaction parameters between K^+ and the neptunyl carbonato complexes. Although the solutions will be very basic, carbonato complexation will dominate over hydrolysis (reactions with hydroxide). Neptunyl hydroxide solid was added to reaction vessels containing K_2CO_3 at concentrations such as 0.01, 0.03, 0.05, 0.07, 0.1, 0.25, 0.5, 0.75, 1.0, 1.5, 2.0, and 5.0m. The solid will convert to potassium neptunyl carbonate and reach steady-state conditions, yielding a value for the standard chemical potential of this solid. The variation of total Np(V) aqueous concentration provides data for confirming the standard chemical potentials of the aqueous species, and determining ion interaction parameters between these species and potassium ion.

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+V Experiment: Solubility of Np(V) in $MgCl_2$ Media

Chloride is one of the most important inorganic anions in WIPP brines. However, the data upon which NpO_2^+-Cl^- ion interaction parameter values are based are limited. Thus, the values for these parameters, which are very important for WIPP brines, are not well established. Experiments measuring the solubility of Np(V) in magnesium chloride media will firmly establish the NpO_2^+-Cl^- ion interaction parameters, eliminate questions regarding the sodium content of the solid phase, and provide evidence for interactions, or lack of interaction, between NpO_2^+ and Mg^{2+}. The experiments will be conducted under conditions such as 0.5, 1.0, and 2.0m $MgCl_2$ for a range of acidic hydrogen ion concentrations. The conditions will be developed through exercising the existing Np(V) model, recognizing that it does not contain magnesium effects. An acidic range will be used to prevent formation of hydrolysis species, and insure the dominant aqueous form of Np(V) is NpO_2^+.

+V Experiment: Solubility of Np(V) in Mixed Na-K-Mg-Cl Electrolytes

The dominant ion interactions for the Np(V) system are expected to be between uncomplexed neptunyl and chloride, and between neptunyl hydrolysis species and sodium, potassium, and magnesium. Complex formation with carbonate is expected to be important as well. Measurements of the solubility of Np(V) in mixed Na-K-Mg-Cl electrolytes will be performed to meet two objectives, depending on the outcomes of the experiments. The conditions for these solubility experiments will be designed using the best existing Np(V) model. If the measured solubilities agree well with the model, the model is demonstrably complete with respect to the major electrolytes in WIPP brines. If the measured solubilities do not agree well, the data from these same experiments will be used to develop the additional ion interaction parameters necessary to describe the data.

+V Experiment: Confirmatory Studies for Np(V) Solubility in Complex Synthetic WIPP Brines

The goal of the Dissolved Concentration subprogram for the WIPP Actinide Source Term is a numerical model predicting dissolved actinide concentrations as a function of the dominant variables in WIPP brines. The ability of the model to predict actinide solubilities in WIPP brines must be demonstrated through comparison with measured values. This activity will measure Np(V) solubility in several synthetic brines pertinent to WIPP Performance Assessment, as a function of hydrogen ion concentration and $CO_2(g)$ pressure. The results of these experiments will be compared directly with model calculations, and will confirm the ability of the model to predict dissolved Np(V) concentrations in the WIPP system.

The +V Actinide Model; Inorganic Interactions Part 2 (Contract AN-1756; Glenn T. Seaborg Institute for Transactinium Science (GTS/ITS))

Modeling the Np(V)-Na-Cl-CO_3-HCO_3-CO_2-H-OH-H_2O System

The WIPP project has obtained data on the solubility of Np(V) in aqueous media as a function of sodium chloride concentration, hydrogen ion concentration, and carbon dioxide gas pressure. These data, from the German nuclear waste research program, are reliable, and thus do not need to be remeasured for the Actinide Source Term. These data are being reinterpreted to be consistent with the Pitzer activity coefficient formalism and the Harvie-Møller-Weare/Felmy-Weare nonactinide data base (Harvie et al., 1984; Felmy and Weare, 1986). Development of this model using this newly available data will improve the numerical representation of the solubility of Np(V) in WIPP brines relative to the preliminary model presented in Novak and Roberts (1994).

Isolating Pu(V) for Oxidation State Analogy Demonstration

Preliminary research of the dissolved concentrations of Pu in several synthetic WIPP brines has demonstrated that the solubility-controlling solid phase contains Pu(V) (Nitsche et al., 1992; 1994). In order to obtain reliable data against which to compare and confirm the Np(V)-Pu(V) oxidation state analogy, the optimal conditions under which to measure Pu(V) chemical behavior must be determined. This activity represents developmental experimental work focused on isolating Pu(V), or minimizing the amounts of other Pu oxidation states, so the oxidation state analogy can be confirmed.

+V Experiment: Confirmatory Studies for the Np(V)-Pu(V) Oxidation State Analogy

The oxidation state analogy for Np(V) and Pu(V) is established in all systems for which there is data, but the number of these systems are limited. These experiments are intended to demonstrate quantitatively similar aqueous and solid chemical behavior for Np(V) and Pu(V) in WIPP-specific brine compositions under a range of hydrogen ion concentrations and $CO_2(g)$ pressures. The literature indicates that +V is a prevalent oxidation state for aqueous plutonium in the environment. However, because the potential differences separating plutonium oxidation states are small, it is difficult to isolate Pu(V) in laboratory experiments. These confirmatory studies will rely on the developmental work of the immediately preceding activity, and will implement these studies to confirm the Pu(V)-Np(V) oxidation state analogy and thus the Pu(V) dissolved concentration model.

Data to confirm the Am(V)-Np(V) oxidation state analogy in NaCl media with CO2(g) as a function of hydrogen ion concentration is available in the German literature. This work is being translated into English for use in WIPP Actinide Source Term Model Development.

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The +VI Actinide Model; Inorganic Interactions (Contract AF-3341; LLNL)

The prototype actinide for the +VI model is U(VI). The other +VI elements, Pu(VI) and Np(VI), have similar chemical behavior, and all available information indicates that a U(VI) dissolved concentration model can be applied quantitatively to these elements. As scoping experiments have demonstrated, the chemical behavior of U(VI) is more complicated than that for the +III, +IV, and +V oxidation states. In particular, the scoping experiments demonstrated that the solubility-controlling solids are both a function of the ionic strength and the hydrogen ion concentration. Therefore, the +VI effort was replanned and reprioritized. This section presents the results of this redesign effort.

The literature contains numerous papers about the thermodynamics of aqueous uranyl, UO_2^{2+}, in electrolyte solutions. Historically, these aqueous solutions have predominantly been dilute solutions of single electrolytes such as $NaNO_3$. Dilute solutions were chosen to evaluate standard-state formation constants for a multitude of proposed uranyl solution species using the Debye-Hückel electrolyte theory. Only within the last 20-30 years, have researchers begun to evaluate for standard-state thermodynamic formation constants in solutions of considerably higher ionic strength, a situation relevant to the WIPP Site. Thermodynamics in high ionic strength solutions are not adequately described by the Debye-Hückel theory, so more extensive electrolyte theories like the Pitzer formalism are needed to interpret these data.

Thermodynamic data for uranyl ion in high ionic strength solutions are somewhat scarce. This inadequate amount of data has led to considerable debate as to which uranyl species dominate under various solution conditions. Most research groups conclude their investigations by proposing uranyl solution species that best fit their data, using their chosen form of an electrolyte theory, and then publish evaluated values of the standard-state formation constants for those species. Standard-state formation constants for any species resulting from different studies should be the same independent of the electrolyte theory used in the evaluation of the data.

The difficulty in using the conclusions from studies in single electrolytes with a simplified electrolyte theory is that resulting solution dependent formation constants do not include effects that result in concentrated, multi-component electrolytic solutions, like the brines associated with the WIPP Site. The Pitzer electrolyte theory (Pitzer, 1991) has been shown to describe the aqueous thermodynamics of multi-component systems in solutions that range from very dilute to rather concentrated (Harvie et al., 1984; Felmy and Weare, 1986). This theory requires the evaluation of ion interaction parameters for chemically viable species in single electrolyte systems like solutions of NaCl or K_2SO_4. Parameters obtained from experiments in simple systems can be used to model complicated systems that contain many components.

This Pitzer modeling approach has not yet been applied to the thermodynamics of the uranyl ion in solutions that contain the predominant species found in WIPP brines for two reasons. First, the existing data has never

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been interpreted together in a consistent way using the Pitzer formalism. Second, measurements of uranyl ion chemical interactions with some of the important components in WIPP brines have not been measured. The U(VI) model development work is evaluating uranyl ion interaction parameters by reinterpreting literature data with the Pitzer formalism, and measuring the missing chemical behavior.

Scoping studies conducted at LLNL showed that hydrolysis and carbonate complexation are important components to the understanding of uranyl solubility, especially at pmH near and above neutral. The scoping experiments also demonstrated that complete characterization of the chloride and sulfate systems would be a considerable task. These experiments showed that the solubility controlling solid phase is a function of both pmH and ionic strength, and the solid phases did not match any known, characterized uranium-bearing solid phases.

Develop Matrix of All Possible Interactions, and Prioritize Interactions

Based on these results and the WIPP Disposal Decision Plan, a detailed investigation of uranyl thermodynamics for each WIPP brine component cannot be performed. Therefore, the work has been prioritized to include only those effects that appear to dominate the solution chemistry of U(VI) based on conclusions found in the literature, as summarized in Grenthe et al. (1992).

Important potential interaction parameters were identified started with the possible uranyl complexes species that could form in WIPP brines and that were found to be viable by Grenthe et al. (1992). This review indicated that hydrolysis and carbonate complexation will dominate uranyl ion chemistry in WIPP brines. Carbonate complexation dominates uranyl solution chemistry at near neutral pmH even at atmospheric $CO_2(g)$ partial pressures (Grenthe et al., 1984; Sergeyeva et al., 1972). Below atmospheric $CO_2(g)$ pressures, uranyl hydrolysis dominates. Chloride and sulfate complexes will be present in significantly smaller concentrations. Thus, the first priority is evaluation of interaction parameters between anionic uranyl carbonate complexes and the dominant WIPP cations Na^+, K^+, Mg^{2+}, and Ca^{2+}.

Many of the species listed in Grenthe et al. (1992) need not be included in a complete model using the Pitzer formalism. For example the effects of the UO_2Cl^+ and $UO_2Cl_2(aq)$species are incorporated in existing parameters for UO_2^{2+}-Cl^- interactions. The situation is similar for the sulfate system. The behavior of the hydrolysis species could be explained adequately by a much smaller set of, perhaps three, hydrolysis species and their appropriate ion interaction parameters. This reduces the matrix of needed chemical properties to:

	Cl^-	SO_4^{2-}	CO_3^{2-}	$UO_2(CO_3)_2^{2-}$	$UO_2(CO_3)_3^{4-}$	$(UO_2)_3(CO_3)_6^{6-}$
Na^+	available	available	available	needed	needed	needed
UO_2^{2+}	available	available	possibly needed	not needed	not needed	not needed
UO_2OH^+	needed	possibly needed	not needed	not needed	not needed	not needed
$(UO_2)_2(OH)_2^{2+}$	needed	not needed	not needed	not needed	not needed	not needed
$(UO_2)_3(OH)_5^+$	needed	not needed	not needed	not needed	not needed	not needed
$(UO_2)_4(OH)_7^+$	needed	not needed	not needed	not needed	not needed	not needed

The above matrix lists the ion interaction parameters that are available, that are needed, and that may be needed for the WIPP +VI actinide solubility model. The interactions marked "not needed" have been assumed to be unimportant based on chemical reasoning. This and other assumptions for the +VI model will be tested and challenged using the empirical solubility studies in complex synthetic WIPP brines.

Laboratory experiments will evaluate the interaction parameters for uranyl carbonate complexes with Na^+. Experiments will then be extended to other media, time permitting, to evaluate the parameters with the remaining three cations.

While experiments are being performed under carbon dioxide, we will also model literature data for the hydrolysis of uranyl ion in the absence of carbonate, to obtain interaction parameters between the cationic hydrolytic uranyl species and Cl^-, the dominant anion found in WIPP brines. This will also extend to the evaluation of the interaction between anionic hydrolytic uranyl species, which form above pmH 7, with Na^+.

The carbonate experiments together with the hydrolysis model in NaCl will permit modeling of U(VI) solubility in the multi-component system Na-Cl-CO_2-HCO_3-CO_3-H-OH-H_2O.

Develop Uranyl Hydrolysis Model and Parameters

A summary of data for the aqueous chemical behavior of uranium is given in Grenthe et al. (1992). Based on this and more recent information for the scoping experiments and the literature, the Actinide Source Term team, including all contractors, has reached consensus that sufficient data exist to parameterize the uranyl hydrolysis model from existing information. However, not all literature data are reliable or relevant. Thus, the first part of uranyl hydrolysis model development is an evaluation of the literature data for suitability of use in the integrated uranyl hydrolysis model. All suitable data will then be used to determine uranyl hydrolysis parameters. The resulting model will predict uranyl hydrolysis behavior in concentrated aqueous chloride, sulfate, and perchlorate media.

Design, Conduct, and Analyze Experiments for Uranyl Complexation with Carbonate

The solubility of $UO_2CO_3(s)$ in NaCl solutions (0.01, 0.05, 0.08, 0.2, 0.8, 2.0, 3.0, 4.0, and 5.0 m) will be measured as a function of hydrogen ion concentration, at a constant $CO_2(g)$ pressure of 1 bar. These experiments will yield the standard chemical potentials for UO_2Cl^+, $UO_2CO_3(s)$, $UO_2CO_3(aq)$, $UO_2(CO_3)_2^{2-}$, $UO_2(CO_3)^{4-}$,3), and $(UO_2)_3(CO_3)_6^{6-}$. The multiple NaCl concentrations will allow evaluation of the interaction parameters between Na^+ and uranyl complexes.

The experiment will proceed in three phases. Solutions will be prepared at three ionic strengths, for example, 0.8, 3.0, and 5.0m so at the conclusion of these experiments an interim model can be developed and used to predict the next set of three ionic strength experiments. The results of the second phase will be used to refine the model. The process will be repeated. This iterative process will build confidence in the model by using to predict future experiments and to observe how well the predictions were realized.

Alternative +VI Actinide Model using the SIT Activity Coefficient Formalism (Contract #__-___; LANL)

The +VI numerical model is the least developed of all the actinide oxidation state models. To ensure that WIPP has a +VI model within the time frame of the WIPP Disposal Decision Plan, a parallel track for +VI model development was established. The +VI model development using the Pitzer formalism is the original track, and the +VI model development using the Specific Interaction Theory (SIT) (Grenthe et al., 1992) is the contingency track.

SIT is a thermodynamic formalism for calculating activity coefficients in electrolytes up to ~3.5 molal total ionic strength. Although all WIPP Salado and Castile brines are more concentrated, it may be possible to use SIT to adequately describe U(VI) solubility in 6 to 8 molal ionic strength WIPP brines. Extensive data for U(VI) solubility and speciation in simple electrolytes is available and interpreted using SIT (Grenthe et al., 1992). However, the SIT formalism has not been applied to brine systems. Thus, the situation for the SIT model is the opposite to that for the Pitzer model: for the Pitzer formalism, a good brine model exists, to which WIPP is adding actinides; for the SIT formalism, a good U(VI) data base exists, to which WIPP is adding a brine model.

The steps in the +VI "contingency model" development are:

- modify the EQ3/6 computer program (Wolery, 1992a; 1992b; Wolery and Daveler, 1992) to compute activity coefficients using the SIT (Grenthe et al., 1992) formalism;
- compare brine composition predictions, i.e., without actinides, with predictions from the Pitzer model using the Harvie-Møller-Weare/Felmy-Weare data base (Harvie et al., 1984; Felmy and Weare, 1986);
- predict U(VI) solubility in WIPP brines, and compare with results of empirical solubility measurements conducted under separate WIPP contracts; and
- develop lookup tables for U(VI) solubility for use in WIPP Performance Assessment.

Empirical Studies of the Solubility of +IV, +V, and +VI Actinides in WIPP Brines

A meeting to discuss actinide oxidation states under anticipated WIPP disposal room conditions was held at Sandia on 3 May 1995. Attending the meeting were DOE/CAO personnel, SNL personnel, contractors to the SNL AST work, and other scientists working with actinide elements and nuclear waste disposal issues. One of the conclusions of this meeting was the agreement that empirical solubility experiments in complex synthetic WIPP brines should be accelerated. These experiments, formerly part of the model development plan for each of the oxidation states, have been decoupled from the model development work, and will be performed under a separate contract with Argonne National Laboratory. These experiments will be used to challenge the thermodynamic models for the actinides that are available within the WIPP Disposal Decision Plan, and will also be used to provide empirical solubility measurements for any oxidation state for which a model is not ready within the WIPP Disposal Decision Plan. In addition, these experiments will challenge the oxidation state analogy for the actinides, and demonstrate the degree of accuracy under which the analogy can be applied.

Details about these empirical solubility studies are given in Novak (1995), Appendix B.

The Models for Complexation of Organic Ligands with +III, +IV, +V, and +VI Actinides (Contract AH-5590; FSU)

Organic ligands, which are present in the waste or may form from degradation of cellulosic material in the waste, hold the potential for complexing actinides and thus increasing total dissolved actinide concentrations in the aqueous phase. It is this complexing, also called chelating, property of organic

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ligands for actinides that is used for cleaning up, e.g., actinide spills. Brush (1990) identified the 10 organic ligands acetate, ascorbate, citrate, EDTA, α-hydroxyisobutyrate, lactate, oxalate, oxine, 1,10-phenanthroline, and TTA as organic ligands present in potential WIPP wastes that could potentially impact total dissolved actinide concentrations. These 10 ligands are those represented by "L^{p-}" in Reaction 1. During CY93, the complexation of these ligands with Am(III), Th(IV), Np(V), and U(VI) in 5.0m NaCl was measured to determine whether these organic ligands hold significant potential to increase aqueous actinide concentrations. The results of this experimental work divided these ligands into two categories. The first group, those expected to significantly enhance dissolved actinide concentrations, includes acetate, citrate, EDTA, lactate, and oxalate. The second group, those not expected to significantly enhance dissolved actinide concentrations, includes ascorbate, α-hydroxyisobutyrate, oxine, 1,10-phenanthroline, and TTA. Ascorbate is present in TRU wastes primarily as a redox agent, not for actinide complexation. Because α-hydroxyisobutyrate is chemically very similar to lactate, the models developed for lactate complexation with actinides will be applicable to α-hydroxyisobutyrate as well.

The AST experimental work to quantify the effects of organic ligands on dissolved actinide concentrations fall into three categories: the protonation/deprotonation behavior of the organic ligands, the complexation of actinides with the organic ligands, and the complexation of magnesium with the organic ligands, all as a function of sodium chloride concentration. This division was made for several reasons. Sodium chloride is the dominant electrolyte in WIPP brines, and the concentration of NaCl varies widely in the WIPP system. The Na^+ and Cl^- concentrations influence the concentrations of actinide complexes through specific ion interactions for cation-anion pairs, and this is expected to be a major effect. The protonation/deprotonation, or acid-base, behavior of the organic ligands is also important and is influenced by the NaCl concentration through specific ion interactions. The organic ligands listed above are all organic acid anions, that is, deprotonated organic acids, and it is the deprotonated form that complexes actinides. The protonation/deprotonation behavior of these organic acids, as a function of NaCl concentration, is necessary for a quantitative description of the complexation.

Models for the complexation and acid-base behavior of the organic ligands in NaCl media calculate the enhancement of dissolved actinide concentrations caused by complexation with organic ligands. This model would not include the effects of competitive complexation of magnesium for the ligands, and thus would tend to overestimate dissolved actinide concentrations in high magnesium brines typical of the Salado Formation. Measurement of the complexation behavior of magnesium with the organic ligands is given by the reaction

$$Mg^{2+} + L^{p-} \leftrightarrow MgL^{(2-p)} \qquad (2)$$

In brines that contain large magnesium concentrations, the complexation of Mg with the organic ligands, as given by Reaction 2, will reduce the free concentration of organic ligands, and is expected to effectively prevent the organic

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ligands from reacting with actinides and thus increasing dissolved actinide concentrations. After the models are developed from experimentally simple systems, they will be confirmed in complex synthetic brines based on brines occurring at the WIPP Site. The experiments within the program are designed to quantify these effects.

Actinide Complexation with Organic Ligand Experiments: Acid Constants for Five Organic Ligands

Actinides in WIPP brines are complexed by the deprotonated form of the organic ligands. The organic species deprotonate according to the reaction

$$H_pL(aq) \leftrightarrow H_{p-n}L^{(-n)} + nH^+ \qquad n=1,2,3,\ldots \tag{3}$$

where L is the organic ligand and p=1,2,etc. are for monoprotic acids, diprotic acids, etc. The concentrations of these deprotonated forms depend on the hydrogen ion concentration, the deprotonation, or "acid" constants, and specific ion interactions between the ligands and the major ions in solution. These experiments measure the deprotonation constants as a function of NaCl concentration to determine standard chemical potentials for ligand species and the specific ion interaction parameters between these species and Na^+. These experiments do not require actinides or any radioactivity to perform. Therefore, they are relatively simple and straightforward, except for the complications due to high ionic strength. The parameters from these experiments are equivalent to the Harvie-Møller-Weare parameters for the inorganic model. We require information about the chemical behavior of organic ligands in the absence of actinides in order to predict how actinides will behave in the same system. This activity represents 30 experiments (5 organic ligands at 6 NaCl concentrations).

Actinide Complexation with Organic Ligand Experiments: Magnesium Complexation with Organic Ligands in Concentrated NaCl Media

Chemical interactions of acetate, EDTA, oxalate, citrate, and lactate with Mg^{2+} will reduce the extent of actinide complexation by these ligands and result in lower dissolved actinide concentrations. Magnesium stability constant data with these ligands (in the above order) will be collected in solutions of 0.3, 1.0, 2.0, 3.0, 4.0, and 5.0 m NaCl, and will be used to develop a numerical description of these processes. Nonradioactive solvent extraction techniques similar to these used in the determination of actinide complexation with organic ligands will be employed. These stability constants will provide the data necessary for including Mg^{2+} complexation with organic ligands into the overall actinide dissolution model, and result in lower dissolved actinide concentrations in brines containing Mg^{2+}.

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Actinide Complexation with Organic Ligand Experiments: Complexation with Am(III), Th(IV), Np(V), and U(VI) in NaCl Media

The central group of experiments for organic ligands measures the complexation of the ligands with each of the prototypical actinides as a function of NaCl concentration. All the deprotonated forms of the ligands will be anionic, and thus specific ion interactions with Na^+ are expected to be most important. The complexation of the actinides with the organic ligands in 5.0m NaCl has been measured. Similar experiments at various NaCl concentrations such as 0.1, 0.3, 1.0, 2.0, and 3.0m, will provide the variation of complexation versus NaCl, which in turn will yield specific ion interaction parameters for Na^+ with anionic actinide-ligand complexes, or Cl^- with cationic actinide-ligand complexes. This activity represents 100 experiments (4 actinides with 5 organic ligands at 5 NaCl concentrations).

Actinide Complexation with Organic Ligand Experiments: Confirmatory Studies for Actinide Complexation with Organic Ligands in Complex Synthetic WIPP Brines

As noted for the inorganic models for dissolved actinide concentrations, the combined inorganic/organic model must predict the total dissolved actinide concentrations as a function of expected brine conditions in the WIPP system. These experiments will measure the dissolved actinide concentrations as a function of organic ligand concentration and hydrogen ion concentration in several synthetic WIPP brines. The model will predict the experimental results, and the agreement between the model and the data will confirm the model and support its applicability for predicting actinide concentrations in WIPP brines.

Measurements of Actinide Reduction Behavior for Determining Most Probable Actinide Oxidation States in the WIPP Disposal Room Environment

A meeting to discuss actinide oxidation states under anticipated WIPP disposal room conditions was held at Sandia on 3 May 1995. Attending the meeting were DOE/CAO personnel, SNL personnel, contractors to the SNL AST work, and other scientists working with actinide elements and nuclear waste disposal issues. One of the conclusions of this meeting was the agreement that David L. Clark (LANL), Andrew R. Felmy (PNL), and Dhanpat Rai (PNL) would prepare a joint proposal for work addressing the WIPP information need for actinide oxidation states. Ruth F. Weiner was selected as the Sandian responsible for technical management of this work. A draft proposal has been prepared and is being negotiated among the concerned parties.

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6748	H.W. Papenguth, MS 1320

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SCHEDULES: GANTT CHARTS

+III and +IV Schedule (Contract AF-3339; PNL)

The following pages contain Gantt charts for the +III and +IV actinide chemistry work at PNL. The Gantt charts for the +III and +IV actinides are merged because this work is being done under the same contract. The schedules present experimental work planned through FY96; and are contingent on the Sandia funding request for the Actinide Source Term Dissolved Concentration Model Development effort of $3.6M for FY96.

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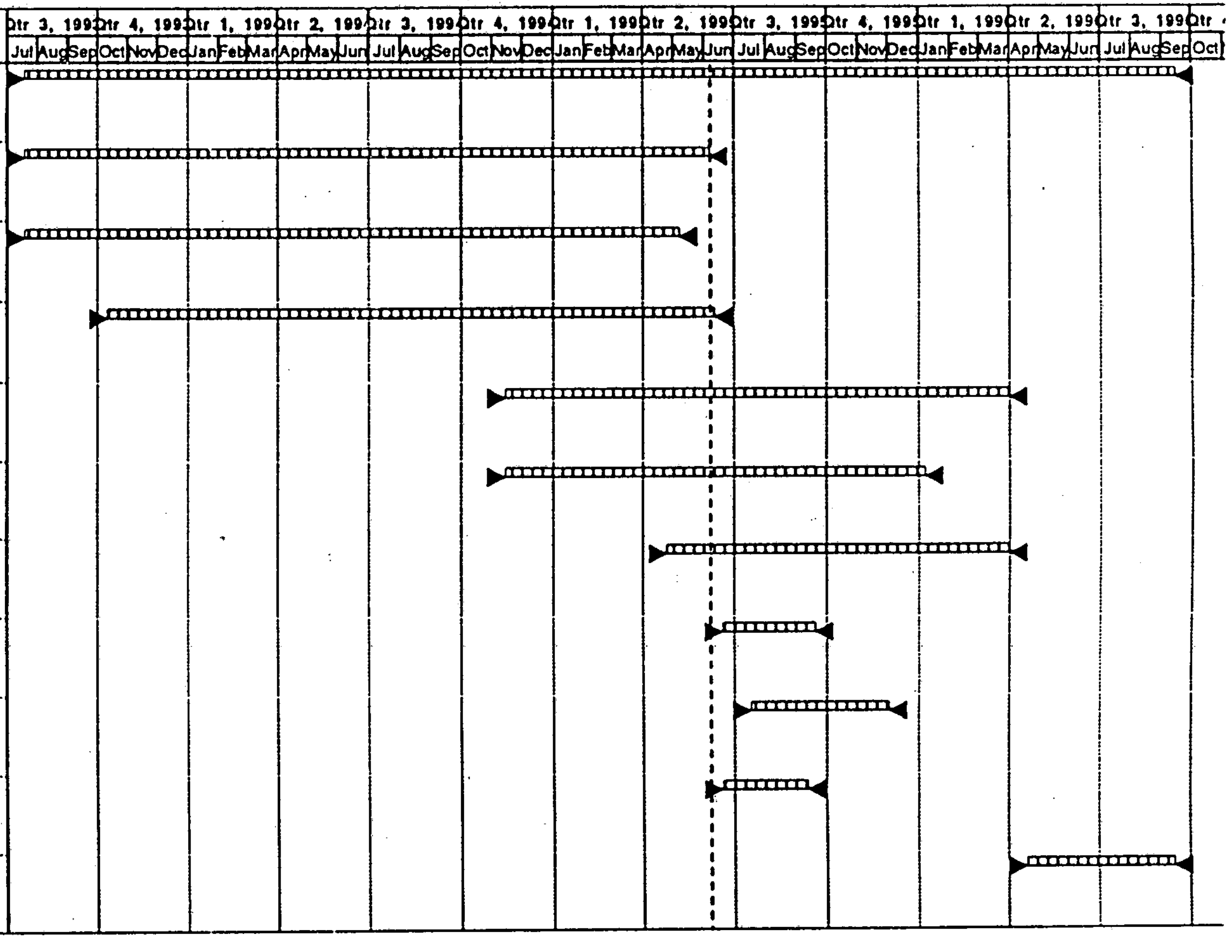
ID	Name	Dura	S. Start	S. Finish	Pred
1	PACIFIC NORTHWEST LAB CONTRACT	837d	7/1/93	10/2/96	
2	pcH MEASUREMENTS	517d	7/1/93	6/23/95	
11	COMPLEXATION OF Nd(III) WITH CO3-- (solubilities and solubility controlling	495d	7/1/93	5/24/95	
17	COMPLEXATION OF U(IV) WITH Cl- (solubilities and solubility controlling solids)	460d	9/23/93	6/28/95	
26	COMPLEXATION OF U(IV) WITH SO4-- (solubilities and solubility controlling solids)	375d	10/27/94	4/17/96	
31	Nd(III) IN COMPLEX BRINES AND MODEL TESTING	315d	10/27/94	1/24/96	
36	COMPLEXATION OF Np(IV) WITH Cl-	260d	4/6/95	4/17/96	
41	Np(IV),U(IV),Pu(IV) IN COMPLEX BRINES AND MODEL TESTING	90d	6/1/95	10/6/95	
46	Pu(III), Am(III) in complex synthetic WIPP brines	120d	6/29/95	12/19/95	
51	Interpretation of data of CO3-2 complexation with Th(IV) and U(IV)	85d	6/1/95	9/29/95	
56	Final Wrap-up Activities	130d	4/1/96	10/2/96	



Project: Pacific Northwest Lab Co
Date: 6/7/95

Critical ▨ Noncritical ■ Progress ▬ Milestone ◆ Summary ▼▭▼ Rolled Up ◇

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+V Schedule, Part 1 (Contract AH-5592; LBL)

The following pages contain Gantt charts for the +V actinide chemistry work at LBL. Related Gantt charts for the work being conducted through the GTS/ITS are in the next section. The schedules present experimental work planned through March 96; and are contingent on the Sandia funding request for the Actinide Source Term Dissolved Concentration Model Development effort of $3.6M for FY96.

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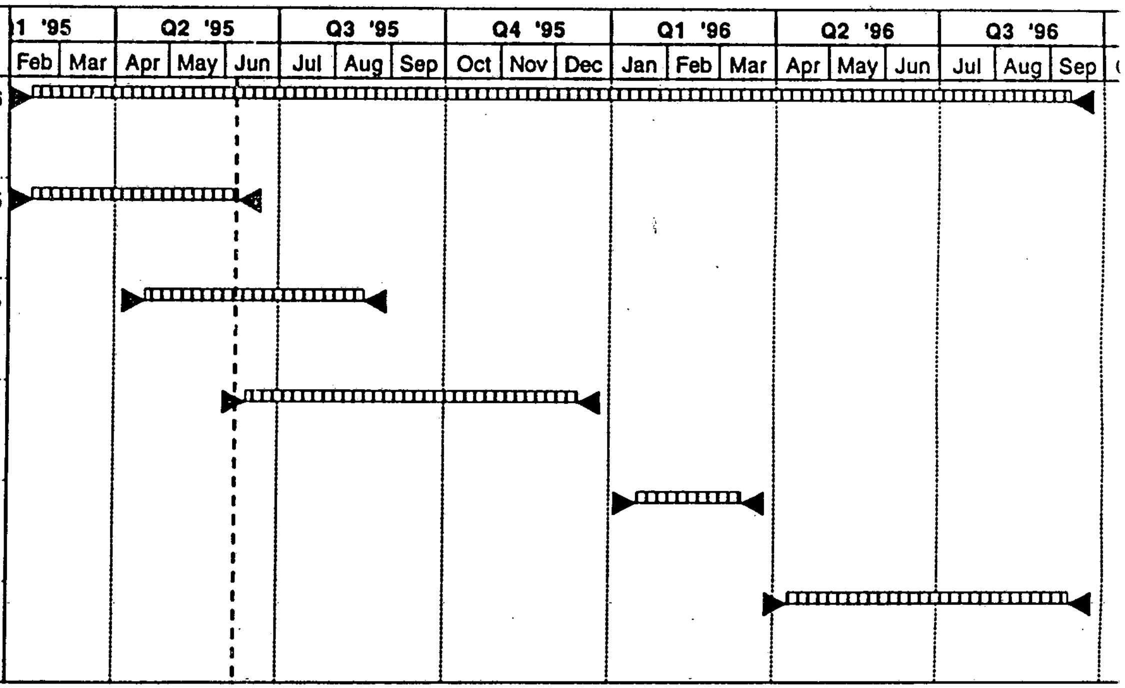

ID	Name	Dura	S. Start	S. Finish
1	Lawrence Berkeley Labs +V work	430d	2/1/95	9/24/96
2	Study Np(V) in K2CO3 experiments	100d	2/1/95	6/20/95
6	Study Np(V) in MgCl2 media	105d	4/5/95	8/29/95
10	Study Np(V) in mixed Na-K-Mg-Cl experiments	150d	5/31/95	12/26/95
14	Confirmatory experiment for Np(V) solubility in complex synthetic brines	60d	1/3/96	3/26/96
18	Final Wrap-up Activities	130d	3/27/96	9/24/96

IP94

Project: LBL +V lab work
Date: 6/7/95

Critical	▨	Progress	▬	Summary	▶▭◀
Noncritical	■	Milestone	◆	Rolled Up	◇

27/37

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+V Schedule, Part 2 (Contract AN-1756; GTS/ITS)

The following pages contain Gantt charts for the +V actinide chemistry work at the GTS/ITS. Related Gantt charts for the work being conducted through LBL are in the previous section. The schedules present experimental work planned through March 96; and are contingent on the Sandia funding request for the Actinide Source Term Dissolved Concentration Model Development effort of $3.6M for FY96.

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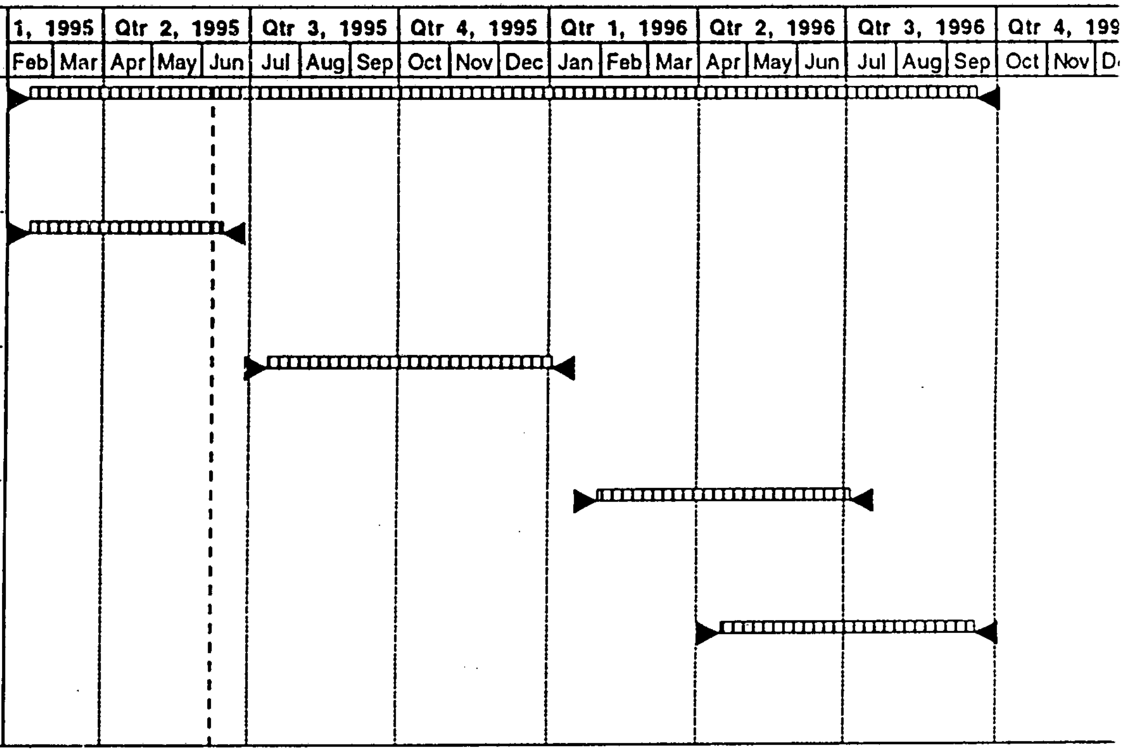
ID	Name	Dura	S. Start	S. Finish
1	Wolfgang Runde Contract work support of +V oxidation state analogy @ LANL	423d	2/1/95	10/2/96
2	Modeling the Np(V)-Na-Cl-CO3-HCO3-H-O system using new data from German Nuclear Waste	105d	2/1/95	6/27/95
7	Isolating Pu(V) for Oxidation Analogy Demonstration	135d	6/28/95	1/16/96
12	Confirmatory studies for Np(V) - Pu(V) oxidation state analogy	130d	1/17/96	7/18/96
17	Final Wrap-up Activities	130d	4/1/96	10/2/96



[illegible]
29/37

Project: Runde contract
Date: 6/7/95

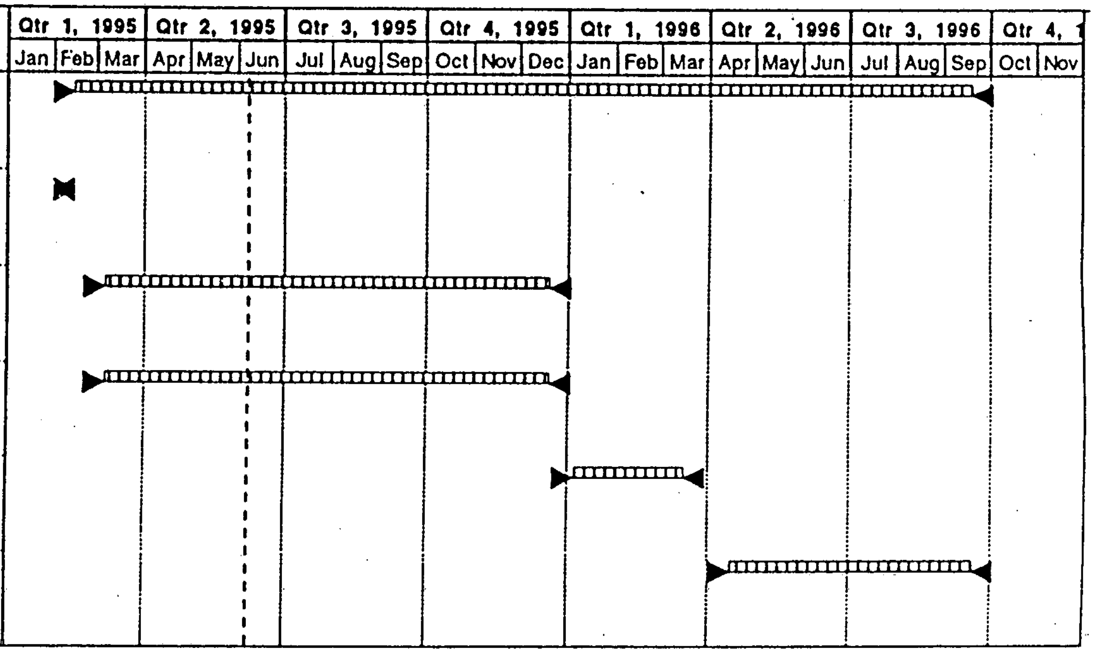
Critical ▨
Noncritical ■
Progress
Milestone ◆
Summary
Rolled Up ◇

B29

+VI Schedule (Contract AF-3341; LLNL)

The following pages contain Gantt charts for the +VI actinide chemistry work at LLNL. The schedules present experimental work planned through March 96; and are contingent on the Sandia funding request for the Actinide Source Term Dissolved Concentration Model Development effort of $3.6M for FY96.

ID	Name	Dura	S. Start	S. Finish	Pred
1	Lawrence Livermore Lab +VI work	424d	1/31/95	10/2/96	
2	Literature survey and experimental design	10d	1/31/95	2/13/95	
5	Hydrolysis Model Effort	217d	2/20/95	1/2/96	
10	Carbonate Experimental and Model Effort	217d	2/20/95	1/2/96	
17	Experiments to Confirm Prioritization	65d	12/22/95	3/29/96	
20	Final Wrap-up Activities	130d	4/1/96	10/2/96	



TP98
31/37

Project: LLNL +VI lab
Date: 6/7/95

Critical	▨	Progress	▬	Summary	▶▭◀
Noncritical	■	Milestone	◆	Rolled Up	◇

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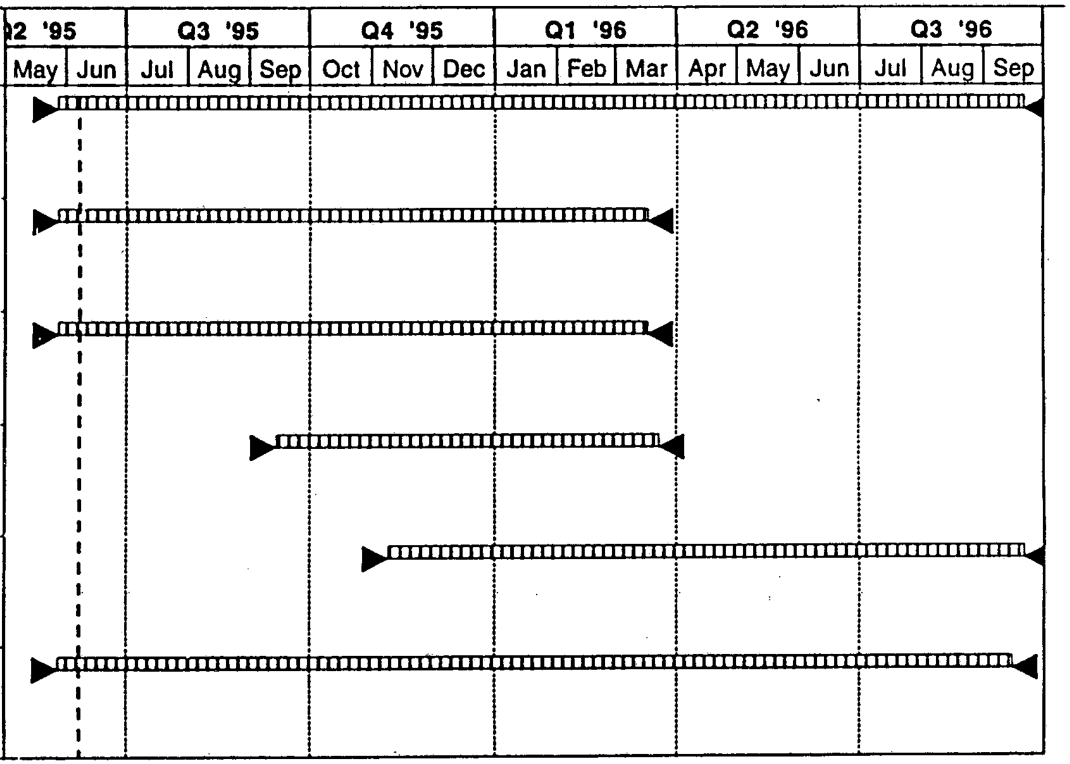
+Schedule for Alternative +VI Model (Contract __-____; LANL)

The following pages contain Gantt charts for the thermodynamic interpretation of +VI data using the SIT formalism, is being conducted at LANL. The schedules present experimental work planned through March 96; and are contingent on the Sandia funding request for the Actinide Source Term Dissolved Concentration Model Development effort of $3.6Mfor FY96.

