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CONTENTS

	Page
1. PURPOSE	5
2. METHOD.....	5
3. ASSUMPTIONS	5
4. USE OF COMPUTER SOFTWARE AND MODELS	7
4.1 SOFTWARE.....	7
4.1.1 MCNP.....	7
4.2 SOFTWARE ROUTINES	7
4.2.1 Excel.....	7
4.3 MODELS	7
5. CALCULATION.....	8
5.1 CRITICALITY IN THE NEAR-FIELD ZONE	8
5.2 CRITICALITY IN THE FAR-FIELD ZONE	9
5.3 PROBABILITY OF EXTERNAL CRITICALITY, UPPER BOUND	11
6. RESULTS.....	13
6.1 EXTERNAL CRITICALITY IN THE INVERT.....	13
6.2 CRITICALITY IN A FRACTURED MATRIX OF TUFF	13
6.3 PROBABILITY OF EXTERNAL CRITICALITY, UPPER BOUND	14
7. REFERENCES.....	16
8. ATTACHMENTS	18

FIGURES

	Page
Figure 5-1. Invert Dimension (not to scale).....	9
Figure 5-2. Tuff Matrix with Fracture (not to scale)	10

TABLES

	Page
Table 5-1. Tuff Composition.....	8
Table 5-2. EQ3/6 Results for U and Pu Accumulations from Ceramic Bearing Waste Packages	9
Table 5-3. U and Pu Accumulations in the Far-Field Zone from Ceramic Bearing Waste Packages.....	11
Table 6-1. k_{eff} Values for the Invert.....	13
Table 6-2. k_{eff} Values for the Tuff Matrix.....	14
Table 8-1. List of Attachments.....	18
Table 8-2. Files Contained in Attachment I.....	18

1. PURPOSE

There are two principal objectives of this calculation: (1) evaluate the criticality of those significant accumulations of fissile material external to the waste package (WP) containing Plutonium Disposition waste forms, and (2) estimate the probability of those configurations (fissile accumulations) found to be critical. The scope of this calculation is limited to the fissile accumulations in the invert and external drift regions that have been defined by the calculations using the geochemistry-transport codes (References 3 and 4). This calculation will be used to support the analysis that will be done to demonstrate concept viability related to use in Monitored Geologic Repository environment. This calculation is in accordance with Technical Document Development Plan TDP-DCC-MD-000001 REV 00, item 7 (Reference 1).

This calculation has been prepared according to Procedure AP-3.12Q, *Calculations* (Reference 19).

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2. METHOD

Criticality is characterized by the effective neutron multiplication factor (k_{eff}), which is calculated using the Monte Carlo neutron transport code MCNP Version 4B2 code (Reference 2). The calculation was performed using continuous energy cross-section libraries from the Evaluated Nuclear Data File (ENDF). The concentrations of fissile material outside of the WP are given in Reference 3 for the near-field zone and Reference 4 for the far-field zone.

With regard to the development of this calculation, the control of electronic management of data was evaluated in accordance with AP-SV.1Q, *Control of the Electronic Management of Information* (Reference 20). The evaluation (Reference 18) determined that current work processes and procedures are adequate for the control of the electronic management of data for this activity.

3. ASSUMPTIONS

- 3.1 The invert is composed of crushed tuff (Reference 13, Page 6-47) with an average void space of 35% (Reference 16, Figure 9.4.9). It is assumed that the void space is fully saturated with water. The rationale for this assumption is that it is conservative since moderation is favorable to criticality. This assumption is used in Sections 5.1 and 6.1.
- 3.2 The tuff has a porosity of 11% (Reference 6, Table 1, Units TMN, TLL, and TM2) and it is assumed to be fully saturated with water. It is also assumed that the tuff matrix apertures (1 mm wide and 10 cm apart, Section 5.2) are filled with full density water. The rationale for

these assumptions is that they are conservative since moderation is favorable to criticality. This assumption is used in Sections 5.2 and 6.2.

