

CESIUM AND STRONTIUM FRACTIONATION FROM HIGH-LEVEL WASTE TO REDUCE
THERMAL STRESS IN A GEOLOGIC REPOSITORY

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ABSTRACT

Results are described for a study that assessed the benefits and costs of fractionating the cesium and strontium components in commercial high-level waste (HLW) to a separate waste stream for the purpose of reducing geologic repository thermal stresses in the region of the HLW.

System costs are developed for a broad range of conditions comparing the Cs/Sr fractionation concept with disposal of 10-year-old vitrified HLW and vitrified HLW aged to achieve (through decay) the same heat output as the fractionated high-level waste (FHLW). All comparisons are based on a 50,000 metric ton equivalent (MTE) system. The FHLW and the Cs/Sr waste are both disposed of as vitrified waste but emplaced in separate areas of a basalt repository. The FHLW is emplaced in high integrity packages at relatively high waste loading but low heat loading, while the Cs/Sr waste is emplaced in minimum integrity packages at relatively high heat loading in a separate region of the repository. System cost comparisons are based on minimum cost combinations of canister diameter, waste concentration, and canister spacing in a basalt repository. The effects on both long- and near-term safety considerations are also addressed.

The major conclusion is that the Cs/Sr fractionation concept offers the prospect of a substantial total system cost advantage for HLW disposal if reduced HLW package temperatures in a basalt repository are desired. However, there is no cost advantage if currently designated maximum design temperatures are acceptable. Aging the HLW for 50 to 100 years can accomplish similar results at equivalent or lower costs.

INTRODUCTION

A primary concern in developing deep geologic repository designs for commercial fuel cycle high-level waste (HLW) is that the heat and radiation produced by the HLW be dissipated in a manner that will not disrupt the stability of either the waste containers or the geologic formation and allow release of radioactivity to the accessible environment. If spent fuel is reprocessed to recover the uranium and plutonium, this provides an opportunity for additional processing to separate other components to enhance disposal integrity. Cesium-137 and strontium-90, both with half lives of approximately 30 years, and their short-lived decay products produce 70 to 90 percent of the high-level waste heat output and a larger percentage of the gamma activity of 10- to 100-year-old high-level waste. At the "Waste Management 81" meeting here in Tucson, Platt and Eschbach⁽¹⁾ suggested fractionating cesium and strontium from the HLW as a means of reducing the thermal stresses and gamma activity in a geological repository. More recently, Northrup et al.⁽²⁾ discussed this concept at a 1982 IAEA symposium.

The U.S. Department of Energy (DOE) contracted with the Pacific Northwest Laboratory (PNL) to evaluate the feasibility of this concept for processing commercial HLW to fractionate the cesium and strontium components to a separate waste stream for waste management purposes. The primary objective was to determine if fractionation could provide an economic means, relative to other alternatives, for controlling thermal stresses in a geologic repository. The primary incentive was the prospect of enhancing the ability to license such a repository. There is also the possibility that there are useful applications for separated cesium and strontium. This aspect was not

evaluated in this study but is the subject of other papers in this symposium.

Alternatives to cesium/strontium (Cs/Sr) fractionation that could be used to reduce thermal stress include:

1. Disperse the HLW canisters over a larger area at the expense of increased area and mining requirements.
2. Dilute the HLW to reduce its heat output and, in addition, disperse it over a larger area. This increases the volume of HLW glass and the number of canisters, as well as the area and mining requirements.
3. Use smaller canisters to limit their heat output and, in addition, disperse them over a larger area. This increases the number of canisters but not the waste volume, and also increases area mining requirements.
4. Store the HLW for a number of years to achieve thorough radioactive decay, a result similar to fractionation, at the expense and risks involved with the storage operations.

The fractionation concept presents the problem of disposing of the Cs/Sr waste. It could be stored until the heat decays to lower levels, but this has similar disadvantages to storing the entire HLW component. It might be preferable to simply store the unfractionated HLW and allow the Cs/Sr component to decay in that form. If the Cs/Sr waste is sent to geologic disposal, this adds heat back into the repository and there might be no net gain.

(a) Operated for the Department of Energy by Battelle Memorial Institute.

Considering these alternatives, the analysis was focused on the three basic alternatives illustrated in Fig. 1. Alternative I is the reference concept where 10-year-old HLW is vitrified and sent to a geologic repository. Alternative II is the Cs/Sr fractionation concept in which the HLW is separated into two components: a fractionated high-level waste (FHLW) that is vitrified and sent to geologic disposal, and the Cs/Sr component that is also assumed to be vitrified and may either be stored prior to disposal to allow decay to lower heat output levels, or be sent directly to a geologic repository. In the third alternative, 10-year-old high-level waste is also vitrified but is stored in a surface, monitored-retrievable-storage facility for a period of approximately 50 years. The storage accomplishes, through decay, the same reduction in heat output as fractionation of the Cs/Sr, after which time it is then sent to a geologic repository. The alternatives were compared on the basis of achieving specified thermal objectives in each alternative with a minimum cost combination of 1) waste concentration adjusted by dilution with inert glass, 2) canister diameter, and 3) spacing or pitch in the repository.

