ENGINEERING CHANGE NOTICE

Page 1 of 2

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ENGINEERING CHANGE NOTICE	Page 2 of 2	1. ECN (use no. from pg. 1) 644554

16. Design Verification Required ☐ Yes ☒ No	17. Cost Impact ENGINEERING: Additional ☐ $ / Savings ☐ $ CONSTRUCTION: Additional ☐ $ / Savings ☐ $	18. Schedule Impact (days) Improvement ☐ Delay ☐

19. Change Impact Review: Indicate the related documents (other than the engineering documents identified on Side 1) that will be affected by the change described in Block 13. Enter the affected document number in Block 20.

SDD/DD ☐
Functional Design Criteria ☒
Operating Specification ☐
Criticality Specification ☐
Conceptual Design Report ☐
Equipment Spec. ☐
Const. Spec. ☐
Procurement Spec. ☐
Vendor Information ☐
OM Manual ☐
FSAR/SAR ☐
Safety Equipment List ☐
Radiation Work Permit ☐
Environmental Impact Statement ☐
Environmental Report ☐
Environmental Permit ☐

Seismic/Stress Analysis ☐
Stress/Design Report ☐
Interface Control Drawing ☐
Calibration Procedure ☐
Installation Procedure ☐
Maintenance Procedure ☐
Engineering Procedure ☐
Operating Instruction ☐
Operating Procedure ☐
Operational Safety Requirement ☐
IEFD Drawing ☐
Cell Arrangement Drawing ☐
Essential Material Specification ☐
Fac. Proc. Samp. Schedule ☐
Inspection Plan ☐
Inventory Adjustment Request ☐

Tank Calibration Manual ☐
Health Physics Procedure ☐
Spares Multiple Unit Listing ☐
Test Procedures/Specification ☐
Component Index ☐
ASME Coded Item ☐
Human Factor Consideration ☐
Computer Software ☐
Electric Circuit Schedule ☐
ICRS Procedure ☐
Process Control Manual/Plan ☐
Process Flow Chart ☐
Purchase Requisition ☐
Tickler File ☐
☐
☐

20. Other Affected Documents: (NOTE: Documents listed below will not be revised by this ECN.) Signatures below indicate that the signing organization has been notified of other affected documents listed below.

Document Number/Revision	Document Number/Revision	Document Number Revision

21. Approvals

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Cog. Mgr.		10/28/97
QA		
Safety		
Environ.		
Other		

	Signature	Date
Design Agent		
PE		
QA		
Safety		
Design		
Environ.		
Other		

DEPARTMENT OF ENERGY

Signature or a Control Number that tracks the Approval Signature

ADDITIONAL

Stabilization of In-Tank Residual Wastes and External-Tank Soil Contamination for the Tank Focus Area, Hanford Tanks Initiative: Applications to the AX Tank Farm

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Abstract: This report investigates five technical areas for stabilization of decommissioned waste tanks and contaminated soils at the Hanford Site AX Farm. The investigations are part of a preliminary evaluation of end-state options for closure of the AX Tanks. The five technical areas investigated are: (1) emplacement of smart cementations grouts and/or other materials, (2) injection of chemical "getters" into contaminated soils surrounding tanks (soil mixing), (3) emplacement of grout barriers under and around the tanks, (4) the explicit recognition that natural attenuation processes do occur, and (5) combined geochemical and hydrological modeling of soil-grout-waste interactions. Research topics are identified in support of "key areas of technical uncertainty" in each of the five areas. Detailed cost/benefit analyses of the technologies are not provided. This investigation was conducted by Sandia National Laboratories, Albuquerque, New Mexico, during FY 1997 by tank Focus Area (EM-50) funding.

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RECORD OF REVISION	(1) Document Number HNF-SD-HTI-ES-004	Page 1

(2) Title

Stabilization of In-Tank Residual Wastes and External-Tank Soil Contamination for the Tank Focus Area, Hanford Tanks Initiative: Applications to the AX Tank Farms

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Stabilization of In-Tank Residual Wastes and External-Tank Soil Contamination For the Tank Focus Area, Hanford Tank Initiative: Applications to the AX Tank Farm

1. INTRODUCTION

Five technical areas are considered for the stabilization of decommissioned waste tanks and contaminated soils at the Hanford AX tank farm: (1) emplacement of smart cementitous grouts and/or other materials, (2) injection of chemical "getters" into contaminated soils surrounding tanks (soil mixing), (3) emplacement of grout barriers under and around the tanks, (4) the explicit recognition that natural attenuation processes do occur, and (5) combined geochemical and hydrologic modeling of soil-grout-waste interactions. Detailed tabulations of stabilization options for tanks and contaminated soils have been recently proposed (e.g., Kincaid et al., 1993; Treat et al., 1995), and a summary of our recommendations in these areas may be found in Sections 2-5 of this report. Solidification and stabilization technology is currently being used to treat a wide variety of wastes, and advancements taking place everywhere in the cement industry are contributing to continuous integration of new technologies at sites throughout the United States. The engineering-related recommendations presented in this report are therefore posed as general guidelines, and will assuredly require adjustments in light of these changing technologies.

Yet, despite the apparent rapid evolution of solidification and stabilization cement technologies, considerable uncertainty remains regarding radioactive waste stabilization and the chemical processes that occur as the various components of a disposal package interact. In Section 6 we approach this problem through an integrated, site specific study of tank AX-102, considering, (1) the effect of ground water incursion on the estimated waste inventory, (2) cement-waste package effects on actinide solubilities, and (3) the effect of adding specific minerals as chemical getters to the waste backfill. The chemistry of tank AX-102 waste is based on an estimated inventory following retrieval of 90% of the waste (Ramsower, 1997). Ground water infiltration rates are taken from studies of the local hydrogeology (Fayer and Walters, 1995). Predicted actinide solubilities were performed using the program "React" (Bethke, 1994), which calculates aqueous and surface speciations in an assumed baseline condition and in three assumed closure scenarios involving cement grout and soil backfill materials. The models are accurate only to the degree to which the baseline chemical inventories of Hanford tanks have been estimated. Additional information regarding phase mineralogy and radionuclide speciation may be needed to more fully understand the chemical equilibria of initial fluid-mineral system(s) for each tank. However, in the absence of these data we contend that the models in Section 6 are reasonable first-order estimates of the solubilities and retardation factors that can be expected in tank AX-102 under the prescribed conditions.

2. EMPLACEMENTS WITHIN TANKS

2.1. *Filler, Design*

A review of the substantial literature on Hanford tank closure options revealed that mechanical stabilization is best achieved by backfilling with an inexpensive coherent filler material. Candidate materials range from crushed and sieved basalt to grouts (Smyth et al., 1992). However, as long as tank integrity is maintained there is the possibility that water will collect and create a reservoir of fluid in contact with residual waste. Minimization of filler pore space and reducing pore diameter to the point where capillary forces will hold the fluid in place would minimize fluid-waste contact. In addition, the use of multiple sealant layers above the waste would reduce the possibility for formation of a single crack from top to bottom of the tank.

Tank filling strategies range from emplacement of granular materials to pumping of a semi-fluid material. Granular material emplacement necessitates (1) the regular distribution of material into the tank, and (2) compaction of the material to its "final" density in the time allotted for the closure process (Smyth et al., 1992). The engineering challenges of achieving these two goals are considerable. Rather, we prefer injection of semi-fluid materials that would flow over the irregular surfaces in each tank. Two cementitous grouts are identified with the requisite longevity and low cost; Portland cement based and bentonite based. Several commercially available bentonites may be emplaced as fluids and induced to seal, such as Volclay® and Benseal®. These are common components of subsurface barriers and the emplacement technologies are well established (Rumer and Mitchell, 1995). Costs are on the order of $125 per ton of material. Wakeley and Ernzen (1992) developed a Portland cement-based grout with the appropriate rheological and thermal properties for "mass emplacement." Components of the grout are those typical of most Portland cements so the cost of the materials will be similar to those of normal concrete. Because the material is designed to fill vaults, emplacement costs are not excessive. This material should serve as a starting point for additional developments in the field (e.g., incorporation of fiber reinforcement that would inhibit crack propagation). A variety of fibrous materials (steel, glass, cellulose etc.) have been evaluated (Kosmatka and Panarese, 1990) and additional research should examine novel materials such as graphite fiber, which might have a longer lifetime in the tank environment.

We propose a layered, cementitous grout backfill structure rather than filling the tank with a single massive grout formulation (figure 1). Successful placement of multiple 16 inch layers of grout (dark lines in the blue field in figure 1) under simulated Hanford Grout Vault Program conditions has been demonstrated (Wakeley and Ernzen, 1992). We also recommend a similar lift pouring strategy with sufficient intervening time to allow for the heat liberated in setting to dissipate. A 2 foot thick Benseal® layer about 8 feet from the top of the stack is also recommended to help seal the underlying grout layers and to inhibit the formation of long, through-tank cracks. Consideration was given to placing bentonite layers further down as well, but it is not clear that the bentonite would support a greater thickness of grout, particularly if the tank walls were to fail and some of the clay was extruded into the surrounding formation.

2.2. *Filler, Function*

The function of this package is hydrologic and is intended to divert water away from the tank and preclude undue wetting of the waste (Kincaid et al., 1993; section 5.2 therein). The propagation of cracks through the grout layers as a result of settling, seismic disturbances, etc. would encounter the plastic Benseal® or Volclay® layers and be arrested. Conversely, any contraction of Benseal® or Volclay® layer would create a limited zone of porosity before encountering a rigid grout member. The hydraulic conductivity of a "tight" grout or concrete is typically at least three orders of magnitude less than soil, and the pore diameters in grout are also significantly less than that of the poorly sorted gravel fill surrounding the tanks (Brendel, 1997). Once this coarse material becomes dampened, its hydraulic conductivity will increase (Wierenga, 1995) and it will become the preferred path for draining the top of the tank. Thus, if a sustained

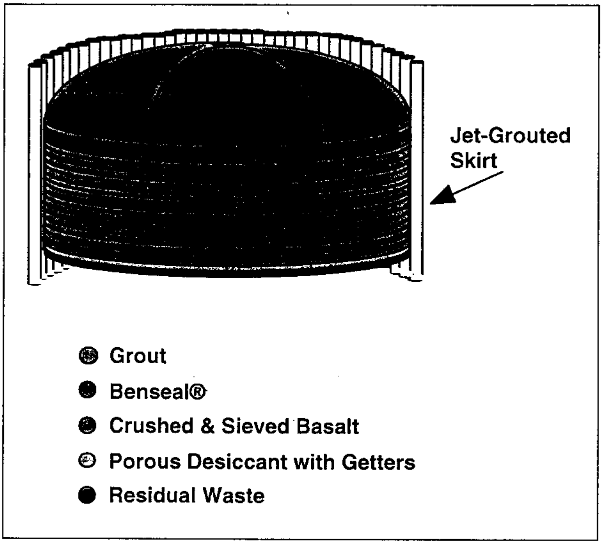


Figure 1. Proposed tank stabilization scheme (not to scale).

recharge managed to pass through the Resource Conservation and Recovery Act (RCRA) cap, the gross effect of the design would be to divert the flow around the tank and the waste inside.

