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HIGH-LEVEL WASTE IMMOBILIZATION

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ABSTRACT

Analysis of risks, environmental effects, process feasibility, and costs for disposal of immobilized high-level wastes in geologic repositories indicates that the disposal system safety has a low sensitivity to the choice of the waste disposal form.

WASTE FORM SELECTION

For the past three years, the United States program for the immobilization of high-level nuclear reprocessing wastes has focused primarily on two items, the choice of a waste form and the design of the first U.S. waste forms plant, the DWPF at Savannah River. Over the same period, a general consensus has been reached in the overall U.S. nuclear waste program that the preferred method for disposing of high-level wastes is multibarrier isolation of high-integrity immobilization forms in an engineered repository. (However, since I'm at Hanford, I'd better inject a quick aside that there might be exceptions to this overall approach. That is, in-place engineered isolation may be a better option for some of the historical wastes, such as the low-activity waste from the original Hanford operations.)

We will be hearing detailed talks on waste form development and comparison in Session 8, on DWPF design in Session 11, and on multibarrier repository disposal in Session 16. What I want to do in this introductory talk is look at the high-level waste form selection in light of all this work, and present the thesis that this apparently difficult decision which has occupied us for so many years is really a straightforward one -- a sort of Lady and the Tiger choice where nature decided that tigers are an endangered species and substituted a tabby cat. I will be drawing most of my examples from the DWPF, where the waste form decision is currently being made, but will try to extend them to the general high-level waste form problem. Most of the studies referred to are being issued by the original authors as background to the DWPF waste form selection.

Let me start very traditionally in Figure 1 with a diagram of the waste hazards versus time.[1] As most of you know, this sort of diagram has recently been complicated by proposed reductions in hazards in the uranium decay chain products, and proposed increases in hazards for some of the waste components such as neptunium. The older hazards listings are used in the ICRP-2 curves and the newer in the ICRP-30 curves. Unfortunately, the ICRP-30 curves eliminate the older contention that the waste hazards fall below uranium ore hazards in less than a thousand years, but they do it in a very nice way by reducing both the waste hazards and ore hazards below their previous levels. The general conclusions remain the same for either set of curves. For the first several years after reprocessing, hazards are dominated by short-lived radionuclides, for the next 300 years by Sr-90, and finally, at a hazard level several orders of magnitude lower, by actinides, uranium decay products, and very long-lived fission products. From the standpoint of the waste form decision, the key points are that the radionuclide hazards are always in the range of a variety of natural hazards and that, although they do extend over very long time periods, they are indeed close to uranium ore values after a few hundred years.

In the closely related data on waste heat production, essentially the same type curves apply, with Cs-137 plus Sr-90 being the chief intermediate term heat producers. Currently, Ce-144, Sr-90, Cs-137, and the miscellaneous, mainly short-lived radionuclides each produce roughly equal contributions to the SRP high-level waste heat load. When these wastes are ready for repository disposal, almost all the heat will be from Sr-90 and Cs-137 and will amount to about 2 megawatts. This heat load is equivalent to the waste heat load from reprocessing about 3000 metric tons of power reactor fuel and aging it similarly. Again, from the standpoint of the waste decision, these are not overwhelmingly large numbers.

After radionuclide hazards and heat production, the next most important data on the high-level wastes are their chemical compositions and radionuclide loading densities. These data are given in Table 1 for the existing U.S. high-level wastes at Hanford, Idaho, Savannah River, and West Valley,[2] as well as for possible future high-level wastes from U.S. power reactor fuels reprocessing. The values for the existing waste are after concentration by sludge-salt and radionuclide separation for the alkaline Hanford, Savannah River, and West Valley wastes, and by calcination for the acid Idaho wastes. Without concentration, the heat loadings are up to 30-fold smaller. The values for the future commercial waste are for possible direct production in concentrated form. Compositions for the existing waste all lead to very low heat loadings in the waste forms made from these wastes. However, potential heat loadings for the commercial waste forms can be as much as 100 times higher than for current waste forms.

As will be described in a subsequent paper by Ray Walton and Bruce Wilson, the National U.S. high-level waste forms program in 1981 narrowed the high-level waste forms choice from some seventeen forms to two, borosilicate glass and crystalline ceramics such as Synroc. These two forms were picked, not only because they rated highest on a combined properties-processability analysis, but because they represent very different material types. Table 2 describes borosilicate glass and Synroc-D forms for SRP high-level waste as they might be produced in the DWPF.[3] Aside from the differences in borosilicate glass and Synroc properties, a cardinal difference between the two forms is that Synroc has about a factor of three higher volumetric radionuclide loading due both to a higher allowed waste loading and a higher density.

The essence of the waste form decision is how the forms perform in the various disposal system steps of form preparation, interim storage, transport, repository emplacement, and repository disposal.

Simpler and well developed waste form production is the chief advantage of borosilicate glass. Figure 2 compares the manufacturing processes for borosilicate glass and Synroc in the DWPF.[4] Borosilicate glass preparation comes close to being a one-step process involving continuous melting of a waste/glass-frit slurry in a ceramic-lined melter and pouring the molten glass into the waste canisters. Synroc preparation is inherently a multistep process involving intimate mixing of the waste and Synroc additives, calcination, compact forming, compact hot pressing, and canisterization. However, there seems little doubt that the Synroc process, like the much better demonstrated borosilicate glass process, can be made to work, and some process simplifications will probably be possible.

