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subject: Estimate WIPP Waste Particle Sizes Based on Expert Elicitation Results: Revision 1

Introduction

An expert elicitation has been conducted on the particle size distribution(s) of transuranic waste in the Waste Isolation Pilot Plant (WIPP) over the 10,000-year regulatory post-closure period [1]. This memorandum presents calculations of waste particle size distributions, based on the expert elicitation. The range of mean particle sizes developed based on expert elicitation are 1 mm (minimum) to 10 cm (maximum). The use of this distribution in the Environmental Protection Agency (EAP)-mandated performance assessment verification test (PAVT) to develop the TAUFAIL parameter is discussed in Wang and Larson (1997) [2]. Because spall volumes are sampled directly in the EPA PAVT, the particle size distributions are not used to determine spallings volume (although it is noted that the distribution derived from the expert elicitation falls in the range of values (40 μm - 20 cm) used in the WIPP Compliance Certification Application (CCA) (see CCA, PAR-115), indicating that the CCA range encompassed more uncertainty, especially for small particles and higher releases, than is presently indicated). This memorandum conprises two parts: a summary of expert elicitation results, and a description of the use of those results to determine a range of mean particle diameters appropriate for use in CCA/EPA PAVT performance assessment models. Calculations in this memorandum have been performed using Microsoft EXCEL 7.0 spreadsheet on IBM ThinkPad under MS-DOS 6.22 and Windows 95.

This memorandum is a revision of a previous memorandum dated June 27, 1997. The revision is made based on the discussion with EPA representatives and an EEG observer in July, 1997. The revision only adds more clarifications, and all the calculations and conclusions documented in the previous memorandum have not been changed.

Summary of Expert Elicitation Results

According to chemical characteristics, the expert panel categorized WIPP transuranic waste into four groups [1]: (1) iron- and aluminum-based metal/alloys, (2) cellulosics/solidified organics, (3) rubber/plastics, and (4) other metals/inorganic/vitrified/soils/cements/solidified inorganics. The initial distributions of particle size and particle number in each group have been documented in the expert elicitation report [1] (p. 54-57) and are repeated here in the first two columns in Table 1. The expert panel included MgO backfill and salt as two additional waste groups, although those materials are not real wastes. Since the MgO pellets used for recent experimental studies have a size range of 0.5 to 4 mm [3], the panel estimated that MgO backfill has a constant particle size of $\sim 10^{-9}$ m^3. The panel also estimated that the particle size of salt ranges from dust (1 μm in diameter) to half room size slabs (2 m thick), with an average value of 10^{-5} m^3. Salt will be introduced into the repository by mechanic separation from the roof and walls during room closure. The panel used the following complementary cumulative distribution function (CCDF) to describe the salt particle size distribution [1] (p. 55):

$$1 - F(v) = \frac{10^{-18}}{v} \tag{1}$$

where $F(v)$ is the cumulative distribution function (CDF) and v is the particle size (m^3). Note that the average salt particle size estimated by the panel is not consistent with the one that can be calculated from Equation (1) and the latter is much smaller. To be conservative, only Equation (1) will be used in this memo to estimate salt particle size distribution. The relative amount of each waste group is listed in Table 2. Among the waste groups, group 4 and salt are considered to be chemically inert and their particle sizes will remain unchanged over the regulatory period.

The expert panel also identified the chemical and physical processes that can modify the particle sizes of the waste over the 10,000 year regulatory post-closure period [1] (p. 10-11). Those processes include crushing, anoxic metal corrosion, organic material biodegradation, precipitation/cementation, encapsulation, and fragmentation. Room creep closure will tend to crush the waste containers, exposing their contents to the surrounding environment to a variable extent. Crushing could result in mechanical bonding of waste as some components deform into interlocking shapes. Metal in the waste, predominantly steel, will anoxically react with water, generating H$_2$ gas and iron hydroxides. Steel corrosion strongly depends on the availability of brine in the repository [4]. It is unlikely that anoxic corrosion will consume all the steel initially present in the repository. Biodegradation consumes organic materials and generates CO$_2$ and CH$_4$ gases as its final reaction products. In the WIPP performance assessment (PA) calculations,

it is assumed that biodegradation occurs in half of the realizations, out of which there is a 50% chance for rubbers and plastics to be biodegraded [4]. Biomass could form fine particles, but the effects are expected to be negligible under WIPP conditions [1] (p. 39). During room closure, salt may creep locally into the voids in the waste, encapsulating waste particles. Such encapsulation occurs mainly near the salt-waste boundary. Fragmentation caused by the mechanical action of drill bit is mentioned, but not further discussed in the expert elicitation report.

Secondary mineral precipitation and cementation are the two most important processes controlling the evolution of waste particle size in the repository. The chemical reactions involved in the two processes include:

$$Fe + 2H_2O = Fe(OH)_2 + H_2 \tag{2}$$
$$MgO + H_2O = Mg(OH)_2 \tag{3}$$
$$Mg(OH)_2 + CO_2 = MgCO_3. \tag{4}$$

Water consumption in the above reactions may also cause salt precipitation out of brine. The expert panel estimated that most corrosion and MgO hydration products will precipitate out as cementation agents, with the remainder precipitating as free particles. The panel has assessed that a maximum of 25% of the reaction products will precipitate out as free particles, and that this percentage will decrease to zero as available porosity approaches zero. The free particles will range in size from 0.1 to 10 μm, with an average value of 2 μm. The expert panel suggested that smaller particles are likely to be aggregated by cement, with the likelihood approximately inversely proportional to particle volume. The panel estimated that the particle size will approach to room size (i.e. a cemented mass) as the cement volume approaches about 40% of pore space (assuming about a 25% porosity prior to cementation) and that the range of particle size will also decrease [1] (p. 58). However, the panel did not provided any guidance for other initial waste porosities, nor has it provided a quantitative measure for the degree of cementation as a function of the filled void volume.

