

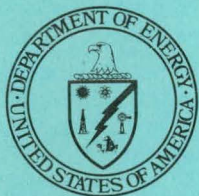
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# Evaluation of Increased Cesium Loading on Submerged Demineralizer System (SDS) Zeolite Beds

*DOE-SDS Task Force*

May 1981



U.S. Department of Energy  
Assistant Secretary for Nuclear Energy  
Office of Waste Operations and Technology

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**U.S. Department of Energy**  
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# EVALUATION OF INCREASED CESIUM LOADINGS ON SUBMERGED DEMINERALIZER SYSTEM ZEOLITE BEDS

## EXECUTIVE SUMMARY

A Submerged Demineralizer System (SDS) is being installed at the Three Mile Island Unit 2 (TMI-2) Nuclear Power Station for decontamination of the Containment Building (CB) sump water and Reactor Coolant System (RCS) water.

A Department of Energy (DOE) Task Force was assembled to evaluate the relative technical and financial benefits in storing, shipping, treating, and disposing of SDS zeolite liners, assuming that the liners will be loaded to a level higher than that (10,000 Ci/liner) originally planned by General Public Utilities (GPU).

The DOE-SDS Task Force concludes that it is technically feasible to load the zeolite liners used in the SDS to levels up to 60,000 Ci of cesium per liner without additional preoperational testing. This would result in approximately ten such liners. The Task Force further concludes that these liners can be safely handled, stored, transported, and vitrified. Moreover, the Task Force acknowledges that it may be technically feasible to load them to even higher levels.

Loading the SDS zeolite liners up to 60,000 Ci of cesium would result in approximately 17 additional strontium-loaded liners if Ionsiv IE-95 ( $\text{Na}^+$  form) is used. The Task Force concludes that these liners can also be safely handled, stored, transported, and disposed of; however, the choice of the form for ultimate disposal of these wastes is beyond the scope of this exercise.

The Task Force addressed only the zeolite portion of the SDS and did not consider the final purification portions of the system, since they have no bearing on the Task Force conclusions. The findings in each of the areas to support the Task Force conclusions are summarized below.

#### Process Flowsheet

Laboratory-scale process tests were conducted at Oak Ridge National Laboratory (ORNL) with actual TMI-2 Containment Building water. These tests verified that the SDS liners could be loaded to 200 bed volumes (approximately 10,000 Ci of cesium per liner), in accordance with the reference flowsheet, without cesium or strontium breakthrough. These tests also demonstrated that SDS liners could be loaded to about 500 bed volumes (22,000 Ci/liner) with less than 1% strontium breakthrough. Further ORNL testing indicated that SDS liners could be loaded to at least 1000 bed volumes of zeolite (42,000 Ci/liner) without cesium breakthrough, but with significant breakthrough of strontium, which could be removed by additional downstream liners. Extrapolation of these experimental data indicates that increasing the loadings to 60,000 Ci, or more, of cesium per liner should be readily achievable.

The ORNL information served as a basis for establishing several options used to evaluate the relative merits of higher SDS liner loadings. The approximate number of zeolite liners for each option is listed in Table 1.1. The reference case of 10,000 Ci per liner requires a total of 60 liners. All other cases considered result in a twofold reduction in the total number of zeolite liners.



Table 1.1. SDS Liners Required as a Function of Cesium Loading per Liner  
(Based on ORNL-Provided Data)

Case	Cesium Loading per Liner (Ci)	Number of SDS Liners with High-Level Loadings of Cesium <sup>1</sup>	Number of SDS Liners with Low-Level Loadings of Strontium	Total Number of Liners	Number of SDS Liners Requiring Vitrification of Zeolite	Approximate Number of Vitrified Waste Canisters <sup>2</sup> Generated
Ia	120,000	5	21	26	26	46
Ib <sup>3</sup>	120,000	5	21	26	5	9
IIa	60,000	10	17	27	27	48
IIb <sup>3</sup>	60,000	10	17	27	10	18
III	22,000	25	0	25	25	44
IV (Reference Case)	10,000	60	0	60	60	105

<sup>1</sup>These numbers have about 20% margin. Also, the liners in this column will have curie loadings to the level specified in the second column.

<sup>2</sup>Each SDS liner that is vitrified generates 1.75 canister per liner.

<sup>3</sup>This case assumes that only the cesium loaded liners are vitrified.

### Stability of Loaded Zeolite Liners

There is substantive evidence which provides the basis for confidence that the SDS liners containing Ionsiv IE-95 can be loaded to 60,000 Ci of cesium each and stored for a period of 10 years (i.e., a dose of  $1.7 \times 10^{11}$  rads) without hazard to public health and safety and in a manner which provides a basis for their ultimate disposal. This evidence can be summarized as follows:

- (1) In two laboratory-scale tests performed in 1965, zeolite (Linde Type 4A) was loaded up to 10 Ci of  $^{90}\text{Sr}$  per  $\text{cm}^3$ , irradiated to exposures as high as  $4 \times 10^{11}$  rads during a 2-year period, and then in one case washed with water and eluted with nitric acid. In these tests the irradiated zeolite did not undergo gross degradation (i.e., alteration of flow properties or decreased affinity for sorbed strontium).
- (2) Ionsiv IE-95 is known from recent laboratory tests to retain both its crystal structure and its affinity for cesium at an absorbed dose of  $1.1 \times 10^{10}$  rads.
- (3) From a theoretical standpoint, there is good reason to believe that Ionsiv IE-95 will not suffer significant radiation damage during a 10-year storage period. Even if it did, the principal expected effect is a very slow conversion of some of the crystalline chabazite at the center of the liner to an amorphous form without significant loss of cesium. The resulting amorphous form would be expected to be suitable for eventual conversion to a glass.
- (4) The SDS liner vessel has been designed to be the primary protective barrier to the environment and public. This is accomplished by

(a) designing the vessel with a pressure rating of 350 psi, which prevents pressurization failure of a closed vessel; (b) providing an off-gas system adequate to remove any radioisotopes which may be released; and (c) using low-carbon austenitic stainless steel (316L), which provides corrosion resistance.

SDS liners loaded with 60,000 to 120,000 Ci of cesium and strontium are expected to be thermally stable during storage. Peak centerline temperatures of 700 and 1290°F are calculated on the basis that all radiation from the cesium and strontium would be dissipated as heat energy within the liner. In reality, some radiation escapes from the liner. This results in temperatures well below the calculated values. At temperatures of 700 to 1290°F little, if any,  $^{137}\text{Cs}$  is expected to volatilize during storage of cesium-loaded SDS liners.

Loaded SDS zeolite liners will contain considerable amounts of water; radiolysis of the water during storage will generate hydrogen and oxygen. Evolved hydrogen and oxygen gases can be dispersed by venting the stored SDS liners to an appropriate off-gas system, as presently planned.

#### On-Site Handling

Effects upon personnel exposure during the handling of higher loaded liners at 60,000 Ci (and 120,000 Ci) are expected to be of no consequence and, in fact, may reduce exposure by reducing transfers to the storage module. In addition, the handling of the strontium-predominated liners does not appear to present any technical difficulties. The Chem-Nuclear Systems, Inc. (CNS) 1-13C cask appears to be suitable for handling liners with loadings up to 60,000 Ci at TMI.

The use of alternative casks is possible, but efforts would have to be made to modify the pool loading rack, and actions would be necessary to address uncertainties with regard to the Certificate of Compliance.

#### Transportation

SDS liners containing as much as 120,000 Ci of cesium, and the resulting vitrified waste canisters, can be safely transported in compliance with Department of Transportation (DOT) and Nuclear Regulatory Commission (NRC) regulations.

The cost for shipping liners with 60,000 Ci is appreciably less than that for the reference case. Depending on shipping destinations, including the return of the vitrified waste, it would be possible to realize cost savings up to \$1 million if the SDS liners are loaded to 60,000 Ci instead of 10,000 Ci.

The CNS 1-13C cask has been identified as the most appropriate shipping container because of its apparent suitability for such service.

#### Waste Form Processing

Waste form processing considerations utilized vitrification as a reference immobilization process. Loading to 60,000 Ci per liner would result in approximately 18 waste glass canisters, each containing ~34,000 Ci. [One SDS liner will result in 1.75 waste glass canisters (see Table 1.1).] Battelle Pacific Northwest Laboratory (PNL) has successfully vitrified zeolite which simulated TMI wastes up to an equivalent of 70,000 Ci of cesium per liner. These laboratory vitrification tests were conducted without any observable cesium volatility. If higher loadings are used, the vitrification tests should be repeated for

120,000 Ci per liner to confirm that no unexpected problems are experienced with the TMI waste at this loading. It should be noted that spent fuel assemblies have been processed and successfully vitrified at PNL. The total curie content of the waste glass canisters which were generated from the vitrification of high-level waste from spent fuel was approximately 265,000 Ci.

#### Incremental Costs

A summary of the costs associated with the processing of SDS liners is presented in Table 1.2. The data given in this table indicate that higher loadings of the liners can result in significant cost savings over those of the Reference Case (Case IV). These savings are associated with transportation and vitrification, rather than operations, maintenance, and on-site storage. Waste disposal costs were not estimated, however, since waste forms for a repository have not been defined, and it is not known whether further treatment may be required. Shipping distances also have a marked effect on cost.

Table 1.2. Cost Estimates for Processing SDS Liners

Case	Cesium Loading per Liner (Ci)	Cost (\$000) for Shipping Distance	
		250 Miles	2500 Miles
Ia	120,000	1717	2410
Ib	120,000	718	1050
IIa	60,000	1731	2398
IIb	60,000	955	1341
III	22,000	1611	2232
IV	10,000	3751	5240



EVALUATION OF INCREASED CESIUM LOADINGS ON SUBMERGED DEMINERALIZER  
SYSTEM ZEOLITE BEDS

DOE-SDS Task Force

1.0 INTRODUCTION

A Submerged Demineralizer System (SDS) is being installed at the Three Mile Island Unit 2 (TMI-2) Nuclear Power Station for decontamination of Containment Building (CB) sump water and reactor coolant system (RCS) water. The reference flowsheet that was developed was designed to decontaminate the high-activity-level water (HALW) so that, when mixed with normal plant discharges, the concentrations of all nuclides (except tritium) in the decontaminated water would be <10% of the concentrations listed in the Code of Federal Regulations, Title 10, Part 20 (10 CFR 20). The reference flowsheet includes (1) clarification by filtration, (2) sorption of cesium and strontium (the bulk of the radioactivity) on an inorganic zeolite ion exchanger (Linde Ionsiv IE-95<sup>™</sup>\*), and (3) sorption of the remaining radioactive species (polishing treatment) on organic ion exchangers. The SDS system uses three zeolite beds in series. In the reference case, the operating procedure planned by General Public Utilities (GPU) provides that 200 bed volumes of water will be passed through each zeolite bed while it is in the first position. The loaded zeolite bed will then be removed from the system, the other zeolite columns will be moved forward one position (countercurrent to the water flow), and a new zeolite column will be installed in the third position. This mode of operation would result in the sorption of nearly 10,000 Ci of radioactivity on each loaded zeolite bed.

---

\*Formerly called AW-500.

A DOE-SDS Task Force was assembled to evaluate the relative technical and financial benefits in storing, shipping, treating, and disposing of SDS zeolite liners, assuming that the liners are loaded to a level higher than the 10,000 Ci/liner originally planned by GPU. Loadings of 60,000 and 120,000 Ci/liner were considered by the Task Force. This document is a report of these considerations. Specifically, these considerations included evaluations of the potential impacts of the larger loadings on: (1) the process flowsheet, (2) the storage of the liners, (3) on-site handling of the liners, (4) transportation of the liners from the TMI site and the return of waste forms to the site, (5) processing of the waste into vitrified glass form, and (6) cost of treatment.

Although there are two aspects involved, namely, removal of the bulk of the cesium and strontium on the zeolite columns and removal of the trace residual contaminants, only the first part of the process is considered in this study.

## 2.0 PROCESS FLOWSHEET CONSIDERATIONS

The radiocesium isotopes,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ , are the predominant sources of gamma activity in the high-activity-level (HALW) contaminated water at TMI-2, as shown in Table 2.1. Sorption of this radiocesium onto the fewest number of SDS zeolite columns (containing 60 gal of zeolite per column) may enable benefits to be realized in the subsequent storage, transportation, treatment, and disposal operations. The purpose of this chapter is to assess the impact of modifying the SDS flowsheet to enable higher loadings of cesium on the zeolite ion exchanger columns. Toward this end, cesium loadings of 60,000 and 120,000 Ci per zeolite column were compared with the reference SDS flowsheet, in which the columns are to be loaded to nearly 10,000 Ci.

### 2.1 Reference SDS Flowsheet

#### 2.1.1 Reference Flowsheet Development

The reference flowsheet was specified by the TMI-2 Technical Advisory Group on the basis of brief studies carried out at the Oak Ridge National Laboratory (ORNL) and at the Savannah River Laboratory (SRL). Sorbent selection, which was made during the summer of 1979, was based on a literature survey of similar processing experience and on the results of a few tests which were made with the small-volume sample of Reactor Coolant System (RCS) water that was then available. The studies at ORNL included distribution measurements using RCS water and a variety of sorbents, and small-scale column tests using synthetic solutions traced with either  $^{89}\text{Sr}$  or  $^{137}\text{Cs}$ . The studies at SRL included computer calculations plus confirming tests. The results of these studies

Table 2.1. Concentration and Composition of Bulk Contaminants  
in High-Activity-Level Water at TMI-2

	As of July 1, 1980		Est. as of July 1, 1981 <sup>c</sup>	
	RCS <sup>a</sup>	CB Sump <sup>b</sup>	RCS	CB Sump
Concentration, $\mu\text{Ci/mL}$				
<sup>89</sup> Sr	3	0.53	0.02	0.005
<sup>90</sup> Sr	25	2.3	24	3.6
<sup>134</sup> Cs	6	26	2.4	18
<sup>137</sup> Cs	40	160	22	149
Total Ci <sup>d</sup>				
<sup>89</sup> Sr	1,020	1,200	7	11
<sup>90</sup> Sr	8,520	5,220	8,300	8,800
<sup>134</sup> Cs	2,040	59,000	810	42,800
<sup>137</sup> Cs	13,600	363,000	7,400	361,000

<sup>a</sup>Reactor Coolant System (RCS).<sup>1</sup>

<sup>b</sup>Containment Building (CB) Sump.<sup>2</sup>

<sup>c</sup>Based on radioactive decay plus leakage from RCS to CB sump of ~40,000 gal in 365 days.

<sup>d</sup>Based on volumes of 90,000 gal in RCS, 600,000 gal in CB sump on 7/1/80, and 640,000 gal in CB sump on 7/1/81.<sup>3</sup>

indicated that the zeolite chabazite was the best commercially available, inorganic sorbent for both cesium and strontium. The kinetics of the sorption of strontium were found to be much slower than for cesium; this dictated the required contact time of the water with the zeolite. Also, the equilibrium distribution coefficient ( $K_d$ ) of strontium was found to be much smaller than that of cesium. Thus, the volume of zeolite necessary (the number of

columns to be used) was selected to provide adequate sorption of the strontium. If the water contained only cesium, much higher loadings, and therefore the use of fewer columns, would then be possible.

#### 2.1.2 Description of Reference Flowsheet

The reference SDS process flowsheet, shown in Fig. 2.1, was designed by Allied-General Nuclear Services for Chem-Nuclear Systems, Inc., the prime contractor for fabrication and installation of the equipment. In this flowsheet, the contaminated water is clarified by filtration during transfer into the ion exchange feed tank. The clarified water is pumped through either or both of two duplicate trains of ion exchange columns. Each train consists of a series of three columns containing zeolite (Ionsiv IE-95 in the  $\text{Na}^+$  form). The effluent from either train of zeolite columns is passed through one of two duplicate columns containing an organic cation exchange resin (Nalcite HCR-S™, initially in the  $\text{H}^+$  form). Finally, the effluent from both cation resin columns is combined and passed through a single large polishing column containing layers of cation resin (HCR-S, initially in the  $\text{H}^+$  form), anion resin (Nalcite SBR™, initially in the  $\text{OH}^-$  form), and mixed resin (Nalcite MR-3™, a 1:1 volume mixture of HCR-S and SBR).

The ion exchange columns can be moved easily since they are of modular design. The zeolite and cation resin columns are of the same size, although the volume of cation resin to be used is only one-half of the volume of zeolite that will be used in each column.

The operating procedure provides that 200 bed volumes of water will be passed through each zeolite column while it is in the first position. At



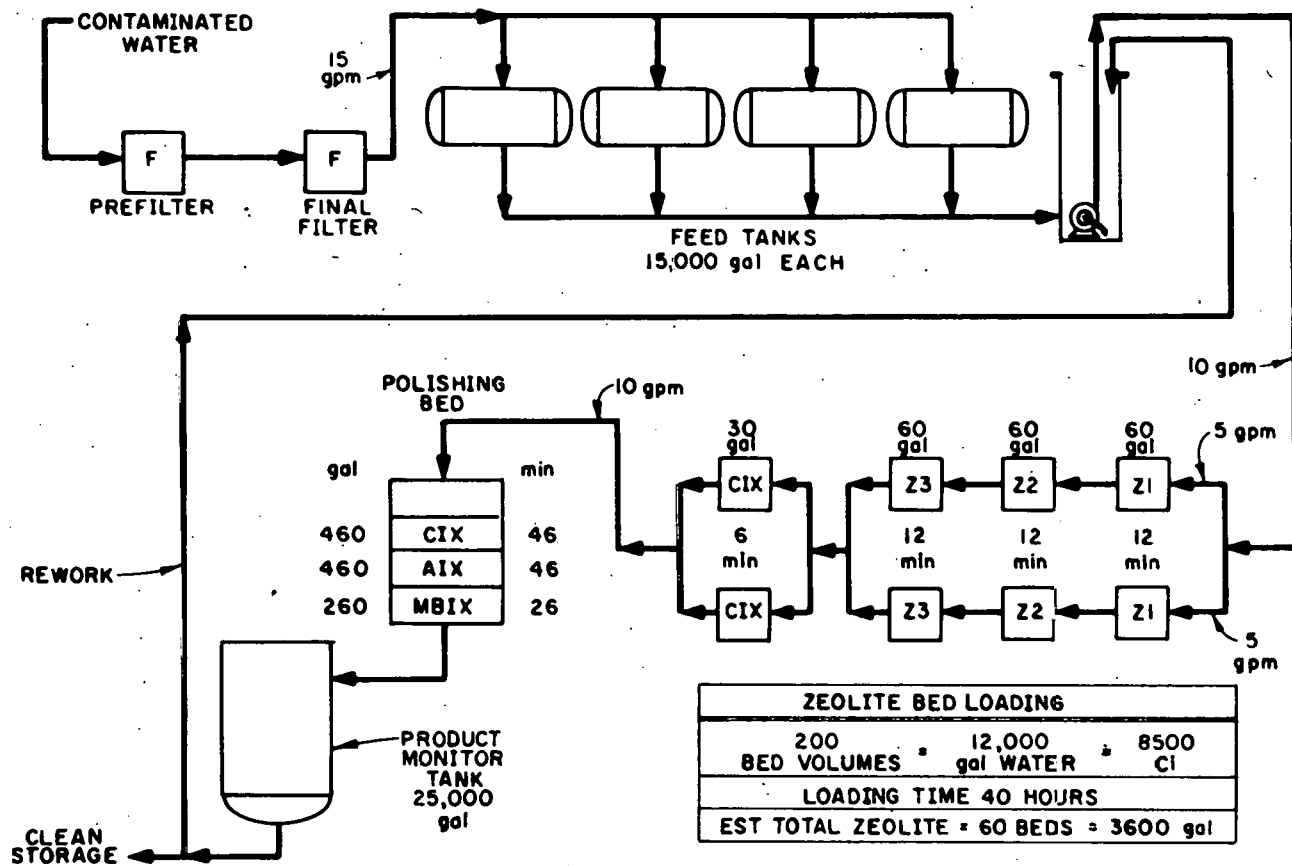


Figure 2.1. Submerged Demineralizer System flowsheet.

that time, the column containing the loaded zeolite will be removed from the system, the other zeolite columns will be moved forward one position (countercurrent to the water flow), and a new zeolite column will be installed in the third position. In this manner, the zeolite columns will sorb the cesium and most of the strontium while in the first position. While in the second and third positions, they will provide the necessary residence time to allow sorption of the residual strontium.

### 2.1.3 Evaluation of the Reference Flowsheet

A series of small-scale column tests ( $10^{-5}$  scale, based on SDS column size) was made at ORNL<sup>2</sup> to provide data for evaluating the reference flowsheet. These tests showed that the reference flowsheet would meet the design objective of decontaminating the HALW sufficiently that, when a reasonable volume of the effluent was mixed with the normal plant discharges, the concentrations of all radionuclides except tritium in the decontaminated water would be less than 10% of the concentrations listed in 10 CFR 20.<sup>4,5</sup>

The reference SDS flowsheet can be considered in two parts: (1) removal of the bulk of the cesium and strontium on the zeolite columns, and (2) removal of the trace residual contaminants. As stated previously, the present study is concerned only with the first part of the process, i.e., the removal of the bulk of the cesium and strontium on the zeolite columns.

## 2.2 Cesium Loading

The cesium concentration ( $^{134}\text{Cs} + ^{137}\text{Cs}$ ) in the CB sump water was expected to be about 186  $\mu\text{Ci/mL}$  (0.70 Ci/gal) as of July 1, 1980, as

shown in Table 2.1. Thus, the 200-bed volume loading specified in the reference flowsheet was expected to load about 10,000 Ci of cesium onto each zeolite bed, whereas approximately 1500 and 3000 bed volumes must be processed through each column in order to obtain loadings of 60,000 and 120,000 Ci, respectively.

In the SDS reference flowsheet evaluation tests, the loading of the first zeolite column was extended to 1000 bed volumes, which is equivalent to a loading of 42,000 Ci on a 60-gal bed. (The loading was not extended to cesium breakthrough.) The cesium concentration profile in the column was determined at the conclusion of the experiment. By this means, it was determined that no more than half of the column was loaded and that the peak-to-average loading ratio was about 4 (75% of the cesium was located on the top 20% of the column). From these data, a reasonable extrapolation indicates that an average cesium loading of 60,000 Ci per 60 gal of zeolite is possible, and that a loading of 120,000 Ci may also be feasible.

Other tests made with synthetic solutions traced with  $^{137}\text{Cs}$  were taken to breakthrough, as shown in Figure 2.2. Reasonable extrapolations can be made from these data for the sodium concentration and zeolite contact time to conditions that are anticipated (1200 ppm Na, 12-min contact time) during TMI-2 HALW cleanup. These extrapolations indicate that the breakthrough volume (10% instantaneous or about 1% cumulative breakthrough) for cesium would be about 7000 bed volumes. Based on these extrapolations, the SDS zeolite beds can be loaded to more than 120,000 Ci per 60 gal of zeolite.

