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Possible Weathering (Dissolution) of Basalt Dome-Filling Material in 241-TY Tank Farm Waste

R. C. Routson
Health, Safety and Environment

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Possible Weathering (Dissolution) of Basalt Dome-Filling Material in 241-TY Tank Farm Waste

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POSSIBLE WEATHERING (DISSOLUTION) OF BASALT DOME-FILLING MATERIAL IN 241-TY TANK FARM WASTE

R. C. Routson
Health, Safety and Environment

March 1983

Prepared for the United States
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EXECUTIVE SUMMARY

Assumed dome collapse must be mitigated if in place disposal of radioactive waste in single-shell tanks is to be a viable, environmentally sound disposal alternative. It has been proposed to fill the domes above the waste with crushed basalt to prevent dome collapse and the possible release of ejected radionuclides. Dissolution of crushed basalt in the tanks' interstitial liquid was foreseen as a possible dome filling problem, resulting in a decrease of dome filling material and an increase of sludge-like clays.

Background issues were evaluated. It was found that basalt would likely settle slowly through the sludge to the bottom of the tanks, but this would present no problem. Based on the solubility data, it was recommended that the minimum size of the basalt fragments be 1.0 cm in diameter.

Maximum basalt solubilities in high-level tank interstitial solutions were estimated from basalt weathering in a surface environment, basalt weathering in a natural watershed, and available thermodynamic data. Maximum expected solubility was estimated to be restricted to a thin weathering rind around any basalt fragment. Thus, no significant solubility effect can be expected as a result of dome filling. A solubility study is proposed to confirm these estimates.

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INTRODUCTION

Basalt is the major rock type available in the Pasco Basin for filling high-level waste tanks. Dome filling would limit the effects of possible dome collapse, the only credible tank-accident scenario that could result in a significant dose to the public (Murthy et al., 1982). To mitigate the possible effects of dome collapse, it is proposed to fill the 241-TY (TY) tanks with rock. Since the tanks contain interstitial liquid, dissolution of the basalt filling material, over the 500 yr the disposal system must endure, may pose a radionuclide release problem. The purpose of this evaluation is to determine if weathering of basalt in the TY Tank Farm interstitial solution, in 500 yr at the maximum temperatures of 170° to 180°C, is likely to compromise the integrity of the proposed in situ disposal system. Research is suggested to verify this evaluation.

BACKGROUND CONSIDERATIONS

BASALT PROPERTIES

Basalt is formed in a basic, geologic environment. The chemical environment of the TY Tank Farm is likewise basic (Delegard and Barney, 1982). In contrast, granite, the other end-member of geologic rock types, is acidic (Grout, 1959) and is formed in an acidic geologic environment. Thus, intuitively, one would expect basalt to be more stable (less soluble) than granite in a high-level waste tank chemical environment. Furthermore, basalt is formed at temperatures and pressures nearer to those of surface temperature and pressure than deep-seated granites and can therefore be expected to be more stable.

Other considerations, however, may suggest that granite tends to be more stable than basalt. Basalt and granite compositions differ chemically (Grout, 1959). In addition, basalt has cooled faster than granite. Basalt is therefore a finer-grained rock than granite. Small particles are more soluble than larger particles of the same composition (Routson, 1970). More importantly, basalt has cooled so rapidly that a major portion (up to half) of basalt is composed of a metastable glass (Nubergall et al., 1968). Glasses can be considered either as an amorphous solid, or as a supercooled, viscous liquid (Nubergall et al., 1968). Glasses have no melting points and thus are amorphous (they also have no x-ray diffraction patterns). Like a liquid, glass assumes the shape of the container it is enclosed in (albeit very slowly). Glass windows in old European buildings are thicker at the bottom than at the top due to this viscous flow. The solubility of glass in water may be expected to be relatively high.

Among these considerations, the environmental and chemical conditions probably outweigh the others. Basic basalt is probably more stable than acidic granite in the TY Tank Farm's interstitial solution. Furthermore, all other rock types are probably of intermediate acidic-basic character and of intermediate stability (Grout, 1959).

