

DISSOLUTION OF HIGH-LEVEL  
NUCLEAR WASTE SOLIDS

by  
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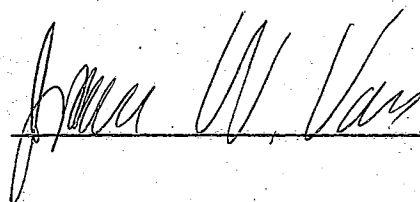
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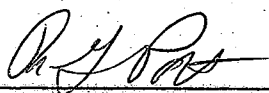
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APPROVAL BY THESIS DIRECTOR

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## ABSTRACT

A work has been done to explore the release of radioisotopes from high-level nuclear waste solids by dissolution. Initially, a generalized expression describing the dissolution of a waste form as a function of time and temperature is derived. Discussion of the element specific dissolution behavior is included. The environmental transport of dissolved radionuclides is next discussed. Equations describing the flow rate of radioisotopes from a waste form, the dilution of radioisotopes as they are transported in a groundwater environment, and the radioactive decay of radioisotopes due to ion exchange with the soils are developed. Finally, the derived equations are used in a sample calculation involving the dissolution of a borosilicate glass high-level waste form and the subsequent environmental transport of the dissolved radionuclides.

## CHAPTER 1

### INTRODUCTION

Commercial nuclear power has been under development for nearly thirty years. During this time period, the recycle of uranium and plutonium has been considered essential to fuel cycle economy and conservation of uranium resources. This strategy has been strengthened by the link between the Light Water Reactor (LWR) fuel cycle and the Fast Breeder Reactor (FBR) fuel cycle.

The LWR and FBR fuel cycle relation is a complex one. Some fuel cycles include a synergistic relation between the two. Others show the FBR as an evolutionary step past the LWR, with the eventual phase out of the LWR systems. Within both strategies, the need for uranium and plutonium recovery by nuclear fuel reprocessing is essential.<sup>1,2</sup>

The need for uranium and plutonium recovery by nuclear fuel reprocessing may be satisfied by several different means. One such method may be the reprocessing of nuclear fuels by government owned installations. Another possibility is for the reprocessing step to be carried out by commercial reprocessing plants. Neither is being done at the present. The choice of government reprocessing of spent nuclear fuel is technically feasible. Commercial fuel reprocessing, while

being technically feasible, is not legally possible at the present, as a variety of regulatory questions restricts the licensing of any commercial nuclear fuel reprocessing plant.

One regulatory issue which is currently being addressed by the Nuclear Regulatory Commission, surrounds the disposition of high-level nuclear wastes. High-level nuclear wastes contain nearly all of the radioactive fission products generated by the fissioning of nuclear fuels. Current reprocessing technology generates these high-level wastes in the form of liquids.

At present, government regulations require that these high-level liquid wastes be solidified within five years of their generation and be transferred to a Federal Government Nuclear Waste Repository within ten years of liquid waste generation.<sup>3</sup> While regulations state that the solidified high-level waste must be "chemically, thermally, and radiolytically stable," it has been determined that specific waste form properties are needed to ensure that the environmental impact of high-level nuclear waste disposal be predictable and environmentally acceptable.

For complete environmental assessment of the impact of nuclear waste disposal, a great deal of information is required about nuclear waste form behavior. Two items which are within this need are the descriptions of waste form dissolution and of environmental transport of radionuclides. This work addresses these two subjects by developing generalized models for each phenomenon.

## CHAPTER 2

### ANALYTIC EXPRESSION RELATING TO RELEASES OF RADIOISOTOPES FROM WASTE FORMS

Radioisotopes may be released from a waste form by dissolution, volatilization, and particulate dispersion. This work analyzes the dissolution of waste solid.

This dissolution mechanism has been referred to as the leaching mechanism. However, the act of leaching implies only the percolation of a liquid about a solid which says nothing of the mechanisms involved in the leaching process. This work views the act of leaching as the solution or dissolution of waste form.

Within this section, one model characterizing the dissolution of solid is developed. A second discussion evolves a model relating the dissolution of a waste form to the environmental transport of dissolved radionuclides.

#### Solution Behavior of Waste Forms

The dissolution of waste forms is a complex phenomenon. The solution process is both temperature and time dependent.<sup>4</sup> In addition, the solution process is element specific.<sup>5</sup> The element specific behavior is discussed first.

It has been observed that alkali metals, being highly soluble, dissolve most readily from waste solids.<sup>5,6</sup> In addition, solubility is seen to decrease as ionic radius increases within a valence state.

These observations are consistent with experimental evidence revealing advanced dissolution behavior of sodium and cesium from waste solids. In contrast, cerium, an abundant element in high-level nuclear wastes, is seen to dissolve from waste solids at a rate much slower than the alkali metals.<sup>5</sup>

Mendel<sup>5</sup> compiled a table demonstrating the preferential dissolution of various elements from potential waste glasses. Mendel studied experimental dissolution results obtained by several different researchers. While several different experimental techniques were used, the comparison of atomic ratios of various elements in the glass forms and in the solutions lead to Mendel's results shown below in Table I.

The time dependent behavior of the dissolution of waste forms has been described by equation (1).<sup>5</sup>

$$L = A \theta^{-1/2} + B \quad (1)$$

where L = solution rate of waste form

$\theta$  = time

A = rate constant related to diffusion of ionic species through waste matrix

B = rate constant related to corrosion of waste matrix

Equation (1) is seen to have a singularity at  $\theta = 0$  sec. Since experimental measurements to which this expression has been fit tend not to be for times less than two hours, the equation is considered not to be valid for these small times.

This expression has been further extended to the arrhenius form in equation (2).<sup>12</sup>

Table I. Relative Dissolution of Elements in Glass

Glass Type	Relative Dissolution	Reference
Silicate glass	Cs>Sr>Fe>Zr	Eliassen and Goldman <sup>7</sup>
Lead glass	Cs>Al>Pb>Sr,Ce	Paige <sup>8</sup>
Phosphate ceramic	Na, Cs>Sr>Zr-Nb>Ru,Ce	Allemann <sup>9</sup>
Borosilicate glass	Cs, Na>Zr-Nb>Ru,Ce	Allemann <sup>9</sup>
Borosilicate glass	Na>Si>B>Cs>Sr>Ce,Tb	Elliot and Auty <sup>6</sup>
Borosilicate glass	Na>B>Si	Heimerl et al. <sup>10</sup>
Phosphate glass	Cs>Na>Sr>Ru>Ce	Mendel and McElroy <sup>11</sup>

$$L = ae^{-\frac{\Delta H_d}{RT}} t^{-1/2} + be^{-\frac{\Delta H_c}{RT}} \quad (2)$$

where a, b = constants (element and solid matrix specific)

$\Delta H_d$  = activation energy of diffusion (element and solid matrix specific)

$\Delta H_c$  = activation energy of corrosion (element and solid matrix specific)

R = gas constant

T = absolute temperature of solid matrix-solution interface.

Equation (2) then predicts that at short times, the diffusion of radionuclide species from the waste surface is the dominant effect. After long times, the corrosion of the waste matrix becomes dominant. The separation between short and long times is on the order of  $10^6$  seconds, based strictly on an order of magnitude argument.

The constants a and b are dependent upon several factors. These include solution velocity, solution pH, solution chemical composition, and surface condition of solid matrix.

Addressing the surface condition in particular, it is observed that the surface energy of particles on a jagged surface is higher than for a smooth surface. Thus, less energy is required to remove those particles on a jagged form. This means that if a piece of a waste form has a very rough surface, it will dissolve faster than a piece with a smooth surface.

This surface condition is of special importance in regard to measurement of solution rates of waste forms. Solution rate measurement methods vary widely, yet nearly all require a high degree of waste form subdivision. This may include the grinding of a waste matrix to

a powder, with subsequent dissolution of a specified set of waste particle sizes. Then, in light of the previous discussion, it is apparent that the degree of subdivision of a waste matrix will have an effect on the results of the solution rate measurement.

The various solution rate measurement methods vary the solution flow rates. Some techniques have static solutions, some have cycled solutions, while others have continuously flowing solutions. This too will affect solution rate measurement results.

Results obtained from the various measurement methods vary. Most methods are reproducible within a factor of ten, yet since the values obtained from different methods on similar waste forms vary, the meaning must be questioned. The differences mentioned above along with many others account for result variations.

Calculations using the results of any solution rate measurements must be performed with caution. Recognition of weaknesses in the test methods and thus solution rates is demanded.

The actual meaning of solution rate measurements is usually quite clear. As most methods tend to create a set of "worst possible" dissolution conditions, measured solution rates generally serve as upper limits. Thus, calculations made using measured values reflect this same upper limit behavior.