With regard to radiation exposure and shielding calculations, it should be understood that the cesium concentration on the zeolite bed depends on

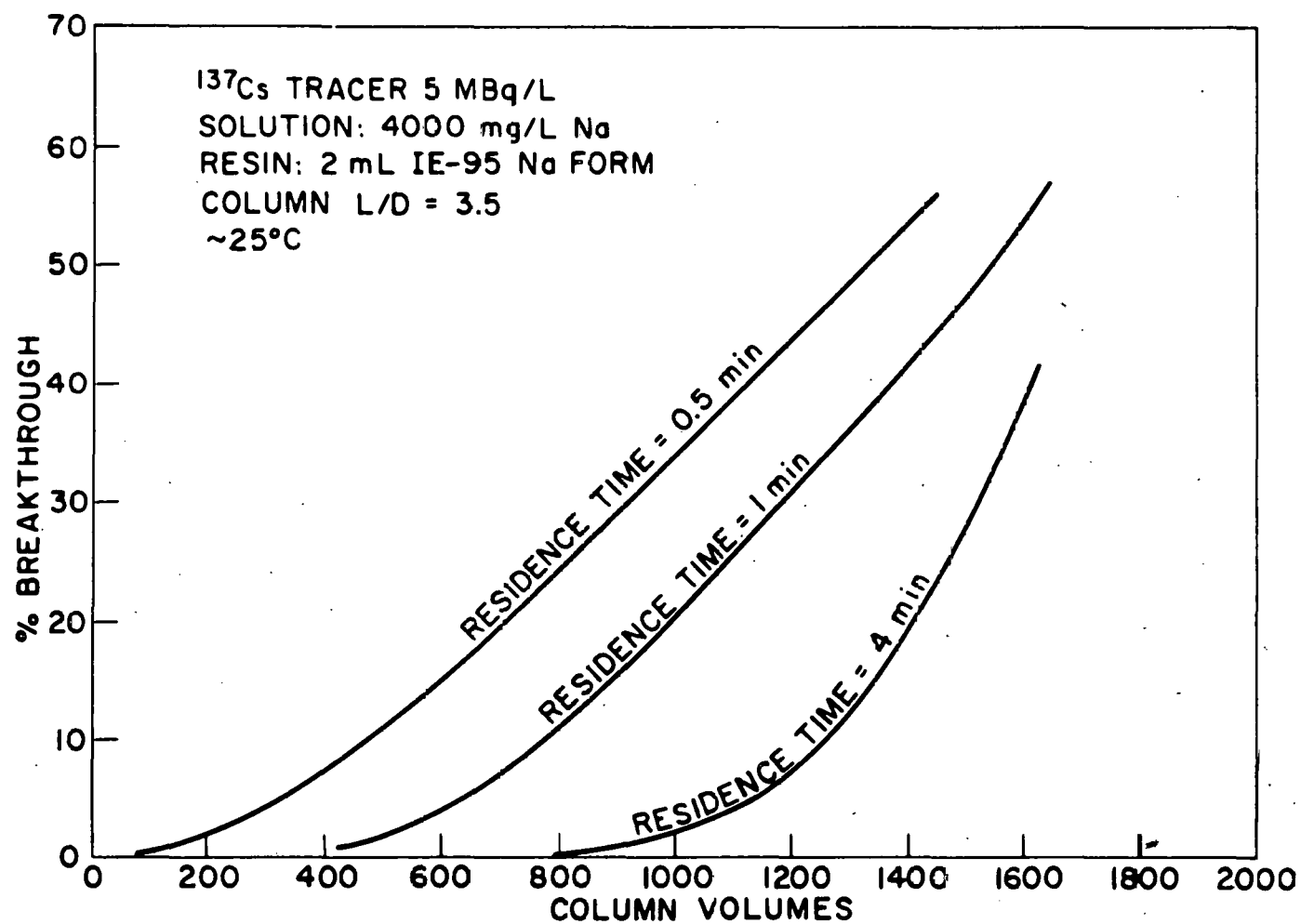


Figure 2.2. Effect of residence time on cesium loading of IE-95 zeolite.

the value of the distribution coefficient and is largely independent of the total amount loaded. In column operations, the volumetric distribution coefficient is defined as being equal to the breakthrough volume at 50% instantaneous breakthrough. Therefore, based on the data described above, the distribution coefficient will be greater than 7000. However, even at that value (7000) and with loadings of 10,000, 60,000, or 120,000 Ci per 60 gal of zeolite, the cesium band will occupy only the top 3.5, 21, or 41% of the SDS zeolite beds, respectively.

### 2.3 Strontium Loading

Strontium is sorbed on Ionsiv IE-95 zeolite to a considerably lesser degree than is cesium (see Figure 2.3). In the reference SDS flowsheet, the total volume of zeolite required (or the total number of SDS columns required) depends on the amount of strontium that must be sorbed, independent of the amount of cesium loaded, unless some other means is provided to remove the strontium. Thus, alternatives which may involve either the use of additional zeolite liners or other absorbents, such as Linde 4A™, sodium titanate,\* or organic resins, may be employed to remove the residual strontium.

The number of SDS zeolite columns required for adequate strontium sorption will probably be about 20-30 liners rather than the 60 liners specified by the reference flowsheet, if the performance of the SDS system scales adequately from the small-scale column tests (scale factor of  $10^5$ ). From the data presented in Figure 2.3, the volume of water that can be processed through the series of three zeolite columns without exceeding 1% breakthrough of strontium is calculated to be 500 ( $\pm$  50) bed volumes. This

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\*See Appendix A.



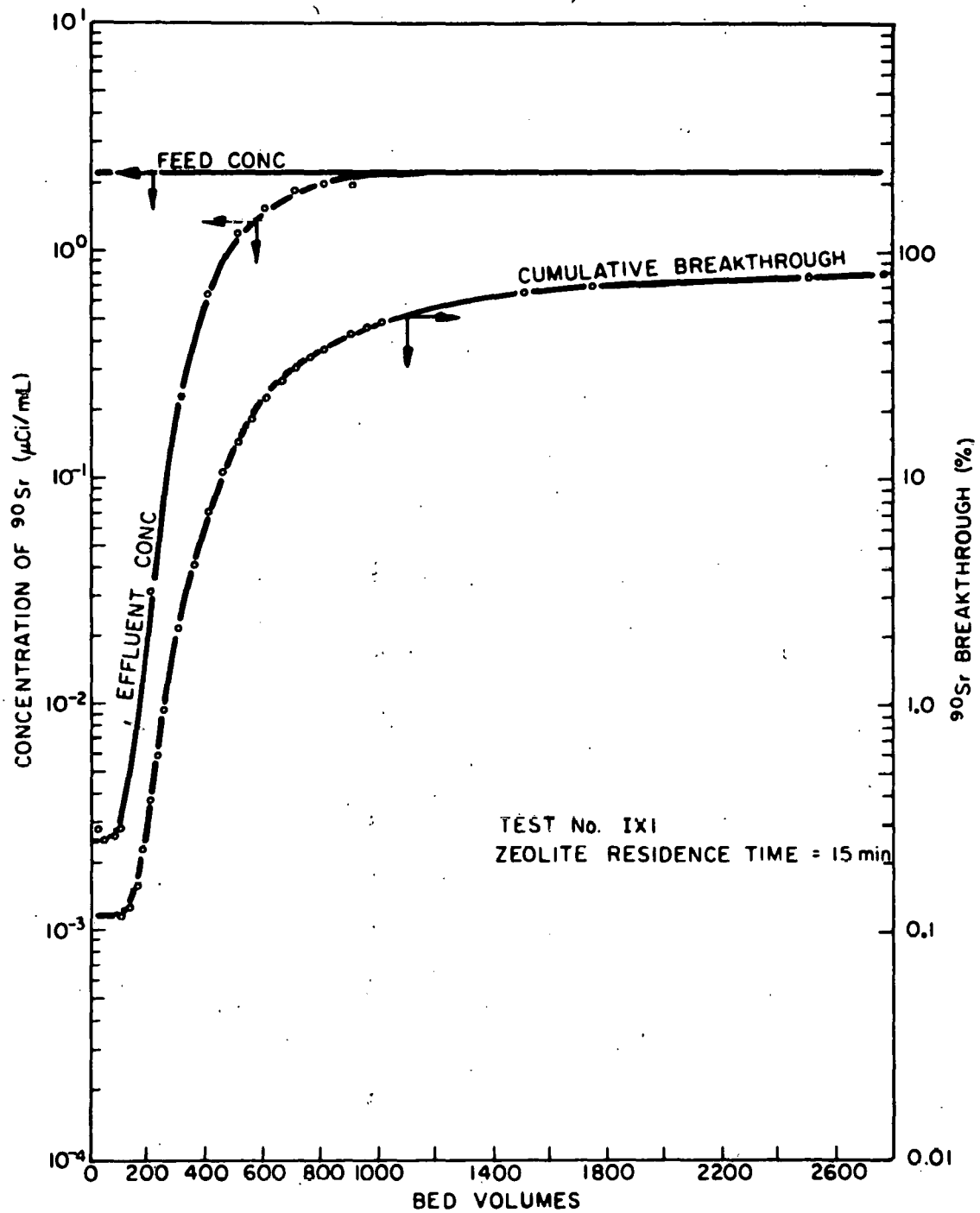


Figure 2.3.  $^{90}\text{Sr}$  concentration in zeolite inlet and outlet streams and cumulative breakthrough as functions of volume of water processed.

effect is shown graphically in Figure 2.4. These calculations are based on a  $^{90}\text{Sr}$  feed concentration of  $2.3 \mu\text{Ci/mL}$ , the concentration in the CB sump water. The volume of zeolite (number of SDS beds) required is not expected to change, even if the  $^{90}\text{Sr}$  feed concentration is higher than  $2.3 \mu\text{Ci/mL}$ , because the strontium loading is kinetically controlled and is not capacity limited.

## 2.4 Potential Flowsheet Modifications

A large number of operational modifications can be made to the 200-bed volume (10,000-Ci) reference case to increase the loading of the cesium on the zeolite. Three representative options which were considered are described in this section.

### 2.4.1 Option A — Increase of Volume of Water Processed in the First Column to 500 Bed Volumes

Based on the experimental data, the volume of water processed through the first of the series of three zeolite columns could be increased to about 500 bed volumes without causing a breakthrough of more than 1% of the strontium from the third zeolite column. However, the breakthrough would be rather sharp. The cesium loading of the beds removed from the first position would be increased to about 22,000 Ci; the number of zeolite columns used for processing the HALW (excluding flush water) would be reduced to about 25.

### 2.4.2 Option B — Increase of Volume of Water Processed in the First Column to 1500 or 3000 Bed Volumes

Increases of the volume of water processed to 1500 and 3000 bed volumes are necessary to achieve cesium loadings of 60,000 and 120,000 Ci per SDS column, respectively. This would reduce the number of columns loaded with

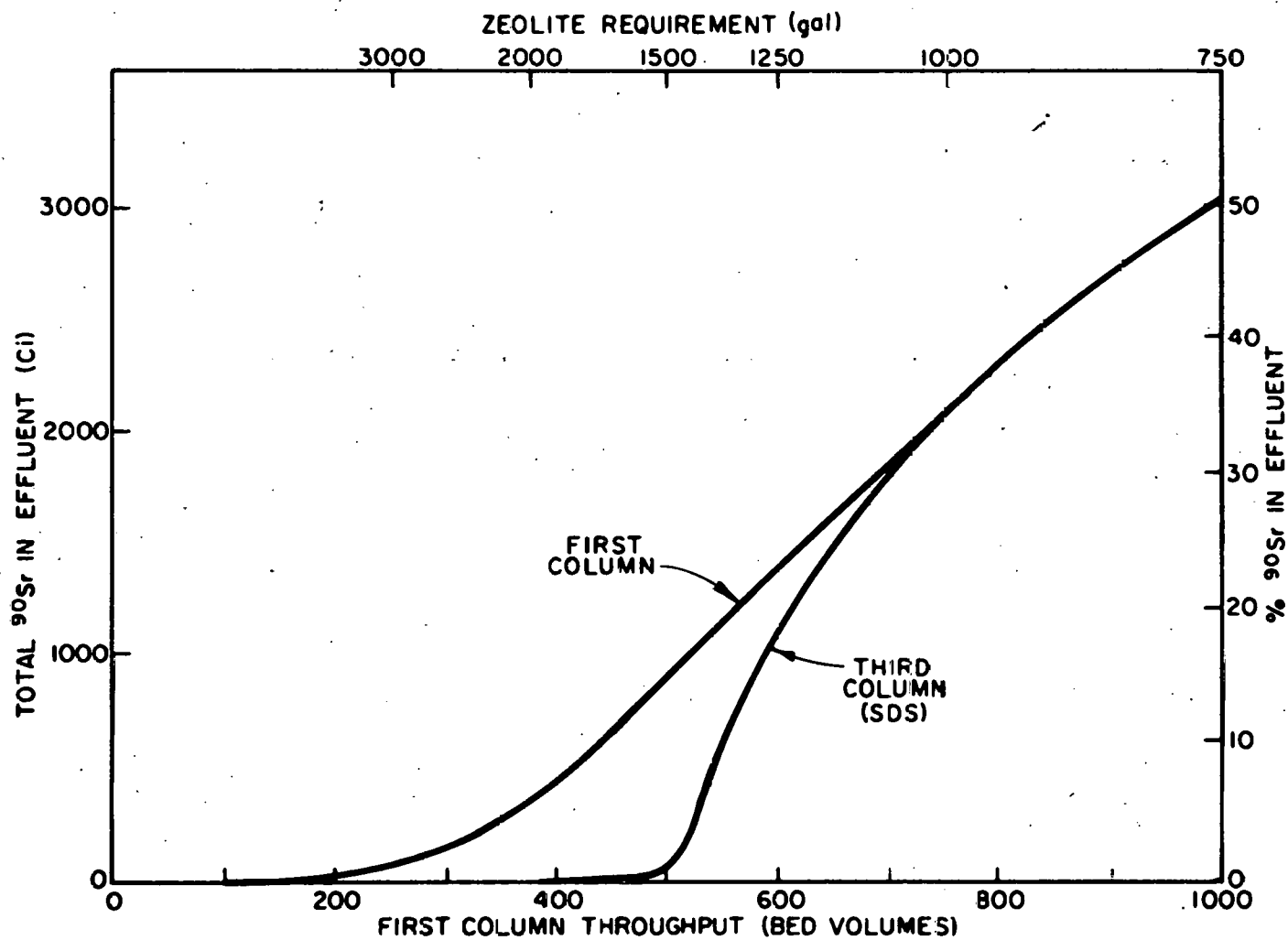


Figure 2.4. Strontium breakthrough in SDS flowsheet.

cesium to about 10 and 5, respectively. However, if the 1500 and 3000 bed volumes are processed through the second and third columns, about 50% and 75%, respectively, of the strontium would remain in the effluent water and would require further decontamination in subsequent operations. Overall, the volume of zeolite used would still depend on strontium removal and could not be reduced further than in Option A unless a sorbent other than Ionsiv IE-95 were employed to remove the strontium.

#### 2.4.3 Option C — Processing of 1500 or 3000 Bed Volumes in the First Zeolite Column with Changeout of the Second and Third Columns

In this case, the zeolite columns in the first position would be changed after processing 1500 or 3000 bed volumes to enable cesium loadings of 60,000 or 120,000 Ci. The columns in the second and third positions would be changed after processing 500 bed volumes. (The second-position column would be removed, the third-position column moved to the second-position, and a new column installed at the third position.) If necessary, the fourth-position column could be used as a zeolite column to provide the original three-column strontium removal capability. Again, the volume of zeolite (number of columns) used for strontium sorption could not be reduced below that in Option A unless a sorbent other than Ionsiv IE-95 were employed to remove the strontium.

### 2.5 Findings

Laboratory-scale process tests conducted at ORNL with actual TMI-2 Containment Building water verified the reference flowsheet, i.e., the processing of 200 bed volumes (10,000 Ci) per liner without cesium or strontium breakthrough. Tests were also conducted which indicated that 500 bed volumes (22,000 Ci/liner) could be processed with less than 1% strontium breakthrough.

Other tests performed at ORNL indicate that loadings of at least 42,000 Ci of cesium (1000 bed volumes) can be achieved without cesium breakthrough, but with significant strontium breakthrough. The residual strontium would thus require other means for its removal.

A reasonable extrapolation of the experimental data indicates that an average cesium loading of 60,000 Ci per 60 gal of zeolite is possible, and that a loading of 120,000 Ci may also be feasible.

The estimated number of zeolite columns that will be required for various cases considered in the remainder of this report are summarized in Table 2.2. The bases for these estimates are the results of the ORNL laboratory-scale tests; however, because of the  $10^5$  scaling factor involved, the numbers of columns required must be considered as approximate.

The reference case requires a total of 60 liners; all of the other cases considered result in a factor of 2 reduction in the total number of liners which contain high-specific-activity levels.

Table 2.2. Estimated requirements for zeolite columns

Case	Maximum Cs loading per liner (Ci)	Number of liners required	
		High activity level (Cs-Sr)	Low activity level (Sr)
I <sup>a</sup>	120,000	5	21
II <sup>a</sup>	60,000	10	17
III	22,000	25	0
Reference	10,000	60	0

<sup>a</sup>Assumes Option C is employed.

## 2.6 References

1. W. N. Bishop, D. A. Nitti, N. P. Jacob, and J. A. Daniel, "Fission Product Release from the Fuel Following the TMI-2 Accident," Proceedings of the 1980 ANS/ENS Topical Meeting, Thermal Reactor Safety, American Nuclear Society, Knoxville, Tenn., April 1980.
2. D. O. Campbell, E. D. Collins, L. J. King, and J. B. Knauer, Evaluation of the Submerged Demineralizer System (SDS) Flowsheet for Decontamination of High-Activity-Level Water at the Three Mile Island Unit 2 Nuclear Power Station, ORNL/TM-7448 (July 1980).
3. C. G. Hitz, Metropolitan Edison Company, private communication (Jan. 29, 1981).
4. Letter from B. C. Rusche to R. C. Arnold, "Submerged Demineralizer System Progress Review by the TMI Technical Advisory Group," dated May 27, 1980.
5. Code of Federal Regulations, Title 10, Part 20, Appendix B, Table II, Column 2.

### 3.0 ZEOLITE STABILITY CONSIDERATIONS

This chapter considers the various issues involved in estimating the stability of  $^{137}\text{Cs}$ -loaded zeolite (Ionsiv IE-95) after extended storage. The  $^{137}\text{Cs}$ -loaded zeolite liners resulting from operation of the TMI-SDS may be stored at the TMI site for 1 to 10 years, and perhaps even longer. Stability is a major consideration in the determination of appropriate radioactive cesium loadings of the SDS zeolite liners.

To provide a suitable basis for comprehensive stability assessments, parametric calculations of cumulative radiation doses and thermal loads were made for SDS-type zeolite liners loaded with 10,000 to 120,000 Ci of  $^{137}\text{Cs}$  and stored for as long as 10 years. Results of these calculations are presented in the early part of this chapter. Subsequent sections address the stability of cesium-loaded liners during storage. Laboratory- and plant-scale experience acquired over the last 10 to 20 years at various Department of Energy (DOE) sites provides a large data base from which to judge the stability of cesium-loaded TMI zeolite liners. Radiation and chemical effects during loading of the SDS zeolite liners are mentioned and briefly discussed.

#### 3.1 Proposed Levels of Loading

A parametric analysis of absorbed radiation dose and heat generation rates in loaded SDS liners was conducted based on data from ion exchange studies at the Oak Ridge National Laboratory (ORNL) (cf. Chapter 2). These studies have shown that the  $^{137}\text{Cs}$  concentration on the Ionsiv IE-95 sorbent can vary from 4000 to 8000 times the  $^{137}\text{Cs}$  concentration in the CB sump water. Estimates of heat and absorbed radiation dose over this range are expected to cover any extremes that could occur in the operation of the SDS system.

Total  $^{137}\text{Cs}$  liner loadings of 10,000, 60,000, and 120,000 Ci were considered in the parametric calculations. Three different concentrations of  $^{137}\text{Cs}$  in the Ionsiv IE-95 sorbent were considered for each total loading level, namely, 4000, 6000, and 8000 times the  $^{137}\text{Cs}$  concentration in the CB sump water. At a  $^{137}\text{Cs}$  loading of 60,000 Ci, these concentrations correspond, respectively, to cesium loading band heights on the SDS liners of 12.7, 8.4, and 6.4 in. In all cases, the concentration of  $^{90}\text{Sr}$  in the Ionsiv IE-95 sorbent was 500 times that in the CB sump water. This radiostrontium concentration corresponds to a strontium loading band height of 30.6 in. for  $^{137}\text{Cs}$  loadings of 60,000 to 120,000 Ci.

A Monte Carlo code developed by Zimmerman and Thomsen<sup>1</sup> was used to calculate absorbed radiation doses at various points in the loaded SDS liners for each of the nine cases considered. All dose calculations were made for liners filled with Ionsiv IE-95 of density 42 lb/ft<sup>3</sup>.

The Monte Carlo-type radiation dose calculations were made on the basis that all electronic contributions were converted to bremsstrahlung photons. The photon spectrum from bremsstrahlung radiation was combined with the photon emission spectrum to obtain a single radiation source term. (This calculational procedure was necessary in order to obtain proper input data to the particular computer code used.) Photons, of course, have a greater chance of escaping from the SDS liner than do electrons. Thus, doses calculated by converting electronic contributions to bremsstrahlung radiation are lower than those which take into account absorbed beta radiation. However, treating the beta dose in this fashion does not seriously impact results of Monte Carlo-type calculations for SDS liners.



The following equation, due to Jaeger et al.,<sup>2</sup> was used in a few instances to calculate absorbed radiation doses:

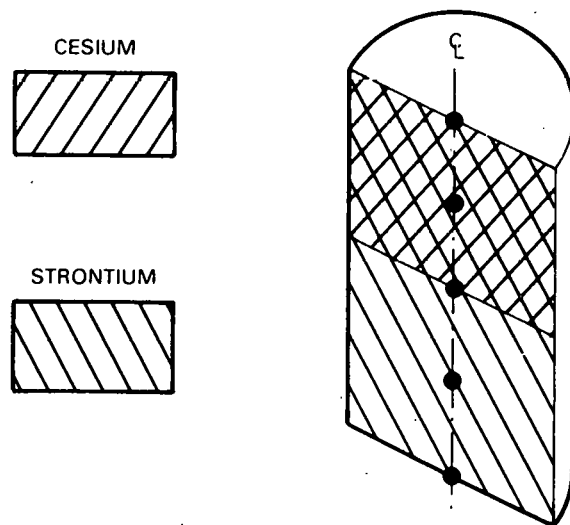
$$\phi = \frac{S_v}{2\mu_s} [G(\mu_s, h_1, b) + G(\mu_s, h_2, b)] , \quad (1)$$

where

- $\phi$  = the dose rate (R/h);
- $S_v$  = the specific strength of the volume source ( $\text{Rcm}^{-1} \text{ h}^{-1}$ );
- $\mu_s$  = the linear attenuation coefficient (total macroscopic cross section) for gamma rays in the radioactive source material,  $\text{cm}^{-1}$ ;
- $h_1$  = height from the top of the bed to the point in the bed where the dose is to be calculated, cm;
- $h_2$  = height from the bottom of the bed to the point where the dose is to be calculated, cm.