As shown in Table 3, remote operation and maintenance, plus multistage containment and filtration in the DWPF, make the offsite risks and occupational hazards of production extremely small for either waste form,[5] although borosilicate glass has a slight advantage because of its simpler process and avoidance of dry powder handling. Overall then, the differences between manufacturing the two waste forms are in time and money, not in safety, or environmental effects. The same conclusion applies to almost any waste form preparation with the possible exception of very complex forms such as coated particles, for which feasibility is still in doubt.

Figure 3 shows an interim storage design for the DWPF.[6] Some such building will be required to provide convenient lag storage between the forms manufacturing plant and the repository in any disposal regime. For the DWPF, it may also have to accommodate the waste forms for several years between the DWPF startup and repository startup. The storage building itself provides protection against most outside events such as tornados, earthquakes, impacts, floods, and sabotage. It also provides passive, natural-convection air cooling with an air intake occupying almost all of one side of the building and an outlet on the other. The waste form needs only to supply contamination containment and backup protection against impacts, volatilization or attempts at dispersal by sabotage. As listed on the figure, the calculated risks for any of the forms, even over a number of years of interim storage, are very small.

Any of the high-level waste forms offer higher integrity in transport than spent fuel, which is already an accepted transport form. Borosilicate glass and Synroc canisters in particular back up the shipping cask by offering low external contamination, low production of dispersible particles in severe impacts, negligible radionuclide volatilization in accidental transport fires, and low leachability if exposed to water. Doses and risks from shipment of SRP waste forms are, therefore, almost exclusively a matter of transport cask design rather than of waste form choice. Synroc waste forms have the advantage in transport that their greater loading density reduces the number of casks to be transported by a factor of three. However, the total amount of radioactivity to be transported is the same in each case. Table 4 assumes both Synroc and borosilicate transport casks are designed to the same external radiation levels and gives Synroc an exposure advantage.[7] However, if the borosilicate casks were shielded identically to the Synroc casks, this advantage would disappear. Here again, therefore, any effects of the waste form decision are on costs, not on risks per se.

Essentially identical considerations apply to waste form emplacement in the repository. Borosilicate glass, Synroc, or any of the other waste forms under consideration have the necessary mechanical strength and accident resistance for emplacement. Only a third as many waste forms will have to be emplaced with Synroc as with borosilicate glass, but three times as much radioactivity will be involved in each placement, so radionuclide risks should be nearly constant during the repository loading phase.

The small differences in risks between the forms in the waste system steps leading to repository disposal, and straightforward analyses required to compare these risks, means that any real significance in the waste form decision must be in the disposal step itself. Here, two reciprocal questions are involved. How does the repository affect the waste form, and how does the waste form affect the repository?

Figure 4 shows a generic design for a salt repository.[8] The insert illustrates how the DWPF waste canisters might be emplaced in twin rows of holes in the floors of the repository rooms. The suggested spacings of about 2.3 meters center-to-center between rows and between holes are close to a minimum after an allowance is made for overpacks and backfills, and for mechanical clearances required for hole drilling and canister emplacement. With a total heat content of about 2 megawatts for the as-emplaced SRP wastes, each DWPF borosilicate glass canister would initially generate an average of 200 watts of heat, and each Synroc canister an average of 600 watts for repository area loadings of ~30 kilowatts/acre and ~90 kilowatts/acre, respectively. Because there will be variation between canisters, the borosilicate glass canister heat loadings were assumed to be ~310 watts for repository temperature calculations.

Results of these calculations are given in Figure 5.[9] They indicate that, under the assumed conditions, canister-repository interface temperatures should remain under 100°C, and that peak temperatures are obtained about 25 years after waste emplacement, dropping to ambient rock temperatures in the 20 to 40°C range in about 100 years. These temperatures were a basic input to the evaluation of the waste forms in the repositories. They apply fairly directly to any of the existing U.S. high-level waste. However, to apply them to possible high-activity commercial high-level waste, the commercial waste needs to be either diluted to existing waste levels, aged for 75-100 years outside the repository, or protected within the repository by impervious barriers until temperatures have dropped into the assumed ranges.

The other basic inputs to the waste form evaluations in the repositories are the assumption that leaching by groundwater is the most likely mechanism for transporting radionuclides from the repository to the human environment, plus the specification of the repository type and its groundwater composition. The most likely repository geologies are assumed to be salt, basalt, tuff, or granite.

Table 5 shows comparative data on waste form leaching for simulated SRP waste.[3] I would like to make four points from this table. First, the measured short term leach rates do not correspond to congruent dissolution of the forms, but vary element by element and differently from form to form, being higher for strontium in Synroc than in glass, but lower for uranium in Synroc than in glass, and about a stand off for cesium. Secondly, the leaching differences between better forms such as these tend to be only one or two orders of magnitude. Third, there are not large differences in the values with different leachants, distilled water being generally the most aggressive leachant. Fourth, the leaching rates generally increase about a factor of 4-10 in going from 40 to 90°C.