One particular measurement of the solution rate of borosilicate glass, done at Battelle Northwest Laboratory,<sup>13</sup> has been performed to determine the time and temperature behavior of the solution rate. The measurement has been done using the so called Dynamic Method in which a

1 gram sample of -45 +60 mesh particles (particles of mean diameter between 0.56mm and 0.42mm) of glass are placed in a stainless steel "tea bag", and suspended in distilled water at various temperatures. The water is continuously agitated about the sample and changed at frequent intervals. Data from this measurement have been fit to equation (2) by the Least Squares Method to determine the various constants. The values obtained are shown below:

$$a = 1.62 \times 10^5 \text{ } \mu\text{g}/(\text{m}^2\text{s}^{1/2})$$

$$b = 2.12 \times 10^3 \text{ } \mu\text{g}/(\text{m}^2\text{s})$$

$$\Delta H_d = 3.07 \times 10^3 \text{ J/mole}$$

$$\Delta H_c = 1.20 \times 10^4 \text{ J/mole}$$

### Temperature Distribution in Waste Solids

The energy balance equation predicts the temperature of a solid with time, as seen in equation (3).

$$\nabla k \nabla T + q''' = \rho c \frac{\partial T}{\partial \theta} \quad (3)$$

where  $T$  = temperature (K)

$\theta$  = time (s)

$K$  = thermal conductivity of solid (W/mK)

$q'''$  = volumetric heat generation rate (W/m<sup>3</sup>)

$\rho$  = solid density (kg/m<sup>3</sup>)

$c$  = solid specific heat (J/kgK)

The surface temperature of the waste solid becomes of fundamental importance in determining the solution rates of waste solids, as expressed earlier. Determination of the surface temperature requires

detailed knowledge of the environment which the waste will be placed into. Even if all necessary information is known, the descriptive solution of equation (3) is not a trivial one.

The energy balance equation has been solved for one particular steady state case. A waste cylinder of varying thermal properties described below is buried in some soil with all canister shell material and overpack neglected.

Figure 1 shows the steady state temperature distribution for a waste cylinder and the close proximity soil. The figure was developed for a waste form with a heat generation rate of  $15,420 \text{ W/m}^3$  which corresponds to the heat rate of a ten year out-of-reactor borosilicate glass form. The radius of the cylinder is 0.1525m, seen to be at the intersection of the two curves. The ambient temperature is 300K. The points labelled 1 corresponding to TEMP1 are for the thermal conductivities of the waste form and soil both 1.0W/mK. The points labelled 2 corresponding to the variable TEMP10 are for a waste form conductivity of 10.0 W/mK and a soil thermal conductivity of 1.0 W/mK.

The energy balance equation was solved for this calculation by first assuming that the steady state condition existed. The geometry considered was that of an infinite right cylinder. Four boundary conditions were used since the problem is of two regions, one region with and one region without heat generation. The first boundary condition is that the first radial distance derivative of temperature is zero at the center of the cylinder. The second and third are continuities of

heat flux and temperature at the cylinder-soil interface. The last boundary condition is the ambient temperature at some distance from the cylinder.

One meaning of this type of calculation, aside from the dissolution implications, is related to phase changes which various waste forms undergo at elevated temperatures. As discussed later, devitrification of a glass waste form may occur at a rapid rate with elevated temperatures, as its rate is described by an arrhenius equation.

Another implication of this calculation is that for heat conduction to the environment, the thermal conductivity of the waste form does not change the interface temperature or the temperature profile in the surrounding environment. In the figure, the two temperature curves are seen to merge at the waste form surface, and coincide from that point on.

All of these factors, phase of waste form, surface temperature of waste form, and temperature profile in the close proximity of the waste form, have a marked effect on the conditions for dissolution. The constants in the dissolution equation will be different for each phase of each form. Also the temperature for dissolution will have direct effect on the dissolution rate of a waste form.

1=TEMP1  
2=TEMP10

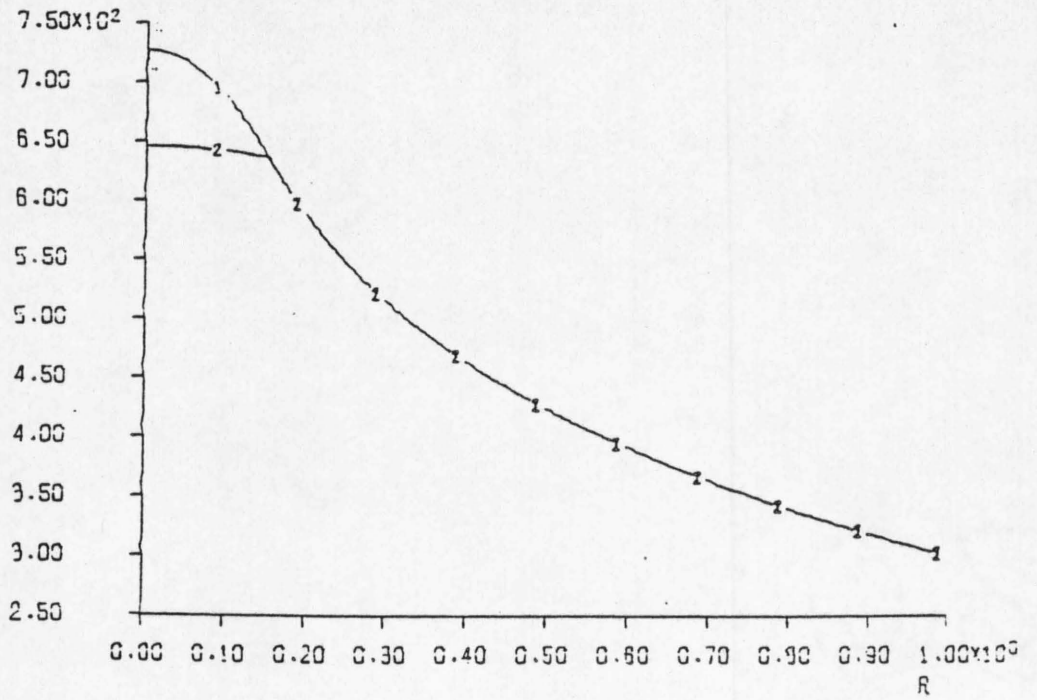


Figure 1. Steady State Temperature (K) versus Radius (M) for Ten Year Old Waste

## CHAPTER 3

### RADIOISOTOPE RELEASE FROM WASTE FORM WITH SUBSEQUENT ENVIRONMENTAL TRANSPORT

The mass flow rate of the radioisotopes into the environment,  $m$ , from a waste form is the product of the solution rate of the solid,  $L$ , and the exposed surface area of the solid,  $A$ , provided the mass flow is independent of the concentration of radioisotopes in the solution.

$$m = L \cdot A \quad (4)$$

Thus, the concentration,  $C_0$ , of radioisotopes in a solution which is leaching the waste form is the quotient of the radioisotope mass flow rate and the volumetric flow rate of the solution,  $V_f$ , as shown in equation (5).

$$C_0 = \frac{m}{V_f} = \frac{L \cdot A}{V_f} \quad (5)$$

It is important to recognize that  $C_0$  will vary with time and waste surface temperature just as the solution rate does. Calculations which follow in this work will consider constant solution rates and thus constant concentration,  $C_0$ . However, recognition of the actual time and temperature variation is made.

Once radioisotopes have been dissolved from waste form, their subsequent environmental transport may be estimated, and analytically described. Equation (5) expresses the concentration of ionic radionuclide species in a dissolving solution as it relates to the solution

rate of a waste form. As the radionuclides are transported through the environment their concentrations are reduced by several interactions, of which only dilution and exponential decay are considered in this work.

Considering radionuclide dilution first, it is seen by equation (6) that the concentration of radioisotopes which reaches man,  $C$ , is expressed as the concentration at the waste form,  $C_0$ , divided by the dilution factor,  $D$ .

$$C = C_0/D \quad (6)$$

This dilution factor is the number of equivalent volumes of uncontaminated water that the radionuclide mixes with during environmental transport.

Concentrations of radionuclides are further reduced as the radioisotopes decay exponentially with time during environmental transport. Thus for the  $i$ th radioisotope, the concentration which reaches man,  $C_i$ , is expressed as in equation (7).

$$C_i = C_{0i} (e^{-\lambda_i \tau_i})/D \quad (7)$$

where  $C_{0i}$  = the concentration of  $i$  at the waste form

$\lambda_i$  = the decay constant of  $i$

$\tau_i$  = the time required for  $i$  to be transported to man

The transport time is found by dividing the distance which radionuclides must travel to reach man by the speed at which the radionuclides move through the environment. The radionuclide rate of travel

is somewhat less than the groundwater speed. The reduction of the speed is caused by the ion exchange of the radionuclides as they travel through some aquifer material.