2.3. Filler, Research

1. *Better definition of the Portland cement and bentonite based grouts to actually be used (engineering design issue).*

2. *Structural engineering of the tank filling: The design inherently incorporates strong (brittle) and weak (plastic) layers that in aggregate will exert a significant pressure on the bottom and sides of the tank. The design should be one that is inherently stable and able to support*

its own weight (engineering design issue).

3. Chemical stability of the bentonite-Portland cement interface: Bentonite seals are typically emplaced with the clay in the Na-exchanged form since it readily disperses in the emplacement water. The bentonite seal will, however, be in contact with the Portland cement based materials on both sides. Bentonite clays used in grouts typically are converted to the CSH and CAH phases indigenous to Portland cements (Rumer and Mitchell, 1995). Even if this does not happen there will be at least some displacement of Na by Ca on the clay exchange sites. This increases the clay permeability significantly and causes the plasticity to drop dramatically (Grimm, 1968). In a marl, the effect of concrete on pore water chemistry is calculated to extend out about 0.2 meters in 12,500 years (Eikenberg and Lichtner, 1992), which suggests that if an unreacted septum of clay is to exist in the interior, the barrier should be on the order of 60 cm thick. Clearly, the process needs to be quantified (e.g., Lee et al., 1995) for the conditions relevant to the planned structure.

4. Continued evaluation of concrete longevity; apply models to site-specific circumstances and catalogue studies on the longevity of buried concrete structures. We note that the short service life of concrete in many PA documents pertains to the near-surface environment in which freeze-thaw and wet-dry cycles significantly enhance degradation rates. In the subsurface environment these effects are absent and we expect to show that significantly longer service lives are appropriate.

2.4. Waste horizon treatment, design

The state of the residual waste is also a concern. Current strategies for pre-closure treatment of the tanks call for repeated sluicing to remove in excess of 90% of the contents and preferably at least 99.9%. A significant fraction of the residuum is expected to be a watery fluid. The remainder will consist of discarded equipment and a pasty to semi-cemented mass of hydrous iron and aluminum hydroxide "sludge" adhering to the interior surfaces of the tank. Radionuclides sorbed on the solid fraction of the sludge will leach with difficulty. However, radionuclides in the sludge pore fluids and the watery fluid overlying the sludge will be highly mobile. The most abundant long-lived radionuclides of concern are: Tc, U, Pu, Np and Am.

A two-part strategy is proposed for treating the residual waste (figure 1). Initially, dry Portland cement (or other low-cost additives such as fly ash and lime) will be added to desiccate the waste. The next layer consists of crushed and sieved basalt with enough pore space to accommodate any of the pasty sludge that is displaced upward by the weight of the grout/bentonite cap. If only a few inches of sludge exists, the crushed stone can be omitted because the desiccant will likely wick the upper few inches of sludge dry and consume the overlying fluid. In either case, the waste heel will be quite porous so little hydrologic benefit can be ascribed to this treatment.

The goal of treating the waste using chemical means is to reduce radionuclide concentrations in solution. This can be accomplished with getters added to the waste prior to the desiccation step. For optimal getter performance, it may be desirable to neutralize the fluid with an acidic solution or by filling the headspace of the tank with an acidic gas, such as carbon dioxide. Getters work by either ion exchange or precipitating a phase containing the radionuclide of concern. A summary of potential getters is presented in Table 1.

Table 1.
Potential Getter Materials For Various Radionuclides

Radionuclide	*Getter Material*	*Cost*
Cs	Crystalline Silicotitantate Titanate (CST) Illitic Clays	Expensive Inexpensive
Sr	CST $SrCO_3$	Expensive Inexpensive
Am	Hydroxycarbonate - coprecipitated with Nd Hydroxyapatite	Inexpensive Inexpensive
Tc	Magnetite or other reducing agent (may require nearly neutral solution to work quickly)	Inexpensive
Np, Pu	Hydroxyapatite Ca-phosphate coprecipitation FeOOH	Inexpensive Inexpensive Inexpensive
U	Magnetite or other reducing agent Ca-phosphate coprecipitation Ca-uraninates/cement phases	Inexpensive Inexpensive Inexpensive
Se	FeOOH Fe-Hydrotalcite (both may require nearly neutral solution to form)	Inexpensive Inexpensive
I	Cu oxides and sulfides Ag compounds	Inexpensive Inexpensive

Ramsower (1997) summarizes the likely radionuclide concentrations in the retrieval scenarios for the four AX farm tanks. Tank AX-102 fluids have the highest radionuclide concentrations, whereas concentrations in the remaining three tanks may be four orders of magnitude lower. In contrast to the mass balance estimates used to derive the values in Table 2, AX-102 supernate analyses were reported for Am and Pu of 1.2×10^{-5} moles/liter and 0.5×10^{-5}

moles/liter, respectively (Ramsower, 1997; see Tables 2-4 therein). Dissolved concentrations of Am are in reasonable agreement with Table 2, whereas the Pu value is ~13x lower. Am is apparently soluble in the tank fluids, but much of the Pu (and probably U and Np as well) may be associated with the solids in the sludge.

Table 2.
Radionuclide Concentrations in Tank AX-102 Rinse Solutions

Radionuclide	*Concentration (mol/L)*
Tc	9×10^{-6}
U	6×10^{-3}
Pu	7×10^{-6}
Am	4×10^{-6}
Np	3×10^{-6}

2.5. Waste Horizon Treatment, Function

Although we have not fully evaluated the function of each getter, the literature contains considerable information to make the following forecasts:

1. Magnetite Fe_3O_4 has been shown to lower Tc concentrations from an initial value of 10^{-6} molar to a final value of 10^{-7} molar (Chu and Eriksen, 1996). However, the reaction is very slow in strongly basic solutions, and the solubility of the TcO_2 increases above a pH of 9.2 due to the formation of the $TcO(OH)^-_3$ complex. At pH ~12 solubility reaches a value of 10^{-4} molar (Eriksen et al., 1992) thus, it may be necessary to temporarily neutralize the residual waste to accommodate Tc removal. Re-release of Tc is very slow; once it is sequestered by magnetite Tc should not go back into solution following grout emplacement. At present we

cannot provide estimates of the quantity of magnetite required to scavenge a given amount of Tc. Further research on Tc sorption/precipitation on magnetite surfaces is needed.

2. A multitude of phases exist that might sequester U. The following discussion focuses on the use of cementitous materials to fix the more mobile U(VI) species. It is generally agreed that U(IV) solubility is exceedingly small. Theoretically, solubilities for U(VI) in a cementitous matrix range from 10^{-4} to 10^{-14} M (Berner, 1990), which is in approximate agreement with the calculated solubility of 10^{-15} M determined from equilibrium state modeling (Bethke, 1994) of a theoretical waste package (see section 6 for details). However, in practice the measured value ranges from 10^{-7} to 10^{-8} molar (Atkins and Glasser, 1990; Moroni and Glasser, 1995), which is significantly lower than the value in Table 2.

3. Pu is problematic because, like uranium, it exhibits a variety of valence states. Further, like uranium the tetravalent state is highly insoluble. The fact that AX tank fluids contain about 10^{-6} moles/liter dissolved Pu indicates that an appreciable fraction exists in the hexavalent state. Therefore Pu behavior should resemble that of uranium, and its solubility should undergo a 10 to 100 fold decrease in a cementitious matrix. This is in part supported by React model calculations for a theoretical waste package (Bethke, 1994; see Section 6 of this report for details), which demonstrate similarity in U and Pu solubilities at pH 13. The Kd for U and Pu is in the 10^3 range, however in the high pH tank environment these values will probably drop to ~100. However, as is noted in section 6, the large drop in U and Pu solubility in the presence of grout-related fluids will greatly enhance whatever loss in apparent Kd occurs at high pH. Secondly, it is known that hydroxyapatite has a Kd for Pu of

about 980 in saturated NaCl brines. When a getter acts by sorbing a radionuclide (rather than precipitating it) the final concentration depends strongly on the amount of getter that is added. Therefore a variety of design issues need to be resolved before an estimation can be made of the decrease in Pu concentration that might result from adding a hydroxyapatite getter.

4. Am presents an interpretation problem. On one hand, the hydroxy-carbonates of trivalent ions are exceedingly insoluble (Felmy et al., 1990; Carroll, 1993). Given the high concentrations of hydroxide and carbonate in these fluids, the solubility of Am should be less than 10^{-7} M. Yet, supernate Am concentrations on the order of 10^{-5} M indicates that there may be other carbonate complexes that have yet to be evaluated. Fortunately, hydroxyapatite appears to have a very high Kd for Am, in excess of 50,000 in the neutral pH range. In the high-pH tank environment we conservatively scale the Kd down an order of magnitude to 5,000 until actual measurements are made. Again, issues of capacity exist as does the possibility that some Am complex will exist that is unique to these fluids.

5. Like Am, it appears that hydroxyapatite has a very high Kd (greater than 59,000) for Np under normal pH conditions. However, like Am, the Kd for Np will certainly drop in the higher pH range, and for this reason we scale the Np Kd back 10 times to a value of 5,900.