The next three figures are for the borosilicate glass waste forms and constitute a sort of sensitivity analysis on the leaching results in Table 5. Figure 6 shows that leach rates tend to fall off with time. In borosilicate glass, this effect is apparently largely due to layer formation on the glass surface; in Synroc it is perhaps more likely to be due to selective leaching of more soluble phases. Figure 7 shows that leaching can vary strongly with pH,[9] but the variation is fairly small over the expected range of repository conditions. This variation, however, does show the need for choosing the repository arrangements so as to prevent pH excursions into undesired ranges. Figure 8 shows that leaching tends to increase with increasing leachant flow rates.[9] Repositories will be chosen so that their groundwaters are at near static conditions, but there has been some concern that the waste forms should also protect against high-flow repository upsets. The risk analysis to be discussed shortly suggests that low leach rates are least meaningful under such repository upsets.

Based on the leaching data we have seen and on effective leaching surfaces for the DWPF forms of about five times the geometrical canister area, leached fractions for the DWPF borosilicate glass are put in the 10^{-5} per year range, and for DWPF Synroc in the 10^{-6} per year range.

These leaching values were compared against parametric repository risk analysis calculations, performed by Pacific Northwest Laboratory and by Lawrence Livermore National Laboratory, to determine whether such leaching rates lead to acceptable risks, and whether there is incentive to work towards still lower leach rates. Typical LLNL risk analysis results are shown in Figure 9. These results indicate that the risks tend to be much more dependent on repository parameters than leach rates for leach rates in the range 10^{-3} to 10^{-6} per year.[10] Although risks are calculated to diminish at leach rates less than 10^{-6} per year, they are already very low, i.e., they are calculated to offer less than one chance of a cancer death in a million years.

Another test of the sensitivity of the waste form decision is how closely it affects meeting the draft criteria and regulations proposed by EPA and NRC for high-level waste disposal as embodied in those agencies' drafts of 40 CFR 191 and 10 CFR 60. This question is addressed in Table 6.[11] NRC puts a few direct criteria on the waste form itself, requiring that it not be liquid, dispersible, combustible, pyrophoric, explosive, chemically toxic, or a criticality hazard. Essentially any of the waste forms examined by DOE meet these criteria. EPA puts system requirements on radionuclide effects from both waste form manufacture and waste form disposal. As shown earlier, DWPF radiation effects can be held to about 10^{-3} of the EPA requirements almost regardless of the waste form. Similarly, for either borosilicate glass or Synroc DWPF forms, "worst case" repository releases would be about 10^{-6} of the allowed EPA health effects. Even over a million years, the best-estimate health effects are still only about 10^{-2} of the EPA 10,000 year values. NRC puts its disposal system requirements in terms of package requirements, asking for zero radionuclide release from the package for the first thousand years, and 10^{-5} per year thereafter. The first requirement is taken to be largely an overpack requirement, but since NRC states that its main purpose is to isolate the waste form from the repository during a high-temperature phase, it should presumably apply only in the special case where high-heat waste forms are used in the repository. The second requirement of 10^{-5} per year leachability is also an overall package requirement; the package designers have placed a 10^{-4} per year requirement on the form alone, easily met by either borosilicate or Synroc.

In summary, selecting a waste form from those developed in DOE's alternative waste form program would meet all the proposed regulatory requirements and lead to very low risks at every stage of the waste disposal process. Further, developing a "best" form would lead to no practical reductions in risks. On these bases then, the decision, at least for the low-heat defense high-level wastes, is not crucial and can be made primarily on practicalities and costs.

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TABLE 1. U.S. High-Level Nuclear Wastes.

Location	Type	Megacuries	Curies/kg*
Hanford Plant	Alkaline Cs/Sr Sources	226 332	0.6 - 2 NA
Idaho Chemical Processing Plant	Acid Liquid Calcine	17 36	0.6 - 3
Savannah River Plant	Alkaline	699	2
West Valley Plant	Alkaline Acid	37 1.7	15 30
Spent LWR Fuel	Not Processed	10,400	175

* After decay to Sr-90 and Cs-137 and concentration for waste form loading.

TABLE 2. Reference DWPF Waste Forms.

Waste Form Characteristics	Borosilicate Glass	Synroc-D Ceramic
Canister dimensions, (d x ℓ), m	0.61 x 3.0	0.61 x 3.0
Radionuclide content, Ci*	150,000	450,000
Heat generation, initial, w*	423	1270
Heat generation, 1000 yr, w*	<1	<2
Leachability, parts per year	$\sim 10^{-5}$	$\sim 10^{-6}$
Fines generation (10 J/m^3 impact), %	0.14 - 0.18	0.16
Volatilization, 900°C, mg/hr	0.002	NA
Compressive strength, MPa	550	280

* Nominal maximum values; average values are half maximum values.

TABLE 3. Off-Site Radiation Exposures from DWPF Forms Production.