For the fractionation alternative we assumed that the FHLW component would be emplaced in the repository in canisters with a relatively low heat loading but high equivalent waste loading, and in high integrity waste disposal packages for very long-term survival and containment of their radionuclides. The Cs/Sr waste would be emplaced in canisters at a relatively high heat loading but with only nominal waste package integrity since, relative to the long-term containment required for the other HLW components, these nuclides decay to negligible levels in relatively short periods of time. Only one ten-thousandth of the initial Cs/Sr remains after 500 years. The relatively rapid decay of the radioactive cesium and strontium, combined with the probable dilution and long time period required for any of this material to reach the biosphere (probably thousands of years for conceivable release scenarios) should reduce the risk from short-lived Cs/Sr canisters to insignificant levels.

REPOSITORY CONCEPT

The analysis centered on disposal in a deep basalt formation repository. The reasons for this choice were the relatively low thermal conductivity of basalt that constrains the dissipation of the HLW heat, and the belief that this would enhance the benefits of the fractionation concept. A further constraint on the heat dissipation in a basalt repository is the possible requirement of an absorbent backfill material such as bentonite clay or a clay/basalt mixture as one of a series of multiple barriers designed to inhibit release of radioactivity. Bentonite or bentonite/basalt mixtures have even lower thermal conductivities than the basalt. Because of the relatively short containment period required, it was assumed that the Cs/Sr canisters would not require the bentonite backfill.

The emplacement concept utilizes vertical bore holes in a single row down the center of the emplacement rooms, with one canister per bore hole. The HLW and FHLW canisters are overpacked with a long-lived canister in a surface facility prior to repository emplacement. The Cs/Sr waste canisters are overpacked in a less expensive, shorter-lived canister. The two emplacement concepts are shown schematically in Fig. 2. To insure stability of the formation near the repository shafts, a 500-acre shaft-pillar exclusion area is required. Emplacement of the HLW and Cs/Sr waste on separate sides of this exclusion area provides a separation barrier between the two waste

types. A higher heat flux in the Cs/Sr emplacement area, but still within acceptable design limits, should not jeopardize the higher integrity objective for the FHLW emplacement area. This concept is illustrated schematically in Fig. 3.

No consideration was given in this analysis to the repository requirements or system costs for disposal of transuranic (TRU) wastes that will accompany the HLW. These were considered to be a fixed requirement in all cases. It was assumed that the TRU waste requirements would not influence the relative advantages of the alternatives evaluated in this study.

ALTERNATIVE REPOSITORY CONCEPTS

A more limited analysis was also carried out for emplacement of HLW and FHLW canisters without the bentonite/basalt backfill material.

A different concept for emplacement of HLW canisters in horizontal bore holes drilled through the pillars separating repository rooms has currently evolved as the reference concept for the basalt waste isolation project (BWIP). In this concept, multiple canisters are placed end to end in each bore hole but the separation of the bore holes is much greater than for single canisters placed in vertical bore holes. In this concept, the backfill material is not added until the waste has cooled for a number of years after it is emplaced. A limited analysis of this emplacement concept indicated that the relative cost advantages for the alternatives would be similar to the vertical bore hole concept without the backfill.

A limited thermal analysis for a repository in a salt formation indicated that the incentive for fractionation would be reduced for this medium.

HLW CHARACTERISTICS

Three factors affect the radioactivity and heat output of the high-level waste from fuel reprocessing: 1) the amount of plutonium recycle involved, which strongly affects the transuranic nuclide content; 2) the time between reactor discharge and reprocessing, which affects the amount of americium-241, which grows in from the decay of plutonium-241; and 3) the age of the waste relative to reactor discharge.

Two high-level waste compositions were considered in this analysis. One of these is the reference waste composition used in developing the Final Environmental Impact Statement -- Management of Commercial Radioactive Waste Management⁽³⁾, which was detailed in Volume 1 of Technology for Commercial Radioactive Waste Management⁽⁴⁾. This represents a high-level waste composition that would evolve after a period of about 20 years of plutonium recycle. The spent fuel was processed one and one-half years after reactor discharge. The effect of cesium and strontium removal on heat output of the high-level waste from this plutonium/uranium recycle waste composition is shown in Table I for waste ages (time from reactor discharge) ranging from 10 to 100 years. The heat output of FHLW at both a 95 percent Cs/Sr removal and at a 99 percent Cs/Sr removal is shown. In this analysis a 95 percent Cs/Sr removal was assumed, although the results of the process analysis in this study now indicate that a 99 percent removal is feasible. For this waste, Table I shows that a waste age on the order of 60 years accomplishes approximately the same heat reduction as the Cs/Sr fractionation.