Ion exchange is a phenomenon in which some dissolved species are held by ionic bonds at sites within a solid; the solid being aquifer materials for the case of radionuclide transport. There is a competition for the dissolved ionic radionuclide species as they are moved through the biosphere.<sup>14</sup> Thus, the dissolved species will be continually exchanged through environmental transport.<sup>15</sup>

The problem at hand is to relate the chemical behavior of the ionic radioisotope species to their transport. If  $M^{+m}$  and  $A^{+n}$  represent ions of radionuclides and naturally occurring exchangeable aquifer minerals respectively carrying charges  $+m$  and  $+n$ , and  $X$  is assumed to be a reactive chemical radical in aquifer material, the equation for the ion exchange reaction is as in equation (8).<sup>16</sup>



The equilibrium constant for this exchange,  $K$ , is defined by equation (9).

$$K = \frac{[MX_m]^n [A^{+n}]^m}{[M^{+m}]^n [AX_n]^m} \quad (9)$$

Since in reality, a very small fraction of the available exchange sites in the minerals is occupied,  $[A^{+n}]$  and  $[AX_n]$  may be assumed constant. This allows the definition of a useful distribution coefficient.

$$K_D = \frac{V}{W} \left[ K \frac{[AX_n]^m}{[A^{+n}]^m} \right]^{1/n} = \frac{V}{W} \frac{[MX_m]}{[M^{+m}]} \quad (10)$$

where  $V$  = the volume of the solution

$W$  = the weight of minerals

Consider, then, a species of radionuclides, initially localized to a waste form. The flow rate of the radionuclides,  $F_a$ , may be expressed as in equation (11).

$$F_a = \frac{F_w}{1 + \rho K_D} \quad (11)$$

where  $F_w$  = the flow rate of groundwater

$\rho$  = the ratio of weight of minerals to volume of water per unit volume of aquifer material

Since  $\rho$  is not known exactly, it is convenient to use a value of 1, the usual value being 4 or 5. Thus, by assuming in addition that the area of flow for the radionuclides and groundwater are equal, equation (11) reduces to equation (12).

$$V' = \frac{V}{1 + K_D} = \frac{V}{I} \quad (12)$$

where  $V'$  = the velocity of the radionuclides

$V$  = the velocity of the groundwater

$I$  = the ion exchange holdup factor

The transport time of the radionuclides,  $\tau$ , is then the distance,  $X$ , which radionuclides must travel to reach man divided by the velocity of the radionuclides in solution.

$$\tau = \frac{X}{V'} = \frac{XI}{V} \quad (13)$$

Thus, the concentration of each radionuclide transported to man may be calculated.

$$C = \frac{AL}{V_f D} e^{-\frac{\lambda(XI)}{V}} \quad (14)$$

Given equation (14), a specific waste form with its dissolution characteristics, and a maximum concentration of radionuclides which may reach man, the required isolation of radioactive wastes may be calculated. This required isolation would be expressed in terms of the distance from man,  $X$ , and depends on the velocity of water through the environment,  $V$ , the sorption characteristics of the environment,  $I$ , and the quantity of water which may flow through the environment,  $V_f \cdot D$ .

A note must be added about this environmental transport model. Many sophisticated computer codes modeling this behavior have been written and are in use. This simple model has been derived for ease in hand calculations. Assumptions have been made in this theory. One is in assuming bulk quantities such as single sorption characteristics of an environment. This may be true in a small increment, but certainly is not true on a large scale. The radionuclides have been modeled as moving only in one direction, while in fact they will tend to spread in a manner similar to material in a plume dispersion. The important factors in constructing this simple model are that hand calculations may be performed, and with proper selection of averaged properties, the results will be conservative.

## CHAPTER 4

### NUMERICAL EXAMPLES

In order to demonstrate the use and meaning of the analytic expressions previously presented, a series of sample calculations will be performed on a typical waste form. The calculations will include an analysis of radioisotope release by solution, and a sample analysis of radioisotope transport through the environment.

The type waste form which will be considered is borosilicate glass produced from PUREX waste. This section will quantitatively define the borosilicate glass and the PUREX waste which, after calcination, is mixed with the glass.

In addition, the borosilicate waste form fitted to the solution rate expression in Chapter 2 is studied in terms of its radioisotope release and subsequent environmental transport. For calculational purposes, it is assumed that the radionuclide content to be specified for the reference waste form will exist in the modeled form.

#### PUREX Waste Description

This analysis considers nuclear wastes generated by the LWR fuel cycle. While in the reactor, the LWR fuel will acquire a fissile fuel burnup of 2.85 TJ/kgU (33000 MWD/MTU) over a three year period.<sup>17</sup> The fuel considered will have been removed from the reactor 160 days prior to reprocessing.

The nuclear fuel is processed in the PUREX process. This PUREX process uses concentrated nitric acid in aqueous solutions and tributyl phosphate in kerosene to separate uranium and plutonium from the fission products in spent reactor fuel. The resulting liquid wastes are nitrates. The chemical content of these liquid wastes is described in Table II.<sup>18</sup>

The high-level PUREX wastes are highly radioactive and self-heating. The total fission product activity in the liquid waste is  $0.451 \text{ EBq/m}^3$  ( $1.22 \times 10^4 \text{ Ci/l}$ ). The total actinide activity, assuming 0.5% loss of uranium and plutonium in reprocessing, is  $4.28 \text{ PBq/m}^3$  ( $115.8 \text{ Ci/l}$ ). Thus the total radioisotope content is  $0.455 \text{ EBq/m}^3$  ( $1.24 \times 10^4 \text{ Ci/l}$ ).<sup>17</sup> The major radioisotope composition in the PUREX waste at 160 days out-of-reactor is listed in Appendix A. In addition, in Table III, the volumetric heat rate of PUREX waste as it varies with time is listed.<sup>19</sup>

The analysis next considers that the PUREX liquid waste is solidified by the calcination process. This process drives the waste nitrates to oxides, removing the nitrates and water as off-gas. The calcined product is then suspended in a glass matrix, specifically in a borosilicate glass matrix. This form is discussed in the next section.

Table II. Chemical Content of PUREX Process Liquid Wastes\*

Component	Concentration (kg/m <sup>3</sup> )
hydrogen	1.058
iron	2.910
nickel	0.265
chromium	0.529
nitrate	174.1
phosphate	2.380
uranium	12.70
plutonium	0.106
neptunium	1.270
americium	0.370
curium	0.106
Total fission products	76.19

\*based on a waste production rate of  $3.78 \times 10^{-4} \text{ m}^3/\text{kgU}$

Table III. Heat Generation Rate of PUREX Waste

Time Out of Reactor (YR)	Heat Generation Rate (W/m <sup>3</sup> )
160 days	5.04 E+04
1	2.73 E+04
5	5.05 E+03
10	2.91 E+03
20	2.06 E+03
30	1.60 E+03
40	1.25 E+03
50	9.80 E+02
60	7.70 E+02
70	6.10 E+02
80	4.80 E+02
90	3.80 E+02
100	3.00 E+02
120	1.90 E+02
140	1.30 E+02
160	8.54 E+01
180	5.95 E+01
200	4.29 E+01
220	3.25 E+01
240	2.58 E+01
260	2.14 E+01
280	1.84 E+01
290	1.74 E+01

Description of Reference Borosilicate Waste Glass

A borosilicate glass contains  $B_2O_3$  and  $SiO_2$  as its major constituents. The borosilicate glass considered in this analysis is Savannah River Plant's Mix #18. Its chemical composition is listed in Table IV.<sup>20</sup>

Table IV. Composition of Reference Borosilicate Glass

Compound	Wt% of Glass Frit
$SiO_2$	52.5
$Na_2O$	22.5
$B_2O_3$	10.0
$TiO_2$	10.0
$CaO$	5.0

Production requirements for borosilicate glass vary from  $1.4 \times 10^{-17} \text{ m}^3/\text{J}$  (1.2 1/1000 MWD) to  $5.8 \times 10^{-7} \text{ m}^3/\text{J}$  (5.0 1/1000 MWD)). This analysis will consider a production rate of  $2.3 \times 10^{-17} \text{ m}^3/\text{J}$  (2.0 1/(1000 MWD)). For this value, waste oxides are about 22 wt% of the borosilicate glass product.

The heat generation rate of the borosilicate product is calculated from the heat generation rate of the PUREX waste as shown

in Table III. The glass heat generation rate is calculated from equation (15).

$$q_b = q_p R_p / (BR_b) \quad (15)$$

where  $q_b$  = the heat generation rate of borosilicate glass ( $W/m^3$ )

$q_p$  = the heat generation rate of PUREX waste ( $W/m^3$ )

$R_p$  = the production rate of PUREX ( $m^3/kgU$ )

$B$  = the fissile fuel burnup of the fuel ( $J/kgU$ )

$R_b$  = the volume of borosilicate glass produced per energy generated ( $m^3/J$ )

In this analysis,  $R_p = 3.78 \times 10^{-4} m^3/kgU$ ,  $B = 2.85 TJ/kgU$  (33000 MWD/MTU), and  $R_b$  is  $2.315 \times 10^{-17} m^3/J$ . Hence,  $q_b$  is described in equation (16).

$$q_b = q_p \left( \frac{W}{m^3 \text{ PUREX waste}} \right) (5.30 \frac{m^3 \text{ PUREX waste}}{m^3 \text{ buro-glass}}) \quad (16)$$

Thus, to determine the heat generation rate of the borosilicate glass product, the data in Table III need be multiplied by 5.30, as shown above.

The reference borosilicate glass has several measured properties which help describe it. Solution rates of this glass under various conditions are shown in Table V.<sup>20,21,22</sup>

The constants for the dissolution equation calculated earlier are used with the dissolution equation to calculate dissolution rates for use in this section. Temperature conditions selected are 298K and 372K as the dissolution temperatures. The time for dissolution is arbitrarily selected to be one day. In this manner, dissolution rates are calculated and shown in Table V.