2.6. Waste Horizon Treatment, Research

1. Explore the systematics of Tc sorption on magnetite in the relevant solutions. This will be an experimental study where typical DSSF-type tank fluids will be diluted, neutralized to

varying degrees with CO_2, and then spiked with Tc. Different forms of magnetite and other Fe^{+3}/Fe^{+2} solid preparations will also be added. For those getters that successfully remove Tc, the desorption kinetics will be determined at pH values likely to exist in cement pore fluids (a pH of about 12).

2. Using U and Nd as a stand-in for Am to experimentally evaluate the systematics of sorption on hydroxyapatite in DSSF-type solutions is proposed (see #1 above). The effectiveness of pre-formed hydroxyapatite in removing these elements will be compared with that of precipitating fresh calcium phosphate in solutions that are pre-spiked with Nd(Am) and U.

3. Resolve the issue of why the Am(Nd) appears to remain in solution when the calculated solubility appears to be so low. This will be a combined computational and experimental study. Two computer codes based on thermodynamic equilibrium are available (React and EQ3/6) and will be applied to the problem.

4. Do a mock-up of the sludge desiccation process with emphasis on characterizing both the chemical and textural features that result, as well as determining the thickness of an artificial sludge heel that can be desiccated.

5. Search the literature and incorporate what the saltcrete programs have learned about the stability and leach characteristics of their waste form.

6. Provide React and EQ3/6 computations to demonstrate that tetravalent U and Pu concentrations should remain much less than 10^{-8} molar over the relevant range of conditions. If possible, do the same for Nd (e.g. Am).

3. GROUT BARRIERS UNDER AND AROUND THE TANK

3.1. Design

Rumer and Mitchell (1995) have edited a definitive work on the construction of subsurface barriers that contains far more information than can summarized here. However some pertinent information such as general costs and dimensions that can be achieved with existing technologies are listed in Table 3. Only inorganic materials were considered for this application because organic "chemical grouts" cannot be demonstrated to have lifetimes commensurate with the required regulatory time frame.

The third part of our design is a skirt to be emplaced against the exterior walls of the tank (figure 1). Because it is desirable to minimize the amount of soil brought to the surface, and because a close bond to the curved outside tank surface is needed, jet grouting is the preferred technology. The skirt would extend some distance beyond the base of the tank, and would have a wall thickness of about a meter. A 3-foot thick skirt that was 50 feet high would contain about 1000 m^3 of grout and cost about $24K (Dwyer, 1994).

3.2. Function

The skirt performs two functions. By extending beyond the base of the tank it diverts the flow of ground water past the base of the tank. Thus, the only remaining mechanism for getting the waste out of the decommissioned tank is diffusion, which is exceedingly slow when the concentration gradient through unsaturated soil is small (the generic function of the getters) and distances of tens of feet are involved.

Table 3.
Representative Costs of Different Types of Barriers

Wall Type	**Width (ft)**	**Depth (ft)**	**Unit Cost ($/ft^2)**	**Production Rate (ft^2/10 hrs)**
Soil Bentonite	2-3	80	2-8	2500-15000
Cement Bentonite	2-3	80	5-18	1000-8000
Biopolymer Drain	2-3	70	7-25	1500-5000
Deep Mixing	2.5	90	6-15	1000-8000
D-mix, Structural	2.5	90	15-30	1000-3000
Jet Grouting	1.5 - 3	200	30-80	200-2500
Grout Curtain	0.5 - 1	200	40-100	200-1000

Secondly, increasing the wall thickness to about a meter would preclude degradation of the wall to the point that radionuclides could exit laterally, or that horizontally moving ground water would enter the tank. Wall thickness determination was based on concrete ionic diffusion coefficients on the order of 10^{-8} cm^2/sec (Walton et al., 1990). A plane source with a zero concentration at the boundary will have a depletion front due to diffusion that moves only about 0.5 meter into the mass over 10,000 years. Although there are many refinements on the calculations (e.g., Haworth et al., 1988; Alcorn et al., 1990; Walton et al., 1990; Lin, 1991; Adenot and Buil, 1992; Buil et al., 1992; Pihlajavaara, 1992) diffusion will control the deterioration rate of concrete, and in the 10^4 year period of concern, maximum ~0.5m of degradation is expected under the most adverse circumstances.

Two points need to be made in regard to this design. First, to take credit for the design will require a specialized fluid flow model able to handle small geometric details. It is not something to be found as a part of the garden-variety PA package. Secondly, the skirt does not have to be a perfect barrier. A reasonable guess might be that the barrier will be 90% effective. This, however, means that the source term for whatever PA package is being used would be only be 10% of that which would exist had not the skirt been constructed. This can make a significant difference in the outcome of such a calculation.

3.3. Research

1. *There is no chemical benefit to this component so the issues associated with its implementation are hydrologic. At least in its simplified form it should be possible to model the flow lines around such a geometry using in-house codes such as TOUGH. Issues such as the optimal extent of the skirt below the tank bottom and the minimum acceptable hydraulic conductivity of the grout and clay layers filling the tank should also be addressed. The impact of increasing rainfall could also be treated. The effect of hypothetical hair line cracks (either in the tank filling plug or between the wall of the tank and the skirt) on overall water flow patterns could also be addressed (e.g., Walton et al., 1990).*

2. Continue to evaluate new barrier technologies, such as are suggested in a just-received Russian report that is currently being evaluated at SNL.

4. CHEMICAL INJECTION OF SOILS (SOIL MIXING)

4.1. Design

Directional drilling is required to get beneath the tanks. The geometry typically proposed is a V-shaped pattern of holes that intersect beneath the tank. This is similar to the design proposed in section 4.3.3 of Treat et al. (1995). Assuming the tank bottom to be at a depth of 55 feet (Kos, 1997) and using a 45° inclined hole, a 190 foot long hole would be required to penetrate to the center of the AX tank farm "fourplex". The depth of the hole at this point would be 135 feet so this intersection would be above the water table. The entire area of the grout panels (including end caps) is about 1.12×10^5 square feet.

Such a barrier was constructed by jet grouting around a small 7,500 l tank at the Hanford Geotechnical Facility in 400 area at a cost of $\$24/m^3$ (Dwyer, 1997). Peterson and Landis (in Rumer and Mitchell, 1995, p. 185-209) discuss such structures in detail and are less encouraging regarding costs: "Jet Grouting. For conventional civil engineering applications, high pressure jet grouting typically costs $15 to $20 per square foot for columnar walls. For thin diaphragm walls using jet grouting, costs are estimated at $10 to $15 per square foot. For estimating purposes, it is assumed that the typical high pressure jet grouted wall has a diameter of roughly 6 feet and the thin diaphragm wall length is 5 feet in both directions. In addition, the cost of directional drilling is estimated at $8 to $25 per square foot for columnar walls drilled on 6-foot centers and $5 to $15 per square foot for thin diaphragm walls drilled on 10-ft centers. The installed cost of a jet grouted columnar floor is estimated at $23 to $45 per square foot and, for thin diaphragm wall floor, $15-$30 per square foot. Not included in these estimates are the costs of the grouting materials, waste disposal, and contingencies." In short, to construct this barrier around the fourplex AX tank farm will cost in excess of a million dollars.

A somewhat simpler application of jet grouting would be to treat the contaminated soils around the tanks. Brendel (1997) estimates that there are 1.63×10^4 cubic yards of material. The heterogeneous distribution of these soils would be difficult to treat with any other mass stabilization process. The process should involve vertical drilling of shallow holes and have an associated cost of approximately \$24/m^3; total cost would therefore approximate \$300K.

4.2. Function

Typically the V-shaped troughs are intended to completely seal the tank off from its surroundings. With the addition of an organic inner liner, this is a tractable problem - at least for the short term (Williams and Ward, 1997). However, given the 10,000 y regulatory period, it is relevant to ask whether this is a desirable objective. A climate shift to a damper mode may cause the liner to become ponded and induce prolonged leaching of the waste. It makes more sense to inject getters without a binder and create a permeable reactive barrier (Shoemaker, et al., in Rumer and Mitchell, 1995, p.301-353). As with the skirt, the effectiveness is proportional to the area actually covered. If the permeable reactive barrier were 90% effective at scavenging radionuclides exiting the tank, the source term would be reduced by another factor of ten. However, given the size of the engineering task, the difficulty of verifying a successful emplacement, and cost, it is likely that the rest of the PA should demonstrate a clear need for this structure before it is seriously proposed.

Jet grouting may be the only economic method of treating contaminated soils in place given their extremely irregular distribution. A combination of getters and grout would both reduce the flux of water that contacts the waste and reduce the solubility of the radionuclides in the small amount of water that does gain access to the waste. Essentially the same computational procedure used to predict flow around the tanks might be used for jet grouted masses of soil, though the dimension would clearly be smaller. However, if the jet grouted masses of soil do not have an uncontaminated rim of grout it would be necessary to consider the diffusion of radionuclides out through the side of the mass in addition to the loss through the base.

4.3. Research

1. Getter research similar to that proposed for the in-tank stabilization plan discussed in section 2.6. Additional engineering-related issues concerning emplacement options and optimal grout-getter mixtures will also be addressed.

5. THE EXPLICIT RECOGNITION OF NATURAL ATTENUATION

5.1. Design

Natural attenuation is not an engineering issue - but rather a recognition of the natural interactions between waste (solutions, usually) and the soils traversed by the waste. The problem of barrier design for natural attenuation is partly one of how the PA package is developed and partly one of getting the appropriate data. Most PA packages employ a Kd approach to model radionuclide retardation so the first step is to get site specific parameters. Thus one “design” for incorporating natural attenuation is to obtain the best Kd values available for the soils underlying the AX tank farm. Kincaid et al. (1993) provide a Kd tabulation for AX tank farm soils, but values given for the actinides other than U seem to be much less than might be expected (e.g., Brady and Kozak, 1995). Thus, one way of designing for natural attenuation will be to improve on the Kd values to be put into the PA calculations.