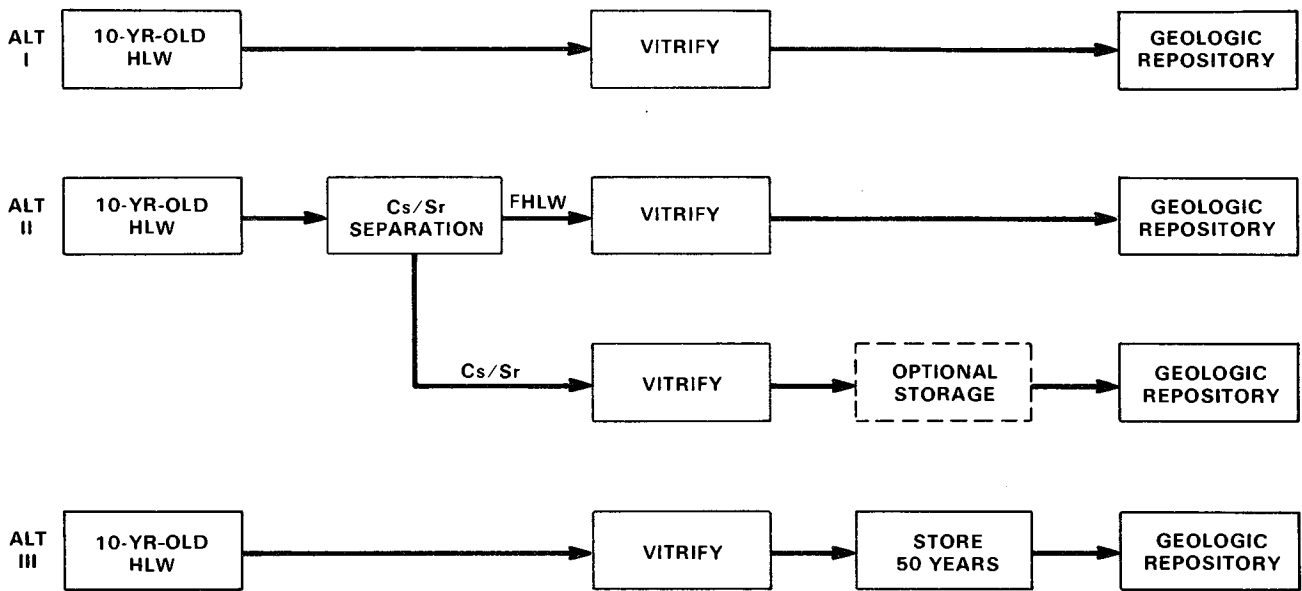


Fig. 1. Three Alternatives Evaluated for the Cs/Sr Fractionation Concept

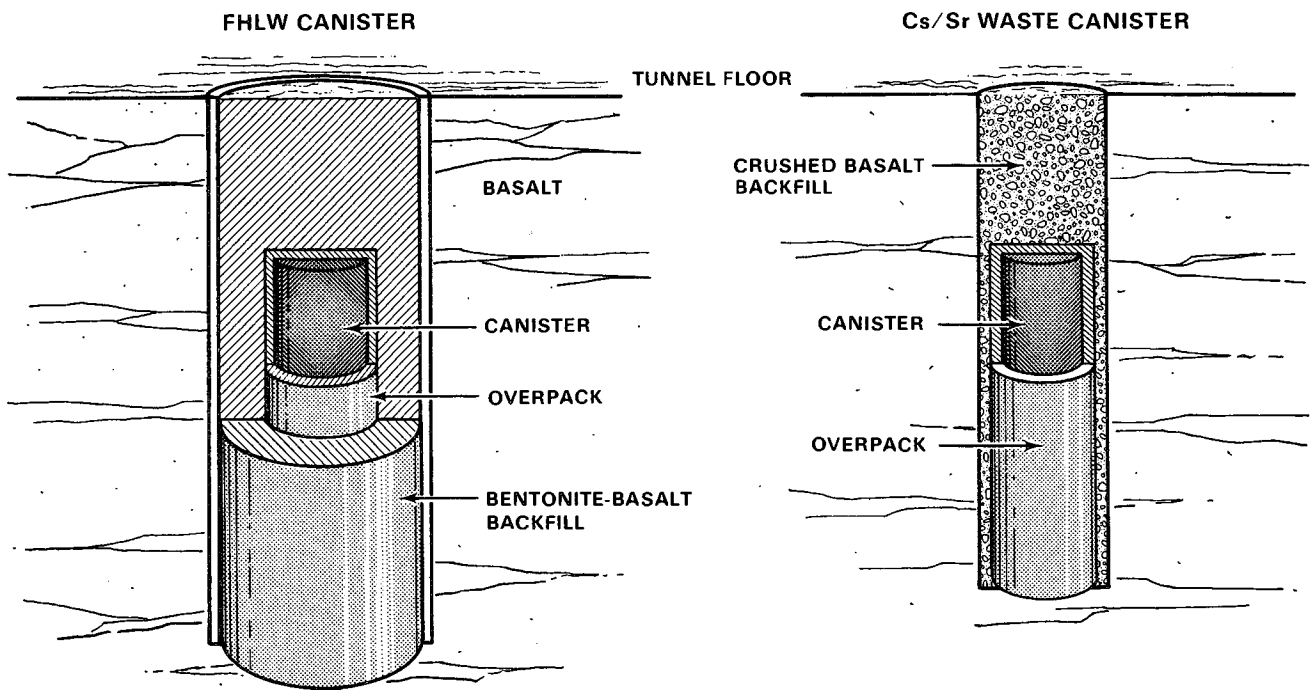


Fig. 2. Waste Emplacement Concepts

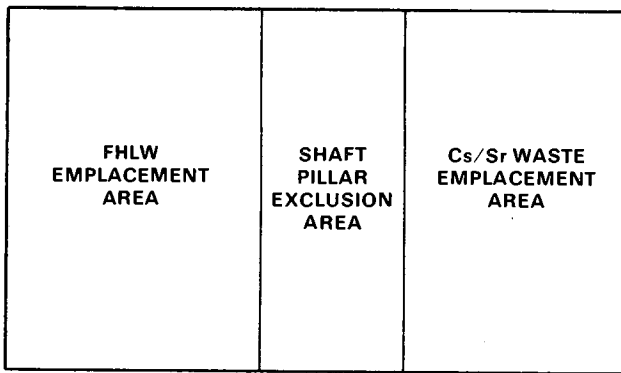


Fig 3. Schematic Basalt Repository Concept

TABLE I. Effect of Cesium and Strontium Removal on Heat Output of HLW in a Pu/U Recycle Fuel Cycle (Watts/MTE)^(a)

	HLW Age (Years)			
	10	50	60	100
<u>HLW</u>				
Fission Products	870	290	220	88
Actinides	210	66	53	32
Total	1080	356	273	120
<u>FHLW (95% Cs/Sr Removed)</u>				
Fission Products	93	22	19	6
Actinides	210	66	53	32
Total	303	88	72	38
<u>FHLW (99% Cs/Sr Removed)</u>				
Total	270	77	64	34

(a) MTE = metric ton equivalent, i.e., HLW from a metric ton of uranium plus plutonium in the original fresh fuel.

The second waste type considered represents the type of high-level waste that can be anticipated during the early years of reprocessing. There will be a large backlog of spent fuel representing once-through fuel cycle operation, and the spent fuel will have been stored for a number of years prior to reprocessing. For this case, the spent fuel was stored for 10 years prior to reprocessing. The heat output of this HLW is shown in Table II. This table also shows the heat output of the FHLW at 95 and 99 percent Cs/Sr removal. At 10 years of age the heat output of this FHLW is only about 60 percent of the heat output of the HLW in the recycle case. However, because of the amount of americium-241 that grows in before reprocessing, at 50 years of age the heat output is almost equal to the recycle waste. It has a higher heat output at any age greater than 60 years.

Most of the gamma activity of the HLW, particularly for older wastes, is associated with the cesium and strontium components. The composite gamma energy emission rates for HLW and FHLW are compared in Table III over waste ages ranging from 10 to 100 years. The gamma energy emission rate is a rough measure of the radiation dose delivered to the repository components in the vicinity of the waste canisters.