- 3.3 For cases with no decay, only Pu-239 is represented in the MCNP cases. For cases with decay, the decayed fraction of the Pu-239 is represented by U-235. For the cases representing decay by one Pu-239 half-life, the U-235 is conservatively represented by a uranium enrichment of 20% (one-half of Pu/ [U + one-half of Pu], Reference 8, Table 3.1) and a conservative uranium enrichment of 34% for cases approximating complete decay of Pu-239. The rationale for this assumption is that it is conservative since the enrichments of the plutonium and uranium are assumed to be higher than the baseline values (Reference 8, Table 3.1) and uranium is not represented in the cases in which it acts only as a parasitic absorber. This assumption is used in Sections 5 and 6.
- 3.4 It is assumed that the plutonium in the ceramic is 100% Pu-239. The rationale for this assumption is that it is conservative (highest possible content of fissile material.) This assumption is used in Sections 5 and 6.
- 3.5 It is assumed that the Weibul distribution parameters derived from the Viability Assessment (VA) WP design (α , β , and θ , given as 12.099, 16.425, and 0, respectively) (Reference 10, Table 5.1.2-1) are appropriate for evaluating the probability distribution of WP breach times for this calculation. The rationale for this assumption is that the plutonium disposition analyses providing input to this calculation have been based on VA design WPs. This assumption is used in Sections 5 and 6.

4. USE OF COMPUTER SOFTWARE AND MODELS

4.1 SOFTWARE

4.1.1 MCNP

The MCNP code is used to calculate the k_{eff} of the near- and far-field configurations of fissile material. The software specifications are as follow:

- Software name: MCNP
- Software version/revision number: Version 4B2
- Computer Software Configuration Item (CSCI): 30033 V4B2LV
- Computer type: Hewlett Packard (HP) 9000 Series Workstations
- Computer processing unit number: Software is installed on the Civilian Radioactive Waste Management System (CRWMS) Management and Operating (M&O) Contractor workstation "Bloom", Tag #700887. Access to the software is through the M&O HP network.

The input and output files for the various MCNP calculations are documented in Attachment I. The calculation files described in Sections 5 and 6 are such that an independent repetition of the software use may be performed. The MCNP software used is appropriate for the calculations documented herein, was obtained from the Software Configuration Management in accordance with appropriate procedures, and used only within the range of validation as documented in Reference 5.

4.2 SOFTWARE ROUTINES

4.2.1 Excel

- Title: Excel
- Version/Revision Number: Microsoft® Excel 97 SR-2
- This software is installed on a personal computer running Microsoft Windows 95.

The Excel spreadsheet programs were used to calculate the isotopic composition of the materials in the waste package as documented in Attachment I (spreadsheets "invert.xls" and "tuff.xls") of this calculation. The user-defined formulas, inputs, and results have been documented according to Procedure AP-SI.1Q, *Software Management* (Reference 21) where used in the referenced spreadsheets. Sufficient detail is included to allow an independent repetition of computations. Only one version of these formulas exists since they were developed for a one-time use.

4.3 MODELS

None used.

5. CALCULATION

This section describes the evaluation of the criticality of significant accumulations of fissile material external to waste packages containing plutonium disposition waste forms, where such accumulations have been defined by the calculations using the geochemistry-transport codes. Two separate zones for fissile material accumulation were considered: near- and far-field zones. The near-field zone criticality investigation is described in Section 5.1. Section 5.2 describes calculations of the far-field zone criticality. Fissile material accumulates in the near-field zone in large interstices between small rock fragments, while it accumulates on fracture walls in the far-field zone. Table 5-1 lists tuff composition and density.

Table 5-1. Tuff Composition (Reference 15 & Spread Sheets "invert.xls")

Element	Weight Percent (wt.%)	Element	wt.%
SiO ₂	76.83	Na ₂ O	3.59
Al ₂ O ₃	12.74	K ₂ O	4.93
FeO	0.84	TiO ₂	0.1
MgO	0.25	P ₂ O ₅	0.02
CaO	0.56	MnO	0.07
Particle Density = 2.54 g/cm ³ (Reference 6 & Spread Sheet "invert.xls")			

5.1 CRITICALITY IN THE NEAR-FIELD ZONE

The near-field zone is the drift region exterior to the WPs. The only location in the drift where there can be a significant accumulation of fissile material is beneath the waste package in the invert. Figure 5-1 gives a representation of the near-field zone as taken from Reference 24, Figure 5. The invert is made up of crushed tuff with a porosity of 35% (see Assumption 3.1). The volume of the invert is calculated in the spread sheet "invert.xls", in the sheet "Volume of Foot Print in Invert". The composition of the tuff is given in Table 5-1 and in the spread sheet "invert.xls", in the sheet "tuff".