All igneous rock types are formed at higher temperatures and pressures than those of the earth's surface. Thus, they presently exist in a different environment than the one in which they were formed. In this new environment, the minerals in the rock tend to convert spontaneously to other, more stable minerals. The minerals in the rock can convert to stable minerals by their components dissolving into solution, or by solid phase conversions. Both conversion types may occur in the chemical environment of the TY Tank Farm. However, the solid phase conversion is generally so slow that in a time frame of 500 yr, it would likely be quantatively insignificant. Thus, only solution-reprecipitation conversions will be considered in this analysis.

BASALT-SLUDGE INTERACTIONS

In the proposed in situ disposal system, basalt fragments would usually be placed upon a sludge, which may be underlain by a salt cake.* In the five tanks containing only sludge, the basalt may be expected to settle slowly to the bottom of the tank. The sludge is essentially a stiff-slurry of clay or near clay-sized hydrous oxides, primarily of iron and aluminum. It has a bulk density of approximately 1.6 g/cm^3. In contrast, basalt has a density of 2.9 (2.4 to 3.1) g/cm^3 (BWIP Staff, 1978). With the density difference, basalt will settle probably slowly to the bottom of the tank. The sludge will tend to fill the interstitial space between the basalt fragments. The upper surface of the sludge will rise; this rise will be a function of how much pore space is present.

At equilibrium (when fragments settle to the bottom of the tanks), single-sized spherodial basalt fragments will tend to close pack. Close packing will result in the minimal pore fraction that can be obtained in this system, 0.26 (Moore, 1965). Irregularly shaped fragments, failure to obtain equilibrium, and a range in fragment sizes will all tend to increase the pore fraction. Based on a pore fraction of 0.26, Table 1 lists the maximum height to which sludge can be expected to rise in the TY Tank Farm tanks. Basalt will more reasonably settle to a porosity of 0.30 to 0.35.

It is difficult to determine how long it will take for basalt fragments to reach or approach equilibrium; the question is best treated empirically. In any event, the time needed is probably a complex function of water content of the sludge. Equilibrium time may be long, relative to the time required to fill the tank with basalt.

Thus, the basalt fill may settle considerably after dome stabilization if equilibrium time is not considered. If that occurred, the tank would still have the unfilled space in its dome. To compensate for this, the tank could be initially, partially filled with basalt, time be allowed to lapse, then the tank refilled.

---

*Salt cake is a solid. Only one of the TY Tank Farm tanks, 241-TY-102, contains a significant amount of salt cake (McCann, 1982), and this tank contains no sludge. Determining whether salt cake will support a column of basalt is a structural, engineering problem to be treated elsewhere.

TABLE 1. Maximum Depth to Which 241-TY Tank Farm Sludge Will Rise if Tanks Are Filled With Basalt (Based on a minimum porosity of 0.26).*

Tank (241-)	Current sludge depth (ft)	Maximum sludge rise (ft)	Distance to dome
TY-101	3.7	14.2	24.8
TY-102	0	0	-
TY-103	5.1	19.6	13.4
TY-104	1.3	5.0	34.
TY-105	7.0	26.9	13.1
TY-106	5.2	20	19

*1 in. = 2750 gallons of high-level tank waste.

Although determining the exact or approximate equilibrium time may be impractical, the impact of dome collapse would probably be sufficiently mitigated by filling the dome to within a few inches of the top. All of the TY Tank Farm tanks contain interstitial liquid. The maximum dose to the general public from dome collapse would occur if a dome collapsed into an air dry sludge (Murthy et al., 1982). This scenario would be mitigated to some degree by a wet sludge, which would limit the amount of contaminated "dust" expected to exit the dome. If the sludge were further covered by several feet of basalt fragments, it is improbable that any sludge would exit the dome opening. The rock-soil cover over the dome possibly would be disrupted, but any significant damage to the cover could be repaired.