However, providing the "correct" site specific Kd values may not really address the issue of incorporating natural attenuation. This follows from the fact that the Kd approach often does not represent the chemical processes that govern the mobility of a particular radionuclide. This typically arises from one of two causes: (1) precipitation/dissolution mechanisms may dominate, and these simply have no analogue in the mathematics of the Kd approach, and (2) where ion exchange is actually the proper mechanism, the desorption step may be so slow that assuming reversible equilibrium greatly overestimates the amount of radionuclides available for leaching. Once the proper data is obtained the next step requires rewriting the PA code to account for this new and more complex chemistry. Reactive transport codes are just now making their appearance and would require a substantial effort to embody into the program. Finally, historical Kd approaches are based on normal rainfall assumptions. From the literature, we are aware of substantial surface leaks at the AX Tank Farm that were followed by extensive water flushing operations that probably lead to far broader initial depth dispersal of radionuclides (Ramsower, 1997; see Appendix A, p.6 therein). These releases will require special modeling assumptions to correctly interpret Kd's.

5.2. Function

The function of incorporating natural attenuation clearly lies in providing more realistic estimates of expected radionuclide migration rates. But, to achieve this first requires improving the PA package that makes the predictions.

5.3. Research

1. Compile a wider list of Hanford Site Kd values and use geochemical modeling codes such as React to interpret the data. In particular, try and determine where ion exchange is likely to have operated and where it is more likely that what limited the dissolved concentration of a radionuclide was the precipitation of a phase. This will at least tell us when we might be

justified in using a Kd approach in modeling the migration of a radionuclide.

2. Obtain AX tank farm sediments and perform batch Kd and column transport experiments using various diluted and neutralized derivatives of the DSSF-type fluids. It is likely that the unique chemistry of the leaked tank fluids will effect Kd values. This is not something that will be in the literature, so it will have to be evaluated experimentally. Again, only a few of the relevant elements can be tested easily but this should give a good idea of how much of an effect this can have.

6. IN-TANK GEOCHEMICAL AND HYDROLOGIC MODELING

6.1. Introduction

The preceding sections summarize a design framework for tank closure that should be economically feasible while also minimizing worker exposure. We expand on specific technical issues by considering basic hydrologic and geochemical aspects of the *near-field* tank environment. Hydrologic calculations provide semiquantiative estimates of the water flux expected to reach the waste given four closure scenarios, and the geochemical calculations address the waste remaining in the tanks and possible chemical retardation that could lower the solubility of various radionuclides, with emphasis on the actinides (Tc is problematic at high pH; see Section 2.5). Finally, these results are combined to provide relative waste release estimates into the far field environment.

Four closure scenarios are evaluated. First a baseline condition is evaluated where a tank is filled with soil and the flux of water through the package is assumed equal to that of the soil column. In the second scenario the waste is covered with a dry cement-getter mix and the tank is backfilled with soil; ground water infiltration rates are estimated for a typical Hanford soil type. The third scenario examines the effect of grouting on the waste form, and assumes that through-going cracks develop, and that fluid is transported to the waste at some fixed rate. The fourth scenario assumes an intact low permeability grout filler, so that the only escape pathway for radionuclides is by diffusion out of the tank bottom. This scenario requires no hydrologic calculations other than the observation that it is likely that the soil will eventually approach saturation for several feet outward from the tank bottom. Our analysis of the problem also considers the impact of placing RCRA caps over individual tanks or the entire AX tank farm, effectively reducing the infiltration rate of ground water into the tanks for an indefinite time.

6.2. Hydrologic Modeling

In this section, the flux of water due to downward infiltration is estimated. This estimate is based on simplifying assumptions, which are described in this section. More detailed description of infiltration at the Hanford site are given by Fayer and Walters (1995) and Gee et al. (1996). General discussions on estimating infiltration are presented in Baer (1979) and Bouwer (1994). The conceptualized vadose flow system for the tanks in shown in Figure 2.

6.3. Infiltration rate

The downward flow and infiltration from the surface can be estimated using the Green and Ampt Equation:

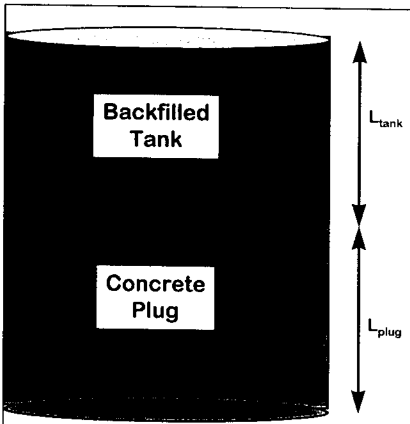
$$V_i = \frac{K(H_w + L_f + h_{cp})}{L_f}$$

V_i = infiltration rate (length/time)
K = hydraulic conductivity of the wetted zone
H_w = depth of water above the soil

L_f = *depth of the wetting front*
h_{cp} = *critical pressure head of soil for wetting*
and
$K = a^*K_{sat}$

The Green and Ampt equation can be written for infiltration through the backfilled tank and the underlying cement plug. The infiltration rates for both zones can be ratioed producing the following equations:

$$V_i(plug) = \frac{K_{sat}(plug)}{K_{sat}(\tan k)} * V_i(\tan k)$$



Properties of Backfilled Tank
Backfill assume to be
similar to silty sandy gravel

K_{sat}=5.2x10^{-3} cm/sec
porosity = 0.26
soil retention capacity - 0.03
density =1.96 gm/cm^2
infiltration rate = 5.9x10^{-4} ft/day
or 1.8x10^{-2} cm/day

Properties of Concrete Plug

assumed to be parallel plate
fractures of various apertures
K_{sat}=a^2/12 cm/sec
where a = the effective aperture of the fracture
infiltration rate through the plug is approximately equal to
(K_{sat}(plug)/K_{sat}(tank)infiltration rate(tank))

Figure 2. Conceptual Backfilled AX Tank and Underlying Cement Plug with Assumed Hydrologic Properties or Relationships.

and for fractured flow (Baer, 1979):

$$K_{sat}(plug) = \frac{a^2}{12}$$

where
a = the effective fracture apertures
values of $K_{sat}(plug)$ are presented in Table 4 for various apertures

The mean saturated hydraulic conductivity for the silty sandy gravel at Hanford is 8.2×10^{-3} cm/sec. Based on the ratioed Green and Ampt equation, if the saturated hydraulic conductivity of the plug is greater than 8.2×10^{-3} cm/sec (apertures of 0.001 cm or greater), then the backfilled tank controls the infiltration rate (therefore, the Hanford infiltration rate of 1.8×10^{-2} cm/day is applicable). If the saturated hydraulic conductivity of the plug is less than 8.2×10^{-3} cm/sec (apertures of less than 0.001 cm), then the cement plug controls the infiltration rate.

6.4. Geochemical Modeling

6.5. Introduction

Radionuclides remaining in the tank will exist either as solid components in the sludge or dissolved components in the fluid phase. Radionuclides associated with the sludge may either exist as their own phases or as sorbed and coprecipitated components on the nonradioactive materials that make up the bulk of the solids (principally iron and aluminum hydroxides). Components such as technetium and cesium will reside almost entirely in the aqueous phase; for others, such as Sr and Am, the opposite will be true. However, for other elements such as the actinides, the distribution is less certain. Thus, a major objective is to obtain basic solubility information on the actinides in order to assess probable distributions among the various phases.

The second objective is to evaluate getter performance; the study of sorption and desorption processes in this unique chemical environment is in its initial stages. In Section 2.4 we recommend several getters (Table 1) in addition to endorsing the use of grout as a tank fill material. In the following discussions we quantitatively evaluate simple grout-waste interactions and examine the effect of adding phosphate to precipitate an insoluble actinide phosphate.

6.5.1. Approach

The React code (Bethke, 1994) was used to perform the requisite solubility calculations. The code uses a thermodynamic approach to calculate the equilibrium configuration of various fluid-solid assemblages based on their bulk chemistry. In setting up the problem it was assumed that tank closure would result in recovering about 90% of the waste. Although the Tri-Party Agreement mandates that 99% of the waste be retrieved, we used a 90% retrieval target as a worst-case scenario. In effect, this results in a 10-fold dilution of the waste fluid. Residual radionuclides concentrations are listed in Table 2, and the resulting trial fluid composition (after the 10-fold dilution) is given in Table 4. The database used in React contains all of the normal components used in geochemical computations, with the exception of oxalate. Oxalate values were added to the database (Pearson et al., 1992) because it is known to be a powerful actinide complexing agent. Speciation diagrams for U(VI), Pu(VI), Np(V) and Am(III) were generated using the React code and modified database (see Appendix A). Four scenarios were tested for each actinide whereby carbonate and phosphate were variably added to the fluid (see discussion below for a detailed explanation). In addition to the normal chemical designations the various figures contain the abbreviation "ox," designating the presence of oxalate. We stress that these calculations and accompanying figures are based on our best-estimates of tank fluid chemistry and grout-getter-fluid interactions. Many of these calculations may require modification as experimental data is accumulated.

Table 4.
Trial Fluid Species and Concentrations

Species	Concentration (molality)
NO_3^-	0.51*
Al^{3+}	0.006
Na^+	0.73#
SiO_2 (aq)	9×10^{-6}
Oxalate	0.042
HCO_3^-/CO_3^{2-}	0.10

*Incorporates NO_3^- and NO_2^-
#Adjusted slightly for charge balance

6.5.2. Baseline Solubility Calculations

This set of calculations was performed by adding 10 grams of each actinide hydroxide to one liter of fluid (Table 5). The pH was adjusted to span a range from pH 6 to pH 13 (for both Pu and U valences there is a general decrease in concentration as the pH rises). Oxalate complexes are only important in the pH 6-8 range. Above pH 8 the carbonate complexes account for most of the dissolved concentrations. Neptunium solubilities are universally high due to carbonate complexing above pH 9 and oxalate complexes below pH 9.

Comparisons with Table 2 indicate that U in the tank fluids is in excess of the saturation limit, however if the pH were slightly lower than 13, U(VI) might well be in equilibrium with the fluid, though the solubility curve is so steep in this situation that it is difficult to arrive at more than a tentative conclusion. The tank fluids (and thus the concentration of the complexing carbonate) were hypothetically diluted by a factor of 10 for this calculation, dropping the radionuclide concentration ranges from ~10^{-6} to ~10^{-7} M. At these concentrations it appears that both Pu and Am may be in approximate equilibrium with a solid phase in the tank, whereas Np is clearly undersaturated by a great amount and no solubility limit is likely to play a role for this radionuclide.