ID	Name	Dura	S. Start	S. Finish
1	SIT Project for LANL (Janecky) U(VI) work at LANL	364d	5/15/95	10/3/96
2	Collect Data	230d	5/15/95	3/29/96
8	Implement Model	230d	5/15/95	3/29/96
15	Define Test Case	155d	9/1/95	4/4/96
21	Define Applications	245d	10/27/95	10/3/96
27	Management, Intergration, Quality Assurance	360d	5/15/95	9/27/96



TP 100
33/37

Project: Janecky @ LANL/SIT
Date: 6/7/95

Critical	▨	Progress	▬	Summary	▶▭◀
Noncritical	■	Milestone	◆	Rolled Up	◇

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Organic Ligand Work (Contract AH-5590;FSU)

The following pages contain Gantt charts for the complexation of +III, +IV, +V, and +VI actinide complexation with organic ligands, being conducted at FSU. The schedules present experimental work planned through March 96; and are contingent on the Sandia funding request for the Actinide Source Term Dissolved Concentration Model Development effort of $3.6M for FY96.

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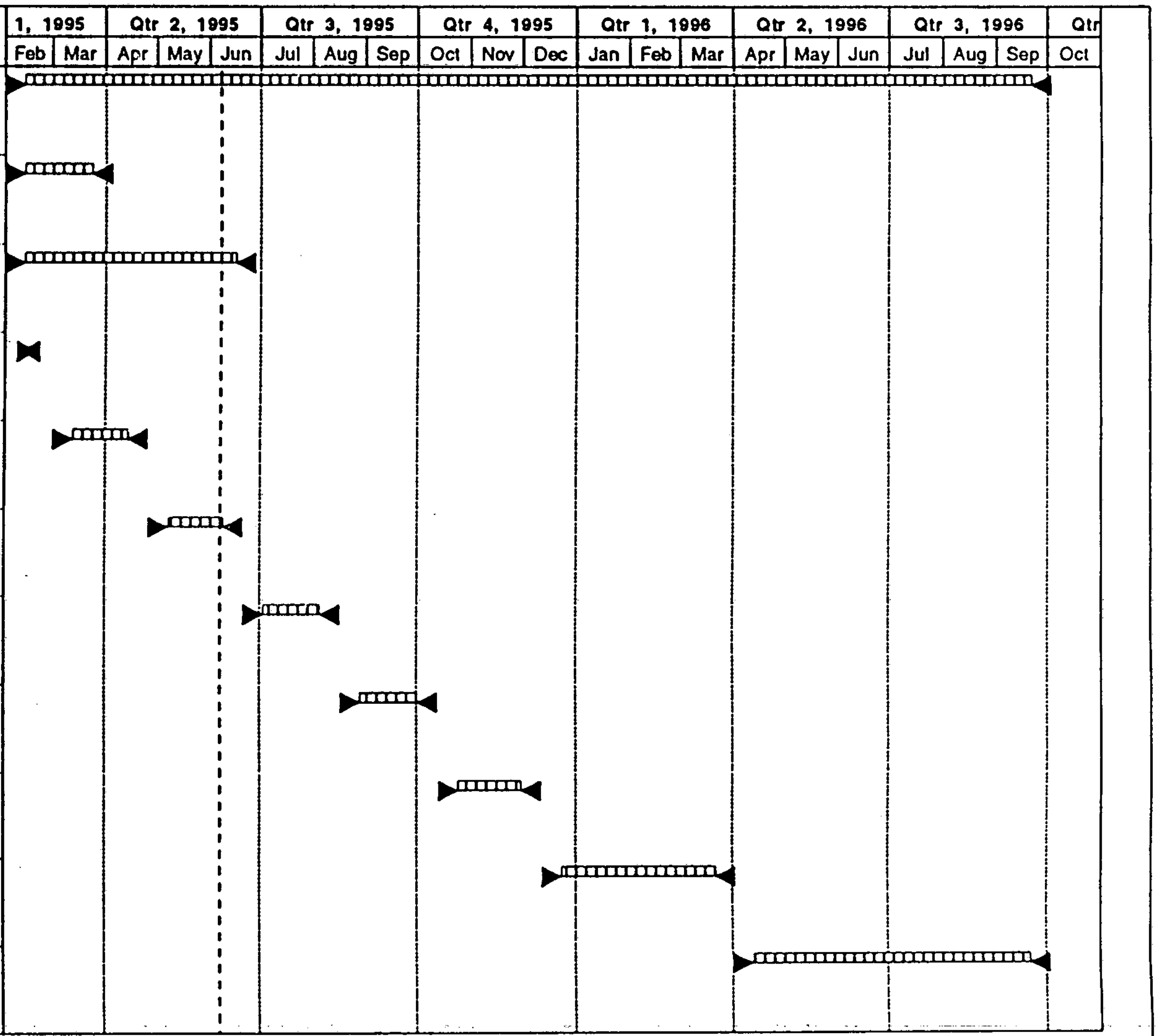
ID	Name	Dura	S. Start	S. Finish	Pred
1	Florida State University Organic Ligand Work	423d	2/1/95	10/2/96	
2	MEASUREMENTS OF LIGAND ACID CONSTANTS AS A FUNCTION OF NaCl	45d	2/1/95	4/4/95	
7	Study of 6 ligand with 3 NaCl concentrations in a Mg++ system with actinides.	105d	2/1/95	6/27/95	
12	QA Audit	10d	2/8/95	2/21/95	
15	4 studies: with each of 4 actinides and acetate in presence of NaCl	40d	3/1/95	4/25/95	
20	4 studies: with each of 4 actinides and oxalate in presence of NaCl	40d	4/26/95	6/20/95	
25	4 studies: with each of 4 actinides and lactate in presence of NaCl	40d	6/21/95	8/16/95	
30	4 studies: with each of 4 actinides and citrate in presence of NaCl	40d	8/17/95	10/12/95	
35	4 studies: with each of 4 actinides and EDTA in presence of NaCl	40d	10/13/95	12/11/95	
40	Experiments to challenge model: actinides + organics in complex synthetic brines	75d	12/12/95	4/2/96	
45	Final Wrap-up Activities	130d	4/1/96	10/2/96	



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TP 10:
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Project: Florida State University
Date: 6/7/95

Critical	▨	Progress	▬	Summary	▶▭◀
Noncritical	■	Milestone	◆	Rolled Up	◇

SNL-Based Work (Showing what the models mean for the WIPP)

The following pages contain Gantt charts for technical work at Sandia, showing the schedules for interpreting contractor information for WIPP Performance Assessment. Chemical information from the contractors needs to be processed at Sandia for WIPP performance assessment scenarios, and examined for implications of backfill additives and engineering alternatives.

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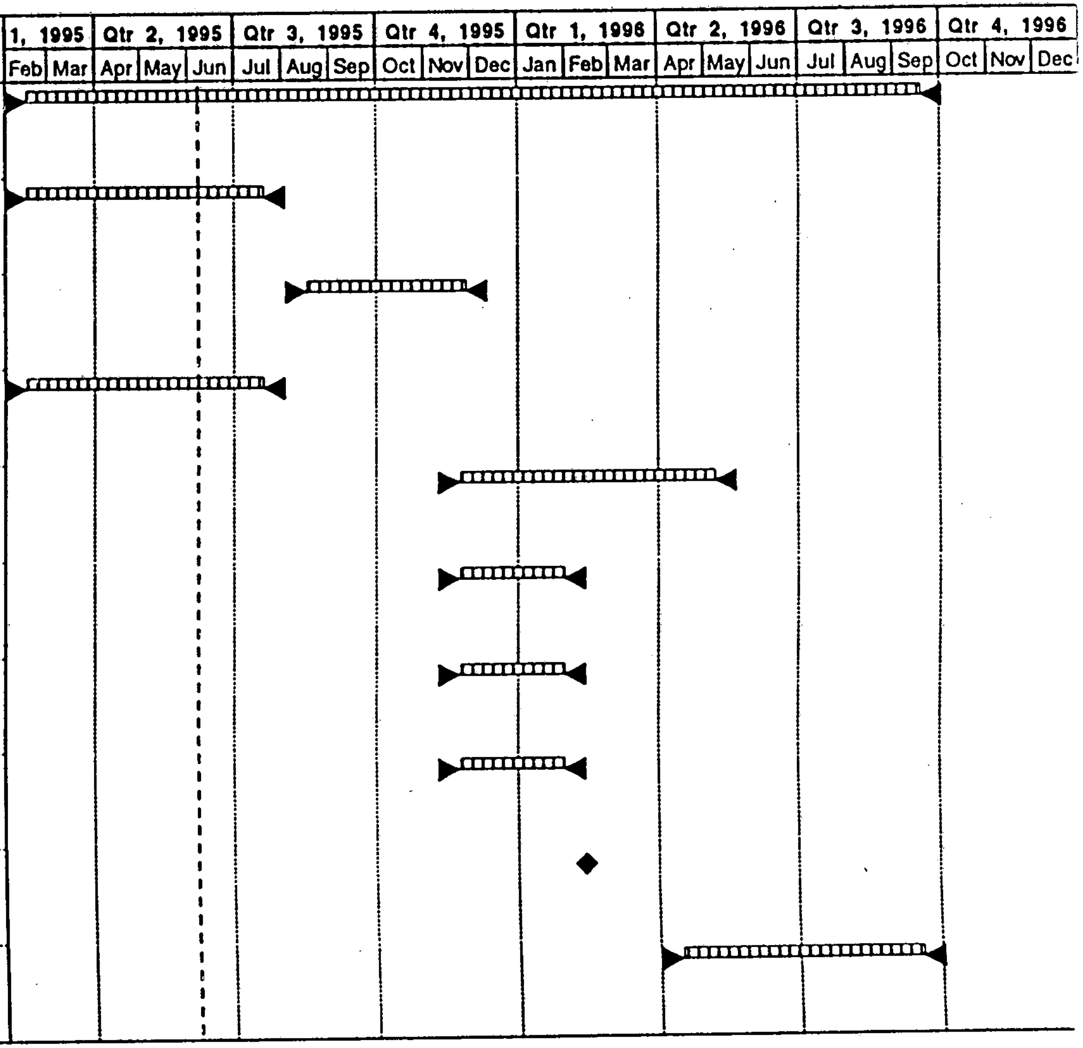
ID	Name	Dura	S. Start	S. Finish	Pred
1	SNL/Contractor information feeds	423d	2/1/95	10/2/96	
2	Interpretation of provisional inorganic +III dissolved concentration model for	130d	2/1/95	8/2/95	
8	Interpretation of provisional inorganic +IV dissolved concentration model for	90d	8/3/95	12/11/95	
14	Interpretation of provisional inorganic +V dissolved concentration model for	130d	2/1/95	8/2/95	
20	Re-interpretation of +III dissolved concentration model for WIPP scenarios,	130d	11/10/95	5/21/96	
25	Re-interpretation of +IV dissolved concentration model for WIPP scenarios,	60d	11/10/95	2/13/96	
30	Re-interpretation of +V dissolved concentration model for WIPP scenarios,	60d	11/10/95	2/13/96	
35	Re-interpretation of +VI dissolved concentration model for WIPP scenarios,	60d	11/10/95	2/13/96	
40	Dissolved Actinic Model development complete	0d	2/13/96	2/13/96	39
41	Final Wrap-up Activities	130d	4/1/96	10/2/96	



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Project: SNL/ASTP
Date: 6/7/95

Critical	▨	Progress	■	Summary	▶▭◀
Noncritical	■	Milestone	◆	Rolled Up	◇

Sandia National Laboratories

MANAGED BY MARTIN MARIETTA CORPORATION
FOR THE U. S. DEPARTMENT OF ENERGY

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date: 6 March 1995

to: Butch Stroud, Dick Lark, DOE/CAO

Craig F Novak

from: Craig F. Novak, Dept. 6119, MS 1320, 848-0619

subject: Experimental and Modeling Program for Determining Mobile Dissolved Actinide Concentrations for Scenarios Relevant to the WIPP: Summary of Program Accomplishments from Inception through 31 January 1995.

This memorandum documents the history and accomplishments of the experimental and modeling program for dissolved actinides in WIPP Performance Assessment scenarios. A companion memo documents the FY95-96 experimental and modeling program for dissolved actinide concentrations (Novak, 1995c).

BACKGROUND

The Actinide Source Term Program (ASTP) was begun in late calendar year 1992. The ASTP was chartered with developing and implementing an experimental and modeling program for estimating potential mobile actinide concentrations for the WIPP. The ASTP has three components, a laboratory/modeling subprogram for dissolved actinides, a laboratory/modeling subprogram for colloidal actinides, and a laboratory subprogram using actual transuranic wastes. R.C. Lincoln was the Sandia manager responsible for the ASTP. E.J. Nowak, then a staff member, was the team leader. C.F. Novak, H.W. Papenguth, and M.L.F. Phillips were staff members assigned part time to each of the subprograms, respectively.

Initial stages of the ASTP were focused on defining the Performance Assessment information needs for the Actinide Source Term, and developing approaches to meet those needs within cost and schedule constraints. Two documents were produced during this stage, the *Technical Requirements for the Actinide Source-Term Waste Test Program* (Phillips and Molecke, 1993) and the "Model Development Plan," which instead of being published was incorporated into the test plans for dissolved and colloidal actinides. This stage involved developing approaches to meet the needs of Performance Assessment, and

TP 10.5

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identifying and interviewing contractors capable of performing the necessary measurements with highly radioactive actinide elements.

Contracts for performing actinide dissolved concentration work at Battelle-Pacific Northwest Laboratory (PNL) and Lawrence Livermore National Laboratory (LLNL) were placed in April 1993. These locations spent approximately three to four months preparing their facilities for the WIPP work, including reconfiguring laboratory and glove box areas, ensuring adequate and appropriate radioactive elements and analytical equipment were available, and developing Quality Assurance documents. Laboratory experiments began at these locations in late FY93.

Contracts for performing actinide dissolved concentration work at Lawrence Berkeley Laboratory (LBL) and Florida State University (FSU) were already in place. It was decided that work at these locations would be continued, but the nature and focus of the work would be reoriented. The transition took place in summer 1993; "new" laboratory experiments began at these locations in late FY93 as well.

ACCOMPLISHMENTS TO DATE

The accomplishments of the Actinide Source Term Dissolved Concentration Subprogram are summarized below, including references to detailed source reports.

The FY94 research conducted for the Actinide Source Term dissolved concentration modeling effort will be published as SAND94-2274, Novak, C.F., editor. 1995. "Actinide Chemistry Research Supporting the Waste Isolation Pilot Plant (WIPP): FY94 Results." This report is currently in Sandia management review and is undergoing final formatting for publication. This report is a compilation of the following articles: Erten et al. (1994), Khalili et al. (1994), Nitsche et al. (1994), Novak and Roberts (1994), Rao et al. (1994), and Reed et al., (1994), which are discussed below.

General

- Experimental methods for determining the hydrogen ion concentration, "pcH," in the concentrated electrolytes NaCl, Na_2SO_4, and $NaCl/Na_2SO_4$ mixtures were developed and documented (Rai et al., 1995). Consistent methods for determining pcH in concentrated electrolytes such as WIPP brines are necessary to interpret data with thermodynamic formalism.

+III Actinides (Pu(III), Am(III))

- Parameter development for Am(III) and Pu(III) solubilities in concentrated carbonate electrolytes (Rao et al., 1994) was completed based on chemical measurements with Nd(III). This work, in addition to work measuring

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the solubility of Am(III) in mixed phosphate/sulfate media (Rai et al., 1994), which was not funded by WIPP, completed the set of binary interaction parameters for the inorganic An(III) model. (Demonstration that ternary interactions are not needed, and measurement of An(III) solubilities in synthetic WIPP brines, are in progress.) The entire model is documented in the memoranda Novak (1995a) and Novak (1995b).

- Measurements of the solubility of Nd(III), a chemical analog for Pu(III) and Am(III), in a synthetic Salado brine (SPC Brine) were completed as a function of hydrogen ion concentration (Khalili et al., 1994). These data provided the solubility-controlling solid phases and preliminary data against which to compare the An(III) model.

+IV Actinides (Th(IV), U(IV), Np(IV), Pu(IV))

- The solubility of Th(IV), as hydrous Th(IV) oxide, was measured in 4.0 and 6.0 molal NaCl and in 1.00, 1.82, and 3.00 molal $MgCl_2$ solutions, as documented in the draft report "The Solubility of Hydrous Th(IV) Oxide in Concentrated NaCl and $MgCl_2$ Solutions," (Contract AF-3339 Monthly Letter Report, 30 December 1994). These data, along with other data in the literature (Felmy et al., 1991; Roy et al., 1992) have been used to develop a preliminary model for the ion interactions between Na^+ and Th^{4+} and between Mg^{2+} and Th^{4+}.

- The solubility of U(IV), as hydrous U(IV) oxide, was measured in 0.2, 1.0, and 6.0 molal NaCl and in 1.0, 2.0, and 3.0 molal $MgCl_2$ solutions, as documented in the draft report "The Solubility of Hydrous U(IV) Oxide in Concentrated NaCl and $MgCl_2$ Solutions," (Contract AF-3339 Monthly Letter Report, 30 December 1994). These data are being interpreted along with other data from the literature (Rai et al., 1990) to develop a preliminary thermodynamic model for U(IV) in these solutions.

+V Actinides (Np(V), Pu(V), Am(V))

- A consistent model for Np(V) solubility in Na-Cl-CO_3 media was developed and applied to solubility measurements in Culebra brines with good agreement (Novak and Roberts, 1994). The model should apply to Castile brines, but solubility data for Np(V) in Castile brines are not available. Effects of magnesium and potassium must be included to render this model applicable to Salado brines.

- Experiments measuring the solubility and oxidation state distributions of plutonium in a synthetic Culebra brine (H-17 Brine) were completed (Nitsche et al., 1994). This work complements measurements of plutonium solubility and oxidation state distribution in synthetic Culebra AISinR (Air Intake Shaft) brine (Nitsche et al., 1992), which was performed before the formal inception of the Actinide Source Term Program. These data demonstrated that, under oxic conditions in synthetic Culebra brines, plutonium exists primarily as Pu(V) and Pu(VI), and the solubility-controlling solid phase is $NaPu(V)O_2CO_3 \cdot xH_2O(s)$.

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Similar work in synthetic Salado SPC Brine is completed, but documentation is incomplete; the draft report "Plutonium(V) and (VI) Solubilities in Synthetic Brines of Interest to the Waste Isolation Pilot Plant (WIPP) Project" was received at Sandia on 23 November 1994.

- Experiments measuring the solubility and oxidation state distributions of neptunium in two synthetic Culebra brines (AISinR and H-17 Brines) and one synthetic Salado brine (SPC Brine) were nearly completed. Final analyses of the solid phases and measurements of the inorganic carbon content of the solutions are in progress. These experiments are complimentary to those for plutonium discussed above, and can be used to demonstrate the oxidation state analogy between Pu(V) and Np(V) chemistry. Documentation of this work is incomplete; the draft report "Neptunium(V) and (VI) Solubilities in Synthetic Brines of Interest to the Waste Isolation Pilot Plant (WIPP) Project" was received at Sandia on 23 November 1994.

+VI Actinides (U(VI), Np(VI), Pu(VI))

- The stability of Pu(VI) in synthetic Salado and Castile brines was demonstrated as part of the Gas Generation Program by Reed et al. (1994). This work, although formally not part of the Actinide Source Term, showed that the carbonate ligand can stabilize plutonium in the +VI oxidation state for long periods of time, preventing it from becoming reduced to lower oxidation states. The implications of this observation are being examined within the Actinide Source Term program.

- Scoping experiments measuring the solubility of U(VI) in 0.001m NaCl, 5.2m NaCl, and 2.0m Na_2SO_4 were completed. These experiments yielded the identities of the solubility-controlling solid phases, and the preliminary dependence of total dissolved uranyl ion concentration on the hydrogen ion concentration and salt content (Contract AF-3341 Monthly Letter Reports: 31 October 1994; 26 August 1994; 27 July 1994). These results are being used to design detailed experiments from which to obtain data for developing thermodynamic modeling parameters.

Complexation with Organic Ligands

- The complexation constants of Th(IV) and U(VI) with oxalate were measured as functions of sodium perchlorate concentration (Erten et al., 1994). This work shows the potential increase in dissolved Th(IV) and dissolved U(VI) caused by the organic ligand oxalate, which is expected to be present in WIPP wastes.

- Measurements of the complexation of Am(III), Th(IV), Np(V), and U(VI) with the organic ligands acetate, ascorbate, citrate, EDTA, α-hydroxyisobutyrate, lactate, oxalate, oxine, 1,10-phenanthroline, and TTA in 5.0m NaCl were completed (Contract AH-5590, Monthly Letter Reports: 29 November 1993; 29 December 1993). This data permitted ranking of the organic ligands in order of

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potential impact on dissolved actinide concentrations under WIPP conditions, and a corresponding prioritization of experimental work.

SUMMARY

Significant progress has been made in measuring data needed to predict dissolved actinide concentrations in WIPP brines for Performance Assessment. Sufficient information is now available to produce preliminary predictions of the impact of waste-brine interactions, gas-brine interactions, and brine-brine (e.g., Salado-Castile) interactions on the dissolved concentrations of Am(III) and Pu(III). Calculations demonstrating this predictive capability are in progress, and are scheduled for completion in June 1995.

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APPENDIX C: DESCRIPTIONS OF THE EXPERIMENTAL PROGRAM FOR EMPIRICAL SOLUBILITY MEASUREMENTS

Moore, R.C. 1995. "Empirical Solubility Measurements for Actinides in WIPP Brines: Descriptions, Deliverables, and Schedule." memorandum to C.F. Novak. 6 June 1995.

Sandia National Laboratories

Albuquerque, New Mexico 87185-1341

date: 6 June 1995

to: C. F. Novak, Org. 6748, MS-1320

from: Robert C. Moore, Org. 6748, MS-1341, 848-0146

subject: **Empirical Solubility Measurements for Actinides in WIPP Brines: Descriptions, Deliverables, and Schedule.**

Definition

The Program for Actinide Stability/Solubility in Simulated WIPP Brines has been initiated as a test program and contingency plan for the Actinide Dissolved Concentration Subprogram (ADCS) of the Actinide Source Term (AST) Program. The Actinide Dissolved Concentration Subprogram represents an experimental and modeling effort to describe the solubility behavior of actinides in concentrated brines. Experimental programs at national laboratories and universities have been designed to measure the activity of actinides in simple salt solutions. The data from these experiments are being integrated at Sandia National Laboratory into a complete actinide solubility model based on the Pitzer treatment for electrolytes. The model must be able to predict total dissolved actinide concentrations as a function of pH, ionic strength, brine composition, and organic ligand concentration.

The Program for Actinide Stability/Solubility in Simulated WIPP Brines is concerned with providing solubility data for actinides in complex brines, similar to WIPP brines, to test and challenge the predictive ability of the Actinide Dissolved Concentration Model. Test data will be generated by performing empirical solubility experiments for actinides in synthetic WIPP brines. Because of the complexity of the system, experiments cannot be performed to simulate all conditions possible in the WIPP disposal room. Therefore, measurements will be taken for three actinides and two synthetic brines at two pmH levels. The effects of carbonate and three organic ligands important to WIPP will

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be determined. The results from this work will indicate which oxidation states are stable for each actinide in the synthetic brines, and for those stable actinides the total concentration as a function of pH and brine composition will be determined. These experimental results will be compared to predictions from the ADCS model.

There are several major differences between the Source Term Test Program (STTP) experiments being conducted at Los Alamos National Laboratory and the solubility experiments in this work. In the STTP additional materials (cellulosics, iron, plastics, microorganisms) have been added to the test vessels to simulate the effects of these substances at the WIPP site. Mechanisms for microbial activity and sorption are not included in the ADCS model. Therefore, direct comparison of results from the STTP with predictions of the ADCS model is difficult. The solubility experiments in this work are being conducted as a function of brine composition, pmH, carbonate concentration, and organic ligand concentration only. The results should serve as a quantitative test for the predictive ability of the ADCS model for actinide solubility in complex brine systems very similar to actual WIPP brines.

The Actinide Stability/Solubility Program will also serve as a contingency plan for the +VI oxidation state part of the Actinide Dissolved Concentration Subprogram. Because of the tight schedule and forthcoming deadlines of the WIPP program, it was deemed necessary to establish a contingency plan for this part of the ADC Subprogram. Unforeseen delays in the experimental programs could delay the development of the complete actinide solubility model. In that event, the Actinide Stability/Solubility Program will be used to provide empirical solubility data for input into the WIPP Compliance Application to the Environmental Protection Agency.

This work is being done independently of the ADCS modeling effort. This independence is important for maintaining the utility of the empirical solubility data for testing the ADC models. These data will not be incorporated into the ADC model. The work for this project will be performed at Argonne National Laboratory.

Actinides

The Actinide Dissolved Concentration Subprogram is focusing on the actinide elements thorium, uranium, neptunium, plutonium, and americium in their various +III, +IV, +V, and +VI oxidation states. The Stability/Solubility Program at Argonne National Laboratory will concentrate on three elements, uranium, neptunium, and plutonium, in the +IV, +V, and +VI oxidation states. Complex brine solubility experiments for thorium and americium and all elements in the +III oxidation state are being done at Pacific Northwest Laboratory.

Synthetic WIPP Brines

Brines present at the WIPP site have various composition and concentration, and no particular brine composition can be considered representative of all WIPP brines. In general, brines can be divided into two categories, Castile and Salado brines, due to their presence in these geological formations at the site. The Castile brines are essentially saturated sodium chloride solutions and the Salado brines primarily composed of sodium chloride and magnesium. For this project, one synthetic Salado brine, G-Seep, and one synthetic Castile brines, ERDA-6, will be used for solubility experiments. The composition of these synthetic brines is given by Brush. (Brush 1990)

Because the Castile and Salado brines are saturated with evaporite minerals, it may be necessary to dilute the synthetic brines by approximately 10%. This would avoid precipitation of salts that would greatly complicate solids analysis. This may be especially important when performing experiments with the addition of carbonate.

Experimental Conditions

After closure, any oxygen sealed in the WIPP disposal room will be depleted by corrosion and aerobic microbial activity. Generation of oxygen by radiolysis is believed negligible. (Lappin et al. 1989) Therefore, all solubility experiments in this work will be conducted under anoxic conditions. Because of corrosion and microbial activity, large amounts of hydrogen and carbon dioxide are expected to be generated. (Brush 1990) The effects of these gases will be taken into account by conducting all solubility experiments in the presence of a hydrogen atmosphere and performing certain solubility experiments with the addition of carbonate.

Experimental Program

The Actinide Stability/Solubility Program is divided into three major Tasks. Task 1 involves oxidation-state-specific solubility studies. This work will evaluate the redox stability and determine steady state concentrations as a function of experimental conditions. Task II involves repeating stability/solubility experiments performed in Task I with the addition of carbonate to the test vessels. Task III involves evaluating the effect of organic complexation on the solubility of actinides.

Task I - Solubility of Actinides in WIPP Synthetic Brines under Anoxic Conditions

Initial experiments will be performed for the +VI actinides. Currently, the +VI program is in the earliest stage of development. Additionally, literature data have indicated the +VI solubility experiments may be very challenging, because the solubility controlling solid phase is a function of both ionic strength and pH (Novak, 1995; Yamazaki et al., 1992; Kramer-Schnabel et al., 1991). Short-term stability experiments will be done first to determine if the +VI actinides are stable with respect to reduction in the synthetic brines under anoxic conditions. It is believed under the reducing conditions of the experiments that some of the +VI actinides will rapidly reduce to the +V and/or +IV state. These will be small-scale experiments conducted in a nitrogen glove box, and should begin June 1995. The results of the stability experiments will be used to plan larger-scale solubility experiments for the redox stable +VI actinides. Following the completion of the +VI work, the +V and then the +IV actinide solubility experiments will be performed. Because actinides in the +V and +IV state are likely to be stable with respect to reduction, short-term stability experiments will not be necessary, and only long-term solubility experiments will be done. However, the oxidation state of actinides will be monitored during all the experiments.

A generalized matrix for the experiments is given in Table 1. Experiments will be performed for uranium, neptunium, and plutonium initially in the +IV and +V oxidation state. Solubility experiments for the +VI oxidation state will depend on the results of the +VI stability experiments. Actinide solubility will be measured at two different pmH levels for each synthetic brine. Experiments will be conducted at pmH 8 and 10 for the Castile brine while the Salado brine experiments will be performed at pmH 5 and 7. The higher pmH values for the synthetic Castile brine were selected to prevent precipitation of magnesium during the experiments.

All experiments will be performed in a temperature controlled glove box maintained at 25°C. The experiments will use existing equipment initially designed and constructed for the WIPP Gas Generation Program. All solubility experiments will be performed in 2 L pressure vessels constructed of Hastelloy with polypropylene liners.

Approximately 1 L of brine solution will be used per experiment. The large volume will allow for sufficient solids to precipitate and be collected for analysis.

The following general experimental approach will be used throughout:

1. Oxidize Pu, Np, and U to the appropriate (+IV, +V, or +VI) oxidation state

2. Set-up and test Hastelloy pressure vessels with polypropylene liners with brine. Overpressure with approximately 1 atm of hydrogen and let sit for approximately 1 week to establish reducing conditions. The gas phase will be analyzed for oxygen content to ensure anoxic conditions.

3. Experiments will be initiated by adding an actinide in a specific oxidation state to the brine.

4. The actinide concentration, composition of the gas phase, and actinide oxidation state speciation will be monitored as a function of time.

5. After steady state has been reached, the vessel will be disassembled in a nitrogen glove box and the solution pH and redox potential measured. Also, solids will be collected and analyzed at this point.

No attempt will be made to control conditions (pH, oxidation state) in the vessels after an experiment has been initiated. Rather, the system will be allowed to change and reach a steady state. A sufficiently large amount of actinide will be added to each vessel to ensure an actinide solid precipitate forms. Experiments will be run until steady state is verified. The preliminary experimental plan calls for solution samples to be taken at 3 days, 1 week, 4 weeks, and 4 months. However, additional samples will be taken if necessary.

Analysis of Samples

The following analysis will be performed when sampling the contents of the experimental vessels:

1. Analysis of gas phase by gas chromatography to establish the anoxicity.

Table 1. Generalized Matrix for the Actinide Stability/Solubility Project

Actinide	Initial Oxidation State	Synthetic Brine	pmH
Uranium	IV	G-Seep	5,7
		ERDA-6	8,10
	V	G-Seep	5,7
		ERDA-6	8,10
	VI	G-Seep	5,7
		ERDA-6	8,10
Neptunium	IV	G-Seep	5,7
		ERDA-6	8,10
	V	G-Seep	5,7
		ERDA-6	8,10
	VI	G-Seep	5,7
		ERDA-6	8,10
Plutonium	IV	G-Seep	5,7
		ERDA-6	8,10
	V	G-Seep	5,7
		ERDA-6	8,10
	VI	G-Seep	5,7
		ERDA-6	8,10

2. Total actinide concentration. Determined by alpha scintillation for plutonium and neptunium. Uranium concentrations will be determined by fluorescence spectrometry.

3. Oxidation-state analysis by spectrometric techniques. This will include a combination of absorption spectrometry, photoacoustic spectroscopy, and if needed, oxidation-state specific titration.

4. pmH measurements as described by Rai. (Rai et al., 1995)

5. Inorganic carbon analysis using a combination of Dohrmann TOC analyzer for the liquid phase and gas chromatography for carbon dioxide in the gas phase. (for experiments with carbonate)

6. Solids analysis by x-ray diffraction.

Task II - Solubility of Actinides in WIPP Synthetic Brines under Anoxic Conditions in the Presence of Carbonate

A substantial amount of carbon dioxide is expected to be generated by microbial activity in the WIPP disposal room. The presence of carbon dioxide will have a significant effect on the pH of the brines and actinide solubility. Task II will repeat the same set of experiments described in Task I with the addition of sodium carbonate to each vessel. During these experiments the gas phase will be monitored for carbon dioxide content and the liquid phase analyzed for total inorganic carbon. Task II experiments will run concurrently with Task I.

Task III - Effect of Organic Complexation of the Stability/Solubility of Actinides

Organic ligands, present in the waste or formed from degradation of cellulosic materials, may complex with actinides and therefore increase the total dissolved actinide concentration. An extensive program is currently underway at Florida State University to determine the effect of 5 organic ligands (acetate, oxalate , lactate, citrate, and

EDTA) on actinide solubility. The data is being used to determine Pitzer parameters for actinide solubility models.

Task III will evaluate the effects of organic complexation on total actinide solubility. In general, the matrix of experiments described in Tasks I and II will be repeated with the addition of individual organic ligands to the test vessels. The ligands that will be investigated are oxalate, citrate, and EDTA. Scoping studies at Florida State University indicate these organics have the greatest effect on actinide solubility. The type and number of experiments will depend on the results from Tasks I and II.

Task III is divided into two parts. Part 1 will determine which actinide-organic complexes are stable in the synthetic brines. These short-term experiments will be used to guide the development of long-term solubility experiments in part 2 of the program. In both types of experiments, actinide solubility will be determined by tracking the absorption spectrum of the actinide as a function of time.

Results and Comparison to the Actinide Dissolved Concentration Subprogram

This work is being done independently of the Actinide Dissolved Concentration Subprogram. No information on actinide solubility or oxidation state stability or distribution is planned on being incorporated into the ADCS model. The results from this work will be used to test and challenge the predictive ability of the model for actinide solubility in complex brines. The results of this work may be used as a contingency plan for the ADCS in the event that the +VI ADCS model is not sufficiently developed to provide input to the WIPP Compliance Application.

Project Schedule (assuming a 6/20/95 start data)

The following schedule has been designed for the stability/solubility program.

1)	Complete stability experiments for +VI actinides FY 1995 Experimental Work	7/19/95
2)	Initiate +VI solubility experiments	8/15/95
3)	Complete first draft of FY 1995 annual report	9/30/95
4)	Initiate short term stability organic interaction experiments	9/30/95
5)	Complete Task I and Task II stability/solubility experiments for +VI oxidation state	12/30/95
6)	Complete short term stability organic interaction experiments	2/28/96
7)	Initiate long term stability/solubility organic interaction experiments and +V actinide stability/solubility experiments	5/1/96
8)	Initiate +IV stability/solubility experiments	8/1/96
9)	Complete long term stability/solubility experiments for organic interactions and +V, and +IV actinides	9/96
10)	Complete first draft of FY 1996 annual report	9/30/96

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Distribution

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APPENDIX D: MEMORANDA DOCUMENTING THE INITIAL DATA BASE FOR THE ACTINIDE SOURCE TERM DISSOLVED CONCENTRATION MODEL

Novak, C.F. 1995. "Creation and Definition of the Database for FMT called "HMW_NP_AM.CHEMDAT," version date 94.01.17." memorandum to N.J. Dhooge. 18 January 1995.

Novak, C.F. 1995. "Correction to the Database for FMT called "HMW_NP_AM.CHEMDAT," to create version date 95.01.31." memorandum to N.J. Dhooge. 31 January 1995.

Moore, R.C. 1995. "Validation of data entries in computer data file HMW_NP_AM.CHEMDAT." memorandum to C.F. Novak, 20 March 1995.

Note: the handwritten corrected entries to the first memo "Creation and Definition..." are documented in the second memorandum "Correction to...."

Sandia National Laboratories

MANAGED BY MARTIN MARIETTA CORPORATION
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Craig F Novak

from: Craig F. Novak, Dept. 6119, MS 1320, 848-0619

subject: Creation and Definition of the Database for FMT called "HMW_NP_AM.CHEMDAT," version date 94.01.17.

A database called "HMW_NP_AM.CHEMDAT," version date 94.01.17, has been created for use with FMT Version 1.1. HMW stands for Harvie-Møller-Weare/Felmy-Weare (Harvie et al., 1984; Felmy and Weare, 1986), journal articles documenting the development of a thermodynamic model for evaporite systems using the Pitzer activity coefficient formalism (Pitzer, 1991). NP stands for neptunium(V), and AM stands for americium(III). This database contains the thermodynamic parameters necessary to model the chemical behavior of Np(V) and Am(III) in brines. Note that the extension of the Pitzer model to higher electrolyte types than in Harvie et al. (1984) and Felmy and Weare (1986) is necessary for Am(III); this is discussed in Pitzer and Silvester (1978).

The purpose of this memorandum is to document the HMW_NP_AM.CHEMDAT database. This includes the sources of numbers in the database, including any necessary mathematical transformations to arrive at the values in HMW_NP_AM.CHEMDAT.

A listing of the file HMW_NP_AM.CHEMDAT is included as Appendix A. A listing of the main output file from FMT v1.1, showing an echo printing of the information from HMW_NP_AM.CHEMDAT is included as Appendix B.

D1

Data Sources

The data for HMW_NP_AM.CHEMDAT can be grouped into three types: parameters for the brine (evaporite) electrolytes, parameters for Np(V) and Np(V) interactions with evaporite electrolytes, and parameters for Am(III) interactions with evaporite electrolytes. There is some overlap among these groups, but this categorization scheme will be used.

Brine Model

Parameters for brine electrolytes come primarily from Harvie et al. (1984) and Felmy and Weare (1986), called Harvie-Møller-Weare/Felmy-Weare, or HMW/FW or simply HMW for short. Harvie et al. (1984) presents a model for the Na-K-Mg-Ca-H-Cl-SO_4-OH-HCO_3-CO_3-CO_2-H_2O system, and Felmy and Weare (1986) extends this model to include boron species. Also included in this category are data from Pitzer (1991) for ion interactions with perchlorate, ClO_4^-. Perchlorate data are included in HMW_NP_AM.CHEMDATT not because perchlorate is important for the WIPP, but because much actinide chemistry was measured in the presence of perchlorate, and therefore parameters parameters are needed to interpret these data. Parameters from Harvie et al. (1984) and Felmy and Weare (1986) were taken directly with no modification, and are not reproduced here. Parameters from Pitzer (1991) were scaled as necessary from the forms in which they were reported to the forms needed for use by FMT. These parameters are documented in Table 1.

Np(V) Model

Parameters for Np(V) interactions in NaCl, $NaClO_4$, and Na_2CO_3 media are taken from Novak and Roberts (1994). This model for Np(V) chemistry in brines is limited to predominantly NaCl, $NaClO_4$, or Na_2CO_3 media, and has not been shown to apply to other media. However, it can provide a first estimate of the solubility behavior of neptunium(V) in predominantly sodium chloride groundwaters containing carbonate, such as brines from the Castile and Rustler Formations in the vicinity of the WIPP Site. Parameters for Np(V) were taken directly from Novak and Roberts (1994) with no modification, as given in Tables 2 and 3.

D2

Am(III) Model

Parameters for Am(III) interactions with chloride, sulfate, phosphate, and carbonate anions, including interactions with several groundwater cations, are taken from Felmy et al. (1990), Felmy et al. (1989), Rai et al. (1992a), Rai et al., (1994), Rai et al. (1992b), and Rao et al. (1994). Although these parameters were developed for Am(III), Pu(III), or Nd(III), we have used the oxidation state analogy for f-elements to apply these parameters to Am(III) as shown here. Although we call it the Am(III) model, this model should apply equally well to Pu(III) and Nd(III).

Felmy et al. (1990) provides the values of standard chemical potential for the americium species shown in Table 4, and the ion interaction parameters shown in Table 5. These parameters were developed from relatively dilute, i.e., less than 0.1 molal, solutions, so they do not necessarily apply to more concentrated solutions, as this discussion of Rao et al. (1994) below shows.

Rai et al. (1992b) use Nd(III) and Am(III) data from Felmy et al. (1990) and Rai et al. (1983) to develop the Nd(III) standard chemical potentials shown in Table 6. These values were converted to standard chemical potentials for the analogous Am(III) species for use within HMW_NP_AM.CHEMDAT as follows. One can write a general chemical reaction Nd^{3+} with a ligand Y^n, where n can be positive or negative, and the analogous reaction for Am^{3+}:

$$Nd^{3+} + yY^n \leftrightarrow NdY_y^{3+yn} \qquad \overset{o}{\mu}_{rxn1} = \overset{o}{\mu}_{NdY_y^{3+yn}} - \overset{o}{\mu}_{Nd^{3+}} - y\,\overset{o}{\mu}_{Yn} \tag{1}$$

$$Am^{3+} + yY^n \leftrightarrow AmY_y^{3+yn} \qquad \overset{o}{\mu}_{rxn2} = \overset{o}{\mu}_{AmY_y^{3+yn}} - \overset{o}{\mu}_{Am^{3+}} - y\,\overset{o}{\mu}_{Yn} \tag{2}$$

Because we are assuming identical chemical behavior for Am(III) and Nd(III), we must have $\overset{o}{\mu}_{rxn1} = \overset{o}{\mu}_{rxn2}$. Therefore, the standard chemical potential for the complex americium species is given by

$$\overset{o}{\mu}_{AmY_y^{3+yn}} = \overset{o}{\mu}_{NdY_y^{3+yn}} + \left[\overset{o}{\mu}_{Am^{3+}} - \overset{o}{\mu}_{Nd^{3+}}\right] \tag{3}$$

D3

Analogous expressions hold for Pu(III). The dimensionless standard chemical potentials for the second and third americium hydrolysis species, calculated using Equation 3, are given in Table 6.

Felmy et al. (1989) gives ion interaction parameters between Pu(III) and Cl^-, as well as the standard chemical potential for $Pu(OH)_3(s)$. The ion interaction parameters are given in Table 7; these are assumed to apply equally well to Am(III) by analogy. The standard chemical potential for $Pu(OH)_3(s)$ is converted to that for $Am(OH)_3(s)$ in Table 8.

Rao et al. (1994) parameterized ion interactions for Nd(III) in concentrated $NaHCO_3$ and Na_2CO_3 media, extending the work for Felmy et al. (1990) to the large carbonate concentrations that could occur in WIPP under disposal scenarios with large $CO_2(g)$ pressures. They identified an additional Nd(III) solid phase that forms under concentrated conditions, and had to modify one of the ion interaction parameters determined in Felmy et al. (1990). The parameters from Rao et al. (1994), converted to apply to Am(III) species, are given in Tables 9 and 10.

Interactions for Nd(III) or Am(III) with phosphate species are discussed in Rai et al. (1992a, 1992b) and Rai et al. (1994). These papers present the standard chemical potentials for H_3PO_4, $H_2PO_4^-$, HPO_4^{2-}, and PO_4^{3-}, as shown in Table 11. The reported standard chemical potentials for H_3PO_4 and $H_2PO_4^-$, taken from Pitzer and Silvester (1976) as the original source, are the same. However, there are slight differences in the standard chemical potentials given for HPO_4^{2-}, and PO_4^{3-}, even though these are all reported as having been taken from Wagman et al. (1982). The differences in these reported values are slight and will have no significant impact on calculations using these numbers. We have gone back to the original Wagman et al. (1982) reference to arrive at the selected values indicated in Table 11. Ion interaction parameters for phosphoric acid, taken from Pitzer and Silvester (1976), are given in Table 12.

The standard chemical potentials for three different Am(III)-phosphate or analogous Nd(III)-phosphate phases are reported in (1992a, 1992b) and Rai et al. (1994). These values, along with conversion from Nd(III) to Am(III) where necessary, are given in Table 13. The difference among the calculated values for

D4

the standard chemical potentials for the americium phosphate solid phase is less than 0.9 units; all values are effectively the same. However, because the value from Rai et al. (1992a) was determined for Am directly, not for Nd, this value was selected for use in HMW_NP_AM.CHEMDAT.

Table 14 contains the ion interaction parameters developed for Nd(III) or Am(III) in Rai et al. (1994).