The expert panel developed a decomposition model to predict waste particle size as a function of the extent of individual physical and chemical processes. Although theoretically sound, the model is difficult to be fully implemented in PA calculations for the following reasons: First, the current BRAGFLO model does not explicitly calculate the extent of all relevant chemical processes, e.g. MgO hydration reaction. Second, the particle size distribution used in this model is based on particle numbers, which was chosen as convenient for the panel to use (i.e., all data was transformed to a common particle number basis) but was not intended to be taken as the physically meaningful measure of particle size for input into PA codes (Mike Gross, Expert Panel Member, personal communication July 1997). As will be shown below, this type of particle size distribution is not appropriate for deriving physically meaningful parameters for caving and spalling models. Third, no quantitative approach is provided in the expert elicitation report to treat the cementation effect for cases in which the waste porosity is larger than

25% or the cement volume is less than 40% of the pore space. The EXCEL spreadsheet provided in Appendix B of the expert elicitation report does provide a treatment for the cementation effect, but it is inconsistent with that proposed in the report. Fourth, the expert panel has discussed at length how to use statistical methods to simulate waste spatial heterogeneity. However, most input parameters required to simulate heterogeneity are not currently available and were not provided by the panel. These points were discussed in July 1997 with Mike Gross, Expert Panel Member, who agrees with them (personal communication). For these reasons, the EXCEL spreadsheet provided by the panel will not be used in the particle size calculations performed below.

Volume-Fraction Based Particle Size Distribution

To be physically meaningful, calculations of particle size distributions must be based on volume fractions instead of particle numbers. Reasons for this are straightforward: First of all, our primary interest is the waste volume, not the number of particles, released by spalling and caving processes. The probability of a given collection of particles being accessed by a borehole is proportional to their relative volume in the repository. Second, the mechanical properties of a mixture of variably-sized particles tend to be determined by the size of the particles that account for the majority of total solid volume. A particle-number-based particle size distribution, however, does not reflect this relationship. For example, suppose that we have a mixture of 10 particles with a diameter of 1 cm and 1000 particles with a diameter of 0.1 cm. According to particle-number-based size distribution, the mixture should behave like smaller particles, although these particles only account for about 10% of total solid volume. These points were also discussed in July 1997 with Mike Gross, Expert Panel Member, who agrees with them (personal communication).

An alternative to use of volume fraction is a mass-fraction-based size distribution. However, a mass-fraction-based size distribution is also not appropriate for our purposes, because particle density and size are taken into account separately in the caving and spalling models and use of the mass-fraction-based size distribution would thus doubly account for the effect of particle density.

The calculation of volume-fraction-based particle size distributions is straightforward for waste groups 1 - 4. The CDF for each waste group is calculated in Table 1 and displayed in Figure 1. The volume-fraction-based particle size distribution for salt is calculated from Equation (1) (from the panel), using the following relationships:

$$f(v) = \frac{dF(v)}{dv} = \frac{10^{-18}}{v^2} \tag{5}$$

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$$G(v) = \frac{\int_{10^{-18}}^{v} vf(v)dv}{\int_{10^{-18}}^{10^{3}} vf(v)dv} = \frac{\log(v) + 18}{21} \qquad (6)$$

where $F(v)$ and $G(v)$ are respectively the particle-number-based and volume-fraction-based CDFs for salt. $G(v)$ is tabulated in Table 3 and shown in Figure 1E. Particle diameters are computed from particle sizes by assuming that all particles are spherical.

Particle Size Distribution of Initial Waste

The particle size distribution in each waste group is shown in Figure 1. This figure shows that particles smaller than 100 μm are present only in waste group 4 and salt. The volume fraction of these particles is small, especially in waste group 4.

The composite particle size distribution of initial waste is summarized in Table 4 and shown in Figure 2. The composite distribution is calculated by combining the particle size distributions in individual waste groups weighted by the volume fraction of each group listed in Table 2. To be consistent with the CCA, spatial heterogeneity of waste properties is ignored here. In the calculation, the particle size of MgO pellets is assumed to be 2 mm in diameter. Figure 2 shows that the particle size of the initial waste ranges from a few μm to 1 m and that particles smaller than 124 μm account for about 4% of total waste volume. Furthermore, most of the fine particles are salt (Table 4), which is not an actual waste component.

Particle Size Reduction due to Dissolution

Waste particles will be reduced in size by various dissolution processes, such as steel corrosion and organic material biodegradation. However, these dissolution processes will not produce any significant volume fraction of fine particles in the repository. This occurs because particle volume ∝ (particle diameter)³ and thus the particles subjected to dissolution will become negligible in volume relative to non-degradable materials (such as waste group 4 and salt), before these particles become very small. For example, suppose that we initially have a certain number of particles with a diameter of 1 cm, which are mixed with other inert particles and account for 50% of total volume. Assume that, due to dissolution, the particle diameter is reduced to 0.1 cm. Accordingly, the volume of those particles is reduced by a factor of 1000. As a result, the volume of those particles after dissolution becomes negligible with respect to total solid volume.

To illustrate the effect of dissolution processes on particle size, the CDF for 96% degraded cellulosics is calculated in Table 5 and presented in Figure 3. It can be seen in Figure 3 that no small particles are produced by cellulosics biodegradation and, as a matter of fact, the biodegradation even eliminates small particles (< 0.01 m) initially present in this waste group.

Cementation Effect

Cementation was identified by the expert panel as an important process leading to the aggregation of waste particles in the repository. The panel estimated that the particle size will approach room size (i.e. a cemented mass) as the cement volume approaches about 40% of the available pore space (assuming about a 25% porosity prior to cementation) [1] (p. 58). In order to quantify the cementation effect, assume that the 40% of cement in pore volume for total cementation can be extended to the whole range of waste porosity. The average porosity of the waste in the repository after room closure is estimated to be between 45 - 50%[1]. This assumption is reasonable. Take sandstone as a natural analog. Following mechanical compaction, unconsolidated sand sediments usually attain a porosity of 40 - 50%, while consolidated sand sediments (sandstone) can have a porosity up to 30% [7]. If we assume that the porosity decrease from mechanically-compacted, unconsolidated sand sediments to sandstone is caused by cementation, then the 40% cement in pore space is sufficient to consolidate sediments which have an initial porosity up to 50%. Note that the minimum amount of cement required for sediment consolidation may be even less.

For simplification, in the calculation performed below only cementation induced by MgO hydration and steel corrosion are considered, and the other processes such as salt precipitation are ignored. To be conservative, it is assumed that 25% of reaction products will precipitate out as free particles with a diameter of 2 μm and the rest will precipitate as cement. Note that those free particles are the smallest particles that can be created in the repository.