STUDY SCOPE

Four major tasks were undertaken in this study. An extensive heat transfer analysis effort established the basis for defining allowable waste emplacement configurations in the repository. A fractionation process study analyzed the process requirements

Table II. Effect of Cesium and Strontium Removal on the Heat Output of HLW in a Once-Through Fuel Cycle (Watts/MTE)

	HLW Age (Years)			
	10	50	60	100
<u>HLW</u>				
Fission Products	927	318	250	96
Actinides	88	61	58	51
Total	1015	379	308	147
<u>FHLW (95% Cs/Sr Removed)</u>				
Fission Products	92	23	17	6
Actinides	88	61	58	51
Total	180	84	75	57
<u>FHLW (99% Cs/Sr Removed)</u>				
Total	145	71	66	54

TABLE III. Effect of Cesium and Strontium Removal on the Gamma Energy Emission Rate (10^{15} Mev/sec/MTE)

	HLW Age (Years)		
	10	50	100
Unfractionated HLW	2.45	0.767	0.234
FHLW (95% Sr/Cs Removed)	0.700	0.075	0.017
% Remaining after 95% Cs/Sr Removed	28.6	9.8	7.3

and established the feasibility of the proposed fractionation process through bench-scale laboratory experiments. The minimum (optimum) system cost basis for comparison of the alternatives was developed in a waste management system cost analysis. Both near-term and long-term relative risks for the three waste types (HLW, FHLW, and Cs/Sr waste) during processing, transportation, storage, and final disposal in a geologic repository were evaluated in a radiation risk analysis.

The emphasis in the thermal analysis was on temperature limits at or near the waste canisters. Previous work had indicated that in a basalt repository these temperatures would be the limiting concern rather than the overall repository loading. In other words, the very near-field thermal loadings tend to limit the far-field total repository loadings.

A computer code called RECON⁽⁵⁾ that models the life cycle construction and operating costs of a geologic repository was used to develop costs for the broad range of repository parameter variations required. To account for varying system requirements, cost scaling relationships were developed to define the costs of fractionation, vitrification, transportation, and storage.

For the cost analysis, alternatives were compared on a constant dollar basis without any cost discounting considerations. Discounting was found to have only a small effect on the relative advantages of the near-term disposal alternatives and comparisons are simplified without it. For the storage alternatives,

the uncertainty in selecting an appropriate discount rate for the long time period was the primary consideration. It is clear, however, that in an alternative that utilizes long-term storage to reduce heat output rates even a small discount rate would substantially reduce the present-worth cost of the repository component of the system cost.

FRACTIONATION PROCESS

A number of processes have been considered for fractionating cesium and strontium from HLW, and one has been used on a plant scale at Hanford (with defense HLW). This process was not chosen for the current study because it would substantially increase the quantity of waste glass required to contain the FHLW. One important process criterion for this study was to minimize the volume of FHLW to gain a maximum benefit from Cs/Sr removal and permit a wide range of waste concentrations to be examined in the overall evaluation. Another criterion was that the transuranic element content of the fractionated Cs and Sr should be kept as low as practicable so that less stringent disposal conditions could be postulated for the Cs/Sr waste stream. Both of these criteria are met by the process used here.

A simplified schematic flowsheet is shown in Fig. 4. The key step of the process is the sorption of cesium and strontium from moderately acidic liquid HLW onto inorganic ion exchange materials (titanium phosphate for the cesium and hydrated antimony pentoxide for the strontium). The remaining HLW constituent, i.e., the FHLW, is incorporated in a glass waste form for disposal. The loaded sorbers are also mixed with suitable glass-forming materials and heated to melt the mixture and produce a glass waste form. Other features of the process are a solution clarification step and a Pu-recovery solvent extraction step, both of which are aimed at minimizing the transuranic element content in the Cs/Sr waste.

SYSTEM COST ANALYSIS

The system costs were developed for a 50,000 metric ton equivalent (MTE) system over a range of maximum waste package temperatures, and for HLW and FHLW emplaced both with and without a bentonite/basalt mixture backfill (bentonite helps seal out water from the waste canister). Other variables included a range of maximum waste concentrations, considering minimum and maximum inert diluents in the HLW and FHLW as determined by the reprocessing flowsheet, and repository size limits of either 50,000 MTE or 2,000 acres. For 2,000-acre repository limits, some cases required several repositories and others required only a fraction of a single repository. The system costs include the fractionation process, vitrification, storage, transport, and disposal. For the bentonite backfill cases, the limiting parameter was the maximum bentonite temperature, while for the cases without bentonite, the limiting parameter was the maximum waste centerline temperature.