Description of the Chemical Data Base File HMW_NP_AM.CHEMDAT

The data file HMW_NP_AM.CHEMDAT is read by program FMT V1.1 using standard FORTRAN free-format read conventions. All character data must be enclosed in single quotes. Data are separated by either a space or a comma. Blank lines are included for (human) readability only, and are ignored by FMT V1.1. The following description of file HMW_NP_AM.CHEMDAT uses the line numbers printed with the file in Appendix A. In the following description, the variable names used within FMT V1.1 are printed in all capitals. Input parameters that are described as unused are not supported in Version 1.1. The character strings FRSR89, FRF90, PS76, P91, RFF92, RFFR92, RFF94, RRFF94, NR94, HMW84, and FW86 indicate the source of the data; the key to citations is at the bottom of the file. These indicators are not read by FMT V1.1, but serve merely to help with human interpretation of the file.

Some of the standard chemical potentials for species, i.e., lines 39-49, 70-72, are described as "arbitrary" in HMW_NP_AM.CHEMDAT. There are several conventions used for this designation. Some species, i.e., lines 39, 40, 70, 72, are physical species but represent the only occurrence for that element within the data file. The standard chemical potentials for these species are given as –999.99. If chemical reactions involving these species are added to the data base in future, these arbitrary values must be replaced with quoted values from a data source. Other species, i.e., lines 45, 46, 49, are nonphysical species that are included to facilitate the running of certain types of problems. These are assigned the arbitrary value of 0 for standard chemical potential. A third type of species, lines 42, 43, 44, 47, 48, are species included for adjusting the hydrogen ion concentration when setting up an input file. These species, assigned the

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arbitrary value of 500. for standard chemical potential, are designed to completely dissociate within an equilibrium problem. The final category, line 77, containing the string DISABLED in the name, is for species that may be added in future, but which are not allowed to form in the current version of the data base (thus, they are "disabled" from forming). These species are assigned the arbitrary value 999.99 for standard chemical potential.

Line 1: Line 2:	DBASE1 DBASE2	Two 78 character lines for naming/describing the data base; echo printed in all output files.
Line 4:	TTLELEM, TTLSPEC, DUMMY, DUMMY2, DUMMY1, DUMMY3	number of elements (whole number); number of species (whole number); unused flag; unused flag; extra echo printing flag (will echo print extra information when specified as 'ECHO'); unused flag;
Line 5:	MAXIT, ACCURCY, MINABU	maximum number of iterations for each equilibrium calculation (whole number); convergence tolerance on equilibrium problem (positive, real); minimum element abundance, abundances below which elements are considered not to be there (positive, real)
Line 6:	DUMMY, TEMPERA(1), P(1), P(2)	character flag indicating the units for standard chemical potentials ('NONE' means dimensionless); temperature in degrees Kelvin (real, positive); unused pressure in atmospheres, unused pressure in atmospheres
Lines 8-10:	ELNAMES()	names of each element
Lines 11-12:	MWELEM()	molecular weights of each element
Lines 14-140:	NAMES(j), FORMULA(i,j), PHASE(j), MU0FORM(j)	name of j^{th} chemical species; stoichiometric number of each element in the species, phase of the species (1=aqueous, 2=solid, 3=gas, but gasses are unsupported); standard chemical potential of the species (in the units indicated by the flag in line 6). Entries in the formula vector usually are, but need not be, integers, and can be positive or negative. The species H_2O must always be the first species in the list. All aqueous species must be declared before any solid species is declared.
Line 142:	DUMMY2, ITEMP	character flag that, when set to 'DG_BYPASS' allows alternate values for the standard chemical potentials to be read from file "bypass" for the first ITEMP species.
Line 143:	DUMMY2	character flag that enables the use of the Pitzer activity coefficient model when equal to 'PITZACT'. Any other character string will disable the Pitzer activity coefficient model.
Line 144:	NCATION	number of cations for which Pitzer activity coefficient model is used (whole number) See note below Line 854, Line 855, and Line 856 for a discussion of the order of the cations, anions, and neutral species.

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Line 145:	NANION	number of anions for which Pitzer activity coefficient model is used (whole number)
Line 146:	NNEUTRL	number of neutral species for which Pitzer activity coefficient model is used (whole number)
Lines 149-420:	SE(i,j,k=1,5)	array for single electrolyte, cation-anion interaction parameters: SE(i,j,1)=1 when either the cation or the anion has a charge of 1, SE(i,j,1)=2 when both the cation and anion have a charge of 3, SE(i,j,1)=3 for all other cases. SE(i,j,2) holds the $\beta^{(0)}$ parameters, SE(i,j,3) holds the $\beta^{(1)}$ parameter, SE(i,j,3) holds the $\beta^{(2)}$ parameter, SE(i,j,4) reads the C^{ϕ} parameter, but this is later converted to C_{MX} (see Eq. A.4b, Harvie et al., 1984. When reading the input file, interaction parameters are read for the first cation for all anions, then the second cation for all anions, etc. The character strings at the end of the numbers indicate the cation-anion pair specified on each line, for ease of human interpretation only.
Lines 423-434:	ME()	cation-cation interactions
Lines 437-455:	ME()	anion-anion interactions
Lines 457-545:	PSI(i,j,k)	for $i>j$, i and j are cation indices, k is the anion index
Lines 547-754:	PSI(i,j,k)	for $i<j$, i and j are anion indices, k is the cation index
Lines 756-761:	NEUCAT(i,j)	neutral-cation binary ion interaction parameters, i=neutral species index, j=cation index
Lines 763-768:	NEUANI(i,j)	neutral-anion binary ion interaction parameters i=neutral species index, j=anion index
Lines 770-852:	PTSTSI(i,j,k)	neutral-cation-anion ternary ion interaction parameters, i=neutral species index, j=cation index, k=anion index
Line 854:	ELMAP(1,i)	cation map
Line 855	ELMAP(2,i)	anion map
Line 856	ELMAP(3,i)	neutral species map
		Note on species order: cationic, anionic, and neutral species are referenced by the order given in ELMAP(,). This is determined as follows: the cations are the second, third, forth, fifth, sixth, seventh, twenty-second, etc., species entered in the species list, i.e., Na$^+$, K$^+$, Ca$^+$, Mg$^+$, MgOH$^+$, H$^+$, MgB(OH)$_4^+$, etc. Similar patterns are used for anions and cations. This allows additional flexibility in reorganizing and rearranging the data input files without having to reenter all of the ion interaction parameters. Although the order is arbitrary, it is very important that a consistent order is maintained throughout the CHEMDAT file. Because all parameters are clearly labeled, the echo printing of the ion interaction parameters makes is easy to determine whether errors were made when changing the CHEMDAT file.

Line 858	DUMMY, NEQACT	character flag that, when equal to 'EQUALACT', indicates that NEQACT (whole number) activity coefficients of "nonPitzer" species will be set equal to activity coefficients of Pitzer species. For example, one could run calculations using both ^{22}Na and ^{23}Na (which would have to be declared as separate elements in the element list), but instead of repeating all the ^{23}Na parameters for ^{22}Na, use the 'EQUALACT' flag. When DUMMY='EQUALACT' this line would be followed by NEQACT pairs of whole numbers, one pair per line, corresponding to species position in the species list. The activity coefficient of the first species in the pair will be set equal to the activity coefficient of the second species of the pair.
Line 860:	RPLWCHG	whole number indicating the element to RePLace with CHarGe. Used to implement the constraint of solution charge neutrality. The 2nd element corresponds to oxygen.
Line 861:	DUMMY, NEHRXN	character flag indicating whether redox reactions are specified (they are when the flag is 'REDOX'). When this flag is used, it will be followed by specifications of NEHRXN (whole number) redox reactions. This feature is unsupported in FMT V1.1
Line 862:	DUMMY, NSBSTPM, NSBSTRX, DUMMY1	Flags and parameters for specifying ion exchange reactions. The value 'IONEX' means that ion exchange reactions are specified on NSBSTPM permanent substrates and NSBSTRX reactive (soluble) substrates. The value for DUMMY1 indicates what type of mass-action model to use for ion exchange calculations. This feature is unsupported in FMT V1.1.

END OF COMPUTER-READ PORTION OF FILE

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Table 1: Ion interaction parameters from Pitzer (1991), converted to values needed for FMT.

	$\beta^{(0)}$	$\beta^{(1)}$	C^{ϕ}	$\beta^{(0)}$	$\beta^{(1)}$	C^{ϕ}
1-1 electrolytes						
Na^+-ClO_4^-				0.0554	0.2755	−0.00118
Na^+-$H_2PO_4^-$				−0.0533	0.0396	0.00795
K^+-$H_2PO_4^-$				−0.0678	−0.1042	0
H^+-ClO_4^-				0.1747	0.2931	0.00819
2-1 electrolytes	$\frac{4}{3}\beta^{(0)}$	$\frac{4}{3}\beta^{(1)}$	$\frac{2^{5/2}}{3}C^{\phi}$	$\beta^{(0)}$	$\beta^{(1)}$	C^{ϕ}
Na^+-HPO_4^{2-}	−0.0777	1.954	0.0554	−0.0583	1.466	0.0294
K^+-HPO_4^{2-}	0.0330	1.699	0.0309	0.0248	1.274	0.0164
Ca^{2+}-ClO_4^-	0.6015	2.342	−0.00943	0.4511	1.756	−0.00500
Mg^{2+}-ClO_4^-	0.6615	2.678	0.01806	0.4961	2.008	0.009578
UO_2^{2+}-Cl^-	0.5698	2.192	−0.06951	0.4274	1.644	−0.03686
UO_2^{2+}-ClO_4^-	0.8151	2.859	0.04089	0.6113	2.144	0.02168
3-1 electrolytes	$\frac{3}{2}\beta^{(0)}$	$\frac{3}{2}\beta^{(1)}$	$\frac{3^{3/2}}{2}C^{\phi}$	$\beta^{(0)}$	$\beta^{(1)}$	C^{ϕ}
Na^+-PO_4^{3-}	0.2672	5.777	−0.1339	0.1781	3.851	−0.05154
K^+-PO_4^{3-}	0.5594	5.958	−0.2255	0.3729	3.972	−0.08680
2-2 electrolytes				$\beta^{(0)}$	$\beta^{(1)}$	C^{ϕ}
UO_2^{2+}-SO_4^{2-}				0.322	1.827	−0.0176

Table 2. Standard chemical potentials for neptunyl(V) species, from Novak and Roberts (1994).

Species	μ_i^o/RT	Species	μ_i^o/RT
NpO_2^+	−369.127	$NaNpO_2CO_3(s)$	−713.707
$NpO_2OH(am)$	−452.642	$NpO_2CO_3^-$	−594.492
$NpO_2OH(aged)$	−454.010	$NpO_2(CO_3)_2^{3-}$	−808.403
$NpO_2OH(aq)$	−438.518	$NpO_2(CO_3)_3^{5-}$	−1019.918
$NpO_2(OH)_2^-$	−505.829		

Table 3. Ion interaction parameters for neptunyl(V) species, from Novak and Roberts (1994).

$\beta^{(0)}_{NpO_2^+\text{-}ClO_4^-}$	0.312	$\beta^{(0)}_{Na^+\text{-}NpO_2CO_3^-}$	0.161	β\s((0),Na$^+$-NpO$_2$(CO$_3$)$_3^{5-}$)	1.97
$\beta^{(0)}_{NpO_2^+\text{-}Cl^-}$	0.169	β\s((0),Na$^+$-NpO$_2$(CO$_3$)$_2^{3-}$)	0.407	β\s((1),Na$^+$-NpO$_2$(CO$_3$)$_3^{5-}$)	16

Table 4. Dimensionless standard chemical potentials for Am(III) species from Felmy et al. (1990)

Species	μ_i^o / RT
Am^{3+}	−241.694
$AmCO_3^+$	−472.06
$Am(CO_3)_2^-$	−695.88
$Am(CO_3)_3^{3-}$	−915.46
AmOHCO3(c)	−569.98

Table 5. Specific ion interaction parameters for Am(III) species from Felmy et al. (1990).

	$\beta^{(0)}$	$\beta^{(1)}$	$\beta^{(2)}$	C^ϕ
Na^+ - ClO_4^-	0.80	5.35	0	−0.0048
Na^+ - $Am(CO_3)_3^{3-}$	0.24*	8.1	0	0
* this value is modified in Table 10				

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Table 6. Dimensionless standard chemical potentials for Nd(III) species from Rai et al. (1992b), with calculated values for analogous Am(III) species.

Nd Species	μ_i^o / RT	Am Species	μ_i^o / RT
Nd^{3+}	−270.926	Am^{3+}	−241.694
$Nd(OH)_2^+$	−422.879	$Am(OH)_2^+$	−393.647*
$Nd(OH)_3^0$	−492.182	$Am(OH)_3^0$	−462.950*
$Nd(OH)_3(gl)$	−527.259	$Am(OH)_3(gl)$	−498.027*
*calculated by Eq. 3			

Table 7. Specific ion interaction parameters for Am(III) species from Felmy et al. (1989), and, by analogy, for Am(III).

	$\beta^{(0)}$	$\beta^{(1)}$	$\beta^{(2)}$	C^{ϕ}
Pu^{+++} - Cl^-	0.6117	5.403	0	−0.0284
Am^{+++} - Cl^-	0.6117	5.403	0	−0.0284

Table 8. Dimensionless standard chemical potentials for Pu(III) species from Felmy et al. (1989), with calculated values for analogous Am(III) species.

Pu Species	μ_i^o / RT	Am Species	μ_i^o / RT
Pu^{3+}	−233.4	Am^{3+}	−241.694
$Pu(OH)_3(s)$	~~−527.259~~	$Am(OH)_3(s)$	~~−535.553~~*
*calculated by Eq. 3			
	−484.0		−492.294

See 31JAN95 Correction memo

Table 9. Dimensionless standard chemical potentials for Nd(III) species from Rao et al. (1994), with calculated values for analogous Am(III) species.

Nd Species	μ_i^o / RT	Am Species	μ_i^o / RT
Nd^{3+}	−270.926	Am^{3+}	−241.694
$NaNd(CO_3)_2 \cdot 6H_2O(c)$	−1425.726	$NaAm(CO_3)_2 \cdot 6H_2O(c)$	−1396.494*
*calculated by Eq. 3			

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Table 10. Specific ion interaction parameters for Nd(III) species from Rao et al. (1994), and, by analogy, for Am(III).

	$\beta^{(0)}$	$\beta^{(1)}$	$\beta^{(2)}$	C^{ϕ}
Na^+ - $Nd(CO_3)_2^-$	0	−8.37	0	0
Na^+ - $Nd(CO_3)_3^{3-}$	−0.94*	8.1	0	0.418
Na^+ - $Am(CO_3)_2^-$	0	−8.37	0	0
Na^+ - $Am(CO_3)_3^{3-}$	−0.94*	8.1	0	0.418
*this value was changed from that given in Felmy et al. (1990)				

Table 11. Standard chemical potentials for phosphate species, with selected values for HMW_Np_Am.CHEMDAT database.

name	Rai et al. (1992a)	Rai et al. (1992b)	Rai et al. (1994)	selected value
$H_3PO_4(aq)$	−460.90	−460.90	−460.90	−460.90
$H_2PO_4^-$	−455.96	−455.960	−455.960	−455.960
HPO_4^{2-}	−439.404	−439.354	−439.354	−439.367
PO_4^{3-}	−410.98	−410.947	−410.947	−410.947

Table 12. Specific ion interaction parameters for $H_3PO_4(aq)$ from Pitzer and Silvester (1976).

	λ
H^+ - $H_3PO_4(aq)$	0.290
K^+ - $H_3PO_4(aq)$	−0.070
HPO_4^{2-} - $H_3PO_4(aq)$	−0.400

Table 13. Standard chemical potentials for Am(III)- or Nd(III)-phosphate solid phases.

Nd Solid Phase	μ_i^o / RT	Am Solid Phase	μ_i^o / RT
Nd^{3+}	−270.926	Am^{3+}	−241.694
		AmPO4•xH2O(am), Rai et al. (1992a)	−709.75
NdPO4(c), Rai et al. (1992b)	−738.166	AmPO4(c)	−708.934*
NdPO4(c), Rai et al. (1994)	−738.63	AmPO4(c)	−709.398*
*calculated by Eq. 3			

Table 14. Specific ion interaction parameters for Nd(III) species from Rai et al. (1994), and, by analogy, for Am(III).

	$\beta^{(0)}$	$\beta^{(1)}$	$\beta^{(2)}$	$C\phi$
Nd^{3+} - SO_4^{2-}	3.0398	0	−2500	0
Nd^{3+} - $H2PO_4^-$	0	0	−92.9	0
Am^{3+} - SO_4^{2-}	3.0398	0	−2500	0
Am^{3+} - $H2PO_4^-$	0	0	−92.9	0