BASALT FRAGMENT SIZE

The best size of basalt fragments to use in dome filling must be determined. In general, the smaller the size of the fragment, the greater the surface area and the greater the solubility (Routson, 1970). Table 2 lists the relationship between particle size (fragment size) and surface area. Surface area increases relatively slowly for coarse particles and rapidly for small particles. Furthermore, the increase in surface area of particles greater than 1.0 cm is essentially insignificant in an absolute sense, having no appreciable effect on the solubility (Routson, 1970). Thus, any basalt fragment size greater than 1.0 cm would be acceptable. The final selection of a size should be based on a cost analysis.

TABLE 2. The Relation of Surface to Particle Size (Baver, 1961).

Diameter of sphere	Textural name	Volume per particle $\left(\frac{1}{6}\pi D^3\right)$	Number of particles $\left(\frac{\pi}{6}\text{ cm}^3\right)$	Total surface (πD^2 x number of particles)
1 cm	Gravel	$\frac{1}{6}\pi(1)^3$	1	3.14 cm^2 (0.49 in.^{2})
0.1 cm (1 mm)	Coarse sand	$\frac{1}{6}\pi\left(\frac{1}{10}\right)^3$	1×10^3	31.42 cm^2 (4.87 in.^{2})
0.05 cm (0.5 mm or 500μ)	Medium sand	$\frac{1}{6}\pi\left(\frac{5}{100}\right)^3$	8×10^3	62.83 cm^2 (9.74 in.^{2})
0.01 cm (0.1 mm or 100μ)	Very fine sand	$\frac{1}{6}\pi\left(\frac{1}{100}\right)^3$	1×10^6	314.16 cm^2 (48.67 in.^{2})
0.005 cm (0.05 mm or 50μ)	Coarse silt	$\frac{1}{6}\pi\left(\frac{5}{1000}\right)^3$	8×10^6	628.32 cm^2 (97.34 in.^{2})
0.002 cm (0.02 mm or 20μ)	Silt	$\frac{1}{6}\pi\left(\frac{2}{1000}\right)^3$	125×10^6	1,570.8 cm^2 (1.69 ft^2)
0.0005 cm (0.005 mm or 5μ)	Fine silt	$\frac{1}{6}\pi\left(\frac{5}{10,000}\right)^3$	8×10^9	6,283.2 cm^2 (6.76 ft^2)
0.0002 cm (0.002 mm or 2μ)	Clay	$\frac{1}{6}\pi\left(\frac{2}{10,000}\right)^3$	125×10^9	15,708 cm^2 (16.9 ft^2)
0.0001 cm (0.001 mm or 1μ)	Clay	$\frac{1}{6}\pi\left(\frac{1}{10,000}\right)^3$	1×10^{12}	31,416 cm^2 (33.8 ft^2)
0.00005 cm (0.0005 mm or 500 mμ)	Clay	$\frac{1}{6}\pi\left(\frac{5}{100,000}\right)^3$	8×10^{12}	62,832 cm^2 (67.6 ft^2)
0.00002 cm (0.0002 mm or 200 mμ)	Colloidal clay	$\frac{1}{6}\pi\left(\frac{2}{100,000}\right)^3$	125×10^{12}	157,080 cm^2 (169 ft^2)
0.00001 cm (0.0001 mm or 100 mμ)	Colloidal clay	$\frac{1}{6}\pi\left(\frac{1}{100,000}\right)^3$	1×10^{15}	314,160 cm^2 (338 ft^2)
0.000005 cm. (0.00005 mm or 50 mμ)	Colloidal clay	$\frac{1}{6}\pi\left(\frac{5}{1,000,000}\right)^3$	8×10^{15}	628,320 cm^2 (676 ft^2)

ASSESSMENT

Three independent methods were used to evaluate the solubility of basalt fragments at 170° to 180°C temperatures for a 500-yr duration: (1) by inferring solubility qualitatively from basalt weathering in a surface environment corrected for temperature; (2) by estimation of basalt solubility in natural watershed under known environmental conditions corrected for temperature; and (3) by calculating solubility from thermodynamically derived considerations. The conclusions of these three methods are compared and a combined estimate of the importance of solubility given. These three methods represent an increasingly scientific rigor for estimating solubility.