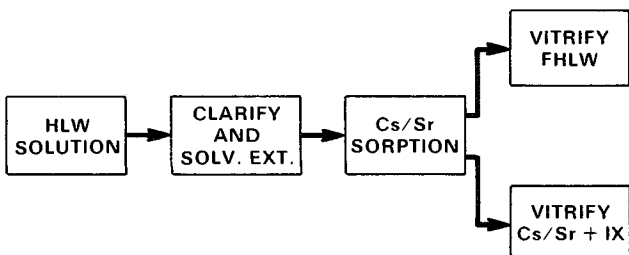


Fig 4. Fractionation Process

Minimum costs for each alternative were developed from considerations of the three primary variables, i.e., canister size, waste concentration, and canister spacing or pitch in the repository. Canister size was varied by changing the canister diameter. For each of four canister diameters (0.5, 1.0, 1.7 and 2.5 feet), three waste-concentration canister-pitch combinations were selected from the thermal analysis results that yielded a specified maximum bentonite or waste centerline temperature. Vitrification, transportation, and waste storage costs (in the case of the storage alternatives) were then determined for each of these three conditions. The minimum system cost was then determined from a plot of total costs versus canister pitch for each of the four canister sizes.

Figure 5 provides an example optimization for a 1-ft diameter HLW waste canister and a 200°C maximum bentonite temperature. To aid in understanding the relationships involved here, two additional abscissa scales are provided. These are the kilowatt per canister loading and the waste volume as cubic feet/MTE. For example, to maintain the 200°C maximum bentonite temperature, the canister heat loading increases as the canister pitch increases. This is accomplished by increasing the waste concentration in the glass, which is reflected in the declining waste volume shown on the bottom scale. Since the canister size is fixed, the total waste volume variations are accommodated by reducing the number of canisters required to dispose of 50,000 MTE. As the canister pitch is reduced, the concentration must be reduced to decrease the heat loading, and increasing numbers of canisters are required. The result is that repository costs pass through a minimum as the canister pitch increases. The costs at first decline because fewer canisters are required, but then they tend to increase because the larger mined volume requirement raises costs more rapidly than the reduction in the number of required canisters reduces the costs. Vitrification and transportation costs both decline as the canister pitch is increased because fewer canisters are required. Adding up the repository, vitrification, and transportation costs yields the total system costs. The minimum point on the system cost is shifted to the right, relative to the repository minimum costs, because of the influence of the vitrification and transportation costs. The kilowatt per canister loading at both the repository and system cost minimums is indicated on the figure.

The minimum system costs at four canister diameters are plotted in Fig. 6 for the same 200°C maximum bentonite temperature example. This shows repository costs to be relatively insensitive to canister size, but total system costs decline sharply with reduced canister sizes. This trend was found in all of the cases evaluated in this study. To aid in understanding this trend the same two additional scales are shown on the abscissa as in the previous figure. This shows that although the canister waste loading increases with increasing canister size, it does not increase nearly in proportion to the increase in waste volume. The increasing waste volume increases both vitrification and transportation costs and results in the higher costs for larger canisters.

The results consistently showed that, from a cost standpoint, the smallest size canisters consistent with the temperature constraints and maximum waste concentrations are desirable. The optimum canister size was determined as the larger of either a 0.5-ft diameter canister or the canister size determined by the temperature objective and the process-limited maximum achievable waste concentration. If canister

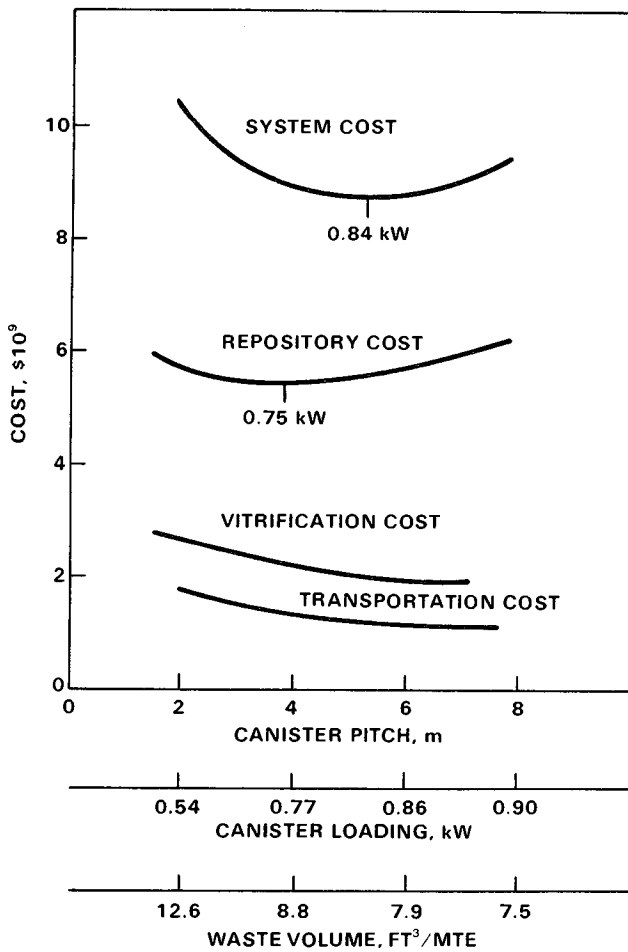


Fig. 5. Example of Disposal System Optimization with a Fixed Canister Size--HLW in 1-ft Diameter Canisters, 200°C Maximum Bentonite Temperature, 50,000 MTE System

sizes smaller than 0.5-ft in diameter are feasible, some of the cost relationships developed in this study would be affected.