Source of Exposure	Borosilicate Glass	Synroc-D Ceramic
Routine Operation		
Maximum individual exposure, mr/yr	0.06	0.06
Maximum population exposure, mrem/yr	1.3	1.3
Accident Consequences		
Maximum individual exposure, mr/yr	0.006*	0.008**
Maximum population exposure, mrem/yr	0.7*	14.0*

* Slurry mix evaporator reaction with frequency of 0.03/yr.
** Calcine bin failure with frequency of 1×10^{-6}/yr.

TABLE 4. Radiation Exposures From DWPF Forms Shipment.*

	Borosilicate Glass	Synroc-D Ceramic
Normal operations, rail transport		
Population exposure, mrem/yr**	79	57
Normal operations, truck transport		
Population exposure, mrem/yr**	700	72
Accident risk, rail transport		
Maximum individual, mr/yr	0.0035	0.0035
Accident risk, truck transport		
Maximum individual, mr/yr	0.0061	0.0061

* Assumed transport of 3000 miles.

** Assumed route population of 1,125,000 receiving 112,500 mrem background dose.

TABLE 5. Leach Rates of DWPF Waste Forms.

	Leach Rates - 28 day MCC-1 Tests Composite SRP Waste - Unit $g/cm^2 \cdot day$					
	Borosilicate Glass			Synroc-D Ceramic		
	Cs	Sr	U	Cs	Sr	U
40°C Tests						
Deionized water	0.05	<0.001	0.01	0.4	0.087	<0.001
Silicate water	0.08	<0.001	0.04	0.2	0.033	<0.001
Brine	0.04	<0.001	<0.7	<0.1	<0.004	——
90°C Tests						
Deionized water	1.5	<0.001	0.05	0.9	0.28	<0.001
Silicate water	0.8	<0.001	0.2	0.4	0.079	<0.001
Brine	0.4	<0.001	<0.02	0.6	<0.1	<0.001

TABLE 6. Regulatory Requirements on High-Level Waste Forms

EPA Requirements — Draft 40 CFR 191 (unpublished)	
• Meet radiation guides in manufacture	• DWPF 10^{-3} below
• Control radionuclide release from repository so - as to produce <1000 premature deaths in 10,000 yrs. - from a 100,000 MTHM repository	• Either glass or ceramic forms calculated at 10^{-6} of EPA limit
NRC Requirements — Draft 10 CFR 60	
• Waste form should not be liquid, dispersible, combustible - Explosive, pyrophoric, chemical-toxic, criticality hazard	• Both glass and ceramic forms meet
• Waste package should not leach for 1000 years	• Overpack requirement
• After 1000 years package leachability $<10^{-5}$/yr	• 10^{-4} or better met by form alone

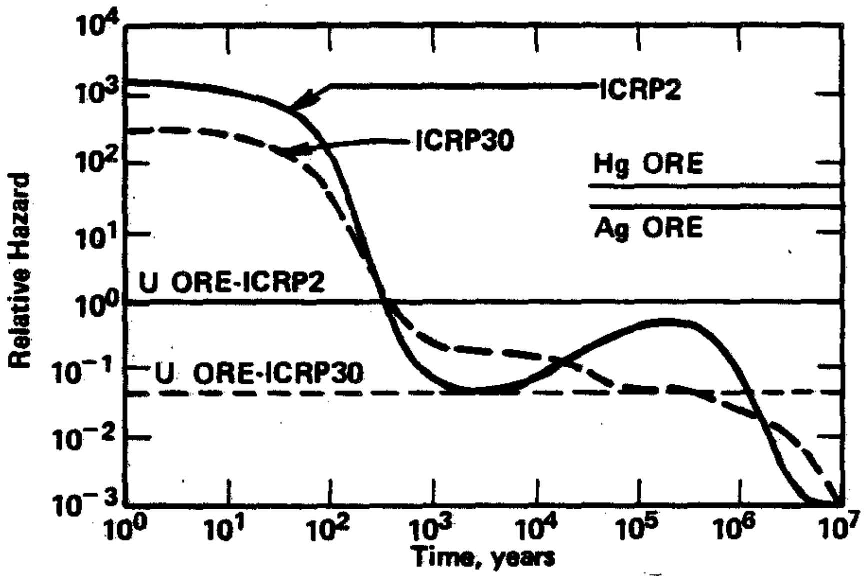
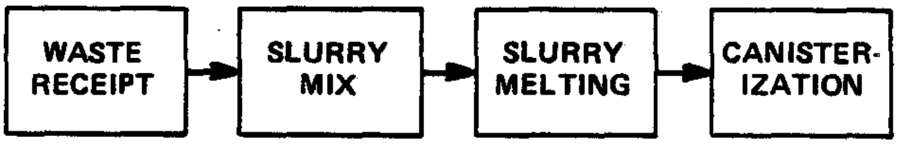