The changes of particle size as a function of the extents of MgO hydration and steel corrosion can be calculated with a linear interpolation method:

$$\log d = \log 2 - \frac{40\% - f_c}{40\%}(\log 2 - \log d_0) \qquad \text{for } f_c \leq 40\%$$

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[1] The average porosity of waste after room closure is estimated from BRAGFLO calculations. In the BRAGFLO, the total room volume is fixed, and the actual porosity is (ϕ) related to the BRAGFLO porosity (ϕ_b) by

$$\phi_b = \phi\left(\frac{1-\phi_0}{1-\phi}\right)$$

where ϕ_0 is the initial waste porosity, which is 0.848 [5] (p. 6-100). From the BRAGFLO calculations, the average value of ϕ_b is 0.12 for disturbed cases and 0.15 for undisturbed cases [6] (fig. 7.1-18).

$$\log d = \log 2 \qquad \text{for } f_c > 40\% \qquad (7)$$

where d_0 is the particle diameter before cementation and d is the particle diameter when a fraction, f_c, of pore space is occupied by cements. The totally cemented waste is assumed to have a particle diameter of 2 m. Notice that the curve described by Equation (7) is always below the interpolation curve qualitatively suggested by the expert panel [1] (fig. 5-7). Therefore, the above interpolation method tends to underestimate the cementation effect and is thus conservative.

The calculation results using Equation (7) are shown in Tables 6 and 7. Table 6 shows the fraction of filled pore space as a function of the extent of the chemical reactions. Table 7 shows the evolution of the smallest particle size as a function of the extent of chemical reactions. After 30% degradation of either MgO or steel, the smallest particle size will exceed 120 μm. In other words, fine particles can be present only when a small fraction of MgO and steel are reacted. The calculation also shows that cementation can effectively aggregate waste particles to a room size, as either MgO hydration or steel corrosion proceeds.

If substantial brine inflow occurs in the repository, MgO hydration will be completed very early in the 10,000 year regulatory time period, because the hydration reaction is usually much faster than any other reactions such as organic material biodegradation and steel corrosion. In this case, fine particles will be cemented into massive units before the repository reaches the conditions (e.g., gas pressure) necessary to result in a significant spalling and caving release. The repository performance then becomes insensitive to the particle size distribution in the initial waste. If brine inflow is minimal, MgO hydration and steel corrosion will be limited and only organic material biodegradation will possibly proceed. In this case, the particle size distribution in the waste will be similar to that in the initial waste.

Particle Size Distribution for Bounding Case

In principle, using Equation (7), we can calculate particle size distributions in degraded waste as a function of the extent of steel corrosion and MgO hydration. This would yield time-dependent estimates of particle sizes as steel corrodes and MgO hydrates. However, constant values for mean particle size were used in the CCA rather than time-dependent particle sizes. As an alternative, in this section, we will construct a bounding case that will provide a lower limit for actual cases in terms of particle size distribution. The bounding case assumes partial steel corrosion, partial MgO hydration, complete degradation of cellulosics, plastics, and rubbers, and discounts some effects of cementation. This bounding case will be used in next section to estimate the lower limit of mean particle size for degraded waste.

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Steel corrosion and MgO hydration will not only generate fine particles but will also produce cements to aggregate those particles (Table 7). Table 7 can be divided into two regions: region I with $f_{MgO} + f_{Fe} \geq 0.8$ and region II with $f_{MgO} + f_{Fe} < 0.8$, where f_{MgO} and f_{Fe} are the fractions of MgO and steel reacted respectively. In region I, particle sizes are close to or larger than drill bit diameter, and thus spalling and caving releases seem unlikely. Thus, little needs to be concerned with region I. Spalling and caving releases will be possible only when $f_{MgO} + f_{Fe} < 0.8$ (region II). The lower limit of particle size distributions in region II can be bounded by a bounding case which is constructed so that the sizes of waste particles are minimized while the relative volume of fine particles in total waste is maximized:

- It is assumed in the bounding case that fine particles precipitated during steel corrosion and MgO hydration remain un-cemented and have a diameter of 2 μm (i.e. the smallest size in region II). Similarly, no particle aggregation is assumed for partially-reacted and non-reactive waste components. The particle sizes of remaining MgO and steels are thus controlled solely by dissolution processes.

- The maximum volume of fine particles that can be produced in region II is calculated by assuming that $f_{MgO} + f_{Fe} = 0.8$, with $f_{MgO} = 0.4$ and $f_{Fe} = 0.4$, and that 25% of reaction products precipitate as free particles, consistent with the expert elicitation. Note that the limit of $f_{MgO} + f_{Fe} = 0.8$ is directly related to cementation effect.

- To further maximize the volume fraction of fine particles, complete degradation of cellulosics, plastics, and rubbers is also assumed in the bounding case. It can be shown that the mean particle sizes of initial organic materials are larger than the one to be calculated in the next section for the overall waste in the bounding case (Table 1). It can also be shown that this size will not be changed significantly during biodegradation (Figure 3). Thus, the assumption of complete biodegradation of organic materials will result in a smaller mean particle size for overall waste.

Apparently, the above bounding conditions, which may not necessarily be encountered in the repository, will result in a very conservative estimate for the lower bound of particle size distribution.

The particle size distribution for the bounding case is shown in Figure 4. It is calculated as follows:

- Calculate the particle diameters and particle size distributions for remaining steel (Table 8) and MgO. For MgO pellets with an initial diameter of 2 mm, after 40% hydration, the remaining pellets have a diameter of 1.69 mm.

- Calculate the volume fractions of waste groups including fine free particles (Table 9).

- Combine particle size distributions of individual waste groups based on their volume fractions (Table 10). For example, the remaining MgO pellets account for 100% of the totoal volume in waste group 6, which in turn

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accounts for 21.8% of the total volume in the whole waste (Table 8, column 6). Therefore, the volume fraction of the remaining MgO pellets in the whole waste is equal to 100% x 21.8% = 21.8% (Table 10, column 3).

Figure 4 shows that, even in this bounding case, particles smaller than 120 μm account for only 16% of total volume. In the bounding calculation, it is assumed that $f_{MgO} + f_{Fe} = 0.8$. However, Table 7 shows that 30% degradation of either steel or MgO will produce enough cements to raise the smallest particle size from 2 μm to > 120 μm. Therefore, in actual worst cases, particles smaller than 120 μm will account for less than 10% of total volume.

Range of Mean Particle Size

As discussed above, a small fraction of <u>individual</u> particles could have a diameter as small as a few microns. This number can not be used as a lower limit for particle size sampling in the spalling and caving models, because it is not physically meaningful. The appropriate particle size range must be estimated based on mean particle size, for the following reasons:

- A small fraction of fine particles can be present in the initial waste. However, there is no conceivable mechanism by which the fine particles will be segregated from coarse particles in space on a multiple-drum scale. Note that waste containers themselves are considered as large particles, that MgO pellets will be emplaced between the drums and on the top of the drums, and that mechanical crushing of the waste after room closure will further mix the waste. Also note that the fine particles (< 120 μm) largely consist of salt fragments falling from the roof and walls of the repository (Table 4) and there is no way for those fine particles to be segregated from large fragments.