Table 5.
Predicted Solubilities in Reference Waste Fluid*

Species	pH 6	pH 11	pH 13
Pu(VI)	-2.4	-6.2	-8.0
Pu(IV)	-2.4	-6.2	-8.0
U(VI)	-1.8	-2.0	-7.5
U(IV)	-7.1	-7.1	-7.1
Np(V)	0.0	-2.0	-3.0
Am(III)	-2.0	-7.4	-8.4

*Values reported as $\log_{10}$.

6.5.3. Consequences of Grouting

Grouting the waste has many chemical as well as mechanical benefits. Grouting will greatly decrease the availability of carbonate in the fluids because calcite ($CaCO_3$) will precipitate when portlandite ($Ca(OH)_2$) in the grout mixes with carbonate in the waste. To model this process identical simulations were run in the absence of carbonate (Table 6). Note first that many of pH 6 solubilities did not change significantly because oxalate complexes predominate in this range. However, Ca oxalate is a relatively insoluble compound (7×10^{-5} moles per liter). In the "real world" adding a source of Ca would also remove this component and diminish the complexation of radionuclides. The effect of carbonate addition to the predicted concentration of actinide species is shown graphically in Appendix A. For the moment, however, an approximate estimate of the importance of removing oxalate can be obtained by noting the concentration of the most abundant complex that does not involve oxalate.

Table 6.
Predicted Solubilities Following Grouting*

Species	pH 6	pH 11	pH 13
Pu(VI)	-2.4 (-7)	-8.8	-11.6
Pu(IV)	-2.4 (-5.5)	-7.2	-9.2
U(VI)	-1.8 (-2.1)	-13.5	-17
U(IV)	-7.1 (-11)	-7.1	-7.1
Np(V)	0.0 (-4.0)	-5.0 (-7)	-7.0
Am(III)	-2.0 (-2)	-8.4	-8.4

*Values reported as $\log_{10}$. ( ) approximate value in the absence of oxalate, only shown for large differences.

6.5.4. Consequences of Phosphate Addition

Hydroxyapatite is listed as a potential getter for several of the actinides in Table 1, indicating that the addition of phosphate to the waste might also have potential benefits. Within the context of the React program, this would be reflected as precipitation of insoluble actinide-containing phosphates. The models were run with a fluid containing 0.1 M phosphate.

Unfortunately, for Np, Pu, and U, phosphate is better at forming aqueous complexes than at precipitating low solubility phases, particularly near pH 13 (Table 7). Fortunately, the equilibrium of portlandite, hydroxyapatite and calcite leave negligible concentrations of both carbonate and phosphate in the dissolved phase and therefore not precluding the use of hydroxyapatite getters.

6.5.5. Summary

Modeling suggests that significant solubility decreases for the actinides can be achieved by grouting the wastes. However, adding soluble phosphate to precipitate insoluble actinide phosphates provides no added benefit.

Table 7.
Predicted Solubilities Following Phosphate Addition*

Species	pH 6	pH 11	pH 13
Pu(VI)	-1.7	-8.5	-10.8
Pu(IV)	-1.7	-7.5	-9.4
U(VI)	-1..5	-1.4	-1.5
U(IV)	-1.3	-2.7	-2.7
Np(V)	-1.8 (-4)	-3.5	-7.0
Am(III)	-1.8	-8.5	-8.4

*Values reported as $\log_{10}$. ( ) approximate value in the absence of oxalate, only shown for large differences.

6.6. Getter Addition

6.6.1. Hydroxyapatite

The prospects for using hydroxyapatite as a getter are more encouraging. Experiments were performed for the Waste Isolation Pilot Project (WIPP) in which 2 grams of hydroxyapatite was added to 250 ml of brine containing 1.7×10^{-7} M Am and 1.1×10^{-7} M Pu.

Significantly, the actinide concentrations in these studies were about a factor of 10 below that in the actual waste. Thus, to achieve a similar decontamination factor in tank wastes requires 20 g of hydroxyapatite per 250 ml of tank fluid, which is not a prohibitive quantity.

Significant differences in the degree of complexing in WIPP brines as compared to the tank fluids may exist. We argue, however, that the oxalate, phosphate and carbonate already present in the tank fluids will be tied up in solid phases when grout is added. In this situation the brine will principally be a $NaNO_3$ fluid which should provide a matrix.

A final consideration concerns the pH difference between the WIPP brine and AX tank pore fluids. The Kd for Pu decreases by roughly a factor of 3 for each increase of two pH units. Thus, between a pH of 9 and 13 the Kd falls by about a factor of 10. Hydroxyapatite could decrease the dissolved concentrations of Pu by a factor of 100 if the waste were just grouted and no getters were added. The pH dependence for Am is less clear, but even a tenfold drop results in a Kd of about $10^{+2.5}$. The same tests failed to find significant U sorption on hydroxyapatite, probably because carbonate complexes had formed. Thus, it is likely that hydroxyapatite will not be a good getter for uranium. From the preliminary data we observed that Np is sorbed about 10 times better than Pu. Finally, allowing both the Pu and Np to equilibrate for several days resulted in significantly higher Kd values. Thus, the values stated here are probably conservative estimates.

6.6.2. Magnetite, etc.

Presently, the best way of reducing Tc concentrations in solution is to reduce the TcO_4^- to the relatively insoluble $TcO_2.nH_2O$. This process has been reported in considerable detail with the following results:

a. Ferrous salts in solution generally do a poor job at removing pertechnetate from solution. A solid reducing agent is needed to simultaneously supply all three electrons required for the process (Chu et al., 1996).

b. Magnetite works well only in reducing environments, and its performance is further enhanced if the solution also contains some ferrous iron that can sorb onto the mineral surface synchronously with pertechnetate. Vandergraaf et al. (1984) reported >95% Tc removal from a mix of 10^{-4} M Tc, 1 g of magnetite and 10 g of synthetic ground water under reducing conditions. With 1 g of powdered magnetite in 7 ml of fluid, the addition of 6 ppm Fe^{+2} results in more than 95% removal of the Tc in 48 hours. The pH was about 8 for these tests and the initial Tc concentration was 10^{-6} M. In the absence of Fe^{+2} only 16% was removed (Byegard et al., 1992).

c. The rate of TcO_4-uptake onto magnetite falls off markedly as the pH increases, and appears to all but stop above a pH of 9.5 (Chu and Eriksen, 1996).

d. Metallic iron works effectively even in contact with atmospheric oxygen (Bostik et al., 1990).

e. Metallic iron is not stable over "geologic" time in subsurface environments but magnetite is.

f. Tc(IV) forms a number of aqueous complexes which, in theory, can elevate the solubility significantly. In carbonate-free systems above pH ~10, formation of $TcO(OH)_3^-$ enhances $TcO_2.nH_2O$ solubility. Below this value the dissolved Tc concentration is about $10^{-8.2}$ M: At pH values of 12 and 13 this complex increases Tc solubility to 10^{-7} and 10^{-6} M respectively. Tc(IV) - carbonate complexes also form. $Tc(OH)_2CO_3(aq)$ predominates below pH of 8, while above this, $Tc(OH)_3CO_3^-$ dominates. In carbonate-rich environments both complexes can significantly increase total dissolved Tc concentrations (Eriksen et al., 1992)

g. Once Tc is immobilized by reduction, its release from a grouted waste form with an inherently reducing matrix is exceedingly slow. (Bostik et al., 1988; Tallent et al., 1988; Brodda and Mingxia, 1989).

h. 100 ppm dissolved phosphate also interferes with the sorption of Tc on magnetite. (Vandergaaf et al., 1984)

The best current estimate of the lowest solubility obtainable by reducing pertechnetate is $\sim 10^{-8}$ M. Magnetite (and by proxy metallic iron metal) is well documented to have significant capacity for reducing Tc. Thus, if getters were added in the amount of 10% of the waste volume, all of the Tc would be consumed. This is a feasible concept because neither the cost of the material nor the amount to be emplaced are large. However, it would be prudent to add both metallic iron, for speed, and magnetite, for longevity.

There are two potential problems with this concept. The first is that grout pore fluids have a pH in the range 12 to 13. In this environment formation of $TcO(OH)_3^-$ is possible. Formation of carbonate and phosphate complexes in a grouted waste form should not be problematic for reasons outlined previously. Thus, the solubility of $TcO_2.nH_2O$ in a cement pore fluid could be almost the same as it was in the diluted waste fluid prior to reduction. A second concern regards the time period over which the waste will remain reducing enough to sequester Tc in the tetravalent form. A quantitative model for oxygen entry into a grouted waste was proposed by Smith and Walton (1993). This will eventually be applied to our site-specific circumstances to assess whether reducing conditions could persist long enough to significantly reduce Tc releases from a decommissioned tank. The current literature does not resolve whether this is a realistic possibility, and therefore Tc is omitted from the following models.

6.6.3. Summary

For the Pu, Am, and Np addition of a hydroxyapatite getter appears most promising and should lower the aqueous solubilities by about a factor of 100 below that expected from just the grouting. For Tc additional work needs to be done before the issue can be resolved. However, it is likely that addition of magnetite plus iron metal can result in solubilities between 10^{-6} to 10^{-8}

M.

6.7. Radionuclide Release Models

From the preceding discussion we now provide a relative measure of radionuclide releases for various tank closure scenarios. From a knowledge of the tank inventory, radionuclide solubilities, retardation factors in the presence of added getters, and water flux, it is possible to predict first-order radionuclide release rates and the time required to completely remove specific radionuclides from a tank. For simplicity we assume fluid accumulation at the base of a tank to be zero, however future models will undoubtedly accommodate more complex hydrologic parameters. Ultimately, the release rates may serve as inputs for traditional performance assessment packages that evaluate the transport and impact of radionuclide releases into the far field.