Appendix A: Computer listing of the file HMW_NP_AM.CHEMDAT

```
'DATABASE:  HMW84/FW86; Np(V)-Na-CO3-OH-Cl-ClO4 (NR94);'
'~~94.01.17~~    Am(III)-Na-Cl-CO3-SO4-PO4 (FRSR89,FRF90,P91,RFFR92,RFF94,RRFF94)'
95.01.31
23 115 'nNEW T' 'nACTCOEF' 'nECHO' 'nABCD'
200 1.d-6 1.d-18
'NONE' 298.15 1.d0 1.d0

'Hydrogen' 'Oxygen' 'Sodium' 'Potassium' 'Magnesium' 'Calcium' 'Chlorine' 'Sulfur' 'Carbon' 'PosIon'
   'NegIon' 'Air' 'Boron' 'Bromine' 'TracerEl' 'Th(IV)' 'Am(III)' 'U(VI)' 'Np(V)' 'ClO4-(EL)'
   'Phosphorus' 'Electron' 'Charge'
1.0079 15.9994 22.98977 39.0983 24.305 40.08 35.453 32.06 12.011 0.0 0.0
   28.84 10.81 79.904 0.0 232.0381 243.0 238.029 237.0482 99.4506 30.974 0.0 0.0

'H2O                               WATER' 2 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0  0   1 -95.6635     HMW84
'Na+                                 Na+' 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0  1   1 -105.651     HMW84
'K+                                   K+' 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0  1   1 -113.957     HMW84
'Ca++                               Ca++' 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2   1 -223.30      HMW84
'Mg++                               Mg++' 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2   1 -183.468     HMW84
'MgOH+                             MgOH+' 1 1 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0  1   1 -251.94      HMW84
'H+                                   H+' 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0  1   1  0.          HMW84

'Cl-                                 Cl-' 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 -1   1 -52.955      HMW84
'SO4=                               SO4=' 0 4 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 -2   1 -300.386     HMW84
'HSO4-                             HSO4-' 1 4 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 -1   1 -304.942     HMW84
'OH-                                 OH-' 1 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 -1   1 -63.435      HMW84
'HCO3-                             HCO3-' 1 3 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 -1   1 -236.751     HMW84
'CO3=                               CO3=' 0 3 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 -2   1 -212.944     HMW84

'CO2(aq)                         CO2(aq)' 0 2 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0  0   1 -155.68      HMW84
'CaCO3(aq)                     CaCO3(aq)' 0 3 0 0 0 1 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0  0   1 -443.5       HMW84
'MgCO3(aq)                     MgCO3(aq)' 0 3 0 0 1 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0  0   1 -403.155     HMW84

'B(OH)3(aq)                   B(OH)3(aq)' 3 3 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0  0   1 -390.81      FW86
'B(OH)4-                         B(OH)4-' 4 4 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 -1   1 -465.20      FW86
'B3O3(OH)4-                   B3O3(OH)4-' 4 7 0 0 0 0 0 0 0 0 0 0 3 0 0 0 0 0 0 0 0 0 -1   1 -963.77      FW86
'B4O5(OH)4=                   B4O5(OH)4=' 4 9 0 0 0 0 0 0 0 0 0 0 4 0 0 0 0 0 0 0 0 0 -2   1 -1239.10     FW86
'CaB(OH)4+                     CaB(OH)4+' 4 4 0 0 0 1 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0  1   1 -692.30      FW86
'MgB(OH)4+                     MgB(OH)4+' 4 4 0 0 1 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0  1   1 -651.89      FW86
'Br-                                 Br-' 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 -1   1 -999.99      arbitrary
'ClO4-                 perchlorate ClO4-' 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 -1   1 -999.99      arbitrary

'NaOH(aq)......to.titrate.base.only' 1 1 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0  0   1  500.        arbitrary
'HCl(aq).......to.titrate.acid.only' 1 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0  0   1  500.        arbitrary
'HClO4(aq).....to.titrate.acid.only' 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0  0   1  500.        arbitrary
'PosIon................POSITIVE.ION' 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0  1   1  0.          arbitrary
'NegIon................NEGATIVE.ION' 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 -1   1  0.          arbitrary
'PosIon(OH)(aq).....to.titrate.base' 1 1 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0  0   1  500.        arbitrary
'HNegIon(aq)........to.titrate.acid' 1 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0  0   1  500.        arbitrary
'Tracer(aq).....conservative.tracer' 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0  0   1  0.          arbitrary

'H3PO4(aq)                     H3PO4(aq)' 3 4 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0  0   1 -460.90      RFF92
'H2PO4-                           H2PO4-' 2 4 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 -1   1 -455.96      RFF92
'HPO4=                             HPO4=' 1 4 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 -2   1 -439.367     RFF94
'PO4=-                             PO4=-' 0 4 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 -3   1 -410.947     RFF94

'NpO2+                             NpO2+' 0 2 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 0  1   1 -369.127     N&R94
'NpO2OH(aq)                   NpO2OH(aq)' 1 3 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 0  0   1 -438.518     N&R94
'NpO2(OH)2-                   NpO2(OH)2-' 2 4 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 0 -1   1 -505.829     N&R94
'NpO2CO3-                       NpO2CO3-' 0 5 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 1 0 0 0 -1   1 -594.492     N&R94
'NpO2(CO3)2=-               NpO2(CO3)2=-' 0 8 0 0 0 0 0 0 2 0 0 0 0 0 0 0 0 0 1 0 0 0 -3   1 -808.403     N&R94
'NpO2(CO3)3==-             NpO2(CO3)3==-' 0 11 0 0 0 0 0 0 3 0 0 0 0 0 0 0 0 0 1 0 0 0 -5  1 -1019.918    N&R94

'Am+++                             Am+++' 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0  3   1 -241.694     FRF90
'AmCO3+                           AmCO3+' 0 3 0 0 0 0 0 0 1 0 0 0 0 0 0 0 1 0 0 0 0 0  1   1 -472.060     FRF90
'Am(CO3)2-                     Am(CO3)2-' 0 6 0 0 0 0 0 0 2 0 0 0 0 0 0 0 1 0 0 0 0 0 -1   1 -695.880     FRF90
'Am(CO3)3=-                   Am(CO3)3=-' 0 9 0 0 0 0 0 0 3 0 0 0 0 0 0 0 1 0 0 0 0 0 -3   1 -915.460     FRF90
'Am(OH)2+                       Am(OH)2+' 2 2 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0  1   1 -393.647     RFFR92
'Am(OH)3(aq)                 Am(OH)3(aq)' 3 3 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0  0   1 -462.950     RFFR92

'Th++++                           Th++++' 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0  4   1 -999.99      arbitrary

'UO2++                        U(VI)O2++' 0 2 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0  2   1 -999.99      arbitrary

'NpO2OH(aged)___________NpO2OH(aged)' 1 3 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 0  0   2 -454.010     N&R94
'NpO2OH(amor)___________NpO2OH(amor)' 1 3 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 0  0   2 -452.642     N&R94
'NaNpO2CO3(s)___________NaNpO2CO3(s)' 0 5 1 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 1 0 0 0  0   2 -713.707     N&R94
'Na3NpO2(CO3)2(s)_DISABLED_DISABLED' 0 8 3 0 0 0 0 0 2 0 0 0 0 0 0 0 0 0 1 0 0 0  0   2  999.99      arbitrary

'AmOHCO3(c)_______________AmOHCO3(c)' 1 4 0 0 0 0 0 0 1 0 0 0 0 0 0 0 1 0 0 0 0 0  0   2 -569.980     FRF90
'Am(OH)3(s)_______________Am(OH)3(s)' 3 3 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0  0   2 ~~-535.553~~     FRSR89   -492.294
'NaAm(CO3)2.6H2O(c)_______________' 12 12 1 0 0 0 0 0 2 0 0 0 0 0 0 0 1 0 0 0 0 0  0   2 -1396.494    RRFF94
'AmPO4(c)___________________AmPO4(c)' 0 4 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 0 1 0  0   2 -709.750     RFF94

'CaSO4____________________Anhydrite'  0 4 0 0 0 1 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0   2 -533.73      HMW84
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File: 95.01.12 FMT DB HMW_NP_AM AppxA

Memo from C.F. Novak
printed June 7, 1995, p. 1

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'NaK3(SO4)2__Aphthitalite/Glaserite'  0  8 1 3 0 0 0 2 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -1057.05     HMW84
'CaCl2.6H2O_____________Antarcticite' 12  6 0 0 0 1 2 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -893.65      HMW84
'CaCO3________________________Aragonite'  0  3 0 0 0 1 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -455.17      HMW84
'K2SO4________________________Arcanite'  0  4 0 2 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -532.39      HMW84
'MgCl2.6H2O_______________Bischofite' 12  6 0 0 1 0 2 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -853.1       HMW84
'Na2Mg(SO4)2.4H2O___________Bloedite'  8 12 2 0 1 0 0 2 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -1383.6      HMW84
'Mg(OH)2__________________________Brucite'  2  2 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -335.4       HMW84
'Na6CO3(SO4)2_________________Burkeite'  0 11 6 0 0 0 0 2 1 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -1449.4      HMW84
'CaCO3__________________________Calcite'  0  3 0 0 0 1 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -455.6       HMW84
'CaCl2.4H2O______CaCl2_Tetrahydrite'  8  4 0 0 0 1 2 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -698.7       HMW84
'Ca4Cl2(OH)6.13H2O__CaOxychloride A' 32 19 0 0 0 4 2 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -2658.45     HMW84
'Ca2Cl2(OH)2.H2O____CaOxychloride B'  4  3 0 0 0 2 2 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -778.41      HMW84
'KMgCl3.6H2O________________Carnallite' 12  6 0 1 1 0 3 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -1020.3      HMW84
'MgSO4.7H2O___________________Epsomite' 14 11 0 0 1 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -1157.83     HMW84
'CaNa2(CO3)2.5H2O_________Gaylussite' 10 11 2 0 0 1 0 0 2 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -1360.5      HMW84
'Na2Ca(SO4)2________________Glauberite'  0  8 2 0 0 1 0 2 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -1047.45     HMW84
'CaSO4.2H2O_______________________Gypsum'  4  6 0 0 0 1 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -725.56      HMW84
'NaCl_____________________________Halite'  0  0 1 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -154.99      HMW84
'MgSO4.6H2O_______________Hexahydrite' 12 10 0 0 1 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -1061.60     HMW84
'KMgClSO4.3H2O__________________Kainite'  6  7 0 1 1 0 1 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -938.2       HMW84
'KHCO3______________________Kalicinite'  1  3 0 1 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -350.06      HMW84
'MgSO4.H2O_________________ Kieserite'  2  5 0 0 1 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -579.80      HMW84
'K2Mg(SO4)2.4H2O_________________Leonite'  8 12 0 2 1 0 0 2 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -1403.97     HMW84
'Na4Ca(SO4)3.2H2O_________Labile_Salt'  4 14 4 0 0 1 0 3 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -1751.45     HMW84
'MgCO3_______________________Magnesite'  0  3 0 0 1 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -414.45      HMW84
'Mg2Cl(OH)3.4H2O______MgOxychloride' 11  7 0 0 2 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -1029.6      HMW84
'KHSO4______________________Mercallite'  1  4 0 1 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -417.57      HMW84
'Na2SO4.10H2O_______________Mirabilite' 20 14 2 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -1471.15     HMW84
'K8H6(SO4)7______________________Misenite'  6 28 0 8 0 0 0 7 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -3039.24     HMW84
'NaHCO3_______________________Nahcolite'  1  3 1 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -343.33      HMW84
'Na2CO3.10H2O_____________________Natron' 20 13 2 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -1382.78     HMW84
'MgCO3.3H2O_______________Nesquehonite'  6  6 0 0 1 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -695.3       HMW84
'K2Mg(SO4)2.6H2O Picromerite/Schoen' 12 14 0 2 1 0 0 2 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -1596.1      HMW84
'Na2Ca(CO3)2.2H2O_________Pirssonite'  4  8 2 0 0 1 0 0 2 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -1073.1      HMW84
'K2MgCa2(SO4)4.2H2O_______Polyhalite'  4 18 0 2 1 2 0 4 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -2282.5      HMW84
'Ca(OH)2_____________________Portlandite'  2  2 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -362.12      HMW84
'K2CO3.3/2H2O___Potassium_Carbonate' 3 4.5 0 2 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -577.37      HMW84
'K8H4(CO3)6.3H2O___K-Sequicarbonate' 10 21 0 8 0 0 0 0 6 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -2555.4      HMW84
'KNaCO3.6H2O___________K-Na-Carbonate' 12  9 1 1 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -1006.8      HMW84
'K2NaH(CO3)2.2H2O___Potassium_Trona'  5  8 1 2 0 0 0 0 2 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -971.74      HMW84
'K3H(SO4)2__Sesquipotassium_Sulfate'  1  8 0 3 0 0 0 2 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -950.8       HMW84
'Na3H(SO4)2_____Sesquisodium_Sulfate'  1  8 3 0 0 0 0 2 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -919.6       HMW84
'Na2CO3.7H2O_____Na2CO3-Heptahydrate' 14 10 2 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -1094.95     HMW84
'KCl______________________________Sylvite'  0  0 0 1 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -164.84      HMW84
'K2Ca(SO4)2.H2O_______________Syngenite'  2  9 0 2 0 1 0 2 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -1164.8      HMW84
'Mg2CaCl6.12H2O__________Tachyhydrite' 24 12 0 0 2 1 6 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -2015.9      HMW84
'Na2SO4_______________________Thenardite'  0  4 2 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -512.35      HMW84
'Na2CO3.H2O_______________Thermonatrite'  2  4 2 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -518.8       HMW84
'Na3H(CO3)2.2H2O_________________Trona'  5  8 3 0 0 0 0 0 2 0 0 0 0 0 0 0 0 0 0 0 0 0  2 -960.38      HMW84
'Na2B4O7.10H2O___________________ Borax' 20 17 2 0 0 0 0 0 0 0 0 0 4 0 0 0 0 0 0 0 0 0  2 -2224.16     FW86
'B(OH)3________________Borix_Acid_Solid'  3  3 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0  2 -390.88      FW86
'KB5O8.4H2O_____K-Pentaborate_(30_C)'  8 12 0 1 0 0 0 0 0 0 0 0 5 0 0 0 0 0 0 0 0 0  2 -1770.26     FW86
'K2B4O7.4H2O____K-Tetraborate_(30_C)'  8 11 0 2 0 0 0 0 0 0 0 0 4 0 0 0 0 0 0 0 0 0  2 -1663.47     FW86
'NaBO2.4H2O_________Sodium_Metaborate'  8  6 1 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0  2 -761.42      FW86
'NaB5O8.5H2O______Sodium_Pentaborate' 10 13 1 0 0 0 0 0 0 0 0 0 5 0 0 0 0 0 0 0 0 0  2 -1854.80     FW86
'NaBO2.NaCl.2H2O___Teepleite_(20_C)'  4  4 2 0 0 0 1 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0  2 -725.77      FW86

'nDG_BYPASS' 0
'PITZACT'
 13 #Cat  Na,K,Ca,Mg,MgOH,H,MgB(OH)4+,CaB(OH)4+,Am+++,AmCO3+,Th++++,UO2++,NpO2+
 20 #Ani  Cl,SO4,HSO4,OH,HCO3,CO3,B(OH)4,B3O3(OH)4,B4O5(OH)4,Br,Am(CO3)2-,Am(CO3)3=-,ClO4-,NpO2(OH)2-
,NpC,NpC2,NpC3,H2PO4,HPO4,PO4
  6 #Neu  CO2,CaCO3,MgCO3,B(OH)3,H3PO4

  1  .0765     .2644    .0     .00127   Na+ Cl-            HMW84 (beta0, beta1, beta2, cphi)
  1  .01958   1.113     .0     .00497   Na+ SO4=           HMW84
  1  .0454     .398     .0     .0       Na+ HSO4-          HMW84
  1  .0864     .253     .0     .0044    Na+ OH-            HMW84
  1  .0277     .0411    .0     .0       Na+ HCO3-          HMW84
  1  .0399    1.389     .0     .0044    Ma+ CO3=           HM284
  1 -.0427     .089     .0     .0114    Na+ B(OH)4-        FW86
  1 -.056     -.910     .0     .0       Na+ B3O3(OH)4-     FW86
  1 -.11      -.40      .0     .0       Na+ B4O5(OH)4=     FW86
  1  .0        .0       .0     .0       Na+ Br-
  1  .0      -8.37      .0     .0       Na+ Am(CO3)2-      RRFF94
  1 -.94      8.1       .0     .418     Na+ Am(CO3)3=-     RRFF94,FRF90
  1  .0554     .2755    .0    -.00118   Na+ ClO4-          P91
  1  .0        .0       .0     .0       Na+ NpO2(OH)2-     NR94
  1  .161      .0       .0     .0       Na+ NpO2CO3-       NR94
  1  .407      .0       .0     .0       Na+ NpO2(CO3)2=-   NR94
  1  1.97     16.       .0     .0       Na+ NpO2(CO3)3==-  NR94
  1 -.0533     .0396    .0     .00795   Na+ H2PO4-         P91
  1 -.0583    1.466     .0     .0294    Na+ HPO4=          P91
  1  .1781    3.851     .0    -.05154   Na+ PO4=-          P91

  1  .04835    .2122    .0    -.00084   K+ Cl-             HMW84
  1  .04995    .7793    .0     .0       K+ SO4=            HMW84
  1 -.0003     .1735    .0     .0       K+ HSO4-           HMW84
  1  .1298     .320     .0     .0041    K+ OH-             HMW84
  1  .0296    -.013     .0    -.008     K+ HCO3-           HMW84
  1  .1488    1.43      .0    -.0015    K+ CO3=            HMW84
  1  .035      .14      .0     .0       K+ B(OH)4-         FW86
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177      1 -.13      .0       .0      .0         K+ B3O3(OH)4-      FW86
178      1 -.022     .0       .0      .0         K+ B4O5(OH)4=      FW86
179      1  .0       .0       .0      .0         K+ Br-
180      1  .0       .0       .0      .0         K+ Am(CO3)2-
181      1  .0       .0       .0      .0         K+ Am(CO3)3=-
182      1  .0       .0       .0      .0         K+ ClO4-
183      1  .0       .0       .0      .0         K+ NpO2(OH)2-
184      1  .0       .0       .0      .0         K+ NpO2CO3-
185      1  .0       .0       .0      .0         K+ NpO2(CO3)2=-
186      1  .0       .0       .0      .0         K+ NpO2(CO3)3==-
187      1 -.0678   -.1042    .0      .0         K+ H2PO4-          P91
188      1  .0248   1.274     .0      .0164      K+ HPO4=           P91
189      1  .3729   3.972     .0     -.08680     K+ PO4=-           P91
190
191      1  .3159   1.614     .0     -.00034     Ca++ Cl-           HMW84
192      2  .20     3.1973  -54.24    .0         Ca++ SO4=          HMW84
193      1  .2145   2.53      .0      .0         Ca++ HSO4-         HMW84
194      1 -.1747   -.2303   -5.72    .0         Ca++ OH-           HMW84
195      1  .4      2.977     .0      .0         Ca++ HCO3-         HMW84
196      2  .0      .0        .0      .0         Ca++ CO3=          HMW84
197      1  .0      .0        .0      .0         Ca++ B(OH)4-       FW86
198      1  .0      .0        .0      .0         Ca++ B3O3(OH)4-    FW86
199      1  .0      .0        .0      .0         Ca++ B4O5(OH)4=    FW86
200      1  .0      .0        .0      .0         Ca++ Br-
201      1  .0      .0        .0      .0         Ca++ Am(CO3)2-
202      3  .0      .0        .0      .0         Ca++ Am(CO3)3=-
203      1  .4511   1.756     .0     -.00500     Ca++ ClO4-         P91
204      1  .0      .0        .0      .0         Ca++ NpO2(OH)2-
205      1  .0      .0        .0      .0         Ca++ NpO2CO3-
206      3  .0      .0        .0      .0         Ca++ NpO2(CO3)2=-
207      3  .0      .0        .0      .0         Ca++ NpO2(CO3)3==-
208      1  .0      .0        .0      .0         Ca++ H2PO4-
209      2  .0      .0        .0      .0         Ca++ HPO4=
210      3  .0      .0        .0      .0         Ca++ PO4=-
211
212      1  .35235  1.6815    .0      .00519     Mg++ Cl-           HMW84
213      2  .2210   3.343   -37.23    .025       Mg++ SO4=          HMW84
214      1  .4746   1.729     .0      .0         Mg++ HSO4-         HMW84
215      1  .0       .0       .0      .0         Mg++ OH-           HMW84
216      1  .329     .6072    .0      .0         Mg++ HCO3-         HMW84
217      2  .0       .0       .0      .0         Mg++ CO3=          HMW84
218      1  .0       .0       .0      .0         Mg++ B(OH)4-       FW86
219      1  .0       .0       .0      .0         Mg++ B3O3(OH)4-    FW86
220      1  .0       .0       .0      .0         Mg++ B4O5(OH)4=    FW86
221      1  .0       .0       .0      .0         Mg++ Br-
222      1  .0      .0        .0      .0         Mg++ Am(CO3)2-
223      3  .0      .0        .0      .0         Mg++ Am(CO3)3=-
224      1  .4961   2.008     .0      .009578    Mg++ ClO4-         P91
225      1  .0      .0        .0      .0         Mg++ NpO2(OH)2-
226      1  .0      .0        .0      .0         Mg++ NpO2CO3-
227      3  .0      .0        .0      .0         Mg++ NpO2(CO3)2=-
228      3  .0      .0        .0      .0         Mg++ NpO2(CO3)3==-
229      1  .0      .0        .0      .0         Mg++ H2PO4-
230      2  .0      .0        .0      .0         Mg++ HPO4=
231      3  .C      .0        .0      .0         Mg++ PO4=-
232
233      1 -.10     1.658     .0      .0         MgOH+ Cl-          HMW84
234      1  .0       .0       .0      .0         MgOH+ SO4=         HMW84
235      1  .0       .0       .0      .0         MgOH+ HSO4-        HMW84
236      1  .0       .0       .0      .0         MgOH+ OH-          HMW84
237      1  .0       .0       .0      .0         MgOH+ HCO3-        HMW84
238      1  .0       .0       .0      .0         MgOH+ CO3=         HMW84
239      1  .0       .0       .0      .0         MgOH+ B(OH)4-
240      1  .0       .0       .0      .0         MgOH+ B3O3(OH)4-
241      1  .0       .0       .0      .0         MgOH+ B4O5(OH)4=
242      1  .0       .0       .0      .0         MgOH+ Br-
243      1  .0       .0       .0      .0         MgOH+ Am(CO3)2-
244      1  .0       .0       .0      .0         MgOH+ Am(CO3)3=-
245      1  .0       .0       .0      .0         MgOH+ ClO4-
246      1  .0       .0       .0      .0         MgOH+ NpO2(OH)2-
247      1  .0       .0       .0      .0         MgOH+ NpO2CO3-
248      1  .0       .0       .0      .0         MgOH+ NpO2(CO3)2=-
249      1  .0       .0       .0      .0         MgOH+ NpO2(CO3)3==-
250      1  .0       .0       .0      .0         MgOH+ H2PO4-
251      1  .0       .0       .0      .0         MgOH+ HPO4=
252      1  .0       .0       .0      .0         MgOH+ PO4=-
253
254      1  .1775    .2945    .0      .0008      H+ Cl-             HMW84
255      1  .0298    .0       .0      .0438      H+ SO4=            HMW84
256      1  .2065    .5556    .0      .0         H+ HSO4-           HMW84
257      1  .0       .0       .0      .0         H+ OH-             HMW84
258      1  .0       .0       .0      .0         H+ HCO3-           HMW84
259      1  .0       .0       .0      .0         H+ CO3=            HMW84
260      1  .0       .0       .0      .0         H+ B(OH)4-         FW86
261      1  .0       .0       .0      .0         H+ B3O3(OH)4-      FW86
262      1  .0       .0       .0      .0         H+ B4O5(OH)4=      FW86
263      1  .0       .0       .0      .0         H+ Br-
264      1  .0       .0       .0      .0         H+ Am(CO3)2-
265      1  .0       .0       .0      .0         H+ Am(CO3)3=-
266      1  .1747    .2931    .0      .00819     H+ ClO4-           P91
267      1  .0       .0       .0      .0         H+ NpO2(OH)2-
268      1  .0       .0       .0      .0         H+ NpO2CO3-
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269      1  .0       .0      .0     .0     H+ NpO2(CO3)2=-
270      1  .0       .0      .0     .0     H+ NpO2(CO3)3==-
271      1  .0       .0      .0     .0     H+ H2PO4-
272      1  .0       .0      .0     .0     H+ HPO4=
273      1  .0       .0      .0     .0     H+ PO4=-
274
275      1  .16      .0      .0     .0     MgB(OH)4+ Cl-       HMW84
276      1  .0       .0      .0     .0     MgB(OH)4+ SO4=      HMW84
277      1  .0       .0      .0     .0     MgB(OH)4+ HSO4-     HMW84
278      1  .0       .0      .0     .0     MgB(OH)4+ OH-       HMW84
279      1  .0       .0      .0     .0     MgB(OH)4+ HCO3-     HMW84
280      1  .0       .0      .0     .0     MgB(OH)4+ CO3=      HMW84
281      1  .0       .0      .0     .0     MgB(OH)4+ B(OH)4-
282      1  .0       .0      .0     .0     MgB(OH)4+ B3O3(OH)4-
283      1  .0       .0      .0     .0     MgB(OH)4+ B4O5(OH)4=
284      1  .0       .0      .0     .0     MgB(OH)4+ Br-
285      1  .0       .0      .0     .0     MgB(OH)4+ Am(CO3)2-
286      1  .0       .0      .0     .0     MgB(OH)4+ Am(CO3)3=-
287      1  .0       .0      .0     .0     MgB(OH)4+ ClO4-
288      1  .0       .0      .0     .0     MgB(OH)4+ NpO2(OH)2-
289      1  .0       .0      .0     .0     MgB(OH)4+ NpO2CO3-
290      1  .0       .0      .0     .0     MgB(OH)4+ NpO2(CO3)2=-
291      1  .0       .0      .0     .0     MgB(OH)4+ NpO2(CO3)3==-
292      1  .0       .0      .0     .0     MgB(OH)4+ H2PO4-
293      1  .0       .0      .0     .0     MgB(OH)4+ HPO4=
294      1  .0       .0      .0     .0     MgB(OH)4+ PO4=-
295
296      1  .12      .0      .0     .0     CaB(OH)4+ Cl-
297      1  .0       .0      .0     .0     CaB(OH)4+ SO4=      HMW84
298      1  .0       .0      .0     .0     CaB(OH)4+ HSO4-     HMW84
299      1  .0       .0      .0     .0     CaB(OH)4+ OH-       HMW84
300      1  .0       .0      .0     .0     CaB(OH)4+ HCO3-     HMW84
301      1  .0       .0      .0     .0     CaB(OH)4+ CO3=      HMW84
302      1  .0       .0      .0     .0     CaB(OH)4+ B(OH)4-
303      1  .0       .0      .0     .0     CaB(OH)4+ B3O3(OH)4-
304      1  .0       .0      .0     .0     CaB(OH)4+ B4O5(OH)4=
305      1  .0       .0      .0     .0     CaB(OH)4+ Br-
306      1  .0       .0      .0     .0     CaB(OH)4+ Am(CO3)2-
307      1  .0       .0      .0     .0     CaB(OH)4+ Am(CO3)3=-
308      1  .0       .0      .0     .0     CaB(OH)4+ ClO4-
309      1  .0       .0      .0     .0     CaB(OH)4+ NpO2(OH)2-
310      1  .0       .0      .0     .0     CaB(OH)4+ NpO2CO3-
311      1  .0       .0      .0     .0     CaB(OH)4+ NpO2(CO3)2=-
312      1  .0       .0      .0     .0     CaB(OH)4+ NpO2(CO3)3==-
313      1  .0       .0      .0     .0     CaB(OH)4+ H2PO4-
314      1  .0       .0      .0     .0     CaB(OH)4+ HPO4=
315      1  .0       .0      .0     .0     CaB(OH)4+ PO4=-
316
317      1  .6117   5.403    .0   -0.0284  Am+++ Cl-              FRSR89
318      3 3.0398    .0   -2500     .0     Am+++ SO4=             RFF94
319      1  .0       .0      .0     .0     Am+++ HSO4-
320      1  .0       .0      .0     .0     Am+++ OH-
321      1  .0       .0      .0     .0     Am+++ HCO3-
322      3  .0       .0      .0     .0     Am+++ CO3=
323      1  .0       .0      .0     .0     Am+++ B(OH)4-
324      1  .0       .0      .0     .0     Am+++ B3O3(OH)4-
325      1  .0       .0      .0     .0     Am+++ B4O5(OH)4=
326      1  .0       .0      .0     .0     Am+++ Br-
327      1  .0       .0      .0     .0     Am+++ Am(CO3)2-
328      3  .0       .0      .0     .0     Am+++ Am(CO3)3=-
329      1  .80     5.35     .0   -0.0048  Am+++ ClO4-             FRF90
330      1  .0       .0      .0     .0     Am+++ NpO2(OH)2-
331      1  .0       .0      .0     .0     Am+++ NpO2CO3-
332      3  .0       .0      .0     .0     Am+++ NpO2(CO3)2=-
333      3  .0       .0      .0     .0     Am+++ NpO2(CO3)3==-
334      1  .0       .0    -92.9    .0     Am+++ H2PO4-            RFF94
335      3  .0       .0      .0     .0     Am+++ HPO4=
336      3  .0       .0      .0     .0     Am+++ PO4=-
337
338      1  .0       .0      .0     .0     AmCO3+ Cl-
339      1  .0       .0      .0     .0     AmCO3+ SO4=
340      1  .0       .0      .0     .0     AmCO3+ HSO4-
341      1  .0       .0      .0     .0     AmCO3+ OH-
342      1  .0       .0      .0     .0     AmCO3+ HCO3-
343      1  .0       .0      .0     .0     AmCO3+ CO3=
344      1  .0       .0      .0     .0     AmCO3+ B(OH)4-
345      1  .0       .0      .0     .0     AmCO3+ B3O3(OH)4-
346      1  .0       .0      .0     .0     AmCO3+ B4O5(OH)4=
347      1  .0       .0      .0     .0     AmCO3+ Br-
348      1  .0       .0      .0     .0     AmCO3+ Am(CO3)2-
349      1  .0       .0      .0     .0     AmCO3+ Am(CO3)3=-
350      1  .0       .0      .0     .0     AmCO3+ ClO4-
351      1  .0       .0      .0     .0     AmCO3+ NpO2(OH)2-
352      1  .0       .0      .0     .0     AmCO3+ NpO2CO3-
353      1  .0       .0      .0     .0     AmCO3+ NpO2(CO3)2=-
354      1  .0       .0      .0     .0     AmCO3+ NpO2(CO3)3==-
355      1  .0       .0      .0     .0     AmCO3+ H2PO4-
356      1  .0       .0      .0     .0     AmCO3+ HPO4=
357      1  .0       .0      .0     .0     AmCO3+ PO4=-
358
359      1  .0       .0      .0     .0     Th++++ Cl-
360      3  .0       .0      .0     .0     Th++++ SO4=
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361      1  .0        .0       .0      .0        Th++++ HSO4-
362      1  .0        .0       .0      .0        Th++++ OH-
363      1  .0        .0       .0      .0        Th++++ HCO3-
364      3  .0        .0       .0      .0        Th++++ CO3=
365      1  .0        .0       .0      .0        Th++++ B(OH)4-
366      1  .0        .0       .0      .0        Th++++ B3O3(OH)4-
367      1  .0        .0       .0      .0        Th++++ B4O5(OH)4=
368      1  .0        .0       .0      .0        Th++++ Br-
369      1  .0        .0       .0      .0        Th++++ Am(CO3)2-
370      3  .0        .0       .0      .0        Th++++ Am(CO3)3=-
371      1  .0        .0       .0      .0        Th++++ ClO4-
372      1  .0        .0       .0      .0        Th++++ NpO2(OH)2-
373      1  .0        .0       .0      .0        Th++++ NpO2CO3-
374      3  .0        .0       .0      .0        Th++++ NpO2(CO3)2=-
375      3  .0        .0       .0      .0        Th++++ NpO2(CO3)3==-
376      1  .0        .0       .0      .0        Th++++ H2PO4-
377      3  .0        .0       .0      .0        Th++++ HPO4=
378      3  .0        .0       .0      .0        Th++++ PO4=-
379
380      1  .4274     1.644    .0      -.03686   UO2++ Cl-            P91
381      2  .322      1.827    .0      -.0176    UO2++ SO4=           P91
382      1  .0        .0       .0      .0        UO2++ HSO4-
383      1  .0        .0       .0      .0        UO2++ OH-
384      1  .0        .0       .0      .0        UO2++ HCO3-
385      2  .0        .0       .0      .0        UO2++ CO3=
386      1  .0        .0       .0      .0        UO2++ B(OH)4-
387      1  .0        .0       .0      .0        UO2++ B3O3(OH)4-
388      1  .0        .0       .0      .0        UO2++ B4O5(OH)4=
389      1  .0        .0       .0      .0        UO2++ Br-
390      1  .0        .0       .0      .0        UO2++ Am(CO3)2-
391      3  .0        .0       .0      .0        UO2++ Am(CO3)3=-
392      1  .6113     2.144    .0      .02168    UO2++ ClO4-          P91
393      1  .0        .0       .0      .0        UO2++ NpO2(OH)2-
394      1  .0        .0       .0      .0        UO2++ NpO2CO3-
395      3  .0        .0       .0      .0        UO2++ NpO2(CO3)2=-
396      3  .0        .0       .0      .0        UO2++ NpO2(CO3)3==-
397      1  .0        .0       .0      .0        UO2++ H2PO4-
398      2  .0        .0       .0      .0        UO2++ HPO4=
399      3  .0        .0       .0      .0        UO2++ PO4=-
400
401      1  0.169     .0       .0      .0        NpO2+ Cl-             NR94
402      1  .0        .0       .0      .0        NpO2+ SO4=
403      1  .0        .0       .0      .0        NpO2+ HSO4-
404      1  .0        .0       .0      .0        NpO2+ OH-
405      1  .0        .0       .0      .0        NpO2+ HCO3-
406      1  .0        .0       .0      .0        NpO2+ CO3=
407      1  .0        .0       .0      .0        NpO2+ B(OH)4-
408      1  .0        .0       .0      .0        NpO2+ B3O3(OH)4-
409      1  .0        .0       .0      .0        NpO2+ B4O5(OH)4=
410      1  .0        .0       .0      .0        NpO2+ Br-
411      1  .0        .0       .0      .0        NpO2+ Am(CO3)2-
412      1  .0        .0       .0      .0        NpO2+ Am(CO3)3=-
413      1  0.312     .0       .0      .0        NpO2+ ClO4-           NR94
414      1  .0        .0       .0      .0        NpO2+ NpO2(OH)2-
415      1  .0        .0       .0      .0        NpO2+ NpO2CO3-
416      1  .0        .0       .0      .0        NpO2+ NpO2(CO3)2=-
417      1  .0        .0       .0      .0        NpO2+ NpO2(CO3)3==-
418      1  .0        .0       .0      .0        NpO2+ H2PO4-
419      1  .0        .0       .0      .0        NpO2+ HPO4=
420      1  .0        .0       .0      .0        NpO2+ PO4=-
421
422
423     -.012 .07  .07   0.   .036 0.   0.  0.  0.  0.  0.  0.    Na:       (cations  2 thru 13)
424      .032 0.   0.   .005 0.    0.   0.  0.  0.  0.  0.        K:        (cations  3 thru 13)
425      .007 0.   .092  0.   0.   0.   0.  0.  0.  0.            Ca:       (cations  4 thru 13)
426        0. .10  0.    0.   0.   0.   0.  0.  0.                Mg:       (cations  5 thru 13)
427        0. 0.   0.    0.   0.   0.   0.  0.                    MgOH:     (cations  6 thru 13)
428        0. 0.   0.    0.   0.   0.   0.                        H :       (cations  7 thru 13)
429        0. 0.   0.    0.   0.   0.                             MgB(OH)4:(cations  8 thru 13)
430        0. 0.   0.    0.   0.                                  CaB(OH)4:(cations  9 thru 13)
431        0. 0.   0.    0.                                       Am+++:    (cations 10 thru 13)
432        0. 0.   0.                                             AmCO3+:   (cations 11 thru 13)
433        0. 0.                                                  Th++++:   (cations 12 thru 13)
434        0.                                                     UO2++:    (cations 13 thru 13)
435
436
437      .02   -.006  -.050  .03   -.02   -.065 .12 .074 0. 0. 0. 0. 0. 0. 0. 0. .1  0. 0. Cl:          (anions  2 thru 20)
438     0.     -.013  .01    .02   -.012  .10   .12  0.  0. 0. 0. 0. 0. 0. 0. 0. 0.  0.    SO4:         (anions  3 thru 20)
439     0.     0.     0.     0.    0.     0.    0.   0.  0. 0. 0. 0. 0. 0. 0. 0. 0.        HSO4:        (anions  4 thru 20)
440     0.     .10    0.     0.    0.     0.    0.   0.  0. 0. 0. 0. 0. 0. 0. 0.           OH:          (anions  5 thru 20)
441    -.04    0.     -.10   -.087 0.     0.    0.   0.  0. 0. 0. 0. 0. 0. 0.              HCO3:        (anions  6 thru 20)
442     0.     0.     0.     0.    0.     0.    0.   0.  0. 0. 0. 0. 0. 0.                 CO3:         (anions  7 thru 20)
443     0.     0.     0.     0.    0.     0.    0.   0.  0. 0. 0. 0. 0.                    B(OH)4:      (anions  8 thru 20)
444     0.     0.     0.     0.    0.     0.    0.   0.  0  0. 0. 0.                       B3O3(OH)4:   (anions  9 thru 20)
445     0.     0.     0.     0.    0.     0.    0.   0.  0. 0. 0.                          B4O5(OH)4:   (anions 10 thru 20)
446     0.     0.     0.     0.    0.     0.    0.   0.  0. 0.                             Br:          (anions 11 thru 20)
447     0.     0.     0.     0.    0.     0.    0.   0.  0.                                Am(CO3)2-:   (anions 12 thru 20)
448     0.     0.     0.     0.    0.     0.    0.   0.                                    Am(CO3)3=-:  (anions 13 thru 20)
449     0.     0.     0.     0.    0.     0.    0.                                         ClO4-:       (anions 14 thru 20)
450     0.     0.     0.     0.    0.     0.                                               NpO2(OH)2-:  (anions 15 thru 20)
451     0.     0.     0.     0.    0.                                                      NpO2CO3-:    (anions 16 thru 20)
452     0.     0.     0.     0.                                                            NpO2(CO3)2=-:(anions 17 thru 20)
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453       0.    0.     0.
454       0.    0.                                                                                         NPC3==-:     (anions 18 thru 20)
455       0.                                                                                               H2PO4-:      (anions 19 thru 20)
456                                                                                                        HPO4=:       (anions 20 thru 20)
457     -.0018 -.010     .0  .0 -.003 .003 .0 .0 .0    .0  .0   .0   .0   .0 0 0 0 0 0 0  Na-K:20anions
458      -.007 -.055     .0  .0   .0   .0  .0 .0 .0    .0  .0   .0   .0   .0 0 0 0 0 0 0  Na-Ca:
459      -.012 -.015     .0  .0   .0   .0  .0 .0 .0    .0  .0   .0   .0   .0 0 0 0 0 0 0  Na-Mg:
460         .0     .0    .0  .0   .0   .0  .0 .0 .0    .0  .0   .0   .0   .0 0 0 0 0 0 0  Na-MgOH:
461      -.004     .0 -.0129 .0   .0   .0  .0 .0 .0    .0  .0   .0   .0   .0 0 0 0 0 0 0  Na-H:
462         .0     .0    .0  .0   .0   .0  .0 .0 .0    .0  .0   .0   .0   .0 0 0 0 0 0 0  Na-MgB(OH)4+
463         .0     .0    .0  .0   .0   .0  .0 .0 .0    .0  .0   .0   .0   .0 0 0 0 0 0 0  Na-CaB(OH)4+
464         .0     .0    .0  .0   .0   .0  .0 .0 .0    .0  .0   .0   .0   .0 0 0 0 0 0 0  Na-Am+++
465         .0     .0    .0  .0   .0   .0  .0 .0 .0    .0  .0   .0   .0   .0 0 0 0 0 0 0  Na-AmCO3+
466         .0     .0    .0  .0   .0   .0  .0 .0 .0    .0  .0   .0   .0   .0 0 0 0 0 0 0  Na-Th++++
467         .0     .0    .0  .0   .0   .0  .0 .0 .0    .0  .0   .0   .0   .0 0 0 0 0 0 0  Na-UO2++
468         .0     .0    .0  .0   .0   .0  .0 .0 .0    .0  .0   .0   .0   .0 0 0 0 0 0 0  Na-NpO2++
469
470     -.025      .0    .0  .0 .0  .0  .0  .0    .0   .0  .0   .0   .0   .0 0 0 0 0 0 0  K -Ca:
471     -.022   -.048    .0  .0 .0  .0  .0  .0    .0   .0  .0   .0   .0   .0 0 0 0 0 0 0  K -Mg:
472       .0       .0    .0  .0 .0  .0  .0  .0    .0   .0  .0   .0   .0   .0 0 0 0 0 0 0  K -MgOH:
473     -.011    .197 -.0265 .0 .0  .0  .0  .0    .0   .0  .0   .0   .0   .0 0 0 0 0 0 0  K -H :
474       .0       .0    .0  .0 .0  .0  .0  .0    .0   .0  .0   .0   .0   .0 0 0 0 0 0 0  K -MgB(OH)4+
475       .0       .0    .0  .0 .0  .0  .0  .0    .0   .0  .0   .0   .0   .0 0 0 0 0 0 0  K -CaB(OH)4+
476       .0       .0    .0  .0 .0  .0  .0  .0    .0   .0  .0   .0   .0   .0 0 0 0 0 0 0  K -Am+++
477       .0       .0    .0  .0 .0  .0  .0  .0    .0   .0  .0   .0   .0   .0 0 0 0 0 0 0  K -AmCO3+
478       .0       .0    .0  .0 .0  .0  .0  .0    .0   .0  .0   .0   .0   .0 0 0 0 0 0 0  K -Th++++
479       .0       .0    .0  .0 .0  .0  .0  .0    .0   .0  .0   .0   .0   .0 0 0 0 0 0 0  K -UO2++
480       .0       .0    .0  .0 .0  .0  .0  .0    .0   .0  .0   .0   .0   .0 0 0 0 0 0 0  K -NpO2++
481
482     -.012 .024  .0  .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  Ca-Mg:
483       .0    .0  .0  .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  Ca-MgOH:
484     -.015   .0  .0  .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  Ca-H :
485       .0    .0  .0  .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  Ca-MgB(OH)4+
486       .0    .0  .0  .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  Ca-CaB(OH)4+
487       .0    .0  .0  .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  Ca-Am+++
488       .0    .0  .0  .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  Ca-AmCO3+
489       .0    .0  .0  .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  Ca-Th++++
490       .0    .0  .0  .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  Ca-UO2++
491       .0    .0  .0  .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  Ca-NpO2++
492
493      .028 .0      .0 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  Mg-MgOH:
494     -.011 .0 -.0178 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  Mg-H :
495       .0  .0      .0 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  Mg-MgB(OH)4+
496       .0  .0      .0 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  Mg-CaB(OH)4+
497       .0  .0      .0 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  Mg-Am+++
498       .0  .0      .0 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  Mg-AmCO3+
499       .0  .0      .0 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  Mg-Th++++
500       .0  .0      .0 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  Mg-UO2++
501       .0  .0      .0 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  Mg-NpO2++
502
503      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  MgOH-H :
504      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  MgOH-MgB(OH)4+
505      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  MgOH-CaB(OH)4+
506      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  MgOH-Am+++
507      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  MgOH-AmCO3+
508      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  MgOH-Th++++
509      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  MgOH-UO2++
510      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  MgOH-NpO2++
511
512      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  H-MgB(OH)4+
513      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  H-CaB(OH)4+
514      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  H-Am+++
515      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  H-AmCO3+
516      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  H-Th++++
517      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  H-UO2++
518      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  H-NpO2++
519
520      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  MgB(OH)4-CaB(OH)4+
521      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  MgB(OH)4-Am+++
522      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  MgB(OH)4-AmCO3+
523      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  MgB(OH)4-Th++++
524      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  MgB(OH)4-UO2++
525      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  MgB(OH)4-NpO2++
526
527      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  CaB(OH)4-Am+++
528      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  CaB(OH)4-AmCO3+
529      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  CaB(OH)4-Th++++
530      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  CaB(OH)4-UO2++
531      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  CaB(OH)4-NpO2++
532
533      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  Am+++-AmCO3+
534      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  Am+++-Th++++
535      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  Am+++-UO2++
536      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  Am+++-NpO2++
537
538      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  AmCO3+-Th++++
539      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  AmCO3+-UO2++
540      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  AmCO3+-NpO2++
541
542      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  Th++++-UO2++
543      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0 0 0 0 0 0 0  Th++++-NpO2++
544
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  .0    .0    .0    .0    .0    .0    .0    .0    .0    .0    .0    .0    .0    .0 0 0 0 0 0 0  UO2++-NpO2++

  .0014    .0 -.018 -.004 .0   .0 .0 .0   .0   .0   .0   .0   .0  Cl-SO4:(Na,K,Ca,Mg,
 -.006     .0    .0     .0 .0 .013 .0 .0   .0   .0   .0   .0   .0  Cl-HSO4:  MgOH,H,MgB(OH)3,
 -.006 -.006 -.025     .0 .0   .0 .0 .0   .0   .0   .0   .0   .0  Cl-OH:    CaB(OH)4,Am+++,
 -.015     .0    .0 -.096 .0   .0 .0 .0   .0   .0   .0   .0   .0  Cl-HCO3:  AmCO3+,Th++++,
  .0085  .004    .0     .0 .0   .0 .0 .0   .0   .0   .0   .0   .0  Cl-CO3:   UO2++,NpO2+)
-.0073     .0    .0     .0 .0   .0 .0 .0   .0   .0   .0   .0   .0  Cl-B(OH)4:
 -.024     .0    .0     .0 .0   .0 .0 .0   .0   .0   .0   .0   .0  Cl-B3O3(OH)4:
  .026     .0    .0     .0 .0   .0 .0 .0   .0   .0   .0   .0   .0  Cl-B4O5(OH)4:
    .0     .0    .0     .0 .0   .0 .0 .0   .0   .0   .0   .0   .0  Cl-Br:
    .0     .0    .0     .0 .0   .0 .0 .0   .0   .0   .0   .0   .0  Cl-Am(CO3)2:
    .0     .0    .0     .0 .0   .0 .0 .0   .0   .0   .0   .0   .0  Cl-Am(CO3)3:
    .0     .0    .0     .0 .0   .0 .0 .0   .0   .0   .0   .0   .0  Cl-ClO4-:
    .0     .0    .0     .0 .0   .0 .0 .0   .0   .0   .0   .0   .0  Cl-NpO2(OH)2-:
    .0     .0    .0     .0 .0   .0 .0 .0   .0   .0   .0   .0   .0  Cl-NpO2CO3-:
    .0     .0    .0     .0 .0   .0 .0 .0   .0   .0   .0   .0   .0  Cl-NpO2(CO3)2=-:
    .0  -.01     .0     .0 .0   .0 .0 .0   .0   .0   .0   .0   .0  Cl-NpO2(CO3)3==-:
    .0     .0    .0     .0 .0   .0 .0 .0   .0   .0   .0   .0   .0  Cl-H2PO4-:
    .0     .0    .0     .0 .0   .0 .0 .0   .0   .0   .0   .0   .0  Cl-HPO4=:
    .0     .0    .0     .0 .0   .0 .0 .0   .0   .0   .0   .0   .0  Cl-PO4=-:

-.0094 -.0677 .0 -.0425 .0 .0  .0   .0   .0   .0   .0   .0   .0  SO4-HSO4:
 -.009 -.050 .0     .0  .0 .0  .0   .0   .0   .0   .0   .0   .0  SO4-OH:
 -.005 .0    .0  -.161  .0 .0  .0   .0   .0   .0   .0   .0   .0  SO4-HCO3:
 -.005 -.009 .0     .0  .0 .0  .0   .0   .0   .0   .0   .0   .0  SO4-CO3:
 .0    .0    .0     .0  .0 .0  .0   .0   .0   .0   .0   .0   .0  SO4-B(OH)4:
 .0    .0    .0     .0  .0 .0  .0   .0   .0   .0   .0   .0   .0  SO4-B3O3(OH)4:
 .0    .0    .0     .0  .0 .0  .0   .0   .0   .0   .0   .0   .0  SO4-B4O5(OH)4:
 .0    .0    .0     .0  .0 .0  .0   .0   .0   .0   .0   .0   .0  SO4-Br:
 .0    .0    .0     .0  .0 .0  .0   .0   .0   .0   .0   .0   .0  SO4-Am(CO3)2:
 .0    .0    .0     .0  .0 .0  .0   .0   .0   .0   .0   .0   .0  SO4-Am(CO3)3:
 .0    .0    .0     .0  .0 .0  .0   .0   .0   .0   .0   .0   .0  SO4-ClO4-:
 .0    .0    .0     .0  .0 .0  .0   .0   .0   .0   .0   .0   .0  SO4-NpO2(OH)2-:
 .0    .0    .0     .0  .0 .0  .0   .0   .0   .0   .0   .0   .0  SO4-NpO2CO3-:
 .0    .0    .0     .0  .0 .0  .0   .0   .0   .0   .0   .0   .0  SO4-NpO2(CO3)2=-:
 .0    .0    .0     .0  .0 .0  .0   .0   .0   .0   .0   .0   .0  SO4-NpO2(CO3)3==-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  SO4-H2PO4-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  SO4-HPO4=:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  SO4-PO4=-:

 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HSO4-OH:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HSO4-HCO3-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HSO4-CO3=:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HSO4-B(OH)4-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HSO4-B3O3(OH)4:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HSO4-B4O5(OH)4:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HSO4-Br-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HSO4-Am(CO3)2-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HSO4-Am(CO3)2=-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HSO4-ClO4-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HSO4-NpO2(OH)2-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HSO4-NpO2CO3-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HSO4-NpO2(CO3)2=-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HSO4-NpO2(CO3)3==-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HSO4-H2PO4-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HSO4-HPO4=:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HSO4-PO4=-:

    .0    .0  .0  .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  OH-HCO3:
 -.017  -.01  .0  .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  OH-CO3:
    .0    .0  .0  .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  OH-B(OH)4:
    .0    .0  .0  .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  OH-B3O3(OH)4:
    .0    .0  .0  .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  OH-B4O5(OH)4:
    .0    .0  .0  .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  OH-Br:
    .0    .0  .0  .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  OH-Am(CO3)2:
    .0    .0  .0  .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  OH-Am(CO3)3:
    .0    .0  .0  .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  OH-ClO4-:
    .0    .0  .0  .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  OH-NpO2(OH)2-:
    .0    .0  .0  .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  OH-NpO2CO3-:
    .0    .0  .0  .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  OH-NpO2(CO3)2=-:
    .0    .0  .0  .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  OH-NpO2(CO3)3==-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  OH-H2PO4-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  OH-HPO4=:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  OH-PO4=-:

.002 .012  .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HCO3-CO3:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HCO3-B(OH)4:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HCO3-B3O3(OH)4:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HCO3-B4O5(OH)4:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HCO3-Br:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HCO3-Am(CO3)2:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HCO3-Am(CO3)3:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HCO3-ClO4-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HCO3-NpO2(OH)2-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HCO3-NpO2CO3-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HCO3-NpO2(CO3)2=-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HCO3-NpO2(CO3)3==-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HCO3-H2PO4-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HCO3-HPO4=:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HCO3-PO4=-:

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 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  CO3-B(OH)4-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  CO3-B3O3(OH)4:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  CO3-B4O5(OH)4:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  CO3-Br:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  CO3-Am(CO3)2:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  CO3-Am(CO3)3:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  CO3-ClO4-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  CO3-NpO2(OH)2-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  CO3-NpO2CO3-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  CO3-NpO2(CO3)2=-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  CO3-NpO2(CO3)3==-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  CO3-H2PO4-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  CO3-HPO4=:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  CO3-PO4=-:

 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B(OH)4-B3O3(OH)4:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B(OH)4-B4O5(OH)4:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B(OH)4-Br:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B(OH)4-Am(CO3)2:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B(OH)4-Am(CO3)3:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B(OH)4-ClO4-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B(OH)4-NpO2(OH)2-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B(OH)4-NpO2CO3-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B(OH)4-NpO2(CO3)2=-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B(OH)4-NpO2(CO3)3==-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B(OH)4-H2PO4-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B(OH)4-HPO4=:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B(OH)4-PO4=-:

 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B3O3(OH)4-B4O5(OH)4:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B3O3(OH)4-Br:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B3O3(OH)4-Am(CO3)2:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B3O3(OH)4-Am(CO3)3:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B3O3(OH)4-ClO4-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B3O3(OH)4-NpO2(OH)2-
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B3O3(OH)4-NpO2CO3-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B3O3(OH)4-NpO2(CO3)2=-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B3O3(OH)4-NpO2(CO3)3==-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B3O3(OH)4-H2PO4-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B3O3(OH)4-HPO4=:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B3O3(OH)4-PO4=-:

 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B4O5(OH)4=Br:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B4O5(OH)4=Am(CO3)2:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B4O5(OH)4=Am(CO3)3:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B4O5(OH)4=ClO4-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B4O5(OH)4=NPO2(OH)2-
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B4O5(OH)4=NpO2CO3-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B4O5(OH)4=NpO2(CO3)2=-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B4O5(OH)4=NpO2(CO3)3==-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B4O5(OH)4=H2PO4-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B4O5(OH)4=HPO4=:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  B4O5(OH)4=PO4=-:

 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Br-Am(CO3)2:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Br-Am(CO3)3:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Br-ClO4-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Br-NpO2(OH)2-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Br-NpO2CO3-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Br-NpO2(CO3)2=-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Br-NpO2(CO3)3==-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Br-H2PO4-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Br-HPO4=:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Br-PO4=-:

 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Am(CO3)2-Am(CO3)3:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Am(CO3)2-ClO4-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Am(CO3)2-NpO2(OH)2-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Am(CO3)2-NpO2CO3-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Am(CO3)2-NpO2(CO3)2=-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Am(CO3)2-NpO2(CO3)3==-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Am(CO3)2-H2PO4-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Am(CO3)2-HPO4=:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Am(CO3)2-PO4=-:

 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Am(CO3)3-ClO4-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Am(CO3)3-NpO2(OH)2-
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Am(CO3)3-NpO2CO3-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Am(CO3)3-NpO2(CO3)2=-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Am(CO3)3-NpO2(CO3)3==-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Am(CO3)3-H2PO4-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Am(CO3)3-HPO4=:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  Am(CO3)3-PO4=-:

 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  ClO4-NpO2(OH)2-
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  ClO4-NpO2CO3-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  ClO4-NpO2(CO3)2=-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  ClO4-NpO2(CO3)3==-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  ClO4-H2PO4-:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  ClO4-HPO4=:
 .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  ClO4-PO4=-:

```

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```
729      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  NpO2(OH)2--NpO2CO3-:
730      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  NpO2(OH)2--NpO2(CO3)2=-:
731      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  NpO2(OH)2--NpO2(CO3)3==-:
732      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  NpO2(OH)2-H2PO4-:
733      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  NpO2(OH)2-HPO4=:
734      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  NpO2(OH)2-PO4=-:
735
736      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  NpO2CO3-NpO2(CO3)2=-:
737      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  NpO2CO3-NpO2(CO3)3==-:
738      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  NpO2CO3-H2PO4-:
739      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  NpO2CO3-HPO4=:
740      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  NpO2CO3-PO4=-:
741
742      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  NpC2-NpO2(CO3)3==-:
743      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  NpC2-H2PO4-:
744      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  NpC2-HPO4=:
745      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  NpC2-PO4=-:
746
747      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  NpO2(CO3)3-H2PO4-:
748      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  NpO2(CO3)3-HPO4=:
749      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  NpO2(CO3)3-PO4=-:
750
751      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  H2PO4-HPO4=:
752      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  H2PO4-PO4=-:
753
754      .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0   .0  HPO4-PO4=-:
755
756      .100 .051  .183 .183 .0  .0  .0  .0  .0  .0  .0  .0  .0  CO2-Cations      HMW84
757        .0   .0    .0   .0 .0  .0  .0  .0  .0  .0  .0  .0  .0  CaCO3-Cations    HMW84
758        .0   .0    .0   .0 .0  .0  .0  .0  .0  .0  .0  .0  .0  MgCO3-Cations    HMW84
759     -.097 -.14    .0   .0 .0  .0  .0  .0  .0  .0  .0  .0  .0  B(OH)3-Cations   FW86
760        .0   .0    .0   .0 .0  .0  .0  .0  .0  .0  .0  .0  .0  NpO2OH-Cations
761        .0 -.07    .0   .0 .0 .29  .0  .0  .0  .0  .0  .0  .0  H3PO4-Cations    PS76
762
763     -.005 .097 -.003   .0 .0  .0  .0  .0  .0  .0  .0  .0  .0  0 0 0 0    0 0 0  CO2-Anions    HMW84
764        .0   .0    .0   .0 .0  .0  .0  .0  .0  .0  .0  .0  .0  0 0 0 0    0 0 0  CaCO3-Anions  HMW84
765        .0   .0    .0   .0 .0  .0  .0  .0  .0  .0  .0  .0  .0  0 0 0 0    0 0 0  MgCO3-Anions  HMW84
766      .091 .018    .0   .0 .0  .0  .0 -.20 .0  .0  .0  .0  .0  0 0 0 0    0 0 0  B(OH)3-Anions FW86
767        .0   .0    .0   .0 .0  .0  .0  .0  .0  .0  .0  .0  .0  0 0 0 0    0 0 0  NpO2OH-Anions
768        .0   .0    .0   .0 .0  .0  .0  .0  .0  .0  .0  .0  .0  0 0 0 0 -.4 0 0  H3PO4-Anions  PS76
769
770     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0   CO2-Cation-Anion Interactions
771     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0     (Cations down, Anions across)
772     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
773     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
774     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
775     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
776     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
777     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
778     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
779     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
780     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
781     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
782     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
783
784     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0   CaCO3-Cation-Anion Interactions
785     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
786     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
787     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
788     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
789     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
790     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
791     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
792     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
793     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
794     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
795     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
796     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
797
798     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0   MgCO3-Cation-Anion Interactions
799     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
800     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
801     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
802     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
803     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
804     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
805     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
806     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
807     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
808     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
809     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
810     .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
811
812     .0      .046 .0 .0 .0 .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0   B(OH)3-Cation-Anion FW86
813     .0      .0   .0 .0 .0 .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0   (Cations down, Anions across)
814     .0      .0   .0 .0 .0 .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
815     .0      .0   .0 .0 .0 .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
816     .0      .0   .0 .0 .0 .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
817     -.0102  .0   .0 .0 .0 .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
818     .0      .0   .0 .0 .0 .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
819     .0      .0   .0 .0 .0 .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
820     .0      .0   .0 .0 .0 .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
```

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```
  .0      .0     .0 .0 .0 .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
  .0      .0     .0 .0 .0 .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
  .0      .0     .0 .0 .0 .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
  .0      .0     .0 .0 .0 .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0

  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0   NpO2OH(aq)-Cation-Anion
  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0

  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0   H3PO4-Cation-Anion Interactions
  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0
  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0  .0 0 0 0

   2  3  4  5  6  7 22 21 43 44 49 50 37                  cation  mapping for ELMAP(1,)
   8  9 10 11 12 13 18 19 20 23 45 46 24 39 40 41 42 34 35 36   anion   mapping for ELMAP(2,)
  14 15 16 17 38 33                                        neutral mapping for ELMAP(3,)
 'nEQUALACT' 0

 2 (Oxygen)
 'nREDOX' 0
 'nIONEX' 0 0 'nMF'



 FRSR89 ==  Felmy, Rai, Schramke, and Ryan (1989) RCA 48 pp.29-35
 FRF90  ==  Felmy, Rai, and Fulton (1990) RCA 50 pp.193-204
 PS76   ==  Pitzer and Silvester (1976) J Soln Chem 5#4 pp.269-278
 P91    ==  Pitzer (1991) CRC Handbook, Activity Coef in Electrolyte Solns
 RFF92  ==  Rai, Felmy, and Fulton (1992)  RCA 56 pp.7-14
 RFFR92 ==  Rai, Felmy, Fulton, and Ryan (1992) RCA 58/59 pp.9-16
 RFF94  ==  Rai, Felmy, and Fulton (1994)
 RRFF94 ==  Rao, Rai, Felmy, and Fulton (1994) SAND94-1948J for RCA

 NR94   ==  Novak and Roberts (1994) SAND94-xxxx MRS/Kyoto
 HMW84  ==  Harvie, Moller, Weare (1984) GCA 48 pp.723-751
 FW86   ==  Felmy and Weare (1986) GCA 50 pp.2771-2783
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Appendix B: Computer listing of a partial output file from FMT V1.1, showing the echo print of the parameters from HMW_NP_AM.CHEMDAT.

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[.FD.TITRATE]NPAM.in; for testing HMW_NP_AM.CHEMDAT file                FMT V1.1
DATABASE:  HMW84/FW86; Np(V)-Na-CO3-OH-Cl-ClO4 (NR94);
~~94.01.17~~  Am(III)-Na-Cl-CO3-SO4-PO4 (FRSR89,FRF90,P91,RFFR92,RFF94,RRFF94)
95.01.31
Inj. and Init. Pressure [=] ATM  1.00000E+00   1.00000E+00
Accuracy of reactions is                1.0000E-06
Minimum elemental abundance is          1.0000E-18
Number of Aqueous Species is              50


Cation-Anion Binary Interaction Parameters

           Cation           Anion        Beta(0)      Beta(1)      Beta(2)      Cphi      Alpha-Values
Na+                  Cl-                0.07650      0.26440      0.           0.00127   (2.0,12)  1-1, 1-2,
Na+                  SO4=               0.01958      1.11300      0.           0.00497   (2.0,12)  1-1, 1-2,
Na+                  HSO4-              0.04540      0.39800      0.           0.        (2.0,12)  1-1, 1-2,
Na+                  OH-                0.08640      0.25300      0.           0.00440   (2.0,12)  1-1, 1-2,
Na+                  HCO3-              0.02770      0.04110      0.           0.        (2.0,12)  1-1, 1-2,
Na+                  CO3=               0.03990      1.38900      0.           0.00440   (2.0,12)  1-1, 1-2,
Na+                  B(OH)4-           -0.04270      0.08900      0.           0.01140   (2.0,12)  1-1, 1-2,
Na+                  B3O3(OH)4-        -0.05600     -0.91000      0.           0.        (2.0,12)  1-1, 1-2,
Na+                  B4O5(OH)4=        -0.11000     -0.40000      0.           0.        (2.0,12)  1-1, 1-2,
Na+                  Br-                0.           0.           0.           0.        (2.0,12)  1-1, 1-2,
Na+                  Am(CO3)2-          0.          -8.37000      0.           0.        (2.0,12)  1-1, 1-2,
Na+                  Am(CO3)3=-        -0.94000      8.10000      0.           0.41800   (2.0,12)  1-1, 1-2,
Na+                  ClO4-              0.05540      0.27550      0.          -0.00118   (2.0,12)  1-1, 1-2,
Na+                  NpO2(OH)2-         0.           0.           0.           0.        (2.0,12)  1-1, 1-2,
Na+                  NpO2CO3-           0.16100      0.           0.           0.        (2.0,12)  1-1, 1-2,
Na+                  NpO2(CO3)2=-       0.40700      0.           0.           0.        (2.0,12)  1-1, 1-2,
Na+                  NpO2(CO3)3==-      1.97000     16.00000      0.           0.        (2.0,12)  1-1, 1-2,
Na+                  H2PO4-            -0.05330      0.03960      0.           0.00795   (2.0,12)  1-1, 1-2,
Na+                  HPO4=             -0.05830      1.46600      0.           0.02940   (2.0,12)  1-1, 1-2,
Na+                  PO4=-              0.17810      3.85100      0.          -0.05154   (2.0,12)  1-1, 1-2,
K+                   Cl-                0.04835      0.21220      0.          -0.00084   (2.0,12)  1-1, 1-2,
K+                   SO4=               0.04995      0.77930      0.           0.        (2.0,12)  1-1, 1-2,
K+                   HSO4-             -0.00030      0.17350      0.           0.        (2.0,12)  1-1, 1-2,
K+                   OH-                0.12980      0.32000      0.           0.00410   (2.0,12)  1-1, 1-2,
K+                   HCO3-              0.02960     -0.01300      0.          -0.00800   (2.0,12)  1-1, 1-2,
K+                   CO3=               0.14880      1.43000      0.          -0.00150   (2.0,12)  1-1, 1-2,
K+                   B(OH)4-            0.03500      0.14000      0.           0.        (2.0,12)  1-1, 1-2,
K+                   B3O3(OH)4-        -0.13000      0.           0.           0.        (2.0,12)  1-1, 1-2,
K+                   B4O5(OH)4=        -0.02200      0.           0.           0.        (2.0,12)  1-1, 1-2,
K+                   Br-                0.           0.           0.           0.        (2.0,12)  1-1, 1-2,
K+                   Am(CO3)2-          0.           0.           0.           0.        (2.0,12)  1-1, 1-2,
K+                   Am(CO3)3=-         0.           0.           0.           0.        (2.0,12)  1-1, 1-2,
K+                   ClO4-              0.           0.           0.           0.        (2.0,12)  1-1, 1-2,
K+                   NpO2(OH)2-         0.           0.           0.           0.        (2.0,12)  1-1, 1-2,
K+                   NpO2CO3-           0.           0.           0.           0.        (2.0,12)  1-1, 1-2,
K+                   NpO2(CO3)2=-       0.           0.           0.           0.        (2.0,12)  1-1, 1-2,
K+                   NpO2(CO3)3==-      0.           0.           0.           0.        (2.0,12)  1-1, 1-2,
K+                   H2PO4-            -0.06780     -0.10420      0.           0.        (2.0,12)  1-1, 1-2,
K+                   HPO4=              0.02480      1.27400      0.           0.01640   (2.0,12)  1-1, 1-2,
K+                   PO4=-              0.37290      3.97200      0.          -0.08680   (2.0,12)  1-1, 1-2,
Ca++                 Cl-                0.31590      1.61400      0.          -0.00034   (2.0,12)  1-1, 1-2,
Ca++                 SO4=               0.20000      3.19730    -54.24000      0.        (1.4,12)  2-2
Ca++                 HSO4-              0.21450      2.53000      0.           0.        (2.0,12)  1-1, 1-2,
Ca++                 OH-               -0.17470     -0.23030     -5.72000      0.        (2.0,12)  1-1, 1-2,
Ca++                 HCO3-              0.40000      2.97700      0.           0.        (2.0,12)  1-1, 1-2,
Ca++                 CO3=               0.           0.           0.           0.        (1.4,12)  2-2
Ca++                 B(OH)4-            0.           0.           0.           0.        (2.0,12)  1-1, 1-2,
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 61   Ca++      B3O3(OH)4-      0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
 62   Ca++      B4O5(OH)4=      0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
 63   Ca++      Br-             0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
 64   Ca++      Am(CO3)2-       0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
 65   Ca++      Am(CO3)3=-      0.        0.        0.         0.        (1.4,50)  2-(n>2)
 66   Ca++      ClO4-           0.45110   1.75600   0.        -0.00500   (2.0,12)  1-1, 1-2,
 67   Ca++      NpO2(OH)2-      0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
 68   Ca++      NpO2CO3-        0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
 69   Ca++      NpO2(CO3)2=-    0.        0.        0.         0.        (1.4,50)  2-(n>2)
 70   Ca++      NpO2(CO3)3==-   0.        0.        0.         0.        (1.4,50)  2-(n>2)
 71   Ca++      H2PO4-          0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
 72   Ca++      HPO4=           0.        0.        0.         0.        (1.4,12)  2-2
 73   Ca++      PO4=-           0.        0.        0.         0.        (1.4,50)  2-(n>2)
 74   Mg++      Cl-             0.35235   1.68150   0.         0.00519   (2.0,12)  1-1, 1-2,
 75   Mg++      SO4=            0.22100   3.34300   -37.23000  0.02500   (1.4,12)  2-2
 76   Mg++      HSO4-           0.47460   1.72900   0.         0.        (2.0,12)  1-1, 1-2,
 77   Mg++      OH-             0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
 78   Mg++      HCO3-           0.32900   0.60720   0.         0.        (2.0,12)  1-1, 1-2,
 79   Mg++      CO3=            0.        0.        0.         0.        (1.4,12)  2-2
 80   Mg++      B(OH)4-         0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
 81   Mg++      B3O3(OH)4-      0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
 82   Mg++      B4O5(OH)4=      0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
 83   Mg++      Br-             0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
 84   Mg++      Am(CO3)2-       0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
 85   Mg++      Am(CO3)3=-      0.        0.        0.         0.        (1.4,50)  2-(n>2)
 86   Mg++      ClO4-           0.49610   2.00800   0.         0.00958   (2.0,12)  1-1, 1-2,
 87   Mg++      NpO2(OH)2-      0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
 88   Mg++      NpO2CO3-        0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
 89   Mg++      NpO2(CO3)2=-    0.        0.        0.         0.        (1.4,50)  2-(n>2)
 90   Mg++      NpO2(CO3)3==-   0.        0.        0.         0.        (1.4,50)  2-(n>2)
 91   Mg++      H2PO4-          0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
 92   Mg++      HPO4=           0.        0.        0.         0.        (1.4,12)  2-2
 93   Mg++      PO4=-           0.        0.        0.         0.        (1.4,50)  2-(n>2)
 94   MgOH+     Cl-            -0.10000   1.65800   0.         0.        (2.0,12)  1-1, 1-2,
 95   MgOH+     SO4=            0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
 96   MgOH+     HSO4-           0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
 97   MgOH+     OH-             0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
 98   MgOH+     HCO3-           0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
 99   MgOH+     CO3=            0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
100   MgOH+     B(OH)4-         0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
101   MgOH+     B3O3(OH)4-      0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
102   MgOH+     B4O5(OH)4=      0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
103   MgOH+     Br-             0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
104   MgOH+     Am(CO3)2-       0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
105   MgOH+     Am(CO3)3=-      0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
106   MgOH+     ClO4-           0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
107   MgOH+     NpO2(OH)2-      0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
108   MgOH+     NpO2CO3-        0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
109   MgOH+     NpO2(CO3)2=-    0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
110   MgOH+     NpO2(CO3)3==-   0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
111   MgOH+     H2PO4-          0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
112   MgOH+     HPO4=           0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
113   MgOH+     PO4=-           0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
114   H+        Cl-             0.17750   0.29450   0.         0.00080   (2.0,12)  1-1, 1-2,
115   H+        SO4=            0.02980   0.        0.         0.04380   (2.0,12)  1-1, 1-2,
116   H+        HSO4-           0.20650   0.55560   0.         0.        (2.0,12)  1-1, 1-2,
117   H+        OH-             0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
118   H+        HCO3-           0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
119   H+        CO3=            0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
120   H+        B(OH)4-         0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
121   H+        B3O3(OH)4-      0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
122   H+        B4O5(OH)4=      0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
123   H+        Br-             0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
124   H+        Am(CO3)2-       0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
125   H+        Am(CO3)3=-      0.        0.        0.         0.        (2.0,12)  1-1, 1-2,
126   H+        ClO4-           0.17470   0.29310   0.         0.00819   (2.0,12)  1-1, 1-2,
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127   H+          NpO2(OH)2-       0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
128   H+          NpO2CO3-         0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
129   H+          NpO2(CO3)2=-     0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
130   H+          NpO2(CO3)3==-    0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
131   H+          H2PO4-           0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
132   H+          HPO4=            0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
133   H+          PO4=-            0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
134   MgB(OH)4+   Cl-              0.16000    0.        0.        0.        (2.0,12)  1-1, 1-2,
135   MgB(OH)4+   SO4=             0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
136   MgB(OH)4+   HSO4-            0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
137   MgB(OH)4+   OH-              0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
138   MgB(OH)4+   HCO3-            0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
139   MgB(OH)4+   CO3=             0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
140   MgB(OH)4+   B(OH)4-          0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
141   MgB(OH)4+   B3O3(OH)4-       0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
142   MgB(OH)4+   B4O5(OH)4=       0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
143   MgB(OH)4+   Br-              0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
144   MgB(OH)4+   Am(CO3)2-        0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
145   MgB(OH)4+   Am(CO3)3=-       0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
146   MgB(OH)4+   ClO4-            0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
147   MgB(OH)4+   NpO2(OH)2-       0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
148   MgB(OH)4+   NpO2CO3-         0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
149   MgB(OH)4+   NpO2(CO3)2=-     0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
150   MgB(OH)4+   NpO2(CO3)3==-    0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
151   MgB(OH)4+   H2PO4-           0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
152   MgB(OH)4+   HPO4=            0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
153   MgB(OH)4+   PO4=-            0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
154   CaB(OH)4+   Cl-              0.12000    0.        0.        0.        (2.0,12)  1-1, 1-2,
155   CaB(OH)4+   SO4=             0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
156   CaB(OH)4+   HSO4-            0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
157   CaB(OH)4+   OH-              0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
158   CaB(OH)4+   HCO3-            0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
159   CaB(OH)4+   CO3=             0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
160   CaB(OH)4+   B(OH)4-          0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
161   CaB(OH)4+   B3O3(OH)4-       0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
162   CaB(OH)4+   B4O5(OH)4=       0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
163   CaB(OH)4+   Br-              0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
164   CaB(OH)4+   Am(CO3)2-        0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
165   CaB(OH)4+   Am(CO3)3=-       0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
166   CaB(OH)4+   ClO4-            0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
167   CaB(OH)4+   NpO2(OH)2-       0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
168   CaB(OH)4+   NpO2CO3-         0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
169   CaB(OH)4+   NpO2(CO3)2=-     0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
170   CaB(OH)4+   NpO2(CO3)3==-    0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
171   CaB(OH)4+   H2PO4-           0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
172   CaB(OH)4+   HPO4=            0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
173   CaB(OH)4+   PO4=-            0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
174   Am+++       Cl-              0.61170    5.40300   0.       -0.02840   (2.0,12)  1-1, 1-2,
175   Am+++       SO4=             3.03980    0.     -2500.       0.        (1.4,50)  2-(n>2)
176   Am+++       HSO4-            0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
177   Am+++       OH-              0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
178   Am+++       HCO3-            0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
179   Am+++       CO3=             0.         0.        0.        0.        (1.4,50)  2-(n>2)
180   Am+++       B(OH)4-          0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
181   Am+++       B3O3(OH)4-       0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
182   Am+++       B4O5(OH)4=       0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
183   Am+++       Br-              0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
184   Am+++       Am(CO3)2-        0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
185   Am+++       Am(CO3)3=-       0.         0.        0.        0.        (1.4,50)  2-(n>2)
186   Am+++       ClO4-            0.80000    5.35000   0.       -0.00480   (2.0,12)  1-1, 1-2,
187   Am+++       NpO2(OH)2-       0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
188   Am+++       NpO2CO3-         0.         0.        0.        0.        (2.0,12)  1-1, 1-2,
189   Am+++       NpO2(CO3)2=-     0.         0.        0.        0.        (1.4,50)  2-(n>2)
190   Am+++       NpO2(CO3)3==-    0.         0.        0.        0.        (1.4,50)  2-(n>2)
191   Am+++       H2PO4-           0.         0.      -92.90000   0.        (2.0,12)  1-1, 1-2,
192   Am+++       HPO4=            0.         0.        0.        0.        (1.4,50)  2-(n>2)
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193  Am+++      PO4=-          0.       0.       0.   0.        (1.4,50)  2-(n>2)
194  AmCO3+     Cl-            0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
195  AmCO3+     SO4=           0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
196  AmCO3+     HSO4-          0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
197  AmCO3+     OH-            0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
198  AmCO3+     HCO3-          0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
199  AmCO3+     CO3=           0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
200  AmCO3+     B(OH)4-        0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
201  AmCO3+     B3O3(OH)4-     0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
202  AmCO3+     B4O5(OH)4=     0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
203  AmCO3+     Br-            0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
204  AmCO3+     Am(CO3)2-      0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
205  AmCO3+     Am(CO3)3=-     0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
206  AmCO3+     ClO4-          0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
207  AmCO3+     NpO2(OH)2-     0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
208  AmCO3+     NpO2CO3-       0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
209  AmCO3+     NpO2(CO3)2=-   0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
210  AmCO3+     NpO2(CO3)3==-  0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
211  AmCO3+     H2PO4-         0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
212  AmCO3+     HPO4=          0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
213  AmCO3+     PO4=-          0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
214  Th++++     Cl-            0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
215  Th++++     SO4=           0.       0.       0.   0.        (1.4,50)  2-(n>2)
216  Th++++     HSO4-          0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
217  Th++++     OH-            0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
218  Th++++     HCO3-          0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
219  Th++++     CO3=           0.       0.       0.   0.        (1.4,50)  2-(n>2)
220  Th++++     B(OH)4-        0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
221  Th++++     B3O3(OH)4-     0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
222  Th++++     B4O5(OH)4=     0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
223  Th++++     Br-            0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
224  Th++++     Am(CO3)2-      0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
225  Th++++     Am(CO3)3=-     0.       0.       0.   0.        (1.4,50)  2-(n>2)
226  Th++++     ClO4-          0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
227  Th++++     NpO2(OH)2-     0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
228  Th++++     NpO2CO3-       0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
229  Th++++     NpO2(CO3)2=-   0.       0.       0.   0.        (1.4,50)  2-(n>2)
230  Th++++     NpO2(CO3)3==-  0.       0.       0.   0.        (1.4,50)  2-(n>2)
231  Th++++     H2PO4-         0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
232  Th++++     HPO4=          0.       0.       0.   0.        (1.4,50)  2-(n>2)
233  Th++++     PO4=-          0.       0.       0.   0.        (1.4,50)  2-(n>2)
234  UO2++      Cl-            0.42740  1.64400  0.  -0.03686   (2.0,12)  1-1, 1-2,
235  UO2++      SO4=           0.32200  1.82700  0.  -0.01760   (1.4,12)  2-2
236  UO2++      HSO4-          0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
237  UO2++      OH-            0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
238  UO2++      HCO3-          0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
239  UO2++      CO3=           0.       0.       0.   0.        (1.4,12)  2-2
240  UO2++      B(OH)4-        0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
241  UO2++      B3O3(OH)4-     0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
242  UO2++      B4O5(OH)4=     0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
243  UO2++      Br-            0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
244  UO2++      Am(CO3)2-      0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
245  UO2++      Am(CO3)3=-     0.       0.       0.   0.        (1.4,50)  2-(n>2)
246  UO2++      ClO4-          0.61130  2.14400  0.   0.02168   (2.0,12)  1-1, 1-2,
247  UO2++      NpO2(OH)2-     0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
248  UO2++      NpO2CO3-       0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
249  UO2++      NpO2(CO3)2=-   0.       0.       0.   0.        (1.4,50)  2-(n>2)
250  UO2++      NpO2(CO3)3==-  0.       0.       0.   0.        (1.4,50)  2-(n>2)
251  UO2++      H2PO4-         0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
252  UO2++      HPO4=          0.       0.       0.   0.        (1.4,12)  2-2
253  UO2++      PO4=-          0.       0.       0.   0.        (1.4,50)  2-(n>2)
254  NpO2+      Cl-            0.16900  0.       0.   0.        (2.0,12)  1-1, 1-2,
255  NpO2+      SO4=           0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
256  NpO2+      HSO4-          0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
257  NpO2+      OH-            0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
258  NpO2+      HCO3-          0.       0.       0.   0.        (2.0,12)  1-1, 1-2,
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NpO2+          CO3=               0.          0.          0.           0.        (2.0,12)  1-1, 1-2,
NpO2+          B(OH)4-            0.          0.          0.           0.        (2.0,12)  1-1, 1-2,
NpO2+          B3O3(OH)4-         0.          0.          0.           0.        (2.0,12)  1-1, 1-2,
NpO2+          B4O5(OH)4=         0.          0.          0.           0.        (2.0,12)  1-1, 1-2,
NpO2+          Br-                0.          0.          0.           0.        (2.0,12)  1-1, 1-2,
NpO2+          Am(CO3)2-          0.          0.          0.           0.        (2.0,12)  1-1, 1-2,
NpO2+          Am(CO3)3=-         0.          0.          0.           0.        (2.0,12)  1-1, 1-2,
NpO2+          ClO4-              0.31200     0.          0.           0.        (2.0,12)  1-1, 1-2,
NpO2+          NpO2(OH)2-         0.          0.          0.           0.        (2.0,12)  1-1, 1-2,
NpO2+          NpO2CO3-           0.          0.          0.           0.        (2.0,12)  1-1, 1-2,
NpO2+          NpO2(CO3)2=-       0.          0.          0.           0.        (2.0,12)  1-1, 1-2,
NpO2+          NpO2(CO3)3==-      0.          0.          0.           0.        (2.0,12)  1-1, 1-2,
NpO2+          H2PO4-             0.          0.          0.           0.        (2.0,12)  1-1, 1-2,
NpO2+          HPO4=              0.          0.          0.           0.        (2.0,12)  1-1, 1-2,
NpO2+          PO4=-              0.          0.          0.           0.        (2.0,12)  1-1, 1-2,

Cation-Cation Ternary Interactions

                  K+         Ca++       Mg++       MgOH+      H+         MgB(OH)4+  CaB(OH)4+  Am+++      AmCO3+     Th++++
Na+              -0.01200    0.07000    0.07000    0.         0.03600    0.         0.         0.         0.         0.
                  UO2++      NpO2+
Na+               0.         0.

                  Ca++       Mg++       MgOH+      H+         MgB(OH)4+  CaB(OH)4+  Am+++      AmCO3+     Th++++     UO2++
K+                0.03200    0.         0.         0.00500    0.         0.         0.         0.         0.         0.
                  NpO2+
K+                0.

                  Mg++       MgOH+      H+         MgB(OH)4+  CaB(OH)4+  Am+++      AmCO3+     Th++++     UO2++      NpO2+
Ca++              0.00700    0.         0.09200    0.         0.         0.         0.         0.         0.         0.

                  MgOH+      H+         MgB(OH)4+  CaB(OH)4+  Am+++      AmCO3+     Th++++     UO2++      NpO2+
Mg++              0.         0.10000    0.         0.         0.         0.         0.         0.         0.

                  H+         MgB(OH)4+  CaB(OH)4+  Am+++      AmCO3+     Th++++     UO2++      NpO2+
MgOH+             0.         0.         0.         0.         0.         0.         0.         0.

                  MgB(OH)4+  CaB(OH)4+  Am+++      AmCO3+     Th++++     UO2++      NpO2+
H+                0.         0.         0.         0.         0.         0.         0.

                  CaB(OH)4+  Am+++      AmCO3+     Th++++     UO2++      NpO2+
MgB(OH)4+         0.         0.         0.         0.         0.         0.

                  Am+++      AmCO3+     Th++++     UO2++      NpO2+
CaB(OH)4+         0.         0.         0.         0.         0.

                  AmCO3+     Th++++     UO2++      NpO2+
Am+++             0.         0.         0.         0.

                  Th++++     UO2++      NpO2+
AmCO3+            0.         0.         0.

                  UO2++      NpO2+
Th++++            0.         0.

                  NpO2+
UO2++             0.


Anion-Anion Ternary Interactions

                  SO4=       HSO4-      OH-        HCO3-      CO3=       B(OH)4-    B3O3(OH)4- B4O5(OH)4= Br-        Am(CO3)2-
Cl-               0.02000   -0.00600   -0.05000    0.03000   -0.02000   -0.06500    0.12000    0.07400    0.         0.
                  Am(CO3)3=- ClO4-      NpO2(OH)2- NpO2CO3-   NpO2(CO3)2 NpO2(CO3)3 H2PO4-     HPO4=      PO4=-
Cl-               0.         0.         0.         0.         0.         0.         0.10000    0.         0.
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                 HSO4-      OH-        HCO3-      CO3=       B(OH)4-    B3O3(OH)4- B4O5(OH)4= Br-        Am(CO3)2-  Am(CO3)3=-
SO4=             0.        -0.01300    0.01000    0.02000   -0.01200    0.10000    0.12000    0.         0.         0.
                 ClO4-      NpO2(OH)2- NpO2CO3-   NpO2(CO3)2 NpO2(CO3)3 H2PO4-     HPO4=      PO4=-
SO4=             0.         0.         0.         0.         0.         0.         0.         0.

                 OH-        HCO3-      CO3=       B(OH)4-    B3O3(OH)4- B4O5(OH)4= Br-        Am(CO3)2-  Am(CO3)3=- ClO4-
HSO4-            0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
                 NpO2(OH)2- NpO2CO3-   NpO2(CO3)2 NpO2(CO3)3 H2PO4-     HPO4=      PO4=-
HSO4-            0.         0.         0.         0.         0.         0.         0.

                 HCO3-      CO3=       B(OH)4-    B3O3(OH)4- B4O5(OH)4= Br-        Am(CO3)2-  Am(CO3)3=- ClO4-      NpO2(OH)2-
OH-              0.         0.10000    0.         0.         0.         0.         0.         0.         0.         0.
                 NpO2CO3-   NpO2(CO3)2 NpO2(CO3)3 H2PO4-     HPO4=      PO4=-
OH-              0.         0.         0.         0.         0.         0.

                 CO3=       B(OH)4-    B3O3(OH)4- B4O5(OH)4= Br-        Am(CO3)2-  Am(CO3)3=- ClO4-      NpO2(OH)2- NpO2CO3-
HCO3-           -0.04000    0.        -0.10000   -0.08700    0.         0.         0.         0.         0.         0.
                 NpO2(CO3)2 NpO2(CO3)3 H2PO4-     HPO4=      PO4=-
HCO3-            0.         0.         0.         0.         0.

                 B(OH)4-    B3O3(OH)4- B4O5(OH)4= Br-        Am(CO3)2-  Am(CO3)3=- ClO4-      NpO2(OH)2- NpO2CO3-   NpO2(CO3)2
CO3=             0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
                 NpO2(CO3)3 H2PO4-     HPO4=      PO4=-
CO3=             0.         0.         0.         0.

                 B3O3(OH)4- B4O5(OH)4= Br-        Am(CO3)2-  Am(CO3)3=- ClO4-      NpO2(OH)2- NpO2CO3-   NpO2(CO3)2 NpO2(CO3)3
B(OH)4-          0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
                 H2PO4-     HPO4=      PO4=-
B(OH)4-          0.         0.         0.

                 B4O5(OH)4= Br-        Am(CO3)2-  Am(CO3)3=- ClO4-      NpO2(OH)2- NpO2CO3-   NpO2(CO3)2 NpO2(CO3)3 H2PO4-
B3O3(OH)4-       0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
                 HPO4=      PO4=-
B3O3(OH)4-       0.         0.

                 Br-        Am(CO3)2-  Am(CO3)3=- ClO4-      NpO2(OH)2- NpO2CO3-   NpO2(CO3)2 NpO2(CO3)3 H2PO4-     HPO4=
B4O5(OH)4=       0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
                 PO4=-
B4O5(OH)4=       0.

                 Am(CO3)2-  Am(CO3)3=- ClO4-      NpO2(OH)2- NpO2CO3-   NpO2(CO3)2 NpO2(CO3)3 H2PO4-     HPO4=      PO4=-
Br-              0.         0.         0.         0.         0.         0.         0.         0.         0.         0.

                 Am(CO3)3=- ClO4-      NpO2(OH)2- NpO2CO3-   NpO2(CO3)2 NpO2(CO3)3 H2PO4-     HPO4=      PO4=-
Am(CO3)2-        0.         0.         0.         0.         0.         0.         0.         0.         0.

                 ClO4-      NpO2(OH)2- NpO2CO3-   NpO2(CO3)2 NpO2(CO3)3 H2PO4-     HPO4=      PO4=-
Am(CO3)3=-       0.         0.         0.         0.         0.         0.         0.         0.

                 NpO2(OH)2- NpO2CO3-   NpO2(CO3)2 NpO2(CO3)3 H2PO4-     HPO4=      PO4=-
ClO4-            0.         0.         0.         0.         0.         0.         0.

                 NpO2CO3-   NpO2(CO3)2 NpO2(CO3)3 H2PO4-     HPO4=      PO4=-
NpO2(OH)2-       0.         0.         0.         0.         0.         0.

                 NpO2(CO3)2 NpO2(CO3)3 H2PO4-     HPO4=      PO4=-
NpO2CO3-         0.         0.         0.         0.         0.

                 NpO2(CO3)3 H2PO4-     HPO4=      PO4=-
NpO2(CO3)2=-     0.         0.         0.         0.

                 H2PO4-     HPO4=      PO4=-
NpO2(CO3)3==-    0.         0.         0.

                 HPO4=      PO4=-
H2PO4-           0.         0.
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391
392                        PO4=-
393   HPO4=                0.
394
395
396   Cation-Cation-Anion Ternary Interactions
397
398                                  Cl-        SO4=       HSO4-      OH-        HCO3-      CO3=       B(OH)4-    B3O3(OH)4  B4O5(OH)4  Br-
399   Na+          K+                -0.00180   -0.01000   0.         0.         -0.00300   0.00300    0.         0.         0.         0.
400   Na+          Ca++              -0.00700   -0.05500   0.         0.         0.         0.         0.         0.         0.         0.
401   Na+          Mg++              -0.01200   -0.01500   0.         0.         0.         0.         0.         0.         0.         0.
402   Na+          MgOH+             0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
403   Na+          H+                -0.00400   0.         -0.01290   0.         0.         0.         0.         0.         0.         0.
404   Na+          MgB(OH)4+         0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
405   Na+          CaB(OH)4+         0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
406   Na+          Am+++             0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
407   Na+          AmCO3+            0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
408   Na+          Th++++            0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
409   Na+          UO2++             0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
410   Na+          NpO2+             0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
411   K+           Ca++              -0.02500   0.         0.         0.         0.         0.         0.         0.         0.         0.
412   K+           Mg++              -0.02200   -0.04800   0.         0.         0.         0.         0.         0.         0.         0.
413   K+           MgOH+             0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
414   K+           H+                -0.01100   0.19700    -0.02650   0.         0.         0.         0.         0.         0.         0.
415   K+           MgB(OH)4+         0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
416   K+           CaB(OH)4+         0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
417   K+           Am+++             0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
418   K+           AmCO3+            0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
419   K+           Th++++            0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
420   K+           UO2++             0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
421   K+           NpO2+             0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
422   Ca++         Mg++              -0.01200   0.02400    0.         0.         0.         0.         0.         0.         0.         0.
423   Ca++         MgOH+             0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
424   Ca++         H+                -0.01500   0.         0.         0.         0.         0.         0.         0.         0.         0.
425   Ca++         MgB(OH)4+         0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
426   Ca++         CaB(OH)4+         0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
427   Ca++         Am+++             0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
428   Ca++         AmCO3+            0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
429   Ca++         Th++++            0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
430   Ca++         UO2++             0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
431   Ca++         NpO2+             0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
432   Mg++         MgOH+             0.02800    0.         0.         0.         0.         0.         0.         0.         0.         0.
433   Mg++         H+                -0.01100   0.         -0.01780   0.         0.         0.         0.         0.         0.         0.
434   Mg++         MgB(OH)4+         0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
435   Mg++         CaB(OH)4+         0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
436   Mg++         Am+++             0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
437   Mg++         AmCO3+            0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
438   Mg++         Th++++            0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
439   Mg++         UO2++             0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
440   Mg++         NpO2+             0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
441   MgOH+        H+                0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
442   MgOH+        MgB(OH)4+         0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
443   MgOH+        CaB(OH)4+         0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
444   MgOH+        Am+++             0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
445   MgOH+        AmCO3+            0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
446   MgOH+        Th++++            0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
447   MgOH+        UO2++             0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
448   MgOH+        NpO2+             0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
449   H+           MgB(OH)4+         0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
450   H+           CaB(OH)4+         0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
451   H+           Am+++             0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
452   H+           AmCO3+            0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
453   H+           Th++++            0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
454   H+           UO2++             0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
455   H+           NpO2+             0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
456   MgB(OH)4+    CaB(OH)4+         0.         0.         0.         0.         0.         0.         0.         0.         0.         0.
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MgB(OH)4+     Am+++         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
MgB(OH)4+     AmCO3+        0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
MgB(OH)4+     Th++++        0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
MgB(OH)4+     UO2++         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
MgB(OH)4+     NpO2+         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
CaB(OH)4+     Am+++         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
CaB(OH)4+     AmCO3+        0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
CaB(OH)4+     Th++++        0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
CaB(OH)4+     UO2++         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
CaB(OH)4+     NpO2+         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Am+++         AmCO3+        0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Am+++         Th++++        0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Am+++         UO2++         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Am+++         NpO2+         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
AmCO3+        Th++++        0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
AmCO3+        UO2++         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
AmCO3+        NpO2+         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Th++++        UO2++         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Th++++        NpO2+         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
UO2++         NpO2+         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.

                            Am(CO3)2- Am(CO3)3= ClO4-  NpO2(OH)2 NpO2CO3- NpO2(CO3) NpO2(CO3) H2PO4-  HPO4=    PO4=-
Na+           K+            0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Na+           Ca++          0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Na+           Mg++          0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Na+           MgOH+         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Na+           H+            0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Na+           MgB(OH)4+     0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Na+           CaB(OH)4+     0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Na+           Am+++         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Na+           AmCO3+        0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Na+           Th++++        0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Na+           UO2++         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Na+           NpO2+         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
K+            Ca++          0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
K+            Mg++          0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
K+            MgOH+         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
K+            H+            0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
K+            MgB(OH)4+     0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
K+            CaB(OH)4+     0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
K+            Am+++         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
K+            AmCO3+        0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
K+            Th++++        0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
K+            UO2++         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
K+            NpO2+         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Ca++          Mg++          0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Ca++          MgOH+         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Ca++          H+            0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Ca++          MgB(OH)4+     0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Ca++          CaB(OH)4+     0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Ca++          Am+++         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Ca++          AmCO3+        0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Ca++          Th++++        0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Ca++          UO2++         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Ca++          NpO2+         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Mg++          MgOH+         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Mg++          H+            0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Mg++          MgB(OH)4+     0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Mg++          CaB(OH)4+     0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Mg++          Am+++         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Mg++          AmCO3+        0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Mg++          Th++++        0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Mg++          UO2++         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
Mg++          NpO2+         0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
MgOH+         H+            0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
MgOH+         MgB(OH)4+     0.       0.       0.       0.       0.       0.       0.       0.       0.       0.
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MgOH+           CaB(OH)4+      0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
MgOH+           Am+++          0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
MgOH+           AmCO3+         0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
MgOH+           Th++++         0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
MgOH+           UO2++          0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
MgOH+           NpO2+          0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
H+              MgB(OH)4+      0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
H+              CaB(OH)4+      0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
H+              Am+++          0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
H+              AmCO3+         0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
H+              Th++++         0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
H+              UO2++          0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
H+              NpO2+          0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
MgB(OH)4+       CaB(OH)4+      0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
MgB(OH)4+       Am+++          0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
MgB(OH)4+       AmCO3+         0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
MgB(OH)4+       Th++++         0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
MgB(OH)4+       UO2++          0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
MgB(OH)4+       NpO2+          0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
CaB(OH)4+       Am+++          0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
CaB(OH)4+       AmCO3+         0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
CaB(OH)4+       Th++++         0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
CaB(OH)4+       UO2++          0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
CaB(OH)4+       NpO2+          0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
Am+++           AmCO3+         0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
Am+++           Th++++         0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
Am+++           UO2++          0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
Am+++           NpO2+          0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
AmCO3+          Th++++         0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
AmCO3+          UO2++          0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
AmCO3+          NpO2+          0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
Th++++          UO2++          0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
Th++++          NpO2+          0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
UO2++           NpO2+          0.          0.          0.          0.          0.          0.          0.          0.          0.          0.

Anion-Anion-Cation Ternary Interactions

                               Na+         K+          Ca++        Mg++        MgOH+       H+          MgB(OH)4+   CaB(OH)4+   Am+++       AmCO3+
Cl-             SO4=           0.00140     0.          -0.01800    -0.00400    0.          0.          0.          0.          0.          0.
Cl-             HSO4-          -0.00600    0.          0.          0.          0.          0.01300     0.          0.          0.          0.
Cl-             OH-            -0.00600    -0.00600    -0.02500    0.          0.          0.          0.          0.          0.          0.
Cl-             HCO3-          -0.01500    0.          0.          -0.09600    0.          0.          0.          0.          0.          0.
Cl-             CO3=           0.00850     0.00400     0.          0.          0.          0.          0.          0.          0.          0.
Cl-             B(OH)4-        -0.00730    0.          0.          0.          0.          0.          0.          0.          0.          0.
Cl-             B3O3(OH)4-     -0.02400    0.          0.          0.          0.          0.          0.          0.          0.          0.
Cl-             B4O5(OH)4=     0.02600     0.          0.          0.          0.          0.          0.          0.          0.          0.
Cl-             Br-            0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
Cl-             Am(CO3)2-      0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
Cl-             Am(CO3)3=-     0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
Cl-             ClO4-          0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
Cl-             NpO2(OH)2-     0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
Cl-             NpO2CO3-       0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
Cl-             NpO2(CO3)2=-   0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
Cl-             NpO2(CO3)3==-  0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
Cl-             H2PO4-         0.          -0.01000    0.          0.          0.          0.          0.          0.          0.          0.
Cl-             HPO4=          0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
Cl-             PO4=-          0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
SO4=            HSO4-          -0.00940    -0.06770    0.          -0.04250    0.          0.          0.          0.          0.          0.
SO4=            OH-            -0.00900    -0.05000    0.          0.          0.          0.          0.          0.          0.          0.
SO4=            HCO3-          -0.00500    0.          0.          -0.16100    0.          0.          0.          0.          0.          0.
SO4=            CO3=           -0.00500    -0.00900    0.          0.          0.          0.          0.          0.          0.          0.
SO4=            B(OH)4-        0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
SO4=            B3O3(OH)4-     0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
SO4=            B4O5(OH)4=     0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
SO4=            Br-            0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
SO4=            Am(CO3)2-      0.          0.          0.          0.          0.          0.          0.          0.          0.          0.
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589	SO4=	Am(CO3)3=-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
590	SO4=	ClO4-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
591	SO4=	NpO2(OH)2-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
592	SO4=	NpO2CO3-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
593	SO4=	NpO2(CO3)2=-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
594	SO4=	NpO2(CO3)3==-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
595	SO4=	H2PO4-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
596	SO4=	HPO4=	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
597	SO4=	PO4=-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
598	HSO4-	OH-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
599	HSO4-	HCO3-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
600	HSO4-	CO3=	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
601	HSO4-	B(OH)4-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
602	HSO4-	B3O3(OH)4-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
603	HSO4-	B4O5(OH)4=	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
604	HSO4-	Br-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
605	HSO4-	Am(CO3)2-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
606	HSO4-	Am(CO3)3=-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
607	HSO4-	ClO4-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
608	HSO4-	NpO2(OH)2-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
609	HSO4-	NpO2CO3-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
610	HSO4-	NpO2(CO3)2=-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
611	HSO4-	NpO2(CO3)3==-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
612	HSO4-	H2PO4-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
613	HSO4-	HPO4=	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
614	HSO4-	PO4=-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
615	OH-	HCO3-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
616	OH-	CO3=	-0.01700	-0.01000	0.	0.	0.	0.	0.	0.	0.	0.
617	OH-	B(OH)4-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
618	OH-	B3O3(OH)4-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
619	OH-	B4O5(OH)4=	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
620	OH-	Br-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
621	OH-	Am(CO3)2-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
622	OH-	Am(CO3)3=-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
623	OH-	ClO4-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
624	OH-	NpO2(OH)2-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
625	OH-	NpO2CO3-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
626	OH-	NpO2(CO3)2=-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
627	OH-	NpO2(CO3)3==-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
628	OH-	H2PO4-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
629	OH-	HPO4=	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
630	OH-	PO4=-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
631	HCO3-	CO3=	0.00200	0.01200	0.	0.	0.	0.	0.	0.	0.	0.
632	HCO3-	B(OH)4-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
633	HCO3-	B3O3(OH)4-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
634	HCO3-	B4O5(OH)4=	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
635	HCO3-	Br-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
636	HCO3-	Am(CO3)2-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
637	HCO3-	Am(CO3)3=-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
638	HCO3-	ClO4-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
639	HCO3-	NpO2(OH)2-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
640	HCO3-	NpO2CO3-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
641	HCO3-	NpO2(CO3)2=-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
642	HCO3-	NpO2(CO3)3==-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
643	HCO3-	H2PO4-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
644	HCO3-	HPO4=	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
645	HCO3-	PO4=-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
646	CO3=	B(OH)4-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
647	CO3=	B3O3(OH)4-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
648	CO3=	B4O5(OH)4=	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
649	CO3=	Br-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
650	CO3=	Am(CO3)2-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
651	CO3=	Am(CO3)3=-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
652	CO3=	ClO4-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
653	CO3=	NpO2(OH)2-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.
654	CO3=	NpO2CO3-	0.	0.	0.	0.	0.	0.	0.	0.	0.	0.

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655  CO3=          NpO2(CO3)2=-   0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
656  CO3=          NpO2(CO3)3==-  0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
657  CO3=          H2PO4-         0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
658  CO3=          HPO4=          0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
659  CO3=          PO4=-          0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
660  B(OH)4-       B3O3(OH)4-     0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
661  B(OH)4-       B4O5(OH)4=     0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
662  B(OH)4-       Br-            0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
663  B(OH)4-       Am(CO3)2-      0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
664  B(OH)4-       Am(CO3)3=-     0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
665  B(OH)4-       ClO4-          0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
666  B(OH)4-       NpO2(OH)2-     0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
667  B(OH)4-       NpO2CO3-       0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
668  B(OH)4-       NpO2(CO3)2=-   0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
669  B(OH)4-       NpO2(CO3)3==-  0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
670  B(OH)4-       H2PO4-         0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
671  B(OH)4-       HPO4=          0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
672  B(OH)4-       PO4=-          0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
673  B3O3(OH)4-    B4O5(OH)4=     0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
674  B3O3(OH)4-    Br-            0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
675  B3O3(OH)4-    Am(CO3)2-      0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
676  B3O3(OH)4-    Am(CO3)3=-     0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
677  B3O3(OH)4-    ClO4-          0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
678  B3O3(OH)4-    NpO2(OH)2-     0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
679  B3O3(OH)4-    NpO2CO3-       0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