Fissile material accumulation in the invert is taken from EQ3/6 geochemistry transport evaluations given in Reference 3. As discussed in Section 6 of Reference 3, significant accumulation in the invert only occurs under waste packages containing the ceramic plutonium disposition waste form. The source term of fissile material from waste packages containing the mixed oxide (MOX) plutonium disposition waste form is very small (Reference 7, Section 6) resulting in insignificant accumulations of fissile material. Thus, k_{eff} values for these cases will not be evaluated. Table 5-2 is a listing of the cases with significant fissile material accumulations taken from Section 6 of Reference 3. The accumulation values are given per liter void, which corresponds to 4.72 kg of tuff (2.86 liters of 35% tuff porosity). In order to calculate the total accumulation under the waste package, the accumulation per liter void is multiplied by the ratio of the invert volume through which the flow is represented to 2.86 liters. All the cases in Table 5-2, except 2a, correspond to focusing of the flow from the waste package onto a horizontal area 1/10th the waste package footprint (full WP footprint ~2x3 m). Such focusing is

likely to occur if all the exit flow from the waste package is from a single hole in the bottom (Reference 3). In the MCNP cases with no Pu-239 decay, the fissile material is conservatively represented as Pu-239 only (Assumption 3.3) and no U-238 from the ceramic material. For the MCNP cases with decay of Pu-239, both U-235 from the Pu-239 decay and U-238 from the ceramic are represented as well as the remaining Pu-239. The atomic densities used in the input are calculated in the spread sheet “invert.xls”, in the sheet “Cases”.

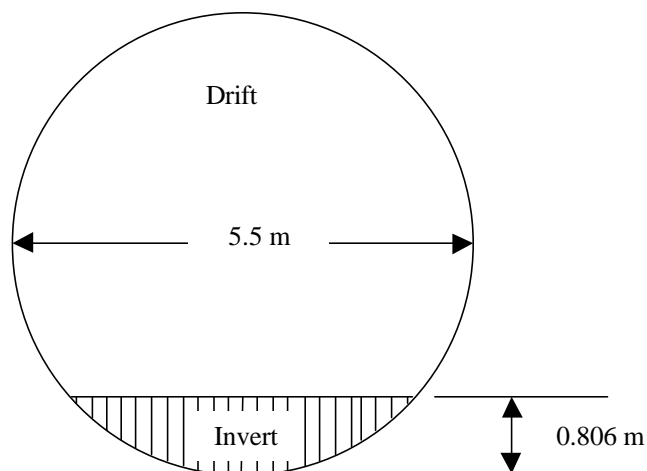


Figure 5-1. Invert Dimension (not to scale)

Table 5-2. EQ3/6 Results for U and Pu Accumulations from Ceramic Bearing Waste Packages

MCNP File Name	Case Code	Max Pu mols/liter Void	Max U mols/liter Void	Comments
011	9o7	0.941	Not Determined	Test effect of using a corrosion rate 10 times the nominal. No decay.
012	2a	0.0104	0.264	Baseline “reducing” case. No decay.
013	4ao	0.104	0.370	Baseline “reducing” case. No decay.
014	6	0.053	0.329	Test effect of pre-decaying system by one ²³⁹ Pu half-life.
N/A ^a	906	0.488	≥0.67	Test of corrosion rate sensitivity - No decay.
N/A	906 [^]	0.246	≥0.59	Test of corrosion rate sensitivity - pre-decay by one ²³⁹ Pu half-life

NOTE: ^a N/A – Not Applicable since case was not used for MCNP calculations.