The function  $G$  and the term  $b$  are defined in Figures 6.4-6.9 of the text by Jaeger et al.<sup>2</sup>

Monte Carlo dose calculations for a 10-year storage period were made along the centerline of loaded SDS liners at the top, midpoint, and bottom of the  $^{137}\text{Cs}$  sorption band and also at the midpoint and bottom of the  $^{90}\text{Sr}$  sorption band (Figure 3.1). Results of these calculations are plotted in Figures 3.2-3.4; the maximum absorbed radiation doses calculated under these conditions are listed in Table 3.1. The computer code used also provided calculated total radiation doses for various regions of the liner. These doses were summed and divided by the volume of Ionsiv IE-95 to yield the average absorbed radiation doses, which are also tabulated in Table 3.1. Because cesium loads onto Ionsiv IE-95 in a tight narrow band, average absorbed (10-year) doses are expected to be less than maximum centerline doses. Note in Table 3.1 the good agreement for the maximum



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Figure 3.1. Absorbed Radiation Doses Calculated at Indicated Points in Sorption Zones.

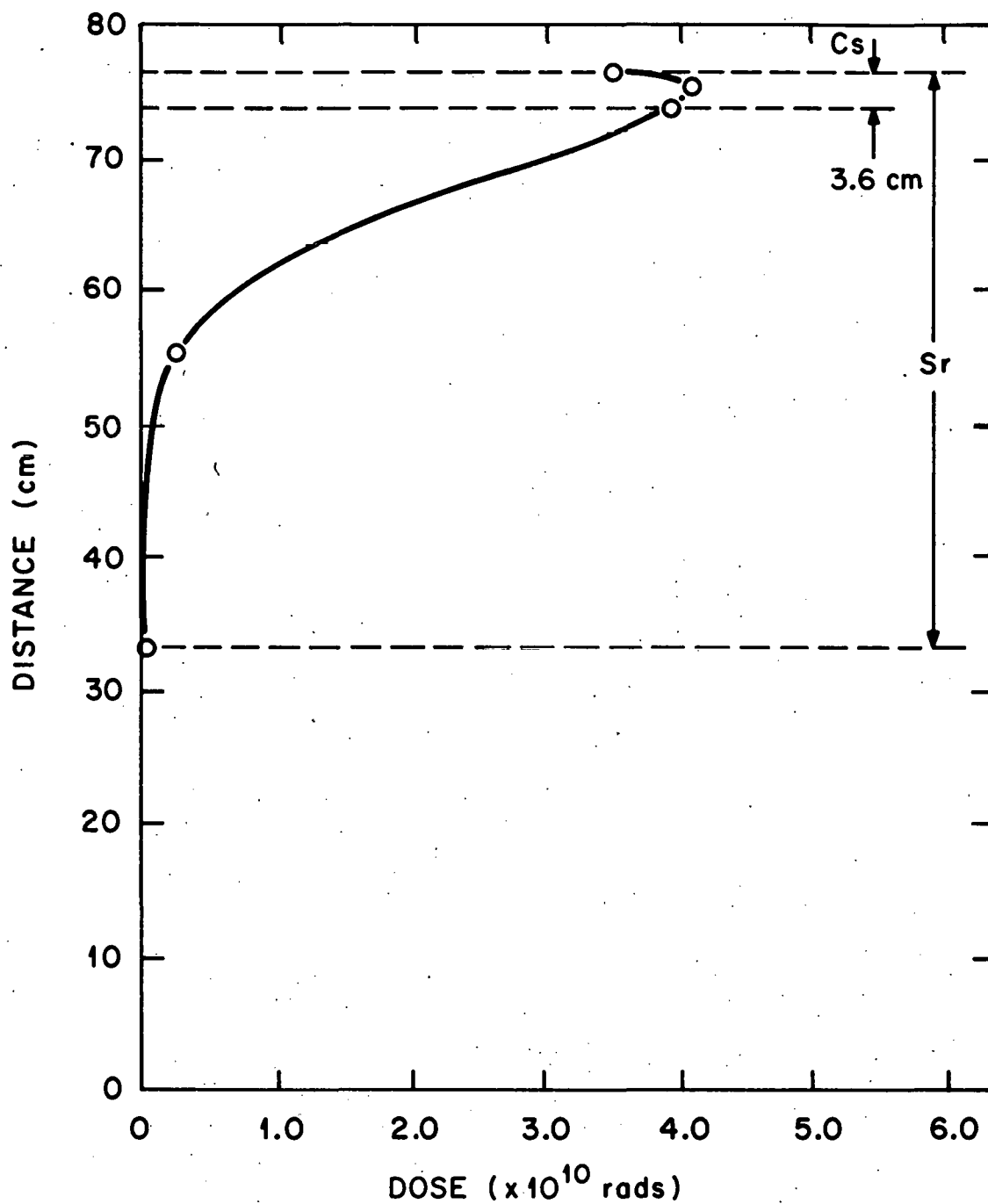


Figure 3.2. Axial Centerline Radiation Dose Profile - 10,000 Ci  $^{137}\text{Cs}$  Loading. (Cesium concentration is 6000 times feed concentration.)

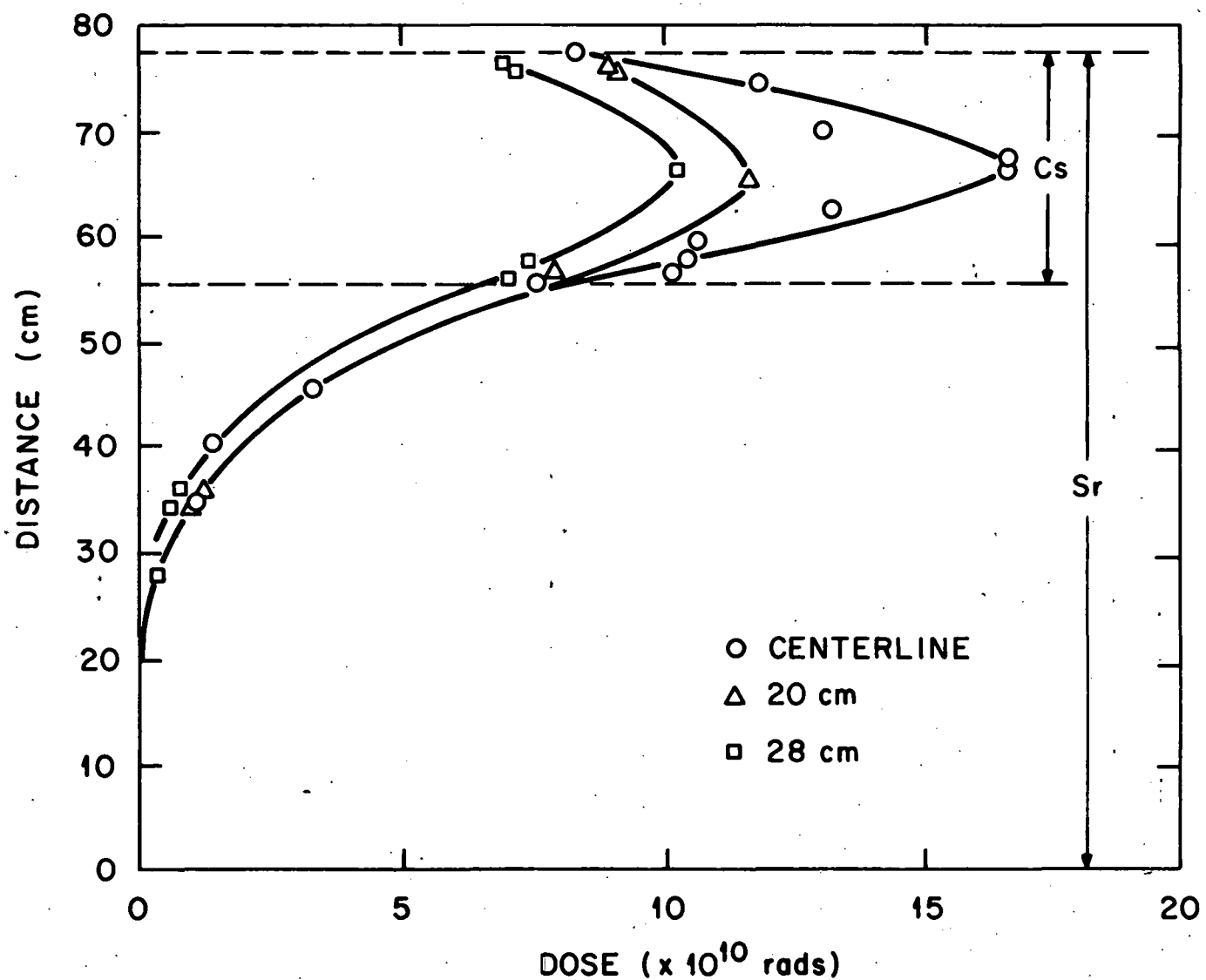


Figure 3.3. Axial Radiation Dose Profile at Various Radial Positions - 60,000 Ci  $^{137}\text{Cs}$  Loading. (Cesium concentration is 6000 times feed concentration.)

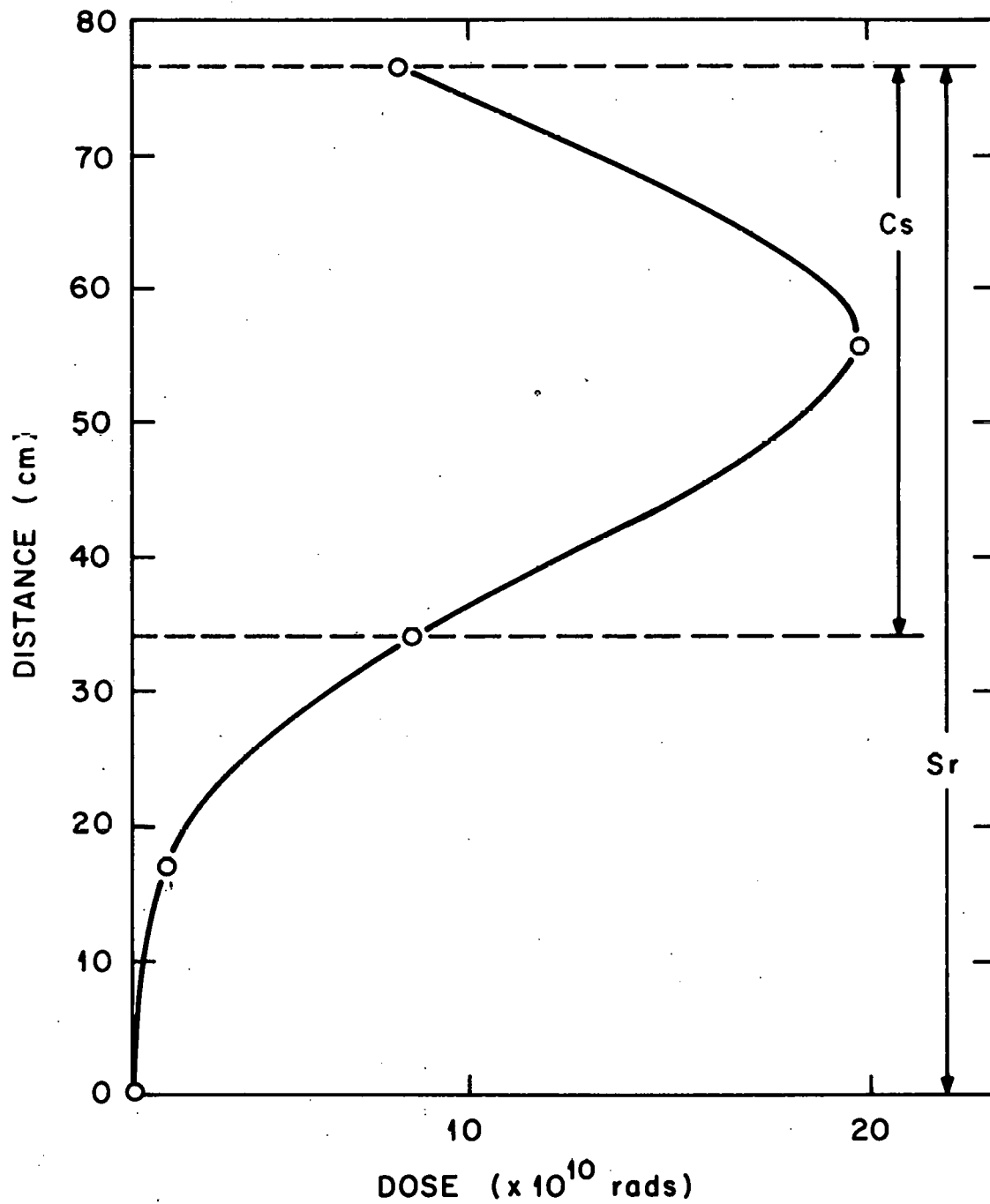


Figure 3.4. Axial Centerline Radiation Dose Profile - 120,000 Ci  $^{137}\text{Cs}$  Loading. (Cesium concentration is 6000 times feed concentration.)

Table 3.1. Estimated Radiation Doses and Heat Generation Rates for Cesium-Loaded SDS Zeolite Liners<sup>a</sup>

Cumulative <sup>137</sup> Cs (Ci)	Total Heat Generation Rate (W)	Absorbed Dose (rads) <sup>b</sup>		
		Loading Parameter <sup>c</sup>	Maximum Dose <sup>d</sup>	Average Liner Dose
10,000	61	4000 x Fd	$5.02 \pm 1.37 \times 10^{10}$	$4.77 \pm 0.01 \times 10^9$
		6000 x Fd	$4.15 \pm 0.22 \times 10^{10}$	$4.42 \pm 0.02 \times 10^9$
		8000 x Fd	$4.24 \pm 0.23 \times 10^{10}$	$4.39 \pm 0.03 \times 10^9$
60,000	363	4000 x Fd	$1.16 \pm 0.08 \times 10^{11}$	$3.76 \pm 0.01 \times 10^{10}$
		6000 x Fd	$1.66 \pm 0.19 \times 10^{11e}$	$3.56 \pm 0.01 \times 10^{10}$
		8000 x Fd	$1.99 \pm 0.13 \times 10^{11}$	$3.44 \pm 0.01 \times 10^{10}$
120,000	725	4000 x Fd	$1.05 \pm 0.05 \times 10^{11}$	$7.85 \pm 0.02 \times 10^{10}$
		6000 x Fd	$1.99 \pm 0.23 \times 10^{11}$	$7.66 \pm 0.02 \times 10^{10}$
		8000 x Fd	$2.35 \pm 0.16 \times 10^{11}$	$7.54 \pm 0.02 \times 10^{10}$

<sup>a</sup>Ionsiv IE-95 bed is 2 ft in diameter and 2.5 ft high.

<sup>b</sup>After a 10-year storage period.

<sup>c</sup>Fd = <sup>137</sup>Cs feed concentration.

<sup>d</sup>Along centerline of liner.

<sup>e</sup>Absorbed dose by Equation (1) is  $1.1 \times 10^{11}$  rads.

Note: A Monte Carlo calculation is a statistical calculation. The + and - figures on the above results are the statistical estimates of the precision of the results. Two results, x and y, can be considered to be equal if the range x - Δx to x + Δx overlaps the range y - Δy to y + Δy.

10-year doses for SDS liners loaded with 60,000 Ci of  $^{137}\text{Cs}$  as calculated by the Monte Carlo method and by Equation (1).

Additional Monte Carlo-type calculations were made (under the same conditions as were used to obtain data in Figures 3.2-3.4) for an SDS liner containing 60,000 Ci of  $^{137}\text{Cs}$  at a concentration about 6000 times that in CB sump water.\* In such a liner, 40 to 50% of the Ionsiv IE-95 would receive a radiation dose of at least  $10^{10}$  rads while 20% of the sorbent would receive a dose of at least  $10^{11}$  rads.

It must be emphasized that the absorbed radiation doses listed in Table 3.1 are for a 10-year storage period. Doses are approximately proportional to storage time. Thus, the maximum radiation dose for an SDS liner loaded with 60,000 Ci of  $^{137}\text{Cs}$  at an average concentration 6000 times the feed concentration will be about  $1.7 \times 10^{10}$  rads after storage for 1 year, about  $3.4 \times 10^{10}$  rads after storage for 2 years, and so on.

Heat generation rates, calculated on the basis that all the decay energy of the sorbed radiocesium and radiostrontium is deposited in the zeolite, are also listed in Table 3.1. These heat generation rates were used to calculate temperature profiles in the SDS liners using the multi-dimensional heat conduction code HEATING5.<sup>3</sup> After Swift,<sup>4</sup> the effective thermal conductivity of dried zeolite was taken as  $0.092 \text{ Btu h}^{-1} \text{ ft}^{-1} \text{ }^{\circ}\text{F}^{-1}$ . It was assumed that loaded SDS liners would be stored in water basins maintained at about  $80^{\circ}\text{F}$  ( $27^{\circ}\text{C}$ ). Peak SDS liner centerline temperatures, calculated at various liner loadings, and on the basis that all radiodecay energy is absorbed in the zeolite, are plotted in Figure 3.5. Actually, at least some of the decay energy will not be sorbed within the liners but

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\*The assumption is made in this report that the average  $^{137}\text{Cs}$  concentration in loaded SDS liners will be about 6000 times that in the CB sump water. Doses calculated at this  $^{137}\text{Cs}$  concentration are cited subsequently in the assessment of the radiation stability of loaded SDS liners.

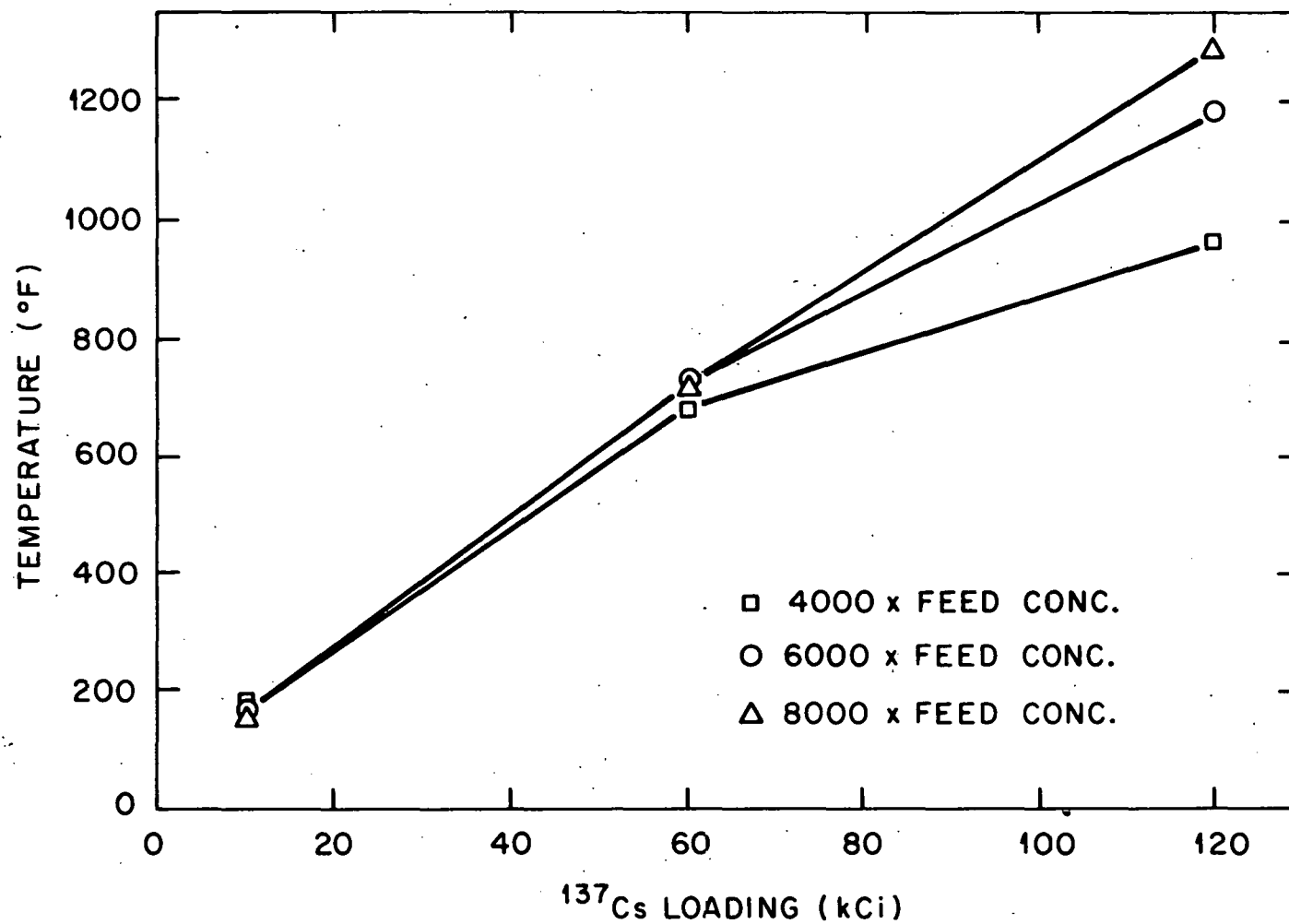


Figure 3.5. Conservative SDS Zeolite Liner Centerline Temperatures. (80 $^{\circ}\text{F}$  Water Storage, Variable Cesium Concentrations, and all Radiodecay Energy Absorbed in the Bed.)



will escape to the storage basin water. Hence, peak centerline temperatures of actual stored SDS liners will be substantially lower than those plotted in Figure 3.5.

### 3.2 Stability Considerations During Extended Storage

#### 3.2.1 Storage Modes

After washing with water, cesium-loaded SDS zeolite liners might be stored wet or dry (i.e., less than 0.4 wt % water). Storage containers loaded with wet SDS zeolite will have to be vented to a suitable off-gas system to remove steam and any radiolytically generated hydrogen and oxygen. In any event, wet SDS zeolite beds loaded with  $^{137}\text{Cs}$  should not be stored for prolonged periods in sealed casks because of the potential for container overpressurization from radiolytic decomposition of all the residual water to hydrogen and oxygen.

It appears technically feasible to ship, immediately after preparation, wet SDS zeolite liners in sealed casks from the TMI site to another location for prompt unloading and conversion to glass or for vented storage. Further analyses are required to ensure that shipments of wet SDS zeolite liners in sealed casks comply with all existing transportation and regulatory requirements. For the record, approximately 77 sealed casks containing kilocurie amounts of  $^{137}\text{Cs}$  were shipped from Hanford to ORNL at various times in the 1960s. Although all but two of the casks pressurized, no incidence of cask overpressurization was ever noted. Some of the casks contained wet AW-500\* zeolite containing 66 to 93 Ci of  $^{137}\text{Cs}$  per liter of sorbent. These concentrations are close to the average 44 Ci/L value for a SDS zeolite liner loaded with 10,000 Ci of  $^{137}\text{Cs}$ .

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\* Former name of Ionsiv IE-95.

### 3.2.2 Radiation Effects

The SDS zeolite liners loaded with 10,000 to 120,000 Ci of  $^{137}\text{Cs}$  and stored for 10 years will be subjected to radiation doses of about  $4.2 \times 10^{10}$  and  $2.0 \times 10^{11}$  rads, respectively (see Table 3.1). Effects of such irradiation on residual water and on the zeolite material itself are discussed in the next two subsections.