Table V. Solution Rates of Reference Borosilicate Glass

Component Leached	Leaching Condition	Solution ( $\mu\text{g}/\text{m}^2\text{s}$ )	Rate ( $\text{atoms}/\text{m}^2\text{s}$ )*
$^{137}\text{Cs}$	1	0.0138	--
$^{90}\text{Sr}$	1	0.0467	--
$^{125}\text{Sb}$	1	0.261	--
alpha actinides	1	0.0201	--
bulk glass	1	605.0	3.96 E+18
$^{137}\text{Cs}$	2	0.0190	--
$^{90}\text{Sr}$	2	0.832	--
$^{125}\text{Sb}$	2	0.230	--
alpha actinides	2	0.0104	--
bulk glass	2	535.0	3.50 E+18
$^{137}\text{Cs}$	3	0.394	--
$^{90}\text{Sr}$	3	0.749	--
$^{125}\text{Sb}$	3	1.01	--
alpha actinides	3	0.0435	--
bulk glass	3	2350.0	1.54 E+19
bulk glass	4	61.2	4.01 E+17
modeled glass	5	176.3	1.15 E+18
modeled glass	6	248.0	1.62 E+18

## Conditions:

- 1 - glass in  $\text{H}_2\text{O}$  at 298K (25°C)
- 2 - glass heated to 773K (500°C) for 1 month, in  $\text{H}_2\text{O}$  at 298K (25°C)
- 3 - glass heated to 873K (600°C) for 1 month (devitrified), in  $\text{H}_2\text{O}$  at 298K (25°C)
- 4 - glass in  $\text{H}_2\text{O}$  at 372K (99°C)
- 5 - glass in  $\text{H}_2\text{O}$  at 298K (25°C) surface temperature for  $\theta = 1$  day
- 6 - glass in  $\text{H}_2\text{O}$  at 372K (99°C) surface temperature for  $\theta = 1$  day

\*Based on a calcine molecular weight of 190g/mole and a waste glass molecular weight of 92g/mole.

A few additional properties of the reference waste glass are known. These are described below.

The density of any waste glass varies with waste oxide content. All data indicate that expected densities will range from 2900 to 3500 kg/m<sup>3</sup>.<sup>23,24,25,26</sup> This work calculates a density of 3000 kg/m<sup>3</sup> for the reference glass.

The thermal conductivity of borosilicate glass varies from 1.0 to 1.4 W/mK.<sup>23,24,25,26</sup>

The thermal stability of borosilicate glass is well known. Glass undergoes two steps to thermal instability, devitrification and mechanical instability.

Devitrification is a phase transformation in glass, in which the supercooled liquid structure of a glass crystallizes. Four main factors directly affect the devitrification of a glass: 1) time, 2) temperature, 3) nucleation, and 4) internal structure.

It has been observed that the rate of devitrification may be expressed in the arrhenius form as in equation (17).<sup>27</sup>

$$\text{Rate} = Ae^{-E/RT} \quad (17)$$

where A = constant

E = activation energy

R = gas constant

T = absolute temperature

Thus, devitrification is a kinetic process which will proceed to some degree at all temperatures. It may be expected that a nuclear waste glass will eventually completely devitrify, even at low temperatures.

The self-heating nature of nuclear wastes provide more advanced temperatures, and thus higher rates of devitrification. It has been observed that at temperatures over 870K, the devitrification of the reference glass proceeds rapidly.<sup>18,23,28</sup> While the effects of devitrification on a glass are varied, data in Table V demonstrate that the reference glass solution rate may increase with devitrification.

Mechanical instability of glass begins at about 973K,<sup>24</sup> as the glass takes on a molten behavior. This behavior becomes more severe with increasing temperature until at 1373K to 1473K, the glass behaves as a viscous liquid.<sup>21</sup>

Waste form geometry is of yet uncertain. This work assumes a canister geometry of 0.30m in diameter by 3.0m long.

#### Primary Radioisotope Release

The radioisotope release by dissolution is to be calculated for the reference borosilicate glass. To demonstrate the possible range of releases which may result from the waste form, fourteen distinct cases will be considered as shown below:

- a - vitreous product with leachant at 298K for cylindrical monolith and for monolith broken into cube particles 0.1 mm on a side, with waste age at one and ten years out-of-reactor;

- b - devitrified product with leachant at 298K for cylindrical monolith and cubic pieces with one and ten years out-of-reactor ages;
- c - vitreous product with leachant at 372K for cylindrical monolith and cubic pieces with and ten years out-of-reactor ages;
- d - the modeled glass in cylindrical monolith form at one day of leaching with the leachant at 298K and 372K.

The calculation of radioisotopic release by solution, utilizing the data of Table V and the previously described analytic expressions, requires some preliminary explanation. Table V lists specific leach rates of  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^{125}\text{Sb}$ , alpha actinides, and bulk glass. Those radioisotopes not specifically mentioned will be assumed to be released from the waste form by corrosion. Thus, the actual radioisotopic release is the release of the waste forms times the weight fraction which those radioisotopes make up of the matrix. These weight fractions are found in Appendices B and C.

The analytic expressions presented describing the concentrations of radioisotopes released from waste forms were of the general form shown below in equation (5).

$$C_o = \frac{m}{V_f} \quad (5)$$

where  $C_o$  = concentrations of radioisotopes  
 $m$  = mass flow rates of radioisotopes  
 $V_f$  = volumetric flow rate of leachant

To assure that calculations are not weighted by the estimation of a volumetric flow rate of a leachant, the mass flow rate of radioisotopes alone will be calculated in the units of (MPC-m<sup>3</sup>/s), from Appendix D.

Thus, the potential mass flow rates of radioisotopes from the reference waste glass are shown below in Table VI with specific releases shown in Appendices E, F, G, and H.

Calculations performed to arrive at values found in Table VII show some additional information of interest. Those radioisotopes which are of major interest at a waste age of ten years are revealed. These are tabulated below in Table VII.

#### Radioisotope Transport Through the Environment

This section will discuss the releases of radioisotopes previously calculated as they are moved through the environment. In particular, radioisotope transport in a river type environment and in a typical desert environment will be considered.

Equation (14) shows that concentrations of radioisotopes at some point in the environment is as shown below.

$$C = \frac{ALe^{-\frac{[IX]\lambda}{V}}}{V_f D} \quad (14)$$

For a river environment, one in which the decay of radioisotopes during the transport may be neglected, this equation may be reduced to the form of equation (18).

$$C = \frac{m}{V_0} \quad (18)$$

Table VI. Radioisotopic Releases from Waste Forms

Waste Geometry	Waste Age (Yr)	Leaching Condition	Mass Flow Rate <sub>3</sub> of Release (MPC-m <sup>3</sup> /s)
cylinder	1	1	9.73 E+2
cylinder	10	1	4.75 E+2
cubes	1	1	5.84 E+3
cubes	10	1	2.85 E+3
cylinder	1	2	2.50 E+5
cylinder	10	2	3.97 E+3
cubes	1	2	1.50 E+6
cubes	10	2	2.38 E+4
cylinder	1	3	6.58 E+3
cylinder	10	3	3.37 E+3
cubes	1	3	3.97 E+4
cubes	10	3	2.02 E+4
cylinder	10	4	7.26 E+3
cylinder	10	5	1.02 E+4

1 - vitreous waste, leachant at 298K

2 - devitrified waste, leachant at 298K

3 - vitreous waste, leachant at 372K

4 - modeled glass, leachant at 298K, one day of leaching

5 - modeled glass, leachant at 372K, one day of leaching

Table VII. Major Radioisotopes Released from Ten Year Old Waste Forms

Waste Geometry	Leaching Condition	Radioisotope	Mass Flow Rate (MPC-m <sup>3</sup> /s)
cylindrical	1	<sup>90</sup> Sr	236.3
		<sup>137</sup> Cs	0.6
		<sup>125</sup> Sb	0.4
		<sup>154</sup> Eu	0.3
cubic	1	<sup>90</sup> Sr	1417.8
		<sup>137</sup> Cs	3.5
		<sup>125</sup> Sb	2.2
		<sup>154</sup> Eu	1.9
cylindrical	2	<sup>90</sup> Sr	1699.0
		<sup>154</sup> Eu	259.4
		<sup>106</sup> Ru	88.3
		<sup>147</sup> Pm	62.1
		<sup>144</sup> Ce	23.0
cubic	2	<sup>137</sup> Cs	7.5
		<sup>90</sup> Sr	10194.0
		<sup>154</sup> Eu	1556.7
		<sup>106</sup> Ru	530.1
		<sup>147</sup> Pm	372.4
		<sup>144</sup> Ce	137.8
		<sup>137</sup> Cs	45.0

Table VII, Continued

Waste Geometry	Leaching Condition	Radioisotope	Mass Flow Rate (MPC-m <sup>3</sup> /s)
cylindrical	3	<sup>90</sup> Sr	1641.6
		<sup>137</sup> Cs	35.5
		<sup>134</sup> Cs	8.6
cubic	3	<sup>90</sup> Sr	9849.8
		<sup>137</sup> Cs	212.8
		<sup>134</sup> Cs	51.3
		<sup>106</sup> Ru	4.4
cylindrical	4	<sup>90</sup> Sr	6841.8
		<sup>137</sup> Cs	149.6
		<sup>154</sup> Eu	9.2
		<sup>134</sup> Cs	3.2
		<sup>106</sup> Ru	3.1
cylindrical	5	<sup>90</sup> Sr	9621.0
		<sup>137</sup> Cs	210.4
		<sup>154</sup> Eu	12.9
		<sup>134</sup> Cs	4.6
		<sup>106</sup> Ru	4.3

1 - vitreous waste, leachant at 298K

2 - devitrified waste, leachant at 298K

3 - vitreous waste, leachant at 372K

4 - modeled glass, leachant at 298K, 1 day of leaching

5 - modeled glass, leachant at 372K, 1 day of leaching

where  $m$  = mass flow rate of radioisotopes

$V_0$  = volumetric flow rate of body of water ( $V_f \cdot D$ )

The concentrations of radionuclides calculated by this equation are contingent on the perfect mixing of released radioisotopes with the volume of flow of water. Thus, using equation (18), and knowing the mass flow rate of radioisotopes from the waste form, the amount of water necessary to achieve proper dilution of radioisotopes may be determined.