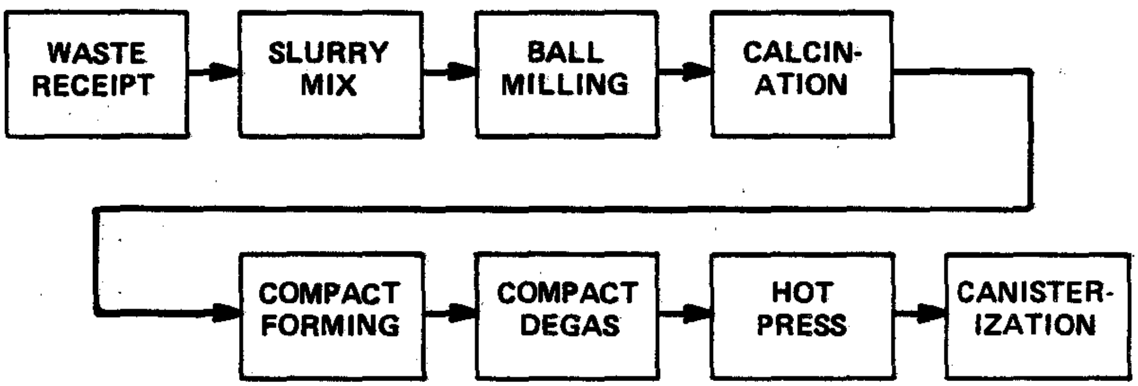
FIGURE 1. SRP High-Level Waste Hazard



BUILDING SIZE – 132′ x 430′

PROCESSABILITY RATING ON SCALE OF 100 = 83

BOROSILICATE GLASS PROCESS

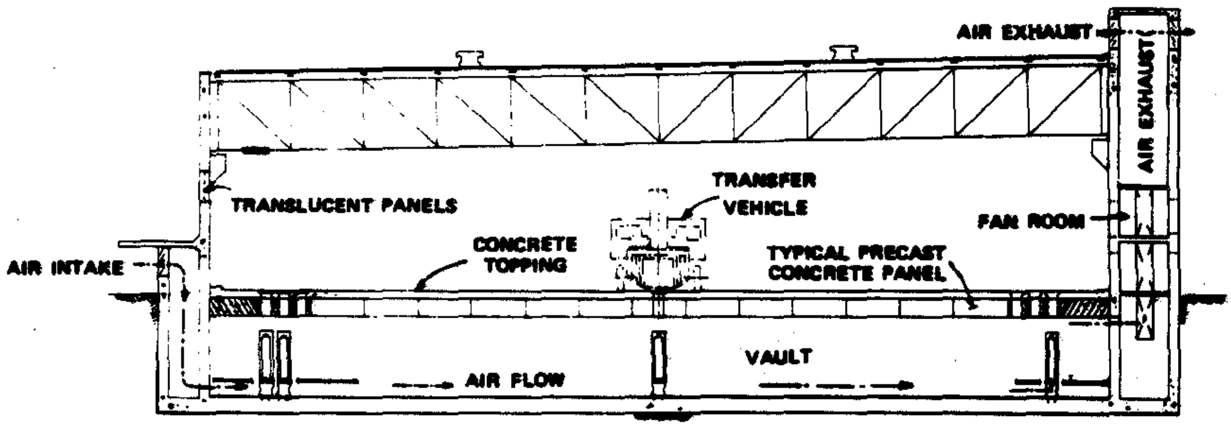


BUILDING SIZE – 132′ x 587′

PROCESSABILITY RATING ON SCALE OF 100 = 42

SYNROC-D CERAMIC PROCESS

FIGURE 2. DWPF Forms Processing Steps



Risks of interim storage of either glass or Ceramic DWPF Waste Forms calculated to be below 3×10^{-5} mr/yr maximum individual, 3×10^{-4} mrem/yr population