3.2.2.1 Radiolysis of Water. Water undergoes radiolytic decomposition to hydrogen and oxygen with a G value on the order of 1 to 2 molecules for each 100 eV of adsorbed energy. Removal of radiolytically generated hydrogen and oxygen will be accomplished by venting the liners to an off-gas system.

3.2.2.2 Laboratory and Plant Experience - Radiation Damage to Zeolites. Roddy et al.<sup>5</sup> have recently compiled and organized the extensive laboratory and plant-scale studies and experience involving irradiation of zeolites and related aluminosilicate materials. Some examples selected from this data base that are particularly relevant to exposures in the range  $1 \times 10^9$  to  $3.7 \times 10^{11}$  rads are briefly highlighted here; Roddy's report<sup>5</sup> should be consulted for details of these and other radiation studies.

Some of the most straightforward evidence of the high resistance of zeolites to attack by ionizing radiation is provided by the results of Fullerton<sup>6</sup> and Wallace.<sup>7</sup> Fullerton irradiated (with  $^{60}\text{Co}$ ) the mineral clinoptilolite to exposures as high as  $8.4 \times 10^9$  rads (absorbed dose). Clinoptilolite is a naturally occurring zeolite with an "open" structure and a high affinity for sorbing cesium. No effects of irradiation were detected by X-ray diffraction, differential thermal analysis, or cesium

distribution measurements. Wallace,<sup>7</sup> at the Savannah River Laboratory, recently irradiated (with  $^{60}\text{Co}$ ) air-dry Ionsiv IE-95 to an absorbed dose of  $1.1 \times 10^{10}$  rads. X-ray diffraction analyses and cesium distribution tests indicated no detectable effects of such irradiation.

Pillay<sup>8</sup> has irradiated, using various radiation sources, both wet ("drip-dry") and air-dry Ionsiv IE-95 zeolite to doses in the range  $1.9 \times 10^8$  to  $2.2 \times 10^9$  rads. Table 3.2 shows types and amounts of gases generated during irradiation of Ionsiv IE-95 containing various amounts of moisture. This information is taken directly from Pillay's report (p. 20). Regarding these data, Pillay<sup>8</sup> states:

The results presented in Table 7\* indicate that the primary source of gases in this matrix is the radiolysis of water within the zeolite matrix. The source of small amounts of methane identified in two samples (#31 and #33) is not well understood. The well known ability of molecular sieves to adsorb significant amounts of gases and the unique ability of zeolites to relatively retain large quantities of hydrogen account for the observed ratio of hydrogen and oxygen in the gas phase.

Pillay also irradiated columns of drip-dried Ionsiv IE-95, contained in aluminum tubes (2 cm diameter by 20 cm long), to a total dose of about  $2.2 \times 10^9$  rads. Flow test measurements showed that such irradiation did not affect the hydraulic behavior of the Ionsiv IE-95 exchanger.

There was much interest at Hanford in the early 1960s in storing kilocurie amounts of recovered and purified  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  on zeolites either for future recovery or for permanent disposal. Several laboratory-scale studies were performed to determine pressurization of sealed containers of  $^{90}\text{Sr}$ -loaded zeolites upon storage and to establish the ability to elute  $^{90}\text{Sr}$  from irradiated zeolites. Some useful guides to the radiation stability of zeolites can be inferred from these studies.

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\*Table 3.2 in this chapter.

Table 3.2. Gas Generation in Synthetic Zeolite (Ionsiv IE-95) during Irradiation<sup>a</sup>

Sample No.	Pretreatment	Pressure (psig)	Gases (%)		
			H <sub>2</sub>	O <sub>2</sub>	N <sub>2</sub>
31 <sup>b</sup>	Drip-dry	20.3	4.5	0.5	90.7
32	Drip-dry	18.0	1.3	32.5	66.2
33 <sup>b</sup>	Drip-dry (Cs-loaded) <sup>c</sup>	20.3	3.1	2.8	92.2
34	Drip-dry (Cs-loaded) <sup>c</sup>	20.0	1.0	36.9	62.1
35	Air-dry	13.9	0.9	29.2	69.9
36	Air-dry	----	0.6	24.1	75.4
37	Air-dry (Cs-loaded)	13.0	1.0	33.1	66.0
38	Air-dry (Cs-loaded)	11.6	0.6	33.4	66.1
Blank	Air-dry zeolite (nonirradiated)	13.9	----	20.2	79.8

<sup>a</sup>Data of Pillay.<sup>8</sup><sup>b</sup>Methane (3 to 4%) was identified in these two samples in addition to H<sub>2</sub>, O<sub>2</sub>, and N<sub>2</sub>.<sup>c</sup>The cesium loading on these samples was 0.6 meq per g of dry zeolite.

Thus, Mercer and Schmidt<sup>9</sup> have reported the details of an experiment performed by Van Tuyl at Hanford in which a sealed 304L stainless steel container (6.25 cm diameter by 38.8 cm high) of zeolite (Linde Type 4A) containing 10,000 Ci of <sup>90</sup>Sr was allowed to age approximately 2 years. The loaded zeolite material was dried at 600°C before the container was sealed. The amount of radiation absorbed by the zeolite is estimated to be about  $4 \times 10^{11}$  rads. (This estimated dose is calculated on the assumption that the zeolite absorbed all the beta decay energy of <sup>90</sup>Sr and its decay product <sup>90</sup>Y.) The sealed container was opened and flushed with about two column volumes of water, which removed less than 0.001% of the <sup>90</sup>Sr. More than 80% of the <sup>90</sup>Sr was eluted with 30 column volumes of 1 M HNO<sub>3</sub>.

Results of these elutions are considered highly significant since they demonstrate that zeolites retain their affinity for  $^{90}\text{Sr}$  even after irradiation to very high doses.

Mercer and Schmidt also discuss the results of another laboratory experiment attributed to DeMier, Martin, and Willingham at Hanford. In this test, a zeolite loaded with  $^{90}\text{Sr}$  was dried to about 1 wt % water, sealed in a 304L stainless steel container, and allowed to stand until the total absorbed dose was about  $10^{11}$  rads. The maximum pressure increase from radiolysis was 40 psig. This latter result signifies that significant breaking of Si-O bonds to release oxygen does not occur even at very high exposures.

Approximately  $3.6 \times 10^6$  Ci of  $^{137}\text{Cs}$  was shipped to ORNL from Hanford between 1961 and 1970. Specially designed stainless steel casks were used to transport the cesium. They contained DeCalso™, an amorphous aluminosilicate gel made by Pfaudler-Permutit (Cask STT-2); Zeolon 900™, a crystalline aluminosilicate zeolite similar to Ionsiv IE-95, but made by the Norton Company (Cask STT-3); and AW-500 (Cask HAP0 IC). The HAP0 IC cask was air-dried before shipment; all other casks were shipped without drying the zeolite. D. O. Campbell at ORNL estimates<sup>5</sup> that the DeCalso and Zeolon 900 beds received cumulative exposures of approximately  $3 \times 10^8$  and  $1.5 \times 10^8$  rads, respectively. Monte Carlo computer code calculations indicate that the AW-500 sorbent in the HAP0 IC cask was irradiated to a dose of about  $7 \times 10^9$  rads.

Finally, at Hanford during 1969 and 1970, a bed of AW-500 zeolite was used to remove approximately  $3.6 \times 10^7$  Ci of  $^{137}\text{Cs}$  from about  $6 \times 10^6$  gal of aged Purex process alkaline waste solution. The AW-500 bed

was 6 ft in diameter and 12 ft high and contained about 300 ft<sup>3</sup> of sorbent. Feed to the bed was at pH 10 to 11 and contained about 4 to 5 M NaNO<sub>3</sub>, 0.01 to 0.1 M NaAl(OH)<sub>4</sub>, 0.15 to 5 Ci of <sup>137</sup>Cs per gallon, and also other radionuclides including <sup>106</sup>Ru, <sup>106</sup>Rh, and <sup>99</sup>Tc. In each load-elution cycle, about 3 x 10<sup>5</sup> to 5 x 10<sup>5</sup> Ci of <sup>137</sup>Cs was loaded onto the bed and then eluted with a few column volumes of concentrated NH<sub>4</sub>OH-(NH<sub>4</sub>)<sub>2</sub>CO<sub>3</sub> solutions; each load-elution cycle required 4 to 5 days to complete. Because of mechanical problems discussed immediately below, the first AW-500 bed was replaced with a new 300 ft<sup>3</sup> of sorbent about midway through the total <sup>137</sup>Cs removal campaign. Each of the two AW-500 beds is estimated by Equation (1) to have been irradiated at the geometrical center of the sorption zone to a cumulative exposure of about 5.9 x 10<sup>8</sup> rads.

The first AW-500 zeolite bed was not replaced because of any perceived radiation degradation but because the retention screen at the bottom of the column ruptured, resulting in loss of the sorbent to load cycle effluent. Exact causes for the failure of the retention screen were not found. Particles of AW-500 sorbent recovered from load cycle effluent were slightly smaller in diameter than virgin AW-500 zeolite.

Other plant-scale experience at Hanford involves the final purification of <sup>137</sup>Cs in the B Plant T-38-5 column. This column, 1 ft in diameter by 12.75 ft high, is filled with Zeolon 900. The chemical composition of the feed to the column is approximately 0.8 M Na<sup>+</sup>, 0.1 M Cs, 0.01 M K<sup>+</sup>, 0.01 M Rb<sup>+</sup>, and 0.9 M NO<sub>3</sub><sup>-</sup>. A total of 7 x 10<sup>5</sup> to 8 x 10<sup>5</sup> Ci of <sup>137</sup>Cs is loaded onto the column in each cesium purification campaign. Each batch of Zeolon 900 processes between 3 x 10<sup>7</sup> and 3.5 x 10<sup>7</sup> Ci of <sup>137</sup>Cs before requiring replacement.

From April 1977 to December 1979, one batch of Zeolon 900 sorbent in the T-38-5 column processed  $3.2 \times 10^7$  Ci of  $^{137}\text{Cs}$  and received a calculated (Equation 1) radiation dose of  $2 \times 10^9$  rads. This estimate is slightly low because the T-38-5 column is in close proximity to a tank containing, at times, purified  $^{137}\text{Cs}$  which also irradiates the Zeolon 900. Some caution is warranted in extrapolating experience with the Hanford Zeolon 900 sorbent to performance of SDS zeolite liners. For example, the Zeolon 900 bed was wet throughout its operating life, while SDS zeolite beds may eventually be dried.

### 3.2.2.3 Theoretical Considerations - Radiation Damage to Zeolites.

Simple theoretical considerations show that inorganic zeolite sorbents should be much less susceptible to damage from fission product radiation than are organic sorbents. Thus, such radiation consists of beta and gamma emissions which interact with electrons, not nuclei, of the sorbent. Displacement of electrons produces radicals, ions, and so forth. Organic materials are not stable to such reactions because bonds are ruptured and new ones form, resulting in major structural changes. In zeolite, however, all metal atoms are bonded by four oxygen bridges; these multiply and form a particular crystal structure and pore configuration that are very resistant to damage by beta and gamma radiation.

Laboratory and plant-scale radiation experience which shows that zeolites are not significantly damaged by irradiation to exposures as high as  $4 \times 10^{11}$  rads is clearly in agreement with the theoretical considerations just enunciated. From a theoretical standpoint also, there is good reason to expect that Ionsiv IE-95 will not suffer significant radiation damage during a 10-year storage period. Even if Ionsiv IE-95 did

begin to undergo observable radiation damage at doses above  $10^{11}$  rads, the principal effect expected would be conversion (metamictization) of some of the crystalline chabazite at the center of the liner to an amorphous material. Transformation from a crystalline structure to an amorphous form would be a very slow process not likely to be accompanied by evolution of gases. The amorphous material would contain essentially the same chemical elements as the original crystalline Ionsiv IE-95 and thus should be suitable for vitrification. Radiocesium and radiostrontium originally trapped in cages in the crystalline lattice may migrate to undamaged Ionsiv IE-95 during the metamictization process. In any event, there is reason to believe that the SDS zeolite will satisfactorily retain all radioactive species for at least a 10-year storage period.

### 3.2.3 Thermal Effects

3.2.3.1 Steam Generation. The heat energy generated in SDS liners loaded with 60,000 to 120,000 Ci of  $^{137}\text{Cs}$  may be sufficient to vaporize any water left in the liner. Work by DeMier<sup>10</sup> demonstrates that "wet" zeolites can retain very large amounts of water. For example, in one particular set of experiments, DeMier saturated and washed zeolite beds ( $0.96 \text{ ft}^3$ ) with water prior to flowing dry air through them. He states:

At this time the test container holds about 11.5 pounds of adsorbed water, 15.6 pounds of interstitial water, 15.6 pounds of "sponge" water (that is, water held within the granules but not actually absorbed) and approximately 10 pounds of "loose" water in the lines and in the container below the support screen.

Storage of wet SDS liners may require equipment to condense and collect water vapor.

3.2.3.2 Thermal Degradation of Ionsiv IE-95. From data plotted in Figure 3.5, peak centerline temperatures of SDS liners loaded with 60,000



and 120,000 Ci of cesium, respectively, would be about 700 and 1290°F if all the radiodecay energy were absorbed in the zeolite. Since, as already noted, at least some of the decay energy will escape the liners, centerline temperatures of actual stored SDS liners will be well below the calculated values.

Ionsiv IE-95 is a molecular sieve made of the crystalline aluminosilicate mineral chabazite. Limited data concerning the thermal stability of molecular sieves indicate that even a temperature of 1290°F is still below that where any kind of structural change starts to occur. Thus, according to Swift:<sup>4</sup>

No particular structural damage occurs to molecular sieves at temperatures up to 1300°F. At 1650°F and above, crystalline structure collapse occurs with an apparent shrinkage of about 50 percent and with surface glazing.

Collapse of the zeolite structure in stored SDS liners, although not anticipated, would likely be beneficial rather than deleterious. Thus, such collapse could entrap and further immobilize  $^{137}\text{Cs}$  in a high leach-resistant ceramic body.

**3.2.3.3 Volatilization of Cesium.** Little, if any, cesium is expected to volatilize during the storage of  $^{137}\text{Cs}$ -loaded SDS liners. This conclusion is based on experimental evidence cited in Chapter 6, which shows that less than 0.1% cesium volatilized when Ionsiv IE-95 loaded with inert cesium was vitrified at 1050°C. Also, any cesium that evaporated from the hottest part of the SDS liner would probably condense on the cooler parts of the zeolite bed or upon liner walls. Also, small amounts of  $^{137}\text{Cs}$  in the vapor space of sealed SDS liners would not present any problem.

### 3.2.4 Liner Corrosion

SDS zeolite liners are cylindrical 316L stainless steel vessels with 3/8-in. walls. Because of the excellent corrosion resistance of 316L stainless steel, no significant corrosion of the liners is expected during their extended storage.

Stainless steel casks used to ship cesium-loaded zeolites to ORNL from Hanford between 1961 and 1970 did not undergo any visible damage.<sup>5</sup> Some of these casks were used for many shipments. But, of course, in each shipment the cesium-loaded zeolite was only in contact with steel surfaces for a few days.

No corrosion failures or problems with 304L stainless steel equipment were ever experienced in any Hanford plant- or laboratory-scale cesium ion exchange column operations.

### 3.3 Stability Considerations During Load Cycle

The principal focus in this chapter is on the stability of loaded SDS liners during storage. It is of interest, nevertheless, to consider briefly some potentially troublesome phenomena (e.g., gassing, column plugging, chemical attack, etc.) which could conceivably occur during the loading of  $^{137}\text{Cs}$  onto the SDS zeolite beds. Experience at Hanford in recovering  $^{137}\text{Cs}$  from alkaline Purex wastes, cited earlier in this chapter, as well as some later Hanford plant-scale experience in purifying recovered  $^{137}\text{Cs}$  by loading on beds of Zeolon 900 (Roddy et al., 1981), is particularly germane.<sup>5</sup>

Excessive evolution of gases frequently occurs during ion exchange separation and recovery of highly active alpha emitters. However, because

of the much lower radiation levels that will prevail during loading of the SDS liners, no gassing problems are anticipated.

What is believed to be chemical, rather than radiolytic, attack has been observed in two different Hanford plant-scale applications of Zeolon 900 for  $^{137}\text{Cs}$  removal. No such attack, however, has ever been observed in plant-scale applications of AW-500 (Ionsiv IE-95) zeolite. Degradation of Zeolon 900 observed at Hanford has manifested itself in deterioration of the amorphous silicate material which binds crystals of mordenite with concomitant decreases in overall sorbent particle size and increases in resistance to flow. At least part of the observed chemical attack is attributed to reaction of the binder material with ethylenediaminetetraacetic acid (EDTA) and hydroxy-N-ethylenediaminetetraacetic acid (HEDTA), two chelating agents which are present to some extent in many Hanford liquid wastes.

The TMI-SDS employs Ionsiv IE-95 exchanger and not Zeolon 900; furthermore, organic complexing agents are not found in the TMI contaminated water. Hence, chemical degradation behavior similar to that noted at Hanford is not expected in SDS operation.

### 3.4 Findings

Various synthetic and naturally occurring zeolites, including Ionsiv IE-95, have been irradiated in laboratory-scale tests to exposures in the range  $10^{10}$  to about  $4 \times 10^{11}$  rads without undergoing either significant structural damage or decreased affinity for sorbed cesium or strontium. Judging from these bench-scale results and also extensive DOE plant-scale experience at lower radiation doses, SDS liners containing 60,000 to

120,000 Ci of radiocesium and radiostrontium are expected to be satisfactorily stable to ionizing radiation for storage times as long as 10 years. Calculated radiation exposures of SDS liners loaded with 60,000 and 120,000 Ci of activity after 10 years of storage are estimated to be about  $1.7 \times 10^{11}$  and  $2.0 \times 10^{11}$  rads, respectively.

Elementary theoretical considerations of the effects of ionizing radiation upon zeolite sorbents support the finding that SDS liners containing 60,000 to 120,000 Ci of  $^{134},^{137}\text{Cs}$  and  $^{90}\text{Sr}$  can be satisfactorily stored for at least 10 years. From a theoretical standpoint there is good reason to believe that Ionsiv IE-95 may not suffer observable radiation damage even at exposures considerably above  $10^{11}$  rads. Even if it did, the principal expected effect would be a very slow conversion of some of the crystalline chabazite at the center of the liner to an amorphous form without significant evolution of any gases. The resulting amorphous form would be expected to be suitable for eventual conversion to a borosilicate glass.

SDS liners loaded with 60,000 to 120,000 Ci of radioactive cesium and strontium are expected to be thermally stable during storage. Calculated maximum centerline temperatures, 700°F and 1290°F, respectively, of liners loaded with 60,000 and 120,000 Ci are below the temperature (1300°F) where structural changes in molecular sieves such as Ionsiv IE-95 are reported to occur. (Although not anticipated, collapse of the zeolite structure in stored SDS liners, which might begin to occur at 1300°F, would likely be beneficial rather than deleterious.) At temperatures of 700° to 1290°F little, if any,  $^{137}\text{Cs}$  is expected to volatilize during the storage of cesium-loaded SDS liners.

Some slight corrosion of the inner liner walls may occur during the storage of  $^{137}\text{Cs}$ -loaded SDS liners. Effects of such corrosion will be minimized because of the selection of highly corrosion-resistant material (316L stainless steel) for construction of the liners and specification of extra thick (3/8-in.) liner walls.

Loaded SDS zeolite liners will contain considerable amounts of water; radiolysis of the water during storage will generate hydrogen and oxygen. Evolved hydrogen and oxygen gases can be dispersed by venting the stored SDS liners to an appropriate off-gas system, as presently planned. Wet SDS liners loaded with strontium and cesium should not be stored for prolonged periods in sealed containers because of the potential for overpressurization by radiolytically generated gases.

## 3.5 References

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## 4.0 ON-SITE HANDLING

### 4.1 Introduction

The objective of this chapter is to examine the handling of highly loaded zeolite SDS liners at TMI-2 in order to identify any constraints which could limit the loading of the liners. However, it should be recognized that there will be additional liners containing mechanical filters, second- and third-stage zeolite filters, and pool containment cleanup filters. With the possible exception of those liners which contain the mechanical filters, all of the liners are expected to contain relatively low amounts of radioactive material. These are not included in this study.

### 4.2 Present Handling Sequence

A discussion of the planned handling sequence will provide a better understanding of the impact of high loadings of the SDS liners.

Initially, six liners are arranged for two flow paths, with three liners in each path. It is anticipated that the first-stage liner(s) will be loaded first and then removed to storage. The second- and third-stage liners will then be advanced and a new liner installed in the third-stage position. After processing, the loaded liners are stored in racks, which can accommodate a total of 60 liners. The rack design calls for 36 liners on the bottom of the spent fuel pool and 24 liners in a second-layer rack.

All of the liners are held within a leakage containment "box" while being loaded; this arrangement helps prevent the spread of radioactivity during coupling and uncoupling. Each containment box has a cleanup ion exchanger.

During processing, a minimum of 15 ft of water will cover each liner. During transfer to storage racks, however, it is necessary to lift the liners approximately 5 ft; thus, the minimum water coverage becomes 10 ft for a short period of time.

Following storage at the bottom of the pool for an undefined period, each liner is transferred (underwater) to a dewatering station for removal of the sponge water (cf. page 41). After being dewatered, the liner is loaded into the cask and the cask lid installed. The cask is removed from the pool, then drained, decontaminated as required, and surveyed. The Fuel Handling Building crane transfers the cask to a suitable location for installation of the shock limiters and then positions it on the transporting vehicle. All of these handling operations will be performed inside the Fuel Handling Building.

A likely alteration in the handling sequence is related to preventing pressurization of the liner due to radiolytic decomposition of water. Venting capability is currently being installed.

#### 4.3 Effects of High-Curie Loading on Operation

An analysis was made to establish radiation levels at the surface of the pool, since this would represent the closest proximity of personnel to the liner. The calculations indicate that an array of more than nine liners is equivalent to an infinite number of liners. The calculations further indicate that, with 10 ft of water above the source, 120,000 Ci/liner will result in a field of less than 1 mR/h. Thus, high-curie loadings on the liners would result in insignificant radiation effects to individuals during normal operations.



#### 4.4 Effects on Maintenance

Maintenance operations conducted above water results in the same exposures as indicated in Section 4.3. However, if underwater maintenance became necessary, removal of the liners in the vicinity of the work area would be required. About 2 ft of water would provide sufficient shielding to reduce the dose rate from a vessel loaded with 120,000 Ci to that of a vessel loaded with 10,000 Ci. It is unlikely that dose rates would be prohibitively high once the vessels are moved from the area; however, if this were the case, lead (plate) shielding could be lowered to protect maintenance personnel.

#### 4.5 Cask Loading and Cask Selection

Calculations presented in Chapter 5 indicate that many available spent fuel casks are suitable to ship the SDS liners. The cask examined in detail for the loading evaluation is the Chem-Nuclear CNS 1-13C cask. The desired features of this cask are: (1) it is relatively lightweight so that truck transport with the two containers per truck may be possible; (2) it appears to be suitable for transporting this type of material (by-product material); (3) the SDS processing system is being constructed, assuming that this particular cask will be used; and (4) Chem-Nuclear is in the process of upgrading the certificate of compliance for this cask. Limitations of this cask include relatively weak shielding and, at least for the present, a maximum permissible heat source of 600 W.