Again recalling equation (14), to demonstrate the use of this equation, a sample calculation has been done. The mass flow rate of major radioisotopes from ten year old devitrified cubic particles is shown in Table VII. The calculation assumes that these radioisotopes are leached in  $10^{-4} \text{ m}^3/\text{s}$  of water, and are undiluted. The radioisotopes are then transported through a typical desert environment. The ion exchange holdup factors used for each of these radioisotopes are listed in Table VIII.<sup>29</sup> Finally, the concentrations of these radioisotopes are listed against the time which the groundwater travels in the environment in Table IX.

Values in Table IX and all other tables in this work are listed in System International or SI units. A listing of a few less common SI units is found in Appendix I.

Table VIII. Ion Exchange Holdup Factors for "Typical" Western U.S. Desert Soil

Element	$I^{-1}$	Element	$I^{-1}$
tritium	1	iodine	1
beryllium	$3 \times 10^{-3}$	cesium	$1 \times 10^{-3}$
carbon	$1 \times 10^{-1}$	cerium	$4 \times 10^{-4}$
sodium	$2 \times 10^{-2}$	promethium	$4 \times 10^{-4}$
chlorine	1	samarium	$4 \times 10^{-4}$
argon	1	europium	$4 \times 10^{-4}$
potassium	$6 \times 10^{-3}$	holmium	$4 \times 10^{-4}$
calcium	$1 \times 10^{-2}$	thallium	$1 \times 10^{-1}$
iron	$3 \times 10^{-4}$	lead	$6 \times 10^{-5}$
cobalt	$3 \times 10^{-3}$	bismuth	$2 \times 10^{-2}$
nickel	$3 \times 10^{-3}$	polonium	$9 \times 10^{-3}$
selenium	$1 \times 10^{-2}$	astatine	1
krypton	1	radon	1
rubidium	$2 \times 10^{-3}$	francium	$1 \times 10^{-3}$
strontium	$1 \times 10^{-2}$	radium	$2 \times 10^{-3}$
yttrium	$1 \times 10^{-4}$	actinium	$2 \times 10^{-4}$
zirconium	$1 \times 10^{-4}$	thorium	$2 \times 10^{-5}$
niobium	$1 \times 10^{-4}$	protactinium	$6 \times 10^{-5}$
molybdenum	$4 \times 10^{-2}$	uranium	$7 \times 10^{-5}$
technetium	1	neptunium	$1 \times 10^{-2}$
ruthenium	$3 \times 10^{-4}$	plutonium	$1 \times 10^{-4}$
palladium	$9 \times 10^{-4}$	americium	$1 \times 10^{-4}$
cadmium	$1 \times 10^{-4}$	curium	$3 \times 10^{-4}$
tin	$9 \times 10^{-4}$	berkelium	$3 \times 10^{-4}$
antimony	$1 \times 10^{-2}$		

Table IX. Concentrations of Radioisotopes Dissolved from Devitrified Reference Waste as They Are Transported through the Environment

$^{90}\text{Sr}$	$^{154}\text{Eu}$	Concentration (MPC)				$^{137}\text{Cs}$	Groundwater Transport Time (Yr)
		$^{106}\text{Ru}$	$^{147}\text{Pm}$	$^{144}\text{Ce}$			
1.02E8	1.56E7	5.30E6	3.72E6	1.38E6	4.50E5	0	
9.95E7	5.28E6	4.90E-4	3.67E3	3.17E-4	3.57E5	0.01	
9.71E7	1.79E6	--	3.55	--	2.83E5	0.02	
9.47E7	6.05E5	--	3.46E-3	--	2.25E5	0.03	
9.24E7	2.05E5	--	--	--	1.79E5	0.04	
9.02E7	6.94E4	--	--	--	1.42E5	0.05	
8.62E7	2.35E4	--	--	--	1.13E5	0.06	
8.58E7	7.95E3	--	--	--	8.92E4	0.07	
8.37E7	2.69E3	--	--	--	7.09E4	0.08	
8.17E7	9.12E2	--	--	--	5.63E4	0.09	
7.97E7	3.09E2	--	--	--	4.46E4	0.10	
6.23E7	6.11E-3	--	--	--	4.43E3	0.20	
4.87E7	--	--	--	--	4.39E2	0.30	
3.80E7	--	--	--	--	4.36E1	0.40	
2.97E7	--	--	--	--	4.33	0.50	
2.32E7	--	--	--	--	4.29E-1	0.60	
1.81E7	--	--	--	--	--	0.70	
1.39E7	--	--	--	--	--	0.80	
1.09E7	--	--	--	--	--	0.90	
8.66E6	--	--	--	--	--	1.00	
7.35E5	--	--	--	--	--	2.00	
6.23E4	--	--	--	--	--	3.00	
5.29E3	--	--	--	--	--	4.00	
4.49E2	--	--	--	--	--	5.00	
3.81E1	--	--	--	--	--	6.00	
3.23	--	--	--	--	--	7.00	
2.74E-1	--	--	--	--	--	8.00	

## CHAPTER 5

### DISCUSSION AND CONCLUSIONS

A study has been performed to develop the importance of the dissolution phenomenon to high-level nuclear waste management. Theory has been presented describing the mechanisms of radioisotope release from an arbitrary waste form.

Typical borosilicate glass waste forms made from PUREX-LWR waste have been considered. The mass flow of radioisotopes from these waste forms has been calculated. Finally, expressions have been developed showing the relation between the solution behavior of waste forms and the environmental transport of radioisotopes, with numerical examples displayed for the typical waste form placed in possible environments.

Several important observations may be made from the analysis. First, regarding specific radioisotope release, it is apparent that the majority of the radioisotopes, which may be potentially released are composed of a few specific fission products, not the actinides. The data in Table VII indicate that for ten year old waste forms,  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^{106}\text{Ru}$ ,  $^{144}\text{Ce}$ ,  $^{147}\text{Pm}$ , and  $^{154}\text{Eu}$  are the radioisotopes of major release. These radioisotopes have half-lives such that after several hundred years, they will have nearly decayed. On the other hand, on

this time scale the summation of all actinides released is a trivial fraction of the total radioisotope concentration released by this mechanism to the environment.

From the analysis comparing vitreous and devitrified products, it is concluded that devitrification is a detrimental phenomenon based on the higher solution rates of devitrified glass forms. Calculations showed that the initial mass flow rate of radioisotopes from a devitrified waste form is greater than  $10^6$  MPC-m<sup>3</sup>/s, while a vitreous waste form in the same condition is about 200 times less soluble.

The statement that a devitrified product is worse than a vitreous form must be made with a note of caution. This analysis is based on the dissolution of waste forms. In the case of the dropping of the waste form, it may be advantageous for the product to be devitrified. Some evidence exists that a devitrified product has a higher impact strength than the vitreous product. However, this work does not address this subject, rather it is noted as a possibility.

On the basis of calculation, it is concluded that it might be advantageous to have waste forms in small shapes rather than in large ones. Data presented that devitrification of a borosilicate glass could result in a 200-fold increase in the solution rate over a vitreous form. From a heat transfer point of view, lower temperatures might be expected with smaller waste forms thus the rate of devitrification would be slower. If the smaller waste form did not cause a surface area increase of greater than 200 times, then from a radioisotopic

release point of view, it would become advantageous to have such forms. Before such a decision could be made, an assessment of the overall environmental impact of such a decision would have to be done to ensure that the overall risk to man would not be increased.

Regarding the release of radioisotopes in some transportation accident scenario placing the waste form into some river type environment, correlations have been developed showing the type of environment flow rate necessary to ensure proper dilution of radioisotopes as a function of radioisotope release rate from waste forms. In terms of the reference borosilicate waste form, it is concluded that the best waste form to ship would be a vitreous form, of at least ten years of age. This statement is made on the basis that this form has the lowest release rate of radioisotope of those considered in the analysis.

Several simplifying assumptions were made in the course of the above mentioned calculations. Of primary importance, it was assumed, for the cylindrically shaped waste form, that the entire surface area of the form was exposed to some leaching environment. In actuality, it is likely that in any transportation type scenario, this condition will not exist. Rather more likely, several decades of reduction of the exposed surface area would exist. This fact alone would greatly reduce the quantities of water which would be necessary to ensure proper dilution of radioisotopes.

In regard to the release of radioisotopes in some disposal scenario, it was shown that relatively high flow rates of ground water carrying radioisotopes would be sufficient to ensure proper hold-up of

radioisotopes in a desert type environment. Calculations showed that for the quantities of  $^{90}\text{Sr}$  released from a ten year old devitrified waste form, if the ground water took about eight years to transport from the nuclear waste disposal site to man's environment, then concentrations of  $^{90}\text{Sr}$  reaching man would be at permissible levels. It was also shown that if the  $^{90}\text{Sr}$  was controlled through its environmental transport, then all other radioisotopes would be similarly controlled.