The residual wastes in the tank following 90% retrieval are predicted to be 26.5 m^3 sludge and 75.44 m^3 saltcake (Ramsower, 1997). No estimates of fluid porosity are provided, however we conservatively assume the sludge contains 50% pore fluids and the saltcake 0%. Our decision to ignore the fluid contribution from the saltcake layer stems from a lack of data regarding the leachability of the saltcake layer. This problem may eventually be mitigated by leach tests or by 99% waste retrieval, whereby 100% of the saltcake layer is predicted to be removed (Ramsower, 1997). Table 8 summarizes the calculations described in detail below.

In the baseline scenario, no chemical getters are applied and the tank fill material consists entirely of a typical Hanford soil (Fayer and Walters, 1995). We bound the ground water infiltration effects by using the cases of (1) no cover but native soil backfill and (2) a RCRA cover and a native soil and/or cementitous backfill. Backfill hydrologic properties can be measured, but the RCRA cover properties are design specific assumptions. These assumptions are made only to limit the range of hydrologic properties. Hanford, INEEL and SRS, for example, have proposed designs for 1000 year covers, hence we do not assess the impact of covers for periods in excess of this time duration.

In the absence of a RCRA cover and at an infiltration rate of 1.8×10^{-2} cm/day (Fayer and Walters, 1995) approximately 27,000 liters of ground water could enter a tank annually. This is sufficient to fill a tank to a depth of ~7 cm per year. In the presence of a RCRA-equivalent barrier, ground water infiltration is reduced to 9.5×10^{-6} cm/day (Fayer and Walters, 1995), which is sufficient to fill a tank to a depth of 3.5×10^{-2} cm per year. The time required to

Table 8.
Predicted Actinide Removal Times

	Treatment			*Moles in Solution*[1]		*Removal Time (y)*	
Scenario	*Backfill*	*Getters*	*Radionuclide*	*Initial*	*After Treating*	*No Cap*	*Cap*
I	soil	none	U, Pu, Np, Am	Table 2	NA	0.49	931
II	soil	added	U	8×10^{-1}	see Appendix B for Kd's	3.7×10^{2}	see text for explanation
	soil	added	Pu	9×10^{-2}		3.7×10^{3}	
	soil	added	Np	4×10^{-2}		2.2×10^{4}	
	soil	added	Am	5×10^{-5}		1.9×10^{4}	
III[2]	grout	none	U	8×10^{-1}	1×10^{-13}	0.49	931
	grout	none	Pu	9×10^{-2}	3×10^{-8}	0.49	931
	grout	none	Np	4×10^{-2}	1×10^{-3}	0.49	931
	grout	none	Am	5×10^{-5}	5×10^{-5}	0.49	931
IV[3]	grout	none	U, Pu, Np, Am	Table 2	Table 6	10^{5}-10^{7}	10^{5}-10^{7}

NA: Not applicable.

[1] Assumes 13,300 L fluid in sludge and fluid pH of 13.

[2] Assumes cracked grout. Infiltration rate equals that of soil backfill.

[3] Non-fractured grout. Assumes diffusion-controlled mechanism. Estimated removal times based on initial flux only.

completely displace the pore fluid in the residual sludge is approximately 179 days in the absence of a RCRA-equivalent barrier. The placement of the RCRA-equivalent barrier increases the displacement period to approximately 931 years.

For the second scenario a layer of wicking material (fly ash, dry grout, etc.) containing getters is placed onto the waste and backfilled with soil. Here, mineral Kd's define the retardation times for each radionuclide (Appendix B). The addition of hydroxyapatite to the getter inventory will effectively retard neptunium and americium. Kd's on the order of 5-6 x 10^3 for Np and Am (pH corrected) will increase the time required for complete removal to between 19,000 and 21,000 years in the absence of a RCRA-equivalent barrier. Because of these long durations an assessment with RCRA barriers was not performed. Uranium and plutonium have lower Kd's (100 and 1000, respectively) and therefore should flush out earlier. The time required for complete removal is estimated to be about 400 years for uranium and 4,000 years for plutonium in the absence of a RCRA-equivalent barrier. Again, assessments were not made for situations with RCRA barriers.

The third scenario examines the influence of a tank filled with cracked grout, where the grout effectively lowers the solubility of some radionuclides according to the React calculations. With a grid of cracks spaced at 1 m intervals and with apertures of 0.001 cm or greater, it was found that the grouted plug would transfer a volume of fluid equivalent to that of the soil column (see Section 6.2). However, the storage capacity of the tank (prior to having incurred a leak) would be much diminished. However, by virtue of the chemical environment imposed by the grout, the solubility of the radionuclides would be diminished from the values in Table 5 (diluted by 10) to those in Table 6. In this scenario although sorption is not the primary retardation mechanism, once a particular radionuclide is precipitated it is effectively removed from fluid transport. Americium solubility is unaffected by grouting, and will therefore remain in the fluid phase in the absence of getters, and should be flushed out of the waste in the time periods calculated in the first scenario. Uranium(VI) displays a sizable solubility drop of ~3 x 10^6 M following interaction with grout, and therefore becomes chemically fixed in the presence of grout-related fluids and high pH. A large drop in pH would dramatically increase U(VI) solubility, however the presence of a large cementitous mass should effectively buffer the pH. The solubility of neptunium and plutonium is calculated to lessen by a factor of between 10^3 to 10^4 M in the presence of grout. In this case, addition of getters (specifically hydroxyapatite) will reduce the solubility of Np, Pu and Am by an additional factor of 100.

In this scenario it is also relevant to question whether the leaching of radionuclides from the grouted waste would impose a significant barrier. Leaching would involve diffusion from within the grouted mass to the fluid channels. The penetration depth for the leach front is approximated by:

$$x(cm) = 2(DT)^{\frac{1}{2}}$$

With a diffusion coefficient of 10^{-8} cm^2/sec, about 1 cm of penetration is expected in the first year, and 10 cm after 100 years. It unlikely that grouting the waste will produce a single solid mass of concrete. Thus, the sizes of the individual pieces in the grouted waste will probably not be more than a 10 to 20 cm. Thus, on the time scale of relevance, leach rates will not provide a significant barrier to radionuclide releases. In short, in this scenario the flux of water is not decreased but actinide solubilities are considerably diminished.

The fourth scenario examines a tank containing an intact plug of low-permeability grout. As crack apertures fall below 10^{-3} cm, the flux of water down through the grouted top of the tank approaches zero and diffusion out of the tank bottom becomes the dominant release mechanism. If the cementitous skirt extends a meter beyond the bottom of the tank then this defines the path length, L. The release rate then is defined by the equation:

$$Flux\left(\frac{moles}{cm^2 s}\right) = D\frac{C_1 - C_2}{L}$$

where again D is less than 10^{-8} cm^2/sec and the concentration at the bottom of the skirt is assumed to be almost zero. C_1, the radionuclide concentration in the tank, is fixed by the in-tank chemistry (grout ± getters). This scenario represents the slowest release mechanism. Assuming the largest concentration gradient (where $C_2 = 0$), calculated fluxes are exceptionally low, ranging from 10^{-20} to 10^{-30} mol/cm^2 s for the initial fluxes (Appendix B). These fluxes represent maximum values because flux as a function of time will vary according to reservoir concentration (which will continuously be falling) and the diffusion coefficient, which likely will vary over time. The calculations required to determine times required for complete removal of radionuclides in simple diffusion settings are underway, however we estimate these times will be on the order of 10^5 to 10^7 y.

7. CONCLUSIONS

Even in the highly simplified context of the models derived for this report it is evident that a wide range of near field source terms can be justified. Each of these scenarios involves differences in cost and worker exposure. However, by applying these different release models as input to a standard performance assessment package it should be possible to determine what level of expenditure is technically justified in decommissioning each tank.

8. ACKNOWLEDGMENTS

Hank Westrich and Larry Bustard (both at Sandia) provided critical reviews of early versions of the report. Tom Burford (Sandia), Sam Bendamere (Denver Grouting, Inc.) and Steven Phillips (AGEC, Inc.) supplied us with useful information and contacts regarding cement grout emplacement technologies, grout formulations and tank filling advice. Thanks to Ernst Ahrens (Sandia) for discussions about tank filler strategies and the use of micro-fine cement grouts. We also thank Dave Becker (Hanford) for technical discussions and Ed Fredenburg (Hanford) for programmatic advice, the combination of which helped establish the guidelines for this work, and clarified objectives for future research within the Tank Focus Area.

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10. APPENDIX A. ACTINIDE SOLUBILITY PLOTS

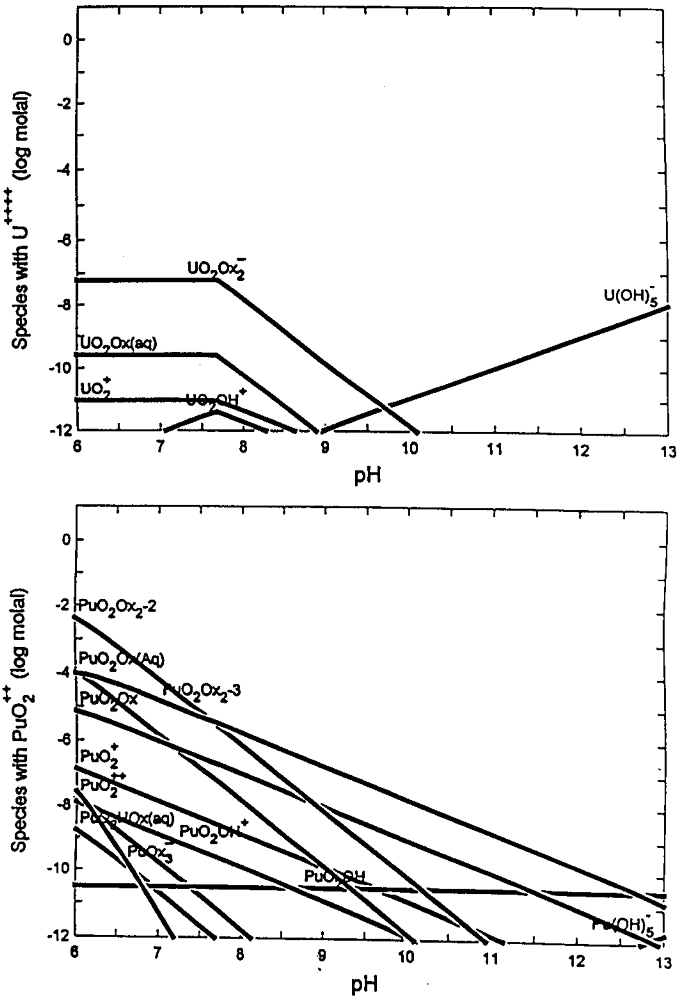


Figure A1. Speciation of U(VI) and Pu(VI) in a tank waste fluid with no carbonate or phosphate added (see Table 2 and Table 5 for trial fluid composition).