Results for the bentonite backfill cases are summarized in Fig. 7. Each point on these curves represents a minimum cost combination of canister size, canister pitch (spacing) in the repository, and waste concentration. The FHLW disposal cases include the requirements for disposal of the Cs/Sr waste component in a separate section of the same repository. The approximate maximum waste centerline temperature is shown in addition to the maximum bentonite temperature. At the higher bentonite temperatures, the cost ranges for the FHLW and HLW disposal system alternatives overlap, indicating no advantage for the fractionation concept. However, as the bentonite temperature or waste centerline temperature is reduced, the fractionation concept begins to show a significant cost advantage. At the 100°C bentonite temperature the FHLW system costs are approximately one-half the HLW system costs. The fractionation system costs increase only modestly as the bentonite temperature is reduced from 250°C to about 150°C, indicating that substantial temperature reductions could be realized with the fractionation concept at a relatively small increase in total system costs.

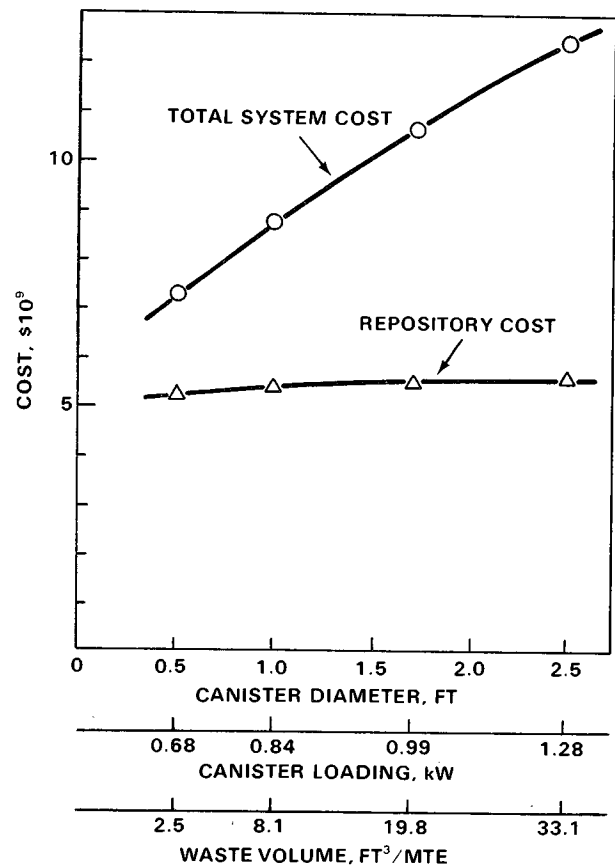


Fig. 6. Example Showing Declining System Costs as Canister Size is Reduced--200°C Maximum Bentonite Temperature, 50,000 MTE System

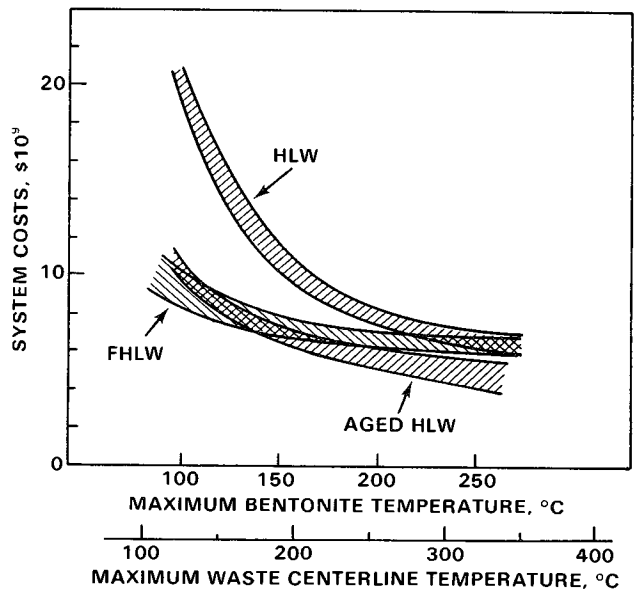


Fig 7. Total 50,000 MTE System Cost Comparison for HLW, Aged HLW and Fractionated HLW Disposal with Bentonite Backfill

The costs for the aged high-level waste disposal alternative show a substantial advantage relative to the HLW disposal over the entire temperature range. This indicates that, from a cost standpoint, aged HLW

may be the most attractive alternative. However, at the low end of the maximum temperature range, the cost for aged HLW disposal is equal to or greater than the costs for the FHLW disposal. If a cost-of-money discount rate is applied, even if only a very small one, the repository costs for the aged HLW case would be reduced even more. The major disadvantage of the aged HLW alternative is the delayed disposal and the long period of surface storage surveillance required.