As previously discussed, simplifying assumptions were made en route to these conclusions. The exposed surface area simplification would force calculated values to be lower. In addition, this analysis assumed very little radioisotope dilution. In reality, it is anticipated that as some leachant moved through the environment, it would be greatly diluted. Thus, calculated values would again be lowered in reality.

On the basis of the theory and numerical examples, it is concluded that two factors dominate the concentrations of radioisotopes which may reach man from solidified HLW. The first is the temperature of the waste form. This is of importance because of the arrhenius relation describing the dissolution of a waste form. As the temperature for dissolution increases, the dissolution rate is seen to increase at an exponential rate.

The second factor of importance is the waste form isolation distance from man and the ion exchange holdup encountered in that distance allowing radioactive decay to reduce the isotope concentration. The initial concentration of radionuclides has very little effect on

the isolation distance required. As a sensitivity analysis, the volume flow rate of ground water used in the calculation of Table IX was increased by a factor of  $10^4$ . As this greatly reduced the initial concentration of the radioisotopes, it was believed that the isolation distance from man would also be greatly reduced. However, the decrease was found to be only a factor of ten. This is reasonable since the isolation distance is a function of the natural logarithm of the initial concentration, and the natural log of  $10^4$  is about 10.

The proper isolation of a waste form may then be described as both the temperature control of the waste form to ensure lower dissolution rates, and geometric isolation of the waste form in a carefully selected environment with favorable sorption characteristics. Thus, it is concluded that with the temporary storage of a solidified waste for several years to lower heat generation rates and with careful site selection for the waste repository, high-level nuclear waste disposal may be carried out ensuring minimum exposure of man to radionuclides.

APPENDIX A

LISTING OF MAJOR RADIOISOTOPE COMPOSITION  
IN PUREX WASTES

Isotope	Ci%	Wt%
FISSION PRODUCTS		
$^3\text{H}$	8.97E-3	1.54E-3
$^{129}\text{I}$	6.49E-7	6.39E-6
$^{131}\text{I}$	3.07E-5	4.25E-6
$^{89}\text{Sr}$	1.76	1.03E-1
$^{90}\text{Sr}$	1.74	2.10E+1
$^{90}\text{Y}$	1.74	5.50E-3
$^{91}\text{Y}$	3.63	2.55E-1
$^{95}\text{Zr}$	6.36	5.13E-1
$^{95\text{m}}\text{Nb}$	1.35E-1	6.28E-4
$^{95}\text{Nb}$	1.19E+1	5.21E-1
$^{103}\text{Ru}$	2.25	1.21E-1
$^{103\text{m}}\text{Rh}$	2.24	1.19E-4
$^{106}\text{Ru}$	1.06E+1	5.35
$^{106}\text{Rh}$	1.06E+1	5.10E-6
$^{110\text{m}}\text{Ag}$	5.91E-2	2.12E-2
$^{119\text{m}}\text{Sn}$	6.29E-4	2.45E-2
$^{123\text{m}}\text{Sn}$	4.57E-2	1.00E-2

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Isotope	Ci%	Wt%
$^{125}\text{Sb}$	1.52E-1	2.45E-1
$^{125\text{m}}\text{Te}$	4.78E-2	4.54E-3
$^{127\text{m}}\text{Te}$	7.61E-2	1.33E-2
$^{127}\text{Te}$	7.49E-2	4.84E-5
$^{129\text{m}}\text{Te}$	1.13E-1	6.28E-3
$^{129}\text{Te}$	1.13E-1	1.08E-5
$^{134}\text{Cs}$	4.22	5.68
$^{136}\text{Cs}$	7.13E-4	1.65E-5
$^{137}\text{Cs}$	2.45	4.82E+1
$^{137\text{m}}\text{Ba}$	2.26	7.31E-6
$^{140}\text{Ba}$	7.22E-3	1.69E-4
$^{140}\text{La}$	8.30E-3	2.55E-5
$^{141}\text{Ce}$	1.49	8.94E-2
$^{144}\text{Ce}$	1.62E+1	8.70
$^{143}\text{Pr}$	1.27E-2	3.26E-4
$^{144}\text{Pr}$	1.62E+1	3.67E-4
$^{147}\text{Nd}$	1.02E-3	2.17E-5
$^{147}\text{Pm}$	2.94	5.56
$^{148}\text{Pm}$	4.71E-3	4.91E-5
$^{151}\text{Sm}$	7.57E-3	4.91E-1
$^{154}\text{Eu}$	1.80E-1	2.12
$^{155}\text{Eu}$	7.75E-2	9.75E-2
$^{160}\text{Tb}$	3.11E-3	4.76E-4

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Isotope	Ci%	Wt%
ACTINIDES		
$^{235}\text{U}$	4.00E-8	2.47E-1
$^{238}\text{U}$	7.70E-7	3.06E+1
$^{238}\text{Np}$	2.43E-3	1.23E-7
$^{238}\text{Pu}$	1.69	1.28
$^{239}\text{Pu}$	1.63E-1	3.52E+1
$^{240}\text{Pu}$	3.06E-1	1.78E+1
$^{241}\text{Pu}$	7.70E+1	9.10
$^{242}\text{Pu}$	1.46E-3	4.94
$^{241}\text{Am}$	1.31E-1	5.34E-1
$^{242}\text{Am}$	2.43E-3	3.97E-8
$^{242}\text{Cm}$	1.91E+1	7.66E-2
$^{243}\text{Cm}$	1.17E-2	3.35E-3
$^{244}\text{Cm}$	2.25	2.56E-1

## APPENDIX B

### WEIGHT FRACTIONS OF MAJOR FISSION PRODUCT RADIOISOTOPES

The absolute solution in Borosilicate Glass rate of each isotope from borosilicate glass, unless otherwise determined, is the product of the bulk waste solid leach rate times the fractional mass which that isotope constitutes of the waste.

In this analysis, the waste oxides constitute about 22 wt% of the total glass mass. In the waste oxides, the actual waste isotopes are about 80 wt% of the total while the oxide is about 20 wt%. This means that the actual waste radioisotope content is 17.6 wt%.

Appendix A presents the weight fraction of major fission product isotopes in wastes. The product of the weight fraction of each isotope of all radioisotopes times the 0.176 mentioned above is the weight fraction of each isotope in glassified wastes. These fractions are presented in Table B-1.

Table B-1. Weight Fractions of Major Fission Product Radioisotopes in Borosilicate Glass

Isotope	Weight Fraction	Isotope	Weight Fraction
$^{89}\text{Sr}$	1.81E-4	$^{129\text{m}}\text{Te}$	1.11E-5
$^{90}\text{Sr}$	3.70E-2	$^{129}\text{Te}$	1.90E-8
$^{90}\text{Y}$	9.68E-6	$^{134}\text{Cs}$	1.00E-2
$^{91}\text{Y}$	4.49E-4	$^{136}\text{Cs}$	2.90E-8
$^{91\text{m}}\text{Nb}$	1.11E-6	$^{137}\text{Cs}$	8.50E-2
$^{95}\text{Nb}$	9.17E-4	$^{137\text{m}}\text{Ba}$	1.29E-8
$^{103}\text{Ru}$	2.13E-4	$^{140}\text{Ba}$	2.97E-7
$^{103\text{m}}\text{Rh}$	2.09E-7	$^{140}\text{La}$	4.49E-8
$^{106}\text{Ru}$	9.42E-3	$^{141}\text{Ce}$	1.57E-4
$^{106}\text{Rh}$	8.98E-9	$^{144}\text{Ce}$	1.50E-2
$^{110\text{m}}\text{Ag}$	3.73E-5	$^{144}\text{Pr}$	6.46E-7
$^{119\text{m}}\text{Sn}$	4.31E-5	$^{147}\text{Nd}$	3.82E-8
$^{123\text{m}}\text{Sn}$	1.76E-5	$^{147}\text{Pm}$	9.79E-3
$^{125}\text{Sb}$	4.31E-4	$^{148}\text{Pm}$	8.64E-6
$^{125\text{m}}\text{Te}$	7.99E-6	$^{151}\text{Sm}$	8.64E-4
$^{127\text{m}}\text{Te}$	2.34E-5	$^{154}\text{Eu}$	3.73E-3
$^{127}\text{Te}$	8.52E-8	$^{155}\text{Eu}$	1.72E-4
		$^{160}\text{Tb}$	8.38E-7

## APPENDIX C

### WEIGHT FRACTION OF ACTINIDES IN BOROSILICATE GLASS

Table II shows the ratio of actinides to fission products in PUREX liquid waste to be 0.190. In Appendix B, it is stated that the fission product isotopes constitute 17.6 wt% of the borosilicate glass solid. This means actinides are  $(0.176) \times (0.190)$  or 3.36 wt% of the borosilicate wastes. If this figure is multiplied by the values of weight each actinide constitutes of all actinides, the weight fraction of the borosilicate waste each actinide represents is determined. These figures are shown in Table C-1.