5.2 CRITICALITY IN THE FAR-FIELD ZONE

The far-field zone is just several meters of rock immediately beneath the drift. The composition of the tuff rock is given in Table 5-1. The porosity of the tuff below the repository is 11 % (see Assumption 3.2). A representation of the aperture matrix in the tuff is given in Figure 5-2. The size of the matrix (W) varies between 1.6 m to 5.58 m depending on the accumulation

calculations documented in Reference 4. The width of the fractures is held constant at 1 mm and spaced at a constant 10 cm interval. The fractures are represented in a two-dimensional grid and filled with fissile material (Pu-239 and/or U-235) and water. As discussed in Section 6 of Reference 4, significant accumulation in the far-field zone only occurs under waste packages containing plutonium disposition ceramic waste form. The source term of fissile material from waste packages containing the plutonium disposition waste form, MOX, is very small (Section 6, Reference 7) resulting in insignificant accumulations of fissile material. Thus, k_{eff} values for these cases will not be evaluated. Table 5-3 is a listing of the cases with significant fissile material accumulation taken from Reference 4. Criticality cases for the far-field zone were all taken from Reference 4; Cases 201-203 from Table 6-6 (see spread sheet “tuff.xls”, sheet “1,000,000”), cases 204-206 from Table 6-5 (see spread sheet “tuff.xls”, sheet “20,000”), and case 207 from Table 6-7 (see spread sheet “tuff.xls”, sheet “20,000 1x”). In the MCNP cases with no Pu-239 decay, the fissile material is conservatively represented as Pu-239 only (Assumption 3.3) and no U-238 from the ceramic material. For the MCNP cases with decay of Pu-239, both U-235 from the Pu-239 decay and U-238 from the ceramic are represented as well as the remaining Pu-239.

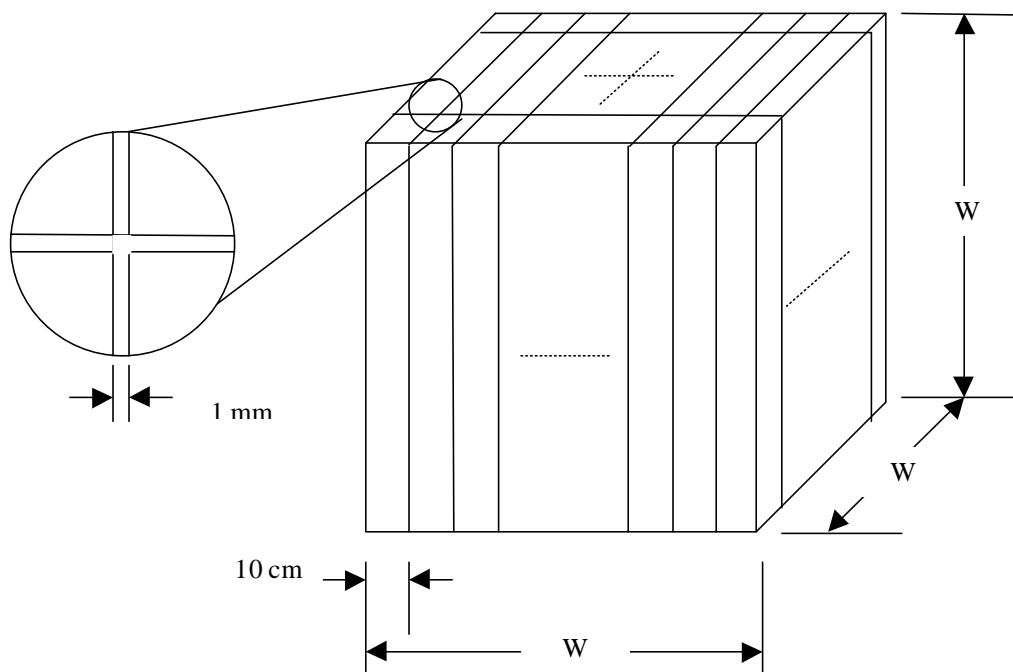


Figure 5-2. Tuff Matrix with Fracture (not to scale)