- The fine particles produced by MgO hydration and steel corrosion can be present only when MgO and steel are partially reacted, and thus those particles will be always mixed with remaining MgO and steel particles.

- Therefore, the fine particles not only account for a small fraction of total solid volume but will also remain mixed with large particles. From a mechanistic point of view, small particles can not be eroded unless large particles become mobile [8].

The conventional characterization of a mixture of different size granular materials is carried out in terms of an appropriately defined mean size [9, 10, 11, 12]. There are various ways to define mean particle sizes. The most commonly used definitions are the arithmetic mean, the geometric mean, and the median. The three means have been calculated for the lower bounding case (Table 10). Because the range of particle diameters extends over about 6 orders of magnitude, the arithmetic mean is significantly skewed towards to large particle sizes. The geometric mean and median give reasonable descriptions of average particle sizes. Since all three means calculated for the lower bounding case are larger than 1 mm, the choice of 1 mm as a lower limit of mean particle diameter for degraded waste is

reasonable and conservative. Since cementation can effectively aggregate waste particles to a room size, and also because particles larger than the drill bit diameter can not be released through a borehole, 0.1 m is recommended as the upper limit of mean particle size for the spalling and caving models.

Conclusions

1. The results of the expert panel elicitation on WIPP particle size distribution have been summarized. Because of the difficulty with implementation, the EXCEL spreadsheet provided by the expert panel is not used in the calculations performed in this memorandum.
2. To be physically meaningful, particle size distributions must be calculated based on volume fractions instead of particle numbers.
3. The particle size in the initial waste ranges from a few μm to 1 m and particles smaller than 120 μm account for about 4% of total waste volume.
4. Dissolution processes cannot produce a significant amount of fine particles in the repository.
5. Cementation induced by MgO hydration and steel corrosion can effectively aggregate waste particles to a room size.
6. In actual worst cases, particles smaller than 120 μm account for less than 10% of total volume in degraded waste.
7. Mean particle size is appropriate for use in CCA and PAVT models. The range of mean particle size is recommended to be 1 mm to 10 cm.

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Table 1. Initial Waste Characteristics [1]. Column 3 = column 1 x column 2. Column 5 = column 3/total volume. The expert elicitation report does not provide density data for waste group 1. Since this group mainly includes iron metals/alloys, the (arithmetic) mean density of this waste group is chosen to be that of steel [13]. Particle diameters are calculated from particle sizes by assuming that all particles are spherical.

Group 1: Iron- and aluminum-Base Metal/Alloys

Size (m^3)	Particle number	Volume (m^3)	Diameter (m)	Volume fraction	CDF	Density
2.50E-07	4.00E+06	1	0.00782	8.06E-05	8.06E-05	
1.00E-06	1.50E+07	15	0.012412	0.001209	0.00129	
1.00E-05	8.50E+07	850	0.02674	0.068527	0.069817	
1.00E-04	3.50E+07	3500	0.057605	0.282169	0.351986	
1.00E-03	1.10E+06	1100	0.124096	0.088682	0.440667	
4.50E-03	7.30E+05	3285	0.204868	0.264836	0.705503	
5.00E-02	4.46E+03	223	0.457114	0.017978	0.723481	
6.70E-02	4.46E+03	298.82	0.503951	0.024091	0.747572	
8.30E-02	7.10E+03	589.3	0.541236	0.047509	0.795081	
3.58E-01	7.10E+03	2541.8	0.880982	0.204919	1	
		Total volume 12403.92				Mean 7.9

Group 2: Cellulosics/solidified organics

Size (m^3)	Particle number	Volume (m^3)	Diameter (m)	Volume fraction	CDF	Density
1.00E-11	9.80E+13	980	0.000268	0.085954	0.085954	2
1.00E-06	1.72E+08	172	0.012412	0.015086	0.10104	2
2.00E-06	2.56E+08	512	0.015638	0.044907	0.145947	0.9
2.70E-05	3.79E+07	1023.3	0.037233	0.089752	0.235699	0.9
4.70E-05	3.27E+07	1536.9	0.044789	0.134799	0.370499	0.9
9.10E-05	1.13E+07	1028.3	0.055822	0.090191	0.460689	0.9
2.90E-04	5.30E+06	1537	0.082144	0.134808	0.595498	0.9
3.90E-04	2.63E+06	1025.7	0.090669	0.089963	0.68546	0.9
7.50E-04	1.37E+06	1027.5	0.11275	0.090121	0.775581	0.9
4.50E-03	3.41E+05	1534.5	0.204868	0.134589	0.91017	0.9
6.30E-03	8.13E+04	512.19	0.229181	0.044923	0.955093	0.9
2.00E-02	2.56E+04	512	0.336815	0.044907	1	0.9
		Total volume 11401.39	Arithmetic mean: 0.1 Geometric mean: 0.05 Median: 0.06			Mean 1.01

Table 1. (cont.)

Group 3: Rubber and plastics

Size (m^3)	Particle number	Volume (m^3)	Diameter (m)	Volume fraction	CDF	Density
1.00E-06	1.00E+06	1	0.012412	0.0001	0.0001	1.14
1.00E-05	1.46E+07	146	0.02674	0.014643	0.014743	1
1.00E-04	4.00E+06	400	0.057605	0.040118	0.054862	1.14
1.00E-04	2.19E+07	2190	0.057605	0.219648	0.27451	1
1.50E-04	7.30E+05	109.5	0.06594	0.010982	0.285492	0.5
4.50E-04	8.90E+03	4.005	0.095098	0.000402	0.285894	0.5
1.00E-03	1.00E+06	1000	0.124096	0.100296	0.38619	1.14
1.00E-03	1.46E+06	1460	0.124096	0.146432	0.532621	1
4.00E-03	1.00E+04	40	0.196981	0.004012	0.536633	1.14
7.70E-03	6.00E+05	4620	0.245033	0.463367	1	1
		Total volume 9970.505 9970.505	Arithmetic mean: 0.16 Geometric mean: 0.14 Median: 0.12			Mean 1.01