Table C-1. Weight Fractions of Actinides in Borosilicate Glass

Isotope	Weight Fraction	Isotope	Weight Fraction
$^{235}\text{U}$	8.30E-5	$^{241}\text{Pu}$	3.06E-3
$^{238}\text{U}$	1.03E-2	$^{242}\text{Pu}$	1.66E-3
$^{238}\text{Np}$	4.13E-11	$^{241}\text{Am}$	1.80E-4
$^{238}\text{Pu}$	4.30E-4	$^{242}\text{Am}$	1.33E-11
$^{239}\text{Pu}$	1.18E-2	$^{242}\text{Cm}$	2.57E-5
$^{240}\text{Pu}$	5.98E-3	$^{243}\text{Cm}$	1.13E-6
		$^{244}\text{Cm}$	8.61E-5

APPENDIX D

TABULATION OF MAXIMUM PERMISSIBLE CONCENTRATIONS  
(MPC) OF VARIOUS ISOTOPES IN WATER<sup>30</sup>

Isotope	MPC (Bq/m <sup>3</sup> )	Isotope	MPC (Bq/m <sup>3</sup> )
<sup>3</sup> H	1.81E8	<sup>110m</sup> Ag	1.11E6
<sup>85</sup> Kr	N/A	<sup>119m</sup> Sn	1.11E6
<sup>131m</sup> Xe	N/A	<sup>123m</sup> Sn	1.11E6
<sup>129</sup> I	2.22E3	<sup>125</sup> Sb	3.70E6
<sup>131</sup> I	1.11E4	<sup>125m</sup> Te	7.40E6
<sup>89</sup> Sr	1.11E5	<sup>127m</sup> Te	2.22E6
<sup>90</sup> Sr	1.11E4	<sup>127</sup> Te	1.11E7
<sup>90</sup> Y	7.40E5	<sup>129m</sup> Te	1.11E6
<sup>91</sup> Y	1.11E6	<sup>129</sup> Te	2.96E7
<sup>95</sup> Zr	2.22E6	<sup>134</sup> Cs	3.70E6
<sup>95m</sup> Nb	3.70E6	<sup>136</sup> Cs	3.33E6
<sup>95</sup> Nb	3.70E6	<sup>137</sup> Cs	7.70E5
<sup>103</sup> Ru	2.96E6	<sup>137m</sup> Ba	7.40E5
<sup>103m</sup> Rh	3.70E8	<sup>140</sup> Ba	1.11E6
<sup>106</sup> Ru	3.70E5	<sup>140</sup> La	7.40E5
<sup>106</sup> Rh	3.70E5	<sup>141</sup> Ce	3.33E6
<sup>144</sup> Ce	3.70E5	<sup>238</sup> U	1.48E7

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Isotope	MPC (Bq/m <sup>3</sup> )	Isotope	MPC (Bq/m <sup>3</sup> )
<sup>143</sup> Pr	1.85E6	<sup>238</sup> Np	1.11E3
<sup>144</sup> Pr	3.70E5	<sup>238</sup> Pu	1.85E5
<sup>147</sup> Nd	2.22E6	<sup>239</sup> Pu	1.85E5
<sup>147</sup> Pm	7.40E6	<sup>240</sup> Pu	1.85E5
<sup>148</sup> Pm	1.85E6	<sup>241</sup> Pu	7.40E6
<sup>151</sup> Sm	1.48E7	<sup>242</sup> Pu	1.85E5
<sup>154</sup> Eu	7.40E5	<sup>241</sup> Am	1.48E5
<sup>155</sup> Eu	7.70E6	<sup>242</sup> Am	1.48E5
<sup>160</sup> Tb	1.48E6	<sup>242</sup> Cm	7.40E5
<sup>235</sup> U	1.11E7	<sup>243</sup> Cm	1.85E5
		<sup>244</sup> Cm	2.59E5

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APPENDIX E

RADIOISOTOPIC RELEASES FROM WASTE FORMS VITREOUS  
FORM, LEACHANT AT 298K

Isotope	Mass Flow Rate from Waste Form (MPC-m <sup>3</sup> /s)			
	Cylinder 1 Year	Cylinder 10 Years	Cubes 1 Year	Cubes 10 Years
<sup>89</sup> Sr	28.9	--	173.3	--
<sup>90</sup> Sr	294.7	236.3	1767.9	1417.8
<sup>90</sup> Y	294.7	236.3	1767.9	1417.8
<sup>91</sup> Y	61.9	--	371.4	--
<sup>95</sup> Zr	5.4	--	32.2	--
<sup>95m</sup> Nb	0.1	--	0.4	--
<sup>95</sup> Nb	6.1	--	36.6	--
<sup>103</sup> Ru	1.4	--	8.6	--
<sup>103m</sup> Rh	0.01	--	0.1	--
<sup>106</sup> Ru	53.9	0.1	323.2	0.6
<sup>106</sup> Rh	53.9	0.1	323.2	0.6
<sup>110m</sup> Ag	0.1	--	0.6	--
<sup>119m</sup> Sn	0.1	--	0.6	--
<sup>123m</sup> Sn	0.1	--	0.5	--
<sup>125</sup> Sb	3.7	0.4	22.3	2.2

Isotope	Cylinder 1 Year	Cylinder 10 Years	Cubes 1 Year	Cubes 10 Years
$^{125m}\text{Te}$	0.1	--	0.7	--
$^{127m}\text{Te}$	0.1	--	0.4	--
$^{127}\text{Te}$	0.01	--	0.1	--
$^{129m}\text{Te}$	0.02	--	0.1	--
$^{129}\text{Te}$	0.01	--	0.1	--
$^{134}\text{Cs}$	2.6	0.1	16.5	0.8
$^{136}\text{Cs}$	--	--	--	--
$^{137}\text{Cs}$	0.7	0.6	4.3	3.5
$^{137m}\text{Ba}$	0.7	0.6	4.3	3.5
$^{140}\text{Ba}$	0.01	--	0.1	--
$^{140}\text{La}$	0.02	--	0.1	--
$^{141}\text{Ce}$	0.8	--	5.1	--
$^{144}\text{Ce}$	80.7	0.03	484.5	0.2
$^{143}\text{Pr}$	0.01	--	0.1	--
$^{144}\text{Pr}$	80.7	0.03	484.5	0.2
$^{147}\text{Nd}$	--	--	--	--
$^{147}\text{Pm}$	0.7	0.07	4.5	0.4
$^{148}\text{Pm}$	0.5	--	2.9	--
$^{151}\text{Sm}$	--	--	--	--
$^{154}\text{Eu}$	0.5	0.3	2.7	1.9
$^{155}\text{Eu}$	0.02	--	0.1	--
$^{160}\text{Tb}$	--	--	0.02	--

Isotope	Cylinder 1 Year	Cylinder 10 Years	Cubes 1 Year	Cubes 10 Years
$^{235}\text{U}$	--	--	--	--
$^{238}\text{U}$	--	--	--	--
$^{238}\text{Np}$	--	--	0.02	--
$^{238}\text{Pu}$	0.01	0.01	0.07	0.07
$^{239}\text{Pu}$	1.2E-3	1.2E-3	7.0E-3	7.0E-3
$^{240}\text{Pu}$	2.2E-3	2.2E-3	0.01	0.01
$^{241}\text{Pu}$	0.01	8.6E-3	0.08	0.05
$^{242}\text{Pu}$	1.0E-5	1.0E-5	6.3E-5	6.3E-3
$^{241}\text{Am}$	1.2E-3	1.2E-3	7.0E-3	6.9E-3
$^{242}\text{Am}$	2.2E-5	--	1.3E-4	--
$^{242}\text{Cm}$	0.03	--	0.2	--
$^{243}\text{Cm}$	8.3E-5	6.8E-5	5.0E-4	4.1E-4
$^{244}\text{Cm}$	8.2E-3	5.8E-3	0.05	0.03

APPENDIX F

RADIOISOTOPIC RELEASES FROM WASTE FORMS DEVITRIFIED  
FORM, LEACHANT AT 298K

Isotope	Mass Flow Rate from Waste Form (MPC-m <sup>3</sup> /s)			
	Cylinder 1 Year	Cylinder 10 Years	Cubes 1 Year	Cubes 10 Years
<sup>89</sup> Sr	207.2	--	1243.3	--
<sup>90</sup> Sr	2118.1	1699.0	12710	10194
<sup>90</sup> Y	2118.1	1699.0	12710	10194
<sup>91</sup> Y	5183.2	--	31099	--
<sup>95</sup> Zr	4500.2	--	27000	--
<sup>95m</sup> Nb	58.0	--	347.8	--
<sup>95</sup> Nb	5120.1	--	30720	--
<sup>103</sup> Ru	1193.9	--	7163.5	--
<sup>103m</sup> Rh	1193.9	--	7163.5	--
<sup>106</sup> Ru	45230	88.3	271400	530.1
<sup>106</sup> Rh	45230	88.3	271400	530.1
<sup>110m</sup> Ag	83.8	0.01	502.8	0.05
<sup>119m</sup> Sn	89.0	0.01	533.8	0.06
<sup>123m</sup> Sn	64.9	--	389.2	--
<sup>125</sup> Sb	64.3	6.4	385.7	38.2
<sup>125m</sup> Te	10.2	--	61.0	--
<sup>127m</sup> Te	54.1	--	324.8	--