FIGURE 3. DWPF Interim Storage Facility

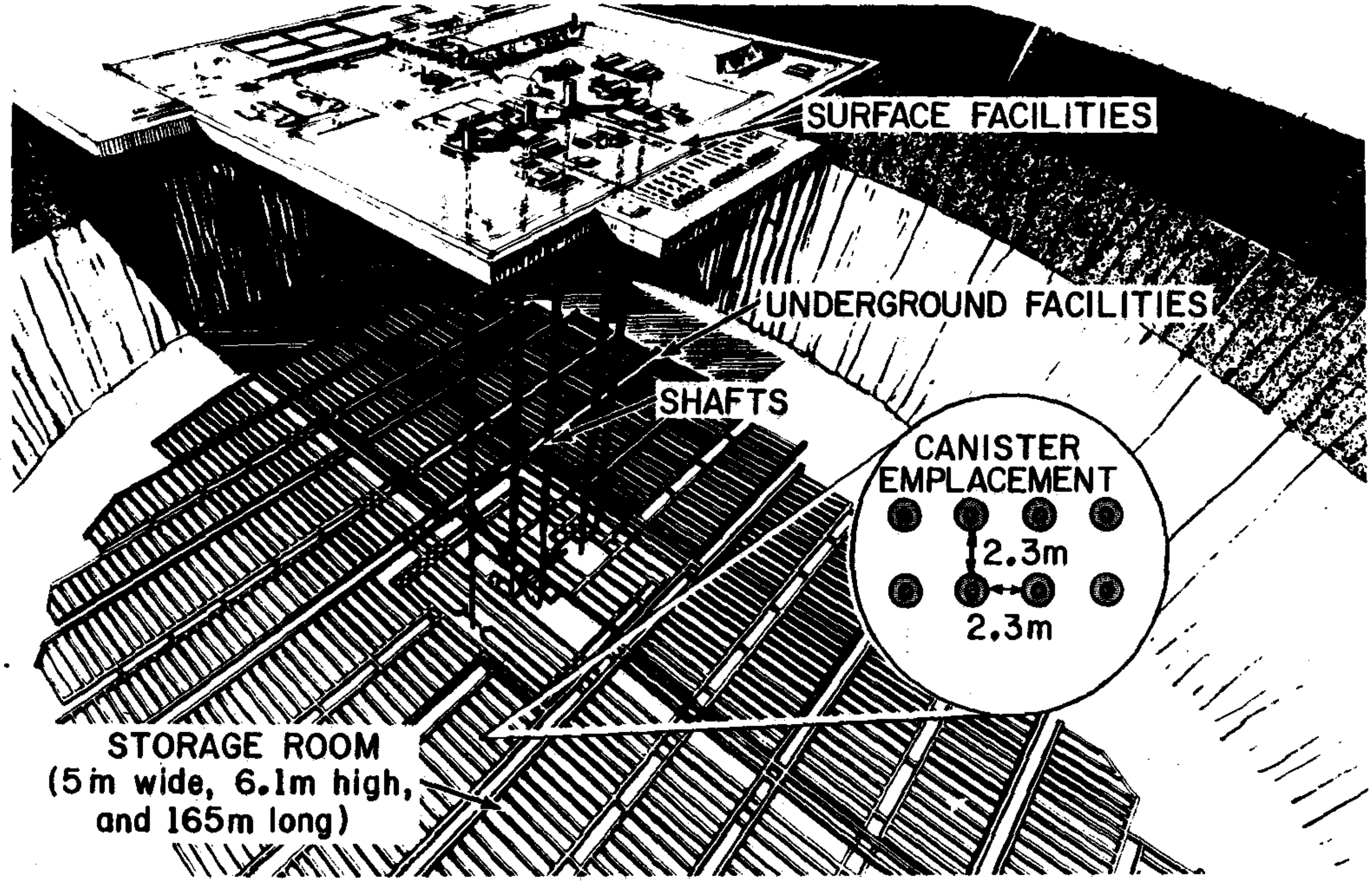


FIGURE 4. Generic High-Level Waste Repository

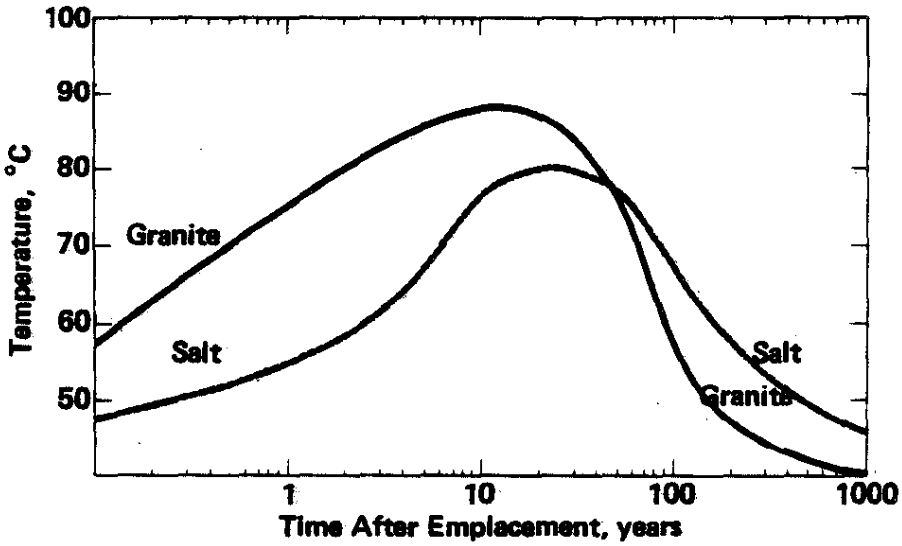


FIGURE 5. Surface Temperatures for DWPF Glass Canisters in Geologic Repositories