Group 4: Other metals/inorganic/vitrified/soils/cements/solidified inorganics

Size (m^3)	Particle number	Volume (m^3)	Diameter (m)	Volume fraction	CDF	Density
1.25E-16	3.20E+16	4	6.21E-06	0.000196	0.000196	2.5
1.00E-12	1.38E+14	138	0.000124	0.006751	0.006946	11.3
1.00E-12	6.00E+13	60	0.000124	0.002935	0.009881	2.5
1.00E-12	3.37E+15	3370	0.000124	0.164854	0.174736	2.2
8.00E-12	8.89E+14	7112	0.000248	0.347906	0.522642	1.3
8.00E-12	3.82E+14	3056	0.000248	0.149494	0.672136	2.8
8.00E-06	4.90E+07	392	0.024823	0.019176	0.691312	2.5
2.13E-04	8.53E+06	1816.89	0.074115	0.088879	0.780191	2.5
2.88E-04	1.13E+06	325.44	0.081955	0.01592	0.796111	8
1.00E-03	3.22E+05	322	0.124096	0.015752	0.811863	11.3
5.00E-03	9.20E+04	460	0.21219	0.022502	0.834365	11.3
3.70E-02	7.75E+03	286.75	0.413466	0.014027	0.848393	11.3
1.49E-01	2.08E+04	3099.2	0.65778	0.151607	1	3
		Total volume 20442.28				Mean 2.76

Table 2. Relative amount of each waste group in initial waste. Data are taken from [1] p. 58

Waste group	Material	Wt%	Volume (%)
1	Fe- and Al-base metals	28.9	14.3
2	Cellulosics and solidified organics	4.0	13.1
3	Rubbers and plastics	4.6	11.5
4	Other metals, inorganic, vitrified, soils, cements, and solidified inorganics	22.1	23.5
5	Salt	9.1	12.0
6	MgO backfill	31.3	25.5

Table 3. Particle size distribution of salt calculated from Equation (6). Volume fraction = G(right end of discretization interval) - G(left end of discretization interval).

Size (m^3)	Diameter (m)	Volume fraction	CDF	Discretization interval (m^3)		Density
1.00E-17	2.68E-06	0.095238	0.095238	1.00E-18	1.00E-16	
1.00E-15	1.24E-05	0.095238	0.190476	1.00E-16	1.00E-14	
1.00E-13	5.76E-05	0.095238	0.285714	1.00E-14	1.00E-12	
1.00E-11	0.000268	0.095238	0.380952	1.00E-12	1.00E-10	
1.00E-09	0.001242	0.095238	0.47619	1.00E-10	1.00E-08	
1.00E-07	0.005762	0.095238	0.571429	1.00E-08	1.00E-06	
1.00E-05	0.02674	0.095238	0.666667	1.00E-06	1.00E-04	
1.00E-03	0.124096	0.095238	0.761905	1.00E-04	1.00E-02	
1.00E-01	0.575914	0.095238	0.857143	1.00E-02	1.00E+00	
1.00E+00	1.240674	0.142857	1	1.00E+00	1.00E+03	
						Mean 2.16

Table 4. The composite particle size distribution of initial waste. Volume fractions are calculated from the volume fractions in Table 1 and Table 3 for individual waste groups weighted on the relative amount of each waste group in Table 2.

Diameter (m)	Volume fraction	CDF	Waste group	Diameter (m)	Volume fraction	CDF	Waste group
2.68E-06	0.011429	0.011429	5	0.124096	0.003702	7.52E-01	4
6.21E-06	4.60E-05	1.15E-02	4	0.124096	0.011429	7.63E-01	5
1.24E-05	0.011429	2.29E-02	5	0.196981	0.000461	7.64E-01	3
5.76E-05	0.011429	3.43E-02	5	2.05E-01	0.037871	8.01E-01	1
0.000124	0.001586	3.59E-02	4	0.204868	0.017631	8.19E-01	2
0.000124	0.00069	3.66E-02	4	0.21219	0.005288	8.24E-01	4
0.000124	0.038741	7.54E-02	4	0.229181	0.005885	8.30E-01	2
0.000248	0.081758	1.57E-01	4	0.245033	0.053287	8.84E-01	3
0.000248	0.035131	1.92E-01	4	0.336815	0.005883	8.89E-01	2
0.000268	0.01126	2.03E-01	2	0.413466	0.003296	8.93E-01	4
0.000268	0.011429	2.15E-01	5	0.457114	0.002571	8.95E-01	1
0.001242	0.011429	2.26E-01	5	0.503951	0.003445	8.99E-01	1
2.00E-03	0.255	4.81E-01	6	0.541236	0.006794	9.06E-01	1
0.005762	0.011429	4.93E-01	5	0.575914	0.011429	9.17E-01	5
0.00782	1.15E-05	4.93E-01	1	0.65778	0.035628	9.53E-01	4
0.012412	0.000173	4.93E-01	1	0.880982	0.029303	9.82E-01	1
0.012412	0.001976	4.95E-01	2	1.240674	0.017143	9.99E-01	5
0.012412	1.15E-05	4.95E-01	3				
0.015638	0.005883	5.01E-01	2				
0.024823	0.004506	5.05E-01	4				
0.02674	0.009799	5.15E-01	1				
0.02674	0.001684	5.17E-01	3				
0.02674	0.011429	5.28E-01	5				
0.037233	0.011758	5.40E-01	2				
0.044789	0.017659	5.58E-01	2				
0.055822	0.011815	5.69E-01	2				
0.057605	0.04035	6.10E-01	1				
0.057605	0.004614	6.14E-01	3				
0.057605	0.02526	6.40E-01	3				
0.06594	0.001263	6.41E-01	3				
0.074115	0.020887	6.62E-01	4				
0.081955	0.003741	6.66E-01	4				
0.082144	0.01766	6.83E-01	2				
0.090669	0.011785	6.95E-01	2				
0.095098	4.62E-05	6.95E-01	3				
0.11275	0.011806	7.07E-01	2				
0.124096	0.012681	7.20E-01	1				
0.124096	0.011534	7.31E-01	3				
0.124096	0.01684	7.48E-01	3				

Table 5. Calculation of particle size distribution for remaining cellulosics. Assume that particle diameters are reduced by 0.01 m (corresponding to about 96% biodegradation). Column 5 = (column 4/column 1)^3 x column 2. Column 6 = column 5/total volume.