INFERRED BASALT SOLUBILITY

It has been observed that chemical weathering is a powerful agent in the destruction of some surface rock. Solubilization of ions from silicate mineral surfaces frees ions for further transport away from the mineral being solubilized. Weak gaps are exposed, and the rock weathers further. Dated masonry surfaces, statues, and reliefs have been observed to be affected (Winkler, 1973). As a relevant example, Cernohouc and Salc (1966) attempted to quantify the weathering rate of basalt in a castle in Bohemia. Figure 1 shows an attempt to generalize weathering as a function of time; penetration of weathering was estimated to have occurred to a depth of approximately 3.5 mm in 500 yr. Probably less than 10% of the minerals in the weathered basalt rind were dissolved, or the weathered rock would have disintegrated. Even considering the effect of temerature, limited total dissolution can probably be expected to occur in a 500-yr time frame. Since none of the basalt actually disintegrated, and the partially weathered basalt would probably support the dome, no significant dissolution has occurred. Furthermore, this weathering value is probably exaggerated due to the low pH of the environment compared with the high pH of a waste tank.

WEATHERING IN NATURAL WATERSHEDS

Mineral weathering was evaluated in an arid watershed containing mixed basaltic, felsic parent material at Hanford (Routson et al., 1977). The dissolution of maphic minerals, including basalt-derived pyroxene, plagioclase, and biotite, was found to control the chemical composition of a spring draining the watershed. It should be noted that basaltic minerals controlled the release of chemical components even though a significant percentage of basalt is composed of glass (Garrels, 1967). Table 3 indicates the relative molar contribution each mineral contributed to the spring water composition. Unfortunately, since the Hanford Study did not include a water balance, the amount of basalt dissolved cannot be calculated. However, basalt dissolution can be estimated by assuming that water discharged from a drainage is proportional to precipitation. Another similar study was made in approximately the same size watershed with approximately the same slope (Bricker et al., 1968). Based on this discharge assumption, the weight of basalt dissolved in 500 years is 1.3×10^6 g, which is equal to 8×10^{-8}% of the weatherable basalt (upper 6 in. of soil). Even this minute percentage is high, due to the difference in acidity in the two environments.