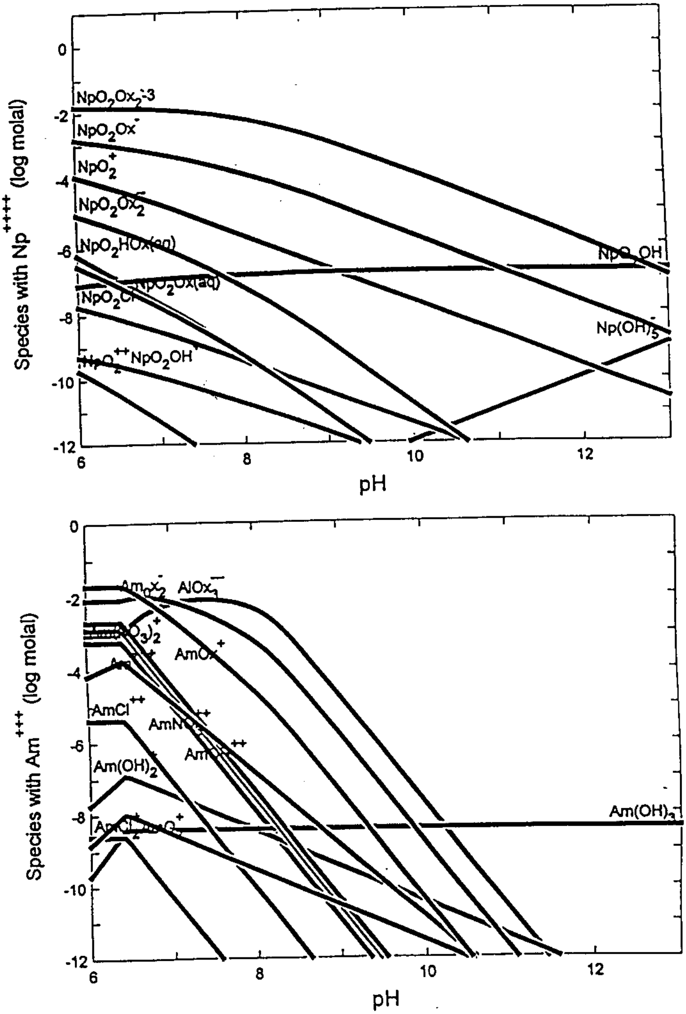


Figure A2. Speciation of Np(V) and Am(III) in a tank waste fluid with no carbonate or phosphate added (see Table 2 and Table 5 for trial fluid composition).

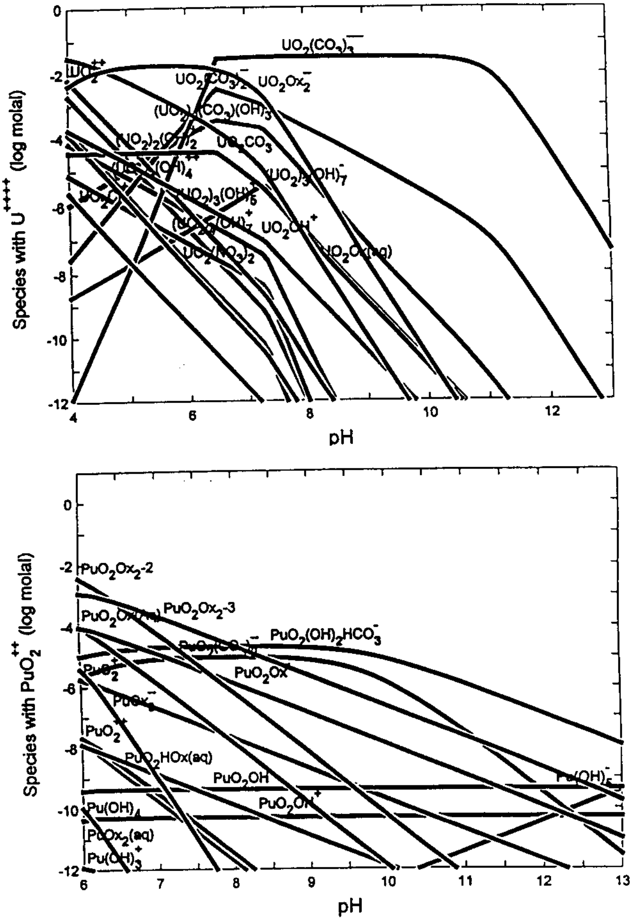


Figure A3. Speciation of U(VI) and Pu(VI) in a tank waste fluid with carbonate added but no phosphate (see Table 2 and Table 5 for trial fluid composition).

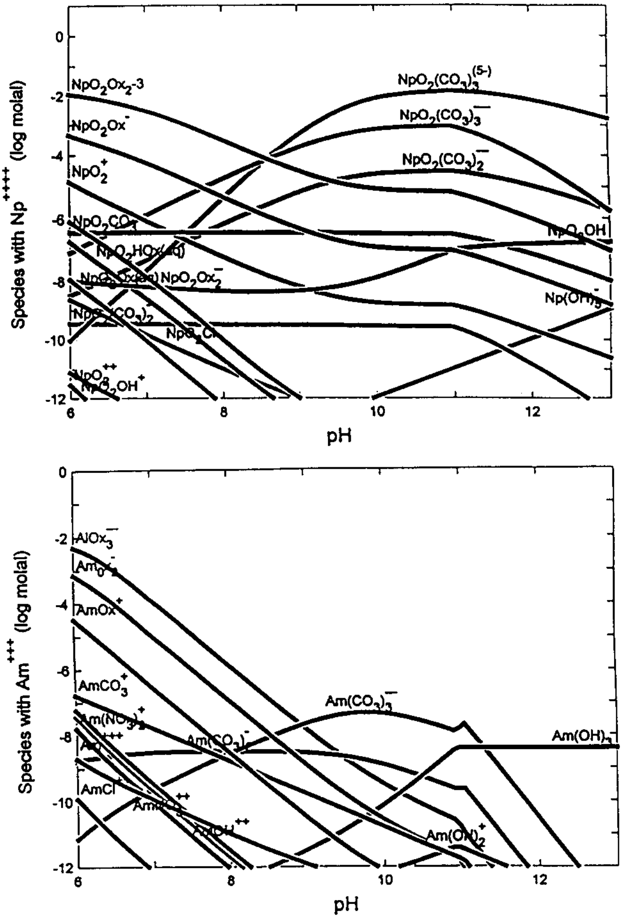


Figure A4. Speciation of Np(V) and Am(III) in a tank waste fluid with carbonate added but no phosphate (see Table 2 and Table 5 for trial fluid composition).

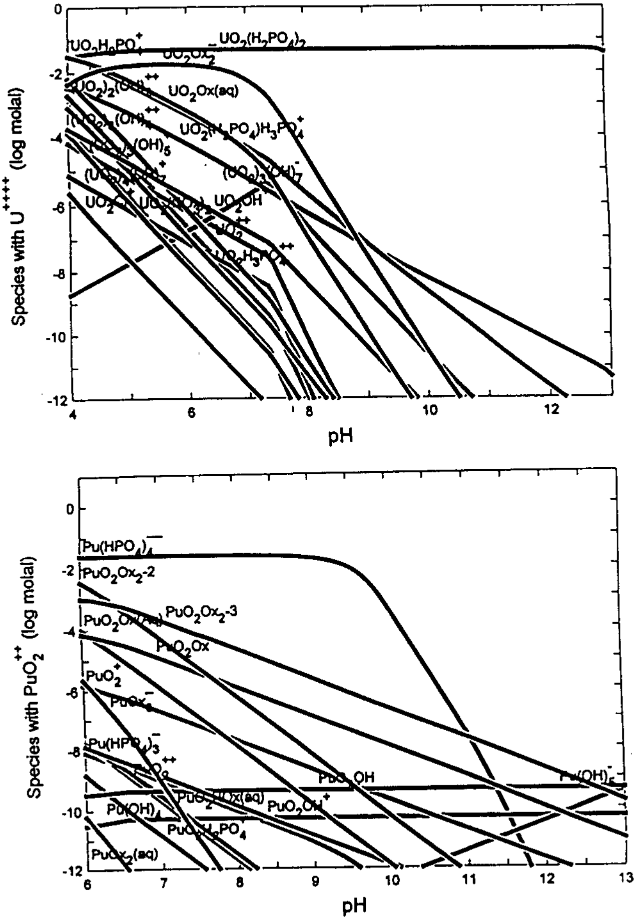


Figure A5. Speciation of U(VI) and Pu(VI) in a tank waste fluid with phosphate added but no carbonate (see Table 2 and Table 5 for trial fluid composition).