Initial cellulosics			Remaining cellulosics			
Diameter (m)	Volume fraction	CDF	Diameter (m)	Remaining volume (m^3)	Volume fraction	CDF
0.000268	0.085954	0.085954	0	0	0	0
0.012412	0.015086	0.10104	0	0	0	0
0.015638	0.044907	0.145947	0	0	0	0
0.037233	0.089752	0.235699	0	0	0	0
0.044789	0.134799	0.370498	0	0	0	0
0.055822	0.090191	0.460689	0	0	0	0
0.082144	0.134808	0.595497	0	0	0	0
0.090669	0.089963	0.68546	0	0	0	0
0.11275	0.090121	0.775581	0.01275	0.00013	0.003115	0.003115
0.204868	0.134589	0.91017	0.104868	0.018052	0.431488	0.434603
0.229181	0.044923	0.955093	0.129181	0.008045	0.192301	0.626904
0.336815	0.044907	1	0.236815	0.015609	0.373096	1
				Total remaining volume 0.041836		

* Given 1 m^3 of cellulosics, the total remaining volume after the particle diameters reduced by Δd due to

$$\text{dissolution} = \sum_{d_i > \Delta d} f(d_i) \left(\frac{d_i - \Delta d}{d_i} \right)^3$$, where d_i is the diameter of initial particles (column 1) and $f(d_i)$ is the volume fraction of the initial particles with a diameter of d_i (column 2).

Table 6. Fractions of pore space occupied by Mg(OH)$_2$ and Fe(OH)$_2$ cements (f_c) as a function of the extents of metal corrosion (rows, f_{Fe}) and MgO hydration (columns, f_{MgO})*. A 50% porosity prior to cementation is assumed. Note that the solid volume in reactions (2) and (3) is expanded by 373.4 % [13] and 219.0% [14].

	0	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	1
0	0	0.041878	0.083757	0.125635	0.167514	0.209392	0.251271	0.293149	0.335028	0.376906	0.418784
0.1	0.04005	0.081928	0.123807	0.165685	0.207564	0.249442	0.29132	0.333199	0.375077	0.416956	0.458834
0.2	0.0801	0.121978	0.163856	0.205735	0.247613	0.289492	0.33137	0.373249	0.415127	0.457006	0.498884
0.3	0.120149	0.162028	0.203906	0.245785	0.287663	0.329542	0.37142	0.413298	0.455177	0.497055	0.538934
0.4	0.160199	0.202077	0.243956	0.285834	0.327713	0.369591	0.41147	0.453348	0.495227	0.537105	0.578984
0.5	0.200249	0.242127	0.284006	0.325884	0.367763	0.409641	0.451519	0.493398	0.535276	0.577155	0.619033
0.6	0.240299	0.282177	0.324055	0.365934	0.407812	0.449691	0.491569	0.533448	0.575326	0.617205	0.659083
0.7	0.280348	0.322227	0.364105	0.405984	0.447862	0.489741	0.531619	0.573497	0.615376	0.657254	0.699133
0.8	0.320398	0.362277	0.404155	0.446033	0.487912	0.52979	0.571669	0.613547	0.655426	0.697304	0.739183
0.9	0.360448	0.402326	0.444205	0.486083	0.527962	0.56984	0.611719	0.653597	0.695475	0.737354	0.779232
1	0.400498	0.442376	0.484255	0.526133	0.568011	0.60989	0.651768	0.693647	0.735525	0.777404	0.819282

* The fractions of pore space filled with cements are calculated as follows:

1. Given a porosity of 50%, the pore space (V_p) associated with 1 m^3 of solids or 2 m^3 of bulk waste = 1 m^3.
2. 1 m^3 of solids initially contains 0.143 m^3 of waste group 1 and 0.255 m^3 of MgO (Table 2). The cement volume (V_c) = (0.143 x 373.4% x f_{Fe} + 0.255 x 219.0% x f_{MgO}) x 75%, where f_{Fe} and f_{MgO} are the fractions of waste group 1 and MgO reacted, respectively. It is assumed here that the solid volumes in Reactions (2) and (3) are respectively expanded by 373.4 % and 219.0 % and that 75% of the reaction products will precipitate as cements.
3. The fraction of pore space filled with cements = V_c/V_p.

Table 7. The smallest particle size (m) in the waste calculated with Equation (7) as a function of the extents of metal corrosion (rows) and MgO hydration (columns). The initial smallest particle size is assumed to be 2 μm in diameter. The fractions of cements in pore space used in the calculation are from Table 6. Table 7 can be divided into two regions: region I with $f_{MgO} + f_{Fe} \geq 0.8$ and region II with $f_{MgO} + f_{Fe} < 0.8$, where f_{MgO} and f_{Fe} are the fractions of MgO and steel reacted respectively. In region I, particle sizes are close to or larger than drill bit diameter, and spalling and caving releases seem unlikely. Thus, little needs to be concerned with region I. Spalling and caving releases will be possible only when $f_{MgO} + f_{Fe} < 0.8$ (region II).

	0	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	1
0	2E-06	8.5E-06	3.61E-05	0.000153	0.000651	0.002766	0.011751	0.049919	0.212053	0.900783	2
0.1	7.98E-06	3.39E-05	0.000144	0.000611	0.002597	0.011032	0.046864	0.199073	0.845648	2	2
0.2	3.18E-05	0.000135	0.000574	0.002438	0.010357	0.043995	0.186889	0.793889	2	2	2
0.3	0.000127	0.000539	0.002289	0.009723	0.041302	0.17545	0.745297	2	2	2	2
0.4	0.000506	0.002149	0.009128	0.038774	0.164711	0.69968	2	2	2	2	2
0.5	0.002017	0.008569	0.036401	0.154629	0.656854	2	2	2	2	2	2
0.6	0.008045	0.034173	0.145165	0.61665	2	2	2	2	2	2	2
0.7	0.032082	0.13628	0.578907	2	2	2	2	2	2	2	2
0.8	0.127939	0.543473	2	2	2	2	2	2	2	2	2
0.9	0.510209	2	2	2	2	2	2	2	2	2	2
1	2	2	2	2	2	2	2	2	2	2	2

Table 8. Calculation of particle size distribution for remaining iron- and aluminum-based metals and alloys. It is calculated that the particle diameters need to be reduced by 0.01975 m for 40% corrosion (column 3)[*]. Column 4 = (column 3/column 1)^3 x column 2. Column 5 = column 4/total volume.