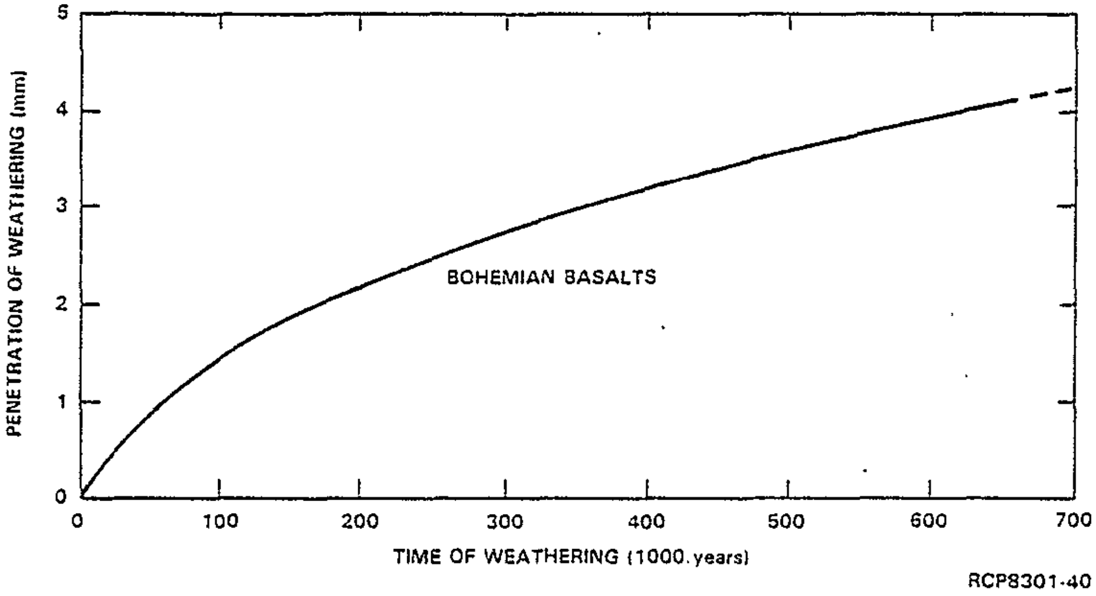

FIGURE 1. Basalt Weathering as a Function of Time (Cernohouc and Salc, 1966).

TABLE 3. Control of a Hanford Spring's Water Composition by Maphic Mineral Dissolution (Routson et al., 1977).

Mineral	SiO_2 (%)	Na^+ (%)	Ca^{+2} (%)	Mg^{+2} (%)	K^+ (%)	HCO_3^- (%)
Pyroxene	82	0	0	85	0	41
Plagioclase	6	100	100	-15	0	43
Biotite	12	0	0	29	100	16

SOLUBILITY BASED ON THERMODYNAMIC DATA

Both equilibrium and kinetic thermodynamic data are available for estimating the solubility of basalt. Stability relationships in the system K_2O-Na_2O-Al_2O_3-SiO_2-H_2O can be depicted graphically as a function of the parameters log K^+/H^+, log Na^+/H^+ and log $H_4SiO_4^0$-(SiO_2^0). Only the standard free energy of formation (ΔF) of the stable minerals in the system are required to construct graphs of the mineral relationships. These ΔF values can be determined either from solubility data (Routson, 1970) or from calorimetric data (Moore, 1965). Figure 2 is a three-dimensional graph of minerals in the NaO_2-K_2O-SiO_2-Al_2O_3-H_2O system. Figures 3 and 4 are graphs for the K^+ and Na^+ components of the system, respectively (Hess, 1966). Stable silicate minerals in the system are listed in Table 4.

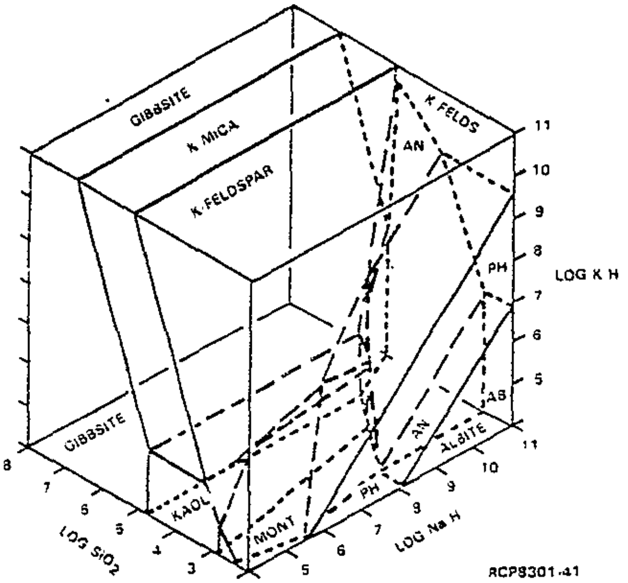

FIGURE 2. Stability Relationships of Minerals in the Na_2O-K_2O-SiO_2-Al_2O_3-H_2O System (KAOL = kaolinite, MORT = montmorillonite, PH - phillipsite, AN = analcite, AB - albite) (Hess, 1966).