Several other truck-mounted casks can accommodate the SDS liners. Some of the problems associated with the employment of these casks are identified in Appendix B.

As demonstrated in Chapter 5, an alternate cask for the SDS liners may be required only for loadings in excess of 60,000 Ci. Furthermore, the use of other casks, such as CNS 4-45 and CNS 3-55, would require modifications to the TMI-2 pool equipment, due to their lengths. These modifications are possible (if completed before SDS startup) and would cost approximately \$25,000, but would not be made unless it was decided to load to 120,000 Ci per liner.

#### 4.6 Findings

Effects upon personnel exposure during the handling of more highly loaded liners are not expected to be of any significant consequence; in fact, exposure may be decreased by reducing the required number of transfers of the liners to the storage modules. The strontium-loaded liners likewise do not appear to present any handling problems.

The CNS 1-13C cask appears to be suitable for the shipment of liners loaded to 60,000 Ci. The use of other casks is possible, but efforts would have to be made to modify the pool loading rack and to address uncertainties associated with the Certificates of Compliance of the alternate casks.

## 5.0 TRANSPORTATION OPTIONS

### 5.1 Introduction

The effects of the extent of loading of the SDS liners on transportation are discussed in this chapter. Four separate transportation segments are considered; two different distances are assumed, and two transportation modes are assessed, for each segment. The four segments are as follows:

- (1) TMI reactor to waste processor (WP),
- (2) WP to spent fuel away-from-reactor (AFR) storage facility or return to TMI reactor for storage,
- (3) storage to terminal geologic repository, and
- (4) WP to terminal geologic repository.

Four liner loading levels and two disposal methods are considered.

Since specific sites for the WP, AFR, or geologic repository have not been specified, a "near" shipping distance of 250 miles and a "far" shipping distance of 2500 miles have been arbitrarily assumed. An assessment of the impact of specific routings on the logistics of movement, costs, and public exposure was not possible; therefore, such assessments were made only on a generic basis using the near and far distances. Finally, for each shipping segment and distance, the availability of either truck or rail mode was assumed so as to not preclude the use of any shipping systems which might be available.

Only existing hardware was included for the study. If the time period when shipments are made extends beyond the near term, other systems may become available or may be designed, certificated, and fabricated for this specific purpose; it is also possible that some of the systems presently in existence which are identified in this study

might become unavailable. The assessment was not limited to waste casks, but also included existing spent fuel casks which could be certificated by amendment to carry the waste forms under consideration.

In the detailed assessment of transportation, the following factors were considered:

- o Applicability
- o Licensability
- o Availability
- o Cask/plant Interface
- o Personnel Exposure
- o Public Exposure
- o Schedule
- o Cost

## 5.2 Cask Selection

Four principal constraints govern the applicability of a given cask: (1) the package must be currently certificated by NRC as type B, (2) the dimensions of the containment cavity must accept the SDS zeolite liners or the vitrified waste canisters, (3) the shielding must be sufficient for at least the 10,000 Ci-per-liner loading level, and (4) the cask must be compatible with shipping and receiving facilities.

The first constraint, coupled with the second constraint, narrowed the list of possible casks to approximately 20. To assess the shielding adequacy of candidate casks, an estimate of dose rate from the loaded SDS liners and from the vitrified waste canisters was needed. The shielding calculations for the high-activity-level strontium/cesium liners and vitrified waste canisters are summarized in Sections 5.2.1 and 5.2.2.

### 5.2.1 One-Dimensional Shielding Assessment

Initially, simplified one-dimensional calculations were made to expedite the cask assessment and selection. The isotope composition reported elsewhere,<sup>1</sup> which is for July 1980, was used in these calculations. As a result, these calculations overestimate the shielding requirements.

Despite these limitations, the one-dimensional calculations provide an insight into which parameters are important. SDS liner loadings of 10,000, 60,000, and 120,000 Ci were assumed, and vitrified waste canisters, 8 in. diam by 8 ft long, were assumed, where 1.75 canisters result from processing one liner. The following composition was assumed for each 10,000 Ci in a liner:

<sup>89</sup> Sr	30
<sup>90</sup> Sr	120
<sup>134</sup> Cs	1,410
<sup>137</sup> Cs	8,440
	<hr/>
	10,000 Ci

Also, based on the results reported previously,<sup>1</sup> it was assumed that 75% of the loading is in a 6-in.-long band at the top of the zeolite bed.

For the vitrified waste canisters, the fission products were assumed to exist in the glass as a homogeneous mixture.

The SANDIA-ORIGEN code<sup>2</sup> was used to obtain estimates for the radiation source characteristics of the zeolite and vitrified waste forms.

The ANISN discrete ordinates transport code<sup>3</sup> was utilized to obtain estimates of the thicknesses of lead required to obtain regulatory dose

rate levels external to the cask of either 200 mR/h at the surface or 10 mR/h at 2 m from the surface.

It is reemphasized that these shield thickness estimates for the SDS liners are conservative. First, in each shielding calculation, the source region was modeled as a radiation-emitting air space. Thus, shielding effects of the waste itself, although probably small, were ignored. Second, the calculations were done in one-dimensional cylindrical geometry. Modeling the problem in this geometry, in effect, transforms the radiation source into an infinitely long cylinder. This results in the greatest inaccuracies for the SDS liners. More realistic, two-dimensional shielding calculations would result in shield thicknesses approximately 1 in. less than those calculated with the one-dimensional model. Since the SDS vitrified waste canisters have a large length-to-diameter ratio, the infinite-cylinder effect is probably of little importance in this case. Third, the age of the waste could be significant since the major constituents are  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ , which have significantly different half-lives (2.062 years for  $^{134}\text{Cs}$  and 30.17 years for  $^{137}\text{Cs}$ ). As the waste ages, the composition of the isotopic mixture will change and the shielding requirements will decrease.

Table 5.1 summarizes the results of the one-dimensional calculations for the three liner loading levels considered. An estimated realistic value of the required shield thickness for the SDS liner is shown. It was obtained by subtracting 1 in. from the conservatively calculated value.

Table 5.1. Estimated Lead Shielding Requirements to Reduce Dose Rate to 10 mR/h at a Distance 2 m from the Lead Surface<sup>a</sup>

SDS Liner Loading (Ci/liner)	Lead Shielding Requirements (in.)	
	SDS Liner	Vitrified Waste Canister
20,000	6.0	5.0
60,000	7.4	6.4
120,000	7.9	6.8

<sup>a</sup>Uses July 1980 isotopic mixture of strontium and cesium. Based on one-dimensional calculations.

### 5.2.2 Two-Dimensional Shielding Assessment

As the efforts of the Task Force progressed, the need was established for a clearer shielding definition, using more realistic values of isotopic mixes, cesium band width, and two-dimensional calculations. Therefore, the loading scenarios described in Chapter 2.0 were used to more accurately represent the loading situations that might occur.

It was assumed that the strontium-cesium mixture shown in Table 2.1, given as of July 1981, would be loaded in the liners. The resulting loaded liners would then be stored until January 1982 prior to shipping. The isotopic compositions for a liner loaded to a total of 10,000 Ci in July 1981 and aged to January 1982 is shown in Table 5.2. Liners loaded to other levels (22,000, 60,000, or 120,000 Ci) would have compositions scaled linearly with the loading level.

The gamma spectrum for this isotopic mixture was determined by hand calculation. The nuclides <sup>89</sup>Sr, <sup>90</sup>Sr, and the daughter <sup>90</sup>Y produce no

gammas upon decay.<sup>4</sup> The significant gamma energies produced by the disintegration of the cesium isotopes are indicated in Table 5.3.

Using the energy structure of an available P3 eight-group cross-section set, the gamma source spectrum as of January 1, 1982, for an SDS liner originally loaded to 10,000 Ci on July 1, 1981, is given in Table 5.4. Also included in the table are the corresponding dose-rate conversion factors by energy group used in the two-dimensional calculations.

Table 5.2. Isotopic Composition of an SDS Liner Loaded to 10,000 Ci on July 1, 1981, and Shipped on January 1, 1982

Isotope	Estimated SDS Liner Content (Ci)	
	When Loaded (July 1, 1981)	When Shipped (January 1, 1982)
<sup>89</sup> Sr	0.5	0.04
<sup>90</sup> Sr	398.5	393.8
<sup>134</sup> Cs	1,016	858.8
<sup>137</sup> Cs	8,585	8,487
	10,000	9,740

Table 5.3. Gamma Yield Data for SDS Liner Cs-Isotopes

Isotope	Gamma Energy (MeV)	Yield (Gammas per Disintegration)
<sup>134</sup> Cs	1.37	0.03
	1.17	0.02
	0.80	0.90
	0.605	0.98
	0.57	0.24
<sup>137</sup> Cs (via <sup>137</sup> Ba)	0.662	0.95



Table 5.4. SDS Liner Gamma Source Spectrum<sup>a</sup> and Dose Rate Conversion Factors

Gamma Average Energy (MeV)	Gamma Emissions per Second	Dose Rate Factor (mR/h-gamma-cm <sup>2</sup> /s)
1.25	$9.534 \times 10^{11}$	$2.32 \times 10^{-3}$
1.025	$6.356 \times 10^{11}$	$2.01 \times 10^{-3}$
0.80	$2.860 \times 10^{13}$	$1.68 \times 10^{-3}$
0.70	$2.983 \times 10^{14}$	$1.52 \times 10^{-3}$
0.60	$3.877 \times 10^{13}$	$1.36 \times 10^{-3}$
0.50		$1.15 \times 10^{-3}$
0.40		$9.85 \times 10^{-4}$
0.30		$7.59 \times 10^{-4}$
Total	$3.673 \times 10^{14}$	

<sup>a</sup>Source is for 10,000 Ci of strontium-cesium mixture loaded on July 1, 1981, and decayed to January 1, 1982.

The MORSE-SGC Monte Carlo Code<sup>5</sup> was used to perform the two-dimensional transport calculations. This code has the advantage of allowing a very detailed modeling of the actual geometry under consideration without the core storage constraints which result in limitations when two-dimensional discrete ordinates codes are used. A disadvantage is that MORSE results are statistical in nature; and, for deep penetration (thick shield) problems, statistically significant results are difficult to obtain in reasonable run times. This difficulty is enhanced when the energy of the radiation being considered is small (approximately 1 MeV).

The geometry considered in the two-dimensional calculations is essentially that of the CNS 1-13C cask. The outer dimensions of this cylindrical cask, as utilized in the numerical model, were 68.5 in. high by 39 in. in diameter. The cask shielding thickness used, in terms of lead equivalent, were 5.8 in. on the side, 6.6 in. on the top, and 6.8 in. on the bottom. These shield thicknesses result in a 55-in.-high, 27.5-in.-diam cask cavity in the model (which is larger than the cavity of the actual cask). This disparity is of no significant consequence since only dose rates external to the cask are significant. For modeling purposes, the walls of the cask were taken to be lead, and the cavity and the cask exterior were assumed to be air.

The cylindrical source zone in the cask cavity was 24 in. in diameter, and its upper surface was 30.8 in. from the bottom of the cask cavity. The depth of the source varied with liner loading (see Chapter 2). The depths used, as a function of initial loading, are given in Table 5.5.

Table 5.5. SDS Liner Source Zone Depth<sup>a</sup> for Two-Dimensional Shielding Calculations

Initial Loading (Ci)	Percent of Zeolite Bed Length	Depth of Source Zone (in.)
10,000	3	0.92
22,000	8	2.45
60,000	21	6.43
120,000	41	12.56

<sup>a</sup>Source zone is cylindrical with a diameter of 24 in.

Dose rates were estimated both at the surface and at 2 m from the surface on the side, top, and bottom of the cask. The statistically significant results, as a function of liner loading, are given in Table 5.6. It is recommended that a safety factor of 2, relative to dose rate, be used in interpreting these results. For instance, when the liner loading is 60,000 Ci initially, the surface and 2-m dose rates at the side of the cask should be taken as 160 mR/h and 7 mR/h, respectively, rather than 80 and 3.5 mR/h as reported in Table 5.6.

The results of the shielding calculations for the liners are summarized in Table 5.7. Cask selection for the liners is based on the results of the two-dimensional calculations.

Cask selection for the vitrified waste canisters is based on the conservative values shown in Table 5.1, where shielding requirements have been adjusted by using the results of the detailed liner calculations. It is estimated that 5 in. of lead equivalent shielding will be sufficient for shipment of any of the vitrified waste canisters.

Table 5.6. Estimated Dose Rates (mR/h) for One SDS Liner in the CNS 1-13C Cask

Initial Liner Loading (Ci)	Cask Location			
	Side Surface	2 m <sup>a</sup>	Top Surface	Bottom Surface
10,000	13	0.4	1.3	0.1
22,000	31	0.9	3.1	1.2
60,000	80	3.5	11	1.9
120,000	120	6.5	35	17

<sup>a</sup>Dose rate given 2 m from the external surface of the cask opposite the center of the cesium source zone.

Table 5.7. Comparison of Estimated Lead Shielding Requirements for SDS Liners

Initial SDS Liner Loading (Ci/liner)	Type of Calculation	Depth of Source Zone (in.)	Date When Liner Loaded	Date When Liner Shipped	Lead Shielding Requirements (in.)
10,000	One-D	6 <sup>a</sup>	7/80	7/80	6.1
10,000	Two-D	0.92	7/81	1/82	5.0
22,000	Two-D	2.45	7/81	1/82	5.5
60,000	One-D	6 <sup>a</sup>	7/80	7/80	7.4
60,000	Two-D	6.43	7/81	1/82	5.7
120,000	One-D	6 <sup>a</sup>	7/80	7/80	7.9
120,000	Two-D	12.56	7/81	1/82	6.0

<sup>a</sup>Assumed 75% of loading in source zone.

### 5.2.3 Shielding for Low-Level Strontium Liners

A final consideration is the shielding requirements of liners uniformly loaded with a  $^{89}\text{Sr}$ - $^{90}\text{Sr}$  mixture in the ratio of 1:800. Two different total initial loadings of strontium, 500 and 2000 Ci, were assumed. It is assumed that liners with these loadings will be aged 6 months before being shipped off-site. Table 5.8 shows the estimated isotopic content of a liner for each of these initial loadings.

The  $^{89}\text{Sr}$ ,  $^{90}\text{Sr}$ , and the daughter  $^{90}\text{Y}$  isotopes disintegrate only by beta decay. The maximum beta energy for each isotope is, respectively, 1.46, 0.544, and 2.27 MeV.<sup>4</sup>

Table 5.8. Isotopic Contents of Strontium-Contaminated SDS Liners

Isotope	Initial Loading (Ci)	6-Month Loading (Ci)	Total Sr Activity (Ci)	
			Initial	After 6 Months
$^{89}\text{Sr}$	0.6	0.05		
$^{90}\text{Sr}$	499.4	493.4	500	493.5
$^{90}\text{Y}$	499.4	493.4		
$^{89}\text{Sr}$	2.5	0.2		
$^{90}\text{Sr}$	1997.5	1973.8	2000	1974
$^{90}\text{Y}$	1997.5	1973.8		

$^{90}\text{Y}$  has a half-life of 64 h and is in equilibrium with  $^{90}\text{Sr}$ .

The shielding requirements for liners loaded with these beta emitters are determined by two factors: the shielding necessary to eliminate the beta radiation, and that required to attenuate the bremsstrahlung radiation. A thin layer of steel or lead would effectively stop all the beta radiation. It is believed that about 2 in. of lead would be sufficient to reduce the radiation flux, for either liner loading, to acceptable levels. This conclusion is based on the observation that the average energy of the bremsstrahlung generated as an electron slows down is only about 10% of the initial energy of the electron. Only a small fraction of the electrons emitted during beta decay has the maximum energy associated with the decay. Thus, for the present situation, most of the bremsstrahlung will have energies of only a few hundred kilovolts. Two inches of lead should be sufficient to attenuate this radiation.

#### 5.2.4 Casks for Each Waste Form

The results of the preceding shielding calculations demonstrate that a cask must have a side-wall shielding capability of at least 5.0 in. in order to be capable of transporting SDS liners loaded to the 10,000-Ci level.

Casks that are capable of carrying SDS liners loaded to 10,000 Ci are listed in Table 5.9. This list was developed on the basis that the cask has sufficient cavity volume and the necessary effective shielding capability.

Table 5.9. Casks Capable of Transporting 10,000-Ci SDS Liners

Cask	USA Certificate Number	Cavity ID (in.)	Cavity Length (in.)	Equivalent Lead Thickness (in.)
CNS 1-13G	9044/B( )F	26.5	54.0	7.0
CNS 1-13C	9081/B( )	26.5	54.0	5.7
Vandenburgh Cask, CNS 3-55	5805/B( )F	36.0	116.75	7.0
CNS 4-45	6375/B( )F	26.0	159.0	6.5 (ends) 7.5 (side)
PB-1	6375/B( )F	26.0	159.0	6.5 (ends) 7.5 (side)
GE IF-300	9001/B( )F	37.5	180.2	6.1
NLI-10/24	9023/B( )F	45.0	179.5	7.7

Another factor which must be considered in selecting a cask is its capability for rejecting decay heat. The estimated heat output of the highly loaded cesium/strontium liners has been presented in Table 3.1.

Casks capable of carrying the SDS liners at different loadings are summarized in Table 5.10. All of the casks shown in these tables are discussed in detail in Appendix B.

Table 5.10. Effect of Liner Loading on Cask Capability  
Based on Shielding and Heat Output Requirements<sup>a</sup>

Cask	Ci per Liner			
	10,000	22,000	60,000	120,000
CNS 1-13G	Y	Y	Y	N (2)
CNS 1-13C	Y	Y	Y	N (1,2)
Vandenburgh Cask CNS 3-55	Y	Y	Y	Y
CNS 4-45	Y	Y	Y	Y
PB-1	Y	Y	Y	Y
GE IF-300	Y	Y	Y	I
NLI-10/24	Y	Y	Y	Y

<sup>a</sup>Y = Yes; cask has sufficient shielding.

I = Insert required; cask could be used if shielding liner insert is added.

N = No; cannot be used because:

- (1) shielding capability is exceeded,
- (2) heat output capability is exceeded.

These results show that liners loaded to 60,000 Ci could be transported in all of the casks listed, without the addition of shielding. Liners loaded to 120,000 Ci could be carried in the CNS 3-55, CNS 4-45, PB-1, and NLI-10/24 casks without additional shielding.

Based on the four principal constraints presented earlier, the best choices for the high-level cesium/strontium SDS liners appear to be either the CNS 1-13C or the CNS 4-45 (or PB-1). Before shipments can be allowed, amendments to certificates will be required for all the casks. The CNS 1-13C case is most readily adaptable to the TMI pool, but the CNS 4-45 cask could carry two liners.

Any of the casks listed in Table 5.9 could be used to transport the low-activity-level liners. In addition, the Hittman HN-200, CNS 4-85, CNS 8-120, and CNS 14-190 casks could be used to transport these liners. Adequate shielding is present in all cases. The CNS 3-55, CNS 4-45 (PB-1), HN-200, CNS 8-120, and CNS 14-190 could each transport two liners; the GE IF-300 and NLI-10/24 could each transport three liners.

The casks that could be used for transporting the vitrified waste canisters are shown in Table 5.11. Each of these has sufficient shielding (5 in. of lead) to allow transportation of waste canisters resulting from processing liners containing up to 120,000 Ci.

One final consideration in cask selection concerns the weight of the loaded system. The weight affects two factors: (1) plant interfacing capabilities (depending on crane capacity), and (2) in the case of truck transport, whether the shipment is legal-weight or overweight.

For the SDS liner, legal-weight shipments can only be made with either CNS 1-13C or CNS 1-13G casks. For the vitrified waste



canisters, legal-weight shipments can only be made with either NFS-4/NAC-1 or NLI-1/2 casks.

Table 5.11. Casks Capable of Transporting Vitrified Waste Canisters

Cask	USA Certificate Number	Cavity ID (in.)	Cavity Length (in.)	Equivalent Lead Thickness (in.)
Vandenberg Cask, CNS 3-55	5805/B( )F	36.0	116.75	7.0
CNS 4-45	6375/B( )F	26.0	159.0	6.5 (ends) 7.5 (side)
PB-1	6375/B( )F	26.0	159.0	6.5 (ends) 7.5 (side)
NSF-4/NAC-1	6698/B( )F	13.5	178.0	6.0
NLI-1/2	9010/B( )F	12.6	178.0	6.2
FSV-1	6346/B( )F	16.6	187.6	5.0
TN-8	9015/B( )F	9.06	168.5	6.0
GE IF-300	9001/B( )F	37.5	180.2	6.1
NLI-10/24	9023/B( )F	45.0	179.5	7.7

Rail shipment could be made using the GE IF-300 or the NLI-10/24, but it appears that this mode would be less cost effective due to higher cask lease costs (resulting from higher lease rates and longer round-trip times) and higher tariffs by rail than by truck.

### 5.3 Cask/Plant Interface

It is assumed that the casks would be loaded at TMI in the fuel pool using methods similar to those normally used for spent fuel.

Personnel dose during these loading operations would be minimal unless the pool is excessively contaminated, which could result in extensive cask decontamination. Details of this interface are presented in Chapter 4.0.

It is further assumed that cask handling facilities are available at the other sites, such that any interface problems encountered will be minimal.

#### 5.4 Shipping Scenarios

The shipment scenario assumptions shown in Table 5.12 were used in projecting personnel exposure, schedules, and costs. Where possible, legal-weight truck shipments were assumed; otherwise, overweight truck shipments were assumed. Two liners per cask were assumed to be loaded for shipment in the CNS 4-45. For the shipment of low-level strontium liners to a repository, it was assumed that any overpacking of the liners for disposal would occur at the repository. The costs associated with overpacking have not been considered.

#### 5.5 Personnel Exposure

The quantification of personnel and public exposure is beyond the scope of this study. However, estimates were made of the relative exposure levels by simply considering external dose rates and package miles for each case, along with the number of packages requiring decontamination.

For external dose rates, the approximate dose commitment relative to the worst case for each shipment leg is shown in Table 5.13. For

Table 5.12. Principal Shipment Scenarios Assumed

	Case					
	Ia	Ib	IIa	IIb	III	IV (Reference)
Initial Liner Loading (Ci)	120,000	120,000	60,000	60,000	22,000	10,000
Number of High-Activity-Level Cs/Sr Liners Shipped to Processor	5	5	10	10	25	60
Cask Used for High-Activity-Level Cs/Sr Liner Shipments	CNS 4-45	CNS 4-45	CNS 1-13C	CNS 1-13C	CNS 1-13C	CNS 1-13C
Number of Low-Activity-Level Sr Liners Shipped to:						
(a) Processor	21	0	17	0	0	0
(b) Disposal	0	21	0	17	0	0
Cask Used for Low-Activity-Level Sr Liner Shipments	CNS 1-13C	CNS 1-13C	CNS 1-13C	CNS 1-13C	N/A	N/A
Number of High-Activity-Level Vitrified Waste Canisters Shipped to Storage or Disposal	9	9	18	18	44	105
Cask Used for Vitrified Canister Shipments	NLI-1/2	NLI-1/2	NLI-1/2	NLI-1/2	NLI-1/2	NLI-1/2
Number of Low-Activity-Level Vitrified Waste Canisters Shipped to Storage or Disposal	37	0	29	0	0	0
Cask Used for Low-Activity-Level Vitrified Canister Shipments	NLI-1/2	N/A	NLI-1/2	N/A	N/A	N/A

Note: CNS 4-45 is interchangeable with PB-1.  
NLI-1/2 is interchangeable with NFS-4/NAC-1.  
N/A - Not applicable.

the shipment of low-activity-level liners and canisters, it was assumed that the dose rates external to the package would be very small, approaching zero, relative to the dose rates from packages of high-activity-level liners or canisters. For liners loaded to 120,000 Ci, shipment in an upgraded cask (having greater shielding capability) is required. This results in reduced dose commitments in all cases relative to shipments of 60,000-Ci liners. The dose commitments have been normalized to the highest case.