Initial iron- and aluminum-based metals and alloys		Remaining iron- and aluminum-based metals and alloys		
Diameter (m)	Volume fraction	Diameter (m)	Remaining volume (m^3)	Volume fraction
0.00782	8.06E-05	0	0	0
0.012412	0.001209	0	0	0
0.02674	0.068527	0.00699	0.001224	0.002038
0.057605	0.282169	0.037855	0.080075	0.133355
0.124096	0.088682	0.104346	0.052722	0.087801
0.204868	0.264836	0.185118	0.195389	0.325397
0.457114	0.017978	0.437364	0.015747	0.026225
0.503951	0.024091	0.484201	0.021368	0.035586
0.541236	0.047509	0.521486	0.042496	0.070772
0.880982	0.204919	0.861232	0.191444	0.318827
	Total initial volume 1.00		Total remaining volume 0.60	

[*] Given 1 m^3 of the group 1 waste, the total remaining volume after the particle diameters reduced by Δd due to dissolution = $\sum_{d_i > \Delta d} f(d_i)\left(\dfrac{d_i - \Delta d}{d_i}\right)^3$, where d_i is the diameter of initial particles (column 1) and $f(d_i)$ is the volume fraction of the initial particles with a diameter of d_i (column 2). By trial and error, it is found that the particle diamaters need to be reduced by 0.01975 m for 40% corrosion (equivalent to 60% remaining).

Table 9. Volume fractions of waste groups for the bounding case. Column 4 is calculated for 1 m³ of initial waste. A 50% porosity prior to cementation is assumed. Note that the solid volume in reactions (2) and (3) is expanded by 373.4 % [13] and 219.0% [14]. Column 5 = column 4/1.031; column 6 = column 4/0.703.

Waste group	Material	Initial waste Volume fraction (m³)	Degraded waste Volume (m³)	Degraded waste Volume fraction (cements included)	Degraded waste Volume fraction (cements not included)
1	Fe- and Al-base metals	0.143	0.0858[a]	0.0833	0.122
2	Cellulosics and solidified organics	0.131	0	0	0
3	Rubbers and plastics	0.115	0	0	0
4	Other metals, inorganic, vitrified, soils, cements, and solidified inorganics	0.235	0.235	0.228	0.334
5	Salt	0.12	0.12	0.116	0.171
6	MgO backfill	0.255	0.153	0.148	0.218
7	Free particles precipitated from metal corrosion		0.0533[b]	0.052	0.076
7	Free particles precipitated from MgO hydration		0.0558	0.054	0.079
	Cement formed from metal corrosion		0.1600[c]	0.155	
	Cement formed from MgO hydration		0.1675	0.163	
			Total volume (cements included): 1.031		
			Total volume (cements not included): 0.703		

a. 0.143 m³ x (1 - 40%) = 0.0858 m³. b. 0.143 m³ x 40% x 25% x 373.4% (volume expansion) = 0.0533 m³. c. 0.143 m³ x 40% x 75% x 373.4% = 0.1600 m³. The volumes in column 4 for MgO and its reaction products can be calculated following the same procedure.

Table 10. Particle size distribution for the lower bounding case. Waste group 7 represents the fine free particles precipitated from steel corrosion and MgO hydration. When cements are included in total solid volume, they are assumed to have the same particle size distribution as that in waste group 4 and salt.

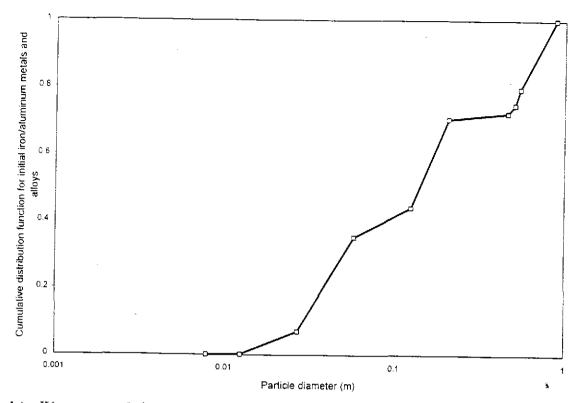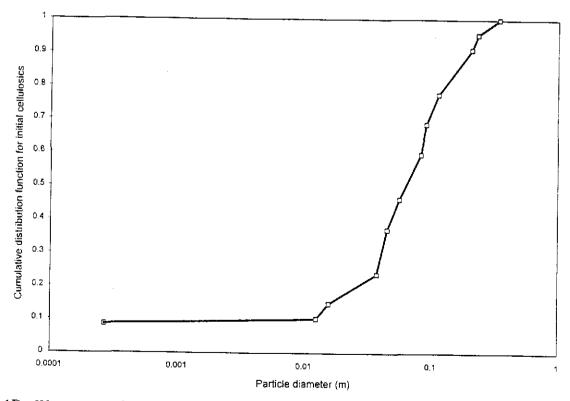| Waste group	Diameter (m)	Cement volume not included		Cement volume included	
		Volume fraction	CDF	Volume fraction	CDF
7	2.00E-06	0.155295	0.155295	0.105939	0.105939
5	2.68E-06	0.016256	0.171551	0.017206	0.123145
4	6.21E-06	6.54E-05	0.171616	6.92E-05	0.123215
5	1.24E-05	0.016256	0.187872	0.017206	0.14042
5	5.76E-05	0.016256	0.204128	0.017206	0.157626
4	0.000124	0.002256	0.206384	0.002388	0.160014
4	0.000124	0.000981	0.207365	0.001038	0.161052
4	0.000124	0.055104	0.262469	0.058324	0.219376
4	0.000248	0.116291	0.37876	0.123086	0.342462
4	0.000248	0.04997	0.42873	0.05289	0.395351
5	0.000268	0.016256	0.444986	0.017206	0.412557
5	0.001242	0.016256	0.461242	0.017206	0.429763
6	1.69E-03	0.217625	0.678867	0.230341	0.660104
5	0.005762	0.016256	0.695123	0.017206	0.67731
1	0.00699	2.49E-04	0.695372	0.000264	0.677573
4	0.024823	0.00641	0.701782	0.006785	0.684358
5	0.02674	0.016256	0.718038	0.017206	0.701564
1	0.037855	0.016287	0.734325	0.017239	0.718802
4	0.074115	0.029709	0.764034	0.031445	0.750247
4	0.081955	0.005321	0.769355	0.005632	0.755879
1	0.104346	0.010724	0.780079	0.011351	0.76723
4	0.124096	0.005265	0.785344	0.005573	0.772802
5	0.124096	0.016256	0.8016	0.017206	0.790008
1	0.185118	0.039742	0.841342	0.042064	0.832072
4	0.21219	0.007522	0.848864	0.007962	0.840034
4	0.413466	0.004689	0.853553	0.004963	0.844997
1	0.437364	0.003203	0.856756	0.00339	0.848387
1	4.84E-01	0.004346	0.861102	0.0046	0.852987
1	0.521486	0.008644	0.869746	0.009149	0.862136
5	0.575914	0.016256	0.886002	0.017206	0.879342
4	0.65778	0.050676	0.936678	0.053637	0.932978
1	0.861232	0.03894	0.975618	0.041215	0.974194
5	1.240674	0.024384	1.000002	0.025809	1.000002