680  B3O3(OH)4-    NpO2(CO3)2=-   0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
681  B3O3(OH)4-    NpO2(CO3)3==-  0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
682  B3O3(OH)4-    H2PO4-         0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
683  B3O3(OH)4-    HPO4=          0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
684  B3O3(OH)4-    PO4=-          0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
685  B4O5(OH)4=    Br-            0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
686  B4O5(OH)4=    Am(CO3)2-      0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
687  B4O5(OH)4=    Am(CO3)3=-     0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
688  B4O5(OH)4=    ClO4-          0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
689  B4O5(OH)4=    NpO2(OH)2-     0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
690  B4O5(OH)4=    NpO2CO3-       0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
691  B4O5(OH)4=    NpO2(CO3)2=-   0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
692  B4O5(OH)4=    NpO2(CO3)3==-  0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
693  B4O5(OH)4=    H2PO4-         0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
694  B4O5(OH)4=    HPO4=          0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
695  B4O5(OH)4=    PO4=-          0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
696  Br-           Am(CO3)2-      0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
697  Br-           Am(CO3)3=-     0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
698  Br-           ClO4-          0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
699  Br-           NpO2(OH)2-     0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
700  Br-           NpO2CO3-       0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
701  Br-           NpO2(CO3)2=-   0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
702  Br-           NpO2(CO3)3==-  0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
703  Br-           H2PO4-         0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
704  Br-           HPO4=          0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
705  Br-           PO4=-          0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
706  Am(CO3)2-     Am(CO3)3=-     0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
707  Am(CO3)2-     ClO4-          0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
708  Am(CO3)2-     NpO2(OH)2-     0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
709  Am(CO3)2-     NpO2CO3-       0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
710  Am(CO3)2-     NpO2(CO3)2=-   0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
711  Am(CO3)2-     NpO2(CO3)3==-  0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
712  Am(CO3)2-     H2PO4-         0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
713  Am(CO3)2-     HPO4=          0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
714  Am(CO3)2-     PO4=-          0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
715  Am(CO3)3=-    ClO4-          0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
716  Am(CO3)3=-    NpO2(OH)2-     0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
717  Am(CO3)3=-    NpO2CO3-       0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
718  Am(CO3)3=-    NpO2(CO3)2=-   0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
719  Am(CO3)3=-    NpO2(CO3)3==-  0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
720  Am(CO3)3=-    H2PO4-         0.  0.  0.  0.  0.  0.  0.  0.  0.  0.
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Am(CO3)3=-      HPO4=           0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
Am(CO3)3=-      PO4=-           0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
ClO4-           NpO2(OH)2-      0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
ClO4-           NpO2CO3-        0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
ClO4-           NpO2(CO3)2=-    0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
ClO4-           NpO2(CO3)3==-   0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
ClO4-           H2PO4-          0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
ClO4-           HPO4=           0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
ClO4-           PO4=-           0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
NpO2(OH)2-      NpO2CO3-        0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
NpO2(OH)2-      NpO2(CO3)2=-    0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
NpO2(OH)2-      NpO2(CO3)3==-   0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
NpO2(OH)2-      H2PO4-          0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
NpO2(OH)2-      HPO4=           0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
NpO2(OH)2-      PO4=-           0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
NpO2CO3-        NpO2(CO3)2=-    0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
NpO2CO3-        NpO2(CO3)3==-   0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
NpO2CO3-        H2PO4-          0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
NpO2CO3-        HPO4=           0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
NpO2CO3-        PO4=-           0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
NpO2(CO3)2=-    NpO2(CO3)3==-   0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
NpO2(CO3)2=-    H2PO4-          0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
NpO2(CO3)2=-    HPO4=           0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
NpO2(CO3)2=-    PO4=-           0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
NpO2(CO3)3==-   H2PO4-          0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
NpO2(CO3)3==-   HPO4=           0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
NpO2(CO3)3==-   PO4=-           0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
H2PO4-          HPO4=           0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
H2PO4-          PO4=-           0.     0.     0.     0.     0.     0.     0.     0.     0.     0.
HPO4=           PO4=-           0.     0.     0.     0.     0.     0.     0.     0.     0.     0.

                                Th++++ UO2++  NpO2+
Cl-             SO4=            0.     0.     0.
Cl-             HSO4-           0.     0.     0.
Cl-             OH-             0.     0.     0.
Cl-             HCO3-           0.     0.     0.
Cl-             CO3=            0.     0.     0.
Cl-             B(OH)4-         0.     0.     0.
Cl-             B3O3(OH)4-      0.     0.     0.
Cl-             B4O5(OH)4=      0.     0.     0.
Cl-             Br-             0.     0.     0.
Cl-             Am(CO3)2-       0.     0.     0.
Cl-             Am(CO3)3=-      0.     0.     0.
Cl-             ClO4-           0.     0.     0.
Cl-             NpO2(OH)2-      0.     0.     0.
Cl-             NpO2CO3-        0.     0.     0.
Cl-             NpO2(CO3)2=-    0.     0.     0.
Cl-             NpO2(CO3)3==-   0.     0.     0.
Cl-             H2PO4-          0.     0.     0.
Cl-             HPO4=           0.     0.     0.
Cl-             PO4=-           0.     0.     0.
SO4=            HSO4-           0.     0.     0.
SO4=            OH-             0.     0.     0.
SO4=            HCO3-           0.     0.     0.
SO4=            CO3=            0.     0.     0.
SO4=            B(OH)4-         0.     0.     0.
SO4=            B3O3(OH)4-      0.     0.     0.
SO4=            B4O5(OH)4=      0.     0.     0.
SO4=            Br-             0.     0.     0.
SO4=            Am(CO3)2-       0.     0.     0.
SO4=            Am(CO3)3=-      0.     0.     0.
SO4=            ClO4-           0.     0.     0.
SO4=            NpO2(OH)2-      0.     0.     0.
SO4=            NpO2CO3-        0.     0.     0.
SO4=            NpO2(CO3)2=-    0.     0.     0.
SO4=            NpO2(CO3)3==-   0.     0.     0.
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787   SO4=      H2PO4-           0.     0.     0.
788   SO4=      HPO4=            0.     0.     0.
789   SO4=      PO4=-            0.     0.     0.
790   HSO4-     OH-              0.     0.     0.
791   HSO4-     HCO3-            0.     0.     0.
792   HSO4-     CO3=             0.     0.     0.
793   HSO4-     B(OH)4-          0.     0.     0.
794   HSO4-     B3O3(OH)4-       0.     0.     0.
795   HSO4-     B4O5(OH)4=       0.     0.     0.
796   HSO4-     Br-              0.     0.     0.
797   HSO4-     Am(CO3)2-        0.     0.     0.
798   HSO4-     Am(CO3)3=-       0.     0.     0.
799   HSO4-     ClO4-            0.     0.     0.
800   HSO4-     NpO2(OH)2-       0.     0.     0.
801   HSO4-     NpO2CO3-         0.     0.     0.
802   HSO4-     NpO2(CO3)2=-     0.     0.     0.
803   HSO4-     NpO2(CO3)3==-    0.     0.     0.
804   HSO4-     H2PO4-           0.     0.     0.
805   HSO4-     HPO4=            0.     0.     0.
806   HSO4-     PO4=-            0.     0.     0.
807   OH-       HCO3-            0.     0.     0.
808   OH-       CO3=             0.     0.     0.
809   OH-       B(OH)4-          0.     0.     0.
810   OH-       B3O3(OH)4-       0.     0.     0.
811   OH-       B4O5(OH)4=       0.     0.     0.
812   OH-       Br-              0.     0.     0.
813   OH-       Am(CO3)2-        0.     0.     0.
814   OH-       Am(CO3)3=-       0.     0.     0.
815   OH-       ClO4-            0.     0.     0.
816   OH-       NpO2(OH)2-       0.     0.     0.
817   OH-       NpO2CO3-         0.     0.     0.
818   OH-       NpO2(CO3)2=-     0.     0.     0.
819   OH-       NpO2(CO3)3==-    0.     0.     0.
820   OH-       H2PO4-           0.     0.     0.
821   OH-       HPO4=            0.     0.     0.
822   OH-       PO4=-            0.     0.     0.
823   HCO3-     CO3=             0.     0.     0.
824   HCO3-     B(OH)4-          0.     0.     0.
825   HCO3-     B3O3(OH)4-       0.     0.     0.
826   HCO3-     B4O5(OH)4=       0.     0.     0.
827   HCO3-     Br-              0.     0.     0.
828   HCO3-     Am(CO3)2-        0.     0.     0.
829   HCO3-     Am(CO3)3=-       0.     0.     0.
830   HCO3-     ClO4-            0.     0.     0.
831   HCO3-     NpO2(OH)2-       0.     0.     0.
832   HCO3-     NpO2CO3-         0.     0.     0.
833   HCO3-     NpO2(CO3)2=-     0.     0.     0.
834   HCO3-     NpO2(CO3)3==-    0.     0.     0.
835   HCO3-     H2PO4-           0.     0.     0.
836   HCO3-     HPO4=            0.     0.     0.
837   HCO3-     PO4=-            0.     0.     0.
838   CO3=      B(OH)4-          0.     0.     0.
839   CO3=      B3O3(OH)4-       0.     0.     0.
840   CO3=      B4O5(OH)4=       0.     0.     0.
841   CO3=      Br-              0.     0.     0.
842   CO3=      Am(CO3)2-        0.     0.     0.
843   CO3=      Am(CO3)3=-       0.     0.     0.
844   CO3=      ClO4-            0.     0.     0.
845   CO3=      NpO2(OH)2-       0.     0.     0.
846   CO3=      NpO2CO3-         0.     0.     0.
847   CO3=      NpO2(CO3)2=-     0.     0.     0.
848   CO3=      NpO2(CO3)3==-    0.     0.     0.
849   CO3=      H2PO4-           0.     0.     0.
850   CO3=      HPO4=            0.     0.     0.
851   CO3=      PO4=-            0.     0.     0.
852   B(OH)4-   B3O3(OH)4-       0.     0.     0.
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853   B(OH)4-        B4O5(OH)4=       0.         0.         0.
854   B(OH)4-        Br-              0.         0.         0.
855   B(OH)4-        Am(CO3)2-        0.         0.         0.
856   B(OH)4-        Am(CO3)3=-       0.         0.         0.
857   B(OH)4-        ClO4-            0.         0.         0.
858   B(OH)4-        NpO2(OH)2-       0.         0.         0.
859   B(OH)4-        NpO2CO3-         0.         0.         0.
860   B(OH)4-        NpO2(CO3)2=-     0.         0.         0.
861   B(OH)4-        NpO2(CO3)3==-    0.         0.         0.
862   B(OH)4-        H2PO4-           0.         0.         0.
863   B(OH)4-        HPO4=            0.         0.         0.
864   B(OH)4-        PO4=-            0.         0.         0.
865   B3O3(OH)4-     B4O5(OH)4=       0.         0.         0.
866   B3O3(OH)4-     Br-              0.         0.         0.
867   B3O3(OH)4-     Am(CO3)2-        0.         0.         0.
868   B3O3(OH)4-     Am(CO3)3=-       0.         0.         0.
869   B3O3(OH)4-     ClO4-            0.         0.         0.
870   B3O3(OH)4-     NpO2(OH)2-       0.         0.         0.
871   B3O3(OH)4-     NpO2CO3-         0.         0.         0.
872   B3O3(OH)4-     NpO2(CO3)2=-     0.         0.         0.
873   B3O3(OH)4-     NpO2(CO3)3==-    0.         0.         0.
874   B3O3(OH)4-     H2PO4-           0.         0.         0.
875   B3O3(OH)4-     HPO4=            0.         0.         0.
876   B3O3(OH)4-     PO4=-            0.         0.         0.
877   B4O5(OH)4=     Br-              0.         0.         0.
878   B4O5(OH)4=     Am(CO3)2-        0.         0.         0.
879   B4O5(OH)4=     Am(CO3)3=-       0.         0.         0.
880   B4O5(OH)4=     ClO4-            0.         0.         0.
881   B4O5(OH)4=     NpO2(OH)2-       0.         0.         0.
882   B4O5(OH)4=     NpO2CO3-         0.         0.         0.
883   B4O5(OH)4=     NpO2(CO3)2=-     0.         0.         0.
884   B4O5(OH)4=     NpO2(CO3)3==-    0.         0.         0.
885   B4O5(OH)4=     H2PO4-           0.         0.         0.
886   B4O5(OH)4=     HPO4=            0.         0.         0.
887   B4O5(OH)4=     PO4=-            0.         0.         0.
888   Br-            Am(CO3)2-        0.         0.         0.
889   Br-            Am(CO3)3=-       0.         0.         0.
890   Br-            ClO4-            0.         0.         0.
891   Br-            NpO2(OH)2-       0.         0.         0.
892   Br-            NpO2CO3-         0.         0.         0.
893   Br-            NpO2(CO3)2=-     0.         0.         0.
894   Br-            NpO2(CO3)3==-    0.         0.         0.
895   Br-            H2PO4-           0.         0.         0.
896   Br-            HPO4=            0.         0.         0.
897   Br-            PO4=-            0.         0.         0.
898   Am(CO3)2-      Am(CO3)3=-       0.         0.         0.
899   Am(CO3)2-      ClO4-            0.         0.         0.
900   Am(CO3)2-      NpO2(OH)2-       0.         0.         0.
901   Am(CO3)2-      NpO2CO3-         0.         0.         0.
902   Am(CO3)2-      NpO2(CO3)2=-     0.         0.         0.
903   Am(CO3)2-      NpO2(CO3)3==-    0.         0.         0.
904   Am(CO3)2-      H2PO4-           0.         0.         0.
905   Am(CO3)2-      HPO4=            0.         0.         0.
906   Am(CO3)2-      PO4=-            0.         0.         0.
907   Am(CO3)3=-     ClO4-            0.         0.         0.
908   Am(CO3)3=-     NpO2(OH)2-       0.         0.         0.
909   Am(CO3)3=-     NpO2CO3-         0.         0.         0.
910   Am(CO3)3=-     NpO2(CO3)2=-     0.         0.         0.
911   Am(CO3)3=-     NpO2(CO3)3==-    0.         0.         0.
912   Am(CO3)3=-     H2PO4-           0.         0.         0.
913   Am(CO3)3=-     HPO4=            0.         0.         0.
914   Am(CO3)3=-     PO4=-            0.         0.         0.
915   ClO4-          NpO2(OH)2-       0.         0.         0.
916   ClO4-          NpO2CO3-         0.         0.         0.
917   ClO4-          NpO2(CO3)2=-     0.         0.         0.
918   ClO4-          NpO2(CO3)3==-    0.         0.         0.
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ClO4-           H2PO4-          0.         0.         0.
ClO4-           HPO4=           0.         0.         0.
ClO4-           PO4=-           0.         0.         0.
NpO2(OH)2-      NpO2CO3-        0.         0.         0.
NpO2(OH)2-      NpO2(CO3)2=-    0.         0.         0.
NpO2(OH)2-      NpO2(CO3)3==-   0.         0.         0.
NpO2(OH)2-      H2PO4-          0.         0.         0.
NpO2(OH)2-      HPO4=           0.         0.         0.
NpO2(OH)2-      PO4=-           0.         0.         0.
NpO2CO3-        NpO2(CO3)2=-    0.         0.         0.
NpO2CO3-        NpO2(CO3)3==-   0.         0.         0.
NpO2CO3-        H2PO4-          0.         0.         0.
NpO2CO3-        HPO4=           0.         0.         0.
NpO2CO3-        PO4=-           0.         0.         0.
NpO2(CO3)2=-    NpO2(CO3)3==-   0.         0.         0.
NpO2(CO3)2=-    H2PO4-          0.         0.         0.
NpO2(CO3)2=-    HPO4=           0.         0.         0.
NpO2(CO3)2=-    PO4=-           0.         0.         0.
NpO2(CO3)3==-   H2PO4-          0.         0.         0.
NpO2(CO3)3==-   HPO4=           0.         0.         0.
NpO2(CO3)3==-   PO4=-           0.         0.         0.
H2PO4-          HPO4=           0.         0.         0.
H2PO4-          PO4=-           0.         0.         0.
HPO4=           PO4=-           0.         0.         0.

Neutral-Ion Binary Interactions

                CO2(aq)    CaCO3(aq)  MgCO3(aq)  B(OH)3(aq) NpO2OH(aq) H3PO4(aq)
Na+              0.10000   0.         0.         -0.09700   0.          0.
K+               0.05100   0.         0.         -0.14000   0.         -0.07000
Ca++             0.18300   0.         0.          0.        0.          0.
Mg++             0.18300   0.         0.          0.        0.          0.
MgOH+            0.        0.         0.          0.        0.          0.
H+               0.        0.         0.          0.        0.          0.29000
MgB(OH)4+        0.        0.         0.          0.        0.          0.
CaB(OH)4+        0.        0.         0.          0.        0.          0.
Am+++            0.        0.         0.          0.        0.          0.
AmCO3+           0.        0.         0.          0.        0.          0.
Th++++           0.        0.         0.          0.        0.          0.
UO2++            0.        0.         0.          0.        0.          0.
NpO2+            0.        0.         0.          0.        0.          0.

                CO2(aq)    CaCO3(aq)  MgCO3(aq)  B(OH)3(aq) NpO2OH(aq) H3PO4(aq)
Cl-             -0.00500   0.         0.          0.09100   0.          0.
SO4=             0.09700   0.         0.          0.01800   0.          0.
HSO4-           -0.00300   0.         0.          0.        0.          0.
OH-              0.        0.         0.          0.        0.          0.
HCO3-            0.        0.         0.          0.        0.          0.
CO3=             0.        0.         0.          0.        0.          0.
B(OH)4-          0.        0.         0.          0.        0.          0.
B3O3(OH)4-       0.        0.         0.         -0.20000   0.          0.
B4O5(OH)4=       0.        0.         0.          0.        0.          0.
Br-              0.        0.         0.          0.        0.          0.
Am(CO3)2-        0.        0.         0.          0.        0.          0.
Am(CO3)3=-       0.        0.         0.          0.        0.          0.
ClO4-            0.        0.         0.          0.        0.          0.
NpO2(OH)2-       0.        0.         0.          0.        0.          0.
NpO2CO3-         0.        0.         0.          0.        0.          0.
NpO2(CO3)2=-     0.        0.         0.          0.        0.          0.
NpO2(CO3)3==-    0.        0.         0.          0.        0.          0.
H2PO4-           0.        0.         0.          0.        0.         -0.40000
HPO4=            0.        0.         0.          0.        0.          0.
PO4=-            0.        0.         0.          0.        0.          0.

Neutral-Cation-Anion Ternary Interactions
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 985                                 CO2(aq)    CaCO3(aq) MgCO3(aq) B(OH)3(aq NpO2OH(aq H3PO4(aq)
 986  Na+          Cl-               0.         0.        0.        0.        0.        0.
 987  Na+          SO4=              0.         0.        0.        0.04600   0.        0.
 988  Na+          HSO4-             0.         0.        0.        0.        0.        0.
 989  Na+          OH-               0.         0.        0.        0.        0.        0.
 990  Na+          HCO3-             0.         0.        0.        0.        0.        0.
 991  Na+          CO3=              0.         0.        0.        0.        0.        0.
 992  Na+          B(OH)4-           0.         0.        0.        0.        0.        0.
 993  Na+          B3O3(OH)4-        0.         0.        0.        0.        0.        0.
 994  Na+          B4O5(OH)4=        0.         0.        0.        0.        0.        0.
 995  Na+          Br-               0.         0.        0.        0.        0.        0.
 996  Na+          Am(CO3)2-         0.         0.        0.        0.        0.        0.
 997  Na+          Am(CO3)3=-        0.         0.        0.        0.        0.        0.
 998  Na+          ClO4-             0.         0.        0.        0.        0.        0.
 999  Na+          NpO2(OH)2-        0.         0.        0.        0.        0.        0.
1000  Na+          NpO2CO3-          0.         0.        0.        0.        0.        0.
1001  Na+          NpO2(CO3)2=-      0.         0.        0.        0.        0.        0.
1002  Na+          NpO2(CO3)3==-     0.         0.        0.        0.        0.        0.
1003  Na+          H2PO4-            0.         0.        0.        0.        0.        0.
1004  Na+          HPO4=             0.         0.        0.        0.        0.        0.
1005  Na+          PO4=-             0.         0.        0.        0.        0.        0.
1006  K+           Cl-               0.         0.        0.        0.        0.        0.
1007  K+           SO4=              0.         0.        0.        0.        0.        0.
1008  K+           HSO4-             0.         0.        0.        0.        0.        0.
1009  K+           OH-               0.         0.        0.        0.        0.        0.
1010  K+           HCO3-             0.         0.        0.        0.        0.        0.
1011  K+           CO3=              0.         0.        0.        0.        0.        0.
1012  K+           B(OH)4-           0.         0.        0.        0.        0.        0.
1013  K+           B3O3(OH)4-        0.         0.        0.        0.        0.        0.
1014  K+           B4O5(OH)4=        0.         0.        0.        0.        0.        0.
1015  K+           Br-               0.         0.        0.        0.        0.        0.
1016  K+           Am(CO3)2-         0.         0.        0.        0.        0.        0.
1017  K+           Am(CO3)3=-        0.         0.        0.        0.        0.        0.
1018  K+           ClO4-             0.         0.        0.        0.        0.        0.
1019  K+           NpO2(OH)2-        0.         0.        0.        0.        0.        0.
1020  K+           NpO2CO3-          0.         0.        0.        0.        0.        0.
1021  K+           NpO2(CO3)2=-      0.         0.        0.        0.        0.        0.
1022  K+           NpO2(CO3)3==-     0.         0.        0.        0.        0.        0.
1023  K+           H2PO4-            0.         0.        0.        0.        0.        0.
1024  K+           HPO4=             0.         0.        0.        0.        0.        0.
1025  K+           PO4=-             0.         0.        0.        0.        0.        0.
1026  Ca++         Cl-               0.         0.        0.        0.        0.        0.
1027  Ca++         SO4=              0.         0.        0.        0.        0.        0.
1028  Ca++         HSO4-             0.         0.        0.        0.        0.        0.
1029  Ca++         OH-               0.         0.        0.        0.        0.        0.
1030  Ca++         HCO3-             0.         0.        0.        0.        0.        0.
1031  Ca++         CO3=              0.         0.        0.        0.        0.        0.
1032  Ca++         B(OH)4-           0.         0.        0.        0.        0.        0.
1033  Ca++         B3O3(OH)4-        0.         0.        0.        0.        0.        0.
1034  Ca++         B4O5(OH)4=        0.         0.        0.        0.        0.        0.
1035  Ca++         Br-               0.         0.        0.        0.        0.        0.
1036  Ca++         Am(CO3)2-         0.         0.        0.        0.        0.        0.
1037  Ca++         Am(CO3)3=-        0.         0.        0.        0.        0.        0.
1038  Ca++         ClO4-             0.         0.        0.        0.        0.        0.
1039  Ca++         NpO2(OH)2-        0.         0.        0.        0.        0.        0.
1040  Ca++         NpO2CO3-          0.         0.        0.        0.        0.        0.
1041  Ca++         NpO2(CO3)2=-      0.         0.        0.        0.        0.        0.
1042  Ca++         NpO2(CO3)3==-     0.         0.        0.        0.        0.        0.
1043  Ca++         H2PO4-            0.         0.        0.        0.        0.        0.
1044  Ca++         HPO4=             0.         0.        0.        0.        0.        0.
1045  Ca++         PO4=-             0.         0.        0.        0.        0.        0.
1046  Mg++         Cl-               0.         0.        0.        0.        0.        0.
1047  Mg++         SO4=              0.         0.        0.        0.        0.        0.
1048  Mg++         HSO4-             0.         0.        0.        0.        0.        0.
1049  Mg++         OH-               0.         0.        0.        0.        0.        0.
1050  Mg++         HCO3-             0.         0.        0.        0.        0.        0.
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1051   Mg++        CO3=             0.    0.    0.    0.          0.    0.
1052   Mg++        B(OH)4-          0.    0.    0.    0.          0.    0.
1053   Mg++        B3O3(OH)4-       0.    0.    0.    0.          0.    0.
1054   Mg++        B4O5(OH)4=       0.    0.    0.    0.          0.    0.
1055   Mg++        Br-              0.    0.    0.    0.          0.    0.
1056   Mg++        Am(CO3)2-        0.    0.    0.    0.          0.    0.
1057   Mg++        Am(CO3)3=-       0.    0.    0.    0.          0.    0.
1058   Mg++        ClO4-            0.    0.    0.    0.          0.    0.
1059   Mg++        NpO2(OH)2-       0.    0.    0.    0.          0.    0.
1060   Mg++        NpO2CO3-         0.    0.    0.    0.          0.    0.
1061   Mg++        NpO2(CO3)2=-     0.    0.    0.    0.          0.    0.
1062   Mg++        NpO2(CO3)3==-    0.    0.    0.    0.          0.    0.
1063   Mg++        H2PO4-           0.    0.    0.    0.          0.    0.
1064   Mg++        HPO4=            0.    0.    0.    0.          0.    0.
1065   Mg++        PO4=-            0.    0.    0.    0.          0.    0.
1066   MgOH+       Cl-              0.    0.    0.    0.          0.    0.
1067   MgOH+       SO4=             0.    0.    0.    0.          0.    0.
1068   MgOH+       HSO4-            0.    0.    0.    0.          0.    0.
1069   MgOH+       OH-              0.    0.    0.    0.          0.    0.
1070   MgOH+       HCO3-            0.    0.    0.    0.          0.    0.
1071   MgOH+       CO3=             0.    0.    0.    0.          0.    0.
1072   MgOH+       B(OH)4-          0.    0.    0.    0.          0.    0.
1073   MgOH+       B3O3(OH)4-       0.    0.    0.    0.          0.    0.
1074   MgOH+       B4O5(OH)4=       0.    0.    0.    0.          0.    0.
1075   MgOH+       Br-              0.    0.    0.    0.          0.    0.
1076   MgOH+       Am(CO3)2-        0.    0.    0.    0.          0.    0.
1077   MgOH+       Am(CO3)3=-       0.    0.    0.    0.          0.    0.
1078   MgOH+       ClO4-            0.    0.    0.    0.          0.    0.
1079   MgOH+       NpO2(OH)2-       0.    0.    0.    0.          0.    0.
1080   MgOH+       NpO2CO3-         0.    0.    0.    0.          0.    0.
1081   MgOH+       NpO2(CO3)2=-     0.    0.    0.    0.          0.    0.
1082   MgOH+       NpO2(CO3)3==-    0.    0.    0.    0.          0.    0.
1083   MgOH+       H2PO4-           0.    0.    0.    0.          0.    0.
1084   MgOH+       HPO4=            0.    0.    0.    0.          0.    0.
1085   MgOH+       PO4=-            0.    0.    0.    0.          0.    0.
1086   H+          Cl-              0.    0.    0.   -0.01020     0.    0.
1087   H+          SO4=             0.    0.    0.    0.          0.    0.
1088   H+          HSO4-            0.    0.    0.    0.          0.    0.
1089   H+          OH-              0.    0.    0.    0.          0.    0.
1090   H+          HCO3-            0.    0.    0.    0.          0.    0.
1091   H+          CO3=             0.    0.    0.    0.          0.    0.
1092   H+          B(OH)4-          0.    0.    0.    0.          0.    0.
1093   H+          B3O3(OH)4-       0.    0.    0.    0.          0.    0.
1094   H+          B4O5(OH)4=       0.    0.    0.    0.          0.    0.
1095   H+          Br-              0.    0.    0.    0.          0.    0.
1096   H+          Am(CO3)2-        0.    0.    0.    0.          0.    0.
1097   H+          Am(CO3)3=-       0.    0.    0.    0.          0.    0.
1098   H+          ClO4-            0.    0.    0.    0.          0.    0.
1099   H+          NpO2(OH)2-       0.    0.    0.    0.          0.    0.
1100   H+          NpO2CO3-         0.    0.    0.    0.          0.    0.
1101   H+          NpO2(CO3)2=-     0.    0.    0.    0.          0.    0.
1102   H+          NpO2(CO3)3==-    0.    0.    0.    0.          0.    0.
1103   H+          H2PO4-           0.    0.    0.    0.          0.    0.
1104   H+          HPO4=            0.    0.    0.    0.          0.    0.
1105   H+          PO4=-            0.    0.    0.    0.          0.    0.
1106   MgB(OH)4+   Cl-              0.    0.    0.    0.          0.    0.
1107   MgB(OH)4+   SO4=             0.    0.    0.    0.          0.    0.
1108   MgB(OH)4+   HSO4-            0.    0.    0.    0.          0.    0.
1109   MgB(OH)4+   OH-              0.    0.    0.    0.          0.    0.
1110   MgB(OH)4+   HCO3-            0.    0.    0.    0.          0.    0.
1111   MgB(OH)4+   CO3=             0.    0.    0.    0.          0.    0.
1112   MgB(OH)4+   B(OH)4-          0.    0.    0.    0.          0.    0.
1113   MgB(OH)4+   B3O3(OH)4-       0.    0.    0.    0.          0.    0.
1114   MgB(OH)4+   B4O5(OH)4=       0.    0.    0.    0.          0.    0.
1115   MgB(OH)4+   Br-              0.    0.    0.    0.          0.    0.
1116   MgB(OH)4+   Am(CO3)2-        0.    0.    0.    0.          0.    0.
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1117  MgB(OH)4+   Am(CO3)3=-     0.   0.   0.   0.   0.   0.
1118  MgB(OH)4+   ClO4-          0.   0.   0.   0.   0.   0.
1119  MgB(OH)4+   NpO2(OH)2-     0.   0.   0.   0.   0.   0.
1120  MgB(OH)4+   NpO2CO3-       0.   0.   0.   0.   0.   0.
1121  MgB(OH)4+   NpO2(CO3)2=-   0.   0.   0.   0.   0.   0.
1122  MgB(OH)4+   NpO2(CO3)3==-  0.   0.   0.   0.   0.   0.
1123  MgB(OH)4+   H2PO4-         0.   0.   0.   0.   0.   0.
1124  MgB(OH)4+   HPO4=          0.   0.   0.   0.   0.   0.
1125  MgB(OH)4+   PO4=-          0.   0.   0.   0.   0.   0.
1126  CaB(OH)4+   Cl-            0.   0.   0.   0.   0.   0.
1127  CaB(OH)4+   SO4=           0.   0.   0.   0.   0.   0.
1128  CaB(OH)4+   HSO4-          0.   0.   0.   0.   0.   0.
1129  CaB(OH)4+   OH-            0.   0.   0.   0.   0.   0.
1130  CaB(OH)4+   HCO3-          0.   0.   0.   0.   0.   0.
1131  CaB(OH)4+   CO3=           0.   0.   0.   0.   0.   0.
1132  CaB(OH)4+   B(OH)4-        0.   0.   0.   0.   0.   0.
1133  CaB(OH)4+   B3O3(OH)4-     0.   0.   0.   0.   0.   0.
1134  CaB(OH)4+   B4O5(OH)4=     0.   0.   0.   0.   0.   0.
1135  CaB(OH)4+   Br-            0.   0.   0.   0.   0.   0.
1136  CaB(OH)4+   Am(CO3)2-      0.   0.   0.   0.   0.   0.
1137  CaB(OH)4+   Am(CO3)3=-     0.   0.   0.   0.   0.   0.
1138  CaB(OH)4+   ClO4-          0.   0.   0.   0.   0.   0.
1139  CaB(OH)4+   NpO2(OH)2-     0.   0.   0.   0.   0.   0.
1140  CaB(OH)4+   NpO2CO3-       0.   0.   0.   0.   0.   0.
1141  CaB(OH)4+   NpO2(CO3)2=-   0.   0.   0.   0.   0.   0.
1142  CaB(OH)4+   NpO2(CO3)3==-  0.   0.   0.   0.   0.   0.
1143  CaB(OH)4+   H2PO4-         0.   0.   0.   0.   0.   0.
1144  CaB(OH)4+   HPO4=          0.   0.   0.   0.   0.   0.
1145  CaB(OH)4+   PO4=-          0.   0.   0.   0.   0.   0.
1146  Am+++       Cl-            0.   0.   0.   0.   0.   0.
1147  Am+++       SO4=           0.   0.   0.   0.   0.   0.
1148  Am+++       HSO4-          0.   0.   0.   0.   0.   0.
1149  Am+++       OH-            0.   0.   0.   0.   0.   0.
1150  Am+++       HCO3-          0.   0.   0.   0.   0.   0.
1151  Am+++       CO3=           0.   0.   0.   0.   0.   0.
1152  Am+++       B(OH)4-        0.   0.   0.   0.   0.   0.
1153  Am+++       B3O3(OH)4-     0.   0.   0.   0.   0.   0.
1154  Am+++       B4O5(OH)4=     0.   0.   0.   0.   0.   0.
1155  Am+++       Br-            0.   0.   0.   0.   0.   0.
1156  Am+++       Am(CO3)2-      0.   0.   0.   0.   0.   0.
1157  Am+++       Am(CO3)3=-     0.   0.   0.   0.   0.   0.
1158  Am+++       ClO4-          0.   0.   0.   0.   0.   0.
1159  Am+++       NpO2(OH)2-     0.   0.   0.   0.   0.   0.
1160  Am+++       NpO2CO3-       0.   0.   0.   0.   0.   0.
1161  Am+++       NpO2(CO3)2=-   0.   0.   0.   0.   0.   0.
1162  Am+++       NpO2(CO3)3==-  0.   0.   0.   0.   0.   0.
1163  Am+++       H2PO4-         0.   0.   0.   0.   0.   0.
1164  Am+++       HPO4=          0.   0.   0.   0.   0.   0.
1165  Am+++       PO4=-          0.   0.   0.   0.   0.   0.
1166  AmCO3+      Cl-            0.   0.   0.   0.   0.   0.
1167  AmCO3+      SO4=           0.   0.   0.   0.   0.   0.
1168  AmCO3+      HSO4-          0.   0.   0.   0.   0.   0.
1169  AmCO3+      OH-            0.   0.   0.   0.   0.   0.
1170  AmCO3+      HCO3-          0.   0.   0.   0.   0.   0.
1171  AmCO3+      CO3=           0.   0.   0.   0.   0.   0.
1172  AmCO3+      B(OH)4-        0.   0.   0.   0.   0.   0.
1173  AmCO3+      B3O3(OH)4-     0.   0.   0.   0.   0.   0.
1174  AmCO3+      B4O5(OH)4=     0.   0.   0.   0.   0.   0.
1175  AmCO3+      Br-            0.   0.   0.   0.   0.   0.
1176  AmCO3+      Am(CO3)2-      0.   0.   0.   0.   0.   0.
1177  AmCO3+      Am(CO3)3=-     0.   0.   0.   0.   0.   0.
1178  AmCO3+      ClO4-          0.   0.   0.   0.   0.   0.
1179  AmCO3+      NpO2(OH)2-     0.   0.   0.   0.   0.   0.
1180  AmCO3+      NpO2CO3-       0.   0.   0.   0.   0.   0.
1181  AmCO3+      NpO2(CO3)2=-   0.   0.   0.   0.   0.   0.
1182  AmCO3+      NpO2(CO3)3==-  0.   0.   0.   0.   0.   0.
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1183  AmCO3+     H2PO4-        0.    0.    0.    0.    0.    0.
1184  AmCO3+     HPO4=         0.    0.    0.    0.    0.    0.
1185  AmCO3+     PO4=-         0.    0.    0.    0.    0.    0.
1186  Th++++     Cl-           0.    0.    0.    0.    0.    0.
1187  Th++++     SO4=          0.    0.    0.    0.    0.    0.
1188  Th++++     HSO4-         0.    0.    0.    0.    0.    0.
1189  Th++++     OH-           0.    0.    0.    0.    0.    0.
1190  Th++++     HCO3-         0.    0.    0.    0.    0.    0.
1191  Th++++     CO3=          0.    0.    0.    0.    0.    0.
1192  Th++++     B(OH)4-       0.    0.    0.    0.    0.    0.
1193  Th++++     B3O3(OH)4-    0.    0.    0.    0.    0.    0.
1194  Th++++     B4O5(OH)4=    0.    0.    0.    0.    0.    0.
1195  Th++++     Br-           0.    0.    0.    0.    0.    0.
1196  Th++++     Am(CO3)2-     0.    0.    0.    0.    0.    0.
1197  Th++++     Am(CO3)3=-    0.    0.    0.    0.    0.    0.
1198  Th++++     ClO4-         0.    0.    0.    0.    0.    0.
1199  Th++++     NpO2(OH)2-    0.    0.    0.    0.    0.    0.
1200  Th++++     NpO2CO3-      0.    0.    0.    0.    0.    0.
1201  Th++++     NpO2(CO3)2=-  0.    0.    0.    0.    0.    0.
1202  Th++++     NpO2(CO3)3==- 0.    0.    0.    0.    0.    0.
1203  Th++++     H2PO4-        0.    0.    0.    0.    0.    0.
1204  Th++++     HPO4=         0.    0.    0.    0.    0.    0.
1205  Th++++     PO4=-         0.    0.    0.    0.    0.    0.
1206  UO2++      Cl-           0.    0.    0.    0.    0.    0.
1207  UO2++      SO4=          0.    0.    0.    0.    0.    0.
1208  UO2++      HSO4-         0.    0.    0.    0.    0.    0.
1209  UO2++      OH-           0.    0.    0.    0.    0.    0.
1210  UO2++      HCO3-         0.    0.    0.    0.    0.    0.
1211  UO2++      CO3=          0.    0.    0.    0.    0.    0.
1212  UO2++      B(OH)4-       0.    0.    0.    0.    0.    0.
1213  UO2++      B3O3(OH)4-    0.    0.    0.    0.    0.    0.
1214  UO2++      B4O5(OH)4=    0.    0.    0.    0.    0.    0.
1215  UO2++      Br-           0.    0.    0.    0.    0.    0.
1216  UO2++      Am(CO3)2-     0.    0.    0.    0.    0.    0.
1217  UO2++      Am(CO3)3=-    0.    0.    0.    0.    0.    0.
1218  UO2++      ClO4-         0.    0.    0.    0.    0.    0.
1219  UO2++      NpO2(OH)2-    0.    0.    0.    0.    0.    0.
1220  UO2++      NpO2CO3-      0.    0.    0.    0.    0.    0.
1221  UO2++      NpO2(CO3)2=-  0.    0.    0.    0.    0.    0.
1222  UO2++      NpO2(CO3)3==- 0.    0.    0.    0.    0.    0.
1223  UO2++      H2PO4-        0.    0.    0.    0.    0.    0.
1224  UO2++      HPO4=         0.    0.    0.    0.    0.    0.
1225  UO2++      PO4=-         0.    0.    0.    0.    0.    0.
1226  NpO2+      Cl-           0.    0.    0.    0.    0.    0.
1227  NpO2+      SO4=          0.    0.    0.    0.    0.    0.
1228  NpO2+      HSO4-         0.    0.    0.    0.    0.    0.
1229  NpO2+      OH-           0.    0.    0.    0.    0.    0.
1230  NpO2+      HCO3-         0.    0.    0.    0.    0.    0.
1231  NpO2+      CO3=          0.    0.    0.    0.    0.    0.
1232  NpO2+      B(OH)4-       0.    0.    0.    0.    0.    0.
1233  NpO2+      B3O3(OH)4-    0.    0.    0.    0.    0.    0.
1234  NpO2+      B4O5(OH)4=    0.    0.    0.    0.    0.    0.
1235  NpO2+      Br-           0.    0.    0.    0.    0.    0.
1236  NpO2+      Am(CO3)2-     0.    0.    0.    0.    0.    0.
1237  NpO2+      Am(CO3)3=-    0.    0.    0.    0.    0.    0.
1238  NpO2+      ClO4-         0.    0.    0.    0.    0.    0.
1239  NpO2+      NpO2(OH)2-    0.    0.    0.    0.    0.    0.
1240  NpO2+      NpO2CO3-      0.    0.    0.    0.    0.    0.
1241  NpO2+      NpO2(CO3)2=-  0.    0.    0.    0.    0.    0.
1242  NpO2+      NpO2(CO3)3==- 0.    0.    0.    0.    0.    0.
1243  NpO2+      H2PO4-        0.    0.    0.    0.    0.    0.
1244  NpO2+      HPO4=         0.    0.    0.    0.    0.    0.
1245  NpO2+      PO4=-         0.    0.    0.    0.    0.    0.
1246  using PITZER ACTIVITY COEFFICIENT model
1247  Charge Balance replaces element   Oxygen
1248
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this is a BATCH problem


Ideal Gas Constant is Unity (Dimensionless)
Temperature =        298.15 [=] degree Kelvin


  115 Species                          23 Elements

The Elements and Molecular Weights are
                Hydrogen              1.007900
                Oxygen               15.999400
                Sodium               22.989770
                Potassium            39.098300
                Magnesium            24.305000
                Calcium              40.080000
                Chlorine             35.453000
                Sulfur               32.060000
                Carbon               12.011000
                PosIon                0.000000
                NegIon                0.000000
                Air                  28.840000
                Boron                10.810000
                Bromine              79.904000
                TracerEl              0.000000
                Th(IV)              232.038100
                Am(III)             243.000000
                U(VI)               238.029000
                Np(V)               237.048200
                ClO4-(EL)            99.450600
                Phosphorus           30.974000
                Electron              0.000000
                Charge                0.000000


Species Name                                    MW     Free Energy of Formation
                                                       (dimensionless)

  1 H2O                                WATER(a)  18.02 -9.5663E+01  2  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
  2 Na+                                  Na+(a)  22.99 -1.0565E+02  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  1
  3 K+                                    K+(a)  39.10 -1.1396E+02  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  1
  4 Ca++                                Ca++(a)  40.08 -2.2330E+02  0  0  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  2
  5 Mg++                                Mg++(a)  24.30 -1.8347E+02  0  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  2
  6 MgOH+                              MgOH+(a)  41.31 -2.5194E+02  1  1  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  1
  7 H+                                    H+(a)   1.01  0.0000E+00  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  1
  8 Cl-                                  Cl-(a)  35.45 -5.2955E+01  0  0  0  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0 -1
  9 SO4=                                SO4=(a)  96.06 -3.0039E+02  0  4  0  0  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0 -2
 10 HSO4-                              HSO4-(a)  97.07 -3.0494E+02  1  4  0  0  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0 -1
 11 OH-                                  OH-(a)  17.01 -6.3435E+01  1  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0 -1
 12 HCO3-                              HCO3-(a)  61.02 -2.3675E+02  1  3  0  0  0  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0 -1
 13 CO3=                                CO3=(a)  60.01 -2.1294E+02  0  3  0  0  0  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0 -2
 14 CO2(aq)                          CO2(aq)(a)  44.01 -1.5568E+02  0  2  0  0  0  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0
 15 CaCO3(aq)                      CaCO3(aq)(a) 100.09 -4.4350E+02  0  3  0  0  0  1  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0
 16 MgCO3(aq)                      MgCO3(aq)(a)  84.31 -4.0315E+02  0  3  0  0  1  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0
 17 B(OH)3(aq)                    B(OH)3(aq)(a)  61.83 -3.9081E+02  3  3  0  0  0  0  0  0  0  0  0  0  1  0  0  0  0  0  0  0  0  0  0
 18 B(OH)4-                          B(OH)4-(a)  78.84 -4.6520E+02  4  4  0  0  0  0  0  0  0  0  0  0  1  0  0  0  0  0  0  0  0  0 -1
 19 B3O3(OH)4-                    B3O3(OH)4-(a) 148.46 -9.6377E+02  4  7  0  0  0  0  0  0  0  0  0  0  3  0  0  0  0  0  0  0  0  0 -1
 20 B4O5(OH)4=                    B4O5(OH)4=(a) 191.27 -1.2391E+03  4  9  0  0  0  0  0  0  0  0  0  0  4  0  0  0  0  0  0  0  0  0 -2
 21 CaB(OH)4+                      CaB(OH)4+(a) 118.92 -6.9230E+02  4  4  0  0  0  1  0  0  0  0  0  0  1  0  0  0  0  0  0  0  0  0  1
 22 MgB(OH)4+                      MgB(OH)4+(a) 103.14 -6.5189E+02  4  4  0  0  1  0  0  0  0  0  0  0  1  0  0  0  0  0  0  0  0  0  1
 23 Br-                                  Br-(a)  79.90 -9.9999E+02  0  0  0  0  0  0  0  0  0  0  0  0  0  1  0  0  0  0  0  0  0  0 -1
 24 ClO4-                   perchlorate ClO4-(a)  99.45 -9.9999E+02  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  1  0  0 -1
 25 NaOH(aq)......to.titrate.base.only(a)  40.00  5.0000E+02  1  1  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
 26 HCl(aq).......to.titrate.acid.only(a)  36.46  5.0000E+02  1  0  0  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
 27 HClO4(aq).....to.titrate.acid.only(a) 100.46  5.0000E+02  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  1  0  0  0
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28 PosIon................POSITIVE.ION(a)   0.00  0.0000E+00   0  0  0  0  0  0  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  1
29 NegIon................NEGATIVE.ION(a)   0.00  0.0000E+00   0  0  0  0  0  0  0  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0 -1
30 PosIon(OH)(aq).....to.titrate.base(a)  17.01  5.0000E+02   1  1  0  0  0  0  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0
31 HNegIon(aq)........to.titrate.acid(a)   1.01  5.0000E+02   1  0  0  0  0  0  0  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0
32 Tracer(aq).....conservative.tracer(a)   0.00  0.0000E+00   0  0  0  0  0  0  0  0  0  0  0  0  0  0  1  0  0  0  0  0  0  0  0
33 H3PO4(aq)                H3PO4(aq)(a)  98.00 -4.6090E+02   3  4  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  1  0  0
34 H2PO4-                      H2PO4-(a)  96.99 -4.5596E+02   2  4  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  1  0 -1
35 HPO4=                        HPO4=(a)  95.98 -4.3937E+02   1  4  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  1  0 -2
36 PO4=-                        PO4=-(a)  94.97 -4.1095E+02   0  4  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  1  0 -3
37 NpO2+                        NpO2+(a) 269.05 -3.6913E+02   0  2  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  1  0  0  0  1
38 NpO2OH(aq)              NpO2OH(aq)(a) 286.05 -4.3852E+02   1  3  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  1  0  0  0  0
39 NpO2(OH)2-              NpO2(OH)2-(a) 303.06 -5.0583E+02   2  4  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  1  0  0  0 -1
40 NpO2CO3-                  NpO2CO3-(a) 329.06 -5.9449E+02   0  5  0  0  0  0  0  0  1  0  0  0  0  0  0  0  0  0  1  0  0  0 -1
41 NpO2(CO3)2=-          NpO2(CO3)2=-(a) 389.07 -8.0840E+02   0  8  0  0  0  0  0  0  2  0  0  0  0  0  0  0  0  0  1  0  0  0 -3
42 NpO2(CO3)3==-        NpO2(CO3)3==-(a) 449.07 -1.0199E+03   0 11  0  0  0  0  0  0  3  0  0  0  0  0  0  0  0  0  1  0  0  0 -5
43 Am+++                        Am+++(a) 243.00 -2.4169E+02   0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  1  0  0  0  0  0  3
44 AmCO3+                      AmCO3+(a) 303.01 -4.7206E+02   0  3  0  0  0  0  0  0  1  0  0  0  0  0  0  0  1  0  0  0  0  0  1
45 Am(CO3)2-                Am(CO3)2-(a) 363.02 -6.9588E+02   0  6  0  0  0  0  0  0  2  0  0  0  0  0  0  0  1  0  0  0  0  0 -1
46 Am(CO3)3=-              Am(CO3)3=-(a) 423.03 -9.1546E+02   0  9  0  0  0  0  0  0  3  0  0  0  0  0  0  0  1  0  0  0  0  0 -3
47 Am(OH)2+                  Am(OH)2+(a) 277.01 -3.9365E+02   2  2  0  0  0  0  0  0  0  0  0  0  0  0  0  0  1  0  0  0  0  0  1
48 Am(OH)3(aq)            Am(OH)3(aq)(a) 294.02 -4.6295E+02   3  3  0  0  0  0  0  0  0  0  0  0  0  0  0  0  1  0  0  0  0  0  0
49 Th++++                      Th++++(a) 232.04 -9.9999E+02   0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  1  0  0  0  0  0  0  4
50 UO2++                    U(VI)O2++(a) 270.03 -9.9999E+02   0  2  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  1  0  0  0  0  2
51 NpO2OH(aged)__________NpO2OH(aged)(s) 286.05 -4.5401E+02   1  3  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  1  0  0  0  0
52 NpO2OH(amor)__________NpO2OH(amor)(s) 286.05 -4.5264E+02   1  3  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  1  0  0  0  0
53 NaNpO2CO3(s)__________NaNpO2CO3(s)(s) 352.05 -7.1371E+02   0  5  1  0  0  0  0  0  1  0  0  0  0  0  0  0  0  0  1  0  0  0  0
54 Na3NpO2(CO3)2(s)_DISABLED_DISABLED(s) 458.03  9.9999E+02   0  8  3  0  0  0  0  0  2  0  0  0  0  0  0  0  0  0  1  0  0  0  0
55 AmOHCO3(c)________________AmOHCO3(c)(s) 320.02 -5.6998E+02   1  4  0  0  0  0  0  0  1  0  0  0  0  0  0  0  1  0  0  0  0  0  0
56 Am(OH)3(s)________________Am(OH)3(s)(s) 294.02 ~~-5.3555E+02~~   3  3  0  0  0  0  0  0  0  0  0  0  0  0  0  0  1  0  0  0  0  0  0   -4.9229E+0
57 NaAm(CO3)2.6H2O(c)________________  (s) 494.10 -1.3965E+03  12 12  1  0  0  0  0  0  2  0  0  0  0  0  0  0  1  0  0  0  0  0  0
58 AmPO4(c)_____________________AmPO4(c)(s) 337.97 -7.0975E+02   0  4  0  0  0  0  0  0  0  0  0  0  0  0  0  0  1  0  0  0  1  0  0
59 CaSO4________________________Anhydrite(s) 136.14 -5.3373E+02   0  4  0  0  0  1  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
60 NaK3(SO4)2__Aphthitalite/Glaserite(s) 332.40 -1.0570E+03   0  8  1  3  0  0  0  2  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
61 CaCl2.6H2O_______________Antarcticite(s) 219.08 -8.9365E+02  12  6  0  0  0  1  2  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
62 CaCO3________________________Aragonite(s) 100.09 -4.5517E+02   0  3  0  0  0  1  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0
63 K2SO4__________________________Arcanite(s) 174.25 -5.3239E+02   0  4  0  2  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
64 MgCl2.6H2O_________________Bischofite(s) 203.30 -8.5310E+02  12  6  0  0  1  0  2  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
65 Na2Mg(SO4)2.4H2O_____________Bloedite(s) 334.46 -1.3836E+03   8 12  2  0  1  0  0  2  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
66 Mg(OH)2_________________________Brucite(s)  58.32 -3.3540E+02   2  2  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
67 Na6CO3(SO4)2___________________Burkeite(s) 390.06 -1.4494E+03   0 11  6  0  0  0  0  2  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0
68 CaCO3___________________________Calcite(s) 100.09 -4.5560E+02   0  3  0  0  0  1  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0
69 CaCl2.4H2O_______CaCl2_Tetrahydrite(s) 183.05 -6.9870E+02   8  4  0  0  0  1  2  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
70 Ca4Cl2(OH)6.13H2O__CaOxychloride A(s) 567.47 -2.6584E+03  32 19  0  0  0  4  2  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
71 Ca2Cl2(OH)2.H2O____CaOxychloride B(s) 203.10 -7.7841E+02   4  3  0  0  0  2  2  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
72 KMgCl3.6H2O________________Carnallite(s) 277.85 -1.0203E+03  12  6  0  1  1  0  3  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
73 MgSO4.7H2O____________________Epsomite(s) 246.47 -1.1578E+03  14 11  0  0  1  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
74 CaNa2(CO3)2.5H2O_________Gaylussite(s) 296.15 -1.3605E+03  10 11  2  0  0  1  0  0  2  0  0  0  0  0  0  0  0  0  0  0  0  0  0
75 Na2Ca(SO4)2_______________Glauberite(s) 278.17 -1.0475E+03   0  8  2  0  0  1  0  2  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
76 CaSO4.2H2O_______________________Gypsum(s) 172.17 -7.2556E+02   4  6  0  0  0  1  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
77 NaCl_____________________________Halite(s)  58.44 -1.5499E+02   0  0  1  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
78 MgSO4.6H2O__________________Hexahydrite(s) 228.45 -1.0616E+03  12 10  0  0  1  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
79 KMgClSO4.3H2O__________________Kainite(s) 248.96 -9.3820E+02   6  7  0  1  1  0  1  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
80 KHCO3________________________Kalicinite(s) 100.12 -3.5006E+02   1  3  0  1  0  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0
81 MgSO4.H2O____________________ Kieserite(s) 138.38 -5.7980E+02   2  5  0  0  1  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
82 K2Mg(SO4)2.4H2O________________Leonite(s) 366.68 -1.4040E+03   8 12  0  2  1  0  0  2  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
83 Na4Ca(SO4)3.2H2O__________Labile_Salt(s) 456.24 -1.7515E+03   4 14  4  0  0  1  0  3  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
84 MgCO3_________________________Magnesite(s)  84.31 -4.1445E+02   0  3  0  0  1  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0
85 Mg2Cl(OH)3.4H2O________MgOxychloride(s) 207.15 -1.0296E+03  11  7  0  0  2  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
86 KHSO4__________________________Mercallite(s) 136.16 -4.1757E+02   1  4  0  1  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
87 Na2SO4.10H2O__________________Mirabilite(s) 322.19 -1.4712E+03  20 14  2  0  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
88 K8H6(SO4)7______________________Misenite(s) 991.24 -3.0392E+03   6 28  0  8  0  0  0  7  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
89 NaHCO3__________________________Nahcolite(s)  84.01 -3.4333E+02   1  3  1  0  0  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0
90 Na2CO3.10H2O_______________________Natron(s) 286.14 -1.3828E+03  20 13  2  0  0  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0
91 MgCO3.3H2O_______________Nesquehonite(s) 138.36 -6.9530E+02   6  6  0  0  1  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0
92 K2Mg(SO4)2.6H2O Picromerite/Schoen(s) 402.71 -1.5961E+03  12 14  0  2  1  0  0  2  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
93 Na2Ca(CO3)2.2H2O__________Pirssonite(s) 242.11 -1.0731E+03   4  8  2  0  0  1  0  0  2  0  0  0  0  0  0  0  0  0  0  0  0  0  0
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 94 K2MgCa2(SO4)4.2H2O______Polyhalite(s) 602.92 -2.2825E+03   4 18  0  2  1  2  0  4  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
 95 Ca(OH)2_________________Portlandite(s)  74.09 -3.6212E+02   2  2  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
 96 K2CO3.3/2H2O___Potassium_Carbonate(s) 165.23 -5.7737E+02   3  4  0  2  0  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0
 97 K8H4(CO3)6.3H2O___K-Sequicarbonate(s) 730.92 -2.5554E+03  10 21  0  8  0  0  0  0  6  0  0  0  0  0  0  0  0  0  0  0  0  0  0
 98 KNaCO3.6H2O__________K-Na-Carbonate(s) 230.19 -1.0068E+03  12  9  1  1  0  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0
 99 K2NaH(CO3)2.2H2O___Potassium_Trona(s) 258.24 -9.7174E+02   5  8  1  2  0  0  0  0  2  0  0  0  0  0  0  0  0  0  0  0  0  0  0
100 K3H(SO4)2__Sesquipotassium_Sulfate(s) 310.42 -9.5080E+02   1  8  0  3  0  0  0  2  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
101 Na3H(SO4)2____Sesquisodium_Sulfate(s) 262.09 -9.1960E+02   1  8  3  0  0  0  0  2  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
102 Na2CO3.7H2O____Na2CO3-Heptahydrate(s) 232.10 -1.0950E+03  14 10  2  0  0  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0
103 KCl________________________Sylvite(s)  74.55 -1.6484E+02   0  0  0  1  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
104 K2Ca(SO4)2.H2O___________Syngenite(s) 328.41 -1.1648E+03   2  9  0  2  0  1  0  2  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
105 Mg2CaCl6.12H2O________Tachyhydrite(s) 517.59 -2.0159E+03  24 12  0  0  2  1  6  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
106 Na2SO4__________________Thenardite(s) 142.04 -5.1235E+02   0  4  2  0  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0  0
107 Na2CO3.H2O___________Thermonatrite(s) 124.00 -5.1880E+02   2  4  2  0  0  0  0  0  1  0  0  0  0  0  0  0  0  0  0  0  0  0  0
108 Na3H(CO3)2.2H2O_______________Trona(s) 226.03 -9.6038E+02   5  8  3  0  0  0  0  0  2  0  0  0  0  0  0  0  0  0  0  0  0  0  0
109 Na2B4O7.10H2O_______________ Borax(s) 381.37 -2.2242E+03  20 17  2  0  0  0  0  0  0  0  0  0  4  0  0  0  0  0  0  0  0  0  0
110 B(OH)3____________Borix_Acid_Solid(s)  61.83 -3.9088E+02   3  3  0  0  0  0  0  0  0  0  0  0  1  0  0  0  0  0  0  0  0  0  0
111 KB5O8.4H2O____K-Pentaborate_(30_C)(s) 293.20 -1.7703E+03   8 12  0  1  0  0  0  0  0  0  0  0  5  0  0  0  0  0  0  0  0  0  0
112 K2B4O7.4H2O___K-Tetraborate_(30_C)(s) 305.49 -1.6635E+03   8 11  0  2  0  0  0  0  0  0  0  0  4  0  0  0  0  0  0  0  0  0  0
113 NaBO2.4H2O_______Sodium_Metaborate(s) 137.86 -7.6142E+02   8  6  1  0  0  0  0  0  0  0  0  0  1  0  0  0  0  0  0  0  0  0  0
114 NaB5O8.5H2O_____Sodium_Pentaborate(s) 295.11 -1.8548E+03  10 13  1  0  0  0  0  0  0  0  0  0  5  0  0  0  0  0  0  0  0  0  0
115 NaBO2.NaCl.2H2O___Teepleite_(20_C)(s) 160.27 -7.2577E+02   4  4  2  0  0  0  1  0  0  0  0  0  1  0  0  0  0  0  0  0  0  0  0
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Sandia National Laboratories

MANAGED BY MARTIN MARIETTA CORPORATION
FOR THE U. S. DEPARTMENT OF ENERGY

P. O. BOX 5800
ALBUQUERQUE, NEW MEXICO 87185-1320

date: 31 January 1995

to: Nancy J. Dhooge, Org. 6348, MS 1342, 848-0883

Craig F Novak

from: Craig F. Novak, Dept. 6119, MS 1320, 848-0619

subject: Correction to the Database for FMT called "HMW_NP_AM.CHEMDAT," to create version date 95.01.31.

Several errors were contained in the memorandum 'Creation and Definition of the Database for FMT called "HMW_NP_AM.CHEMDAT," version date 94.01.17,' dated 18 January. Please make the following changes to the memorandum and the computer file "HMW_NP_AM.CHEMDAT."

- the μ_i^0 / RT value for $\mathrm{Pu(OH)3(s)}$ in Table 8 was copied in error from the source report. Replace the "–527.259" with "–484.0" and replace the "–535.553" with "–492.294"
- change the date stamp, line 2 of Appendix A, from "94.01.17" to "95.01.31"
- change the last number in line 80 of Appendix A from "–535.553" to "–492.294"

These changes will correct the error and make the 95.01.31 version of the file "HMW_NP_AM.CHEMDAT." The only changes that this will make to the partial output file in Appendix B are

- all occurrences of "94.01.17" will be replaced with "95.01.31"
- the dimensionless free energy of formation on line 1343 will be changed from "–5.3555E+02" to "–4.9229E+02"

Copies to:

6119	C.F. Novak, MS 1320
6119	E.J. Nowak, MS 1320
6119	H.W. Papenguth, MS 1320
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6348	C.C. Crafts, MS 1342

SWCF WBS 1.1.1.1.4, 1.1.1.2.6, 1.1.5.1.2, 1.1.5.2.2, 1.1.5.1.6: RNC

File: 95.01.31 FMT DB Correction

Memo from C.F. Novak

TP 168

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APPENDIX E: ACTINIDE SOURCE TERM DISSOLVED CONCENTRATION SUBMODEL PREDICTION OF AM(III) CONCENTRATIONS IN A CLOSED SYSTEM OF SALADO SIMULANT SPC BRINE AS A FUNCTION OF HYDROGEN ION CONCENTRATION

Novak, C.F. 1995. "Input/Output Files for an FMT 'TITRATE' Benchmark Problem: The Solubility of $Am(OH)_3(s)$ in Salado SPC Brine in a Closed System from Neutral to Highly Basic Conditions" memorandum to Chris C. Crafts. 16 March 1995.

i

Sandia National Laboratories

MANAGED BY MARTIN MARIETTA CORPORATION
FOR THE U. S. DEPARTMENT OF ENERGY

P. O. BOX 5800
ALBUQUERQUE, NEW MEXICO 87185-1320

date: 16 March 1995

to: Chris C. Crafts, Org. 6348, MS 1342

Craig F Novak

from: Craig F. Novak, Dept. 6119, MS 1320, 848-0619

subject: Input/Output Files for an FMT 'TITRATE' Benchmark Problem: The Solubility of $Am(OH)_3(s)$ in Salado SPC Brine in a Closed System from Neutral to Highly Basic Conditions

This memorandum is part of a series (Novak, 1995a; 1995b; 1995c; 1995d; 1995e; 1995f; 1995g; 1995h) documenting the use of the computer code FMT (current version 1.3) and defining testing and benchmarking problems for the code. It is expected that the information in these memos will be used to complete the FMT user's guide and associated Quality Assurance requirements for the computer code.

The benchmark problem in this memorandum serves to further illustrate the use of the 'TITRATE' option, and also shows the kinds of predictions FMT can make with regard to expected dissolved actinide concentrations in brines relevant to the WIPP. The calculations in this memorandum were made using the HMW_NP_AM.CHEMDAT data base, dated 95.01.31 (Novak, 1995a; 1995b). Detailed instructions for using the 'TITRATE' option of FMT are given in Novak (1995e; 1995h).

The composition of SPC Brine, a synthetic Salado brine, is documented in Novak (1995d). This brine, related to the Salado simulant Brine A, contains a large amount of magnesium and a relatively small amount of total inorganic carbon. The calculations in this memorandum document the process of adding Am(III) to this brine in sufficient quantity to cause precipitation of an Am(III)-bearing solid phase, and then titrating the resulting solution with concentrated NaOH to calculate the changes in dissolved Am(III) concentration as a function

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of hydrogen ion concentration. This problem simulates a closed system of (initial solution + solids + titrant solutions). No equilibrium with $CO_2(g)$ is maintained. Thus, the total inorganic carbon in the system remains constant at the initial value. Titration problems similar to the one presented in this memorandum but in equilibrium with $CO_2(g)$ at various values of fugacity will be the subject of future memoranda.

The input file AM_SPC_BM.IN and the output file AM_SPC_BM.TITRATE are given in Tables 1 and 2, respectively. The concentration profiles for some of the species included in the TITRATE file are plotted as a function of pmH in Figures 1 through 3. The quantity pmH is defined as the negative base ten logarithm of the hydrogen ion molality.

Figure 1 presents the calculated concentrations of each americium species and the total dissolved Am(III) concentration. The complex behavior in the region 8.6<pmH<10 is caused by the changing set of solid phases calculated to be in equilibrium with the solution, as shown in Figure 2. The calculated sets of solid phases change with the pmH to form the following 6 sets.

set 1	set 2	set 3	set 4	set 5	set 6
Am(OH)3(s)	Am(OH)3(s)	Am(OH)3(s)	Am(OH)3(s)	Am(OH)3(s)	Am(OH)3(s)
AmOHCO3(s)					
	MgCO3(s)	MgCO3(s)	MgCO3(s)		
		Mg2Cl(OH)3.3H2O(s)			
			Mg(OH)2(s)	Mg(OH)2(s)	Mg(OH)2(s)
				CaCO3(s)	CaCO3(s)
					Ca(OH)2(s)

At the lowest pmH in the figures, the solid phases $Am(OH)_3(s)$ and $AmOHCO_3(s)$ are in equilibrium with the aqueous phase, and are controlling the total aqueous concentration of Am(III). In the remainder of the pmH range, $Am(OH)_3(s)$ is the solid phase controlling total Am(III) concentration, but other solid phases are present to control the aqueous concentrations of other elements. For example, $Mg(OH)_2(s)$, brucite, is present beginning at pmH of about 9.3. At pmH values greater than this, the aqueous Mg^{2+} molality decreases two log units for every unit increase in the pmH because of the equilibrium

E2

$$\mathrm{Mg^{2+} + 2\,OH^- = Mg(OH)_2(s)}$$

with the corresponding equilibrium expression

$$\ln m_{\mathrm{Mg^{2+}}} + 2 \ln m_{\mathrm{OH^-}} = \frac{-\left(\mu^0_{\mathrm{Mg^{2+}}} + 2\,\mu^0_{\mathrm{OH^-}} - \mu^0_{\mathrm{Mg(OH)_2(s)}}\right)}{RT} - \ln \gamma_{\mathrm{Mg^{2+}}} - 2 \ln \gamma_{\mathrm{OH^-}}$$

where the right hand side is effectively constant in the simulations. Similar expressions can be written for the other solid species.

The simulations predict that, in a closed system such as this, Am(III) dissolved aqueous concentrations decrease as pmH increases, and reach a limiting value of about 2×10^{-13} molal. This behavior is observed for this system because they hydrolysis species $\mathrm{Am(OH)_2^+}$ and $\mathrm{Am(OH)_3^0}$ are the dominant aqueous Am(III) species for pmH>10.8. There is not enough carbonate in the system to compete successfully with hydrolysis. (Note that carbonate complexation is expected to dominate Am(III) behavior in systems open to a gas phase containing $\mathrm{CO_2(g)}$, or in systems with larger concentrations of carbonate (Rao et al., 1994; Felmy et al., 1990)).

The total Am(III) concentrations in Figure 1 demonstrate a "solubility functionality curve" for SPC Brine in a closed system. The table of numbers used to plot this curve is the corresponding "solubility functionality table." This graph or table can be used to answer the question: "What is the expected total concentration of Am(III) in a closed system of SPC Brine at pmH=___?" While closed systems of SPC Brine are of limited use for WIPP Performance Assessment calculations in which there is gas generation, this example demonstrates how "solubility functionality tables or curves" would be generated. Future memoranda will document the calculation of solubility functionality curves and tables for Am(III) in SPC Brine as a function of pmH and $\mathrm{CO_2(g)}$ fugacity, a system of direct relevance to WIPP Performance Assessment.

References

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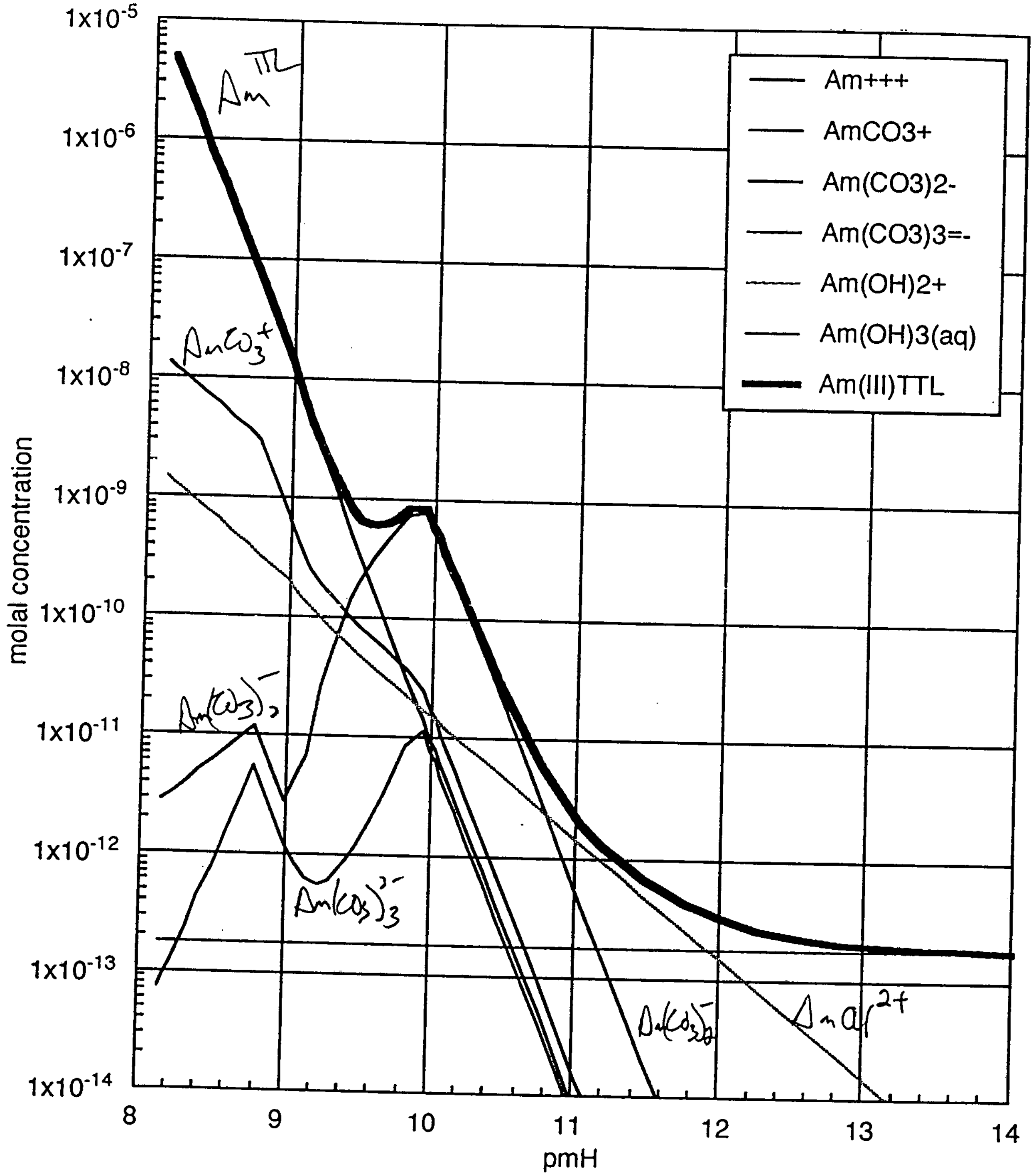
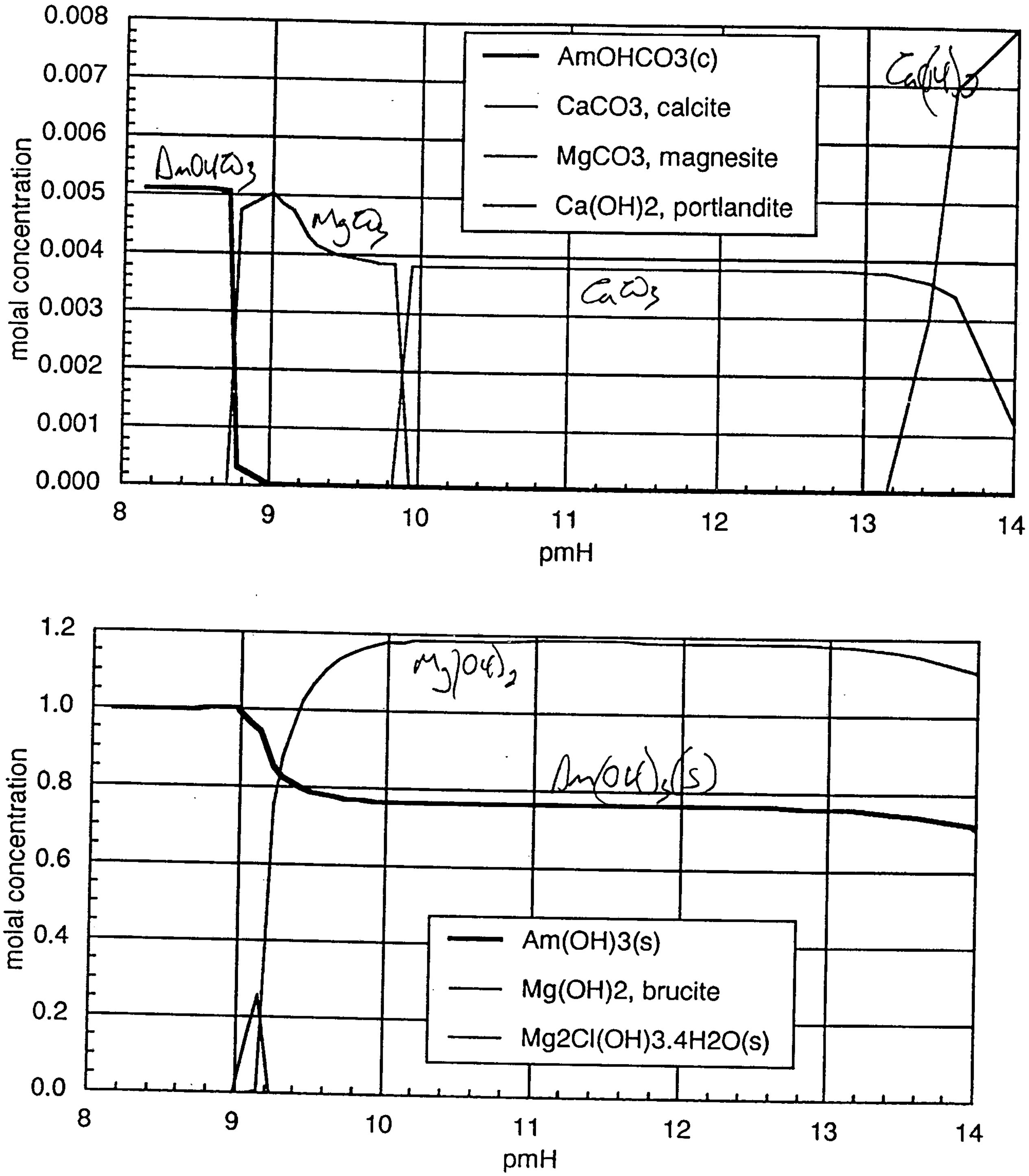
EY

Figure 1. Calculated americium species concentrations in SPC Brine as a function of hydrogen ion concentration in a system closed to mass transfer of $CO_2(g)$



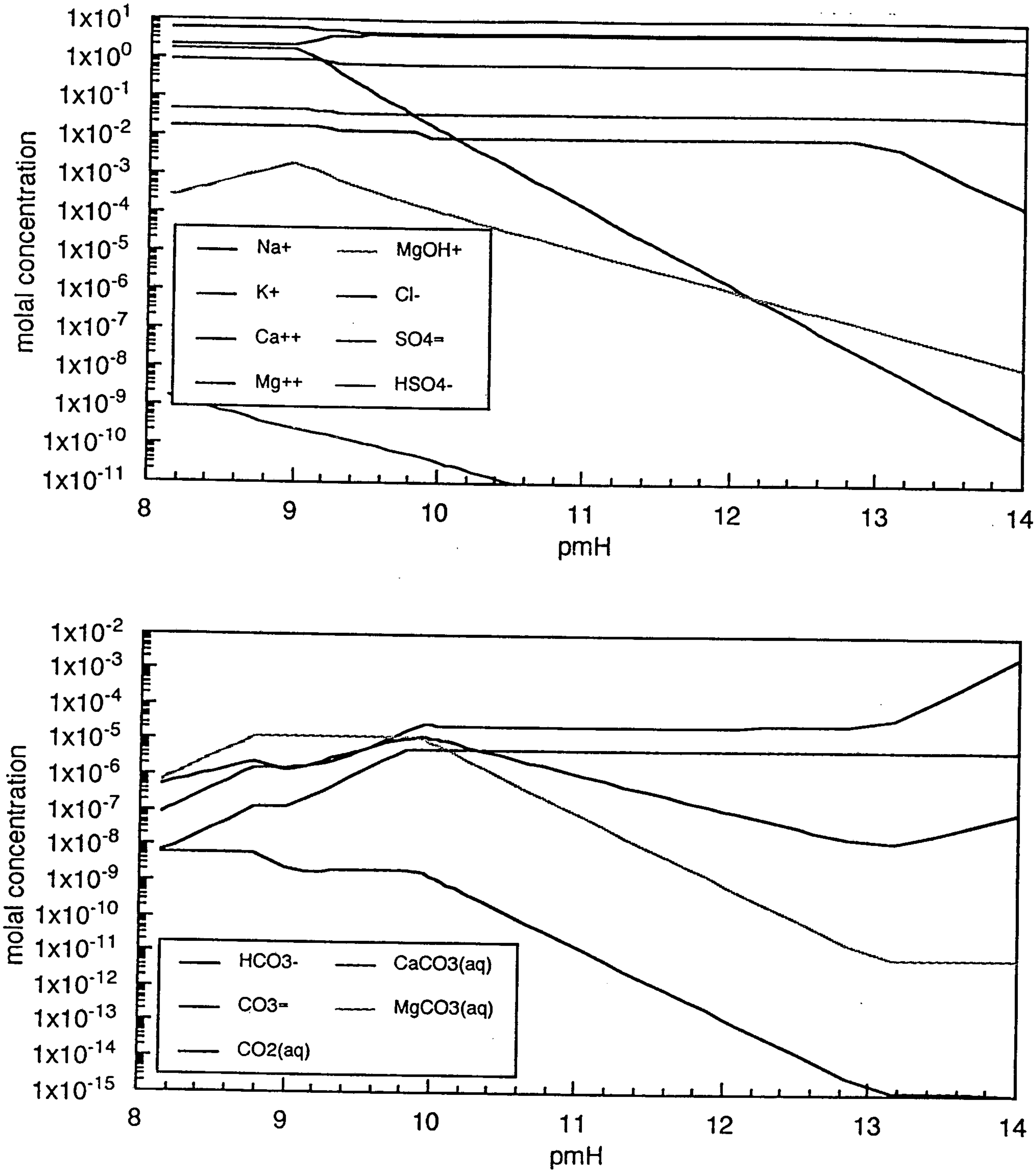
E5

Figure 2. Calculated solid species concentrations in SPC Brine as a function of hydrogen ion concentration in a system closed to mass transfer of $CO_2(g)$



E6

Figure 3. Calculated concentrations for some aqueous species in SPC Brine as a function of hydrogen ion concentration in a system closed to mass transfer of CO_2(g)



E7

Table 1.   Listing of FMT File "AM_SPC_BM.IN"