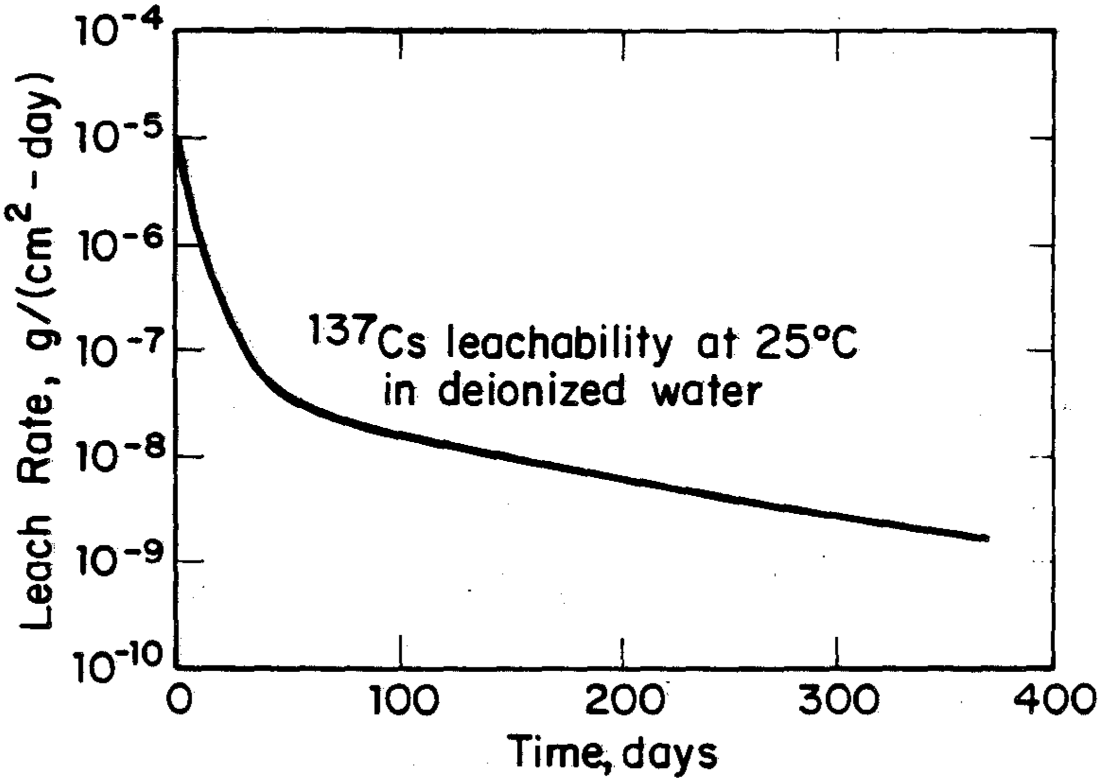


FIGURE 6. Leaching of Borosilicate Glass Versus Time

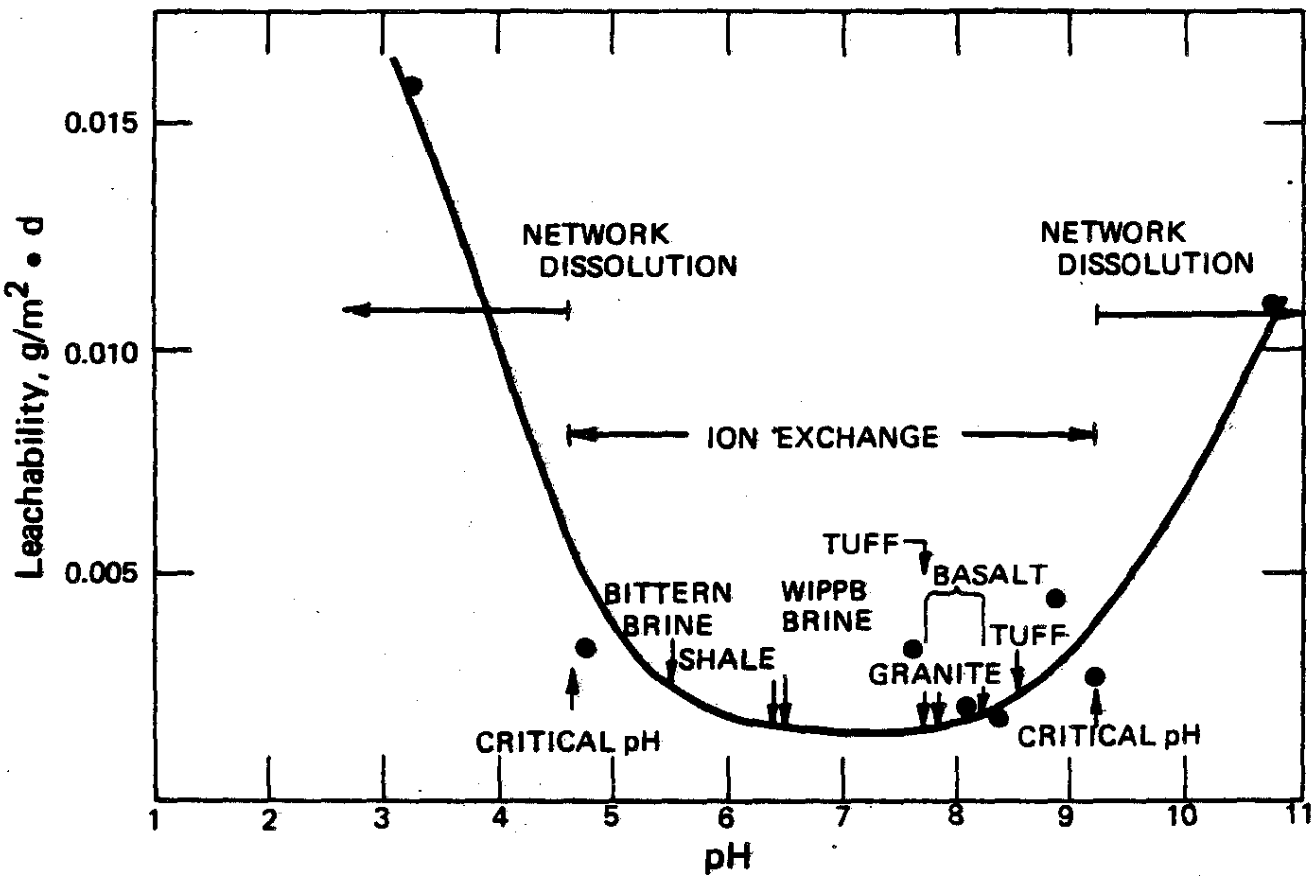
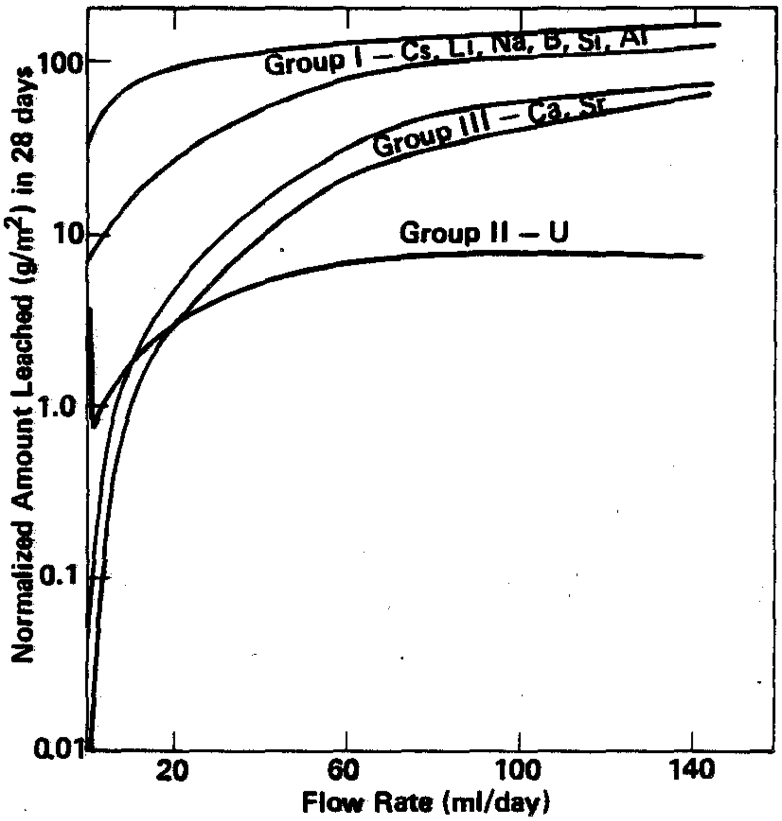
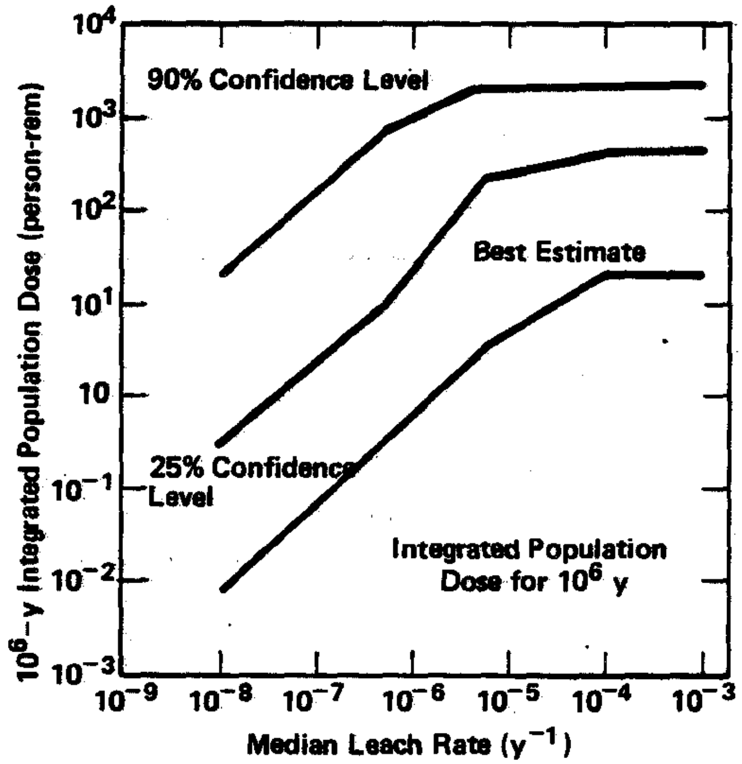


FIGURE 7. Leaching of Borosilicate Glass Versus pH
(Si leachability, 5-day static test, 23°C)



*Tests at 90°C in DI Water, A/V = 0.1 cm^{-1}

FIGURE 8. Effect of Leachant Flow Rate on Cumulative Leaching of Composite Waste Glass*



*For standard deviations of one (σ = 10).

FIGURE 9. Calculated Risks From Disposal of SRP HLW in a Salt Repository*