Table 5-3. U and Pu Accumulations in the Far-Field Zone from Ceramic Bearing Waste Packages

MCNP File Name	Cube Volume (m ³)	Degradation Rate	Decay	Total Accumulation (Moles)	
				Pu	U
201	195.1	10x	No Decay	78.6	39.8
202	195.1	10x	24,100 years	29.3	52.2
203	195.1	10x	Complete Decay of Pu-239	-	124.5
204	4.1	10x	No Decay	85.4	23.2
205	4.1	10x	24,100 years	30.2	36.4
206	4.1	10x	Complete Decay of Pu-239	-	57.4
207	4.1	1x	No Decay	0.32	8.07
208	19.7	10x	No Decay	84.6	25.3

5.3 PROBABILITY OF EXTERNAL CRITICALITY, UPPER BOUND

No critical accumulations in either the near- or far-field zones were identified for the MOX waste form. Critical accumulations from the immobilized ceramic waste form in the external criticality were limited to the highest degradation rates of this waste form. These degradation rates exceeded the maximum rates given in Section 6.1 of Reference 8. For those accumulations found to be critical, an upper bound to the probability of criticality is estimated.

References 3 and 4 have provided estimates of the fissile accumulations in the invert and the far-field zone, respectively, under various assumptions of determining parameters (e.g., waste form degradation rates and environmental parameters inside and outside the waste package). A comprehensive estimate of probability of criticality would require probability distributions for all such parameters. Unfortunately, the only probability distributions presently available are for degradation rates of the waste package materials, and for the environmental parameters relating to water dripping into the waste package. These distributions have been used to develop a model of waste package degradation, implemented by the Monte Carlo code, WAPDEG (Reference 9). This code provides a distribution of waste package breach times.

For plutonium-bearing waste forms, the time of breach is an important determinant of criticality. With increasing time of breach, increasing amounts of Pu-239 decay into U-235. Because the U-235 is less reactive with respect to criticality and is accompanied by neutron absorbing U-238 from the ceramic, a delay of waste package breach will also decrease the probability of criticality because more of the Pu-239 will have decayed to the less reactive U-235. Note that U-238 was also present in the *no-decay* Pu-239 configurations but it was conservative to not take the U-238 into account for those criticality calculations.

The methodology for calculation of the upper bound of criticality probability starts with the recognition of the fact that the amount of Pu accumulated is approximately linearly proportional to the amount of Pu present in the waste package at the time of breach. This correlation is demonstrated for the invert accumulations in Table 5-2 by comparing the accumulation values for Cases 4a0 and 9o6 with the values from Cases 6 and 9o6[^], respectively, in which the waste package does not breach until one half-life of Pu-239. The members of the latter pair are seen to have approximately half the Pu accumulation of the corresponding member of the former pair, which is consistent with their having half the amount of Pu-239 at the time of breach. This is also seen to be approximately true, on a conservative basis, for the far-field accumulations summarized in Table 5-3. Case 204 corresponds to breach at time zero, and Case 205 corresponds to breach at 24,100 years, which is the half-life of Pu-239.

The next step in the methodology is the identification of the minimum amount of fissile accumulation (primarily Pu-239) required for criticality. This identification process for the far-field accumulation is described in Section 6.3. For a criticality to occur, there is a maximum Pu-239 decay time before breach that can not be exceeded before the Pu-239 remaining at breach time is too small to lead to a critical mass accumulation. This maximum decay time will simply be the decay time that reduces the initial Pu inventory by the same ratio as the minimum accumulation needed for criticality divided by the maximum possible accumulation (achieved only if the waste package breach occurs at time zero). Finally, the probability upper bound is estimated as the probability that the waste package breach time is less than this maximum decay time. The application of this concept is given in Section 6.3, below.

The principal quantitative component of this methodology is the probability distribution of waste package breach times. The probability distribution of the natural logarithm of waste package breach times is taken as the same Weibul distribution for the waste package breach times (in years) that was used for the VA waste package design (Assumption 3.5). The VA design Weibul distribution was used for this calculation because the plutonium disposition analyses were based on the VA design waste package. The three-parameter Weibul distribution is characterized by the parameters α , β , and θ , which are given as 12.099, 16.425, and 0, respectively, in Reference 10, Table 5.1.2-1.