```
 'Benchmark:  Titrating Am(III)/SPC with NaOH, Closed System'
'CHEMFILE'
'TITRATE', 'EXPLICIT',

'nMOLES','nEXACT', 10 m NaOH
  1.21017364E+02   Hydrogen
  6.55086820E+01   Oxygen
  1.00000000E+01   Sodium
  0.00000000E+00   Potassium
  0.00000000E+00   Magnesium
  0.00000000E+00   Calcium
  0.00000000E-00   Chlorine
  0.00000000E+00   Sulfur
  0.00000000E+00   Carbon
  0.00000000E+00   PosIon
  0.00000000E+00   NegIon
  0.00000000E+00   Air
  0.00000000E+00   Boron
  0.00000000E+00   Bromine
  0.00000000E+00   TracerEl
  0.00000000E+00   Th(IV)
  0.00000000E+00   Am(III)
  0.00000000E+00   U(VI)
  0.00000000E+00   Np(V)
  0.00000000E+00   ClO4-(EL)
  0.00000000E+00   Phosphorus
  0.00000000E+00   Electron
  0.00000000E+00   Charge

'nMOLES','nEXACT',   SPC Brine with Am(OH)3(s) in Equilibrium
  1.14084063E+02   Hydrogen
  5.87650233E+01   Oxygen
  2.00000000E+00   Sodium
  8.40000000E-01   Potassium
  1.55999951E+00   Magnesium
  1.64000000E-02   Calcium
  5.83000000E+00   Chlorine
  4.36000000E-02   Sulfur
  5.07101504E-03   Carbon
  0.00000000E+00   PosIon
  5.32000000E-02   NegIon
  0.00000000E+00   Air
  2.18000000E-02   Boron
  1.09000000E-02   Bromine
  0.00000000E+00   TracerEl
  0.00000000E+00   Th(IV)
  1.00000000E+00   Am(III)
  0.00000000E+00   U(VI)
  0.00000000E+00   Np(V)
  0.00000000E+00   ClO4-(EL)
  0.00000000E+00   Phosphorus
  0.00000000E+00   Electron
  3.08031097E-15   Charge

50  2.25d3  0.0025d0  1.800001d5 'nDXVARIABLE'
'nDIFFUS',
'CONVEC',
'nSSDIFF',
'nRESTART',
'nPUSHPULL','nMULTINJ',

 20 1 20 'nLOTS' 10
```

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```
 'nTGRAD' 'LINEAR'
 'FRAC FLO', 'nTWO PHASE', 'nMASS TR',
 3
 0.1d0 0.2d0 0.3d0
 1.d-7 0.d0 0.18291d0 0.2d0 0.d0 'RHSFDIF', 'LHSFDIF', vfrc dfrc FPOR mpor dmxt
 'nMOLES', 'nEXACT', Plain old pure H2O
   1.11017364E+02   Hydrogen
   5.55086820E+01   Oxygen
   0.00000000E+00   Sodium
   0.00000000E+00   Potassium
   0.00000000E+00   Magnesium
   0.00000000E+00   Calcium
   0.00000000E-00   Chlorine
   0.00000000E+00   Sulfur
   0.00000000E+00   Carbon
   0.00000000E+00   PosIon
   0.00000000E+00   NegIon
   0.00000000E+00   Air
   0.00000000E+00   Boron
   0.00000000E+00   Bromine
   0.00000000E+00   TracerEl
   0.00000000E+00   Th(IV)
   0.00000000E+00   Am(III)
   0.00000000E+00   U(VI)
   0.00000000E+00   Np(V)
   0.00000000E+00   ClO4-(EL)
   0.00000000E+00   Phosphorus
   0.00000000E+00   Electron
   0.00000000E+00   Charge

 1.d-12 1.d-20 (fracture, matrix permeabilities)
 'VPOROS','FRFLASH', (NOFLASH or FRFLASH, default is all flash)
 'VAR_AQ_RHO' 1074.9d0
 'nNO X DIFF',
 'UNIFORM',0

 'TITRATE','ASREAD',0.5,283.,'INJSOLIDS'
 5.00000E-02      7.7047
 0.10000          7.7470
 0.20000          7.8327
 0.30000          7.9207
 0.40000          8.0124
 0.50000          8.1090
 0.60247          8.2140
 1.5302          8.2934
 1.8
 3.2257          8.5094
 43.867          8.6179
 76.741          8.7135
 161.77          8.8169
 194.92          8.8921
 234.87          9.0300
 246.90          9.0948
 258.94          9.1859
 266.96          9.2769
 270.97          9.3417
 274.98          9.4335
 278.99          9.5462
 280.00          9.5997
 280.50          9.6320
 281.00          9.6697
 281.50          9.7151
 282.00          9.7722
 282.50          9.8492
```

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127	283.00	9.9671
128	283.25	
129	283.55	10.271
130	283.62	
131	283.75	10.644
132	283.80	
133	283.85	10.928
134	283.92	
135	284.00	11.218
136	284.12	
137	284.25	11.487
138	284.40	
139	284.55	11.680
140	285	
141	286	
142	286.55	12.212
143	288.55	12.445
144	294.55	12.767
145	304.55	13.048
146	314.55	13.229
147	359.90	13.643
148	429.90	13.944

E10

Table 2. Partial Listing of FMT File "AM_SPC_BM.TITRATE" (many species with zero concentrations throughout have been deleted; occurrances of "0.00000E+00" have been replaced with "0.0")

```
Benchmark:  Titrating Am(III)/SPC with NaOH, Closed System                FMT V1.3
DATABASE:  HMW84/FW86; Np(V)-Na-CO3-OH-Cl-ClO4 (NR94);
95.01.31   Am(III)-Na-Cl-CO3-SO4-PO4 (FRSR89,FRF90,P91,RFFR92,RFF94,RRFF94)

Titrant Volumes per Grid Block, in milliliters
  1              0.000000 mL
  2              0.050000 mL
  3              0.100000 mL
  4              0.200000 mL
  5              0.300000 mL
  6              0.400000 mL
  7              0.500000 mL
  8              0.602470 mL
  9              1.530200 mL
 10              1.800000 mL
 11              3.225700 mL
 12             43.867000 mL
 13             76.741000 mL
 14            161.770000 mL
 15            194.920000 mL
 16            234.870000 mL
 17            246.900000 mL
 18            258.940000 mL
 19            266.960000 mL
 20            270.970000 mL
 21            274.980000 mL
 22            278.990000 mL
 23            280.000000 mL
 24            280.500000 mL
 25            281.000000 mL
 26            281.500000 mL
 27            282.000000 mL
 28            282.500000 mL
 29            283.000000 mL
 30            283.250000 mL
 31            283.550000 mL
 32            283.620000 mL
 33            283.750000 mL
 34            283.800000 mL
 35            283.850000 mL
 36            283.920000 mL
 37            284.000000 mL
 38            284.120000 mL
 39            284.250000 mL
 40            284.400000 mL
 41            284.550000 mL
 42            285.000000 mL
 43            286.000000 mL
 44            286.550000 mL
 45            288.550000 mL
 46            294.550000 mL
 47            304.550000 mL
 48            314.550000 mL
 49            359.900000 mL
 50            429.900000 mL


Titration Results, molal
```

	H2O	Na+	K+	Ca++	Mg-+
1)	4.38782E+01	2.00000	8.39999E-01	1.62646E-02	1.55135
2)	4.38806E+01	2.00044	8.39952E-01	1.62574E-02	1.55085
3)	4.38831E+01	2.00088	8.39906E-01	1.62502E-02	1.55035
4)	4.38879E+01	2.00177	8.39813E-01	1.62359E-02	1.54933
5)	4.38928E+01	2.00265	8.39721E-01	1.62219E-02	1.54831
6)	4.38976E+01	2.00354	8.39628E-01	1.62081E-02	1.54729
7)	4.39024E+01	2.00442	8.39535E-01	1.61947E-02	1.54626
8)	4.39074E+01	2.00533	8.39440E-01	1.61816E-02	1.54519
9)	4.39525E+01	2.01353	8.38580E-01	1.61571E-02	1.53826
10)	4.39652E+01	2.01592	8.38337E-01	1.61361E-02	1.53560
11)	4.40178E+01	2.02925	8.37335E-01	1.61155E-02	1.52331
12)	4.55171E+01	2.39606	8.09753E-01	1.55411E-02	1.18502
13)	4.67463E+01	2.67461	7.88462E-01	1.50876E-02	9.29541E-01
14)	5.17350E+01	3.21494	7.12432E-01	1.35643E-02	5.57930E-01
15)	5.33450E+01	3.41972	6.90930E-01	1.30985E-02	3.91494E-01
16)	5.52853E+01	3.65066	6.66681E-01	1.25375E-02	2.04441E-01
17)	5.58696E+01	3.71706	6.59709E-01	1.23619E-02	1.50909E-01
18)	5.64543E+01	3.78214	6.52875E-01	1.21774E-02	9.86490E-02
19)	5.68439E+01	3.82475	6.48402E-01	1.20453E-02	6.46119E-02
20)	5.70386E+01	3.84583	6.46188E-01	1.19744E-02	4.73503E-02
21)	5.72334E+01	3.86678	6.43989E-01	1.18977E-02	3.12884E-02
22)	5.74281E+01	3.88758	6.41805E-01	8.21339E-03	1.85913E-02
23)	5.74772E+01	3.89279	6.41257E-01	8.19392E-03	1.45207E-02
24)	5.75014E+01	3.89537	6.40986E-01	8.18386E-03	1.25155E-02
25)	5.75257E+01	3.89795	6.40716E-01	8.17345E-03	1.05183E-02
26)	5.75500E+01	3.90052	6.40446E-01	8.16258E-03	8.53072E-03
27)	5.75743E+01	3.90310	6.40175E-01	8.15112E-03	6.55565E-03
28)	5.75986E+01	3.90567	6.39906E-01	8.13880E-03	4.59827E-03
29)	5.76229E+01	3.90823	6.39636E-01	8.12505E-03	2.67114E-03
30)	5.76350E+01	3.90952	6.39501E-01	8.11724E-03	1.73020E-03
31)	5.76496E+01	3.91106	6.39339E-01	8.10615E-03	6.53682E-04
32)	5.76530E+01	3.91142	6.39302E-01	8.10311E-03	4.34696E-04
33)	5.76593E+01	3.91208	6.39232E-01	8.09722E-03	1.18249E-04
34)	5.76617E+01	3.91234	6.39205E-01	8.09548E-03	6.01448E-05
35)	5.76641E+01	3.91260	6.39178E-01	8.09432E-03	3.20357E-05
36)	5.76675E+01	3.91295	6.39140E-01	8.09333E-03	1.56752E-05
37)	5.76714E+01	3.91336	6.39097E-01	8.09267E-03	8.43438E-06
38)	5.76773E+01	3.91398	6.39033E-01	8.09205E-03	4.24706E-06
39)	5.76836E+01	3.91465	6.38963E-01	8.09158E-03	2.44616E-06
40)	5.76909E+01	3.91541	6.38882E-01	8.09115E-03	1.49499E-06
41)	5.76981E+01	3.91618	6.38801E-01	8.09077E-03	1.00689E-06
42)	5.77200E+01	3.91849	6.38559E-01	8.08980E-03	4.25698E-07
43)	5.77686E+01	3.92360	6.38023E-01	8.08786E-03	1.35104E-07
44)	5.77953E+01	3.92641	6.37728E-01	8.08681E-03	8.79760E-08
45)	5.78924E+01	3.93660	6.36658E-01	8.08295E-03	3.05373E-08
46)	5.81838E+01	3.96697	6.33469E-01	5.25964E-03	7.12839E-09
47)	5.86695E+01	4.01691	6.28225E-01	1.58341E-03	2.05982E-09
48)	5.91552E+01	4.06603	6.23067E-01	7.58494E-04	9.54227E-10
49)	6.13577E+01	4.27904	6.00701E-01	1.70564E-04	1.81825E-10
50)	6.47575E+01	4.57939	5.69164E-01	7.51720E-05	6.37215E-11

	MgOH+	H+	Cl-	SO4=	HSO4-
1)	2.55665E-04	7.11823E-09	5.82999	4.35999E-02	1.53305E-09
2)	2.81591E-04	6.46004E-09	5.82967	4.35975E-02	1.39139E-09
3)	3.10256E-04	5.86061E-09	5.82935	4.35951E-02	1.26237E-09
4)	3.77543E-04	4.81190E-09	5.82870	4.35903E-02	1.03662E-09
5)	4.61964E-04	3.92911E-09	5.82806	4.35855E-02	8.46563E-10
6)	5.70019E-04	3.18149E-09	5.82742	4.35807E-02	6.85577E-10
7)	7.11271E-04	2.54743E-09	5.82677	4.35759E-02	5.49018E-10
8)	9.04951E-04	2.00041E-09	5.82611	4.35709E-02	4.31184E-10
9)	1.07864E-03	1.66820E-09	5.82014	4.35263E-02	3.60048E-10
10)	1.75426E-03	1.02344E-09	5.81835	4.35137E-02	2.20965E-10
11)	1.74458E-03	1.01765E-09	5.80615	4.34617E-02	2.20322E-10

12)	1.47691E-03	8.53033E-10	5.47021	4.20301E-02	2.00261E-10
13)	1.27216E-03	7.20480E-10	5.21729	4.09249E-02	1.81086E-10
14)	8.60518E-04	5.84293E-10	4.94462	3.69786E-02	1.58610E-10
15)	6.66582E-04	5.01404E-10	4.79538	3.58625E-02	1.42933E-10
16)	4.39112E-04	3.72291E-10	4.62708	3.46039E-02	1.12697E-10
17)	3.67028E-04	3.22336E-10	4.57869	3.42420E-02	9.93750E-11
18)	2.88739E-04	2.62575E-10	4.53127	3.38873E-02	8.24470E-11
19)	2.29483E-04	2.13535E-10	4.50022	3.36551E-02	6.78677E-11
20)	1.95714E-04	1.84197E-10	4.48485	3.35402E-02	5.88969E-11
21)	1.56846E-04	1.49293E-10	4.46959	3.34261E-02	4.80214E-11
22)	1.19780E-04	1.15330E-10	4.45443	3.33127E-02	3.73290E-11
23)	1.05621E-04	1.01982E-10	4.45063	3.32843E-02	3.30562E-11
24)	9.79492E-05	9.47046E-11	4.44875	3.32703E-02	3.07190E-11
25)	8.96954E-05	8.68433E-11	4.44687	3.32562E-02	2.81887E-11
26)	8.06887E-05	7.82298E-11	4.44500	3.32422E-02	2.54103E-11
27)	7.06563E-05	6.85961E-11	4.44312	3.32282E-02	2.22962E-11
28)	5.91107E-05	5.74643E-11	4.44125	3.32141E-02	1.86902E-11
29)	4.50036E-05	4.38080E-11	4.43938	3.32001E-02	1.42573E-11
30)	3.62006E-05	3.52615E-11	4.43844	3.31932E-02	1.14790E-11
31)	2.23220E-05	2.17588E-11	4.43732	3.31848E-02	7.08512E-12
32)	1.81313E-05	1.76764E-11	4.43706	3.31828E-02	5.75593E-12
33)	9.45449E-06	9.21908E-12	4.43657	3.31792E-02	3.00166E-12
34)	6.74232E-06	6.57458E-12	4.43639	3.31778E-02	2.14033E-12
35)	4.92043E-06	4.79799E-12	4.43620	3.31764E-02	1.56168E-12
36)	3.44161E-06	3.35589E-12	4.43594	3.31744E-02	1.09197E-12
37)	2.52434E-06	2.46136E-12	4.43564	3.31722E-02	8.00616E-13
38)	1.79108E-06	1.74629E-12	4.43519	3.31688E-02	5.67710E-13
39)	1.35913E-06	1.32504E-12	4.43470	3.31652E-02	4.30507E-13
40)	1.06237E-06	1.03563E-12	4.43414	3.31610E-02	3.36247E-13
41)	8.71736E-07	8.49726E-13	4.43358	3.31568E-02	2.75697E-13
42)	5.66579E-07	5.52132E-13	4.43191	3.31443E-02	1.78770E-13
43)	3.18885E-07	3.10577E-13	4.42818	3.31164E-02	1.00097E-13
44)	2.57191E-07	2.50412E-13	4.42613	3.31011E-02	8.05030E-14
45)	1.51241E-07	1.47089E-13	4.41871	3.30456E-02	4.68551E-14
46)	7.25725E-08	7.05292E-14	4.39658	3.28801E-02	2.19613E-14
47)	3.86807E-08	3.75178E-14	4.36018	3.26079E-02	1.12350E-14
48)	2.60103E-08	2.51216E-14	4.32438	3.23401E-02	7.20758E-15
49)	1.08856E-08	1.03230E-14	4.16915	3.11792E-02	2.45184E-15
50)	6.05116E-09	5.63204E-15	3.95027	2.95423E-02	1.02625E-15

	OH-	HCO3-	CO3=	CO2(aq)	CaCO3(aq)
1)	1.26537E-06	5.40709E-07	8.79182E-08	5.85994E-09	6.51778E-09
2)	1.39415E-06	5.95865E-07	1.06724E-07	5.86072E-09	7.91101E-09
3)	1.53659E-06	6.56882E-07	1.29645E-07	5.86151E-09	9.60889E-09
4)	1.87111E-06	8.00221E-07	1.92235E-07	5.86309E-09	1.42445E-08
5)	2.29107E-06	9.80237E-07	2.88207E-07	5.86468E-09	2.13510E-08
6)	2.82891E-06	1.21086E-06	4.39402E-07	5.86628E-09	3.25445E-08
7)	3.53239E-06	1.51261E-06	6.85107E-07	5.86791E-09	5.07318E-08
8)	4.49755E-06	1.92674E-06	1.11063E-06	5.86960E-09	8.22250E-08
9)	5.38778E-06	2.31540E-06	1.59358E-06	5.87811E-09	1.18426E-07
10)	8.77915E-06	1.42333E-06	1.59449E-06	2.21667E-09	1.18513E-07
11)	8.81863E-06	1.42747E-06	1.59690E-06	2.20723E-09	1.19619E-07
12)	1.01031E-05	1.56646E-06	1.67561E-06	1.93845E-09	1.58597E-07
13)	1.14699E-05	1.71614E-06	1.77027E-06	1.72150E-09	2.05786E-07
14)	1.24931E-05	2.36129E-06	2.00207E-06	1.81459E-09	3.25626E-07
15)	1.38237E-05	2.92380E-06	2.37072E-06	1.87045E-09	4.59611E-07
16)	1.74320E-05	4.21955E-06	3.59873E-06	1.93406E-09	8.65550E-07
17)	1.97253E-05	4.97171E-06	4.53830E-06	1.95238E-09	1.16485E-06
18)	2.37170E-05	6.22395E-06	6.45852E-06	1.97028E-09	1.76802E-06
19)	2.87595E-05	7.75219E-06	9.39579E-06	1.98191E-09	2.68256E-06
20)	3.31073E-05	9.04418E-06	1.23845E-05	1.98762E-09	3.60911E-06
21)	4.05620E-05	1.12294E-05	1.84898E-05	1.99324E-09	5.49648E-06
22)	5.20309E-05	1.27563E-05	2.63991E-05	1.73669E-09	5.55951E-06
23)	5.87377E-05	1.13057E-05	2.62899E-05	1.35984E-09	5.55951E-06