Exposure resulting from cask decontamination was, for simplicity, assumed to be linearly related to the number of casks decontaminated. The relative exposures determined for handling and transport and for cask decontamination cannot be added since they were not quantified. The results of the relative exposure estimates are shown in Table 5.14.

Table 5.13. Approximate Unit Relative Dose Commitment During Transportation for Each Processing Case

Initial Liner Loading (Ci)	Approximate Unit Relative Dose Commitment			
	High-Activity- Level Cs/Sr Liners	Low-Activity- Level Sr Liners	High-Activity- Level Vitrified Canister	Low-Activity- Level Vitrified Canister
10,000	0.12	0.0	0.08	0.0
22,000	0.34	0.0	0.2	0.0
60,000	1.0	0.0	0.5	0.0
120,000	0.4 <sup>a</sup>	0.0	1.0	0.0

<sup>a</sup>Accounts for two liners per cask.

Table 5.14. Relative Exposure During Transportation for Different Liner Loading Scenarios

Case	Initial Liner Loading (Ci)	Relative Personnel and Public Exposure <sup>a</sup>	Relative Personnel Decontamination Exposure <sup>b</sup>
Ia	120,000	0.72	0.42
Ib	120,000	0.72	0.20
IIa	60,000	1.17	0.45
IIb	60,000	1.17	0.27
III	22,000	1.07	0.42
Reference	10,000	1.00	1.00

<sup>a</sup>Normalized to reference case.

<sup>b</sup>Based on number of shipments, and normalized to reference case.

Note: Irrespective of the shipping casks considered, all are well within existing exposure limits of DOT regulations.

## 5.6 Schedule and Costs

The following assumptions are used to develop cask schedules and costs for the various scenarios:

1. Lease rate of legal-weight truck (LWT) casks NFS-4/NAC-1 or NLI-1/2 is approximately \$600 per day.
2. Lease rate of the CNS 1-13C or the CNS 4-45 is \$250 per day.
3. Cask certification fee for SDS liners or waste canisters is \$40,000.
4. Cask in-plant turnaround time is 24 hours at each site.
5. Average speed of legal-weight truck system is 35 mph.
6. Average speed of overweight truck (OWT) system is 15 mph (allowing for daylight travel only).
7. Tariff for legal-weight truck shipments is approximately \$2.00 per mile for short trips (250 miles) and \$1.50 per mile for long trips (2500 miles).
8. Tariff for overweight truck shipment is approximately \$4.50 per mile (includes special permits, etc.)

9. Shipment costs and schedules of the waste canisters are independent of point of origin or destination.

On the basis of the preceding assumptions, the schedule (i.e., round-trip times) and costs for each liner loading are provided for the six cases summarized in Table 5.12.

In the calculation of costs, it is assumed that each shipment leg will involve only one shipping campaign,\* and that one cask will be used to accomplish this campaign.

The shipping schedule and costs for sample campaigns are calculated as follows:

1. Reference Case, 10,000 Ci per high-activity-level cesium/strontium liner, 250-mile trip

- Use of LWT Cask (CNS 1-13C)
- Number of Shipments =  $(60 \text{ liners}) / (1 \text{ liner/trip})$   
= 60 trips
- Duration of Round Trip =  
 $(250 \text{ miles} \times 2) / (35 \text{ miles/h}) + 48 \text{ h} = 63 \text{ h}$
- Duration of Campaign =  $(63 \text{ h/trip})(60 \text{ trips})$   
= 3780 h  
= 158 d
- Cost of Campaign = \$40,000 (Cask Certification)  
+ (158 d) (\$250/d)  
+ (500 miles/trip) (60 trips) (\$2.00/mile)  
= \$140,000

2. Case Ia, 120,000 Ci per liner, high-activity-level vitrified waste canister, 2500-mile trip

- Use of LWT Cask (CNS 1-13C)
- Number of Shipments =  $(9 \text{ waste canisters}) / (1 \text{ canister/trip})$   
= 9 trips
- Duration of Round Trip =  $(2500 \text{ miles} \times 2) / (35 \text{ miles/h})$   
+ 48 h = 191 h

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\*A campaign is defined as continued round-trip use of a shipping cask from one "source" to one "sink" until all of that waste form has been moved.

- Duration of Campaign = (191 h/trip) (9 trips)  
                               = 1719 h  
                               = 72 d
- Cost of Campaign = \$40,000 (Cask Certification)  
                       + (72 d) (\$250/d)  
                       + (5000 miles/trip) (9 trips) (\$1.50/mile)  
                       = \$126,000

The schedules for the various cases are summarized in Table 5.15; the costs are summarized in Table 5.16.

In addition to the schedules and costs shown in Tables 5.15 and 5.16, one other reasonable option appears possible for the vitrified canisters resulting from processing the 10,000-Ci and possibly the 22,000-Ci liners. The TN-8, which has sufficient shielding, could be used to transport three waste canisters simultaneously. It is assumed that the TN-8 lease rate is \$1500 per day. For this case, the calculations for the reference case with a 250 mile one-way distance are:

- Number of shipments = (105 canisters)/(3 canisters/trip)  
                               = 35 trips
- Duration of 500-mile round trip = (250 x 2)/(15 mi/h) + 48  
   = 81 h
- Duration of Campaign = (81 h/trip) (35 trips)  
                               = 2835 h  
                               = 118 d
- Cost of Campaign = \$40,000  
                       + (118 d) (\$1500/d)  
                       + (500 miles/trip) (35 trips) (\$4.50/mile)  
                       = \$296,000

Similarly, for a 2500-mile one-way distance, the duration of the campaign with the TN-8 is 556 d and the cost of the campaign is \$1,662,000. Thus, unless the TN-8 lease rate is significantly below that assumed, the LWT cask is more cost-effective for the 10,000 Ci-per-liner waste canister shipments.

Table 5.15. Summary of Schedule for 250- and 2500-Mile Waste Form Shipping Campaigns

Trip Distance (miles)	Waste Form	Duration of Each Campaign (days) <sup>a</sup>					Reference Case
		Case Ia	Case Ib	Case IIa	Case IIb	Case III	
250	High-Activity-Level Liners	11	11	27	27	66	158
250	Low-Activity-Level Liners	55	55	45	45	0	0
250	High-Activity-Level Waste Canisters	24	24	47	47	116	276
250	Low-Activity-Level Waste Canisters	97	0	76	0	0	0
2500	High-Activity-Level Liners	48	48	80	80	199	478
2500	Low-Activity-Level Liners	167	167	135	135	0	0
2500	High-Activity-Level Waste Canisters	72	72	143	143	350	836
2500	Low-Activity-Level Waste Canisters	294	0	231	0	0	0

<sup>a</sup>Assumes that only one cask is used in each campaign.



Table 5.16. Summary of Costs for 250- and 2500-Mile Waste Form Shipping Campaigns

Trip Distance (miles)	Waste Form	Cost of Each Campaign and Total Cost for Each Case					Reference Case (\$000)
		Case Ia (\$000)	Case Ib (\$000)	Case IIa (\$000)	Case IIb (\$000)	Case III (\$000)	
250	High-Activity-Level Liners	50	50	57	57	82	140
250	Low-Activity-Level Liners	61	61	28 <sup>a</sup>	28 <sup>a</sup>	0	0
250	High-Activity-Level Waste Canisters	43	43	86	86	154	311
250	Low-Activity-Level Waste Canisters	135	0	75 <sup>b</sup>	0	0	0
250	Total	289	154	246	171	236	451
2500	High-Activity-Level Liners	120	120	135	135	277	610
2500	Low-Activity-Level liners	240	240	161 <sup>a</sup>	161 <sup>a</sup>	0	0
2500	High-Activity-Level Waste Canisters	126	126	261	261	258	1,330
2500	Low-Activity-Level Waste Canisters	494	0	356 <sup>b</sup>	0	0	0
2500	Total	980	486	913	557	535	1,940

<sup>a</sup>Cask certification for low-activity-level liners covered by certification for high-activity-level liners.

<sup>b</sup>Cask certification for low-activity-level waste canisters covered by certification for high-activity-level waste canisters.

### 5.7 Other Considerations

One other transportation consideration must be addressed. This concerns the gases that may be present in the liners prior to shipment. The DOT regulations [49CFR173.398(b)(1)] read as follows:

(1) Type A packaging must be so designed and constructed that, if it were subject to the environmental and test conditions prescribed in this paragraph:

- (i) There would be no release of radioactive material from the package;
- (ii) The effectiveness of the packaging would not be substantially reduced; and
- (iii) There would be no mixture of gases or vapors in the package which could, through any credible increase of pressure or an explosion, significantly reduce the effectiveness of the package.

Thus, since type B packagings must meet all type A requirements, consideration must be given prior to shipment of any of the liners to the pressure and explosive potential of any gases in the liner. Venting of such gases prior to shipment may be required.

### 5.8 Findings

The assessment of the transportation of the SDS liners and vitrified waste canisters resulting from alternative processing schemes has shown that significant reductions in personnel and public exposure, and in transportation costs, can result if the loading of the SDS liners is increased above the reference case level of 10,000 Ci per liner.

For Case III, where the liner loading is increased to 22,000 Ci the personnel exposure during cask decontamination could be decreased almost 60% relative to the reference case, and the transportation costs could be decreased by 50 to 56%, depending on the distances between facilities.

For Cases IIa and IIb, where the liner loading is increased to 60,000 Ci, the personnel and public exposure during transport would be increased over the reference case by approximately 20% because the cask would be used near its limit of shielding capability. However, it is predicted that the exposure of personnel during cask decontamination would decrease by 55 to 75%. Similarly, the transportation costs could be decreased 45 to 70%, depending on the distances between facilities and whether the low-activity-level strontium liners are disposed of directly or vitrified prior to disposal. The direct disposal of the low-activity-level liners results in the lowest costs and lowest levels of transport personnel exposure.

For Cases Ia and Ib, where the liner loading is increased to 120,000 Ci, the exposure of personnel and public is the lowest of all the cases considered, and the transportation costs are comparable to, but slightly higher than, cases where the liners are loaded to 60,000 Ci. The significant decrease in exposure arises from the use of upgraded, but overweight, truck casks for shipping high-level liners. This practice reduces external doses and allows two liners to be carried per shipment.

For Cases IIa and IIb, if OWT rather than LWT casks were used to transport the high-level liners, the personnel and public exposure would

be reduced approximately 60 to 70% relative to the reference case, and the transportation costs would be reduced 20 to 55% relative to the reference case. However, the costs would be higher than the case where LWT casks are used for the 60,000-Ci liners and resulting waste canisters. The detailed costs for the LWT/OWT cask trade-off with Cases IIa and IIb are shown in Table 5.17.

Table 5.17. Comparison of Costs of LWT and OWT Casks with 60,000-Ci Liners and Waste Canisters

Distance (miles)	Waste Form	Cost of Each Campaign and Total Cost for Each Case (\$)				Reference Case (\$000)
		Case IIa with LWT Casks (\$000)	Case IIa with OWT Casks <sup>a</sup> (\$000)	Case IIb with LWT Casks (\$000)	Case IIb with OWT Casks <sup>a</sup> (\$000)	
250	High-Activity- Level Liners	57	56	57	56	140
250	Low-Activity- Level Liners	28	68	28	68	0
250	High-Activity- Level Waste Canisters	86	135	86	135	311
250	Low-Activity- Level Waste Canisters	75	115	0	0	0
250	Total	246	374	171	259	451
2500	High-Activity- Level Liners	135	172	135	172	610
2500	Low-Activity- Level Liners	161	201	161	201	0
2500	High-Activity- Level Waste Canisters	261	517	261	571	1,330
2500	Low-Activity- Level Waste Canisters	356	396	0	0	0
2500	Total	913	1,286	557	890	1,940

<sup>a</sup>OWT cask used for high-activity-level liners and waste canisters;  
LWT cask used for low-activity-level liners and waste canisters.

## 5.9 References

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## 6.0 VITRIFICATION OF ZEOLITE

### 6.1 Technology Development

#### 6.1.1 Laboratory-Scale Development

The DOE requested Pacific Northwest Laboratory (PNL) to prepare a program plan outlining the activities required to perform a full-scale demonstration for vitrification of the TMI zeolite. During preparation of the plan, beaker-scale and small laboratory-scale vitrification tests with nonradioactive materials were performed to identify a zeolite/frit composition that would provide a suitable waste form.

The results of the vitrification tests are described in the proposed plan, "TMI Zeolite Vitrification Demonstration Program Plan."<sup>1</sup> The apparatus used to perform the laboratory-scale vitrification tests is sketched in Figure 6.1. A mixture of glass formers and zeolite was placed in the 3-in.-diam canister and placed in the furnace. The canister was then heated to a temperature of 1050°C. The zeolite used in the tests was loaded to the equivalent of ~70,000 Ci/liner, using nonradioactive cesium. Tests indicate that the glass formed was of good quality, based on comparisons of other glasses that have been developed in the National High Level Waste Management Programs. The off-gas from the beaker-scale vitrification tests was passed through successive solutions of nitric acid and caustic. After each test, the solutions were analyzed for cesium content. The connecting tubing between the canister containing the vitrified material and the containers of scrubbing solutions was removed and flushed with dilute nitric acid. This acid flush was also analyzed for cesium content. Five separate tests were conducted, with essentially the same results. Less than 0.1% of the initial cesium loaded on the zeolite volatilized during the melting process. The

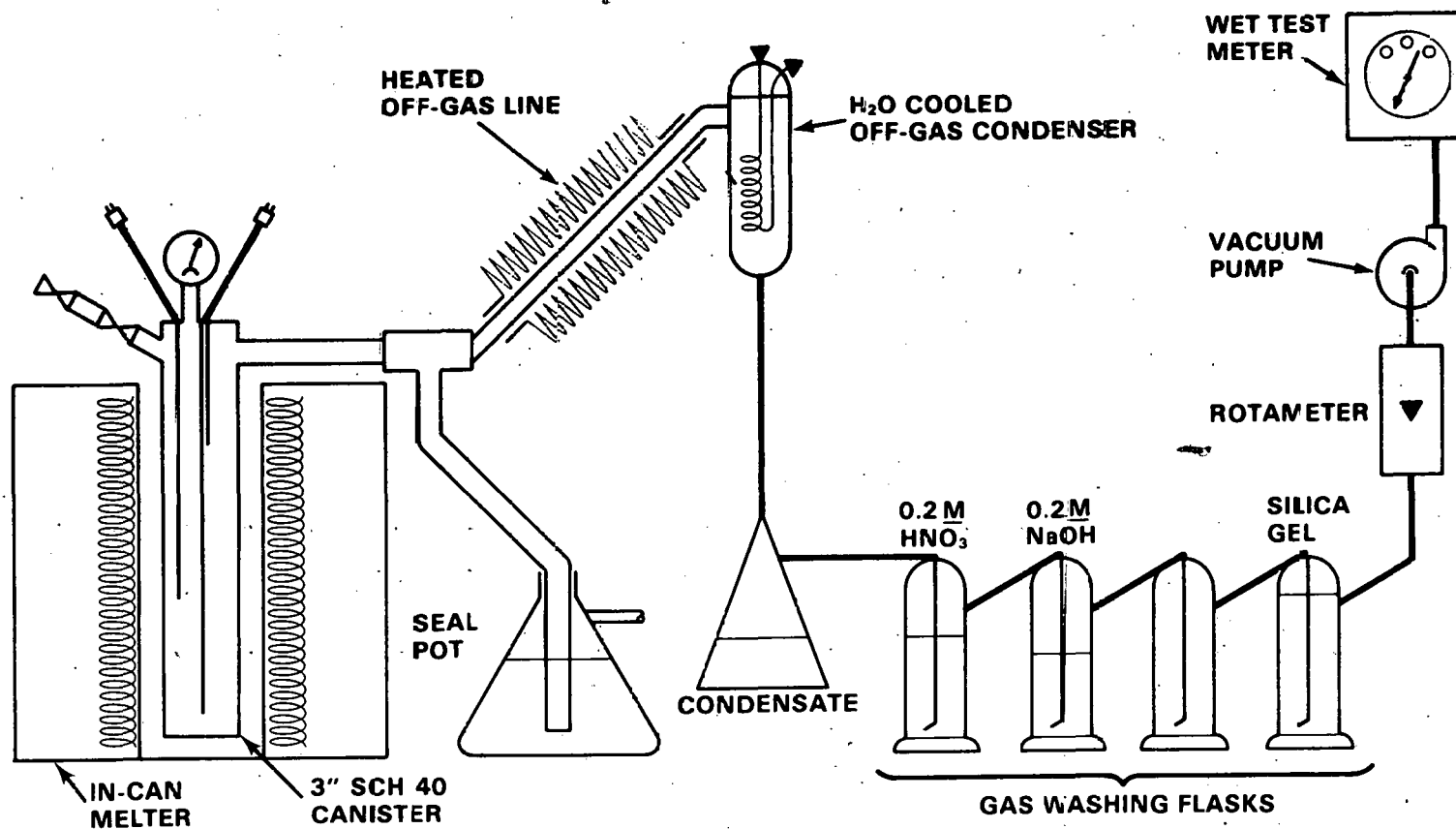


Fig. 6.1. Test Apparatus for Vitrifying TMI Zeolite.

volatilized cesium was trapped in the scrubbing solutions. No cesium was detected downstream of the scrubber; thus, based on laboratory-scale equipment, the cesium volatility is minimal and can be controlled by the use of appropriate off-gas scrubbers. It must be realized, however, that tests were not conducted at cesium levels above 70,000 Ci/liner equivalent, so that results are not available at the 120,000-Ci/liner loading level. Tests at the 120,000-Ci/liner loading levels need to be conducted to provide verification of the vitrification system, including the effluent cleanup efficiencies.

However, during the Nuclear Waste Vitrification Project, six fuel assemblies irradiated at the Point Beach Nuclear Facility were processed in B-Cell of the 324 Building.<sup>2</sup> These bundles contained approximately  $1.2 \times 10^5$  Ci of activity per assembly. Processing (including dissolution, boildown, and vitrification of the high-level waste solution) was completed without any off-limit environmental release of radionuclides. The two 8-in.-diam canisters of vitrified high-level glass from this processing contained  $1.05 \times 10^5$  and  $2.64 \times 10^5$  Ci, respectively.

If sodium titanate is used to obtain improved removal of strontium, additional laboratory tests must be completed to confirm the vitrification and effluent characterization of the zeolite-titanate mixture.

#### 6.1.2 Potential Volume Reduction with a Vitrification System

In addition to the extreme stability of a vitrified waste glass, the volume reduction factor is also very important.

During the laboratory-scale, nonradioactive tests mentioned above, a volume reduction of the glass frit/zeolite mixture was measured as it melted



into the glass product. Using the results of these tests, calculations were made relating the volume of CB sump water at TMI (about 600,000 gal) to the number of canisters of vitrified product. These results are summarized in Table 6.1. The results are based on a 75% weight loading of zeolite in the glass product, which was the highest waste loading that could be used and still produce a high-quality product. The final vitrified product was assumed to be contained in 8-in.-OD canisters filled to a level of 7 ft. An outside diameter of 8 in. was chosen so that the filled canisters could be stored in an AFR, utilizing existing densified fuel storage racks.

The results shown in Table 6.1 represent an ideal situation — that is, one in which the zeolite loading and glass-to-zeolite ratio are controlled at the maximum values. In actual operation, average values will probably be somewhat lower to provide a safety margin allowing for normal operational variations.

Table 6.1. Reduction of Waste Volume

Initial Contaminated Waste Volume, <sup>a</sup> (ft <sup>3</sup> )	Ci per Liner	Product (ft <sup>3</sup> )	Total Canisters of High-Level Cs <sup>b</sup>
90,000	10,000	232	105
90,000	60,000	39	18
90,000	120,000	20	9

<sup>a</sup>The initial waste volume is based on approximately 600,000 gals of contaminated liquid, which is equivalent to about 90,000 ft<sup>3</sup> of liquid.

<sup>b</sup>Canisters are 8 in. OD x 7-ft fill height.

## 6.2 Findings

Laboratory vitrification tests have been conducted at cesium levels (loaded on zeolite) up to about 70,000 Ci/liner equivalent. It is anticipated that the increased loading should have no measurable effect.

The heat generation rate of a canister should not present a problem if the canister is to be disposed of in a geologic repository. At a cesium loading of 120,000 Ci per liner, a waste canister would generate less than 1 kW of heat.

If sodium titanate is used, to further improve strontium removal, additional laboratory-scale vitrification studies would be required.

### 6.3 References

1. D. H. Siemens and H. H. Hollis, "Facilities for Development of Chemical and Nuclear Processes at the Pacific Northwest Laboratory," Pacific Northwest Laboratory, Richland, Washington (undated).
2. E. J. Wheelwright, W. J. Bjorklund, L. M. Browne, G. H. Bryan, L. K. Holton, E. R. Irish, and D. H. Siemens, Technical Summary Nuclear Waste Vitrification Project, PNL-3038, Pacific Northwest Laboratory, Richland, Washington (May 1979).

## 7.0 INCREMENTAL COSTS

### 7.1 Components of Cost and Scenarios Evaluated

The cost components that were considered by the Task Force consist of the following:

- (1) operation and maintenance of the SDS,
- (2) storage of SDS liners on-site,
- (3) shipment of SDS liners to a vitrification site,
- (4) conversion of zeolite to glass,
- (5) shipment of the vitrified waste to a storage site,
- (6) waste disposal.

#### 7.1.1 Operation and Maintenance of the SDS

Cost estimates for the operation and maintenance of the SDS are not yet available; however, as is noted in Table 7.1, the total number of liners needed for the job is approximately the same for Cases I, II, and III (26, 27, and 25, respectively). The reference case (60 liners) would require more materials (liners and zeolite ion exchange material), but labor savings are thought to be negligible. The assumptions are made (1) that the time required for processing all the water is about the same, regardless of the way the system functions; (2) that two lines will still be required (only one in operation at a time); and (3) that the same work force would be needed to operate the system and change liners.

#### 7.1.2 Storage of SDS Liners On-Site

In the reference case, all of the liners are stored in the fuel pool. When they are shipped, they will be unloaded from the pool into the designated cask.