6. RESULTS

The results of external criticality evaluation due to fissile material accumulation from waste packages containing MOX and ceramic plutonium disposition waste forms in the near- and the far-field zones are presented in Sections 6.1 and 6.2, respectively. The k_{eff} results represent the average collision, absorption, and track length estimator from the MCNP calculations. The standard deviation (σ) represents the standard deviation of k_{eff} about the average combined collision, absorption, and track length estimate due to Monte Carlo calculation statistics. The average energy of neutrons causing fission (AENCF) in MeV is also given in Sections 6.1 and 6.2.

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6.1 EXTERNAL CRITICALITY IN THE INVERT

This Section presents the results of the calculation described in Section 5.1. The various amount of fissile material mix with tuff and water is homogenized over the invert volume. Results are listed in Table 6-1.

Table 6-1. k_{eff} Values for the Invert

File Name	Case Code	Density (g/cm ³) Total accumulation (kg)			k_{eff}	2s	AENCF (MeV)
		Pu-239	U-235	U-238			
011	9o7	0.0787 51.66	-	-	1.30663	0.00260	0.02420
012	2a	0.00087 5.709	-	-	0.28094	0.00050	0.00146
013	4ao	0.0087 5.709	-	-	0.88944	0.00188	0.00411
014	6	0.0044 2.91	0.0054 3.55	0.0219 14.39	0.81970	0.00198	0.00604

6.2 CRITICALITY IN A FRACTURED MATRIX OF TUFF

This section presents the results of the calculations described in Section 5.2. The fissile material is Pu, U, or a combination of both with the matrix size varied to accommodate the quantity of accumulated material. Descriptions and results of the different cases investigated are listed in Table 6-2.

Table 6-2. k_{eff} Values for the Tuff Matrix

File Name	Density in apertures (g/cm ³) Total accumulation (kg)			k_{eff}	2s	AENCF ^b (MeV)
	Pu-239	U-235	U-238			
201	0.0048 18.79	-	-	0.05297	0.00010	0.00289
202	0.0018 7.00	0.0006 2.45	0.0026 9.94	N/A ^a	N/A	N/A
203	-	0.0026 9.95	0.0050 19.56	0.01582	0.00004	0.00326
204	0.2505 20.42	-	-	0.96906	0.00182	0.00541
205	0.0886 7.22	0.0210 1.71	0.0850 6.93	0.59946	0.00126	0.00414
206	-	0.0563 4.59	0.1106 9.02	0.24524	0.00058	0.00442
207	0.0009 0.08	-	-	N/A	N/A	N/A
208	0.0516 20.22	-	-	0.42083	0.00076	0.00250

NOTES: ^a N/A – Not Applicable. Fissile material concentration was so low that MCNP could not estimate k_{eff} .

^b Average Energy of Neutrons Causing Fission.

6.3 PROBABILITY OF EXTERNAL CRITICALITY, UPPER BOUND

Using the methodology described in Section 5.3, the upper bound to the probability of criticality can be computed for extreme source terms from the ceramic waste form. There is no possibility of criticality for source terms based on the nominal ceramic degradation rate as shown by the results listed in Table 6-2, Case 207 and Table 6-1, Case 013. Case 207, with the highest far-field accumulation from the source term produced by the nominal degradation rate, resulted in a very low fissile material concentration such that MCNP could not estimate the k_{eff} . Case 013, with the highest invert accumulation for the nominal source term, resulted in a k_{eff} of 0.8894.