Additional details of canister size and waste loadings, as well as a cost breakdown for the main system components, are compared for an example data set in Table IV at two backfill temperatures. These data illustrate the tradeoffs involved in the analysis. The range of costs shown in Fig. 7 relative to the example in Table IV results from consideration of the effects of plutonium recycle, the effects of a range of maximum achievable waste concentrations and the effects of possible repository size limits.

Another way of looking at the fractionation comparison is to ask what it would cost and what would be achieved if the FHLW were emplaced in the same size canisters at the same pitch as the HLW configuration for a 250°C bentonite temperature. The Table IV comparison approximates this case (column 4 compared to column 2). At a cost of approximately \$2 billion, or \$40,000/MTE, the maximum bentonite temperature is reduced from 250°C to 100°C.

Results of the cost comparisons when the bentonite/basalt backfill is not used (not presented here) are roughly similar to the results with the backfill when compared on the basis of maximum waste centerline temperatures, except that the differences between HLW and FHLW costs are somewhat smaller.

The major conclusions relative to the potential advantages of the Cs/Sr fractionation concept can be summarized as follows:

The fractionation concept offers the prospect of achieving reduced temperatures in and near the high-level waste canisters at a substantially lower cost than that achieved by simply dispersing

the unfractionated high-level waste in more canisters over a larger repository area to attain equivalent temperatures.

- Aging high-level waste for 50 years or more can accomplish a similar result at equivalent or lower costs. However, to achieve these lower costs, the storage must be planned for and committed at the time the HLW canisters are produced because a higher waste loading or larger waste canisters must be utilized than would be possible without aging. The disadvantages of this concept are the delayed disposal and the long period of surveillance required.
- Storing the Cs/Sr component of the waste for 50 years prior to disposal in the fractionation concept was found to cost about the same as immediate disposal in a separate section of the repository. The reduced disposal cost is offset by the cost of storage. Furthermore, this option has the same disadvantages as HLW aging, thus is not considered an attractive alternative.
- If repository waste emplacements are designed for currently designated maximum acceptable temperatures, there appears to be no cost advantage for the fractionation concept using the process and disposal assumptions used in this analysis.
- Results of both the long- and short-term radiation risk analyses have indicated that relative risk does not provide a basis for discriminating between these disposal alternatives.

Several conclusions were reached that relate to general repository design considerations. These include:

- Optimization of canister size with respect to the entire waste management system is an important factor that should be considered in repository design concept. It can mitigate the impact of such factors as an expensive canister or overpack material, or a more dilute waste concentration.

TABLE IV. Example Comparison of Tradeoffs Involved in Cs/Sr Fractionation, Conventional HLW Disposal, and HLW Disposal after Aging^(a)

	100°C Maximum Backfill Temperature			250°C Maximum Backfill Temperature		
	HLW	FHLW	Aged HLW	HLW	FHLW	Aged HLW
Canister Diameter (ft)	0.50	0.50	0.50	0.50	1.15	0.92
Canister Loading (kW)	0.16	0.16	0.22	0.98	1.22	1.13
Canisters per 50,000 MTE (10 ³)	338	56	68	55	7.4	13
Cs/Sr Canisters per 50,000 MTE (10 ³) ^(b)	---	17	---	---	17	---
Areal Heat Load (kW per acre)	12	12	6.4	38	33	39
Costs, 50,000 MTE System (\$10 ⁹):						
Fractionation	---	0.9	---	---	0.9	---
Vitrification	4.7	1.4	1.4	1.2	0.9	0.7
Transportation	2.0	0.6	0.4	0.3	0.4	0.2
Storage	---	---	2.0	---	---	0.8
Repository	12.0	5.3	5.8	4.6	3.8	3.2
Total Costs	18.7	8.2	9.6	6.1	6.0	4.9

(a) Example based on once-through cycle spent fuel and a high-range waste concentration.

(b) The Cs/Sr canisters are 0.95-ft diameter with 2.25 kW per canister at 56 kW/acre as limited by a 300°C maximum basalt temperature.

- To achieve *minimum waste disposal system costs*, the maximum possible waste concentration should always be utilized; any required adjustments for thermal limits in the repository should be achieved through adjustment of the canister size and spacing in the repository.

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