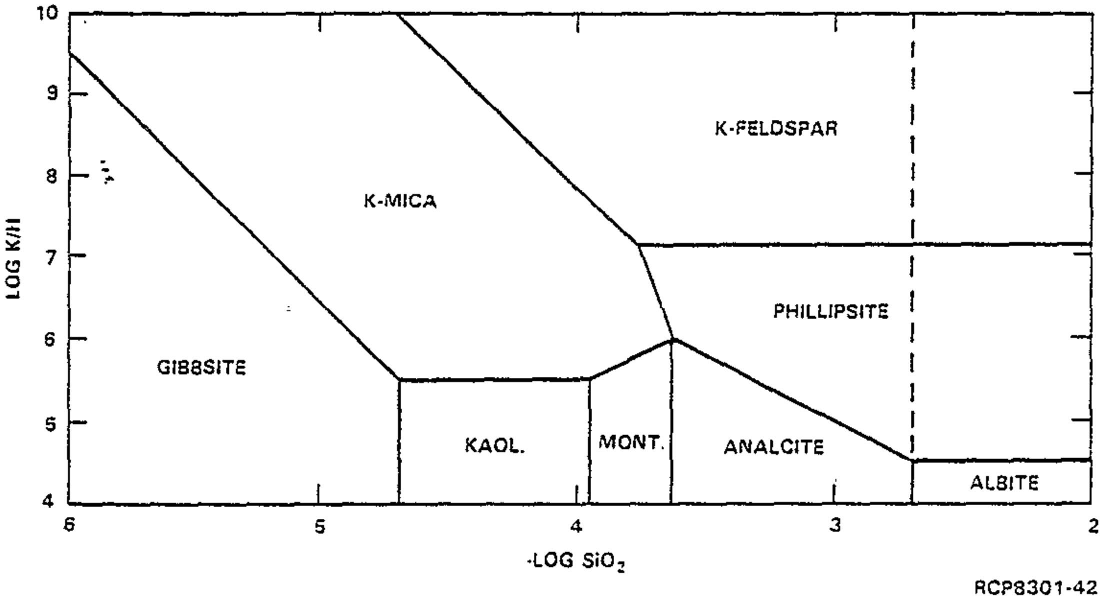

FIGURE 3. Isoplethic Section at log Na/H, 8.5, in the Stability Relationship of the Na_2O-K_2O-Al_2O_5-SiO_2-H_2O System (Hess, 1966).

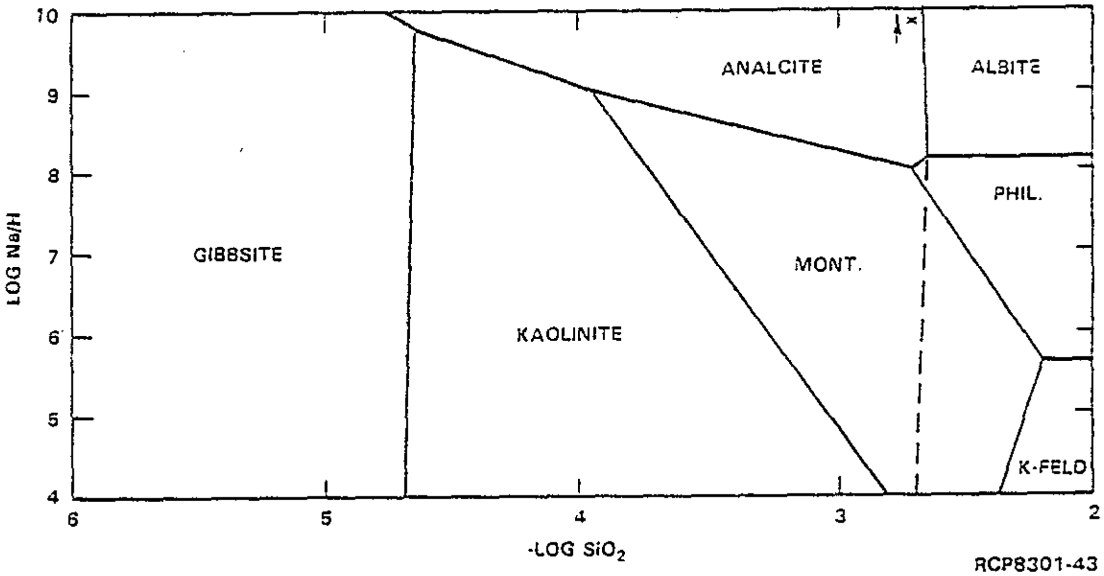

FIGURE 4. Isoplethic Section at log K/H, 4, in the Stability Relationship of the Na_2O-K_2O-Al_2O_3-SiO_2-H_2O System (Hess, 1966).