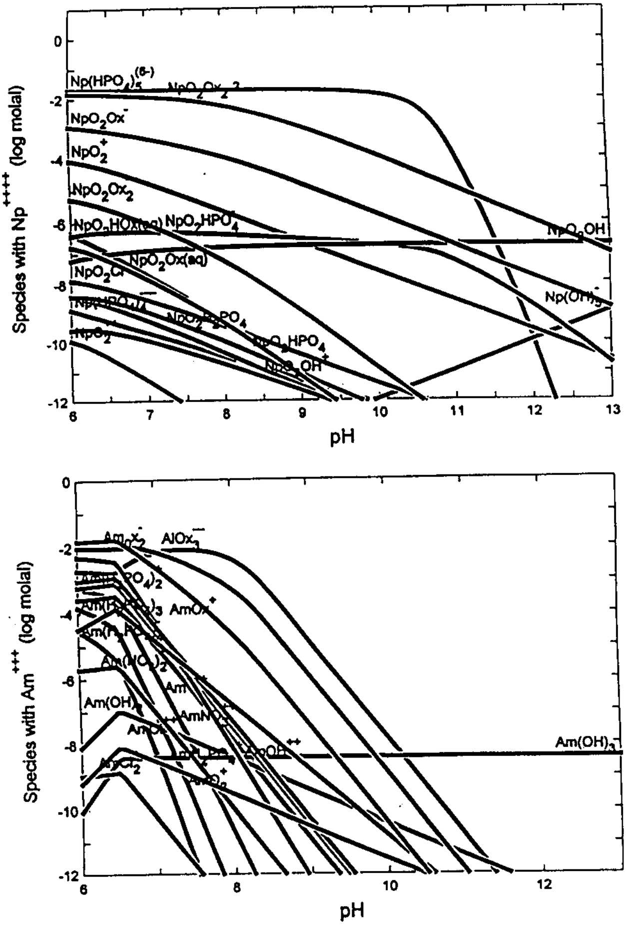


Figure A6. Speciation of Np(V) and Am(III) in a tank waste fluid with phosphate added but no carbonate (see Table 2 and Table 5 for trial fluid composition).

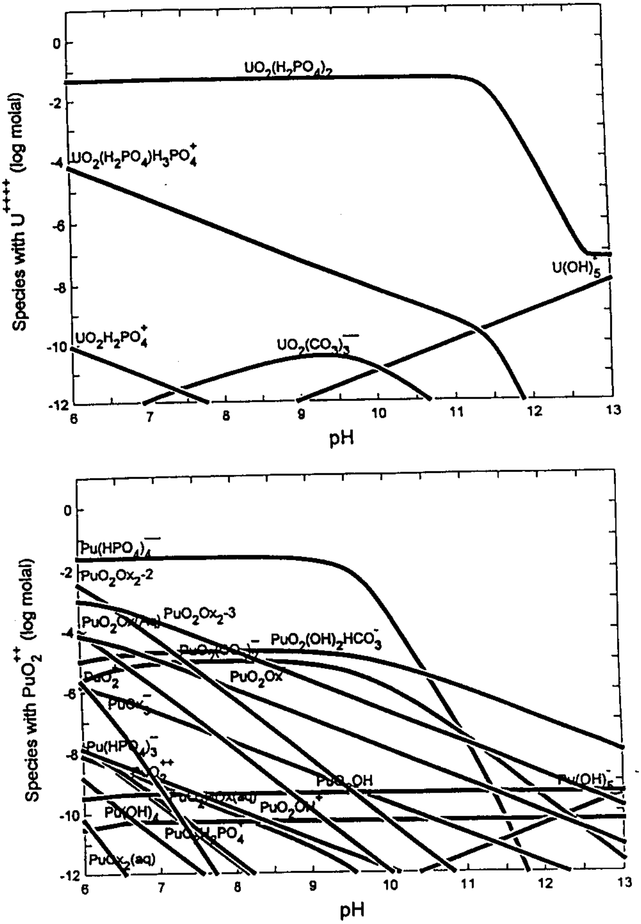


Figure A7. Speciation of U(VI) and Pu(VI) in a tank waste fluid with both phosphate and carbonate added (see Table 2 and Table 5 for trial fluid composition).

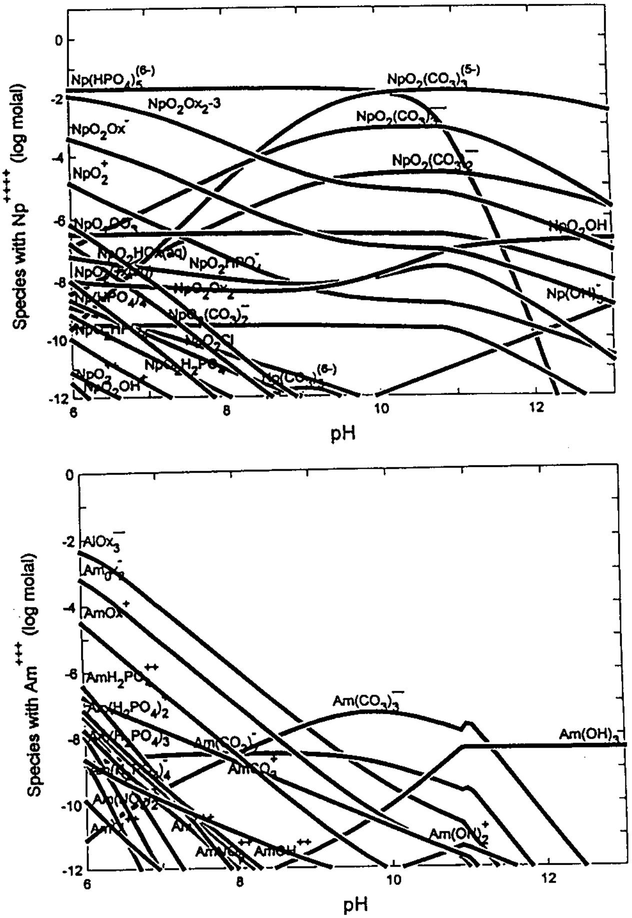


Figure A8. Speciation of Np(V) and Am(III) in a tank waste fluid with phosphate and carbonate added (see Table 2 and Table 5 for trial fluid composition).

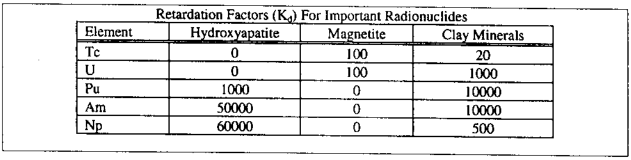
11. APPENDIX B. RADIOACTIVE RELEASE MODELS SPREADSHEET

Scenario I.

Tank filled with soil. No getters. Calculate time required to displace sludge fluid (assuming no ponding).

		RCRA Cap	
		No	Yes
Tank radius	(cm)	1143	1143
Tank area	(m^2)	410	410
Infiltration rate v(f)	(mm/day)	1.8E-01	9.5E-05
	(cm/day)	1.8E-02	9.5E-06
	(m^3/day)	7.4E-02	3.9E-05
Waste vol.	(cm^3)	2.7E+07	2.7E+07
Fluid vol.	(m^3)	13.3	13.3
Displ. T/ fluid vol	(days)	179	339993
Displ. T/ fluid vol	(yrs)	0.49	931

Assumptions		
Sludge height	cm	6.5
Sludge vol. after 90% removal	(m^3)	26.5
Sludge porosity (σ)	(%)	0.5
Soil density (σ)	(g/cm^3)	1.96
Soil porosity (θ)	(%)	0.26
σ/θ		7.5



Retardation Factors (K_d) For Important Radionuclides

Element	Hydroxyapatite	Magnetite	Clay Minerals
Tc	0	100	20
U	0	100	1000
Pu	1000	0	10000
Am	50000	0	10000
Np	60000	0	500

Scenario II.

Tank filled with soil. Getters added. Calculate time required to displace radionuclides (assumes no ponding) from the following expression:

$$\boxed{\frac{v(i)}{v(f)} = \frac{1}{1 + \frac{\sigma}{\theta} K_D}}$$

where v(i) is the rate of radionuclide transport and v(f) is the fluid flow rate.

Note that Kd's for americium and neptunium have been adjusted down by a factor of 10 to accommodate the expected drop in retardation at pH 13.

Americium K_D	**5000**	RCRA Cap	
		No	Yes
Infiltration rate v(f)	(mm/day)	1.8E-01	9.5E-05
Radionuclide rate v(i)	(mm/day)	4.8E-06	2.5E-09
Time to remove	(days)	6.8E+06	1.3E+10
Time to remove	(yrs)	1.9E+04	3.5E+07

Neptunium K_D	**6000**	RCRA Cap	
		No	Yes
Infiltration rate v(f)	(mm/day)	1.8E-01	9.5E-05
Radionuclide rate v(i)	(mm/day)	4.0E-06	2.1E-09
Time to remove	(days)	8.1E+06	1.5E+10
Time to remove	(yrs)	2.2E+04	4.2E+07

Plutonium K_D	**1000**	RCRA Cap	
		No	Yes
Infiltration rate v(f)	(mm/day)	1.8E-01	9.5E-05
Radionuclide rate v(i)	(mm/day)	2.4E-05	1.3E-08
Time to remove	(days)	1.4E+06	2.6E+09
Time to remove	(yrs)	3.7E+03	7.0E+06

Uranium K_D	**100**	RCRA Cap	
		No	Yes
Infiltration rate v(f)	(mm/day)	1.8E-01	9.5E-05
Radionuclide rate v(i)	(mm/day)	2.4E-04	1.3E-07
Time to remove	(days)	1.4E+05	2.6E+08
Time to remove	(yrs)	3.7E+02	7.0E+05

Scenario III.

Tank filled with grout. No getters added. Assume 1 m crack grid with apertures >0.001 cm.

Radionuclide solubility at pH 13 (from React)

	Grouting			
	Prior	After	Ratio	mol in sol[n]
Pu	7.0E-06	2.5E-12	2.8E+06	3.3E-08
U	6.0E-03	1.0E-17	6E+14	1.3E-13
Np	3.0E-06	1.0E-07	30	1.3E-03
Am	4.0E-06	4.0E-09	1005	5.3E-05

Scenario IV.

Tank filled with grout. Assume 1 m crack grid with apertures <0.001 cm. Assume 1 m thick skirt

Diffusion coef. (D)	cm^2/s	1.E-08	
Calculated flux	mol/cm^2 s	2.5E-25	Pu
		1.0E-30	U
		1.0E-20	Np
		4.0E-22	Am

Flux = $D(C_1-C_2)/L$

C_1 = radionuclide conc. in tank after grouting

C_2 = radionuclide conc. at bottom of skirt (assumed zero)

L = path length, defined as 100 cm

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Project Title/Work Order HTI - Stabilization of In-Tank Residual Wastes and External-Tank Soil Contamination for the Tank Focus Area, Hanford Tanks Initiative: Applications to the AX Tank Farm HNF-SD-HTI-ES-004, Rev. 1		EDT No. N/A
		ECN No. 644554

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