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24)	6.31962E-05	1.05114E-05	2.62379E-05	1.17357E-09	5.55951E-06
25)	6.88570E-05	9.65090E-06	2.61875E-05	9.87613E-10	5.55951E-06
26)	7.63723E-05	8.70507E-06	2.61391E-05	8.02108E-10	5.55951E-06
27)	8.70228E-05	7.64375E-06	2.60936E-05	6.17303E-10	5.55951E-06
28)	1.03791E-04	6.41308E-06	2.60520E-05	4.33670E-10	5.55951E-06
29)	1.36031E-04	4.89751E-06	2.60171E-05	2.52360E-10	5.55951E-06
30)	1.68931E-04	3.94606E-06	2.60045E-05	1.63625E-10	5.55951E-06
31)	2.73632E-04	2.43870E-06	2.59997E-05	6.23797E-11	5.55951E-06
32)	3.36794E-04	1.98206E-06	2.60024E-05	4.11838E-11	5.55951E-06
33)	6.45671E-04	1.03487E-06	2.60183E-05	1.12124E-11	5.55951E-06
34)	9.05363E-04	7.38384E-07	2.60295E-05	5.70467E-12	5.55951E-06
35)	1.24060E-03	5.39128E-07	2.60423E-05	3.03934E-12	5.55951E-06
36)	1.77373E-03	3.77344E-07	2.60609E-05	1.48762E-12	5.55951E-06
37)	2.41840E-03	2.76971E-07	2.60820E-05	8.00692E-13	5.55951E-06
38)	3.40882E-03	1.96723E-07	2.61134E-05	4.03353E-13	5.55951E-06
39)	4.49272E-03	1.49446E-07	2.61473E-05	2.32419E-13	5.55951E-06
40)	5.74847E-03	1.16964E-07	2.61861E-05	1.42115E-13	5.55951E-06
41)	7.00648E-03	9.60974E-08	2.62250E-05	9.57630E-14	5.55951E-06
42)	1.07845E-02	6.26947E-08	2.63414E-05	4.05461E-14	5.55951E-06
43)	1.91784E-02	3.55829E-08	2.66010E-05	1.29094E-14	5.55951E-06
44)	2.37904E-02	2.88310E-08	2.67445E-05	8.42101E-15	5.55951E-06
45)	4.05274E-02	1.72390E-08	2.72716E-05	2.94170E-15	5.55951E-06
46)	8.44173E-02	1.33335E-08	4.38633E-05	1.07195E-15	5.55951E-06
47)	1.58534E-01	2.55282E-08	1.57594E-04	1.06129E-15	5.55951E-06
48)	2.37202E-01	3.86993E-08	3.59049E-04	1.04953E-15	5.55951E-06
49)	5.82483E-01	9.99097E-08	2.33879E-03	9.98330E-16	5.55951E-06
50)	1.07428	7.59185E-08	3.43378E-03	3.57974E-16	2.14224E-06

	MgCO3(aq)	B(OH)3(aq)	B(OH)4-	B3O3(OH)4-	B4O5(OH)4=
1)	6.87083E-07	1.04858E-02	2.69612E-03	2.47784E-05	5.25513E-06
2)	8.33992E-07	9.96466E-03	2.82280E-03	2.34207E-05	5.20045E-06
3)	1.01303E-06	9.44578E-03	2.94911E-03	2.19799E-05	5.09878E-06
4)	1.50188E-06	8.41729E-03	3.19995E-03	1.89268E-05	4.76371E-06
5)	2.25131E-06	7.40588E-03	3.44715E-03	1.57738E-05	4.27658E-06
6)	3.43178E-06	6.41875E-03	3.68885E-03	1.26721E-05	3.67630E-06
7)	5.34978E-06	5.46620E-03	3.92240E-03	9.76611E-06	3.01239E-06
8)	8.67073E-06	4.54180E-03	4.14930E-03	7.12806E-06	2.32563E-06
9)	1.24349E-05	3.93649E-03	4.30568E-03	5.54564E-06	1.87046E-06
10)	1.24349E-05	2.60171E-03	4.63617E-03	2.60543E-06	9.45585E-07
11)	1.24349E-05	2.60535E-03	4.65684E-03	2.61550E-06	9.46721E-07
12)	1.24349E-05	2.69623E-03	5.31677E-03	2.93276E-06	9.99342E-07
13)	1.24349E-05	2.74312E-03	5.98578E-03	3.23344E-06	1.07457E-06
14)	1.24349E-05	2.91505E-03	6.90539E-03	4.04073E-06	1.23624E-06
15)	1.24349E-05	2.99128E-03	7.78130E-03	4.71871E-06	1.47639E-06
16)	1.24349E-05	2.90228E-03	9.45651E-03	5.34437E-06	1.81932E-06
17)	1.24349E-05	2.77606E-03	1.02194E-02	5.27785E-06	1.88032E-06
18)	1.24349E-05	2.53706E-03	1.12145E-02	4.83554E-06	1.83184E-06
19)	1.24349E-05	2.25697E-03	1.20880E-02	4.12543E-06	1.65018E-06
20)	1.24349E-05	2.04888E-03	1.26278E-02	3.55219E-06	1.46933E-06
21)	1.24349E-05	1.75902E-03	1.32779E-02	2.75358E-06	1.18565E-06
22)	1.08050E-05	1.45125E-03	1.40749E-02	1.98747E-06	8.98277E-07
23)	8.45467E-06	1.30851E-03	1.43252E-02	1.64455E-06	7.54654E-07
24)	7.29413E-06	1.22771E-03	1.44602E-02	1.46138E-06	6.76109E-07
25)	6.13630E-06	1.13808E-03	1.46046E-02	1.26835E-06	5.91958E-07
26)	4.98208E-06	1.03711E-03	1.47609E-02	1.06458E-06	5.01578E-07
27)	3.83297E-06	9.20833E-04	1.49330E-02	8.49027E-07	4.04212E-07
28)	2.69190E-06	7.82161E-04	1.51276E-02	6.20543E-07	2.98941E-07
29)	1.56598E-06	6.05954E-04	1.53593E-02	3.78136E-07	1.84748E-07
30)	1.01521E-06	4.92415E-04	1.54997E-02	2.51983E-07	1.24172E-07
31)	3.86976E-07	3.08202E-04	1.57134E-02	1.00069E-07	4.99635E-08
32)	2.55480E-07	2.51400E-04	1.57758E-02	6.68455E-08	3.35047E-08
33)	6.95553E-08	1.32185E-04	1.59016E-02	1.86265E-08	9.40972E-09
34)	3.53895E-08	9.44983E-05	1.59398E-02	9.54212E-09	4.83217E-09
35)	1.88557E-08	6.90740E-05	1.59651E-02	5.10627E-09	2.59007E-09

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36)  9.22973E-09  4.83749E-05  1.59851E-02  2.50756E-09  1.27363E-09
37)  4.96823E-09  3.55082E-05  1.59971E-02  1.35202E-09  6.87314E-10
38)  2.50313E-09  2.52076E-05  1.60062E-02  6.81751E-10  3.46834E-10
39)  1.44257E-09  1.91335E-05  1.60110E-02  3.92887E-10  1.99976E-10
40)  8.82231E-10  1.49578E-05  1.60138E-02  2.40147E-10  1.22282E-10
41)  5.94589E-10  1.22743E-05  1.60151E-02  1.61718E-10  8.23714E-11
42)  2.51884E-10  7.97680E-06  1.60154E-02  6.82953E-11  3.48106E-11
43)  8.02925E-11  4.48690E-06  1.60100E-02  2.15972E-11  1.10212E-11
44)  5.24103E-11  3.61744E-06  1.60060E-02  1.40330E-11  7.16530E-12
45)  1.83519E-11  2.12405E-06  1.59898E-02  4.83142E-12  2.47189E-12
46)  6.72627E-12  1.02909E-06  1.61062E-02  1.14137E-12  5.91957E-13
47)  6.72627E-12  5.53603E-07  1.62098E-02  3.31935E-13  1.75288E-13
48)  6.72627E-12  3.70238E-07  1.61285E-02  1.47462E-13  7.86026E-14
49)  6.72627E-12  1.49663E-07  1.55831E-02  2.31021E-14  1.27322E-14
50)  2.59182E-12  8.00622E-08  1.47694E-02  6.20850E-15  3.60463E-15

     CaB(OH)4+    MgB(OH)4+    Br-          ClO4-        NaOH(aq).....
 1)  1.35362E-04  8.38732E-03  1.09000E-02  0.0          0.0
 2)  1.41666E-04  8.77857E-03  1.08994E-02  0.0          0.0
 3)  1.47946E-04  9.16839E-03  1.08988E-02  0.0          0.0
 4)  1.60403E-04  9.94168E-03  1.08976E-02  0.0          0.0
 5)  1.72659E-04  1.07026E-02  1.08964E-02  0.0          0.0
 6)  1.84622E-04  1.14454E-02  1.08952E-02  0.0          0.0
 7)  1.96162E-04  1.21618E-02  1.08940E-02  0.0          0.0
 8)  2.07355E-04  1.28563E-02  1.08927E-02  0.0          0.0
 9)  2.15032E-04  1.32818E-02  1.08816E-02  0.0          0.0
10)  2.31265E-04  1.42761E-02  1.08784E-02  0.0          0.0
11)  2.32356E-04  1.42247E-02  1.08654E-02  0.0          0.0
12)  2.68199E-04  1.27210E-02  1.05075E-02  0.0          0.0
13)  3.05979E-04  1.14136E-02  1.02312E-02  0.0          0.0
14)  3.44781E-04  8.30701E-03  9.24465E-03  0.0          0.0
15)  3.90625E-04  6.74800E-03  8.96563E-03  0.0          0.0
16)  4.77776E-04  4.44207E-03  8.65098E-03  0.0          0.0
17)  5.16957E-04  3.58525E-03  8.56051E-03  0.0          0.0
18)  5.67483E-04  2.60284E-03  8.47184E-03  0.0          0.0
19)  6.11252E-04  1.85239E-03  8.41378E-03  0.0          0.0
20)  6.38007E-04  1.43886E-03  8.38506E-03  0.0          0.0
21)  6.69892E-04  9.93219E-04  8.35652E-03  0.0          0.0
22)  4.92528E-04  6.28100E-04  8.32818E-03  0.0          0.0
23)  5.00702E-04  4.99780E-04  8.32108E-03  0.0          0.0
24)  5.05098E-04  4.35029E-04  8.31756E-03  0.0          0.0
25)  5.09792E-04  3.69431E-04  8.31405E-03  0.0          0.0
26)  5.14859E-04  3.02969E-04  8.31054E-03  0.0          0.0
27)  5.20422E-04  2.35644E-04  8.30704E-03  0.0          0.0
28)  5.26694E-04  1.67512E-04  8.30354E-03  0.0          0.0
29)  5.34124E-04  9.88380E-05  8.30004E-03  0.0          0.0
30)  5.38606E-04  6.46180E-05  8.29829E-03  0.0          0.0
31)  5.45372E-04  2.49426E-05  8.29619E-03  0.0          0.0
32)  5.47325E-04  1.65263E-05  8.29570E-03  0.0          0.0
33)  5.51144E-04  4.53092E-06  8.29479E-03  0.0          0.0
34)  5.52203E-04  2.30979E-06  8.29444E-03  0.0          0.0
35)  5.52804E-04  1.23202E-06  8.29409E-03  0.0          0.0
36)  5.53124E-04  6.03428E-07  8.29360E-03  0.0          0.0
37)  5.53124E-04  3.24824E-07  8.29305E-03  0.0          0.0
38)  5.52827E-04  1.63573E-07  8.29221E-03  0.0          0.0
39)  5.52339E-04  9.41888E-08  8.29130E-03  0.0          0.0
40)  5.51687E-04  5.75375E-08  8.29025E-03  0.0          0.0
41)  5.50988E-04  3.87306E-08  8.28921E-03  0.0          0.0
42)  5.48778E-04  1.63437E-08  8.28607E-03  0.0          0.0
43)  5.43719E-04  5.16336E-09  8.27910E-03  0.0          0.0
44)  5.40927E-04  3.35359E-09  8.27528E-03  0.0          0.0
45)  5.30869E-04  1.15314E-09  8.26139E-03  0.0          0.0
46)  3.32809E-04  2.65430E-10  8.22001E-03  0.0          0.0
47)  9.35434E-05  7.48227E-11  8.15197E-03  0.0          0.0
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  48)  4.11683E-05   3.30238E-11   8.08504E-03   0.0           0.0
  49)  6.35065E-06   5.15793E-12   7.79481E-03   0.0           0.0
  50)  1.67539E-06   1.38477E-12   7.38558E-03   0.0           0.0

       HCl(aq)...... HClO4(aq).... PosIon....... NegIon....... PosIon(OH)(aq
   1)  0.0           0.0           0.0           5.31999E-02   0.0
   2)  0.0           0.0           0.0           5.31970E-02   0.0
   3)  0.0           0.0           0.0           5.31941E-02   0.0
   4)  0.0           0.0           0.0           5.31882E-02   0.0
   5)  0.0           0.0           0.0           5.31823E-02   0.0
   6)  0.0           0.0           0.0           5.31764E-02   0.0
   7)  0.0           0.0           0.0           5.31706E-02   0.0
   8)  0.0           0.0           0.0           5.31645E-02   0.0
   9)  0.0           0.0           0.0           5.31100E-02   0.0
  10)  0.0           0.0           0.0           5.30947E-02   0.0
  11)  0.0           0.0           0.0           5.30312E-02   0.0
  12)  0.0           0.0           0.0           5.12844E-02   0.0
  13)  0.0           0.0           0.0           4.99359E-02   0.0
  14)  0.0           0.0           0.0           4.51207E-02   0.0
  15)  0.0           0.0           0.0           4.37589E-02   0.0
  16)  0.0           0.0           0.0           4.22231E-02   0.0
  17)  0.0           0.0           0.0           4.17815E-02   0.0
  18)  0.0           0.0           0.0           4.13488E-02   0.0
  19)  0.0           0.0           0.0           4.10654E-02   0.0
  20)  0.0           0.0           0.0           4.09252E-02   0.0
  21)  0.0           0.0           0.0           4.07860E-02   0.0
  22)  0.0           0.0           0.0           4.06477E-02   0.0
  23)  0.0           0.0           0.0           4.06130E-02   0.0
  24)  0.0           0.0           0.0           4.05958E-02   0.0
  25)  0.0           0.0           0.0           4.05787E-02   0.0
  26)  0.0           0.0           0.0           4.05616E-02   0.0
  27)  0.0           0.0           0.0           4.05444E-02   0.0
  28)  0.0           0.0           0.0           4.05273E-02   0.0
  29)  0.0           0.0           0.0           4.05103E-02   0.0
  30)  0.0           0.0           0.0           4.05017E-02   0.0
  31)  0.0           0.0           0.0           4.04915E-02   0.0
  32)  0.0           0.0           0.0           4.04891E-02   0.0
  33)  0.0           0.0           0.0           4.04847E-02   0.0
  34)  0.0           0.0           0.0           4.04830E-02   0.0
  35)  0.0           0.0           0.0           4.04813E-02   0.0
  36)  0.0           0.0           0.0           4.04789E-02   0.0
  37)  0.0           0.0           0.0           4.04762E-02   0.0
  38)  0.0           0.0           0.0           4.04721E-02   0.0
  39)  0.0           0.0           0.0           4.04676E-02   0.0
  40)  0.0           0.0           0.0           4.04625E-02   0.0
  41)  0.0           0.0           0.0           4.04574E-02   0.0
  42)  0.0           0.0           0.0           4.04421E-02   0.0
  43)  0.0           0.0           0.0           4.04081E-02   0.0
  44)  0.0           0.0           0.0           4.03894E-02   0.0
  45)  0.0           0.0           0.0           4.03216E-02   0.0
  46)  0.0           0.0           0.0           4.01197E-02   0.0
  47)  0.0           0.0           0.0           3.97876E-02   0.0
  48)  0.0           0.0           0.0           3.94609E-02   0.0
  49)  0.0           0.0           0.0           3.80444E-02   0.0
  50)  0.0           0.0           0.0           3.60471E-02   0.0

       NpO2(CO3)2=-  NpO2(CO3)3==- Am+++         AmCO3+        Am(CO3)2-
   1)  0.0           0.0           4.69972E-06   1.32228E-08   2.78507E-12
   2)  0.0           0.0           3.51331E-06   1.19984E-08   3.07095E-12
   3)  0.0           0.0           2.62361E-06   1.08835E-08   3.38739E-12
   4)  0.0           0.0           1.45255E-06   8.93330E-09   4.13136E-12
   5)  0.0           0.0           7.91003E-07   7.29223E-09   5.06664E-12
   6)  0.0           0.0           4.20051E-07   5.90292E-09   6.26601E-12
   7)  0.0           0.0           2.15690E-07   4.72506E-09   7.83668E-12
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  8)  0.0          0.0          1.04472E-07  3.70926E-09  9.99423E-12
  9)  0.0          0.0          6.04973E-08  3.08447E-09  1.21471E-11
 10)  0.0          0.0          1.39733E-08  7.12478E-10  2.82321E-12
 11)  0.0          0.0          1.36743E-08  6.99353E-10  2.86690E-12
 12)  0.0          0.0          7.11880E-09  4.06674E-10  4.58746E-12
 13)  0.0          0.0          3.92672E-09  2.56172E-10  7.04046E-12
 14)  0.0          0.0          2.04367E-09  1.83639E-10  2.67559E-11
 15)  0.0          0.0          1.24659E-09  1.47272E-10  5.42288E-11
 16)  0.0          0.0          4.91984E-10  1.01395E-10  1.45626E-10
 17)  0.0          0.0          3.16296E-10  8.59203E-11  2.08083E-10
 18)  0.0          0.0          1.69496E-10  6.85360E-11  3.16769E-10
 19)  0.0          0.0          9.06895E-11  5.49777E-11  4.50028E-10
 20)  0.0          0.0          5.80746E-11  4.71050E-11  5.60898E-10
 21)  0.0          0.0          3.08566E-11  3.79242E-11  7.44075E-10
 22)  0.0          0.0          1.42076E-11  2.52921E-11  7.81774E-10
 23)  0.0          0.0          9.81970E-12  1.74709E-11  5.51259E-10
 24)  0.0          0.0          7.86270E-12  1.39857E-11  4.45833E-10
 25)  0.0          0.0          6.06181E-12  1.07802E-11  3.47199E-10
 26)  0.0          0.0          4.43054E-12  7.87793E-12  2.56358E-10
 27)  0.0          0.0          2.98674E-12  5.31021E-12  1.74603E-10
 28)  0.0          0.0          1.75579E-12  3.12163E-12  1.03720E-10
 29)  0.0          0.0          7.77954E-13  1.38329E-12  4.64505E-11
 30)  0.0          0.0          4.05726E-13  7.21534E-13  2.43592E-11
 31)  0.0          0.0          9.53573E-14  1.69634E-13  5.76452E-12
 32)  0.0          0.0          5.11325E-14  9.09699E-14  3.09613E-12
 33)  0.0          0.0          7.25838E-15  1.29147E-14  4.40760E-13
 34)  0.0          0.0          2.63376E-15  4.68575E-15  1.60066E-13
 35)  0.0          0.0          1.02422E-15  1.82185E-15  6.22854E-14
 36)  0.0          0.0          3.50758E-16  6.23690E-16  2.13450E-14
 37)  0.0          0.0          1.38534E-16  2.46211E-16  8.43575E-15
 38)  0.0          0.0          4.95508E-17  8.79964E-17  3.01991E-15
 39)  0.0          0.0          2.16833E-17  3.84737E-17  1.32267E-15
 40)  0.0          0.0          1.03731E-17  1.83869E-17  6.33384E-16
 41)  0.0          0.0          5.74093E-18  1.01657E-17  3.50884E-16
 42)  0.0          0.0          1.58427E-18  2.79673E-18  9.71110E-17
 43)  0.0          0.0          2.85680E-19  5.00875E-19  1.76233E-17
 44)  0.0          0.0          1.50818E-19  2.63434E-19  9.33633E-18
 45)  0.0          0.0          3.13698E-20  5.40536E-20  1.96671E-18
 46)  0.0          0.0          3.70253E-21  9.43062E-21  5.71428E-19
 47)  0.0          0.0          6.25119E-22  4.95746E-21  1.13614E-18
 48)  0.0          0.0          2.11750E-22  3.28194E-21  1.77478E-18
 49)  0.0          0.0          2.47128E-23  1.28543E-21  5.15240E-18
 50)  0.0          0.0          8.08252E-24  2.53402E-22  1.76252E-18

      Am(CO3)3=-   Am(OH)2+     Am(OH)3(aq)  Th++++       UO2++
  1)  7.21634E-14  1.45075E-09  1.80328E-13  0.0          0.0
  2)  9.64966E-14  1.31653E-09  1.80328E-13  0.0          0.0
  3)  1.29173E-13  1.19429E-09  1.80328E-13  0.0          0.0
  4)  2.33146E-13  9.80462E-10  1.80328E-13  0.0          0.0
  5)  4.27837E-13  8.00485E-10  1.80328E-13  0.0          0.0
  6)  8.05119E-13  6.48087E-10  1.80328E-13  0.0          0.0
  7)  1.56694E-12  5.18856E-10  1.80328E-13  0.0          0.0
  8)  3.23308E-12  4.07381E-10  1.80328E-13  0.0          0.0
  9)  5.54081E-12  3.39148E-10  1.80328E-13  0.0          0.0
 10)  1.28181E-12  2.07965E-10  1.80328E-13  0.0          0.0
 11)  1.26510E-12  2.06011E-10  1.80328E-13  0.0          0.0
 12)  8.64432E-13  1.56309E-10  1.80328E-13  0.0          0.0
 13)  6.31870E-13  1.23180E-10  1.80328E-13  0.0          0.0
 14)  5.63179E-13  9.53118E-11  1.80328E-13  0.0          0.0
 15)  6.19175E-13  7.94147E-11  1.80328E-13  0.0          0.0
 16)  9.34248E-13  5.72410E-11  1.80328E-13  0.0          0.0
 17)  1.23373E-12  4.91759E-11  1.80328E-13  0.0          0.0
 18)  1.94827E-12  3.97673E-11  1.80328E-13  0.0          0.0
 19)  3.25357E-12  3.21913E-11  1.80328E-13  0.0          0.0
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     20)  4.80157E-12   2.77065E-11   1.80328E-13   0.0          0.0
     21)  8.54061E-12   2.24071E-11   1.80328E-13   0.0          0.0
     22)  1.15067E-11   1.72749E-11   1.80328E-13   0.0          0.0
     23)  7.86455E-12   1.52672E-11   1.80328E-13   0.0          0.0
     24)  6.26357E-12   1.41740E-11   1.80328E-13   0.0          0.0
     25)  4.80391E-12   1.29940E-11   1.80328E-13   0.0          0.0
     26)  3.49363E-12   1.17020E-11   1.80328E-13   0.0          0.0
     27)  2.34402E-12   1.02582E-11   1.80328E-13   0.0          0.0
     28)  1.37199E-12   8.59121E-12   1.80328E-13   0.0          0.0
     29)  6.05663E-13   6.54774E-12   1.80328E-13   0.0          0.0
     30)  3.15436E-13   5.26961E-12   1.80328E-13   0.0          0.0
     31)  7.40868E-14   3.25114E-12   1.80328E-13   0.0          0.0
     32)  3.97344E-14   2.64104E-12   1.80328E-13   0.0          0.0
     33)  5.64716E-15   1.37728E-12   1.80328E-13   0.0          0.0
     34)  2.05075E-15   9.82149E-13   1.80328E-13   0.0          0.0
     35)  7.98175E-16   7.16707E-13   1.80328E-13   0.0          0.0
     36)  2.73666E-16   5.01243E-13   1.80328E-13   0.0          0.0
     37)  1.08225E-16   3.67594E-13   1.80328E-13   0.0          0.0
     38)  3.87820E-17   2.60756E-13   1.80328E-13   0.0          0.0
     39)  1.70044E-17   1.97819E-13   1.80328E-13   0.0          0.0
     40)  8.15307E-18   1.54580E-13   1.80328E-13   0.0          0.0
     41)  4.52234E-18   1.26804E-13   1.80328E-13   0.0          0.0
     42)  1.25632E-18   8.23418E-14   1.80328E-13   0.0          0.0
     43)  2.29897E-19   4.62522E-14   1.80328E-13   0.0          0.0
     44)  1.22351E-19   3.72633E-14   1.80328E-13   0.0          0.0
     45)  0.0           2.18265E-14   1.80328E-13   0.0          0.0
     46)  0.0           1.03862E-14   1.80328E-13   0.0          0.0
     47)  8.20252E-20   5.45494E-15   1.80328E-13   0.0          0.0
     48)  2.86246E-19   3.60493E-15   1.80328E-13   0.0          0.0
     49)  5.04014E-18   1.40042E-15   1.80328E-13   0.0          0.0
     50)  2.25411E-18   7.09084E-16   1.80328E-13   0.0          0.0

          NpO2OH(aged)_ NpO2OH(amor)_ NaNpO2CO3(s)_ Na3NpO2(CO3)2 AmOHCO3(c)____
      1)  0.0           0.0           0.0           0.0           5.06967E-03
      2)  0.0           0.0           0.0           0.0           5.06917E-03
      3)  0.0           0.0           0.0           0.0           5.06862E-03
      4)  0.0           0.0           0.0           0.0           5.06736E-03
      5)  0.0           0.0           0.0           0.0           5.06577E-03
      6)  0.0           0.0           0.0           0.0           5.06364E-03
      7)  0.0           0.0           0.0           0.0           5.06060E-03
      8)  0.0           0.0           0.0           0.0           5.05584E-03
      9)  0.0           0.0           0.0           0.0           3.14873E-04
     10)  0.0           0.0           0.0           0.0           0.0
     11)  0.0           0.0           0.0           0.0           0.0
     12)  0.0           0.0           0.0           0.0           0.0
     13)  0.0           0.0           0.0           0.0           0.0
     14)  0.0           0.0           0.0           0.0           0.0
     15)  0.0           0.0           0.0           0.0           0.0
     16)  0.0           0.0           0.0           0.0           0.0
     17)  0.0           0.0           0.0           0.0           0.0
     18)  0.0           0.0           0.0           0.0           0.0
     19)  0.0           0.0           0.0           0.0           0.0
     20)  0.0           0.0           0.0           0.0           0.0
     21)  0.0           0.0           0.0           0.0           0.0
     22)  0.0           0.0           0.0           0.0           0.0
     23)  0.0           0.0           0.0           0.0           0.0
     24)  0.0           0.0           0.0           0.0           0.0
     25)  0.0           0.0           0.0           0.0           0.0
     26)  0.0           0.0           0.0           0.0           0.0
     27)  0.0           0.0           0.0           0.0           0.0
     28)  0.0           0.0           0.0           0.0           0.0
     29)  0.0           0.0           0.0           0.0           0.0
     30)  0.0           0.0           0.0           0.0           0.0
     31)  0.0           0.0           0.0           0.0           0.0
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32)  0.0            0.0           0.0          0.0           0.0
33)  0.0            0.0           0.0          0.0           0.0
34)  0.0            0.0           0.0          0.0           0.0
35)  0.0            0.0           0.0          0.0           0.0
36)  0.0            0.0           0.0          0.0           0.0
37)  0.0            0.0           0.0          0.0           0.0
38)  0.0            0.0           0.0          0.0           0.0
39)  0.0            0.0           0.0          0.0           0.0
40)  0.0            0.0           0.0          0.0           0.0
41)  0.0            0.0           0.0          0.0           0.0
42)  0.0            0.0           0.0          0.0           0.0
43)  0.0            0.0           0.0          0.0           0.0
44)  0.0            0.0           0.0          0.0           0.0
45)  0.0            0.0           0.0          0.0           0.0
46)  0.0            0.0           0.0          0.0           0.0
47)  0.0            0.0           0.0          0.0           0.0
48)  0.0            0.0           0.0          0.0           0.0
49)  0.0            0.0           0.0          0.0           0.0
50)  0.0            0.0           0.0          0.0           0.0

     Am(OH)3(s)___  NaAm(CO3)2.6H AmPO4(c)_____ CaSO4_________ NaK3(SO4)2__A
 1)  9.94924E-01    0.0           0.0          0.0           0.0
 2)  9.94871E-01    0.0           0.0          0.0           0.0
 3)  9.94817E-01    0.0           0.0          0.0           0.0
 4)  9.94709E-01    0.0           0.0          0.0           0.0
 5)  9.94601E-01    0.0           0.0          0.0           0.0
 6)  9.94493E-01    0.0           0.0          0.0           0.0
 7)  9.94386E-01    0.0           0.0          0.0           0.0
 8)  9.94278E-01    0.0           0.0          0.0           0.0
 9)  9.97994E-01    0.0           0.0          0.0           0.0
10)  9.98020E-01    0.0           0.0          0.0           0.0
11)  9.96827E-01    0.0           0.0          0.0           0.0
12)  9.63992E-01    0.0           0.0          0.0           0.0
13)  9.38645E-01    0.0           0.0          0.0           0.0
14)  8.48133E-01    0.0           0.0          0.0           0.0
15)  8.22535E-01    0.0           0.0          0.0           0.0
16)  7.93667E-01    0.0           0.0          0.0           0.0
17)  7.85367E-01    0.0           0.0          0.0           0.0
18)  7.77233E-01    0.0           0.0          0.0           0.0
19)  7.71907E-01    0.0           0.0          0.0           0.0
20)  7.69271E-01    0.0           0.0          0.0           0.0
21)  7.66654E-01    0.0           0.0          0.0           0.0
22)  7.64054E-01    0.0           0.0          0.0           0.0
23)  7.63402E-01    0.0           0.0          0.0           0.0
24)  7.63079E-01    0.0           0.0          0.0           0.0
25)  7.62757E-01    0.0           0.0          0.0           0.0
26)  7.62435E-01    0.0           0.0          0.0           0.0
27)  7.62114E-01    0.0           0.0          0.0           0.0
28)  7.61792E-01    0.0           0.0          0.0           0.0
29)  7.61471E-01    0.0           0.0          0.0           0.0
30)  7.61311E-01    0.0           0.0          0.0           0.0
31)  7.61118E-01    0.0           0.0          0.0           0.0
32)  7.61073E-01    0.0           0.0          0.0           0.0
33)  7.60990E-01    0.0           0.0          0.0           0.0
34)  7.60958E-01    0.0           0.0          0.0           0.0
35)  7.60926E-01    0.0           0.0          0.0           0.0
36)  7.60881E-01    0.0           0.0          0.0           0.0
37)  7.60830E-01    0.0           0.0          0.0           0.0
38)  7.60753E-01    0.0           0.0          0.0           0.0
39)  7.60670E-01    0.0           0.0          0.0           0.0
40)  7.60574E-01    0.0           0.0          0.0           0.0
41)  7.60478E-01    0.0           0.0          0.0           0.0
42)  7.60190E-01    0.0           0.0          0.0           0.0
43)  7.59551E-01    0.0           0.0          0.0           0.0
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     44)  7.59200E-01   0.0            0.0            0.0            0.0
     45)  7.57926E-01   0.0            0.0            0.0            0.0
     46)  7.54130E-01   0.0            0.0            0.0            0.0
     47)  7.47887E-01   0.0            0.0            0.0            0.0
     48)  7.41747E-01   0.0            0.0            0.0            0.0
     49)  7.15120E-01   0.0            0.0            0.0            0.0
     50)  6.77576E-01   0.0            0.0            0.0            0.0

          Mg(OH)2______ Na6CO3(SO4)2_  CaCO3________  CaCl2.4H2O___  Ca4Cl2(OH)6.1
      1)  0.0           0.0            0.0            0.0            0.0
      2)  0.0           0.0            0.0            0.0            0.0
      3)  0.0           0.0            0.0            0.0            0.0
      4)  0.0           0.0            0.0            0.0            0.0
      5)  0.0           0.0            0.0            0.0            0.0
      6)  0.0           0.0            0.0            0.0            0.0
      7)  0.0           0.0            0.0            0.0            0.0
      8)  0.0           0.0            0.0            0.0            0.0
      9)  0.0           0.0            0.0            0.0            0.0
     10)  0.0           0.0            0.0            0.0            0.0
     11)  0.0           0.0            0.0            0.0            0.0
     12)  0.0           0.0            0.0            0.0            0.0
     13)  7.28638E-03   0.0            0.0            0.0            0.0
     14)  7.51694E-01   0.0            0.0            0.0            0.0
     15)  8.80080E-01   0.0            0.0            0.0            0.0
     16)  1.02478       0.0            0.0            0.0            0.0
     17)  1.06634       0.0            0.0            0.0            0.0
     18)  1.10702       0.0            0.0            0.0            0.0
     19)  1.13359       0.0            0.0            0.0            0.0
     20)  1.14670       0.0            0.0            0.0            0.0
     21)  1.15969       0.0            0.0            0.0            0.0
     22)  1.17257       0.0            0.0            0.0            0.0
     23)  1.17577       0.0            3.81900E-03    0.0            0.0
     24)  1.17735       0.0            3.81961E-03    0.0            0.0
     25)  1.17892       0.0            3.81998E-03    0.0            0.0
     26)  1.18048       0.0            3.82042E-03    0.0            0.0
     27)  1.18203       0.0            3.82093E-03    0.0            0.0
     28)  1.18357       0.0            3.82156E-03    0.0            0.0
     29)  1.18508       0.0            3.82234E-03    0.0            0.0
     30)  1.18581       0.0            3.82339E-03    0.0            0.0
     31)  1.18664       0.0            3.82409E-03    0.0            0.0
     32)  1.18680       0.0            3.82526E-03    0.0            0.0
     33)  1.18701       0.0            3.82562E-03    0.0            0.0
     34)  1.18703       0.0            3.82631E-03    0.0            0.0
     35)  1.18701       0.0            3.82647E-03    0.0            0.0
     36)  1.18695       0.0            3.82651E-03    0.0            0.0
     37)  1.18688       0.0            3.82643E-03    0.0            0.0
     38)  1.18677       0.0            3.82626E-03    0.0            0.0
     39)  1.18664       0.0            3.82592E-03    0.0            0.0
     40)  1.18649       0.0            3.82551E-03    0.0            0.0
     41)  1.18634       0.0            3.82502E-03    0.0            0.0
     42)  1.18589       0.0            3.82451E-03    0.0            0.0
     43)  1.18490       0.0            3.82297E-03    0.0            0.0
     44)  1.18435       0.0            3.81950E-03    0.0            0.0
     45)  1.18236       0.0            3.81758E-03    0.0            0.0
     46)  1.17644       0.0            3.81060E-03    0.0            0.0
     47)  1.16670       0.0            3.77477E-03    0.0            0.0
     48)  1.15712       0.0            3.62937E-03    0.0            0.0
     49)  1.11559       0.0            3.39676E-03    0.0            0.0
     50)  1.05702       0.0            1.28194E-03    0.0            0.0

          MgSO4.H2O____ K2Mg(SO4)2.4H  Na4Ca(SO4)3.2  MgCO3________  Mg2Cl(OH)3.4H
      1)  0.0           0.0            0.0            0.0            0.0
      2)  0.0           0.0            0.0            0.0            0.0
      3)  0.0           0.0            0.0            0.0            0.0
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E20

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  4)  0.0             0.0             0.0             0.0             0.0
  5)  0.0             0.0             0.0             0.0             0.0
  6)  0.0             0.0             0.0             0.0             0.0
  7)  0.0             0.0             0.0             0.0             0.0
  8)  0.0             0.0             0.0             0.0             0.0
  9)  0.0             0.0             0.0             4.73110E-03     0.0
 10)  0.0             0.0             0.0             5.04540E-03     1.09719E-04
 11)  0.0             0.0             0.0             5.03934E-03     5.35795E-03
 12)  0.0             0.0             0.0             4.87258E-03     1.49861E-01
 13)  0.0             0.0             0.0             4.74375E-03     2.55008E-01
 14)  0.0             0.0             0.0             4.28377E-03     0.0
 15)  0.0             0.0             0.0             4.15290E-03     0.0
 16)  0.0             0.0             0.0             4.00358E-03     0.0
 17)  0.0             0.0             0.0             3.95950E-03     0.0
 18)  0.0             0.0             0.0             3.91447E-03     0.0
 19)  0.0             0.0             0.0             3.88208E-03     0.0
 20)  0.0             0.0             0.0             3.86351E-03     0.0
 21)  0.0             0.0             0.0             3.84006E-03     0.0
 22)  0.0             0.0             0.0             0.0             0.0
 23)  0.0             0.0             0.0             0.0             0.0
 24)  0.0             0.0             0.0             0.0             0.0
 25)  0.0             0.0             0.0             0.0             0.0
 26)  0.0             0.0             0.0             0.0             0.0
 27)  0.0             0.0             0.0             0.0             0.0
 28)  0.0             0.0             0.0             0.0             0.0
 29)  0.0             0.0             0.0             0.0             0.0
 30)  0.0             0.0             0.0             0.0             0.0
 31)  0.0             0.0             0.0             0.0             0.0
 32)  0.0             0.0             0.0             0.0             0.0
 33)  0.0             0.0             0.0             0.0             0.0
 34)  0.0             0.0             0.0             0.0             0.0
 35)  0.0             0.0             0.0             0.0             0.0
 36)  0.0             0.0             0.0             0.0             0.0
 37)  0.0             0.0             0.0             0.0             0.0
 38)  0.0             0.0             0.0             0.0             0.0
 39)  0.0             0.0             0.0             0.0             0.0
 40)  0.0             0.0             0.0             0.0             0.0
 41)  0.0             0.0             0.0             0.0             0.0
 42)  0.0             0.0             0.0             0.0             0.0
 43)  0.0             0.0             0.0             0.0             0.0
 44)  0.0             0.0             0.0             0.0             0.0
 45)  0.0             0.0             0.0             0.0             0.0
 46)  0.0             0.0             0.0             0.0             0.0
 47)  0.0             0.0             0.0             0.0             0.0
 48)  0.0             0.0             0.0             0.0             0.0
 49)  0.0             0.0             0.0             0.0             0.0
 50)  0.0             0.0             0.0             0.0             0.0

      MgCO3.3H2O___   K2Mg(SO4)2.6H   Na2Ca(CO3)2.2   K2MgCa2(SO4)4   Ca(OH)2_______
  1)  0.0             0.0             0.0             0.0             0.0
  2)  0.0             0.0             0.0             0.0             0.0
  3)  0.0             0.0             0.0             0.0             0.0
  4)  0.0             0.0             0.0             0.0             0.0
  5)  0.0             0.0             0.0             0.0             0.0
  6)  0.0             0.0             0.0             0.0             0.0
  7)  0.0             0.0             0.0             0.0             0.0
  8)  0.0             0.0             0.0             0.0             0.0
  9)  0.0             0.0             0.0             0.0             0.0
 10)  0.0             0.0             0.0             0.0             0.0
 11)  0.0             0.0             0.0             0.0             0.0
 12)  0.0             0.0             0.0             0.0             0.0
 13)  0.0             0.0             0.0             0.0             0.0
 14)  0.0             0.0             0.0             0.0             0.0
 15)  0.0             0.0             0.0             0.0             0.0
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E21

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 16)  0.0          0.0          0.0          0.0          0.0
 17)  0.0          0.0          0.0          0.0          0.0
 18)  0.0          0.0          0.0          0.0          0.0
 19)  0.0          0.0          0.0          0.0          0.0
 20)  0.0          0.0          0.0          0.0          0.0
 21)  0.0          0.0          0.0          0.0          0.0
 22)  0.0          0.0          0.0          0.0          0.0
 23)  0.0          0.0          0.0          0.0          0.0
 24)  0.0          0.0          0.0          0.0          0.0
 25)  0.0          0.0          0.0          0.0          0.0
 26)  0.0          0.0          0.0          0.0          0.0
 27)  0.0          0.0          0.0          0.0          0.0
 28)  0.0          0.0          0.0          0.0          0.0
 29)  0.0          0.0          0.0          0.0          0.0
 30)  0.0          0.0          0.0          0.0          0.0
 31)  0.0          0.0          0.0          0.0          0.0
 32)  0.0          0.0          0.0          0.0          0.0
 33)  0.0          0.0          0.0          0.0          0.0
 34)  0.0          0.0          0.0          0.0          0.0
 35)  0.0          0.0          0.0          0.0          0.0
 36)  0.0          0.0          0.0          0.0          0.0
 37)  0.0          0.0          0.0          0.0          0.0
 38)  0.0          0.0          0.0          0.0          0.0
 39)  0.0          0.0          0.0          0.0          0.0
 40)  0.0          0.0          0.0          0.0          0.0
 41)  0.0          0.0          0.0          0.0          0.0
 42)  0.0          0.0          0.0          0.0          0.0
 43)  0.0          0.0          0.0          0.0          0.0
 44)  0.0          0.0          0.0          0.0          0.0
 45)  0.0          0.0          0.0          0.0          0.0
 46)  0.0          0.0          0.0          0.0          0.0
 47)  0.0          0.0          0.0          0.0          2.99495E-03
 48)  0.0          0.0          0.0          0.0          6.95346E-03
 49)  0.0          0.0          0.0          0.0          7.96266E-03
 50)  0.0          0.0          0.0          0.0          1.02636E-02
                                                       1.10333E-02

      Ionic.Streng Eh (=) Volts  Titr Vol, ml       pH
  1)  7.56866       0.0           0.0           7.6626
  2)  7.56794       0.0           5.00000E-02   7.7047
  3)  7.56722       0.0           0.10000       7.7470
  4)  7.56578       0.0           0.20000       7.8327
  5)  7.56432       0.0           0.30000       7.9207
  6)  7.56286       0.0           0.40000       8.0124
  7)  7.56138       0.0           0.50000       8.1090
  8)  7.55985       0.0           0.60247       8.2140
  9)  7.54690       0.0            1.5302       8.2934
 10)  7.54271       0.0            1.8000       8.5057
 11)  7.51802       0.0            3.2257       8.5094
 12)  6.83835       0.0            43.867       8.6179
 13)  6.32590       0.0            76.741       8.7135
 14)  5.66578       0.0            161.77       8.8169
 15)  5.34622       0.0            194.92       8.8921
 16)  4.98713       0.0            234.87       9.0300
 17)  4.88441       0.0            246.90       9.0948
 18)  4.78417       0.0            258.94       9.1859
 19)  4.71895       0.0            266.96       9.2769
 20)  4.68686       0.0            270.97       9.3417
 21)  4.65520       0.0            274.98       9.4335
 22)  4.62405       0.0            278.99       9.5462
 23)  4.61630       0.0            280.00       9.5997
 24)  4.61249       0.0            280.50       9.6320
 25)  4.60870       0.0            281.00       9.6697
 26)  4.60494       0.0            281.50       9.7151
 27)  4.60120       0.0            282.00       9.7722
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E22

765	28)	4.59751	0.0	282.50	9.8492
766	29)	4.59391	0.0	283.00	9.9671
767	30)	4.59217	0.0	283.25	10.061
768	31)	4.59025	0.0	283.55	10.271
769	32)	4.58988	0.0	283.62	10.361
770	33)	4.58949	0.0	283.75	10.644
771	34)	4.58953	0.0	283.80	10.791
772	35)	4.58967	0.0	283.85	10.928
773	36)	4.58994	0.0	283.92	11.083
774	37)	4.59028	0.0	284.00	11.218
775	38)	4.59081	0.0	284.12	11.367
776	39)	4.59140	0.0	284.25	11.487
777	40)	4.59208	0.0	284.40	11.594
778	41)	4.59277	0.0	284.55	11.680
779	42)	4.59482	0.0	285.00	11.868
780	43)	4.59937	0.0	286.00	12.118
781	44)	4.60187	0.0	286.55	12.212
782	45)	4.61095	0.0	288.55	12.445
783	46)	4.62941	0.0	294.55	12.767
784	47)	4.66285	0.0	304.55	13.048
785	48)	4.70438	0.0	314.55	13.229
786	49)	4.89476	0.0	359.90	13.643
787	50)	5.16374	0.0	429.90	13.944

E23