The far-field Pu accumulation required for criticality is determined by interpolating between Cases 204 and 205 that bracket the criticality limit, $k_{\text{eff}} = 0.95$, in the 4.1-m³ volume containing the highest fissile concentration. (The criticality limit of 0.95 derives from the criticality control requirement for emplacement and isolation of radioactive waste for maintaining a minimum 5% margin below unity for the system k_{eff} , after allowing for biases and uncertainties [Reference 17, Section 2.1.1]). This interpolation gives the minimum accumulation of 19.7 kg. The maximum permitted decay time before basket degradation is computed from the formula $\ln(M/m) \cdot T_{1/2}(^{239}\text{Pu})/\ln(2)$, where $M=20.42$ kg is the maximum accumulation (corresponding to the zero decay case, 204), and $m = 19.7$ kg is the minimum Pu accumulation to produce criticality calculated above, $T_{1/2}(^{239}\text{Pu}) = 24,100$ y is the half-life of ²³⁹Pu, and $\ln(2) = 0.693$. The resulting

maximum decay time before waste package breach that can result in possible critical accumulations is approximately 1200 years.

The upper bound for the criticality probability is computed from the Weibul cumulative probability formula, where the random variable is the transformed breach time $\ln(t)$:

$$\Pr\{\text{breach time} \leq t\} = 1 - \exp(-((\ln(t) - \theta)/\alpha)^\beta), \quad (\text{Eq. 1})$$

where α , β , and θ are the Weibul distribution parameters equal to 12.099, 16.425, and 0, respectively as noted in Section 5.3. It should be noted that the Weibul distribution does not require a logarithmic transformation of the random variable, t , as is done for this calculation. The $\ln(t)$ transformation has been used because it is the form used to obtain a least-squares fit to the WAPDEG breach time statistics (Reference 10, Section 5). Hence, it is acceptable to use the $\ln(t)$ transformation, because there is no specific physical significance to the Weibul distribution in this calculation, whatever the functional form of the independent variable. The upper bound for the probability of criticality for a single WP is calculated from Equation 1 as 1.5×10^{-4} . An upper bound to the expected number of criticalities (0.02) is computed by multiplying the upper bound for the probability by the number of waste packages with the ceramic waste form, 128 (Table 2-12 of Reference 14).

It should be noted that using the VA distribution of waste package breach times is very conservative, because the newer Enhanced Design Alternative (EDA) II design gives much longer times to waste package breach. The median breach time (t_m , which makes $\Pr\{\text{breach time} \leq t_m\} = .5$) for the VA waste package is estimated to be 138,000 years, while the median breach time with the EDA II waste package (with Weibul parameters α , β , and θ estimated as 2.347, 4.81, and 10.82 in Reference 10, Table 5.1.2-1) is estimated to be 440,000 years.

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8. ATTACHMENTS

Table 8-1 contains a list of the attachments included in this calculation. Table 8-2 presents a description of the contents of Attachment I.

Table 8-1. List of Attachments

Attachment Number	Description	Pages
I	Compact Disc (CDROM)	N/A

Table 8-2. Files Contained in Attachment I

Name	Date	Time	Size (byte)
011	07/21/00	10:33 AM	1,968
011o	07/21/00	10:33 AM	196,361
012	07/21/00	10:33 AM	1,989
012o	07/21/00	10:33 AM	197,419
013	07/21/00	10:33 AM	2,018
013o	07/21/00	10:33 AM	196,781
014	07/21/00	10:33 AM	2,120
014o	07/21/00	10:33 AM	198,168
201	07/21/00	10:34 AM	2,275
201o	07/21/00	10:34 AM	211,196
202	07/21/00	10:34 AM	2,340
202o	07/21/00	10:34 AM	56,776
203	07/21/00	10:34 AM	2,305
203o	07/21/00	10:34 AM	211,906
204	07/21/00	10:34 AM	2,266
204o	07/21/00	10:34 AM	210,249
205	07/21/00	10:34 AM	2,332
205o	07/21/00	10:34 AM	212,328
206	07/21/00	10:34 AM	2,297
206o	07/21/00	10:34 AM	211,805
207	07/21/00	10:34 AM	2,270
207o	07/21/00	10:34 AM	55,112
208	07/21/00	10:34 AM	2,274
208o	07/21/00	10:34 AM	209,626
invert.xls	09/01/2000	10:16 AM	39,936
tuff.xls	09/01/2000	8:14 AM	50,176