Table 7.1. Number of SDS liners and glass canisters required

Case	No. of Liners			No. of Glass Canisters			Total Glass Canisters Plus Liners
	High- Activity Level	Low- Activity Level	Total	High- Activity Level	Low- Activity Level	Total	
Reference	60	0	60	105	0	105	165
Ia	5	21	26	9	37	46	72
Ib	5	21	26	9	0	9	35
IIa	10	17	27	18	29	47	74
IIb	10	17	27	18	0	18	45
III	25	0	25	44	0	44	69

It is possible that, with the differentiation between high-activity-level and low-activity-level liners, the low-level liners could be stored outside the pool. Such storage outside the pool would be done for reasons other than those of economy. Thus, it is assumed that on-site costs for handling SDS liners would be approximately the same for each case studied.

#### 7.1.3 Disposition of Loaded SDS Liners

Several casks are available for the transfer of liners from TMI to a processing location. In reviewing the various candidate casks, it appears that legal-weight truck (LWT) casks can be used for the reference case as well as Cases II and III. An overweight truck (OWT) cask would be used for Case I.

The cost for shipment was found to be the single most important variable. Table 7.2 shows the total estimated costs for each case. In estimating costs, an assumption is made that the cask will be transported either to a nearby site

(250 miles) or to a distant site (2500 miles). Significant cost differences were identified for the two sites. The cost estimates assume that a single cask is utilized for all shipments.

From Table 7.2, it can be concluded that Cases II and III are about the same and are least costly with respect to shipment of the liners. The costs for Case I are higher due to the use of an OWT cask; should an OWT cask be needed for Case II also, then the associated costs would increase significantly.

#### 7.1.4 Conversion of Zeolite to Glass

For purposes of estimating cost, it is assumed that the SDS liners will be processed and that the zeolite from the liners containing high levels of radioactivity will be incorporated in a glass matrix. The zeolite in the low-activity-level SDS liners may or may not be incorporated in a glass matrix. Cases Ia and IIa assume that all of the liners will be vitrified, whereas in Cases Ib and IIb, vitrification of only the high-activity-level liners is considered. Cost estimates have been developed for vitrification, but not for other solidification processes. For purposes of this study, the cost of solidifying the low-activity-level liners is assumed to be 25% of the cost for vitrification. This estimate is thought to be conservative since other solidification techniques, such as cementation, are quite simple and do not require sophisticated equipment such as a glass melter and furnaces.

A summary of cost estimates for each case is found in Table 7.3. From the table, it appears that Case III has a slight cost advantage over Cases I and II if all the zeolite in the liners must be incorporated in a glass

Table 7.2. Costs for Transporting Liners and Glass Canisters

Case	Number	Liners		Number	Glass Canisters		Total	Total
		Cost,	Cost,		Cost,	Cost,	Cost,	Cost,
		250	2500		250	2500	250	2500
		miles	miles		miles	miles	miles	miles
		(\$000)	(\$000)		(\$000)	(\$000)	(\$000)	(\$000)
Reference	60	140	610	103	311	1330	451	1940
Ia	26	111	360	46	178	620	289	980
Ib	26	111	360	9	43	126	154	486
IIa	27	85	296	47	161	617	246	913
IIb	27	85	296	18	86	261	171	557
III	25	82	277	44	154	258	236	857

matrix. Case Ib is the least costly option if only the high-activity-level liners are incorporated in a glass matrix. In all comparisons, the reference case is the most costly option if all of the liners must be incorporated in a glass matrix, but would be only slightly greater in cost than Cases Ib and IIb if all of the liners could be considered to be low-activity-level waste.

Table 7.3. Costs for Waste Solidification

Case	Liners Vitrified		Liners Solidified		Total Cost (\$000)
	Number	Cost (\$000)	Number	Cost (\$000)	
Reference	60	3300	0	0	3300
Ia	26	1430	0	0	1430
Ib	5	275	21	289	564
IIa	27	1485	0	0	1485
IIb	10	550	17	234	784
III	25	1375	0	0	1375

#### 7.1.5 Shipment of Vitrified Waste to a Storage Site

Once the SDS liners have been processed, and the zeolite vitrified, it is assumed that the waste canisters will be returned to the TMI site for interim storage. LWT casks are used to transport the waste canisters for all cases. Again, as in the shipment of the SDS liners, significant cost differences were identified due to distance. As in Section 7.1.3, the cost estimates assume that a single cask is utilized for all shipments.

From Tables 7.1 and 7.2, it can be concluded that Case Ib is the least costly option, followed by Cases IIb and III.

A comparison of each case has been made with the reference case; the results are presented in Table 7.4. This comparison shows that there is incentive to vitrify only those liners which are considered high-activity-level waste. If this cannot be accomplished, then Case III would be the least costly option.

A comparison was also made between the use of LWT and OWT casks for Cases IIa and IIb. This comparison shows that, if OWT casks are required for Case IIa, costs would increase by \$128,000 (or 52%) for 250 miles and \$373,000 (or 41%) for 2500 miles. For Case IIb, the cost increases would be \$88,000 (or 51%) for 250 miles and \$333,000 (or 60%) for 2500 miles. Most significant is the fact that Case III is more attractive than Case II if an OWT cask is required for Cases IIa and IIb. Case Ib would then be the least costly option.



Table 7.4. Factors for Estimating Transportation Costs When Compared with the Reference Case

Case	250 Miles	2500 Miles
Reference	1	1
Ia	0.64	0.51
Ib	0.34	0.25
IIa	0.54	0.47
IIb	0.38	0.29
III	0.52	0.44

#### 7.1.6 Waste Disposal

Once the SDS liners have been processed, there will be some charges associated with disposal of the zeolite. If it is assumed that all the waste must be stored, pending the establishment of a waste repository, then the volume of waste to be stored is the greatest for the reference case; the other cases are about equal to each other.

If only the vitrified waste is held for a repository, Case Ib would be the least costly and would generate the least volume to be stored. The reference case would generate the greatest volume and would be the most costly. For all cases, the cost for disposal of these liners could not be compared with the cost of storing spent fuel. The activity levels are orders of magnitude less, and the loaded zeolite generates little or no heat.

No estimates are given for waste disposal costs. It should be noted that these costs may differ significantly for each case, depending on the criteria for ultimate disposal.

## 7.2 Findings

A summary of costs for the processing of SDS liners is found in Table 7.5. No cost estimates are assigned for operations and maintenance or for on-site handling and storage. The estimates for these two cost centers are expected to be approximately equal, with the reference-case costs being slightly higher. Also, no cost estimates are given for waste disposal, since waste forms for a repository have not yet been defined and it is not known whether further treatment may be required.

From Table 7.5, it appears that Case Ib (vitrification of only high-activity-level loaded zeolite) will result in the lowest cost option. If all zeolite must be vitrified, then Case I appears to be the lowest. However, if 250-mile shipments are involved, Cases III and IIa are essentially identical. Selection of the appropriate option would be based on other factors.

Table 7.5. Summary of Costs for Disposal of SDS Liners (\$000)

Case	Operations and Maintenance	On-Site Storage	Transport		Solidifi- cation	Waste Disposal	Total	
			250 Miles	2500 Miles			250 Miles	2500 Miles
Reference	a	a	451	1940	3300	---	3751	5240
Ia	a	a	287	980	1430	---	1717	2410
Ib	a	a	154	486	564	---	718	1050
IIa	a	a	246	913	1485	---	1731	2398
IIb	a	a	171	557	784	---	955	1341
III	a	a	236	857	1375	---	1611	2232

<sup>a</sup>Costs not estimated, but thought to be about the same for each case. Costs for reference case may be slightly higher due to the need for additional materials.

## 8.0 CONCLUSIONS

The DOE-SDS Task Force concludes that it is technically feasible to load the zeolite liners used in the SDS to levels up to 60,000 Ci of cesium per liner without additional preoperational testing. This would result in approximately ten such liners. The Task Force further concludes that these liners can be safely handled, stored, transported, and vitrified. Moreover, the Task Force acknowledges that it may be technically feasible to load the liners to even higher levels.

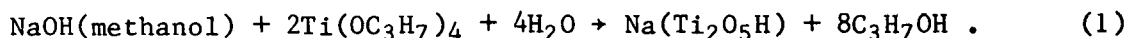
Loading the SDS zeolite liners up to 60,000 Ci of cesium would result in approximately 17 additional strontium loaded liners if Ionsiv IE-95 ( $\text{Na}^+$  form) is used. The Task Force concludes that these liners can also be safely handled, stored, transported, and disposed of, but the choice of the form for ultimate disposal of these wastes is beyond the scope of this exercise.

The Task Force addressed only the zeolite portion of the SDS and did not consider the final purification portions of the system since they have no bearing on the Task Force conclusions.

## APPENDIX A:

## PROPERTIES AND PRODUCTION OF SODIUM TITANATE

The hydrolytic reaction of NaOH and water with titanium tetraisopropyl-oxide yields sodium titanate as a finely divided white powder:



Preparation and properties of sodium titanate have been extensively studied at Sandia Laboratories by Dosch and co-workers<sup>1-6</sup> and at Hanford by Schulz and co-workers.<sup>7,8</sup> These studies demonstrated the very high affinity and capacity of sodium titanate for sorbing <sup>90</sup>Sr<sup>2+</sup> and other multivalent cationic radionuclides from weakly acid, neutral, and even highly alkaline media.

The hydraulic properties of titanate powder made by the reaction shown in Equation (1) are not suitable for large-scale column use. Sandia Laboratory and Rockwell Hanford scientists and engineers, in a cooperative effort extending over several years, conducted and sponsored research to develop consolidated forms of sodium titanate. These efforts succeeded in developing the technology for the manufacture of two acceptable and useful sorbent forms: (1) sodium titanate-loaded (approximately 40 wt % titanate) macroreticular anion exchange resin, and (2) sodium titanate pellets containing 10 to 30 wt % of either an alumina or a calcium aluminate binder and consolidated at temperatures in the range 150 to 320°C. Kilogram amounts of titanate-loaded macroreticular resin have been prepared by Cerac, Inc., Milwaukee, Wisconsin, while the Norton Company has manufactured kilogram quantities of several kinds of titanate pellets.

Mechanical and chemical properties of titanate-loaded resin and titanate pellets were investigated with synthetic waste solutions by Dosch<sup>3</sup> and with actual Hanford defense waste solutions by Schulz.<sup>7,8</sup> These tests affirmed for both titanate forms excellent sorption and capacity properties, coupled with satisfactory hydraulic performance. Kinetics of the uptake of <sup>90</sup>Sr by titanate pellet forms are significantly faster at 60°C than at 25°C.

The sodium titanate powder manufactured by Cerac, Inc., was among the variety of sorbents screened for possible use in the decontamination of the high-activity-level water at TMI-2.<sup>9</sup> Small-column tests (2 mL of sorbent per column) were made using synthetic TMI-2 water containing 3000 ppm boron, as boric acid, and 4000 ppm sodium, as sodium hydroxide. The synthetic water was traced with approximately 125 µCi/L of either <sup>89</sup>Sr or <sup>137</sup>Cs. The sodium titanate powder used in the column tests was screened to obtain a fraction ranging in size from 350 to 420 µm. Three tests were made using sodium titanate as the only sorbent, and two tests were made using a 1-mL layer of sodium titanate on top of a 1-mL layer of AW-500 zeolite (Ionsiv IE-95). The titanate alone was a poor sorbent for cesium (50% breakthrough at 40 bed volumes, while using a 4-min residence time) but was an exceptionally good sorbent for strontium (a DF of 2000 or greater was obtained with no evidence of breakthrough during a 1400-bed volume test using a 2-min residence time). During the tests made with layers of sodium titanate and AW-500, the strontium breakthrough was 5% after 2375 bed volumes when using a 1-min residence time, and 0.1% after 525 bed volumes when using a 20-min residence time. The titanate powder tested had a soft, fragile texture which did not appear to be suitable for use in large columns. Since the conclusion of the column tests, samples of pelletized sodium titanate have been obtained but have not been evaluated.

Kenna<sup>4</sup> irradiated ( $^{60}\text{Co}$ ) samples of sodium titanate powder and sodium titanate-loaded macroreticular anion exchange resin at about  $100^\circ\text{C}$  to doses as high as  $2 \times 10^9$  rads. The strontium exchange capacity of the powder remained constant through a dose of  $3 \times 10^7$  rads; 50% of the exchange capacity was retained even after a dose of  $2 \times 10^9$  rads. [The exchange capacity of unirradiated sodium titanate for strontium in alkaline solution is quite high (10 meq/g titanate) compared with that of conventional organic cation exchange resin.] Kenna believes that the primary loss of capacity of the powder was due to heating during the irradiation; previous work<sup>5</sup> with sodium titanate has demonstrated that continued exposure to elevated temperature causes a continued decrease in cation exchange capacity. Properties of the titanate-loaded anion exchange resin were unchanged after a dose of  $5 \times 10^8$  rads; after a dose of  $2 \times 10^9$  rads, the resin form retained about 30% of its original cation exchange capacity.

## References for Appendix A

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2. R. G. Dosch, The Use of Titanates in Decontamination of Defense Wastes, USDOE Report SAND78-0710, Sandia Laboratories, Albuquerque, New Mexico (1978).
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9. E. D. Collins, Oak Ridge National Laboratory, private communication.



APPENDIX B:  
SHIPPING CASK ALTERNATIVES

The following provides information on available spent fuel and waste shipping casks which could be used to transport SDS liners and vitrified canisters.

CASK: Chem-Nuclear Systems CNS 1-13G (GE-1600)

CERTIFICATE OF COMPLIANCE: 9044/B( )F

TYPE OF CASK: Legal-Weight Truck

CASK DESCRIPTION:

Steel encased lead shielded shipping cask. A double-walled steel cylinder protective jacket encloses the cask during transport. it is bolted to a steel pallet. The cask is closed by a lead-filled flanged plug fitted with a silicone rubber gasket and bolted closure. The cavity is equipped with a drain line.

CASK AVAILABILITY:

1 available from Chem-Nuclear Systems

APPROXIMATE LEASE RATE:

\$250/day

ADDITIONAL INFORMATION (Per GPU input):

- o This cask design is basically the same as the 1-13C except that it has a steel overpack; the steel and air space increase rated shielding from 5.7 inches to 6.2 inches lead equivalent.
- o G. E. and CNS each have one cask.
- o Certificate #9044 expired May 31, 1980 but NRC has permitted one year extension of use under the same certificate limits.
- o Certificate does not cover de-watered resins except for those of low specific activity.
- o This cask's certificate could possibly not be upgraded for de-watered resins without extensive review and a 30-foot drop test (as with 1-13 C).

CASK: Chem-Nuclear Systems CNS 1-13C (CNS-1600)

CERTIFICATE OF COMPLIANCE: 9081/B( )

TYPE OF CASK: Legal-Weight Truck

CASK DESCRIPTION:

A steel encased lead shielded shipping cask. The packaging is a steel double-walled, lead-filled circular cylinder. A steel, plug-type, lead-filled lid is attached with twelve 1-1/4 inch bolts, and a silicone gasket. Outer steel sheets are separated from the cask walls with small diameter wires. The lead shielding is 5 inches in the sides, 6 inches in the base and 5-3/4 inches in the lid. Two bolted-on steel lugs are for lifting only. The lid has a steel U-bar for lifting. The cavity drain line is closed with a plug. The cask is 39 inches in diameter and 68-1/2 inches long. The cavity is 26-1/2 inches in diameter and 54 inches long. The packaging weight is about 20,950 pounds. Decay heat to not exceed 600 watts.

CASK AVAILABILITY:

2 available

APPROXIMATE LEASE RATE:

\$250/day

ADDITIONAL INFORMATION (per GPU input):

- Complete revision to certification package is still under preparation; should be submitted to NRC after completing gamma scans to verify post-drop test integrity.
- Approved certificate expected within approximately three months.
- Basis of 600 watt heat limit was the data package calculation for the CNS 1-13G cask (since CNS 1-13C casks were originally G's but became C's with removal of the overpack).
- Possibility exists for increasing from 600 W to 800 W heat rate for re-certification.
- Still not determined that two loaded casks can be transported on one trailer without being overweight. CNS attempting to reduce cab/trailer weight to permit double transport at legal weight.
- Chem Nuclear has two CNS 1-13C casks but would need commitment on usage before building more casks or promising cask availability.

CASK: Chem-Nuclear Systems Vandenberg Cask (CNS 3-55)

CERTIFICATE OF COMPLIANCE: 5805/B( )F

TYPE OF CASK: Overweight Truck

CASK DESCRIPTION:

The package is steel-encased, lead-shielded cask with crushable impact limiters. The basic cask is a steel cylinder 133-3/4 inches long by 50-1/2 inches in diameter with maximum cavity dimensions of 36 inches in diameter by 116 inches long. Shielding is provided by 6 inches of lead in the sides and closure base plate and 5-1/4 inches in the closed end.

The outside steel encasement is made up of two, 1/2-inch plates on the sides and three plates totaling 2-5/8 inches on the end. The containment vessel is a 1/4-inch thick cylinder with a 1/2-inch end plate. The shells are welded together with the lead shielding poured to fill the annular and end spaces.

The removable, flanged and recessed base plate weldment consists of 0.38-inch and 1-1/4-inch outside plates and a 1/2-inch inside plate. The space between the plates is lead-filled.

The gross weight of the package, excluding the skid and sunshade, is approximately 70,000 pounds. The skid weighs about 4,200 pounds. Decay heat load not to exceed 800 watts.

CASK AVAILABILITY:

One in existence with limited availability

APPROXIMATE LEASE RATE:

\$250/day

ADDITIONAL INFORMATION (per GPU input):

- o CNS has two casks.
- o Old bell-jar design, bottom loading.
- o Certificate #5805 expired December 1980; renewal of certificate was requested.
- o Major refurbishment expected before certificate update would be granted.

CASK: Chem-Nuclear Systems CNS 4-85

CERTIFICATE OF COMPLIANCE: 6244/B( )

TYPE OF CASK: Legal-Weight Truck

CASK DESCRIPTION:

The package consists of a steel and lead shielded cask. The cask is positioned within an overpack constructed of steel and honeycomb material. The gross weight of the package is 46,000 pounds.

The mild steel cask is approximately 111-1/2 inches in length and 58 inches in diameter. The walls, top, and bottom are of 2-inch thick steel plate. Shielding is provided by 2 inches of lead within the walls and 2-inch thick steel walls of the cask. The cask lid is secured to the cask body by twenty-four 3/4-inch diameter bolts and is sealed by compressible polyurethane seal. Lifting devices are attached to the lid and body of the cask.

The cask is positioned within an overpack approximately 139-1/2 inches in overall length and 89-3/8 inches in diameter. Aluminum honeycomb material is confined by an outer steel shell 3/8-inch thick and an inner steel shell of 1/4-inch thickness. The overpack cover is of the same construction as the rest of the overpack and is secured to the walls by eight 5/8-inch diameter bolts. Lifting devices are welded to the outer shell of the overpack cover. Decay heat to not exceed 10 watts.

CASK AVAILABILITY:

1 available

APPROXIMATE LEASE RATE:

\$250/day

CASK: Chem-Nuclear Systems CNS 8-120 (LL-50-100)

CERTIFICATE OF COMPLIANCE: 6601/B( )

TYPE OF CASK: Overweight Truck

CASK DESCRIPTION:

The packaging is a steel-encased, lead shielded shipping cask which weighs approximately 70,000 pounds when loaded. The cask is 73.5 inches in diameter by 92 inches high, with an effective cavity 62 inches in diameter by 75 inches long. Gamma shielding equivalent to 4.5 inches of lead is provided by lead and steel. The outer shell is fabricated of two 3/4-inch thick steel plates and the inner shell of 1/2- and 1/4-inch thick plates. The cavity is closed and sealed by thirty-two 1-3/4 inch bolts and a silicone O-ring within a recessed groove on the flange of the cask. A steel collar encircles the outer shell in the lid area. Shackles are used for lifting the packaging and the lid. Tie-down is accomplished through a steel structure which is not attached to the package. The lid provides several threaded and sealed access plugs and the base has a drain line. Decay heat to not exceed 20 watts.

CASK AVAILABILITY:

2 in existence with fair availability.

2 being fabricated, expected completion date of March 1981.

APPROXIMATE LEASE RATE:

\$250/day

CASK: General Electric IF-300

CERTIFICATE OF COMPLIANCE: 9001/B( )F

TYPE OF CASK: Rail (Heavy-Haul Road-Short Distance)

CASK DESCRIPTION:

A stainless steel-encased, depleted uranium shielded cask. The cask is cylindrical in shape, 64 inches in diameter and a maximum of 210 inches long with maximum cavity dimensions of 37-1/2 inches in diameter by 180-1/4 inches long. Shielding is provided by 4 inches of depleted uranium, 2-1/8 inches of stainless steel and a minimum of 4-1/2 inches of water.

Two closure heads are provided for the shipment of BWR and PWR fuel assemblies. The heads are 304 stainless steel forgings and end plates which encase the 3-inch thick depleted uranium shielding.

The cask has two types of fuel baskets which can be interchanged to accommodate various fuels. The PWR basket holds 7 assemblies; the BWR basket holds 18 assemblies. The BWR fuel basket may be provided with supplementary shielding (depleted uranium) near the cask closure.

The cask is shipped horizontally with the bottom supported in a tipping cradle between two pedestals and the upper end resting in a semi-circular saddle; the upper end is pinned to the saddle. The cask supports are welded to the framing of a 37-1/2-foot long by 8-foot-wide structural steel skid. The skid also holds the cask cooling system which consists of two diesel engines driving two blowers which discharge into common ducting. Four ducts run the length of the cask and direct cooling air to the corrugated surface. Operation of the auxiliary cooling system is not a requirement of this package approval.

The entire cask and cooling system are covered by a retractable aluminum enclosure. Access to the enclosure is via locked panels in the side and a locked door in one end. Although the Model No. IF-300 cask can be transported for short distances on the highway, its principal mode of transportation is by railroad. Decay heat not to exceed 11.7 kW (air coolant), 61.5 kW (water coolant).

AVAILABILITY:

4 available, 1 owned by a utility

APPROXIMATE LEASE RATE:

\$3000/day

CASK: National Lead NLI-10/24

CERTIFICATE OF COMPLIANCE: 9023/B( )F

TYPE OF CASK: Rail

CASK DESCRIPTION:

A lead, water, depleted uranium and high temperature polymer shielded shipping cask, encased in stainless steel, equipped with balsa impact limiters, and mounted to a railcar which is considered to be an integral part of the packaging for normal conditions of transport. The cask body is 204-1/2 inches long by 96 inches outer diameter. The principal shielding consists of 6 inches of lead and 9 inches of water. Depleted uranium plates are encased in the bottom end forging and cask inner closure head. High-temperature polymer sheet is encased in the bottom end and positioned between the inner and outer closure heads at the top end.

The lead shield is bonded between a 3/4-inch stainless steel inner shell and a 2-inch stainless steel outer shell. The outer shell is surrounded by a 3/4-inch stainless steel water jacket shell. The three (3) shells are welded to stainless steel forgings at both ends. Four (4) water expansion tanks are mounted to the railcar, and are connected to the water jacket by a flexible metal hose.

The primary containment vessel is comprised of the 3/4-inch inner shell and the inner closure head. It is 179-1/2 inches long and has a 45-inch inside diameter.