TABLE 4. Stable Minerals in the Na_2O-K_2O-SiO_2-Al_2O_3-H_2O System at 25°C (Hess, 1966).

Mineral	Composition
Gibbsite	Al_2O_3-$3H_2O$
Kaolinite	$H_4Al_2Si_2O_9$
Na-Feldspar	$NaAlSi_3O_8$
K-Feldspar	$KAlSi_3O_8$
K-Mica	$KAl_3Si_3O_{10}\ (OH)_2$
Phillipsite	$K_{0.5}Na_{0.5}AlSi_3O_8 \cdot H_2O$
Analcite	$NaAlSi_2O_6 \cdot H_2O$
Na-Montmorillonite	$Na_{0.33}Al_{2.33}Si_{3.67}O_{10}\ (OH)_2$

By determining the activities of K^+, Na^+, SiO_2, and H^+ ions in the system and plotting them in Figures 3 and 4, the most stable mineral can be determined. Plotting the activities of the components in Table 5, the most stable mineral in this system is the sodium zeolite analsite. Thus, the minerals in basalt should dissolve to form analsite. However, the important question is not if basalt will dissolve, but how fast.

TABLE 5. Approximate Composition of Hanford High-Level Wastes (Delegard and Barney, 1982).

Solution	Concentration (M)
NaOH	4
$NaNO_3$	2
$NaNO_2$	2
$NaAlO_2$	0.5
Na_2CO_3	0.05
Na_2SO_4	0.01
Na_3PO_4	0.01
NaF	0.01

To estimate the rate at which basalt will dissolve, kinetic data for the reaction of basaltic minerals with aqueous solution (dissolution) must be utilized. The empirical rate constant for the hydrolysis of K-feldspar is approximately 10^{-10} moles of plagioclase/cm^2/day$^{1/2}$ (Helgeson, 1971). The reaction rates of biotite, pyroxene, and plagioclase are probably similar (Table 1 of Helgeson, 1971). Rates of mass transfer at supercritical temperatures and pressures are approximately 300 times faster at 200°C than at 25°C. Thus, the 200°C value would be approximately 3 x 10^{-8} moles/cm^2/day$^{1/2}$. If this reaction rate represents the nominal reaction rate of basalt in 500 yr, 0.01 g of basalt would be dissolved in 500 yr if particles have 1.0-cm diameters. This would be 6 x 10^{-5}% of the basalt particle and would represent only a small reaction rind on the smallest recommended basalt fragments.

CONCLUSION AND RECOMMENDATIONS

Only an insignificant amount of dissolution is to be expected on 1.0-cm-diameter basalt fragments in TY Tank Farm high-level wastes at maximum temperatures of 170° to 180°C in 500 yr.

The foregoing analysis shows with some degree of certainty that the dissolution of basalt in the TY Tank Farm high-level waste tanks is probably negligible in 500 yr. If this level of assurance is inadequate for the program, a moderate level of verification may be required. The envisioned research would be kinetic measurements of dissolution of basalt as a function of time in simulated TY Tank Farm waste at 180°C for approximately two years. A consultant would be employed to aid in validly extrapolating the data to 500 yr. Estimated costs are itemized in Table 6.

TABLE 6. Estimated Costs of Basalt Solubility Verification.

Expense item	Cost (x $\$10^3$)
Scientist (1.25 manyear)	70
Technician (1.0 manyear)	48
Consultant	20
Travel	6
Analyses	25
Documentation (2)	5
	174*
Capital equipment	25

*Costs are for 2 yr, or about half of the above costs per year.

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