Isotope	Cylinder 1 Year	Cylinder 10 Years	Cubes 1 Year	Cubes 10 Years
$^{127}\text{Te}$	10.7	--	64.1	--
$^{127\text{m}}\text{Te}$	160.7	--	964.3	--
$^{129}\text{Te}$	7.3	--	44.1	--
$^{134}\text{Cs}$	35.3	1.8	211.8	10.9
$^{136}\text{Cs}$	--	--	--	--
$^{137}\text{Cs}$	9.2	7.5	55.4	45.0
$^{137\text{m}}\text{Ba}$	9.2	7.5	55.4	45.0
$^{140}\text{Ba}$	10.2	--	61.3	--
$^{140}\text{La}$	6.3	--	37.5	--
$^{141}\text{Ce}$	740.5	--	4442.8	--
$^{144}\text{Ce}$	67730	23.0	406400	137.8
$^{143}\text{Pr}$	10.8	--	65.1	--
$^{144}\text{Pr}$	67730	23.0	606400	137.8
$^{147}\text{Nd}$	0.7	--	4.3	--
$^{147}\text{Pm}$	625.7	62.1	3754.0	382.4
$^{148}\text{Pm}$	398.4	--	2390.1	--
$^{151}\text{Sm}$	0.8	0.8	4.8	4.5
$^{154}\text{Eu}$	383.4	259.4	2300.6	1556.7
$^{155}\text{Eu}$	16.6	0.4	99.5	2.5
$^{160}\text{Tb}$	3.3	--	20.0	--
$^{235}\text{U}$	--	--	--	--
$^{238}\text{U}$	--	--	--	--

Isotope	Cylinder 1 Year	Cylinder 10 Years	Cubes 1 Year	Cubes 10 Years
<sup>238</sup> Np	9.0E-4	--	5.4E-3	--
<sup>238</sup> Pu	3.7E-3	3.5E-3	0.02	0.02
<sup>239</sup> Pu	3.6E-4	3.6E-4	2.2E-3	2.2E-3
<sup>240</sup> Pu	6.8E-4	6.8E-4	4.1E-3	4.1E-3
<sup>241</sup> Pu	4.2E-3	2.6E-3	0.03	0.02
<sup>242</sup> Pu	3.2E-6	3.2E-6	1.9E-5	1.9E-5
<sup>241</sup> Am	3.6E-4	3.6E-4	2.2E-3	2.2E-3
<sup>242</sup> Am	6.7E-6	--	4.0E-5	--
<sup>242</sup> Cm	0.01	--	0.06	--
<sup>243</sup> Cm	2.6E-5	2.1E-5	1.5E-4	1.3E-4
<sup>244</sup> Cm	2.5E-3	1.8E-3	0.02	0.01

APPENDIX G

RADIOISOTOPE RELEASE FROM WASTE FORMS VITREOUS  
FORM, LEACHANT AT 372K

Isotope	Mass Flow Rate from Waste Form (MPC-m <sup>3</sup> /s)			
	Cylinder 1 Year	Cylinder 10 Years	Cubes 1 Year	Cubes 10 Years
<sup>89</sup> Sr	199.8	--	1198.5	--
<sup>90</sup> Sr	2049.2	1641.6	12295	9849.8
<sup>90</sup> Y	2049.2	1641.6	12295	9849.8
<sup>91</sup> Y	43.2	--	259.0	--
<sup>95</sup> Zr	37.5	--	224.9	--
<sup>95m</sup> Nb	0.5	--	2.9	--
<sup>95</sup> Nb	42.5	--	254.9	--
<sup>103</sup> Ru	10.0	--	59.9	--
<sup>103m</sup> Rh	0.08	--	0.5	--
<sup>106</sup> Ru	374.8	0.7	2248.9	4.4
<sup>106</sup> Rh	374.8	0.7	2248.9	4.4
<sup>110m</sup> Ag	0.7	--	4.2	--
<sup>119m</sup> Sn	0.7	--	4.5	--
<sup>123m</sup> Sn	0.5	--	3.2	--
<sup>125</sup> Sb	0.5	0.05	3.2	0.3

Isotope	Cylinder 1 Year	Cylinder 10 Years	Cubes 1 Year	Cubes 10 Years
$^{125m}\text{Te}$	0.8	--	5.1	--
$^{127m}\text{Te}$	0.4	--	2.7	--
$^{127}\text{Te}$	0.09	--	0.5	--
$^{129m}\text{Te}$	1.3	--	8.0	--
$^{129}\text{Te}$	0.06	--	0.4	--
$^{134}\text{Cs}$	166.5	8.6	998.8	51.3
$^{136}\text{Cs}$	--	--	0.02	--
$^{137}\text{Cs}$	43.7	35.5	262.1	212.8
$^{137m}\text{Ba}$	43.7	35.5	262.1	212.8
$^{140}\text{Ba}$	0.08	--	0.5	--
$^{140}\text{La}$	0.1	--	0.9	--
$^{141}\text{Ce}$	5.9	--	35.1	--
$^{144}\text{Ce}$	561.9	0.2	3371.7	1.1
$^{143}\text{Pr}$	0.09	--	0.5	--
$^{144}\text{Pr}$	561.9	0.2	3371.7	1.1
$^{147}\text{Nd}$	--	--	0.04	--
$^{147}\text{Pm}$	5.2	0.5	31.3	3.1
$^{148}\text{Pm}$	3.3	--	19.9	--
$^{151}\text{Sm}$	--	--	0.04	0.04
$^{154}\text{Eu}$	0.3	0.2	1.9	1.3
$^{155}\text{Eu}$	0.1	--	0.8	0.02
$^{160}\text{Tb}$	0.3	--	1.7	--

Isotope	Cylinder 1 Year	Cylinder 10 Years	Cubes 1 Year	Cubes 10 Years
$^{235}\text{U}$	--	--	--	--
$^{238}\text{U}$	--	--	--	--
$^{238}\text{Np}$	0.04	--	0.3	--
$^{238}\text{Pu}$	0.2	0.2	1.1	1.0
$^{239}\text{Pu}$	0.02	0.02	0.1	0.1
$^{240}\text{Pu}$	0.03	0.03	0.2	0.2
$^{241}\text{Pu}$	0.2	0.1	1.2	0.8
$^{242}\text{Pu}$	1.5E-4	1.5E-4	9.2E-4	9.2E-4
$^{241}\text{Am}$	0.02	0.02	0.1	0.1
$^{242}\text{Am}$	3.2E-4	--	1.9E-3	--
$^{242}\text{Cm}$	0.5	--	3.0	--
$^{243}\text{Cm}$	1.2E-3	1.0E-3	7.3E-3	6.0E-3
$^{244}\text{Cm}$	0.1	0.08	0.7	0.5

APPENDIX H

RADIOISOTOPIC RELEASES FROM WASTE FORMS, MODELED FORM  
AT ONE DAY OF LEACHING, WASTE AGE -- 10 YEARS

Isotope	Mass Flow Rate from Waste From ( $\text{MPC}\cdot\text{m}^3/\text{s}$ )	
	Cylinder -- $\text{H}_2\text{O}$ at 298K	Cylinder -- $\text{H}_2\text{O}$ at 372K
$^{90}\text{Sr}$	6841.8	9621.0
$^{90}\text{Y}$	105.4	148.1
$^{106}\text{Ru}$	3.09	4.35
$^{106}\text{Rh}$	0.0117	0.0165
$^{125}\text{Sb}$	0.266	0.318
$^{134}\text{Cs}$	3.25	4.6
$^{137}\text{Cs}$	149.6	210.4
$^{137\text{m}}\text{Ba}$	137.9	194.0
$^{147}\text{Pm}$	2.66	3.74
$^{151}\text{Sm}$	0.0265	0.0372
$^{154}\text{Eu}$	9.2	12.9
$^{155}\text{Eu}$	0.0172	0.0242
$^{235}\text{U}$	$2.95\text{E}-10$	$4.15\text{E}-10$
$^{238}\text{U}$	$4.28\text{E}-9$	$6.03\text{E}-9$
$^{238}\text{Pu}$	0.676	0.949
$^{239}\text{Pu}$	0.0723	0.102
$^{240}\text{Pu}$	0.131	0.185

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Isotope	Cylinder -- H <sub>2</sub> O at 298K	Cylinder -- H <sub>2</sub> O at 372K
<sup>241</sup> Pu	0.539	0.756
<sup>242</sup> Pu	6.46E-4	9.10E-4
<sup>241</sup> Am	0.0726	0.102
<sup>242</sup> Am	1.61E-8	2.34E-8
<sup>243</sup> Cm	4.28E-3	6.03E-3
<sup>244</sup> Cm	0.351	0.494

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## APPENDIX I

### NUMERICAL UNITS

Calculations performed in this work are carried out in the Systems International or SI units. This section lists some of these units with the conventional engineering counterparts. In addition, a multiplying factor to convert the SI unit to the conventional unit is included.

SI Unit	Conventional Unit	Multiplying Factor
$\mu\text{g}/(\text{m}^2\text{s})$	$\text{g}/(\text{cm}^2\text{day})$	8.640E-6
W/mK	Btu/fthr $^{\circ}\text{F}$	1.872
$\text{kg}/\text{m}^3$	$\text{g}/\text{cm}^3$	1.0E-3
J/kgK	Btu/lb $_m$ $^{\circ}\text{F}$	7.755E-4
TJ/kgU	MWD/MTU	2.778E5
EBq/m $^3$	Ci/l	2.703E4
PBq/m $^3$	Ci/l	2.703E1

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