Arithmetic mean: 0.133564
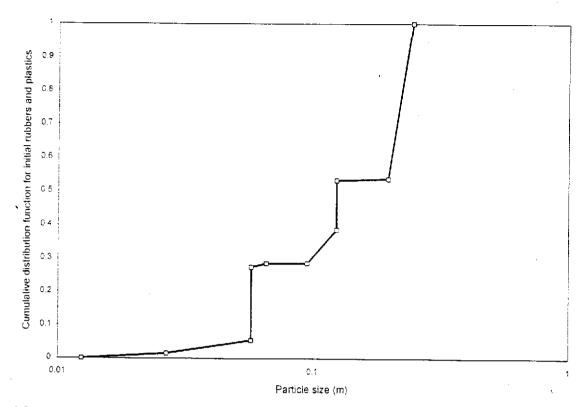Geometric Mean: 0.001317
Median: 0.0015

1A. Waste group 1: iron- and aluminum-based metal/alloys
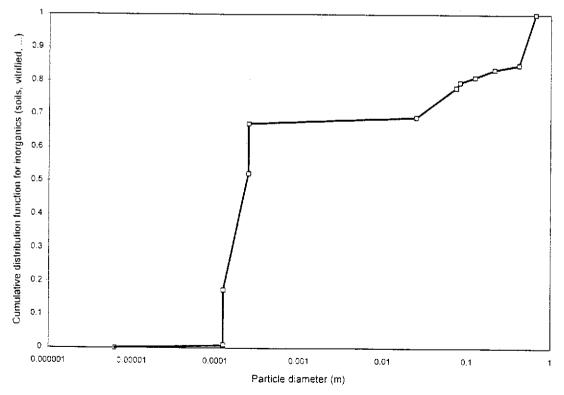

1B. Waste group 2: cellulosics and solidified organics
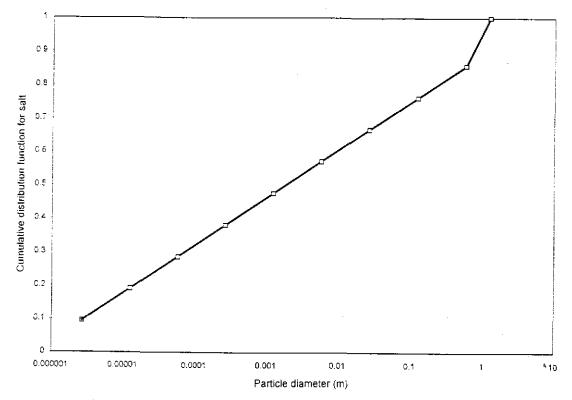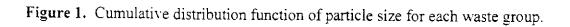
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1C. Waste group 3: rubber and plastics

1D. Waste group 4: other metals. inorganic. vitrified, soils, cements, solidified and inorganics.

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Figure caption axis labels: Cumulative distribution function for salt (y-axis), Particle diameter (m) (x-axis)

1E. Waste group 5: salt

Figure 1. Cumulative distribution function of particle size for each waste group.

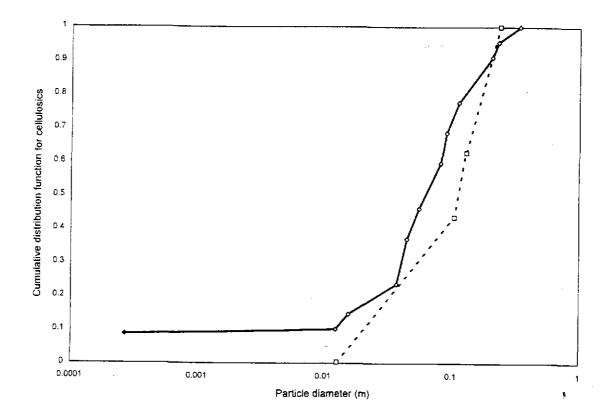**Figure 2.** Cumulative distribution function of particle size for initial waste, including salt and MgO.

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Figure 3. Comparison of particle size distributions between degraded (dashed line) and initial (solid line) cellulosics. The dashed line represents 96% of cellulosics biodegraded and is calculated in Table 5.
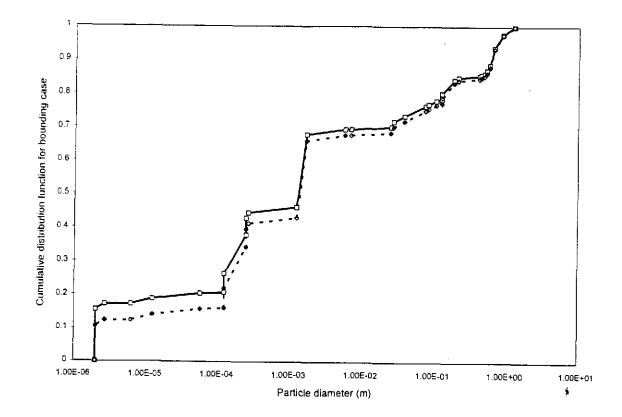
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Figure 4. Cumulative distribution function of particle size for the lower bounding case. Solid line - Cement volume is not accounted in total solid volume. Dashed line - Cement volume is accounted in total solid volume and the cements are assumed to have the same particle size distribution as that in waste group 4 and salt.

Distribution

MS 1320 J. Nowak (Org. 6831)
MS 1322 K. M. Knowles (Org. 6121)
MS 1325 D. S. Coffey (Org. 6811)
MS 1328 H. N. Jow (Org. 6848)
MS 1328 D. R. Anderson (Org. 6849)
MS 1328 M. S. Tierney (Org. 6848)
MS 1335 M. S. Y. Chu (Org. 6801)
MS 1341 J. T. Holmes (Org. 6832)
MS 1341 H. W. Papenguth (Org. 6832)
MS 1395 L. E. Shephard (Org. 6800)
MS 1395 M. G. Marietta (Org. 6821)
MS 1395 S. Y. Pickering (Org. 6811)
MS 1395 K. W. Larson (Org. 6821)
MS 1395 P. E. Sanchez (Org. 6821)
MS 1395 N. F. Tencza (Org. 6811)
MS 1395 Y. Wang (Org. 6821)

CAO/DOE G. T. Basabilvazo

anh 8/25/97

MS 1330 SWCF-A: 1.2.01.5.3.1;~~CO/CCA;QA;PARTICLE SIZE;CAVING;SPALLING~~
PDD; QA; WIPP WASTE PARTICLE SIZE