The fully loaded cask, excluding the railcar, is approximately 194,000 pounds, which includes a maximum gross weight of the cavity contents of 34,100 pounds (fuel, spacers, fuel basket, etc.). Decay heat to not exceed 70 kW.

CASK AVAILABILITY:

2 available

APPROXIMATE LEASE RATE:

\$3000/day



CASK: Nuclear Fuel Services NFS-4  
Nuclear Assurance Corporation NAC-1

CERTIFICATE OF COMPLIANCE: 6698/B( )F

TYPE OF CASK: Legal-Weight Truck

CASK DESCRIPTION:

A steel, lead and water shielded shipping cask. The cask is a right circular cylinder with upper and lower steel-encased balsa impact limiters. The overall dimensions are 214 inches in length and 50 inches in diameter. The gross weight of the cask is approximately 50,000 pounds. The inner cavity is 178 inches long and 13.5 inches in diameter. The thickness of the inner shell is 5/16 inch and 1-1/4 inches for the outer shell. The two stainless steel shells are welded to a 2-inch thick stainless steel shield disc at the bottom. The annulus between the inner and outer shells is filled with lead (maximum lead thickness 6-5/8 inches, minimum 5 inches). Decay heat to not exceed 2.5 kW.

CASK AVAILABILITY:

Seven available, although only two are currently certificated for use on a de-rated basis.

APPROXIMATE LEASE RATE:

Short-term use -- \$1600/day  
Long-term use -- \$650/day

CASK: National Lead NLI-1/2

CERTIFICATE OF COMPLIANCE: 9010/B( )F

TYPE OF CASK: Legal-Weight Truck

CASK DESCRIPTION:

A depleted uranium, water, and lead shielded shipping cask, encased in stainless steel, and equipped with balsa impact limiters. The cylindrical cask body is 195-1/4 inches long by 47-1/8 inches OD. The principal shielding consists of 2-3/4 inches of depleted uranium, 2-1/8 inches of lead, and 5 inches of water.

A 7/8-inch thick stainless steel outer shell is welded to a solid stainless steel forging at each end of the cask. The outer shell of the cask is surrounded by a 1/4-inch-thick steel water jacket that is also attached to the end forgings. A water expansion tank is welded to the water jacket shell. The inner cask cavity is formed by a 1/2-inch thick, stainless steel cylindrical shell, welded at its top end to the upper cask forging and at its bottom end to a circular plate.

There are two separate configurations of the cask.

Configuration (A): The containment vessel is a right circular stainless steel shell, 12-5/8 inches ID by 178 inches inside length by 1/4 inch thick, located within the inner cask cavity. The containment vessel is closed and sealed by a 5-inch thick, composite steel and uranium closure head, twelve 1-inch diameter bolts, and a silver plated, metallic O-ring. Eight of the twelve closure bolts are used to secure the containment vessel to the upper cask forging. Closure of the cask cavity is by a 1-1/2-inch thick steel closure head, eight 1-inch diameter bolts, and an elastomer O-ring. The radioactive contents are positioned and supported within the containment vessel (inner container) by an aluminum basket and internal support structure.

Configuration (B): The containment vessel is the 1/2-inch thick inner cavity shell. The 1/4-inch thick inner container is not used. The cask cavity is closed by two closure heads. The inner head is a 6-inch thick, composite steel and uranium plate secured to the upper cask forging by twelve 1-inch diameter bolts and sealed with a silver plated, metallic O-ring. The outer head is 1-1/2-inch thick forging secured by eight 1-inch diameter bolts and sealed with an elastomer O-ring. The radioactive contents are positioned and supported within the containment vessel (inner cask cavity) by a modified aluminum basket and internal support structure.

The package, including impact limiters, has an overall length of 237 inches and an outside diameter of 75 inches. The maximum weight of the contents is 1600 pounds. The weight of the package is approximately 47,500 pounds. Maximum decay heat to not exceed 10.6 kW.

CASK AVAILABILITY:

5 available

APPROXIMATE LEASE RATE:

Short-term use -- \$1600/day

Long-term use -- \$650/day

CASK: General Atomic Company FSV-1

CERTIFICATE OF COMPLIANCE: 6346/B( )F

TYPE OF CASK: Legal-Weight Truck

CASK DESCRIPTION:

The cask is cylindrical in shape, 208 inches long, and 28 inches in diameter for most of its length except for a flange at the top end which is 11-3/8 inches thick and 31 inches in diameter. Uranium shielding, 3-1/2 inches thick in the walls and 2-1/4 inches in the lid, is encased in stainless steel. The fuel elements are of hexagonal cross section and are loaded into the fuel container, six in one column, which is located inside the cask. Total weight, including contents, is approximately 46,000 pounds. Decay heat to not exceed 4.1 kW.

CASK AVAILABILITY:

3 available, 2 fully committed

APPROXIMATE LEASE RATE:

\$1400/day on long-term (one-year) basis

CASK: Transnuclear TN-8

CERTIFICATE OF COMPLIANCE: 9015/B( )F

TYPE OF CASK: Overweight Truck

CASK DESCRIPTION:

A lead, steel and resin shielded irradiated fuel shipping cask. The cask approximates a right circular cylinder 1,718 mm in diameter and 5,516 mm long. The cavity consists of three (3) stainless steel square pressure vessels welded to an end plate and a circular stepped top flange, separated by a T-shaped copper plate and surrounded with  $B_4C$  + Cu plates. Each cavity is 230 x 230 mm and 4,280 mm long. The main shielding consists of 135 mm of lead, 26 mm of steel, and 150 mm of resin. A wet cement layer is located between the lead and the outer shell. Radial copper fins are welded to the outer shell and cover the surface of the cask between each end drum. Decay heat to not exceed 35.5 kW.

CASK AVAILABILITY:

None available	1/81
One available	1/82

APPROXIMATE LEASE RATE:

\$2200/day

CASK: Chem-Nuclear Systems CNS 14-190

CERTIFICATE OF COMPLIANCE: 5026/B( )

TYPE OF CASK: Overweight Truck Cask

CASK DESCRIPTION:

The packaging is a steel-encased, concrete shielded shipping cask. The cask is 94-1/4 inches in diameter by 103-3/4 inches in length. Reinforced concrete occupies the seven-inch annular space between the shells and the two base plates. The lid is a 4-3/4-inch-thick laminated steel cover held in place by thirty-two high strength 1-1/4-inch-diameter bolts. A silicone O-ring is used to seal the joint between the lid and the cask body. The outer shell and base plate are 1/4 inch thick, while inner shell and base plate are 2 inches thick. The cask is reinforced at the top and bottom with steel rings and is equipped with lifting lugs. The lid is provided with two access ports. Gross weight is about 71,000 pounds. The cavity dimensions are 73 inches diameter by 88-1/4 inches in length. Decay heat to not exceed 20 watts.

CASK AVAILABILITY:

Unknown

APPROXIMATE LEASE RATE:

\$250/day

CASK: Hittman Nuclear HN-200

CERTIFICATE OF COMPLIANCE: 6574/B( )

TYPE OF CASK: Legal Weight Truck

CASK DESCRIPTION:

The packaging consists of a steel-lead-steel annulus cask fabricated in the form of a right circular cylinder and three different types of inner containers. The shielded cask, closed at one end and a lid closure at the other, is 66.25 inches in diameter by 74.5 inches in height. The cask wall consists of a 3/8-inch inner steel shell, 3-3/4 inches of lead shielding, one-inch outer steel shell, and a steel flange connecting the two shells. The cask outer shell is surrounded by a one-inch layer of insulating material and canned in 14-gauge steel. The cavity dimensions are 54 inches in diameter by 62-1/8 inches in height.

The lid, sealed by a Viton O-ring, is of similar construction and is bolted to the cask body. A cylindrical shield plug is located in the center of the cask lid and is sealed by a Viton O-ring. Lifting and tie-down devices are attached to the cask body and impact skirts, consisting of removable rings of shock absorbing foam, are attached to the ends of the cask.

The packaging (empty) weight is 37,325 pounds, and the package (loaded) weight is 44,725 pounds. Decay heat to not exceed 60 watts for a single disposable inner container consisting of a right circular steel cylinder with a positive leaktight closure cap at the top.

CASK AVAILABILITY:

One in existence with limited availability

APPROXIMATE LEASE RATE:

\$250/day

## APPENDIX C:

## PRELIMINARY EVALUATIONS OF VITRIFYING TMI ZEOLITE

Preliminary evaluations were conducted at PNL to determine the feasibility of vitrifying Three Mile Island zeolite. The objectives of these scoping tests were: (1) to develop a glass formulation for the TMI zeolite which optimized the volume of zeolite incorporated into the glass, (2) to demonstrate the vitrification process in laboratory-scale equipment, and (3) to complete these studies with zeolite loaded with nonradioactive isotopes of cesium and strontium to determine whether volatility would be a significant problem.

Glass formulation studies were initiated to identify which chemicals must be added to the zeolite so that it would dissolve completely in a glass. Some of the compositions tested are shown in Tables C-1 and C-2. The results for glasses 80-195 and 80-197 (Table C-1) demonstrated that the glass must contain approximately 12 to 16% alkaline components ( $\text{Na}_2\text{O}$ ,  $\text{Li}_2\text{O}$ , and/or  $\text{K}_2\text{O}$ ) in order to achieve satisfactory dissolution of the zeolite. The results for glasses 80-206 and 80-206A (Table C-2) showed that pulverizing the zeolite aids in its dissolution. Since pulverizing would be a costly and complicated process, it was decided to develop a glass formulation with unpulverized (as-received) material. At this time, the cesium- and strontium-loaded zeolite was used to complete the glass formulation studies so that leach tests could be completed.

Table C-3 gives the results of the glass formulation studies with leach-test results. Two of the glasses (80-215 and 80-216) had 75% zeolite loadings, good appearances, and excellent leach rates. Since the cesium losses were both very low, 80-216 was selected as the glass formulation to be used because of slightly better leach rates and fewer chemical additives. One final series of tests was completed to determine whether the zeolite loading



Table C-1. Glasses<sup>a</sup> with Unpulverized Zeolite

Oxide	Composition (wt %)				
	Glass # 80-195	Glass # 80-196	Glass # 80-197	Glass # 80-198	Glass # 80-199
B <sub>2</sub> O <sub>3</sub>	5.84	--	--	--	--
CaO	2.92	--	--	10.0	2.0
Li <sub>2</sub> O	6.50	--	4.39	--	6.0
Na <sub>2</sub> O	10.40	--	7.69	5.0	2.0
TiO <sub>2</sub>	9.14	--	--	--	--
Na <sub>2</sub> B <sub>4</sub> O <sub>7</sub>	--	18.7	--	--	--
Zeolite (as received)	65.26	81.3	87.91	85.0	90.0

<sup>a</sup>Glass description:

80-195 = smooth, transparent amber glass with no undissolved zeolite detected; melted at 1050°C.

80-196 = only partially melted; most of the zeolite remained as free material.

80-197 = only partially melted at 1050°C; much of the zeolite remained undissolved.

80-198 = did not melt at 1050°C.

80-199 = only partially melted at 1050°C.

Table C-2. Glasses<sup>a</sup> with Pulverized Zeolite

Oxide	Composition (wt %)				
	Glass # 80-202	Glass # 80-203	Glass # 80-204	Glass # 80-205	Glass # 80-206
B <sub>2</sub> O <sub>3</sub>	--	--	--	3.35	--
CaO	--	--	--	1.68	--
K <sub>2</sub> O	--	--	--	4.00	5.00
Li <sub>2</sub> O	--	--	--	3.72	5.00
Na <sub>2</sub> O	--	--	--	3.72	12.00
SiO <sub>2</sub>	--	--	--	17.52	--
TiO <sub>2</sub>	--	--	--	6.00	8.00
Na <sub>2</sub> B <sub>4</sub> O <sub>7</sub>	35.0	30.0	25.0	--	--
Zeolite (pulverized)	65.0	70.0	75.0	60.0	70.0

<sup>a</sup>Glass description:

- 80-202 = melted completely at 1150°C; very viscous; no undissolved zeolite detected.
- 80-203 = melted at 1150°C; much of the zeolite remained undissolved in the glass.
- 80-204 = melted at 1150°C; much of the zeolite remained undissolved in the glass.
- 80-205 = melted completely at 1100°C; very viscous; a few large air bubbles and many small air bubbles trapped in the glass.
- 80-206 = melted completely at 1050°C; viscosity of about 150 poise at 1050°C; transparent amber glass with some white crystalline material but no undissolved zeolite.
- 80-206A = same composition as 80-206 except the zeolite was not pulverized; melted at 1050°C but much of the zeolite remained undissolved in the glass.

Table C-3. Glasses<sup>a</sup> with Cesium-Doped Zeolite

Oxide	Composition (wt %)					
	Glass # 80-213	Glass # 80-214	Glass # 80-215	Glass # 80-216	Glass # 80-217	Glass # 80-218
B <sub>2</sub> O <sub>3</sub>	5.84	5.04	4.20	5.0	--	--
CaO	2.92	2.52	2.10	--	--	--
K <sub>2</sub> O	--	--	--	--	5.0	--
Li <sub>2</sub> O	6.50	5.61	4.68	5.0	5.0	--
Na <sub>2</sub> O	10.40	8.98	7.48	8.0	10.0	10.0
TiO <sub>2</sub>	9.14	7.89	6.57	7.0	10.0	--
Na <sub>2</sub> B <sub>4</sub> O <sub>7</sub>	--	--	--	--	--	20.0
Zeolite with cesium	65.26	70.0	75.0	75.0	70.0	70.0
pH leach test - (wt % lost)	10.64	4.39	0.43	0.32	0.53	27.18
- (g/cm <sup>2</sup> -day)	4.1 x 10 <sup>-4</sup>	1.7 x 10 <sup>-4</sup>	1.7 x 10 <sup>-5</sup>	1.2 x 10 <sup>-5</sup>	2.0 x 10 <sup>-5</sup>	1.0 x 10 <sup>-3</sup>
Soxhlet - (wt % lost)	1.80	1.39	0.99	0.94	1.18	1.14
- (g/cm <sup>2</sup> -day)	6.9 x 10 <sup>-5</sup>	5.4 x 10 <sup>-5</sup>	3.8 x 10 <sup>-5</sup>	3.6 x 10 <sup>-5</sup>	4.5 x 10 <sup>-5</sup>	4.4 x 10 <sup>-5</sup>
Cesium - (wt % lost)	0.45	0.97	0.31	0.48	0.31	0.23
- (g/cm <sup>2</sup> -day)	1.8 x 10 <sup>-5</sup>	3.7 x 10 <sup>-5</sup>	1.2 x 10 <sup>-5</sup>	1.9 x 10 <sup>-5</sup>	1.2 x 10 <sup>-5</sup>	9.0 x 10 <sup>-6</sup>

<sup>a</sup>Glass description:

- 80-213 = melted at 1050°C; transparent amber glass with some white crystalline material (probably titanium); no undissolved zeolite detected.
- 80-214 = melted at 1050°C; transparent amber glass with very little crystalline material; no undissolved zeolite detected.
- 80-215 = melted at 1065°C; transparent amber glass with no crystalline material or undissolved zeolite detected.
- 80-216 = melted at 1065°C; transparent, dark amber glass with many air bubbles trapped in the glass; no undissolved zeolite in the glass.
- 80-217 = melted at 1065°C; transparent amber glass with a great deal of white crystalline material; no undissolved zeolite seen in the glass.
- 80-218 = melted at 1065°C; greenish-yellow glass; some zeolite can be seen in the glass.

could be increased. These results (Table C-4) showed that the zeolite loading limit is less than 80%. With a temperature limit of 1100°C (in-can melter process limit) and the requirement that the zeolite not be pulverized, 80-216 was selected as the optimum glass formulation.

Laboratory-scale process verification tests were initiated upon completion of the glass optimization tests. The zeolite and glass formers were vitrified in a laboratory-scale in-can melter. The process effluents were pulled by vacuum through a condenser, acid scrubber, caustic scrubber, and a filter, as shown in Figure C-1.

At the end of each test, the condensate and the two scrub solutions were analyzed for cesium to obtain volatility data. The initial plan was to melt two canisters of glass product. The first canister was a batch melt. The zeolite and glass formers were mixed and added to the canister. The furnace was heated to 1050°C, where it was held for 2 h. The second canister was a continuous melt. The melter was preheated to 1050°C, and the zeolite-glass formers mixture was fed to the canister. The test apparatus being used was not adequately designed to handle a continuous feeding process. Without a filter on the off-gas line, there was particulate carry-over to the condenser. The off-gas, which was released instantly as the zeolite was added to the hot canister, pressurized the system because the off-gas line was too small to handle the off-gas flow. There was insufficient time to modify the system for zeolite feeding; therefore, two more batch melt tests were conducted to obtain additional volatility data. The results from these four tests are shown in Table C-5. Run 2 shows greater volatility due to the particulate carry-over during this operation. For Runs 3 and 4, the off-gas line from the canister to the condenser was rinsed with acid and

Table C-4. Waste Loading Limitations of Glasses<sup>a</sup>

Oxide	Composition (wt %)		
	Glass # 80-223	Glass # 80-224	Glass # 80-225
B <sub>2</sub> O <sub>3</sub>	4.0	3.0	2.0
Li <sub>2</sub> O	4.0	3.0	2.0
Na <sub>2</sub> O	6.4	4.8	3.2
TiO <sub>2</sub>	5.6	4.2	2.8
Zeolite with cesium	80.0	85.0	90.0

<sup>a</sup>Glass description:

- 80-223 = melted at 1100°C; dark amber glass with many air bubbles and a small amount of undissolved zeolite.
- 80-224 = melted at 1100°C; very dark amber glass with much of the zeolite remaining undissolved.
- 80-225 = did not melt completely at 1100°C; most of the zeolite remained on the top of the glass, unaffected by the added chemicals.

The waste loading limit using the selected optimum glass composition, 80-216 (Table C-3), is about 80%. A 75% waste loading made a glass with good appearance and good leach rate.

"RESIN AND GLASS  
FORMERS ADDITION"

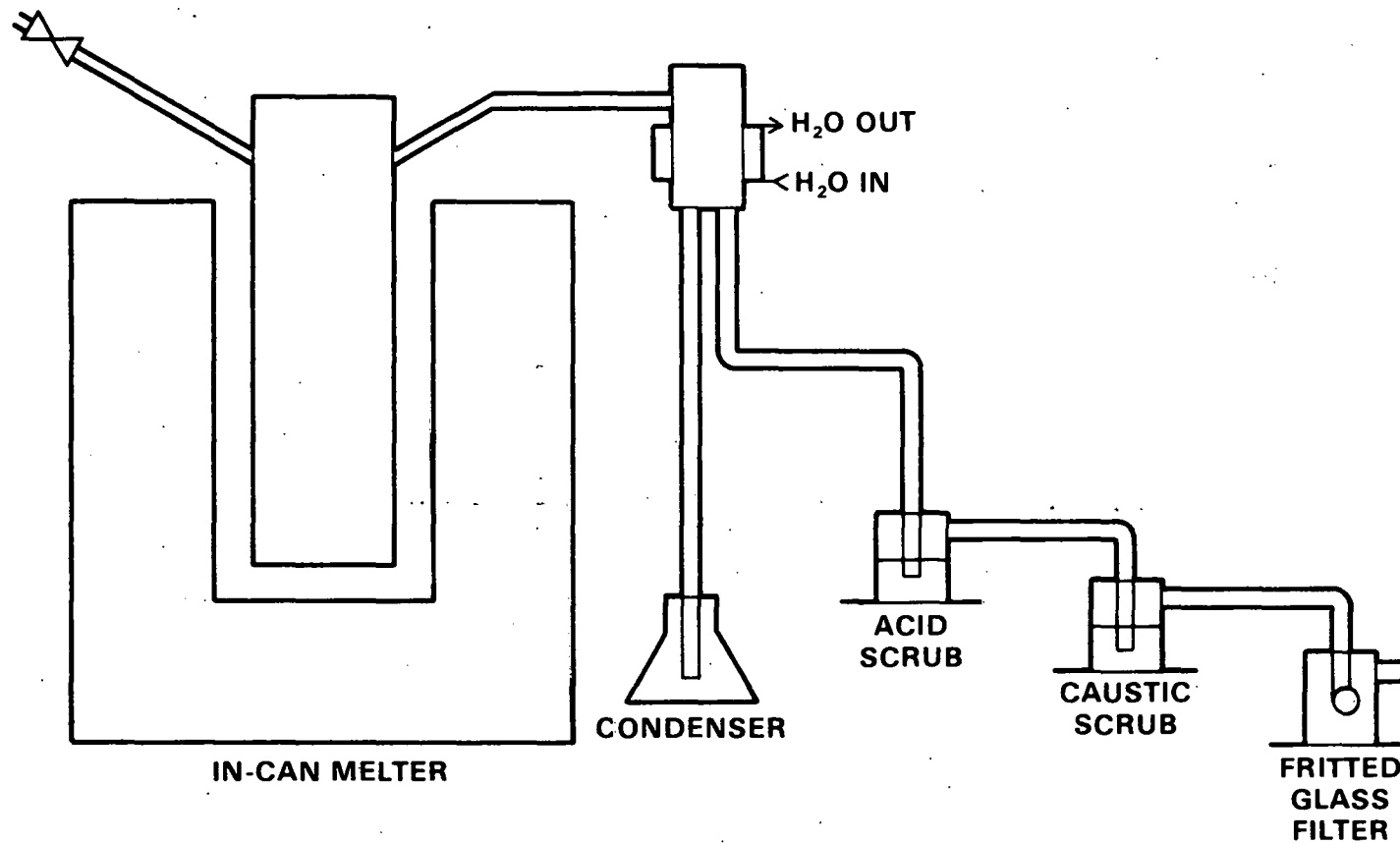


Fig. C-1. Laboratory-Scale Process Verification Process.

Table C-5. Laboratory-Scale TMI Zeolite Vitrification Results

Run No.	Zeolite Vitrified (g)	Cesium Lost (wt %)
1	1741	0.0014
2 <sup>a</sup>	3419	0.063
3	1750	0.026
4	1755	0.027

<sup>a</sup>Due to the significant volume reduction, twice as much zeolite was added during the continuous-feeding run.

water to remove any cesium which had plated out there. This accounts for the higher volatility when compared with Run 1 (Runs 1, 3, and 4 were identical).

These volatility results were so low that one last series of tests was conducted to verify this phenomenon. The cesium-loaded zeolite was mixed with the glass-forming chemicals and added to four crucibles. Each of these crucibles was then heated to 1050°C and held at that temperature for 2 h. The resulting glasses were then analyzed for cesium to determine cesium volatility. In the absence of cesium volatility, the theoretical amount of cesium in the glass would be 1.06 wt % (as  $\text{Cs}_2\text{O}^*$ ). The four glass samples had  $\text{Cs}_2\text{O}$  contents of 1.07, 1.02, 1.06, and 1.04 wt %. Again, cesium volatility was negligible.

The conclusions reached by this series of preliminary evaluations were as follows: (1) glass 80-216 with a 75% zeolite loading would be an optimum formulation; (2) vitrification with the in-can melter process would be feasible; and (3) cesium volatility would not pose any serious problems.

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\*Glass samples must be analyzed for constituents as oxides.