TREATMENT STRATEGIES FOR TRANSURANIC WASTES

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ABSTRACT

This paper presents an analysis of treatment options or strategies for transuranic wastes expected to be generated at a commercial nuclear fuel reprocessing plant. Six potential options were analyzed, ranging from no treatment to maximum volume reduction and high quality waste forms. Economics for the total management of these wastes (treatment, transportation, disposal) indicate life-cycle savings for extensive treatment are as high as $1.7 billion for 70,000 MTU. Evaluations of the waste processing and waste forms support the selection of a number of the extensive waste treatments. It is concluded that there are significant incentives for extensive treatment of transuranic wastes.

INTRODUCTION

This paper presents an evaluation of potential strategies for treatment of transuranic wastes (TRUW) resulting from the reprocessing of commercial spent fuel. The study was performed for the Department of Energy (DOE) as part of the Nuclear Waste Treatment Program at the Pacific Northwest Laboratory.[1] A key objective of the program is to provide needed technology to assure that the availability of waste treatment technology is not an impediment to the implementation of reprocessing and waste management in this country. Although reprocessing is not planned in the foreseeable future in the U.S.A., many of the conclusions may be applicable to treatment before disposal of commercial transuranic wastes that will be generated without reprocessing. Indeed, a similar evaluation is now nearly completed for the DOE on options for treatment of transuranic and high-activity wastes resulting from centralized commercial spent fuel consolidation and packaging, which may be done at a geologic repository or an MRS facility. Early results indicate the conclusions from the second study are similar to those for the study reported on in this paper.

The objective of the study was to evaluate potential alternative treatment strategies of TRUW in the total waste management system. The results of the study have been used to provide DOE with bases for decisions regarding development of transuranic waste treatment technology.

Study Approach

The approach used in this study is illustrated in Fig. 1. The first step was to identify the overall study bases. Data from the Barnwell Nuclear Fuel Plant (BNFP) were used as the primary source for as-generated waste type and waste volume information.[2] The possible treatment methods for each type of waste were then considered. Six objectives were established for selecting treatment processes, and existing regulatory requirements were reviewed.

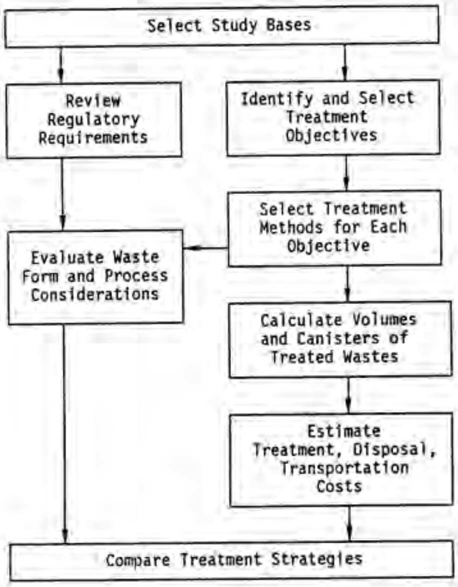

Fig. 1. Approach for Strategy Evaluation

These objectives were then used to select treatment methods for each objective. This selection resulted from numerous meetings of the authors in which the effects of various processing combinations and waste forms were considered. The volumes of treated and packaged wastes for disposal in a repository or a LLW facility were determined for each option. Using these volumes, the costs of transportation and disposal were determined and treatment costs were estimated. The processing, waste form and cost evaluations were then compared.

The study evaluated the waste treatment characteristics, waste form characteristics, and life-cycle costs for treatment (capital and operating), transportation to a repository, and disposal in a repository. It can be noted here that many of the more troublesome wastes generated at a central spent fuel consolidation and packaging facility are similar to the more troublesome TRUW expected at a spent fuel reprocessing plant.

Primary Study Bases

The primary bases used in the study were:

- The Barnwell Nuclear Fuel Plant (BNFP) is the reference reprocessing facility.
- Treatment of all TRUW generated on-site is to be done at the BNFP in incremental treatment facilities.
- The characteristics of the as-generated TRUW expected from the BNFP are the reference waste source characteristics.[2]
- TRUW are disposed of in a deep geologic repository in basalt.
- Waste form requirements for disposal are currently unknown; thus, treatments studied include a broad range of waste form characteristics.
- Life-cycle costs are estimated for processing wastes from 70,000 MTU of spent fuel in two identical reprocessing plants. Costs in this paper are in 1985 dollars.

From these primary bases, other more specific bases were developed for the specific parts of the study.

Initial Waste Quantities

The untreated TRUW volumes, taken from Darr (1981), are given in Table I. These wastes were grouped into 5 categories, based mainly on characteristics for various treatments. The categories are: fuel cladding hulls and hardware failed equipment, HEPA filters, fluorinator solids and general process trash. The TRUW are also divided into remote-handled TRUW (RH-TRUW, with dose rates exceeding 200 mrem/hr) and contact-handled TRUW (CH-TRUW, with dose rates less than 200 mrem/hr), because of the difference in facility and handling needs for the two ranges from radiation levels. In Table I, note the predominance of the RH-TRUW volumes. Also note the predominance in either waste category of fuel hulls and hardware and HEPA filters. (Fuel hardware and HEPA filters are also the major secondary wastes from spent fuel consolidation and packaging.) HEPA filters are difficult to treat chemically because they can consist of stainless steel or wood frames, combustible or noncombustible filter media, up to 1/3 by weight of organic glue, and aluminum spacers. While the general process trash consists of a wide variety of materials (i.e., paper, rags, wood, glass, metals, plastics and ceramics), the quantities are small relative to the HEPA filters and metallic wastes.

The total volume of these untreated wastes can be compared to that for spent fuel assemblies which is about 450m^3/1000 MTU, and from wastes from spent fuel consolidation which is about 470 m^3/1000 MTU as generated.

TABLE I

Volumes of Untreated TRUW

TRUW Type	m^3/1000 MTU Remote-Handled	 Contact-Handled
Hulls and hardware	455	0
Failed equipment	6	16
HEPA filters	94	230
Fluorinator solids	13	0
General process trash	85	74
Totals	653	320

Treatment Objectives and Methods

A variety of treatment objectives was considered for the study. The six objectives selected are summarized in Table II. The overall treatment objectives range from no treatment (Option 1) to relatively extreme treatment to keep volumes low (Options 4, 5 and 6).

TABLE II

Treatment Objectives

Option	Objectives
1	Eliminate treatment, Minimize treatment costs
2	Reduce volume with simple technology
3	Common treatment process to one waste form
4	Minimize final waste volume without decontamination
5	Maximize decontamination to LLW, Minimize final waste volume
6	Minimize final waste volume, Eliminate combustibles.

With waste characteristics and treatment objectives defined, the potential treatment processes were identified. These are shown in Table III, using the treatment categories of no treatment, pre-treatment, intermediate treatments, and immobilization. All of the processes identified are established technology in nonradioactive fields, and some have been used in radioactivity processing applications. None of the processes was considered to require an exorbitant amount of R and D to develop for TRUW treatment applications.

TABLE III

Waste Treatment Methods Considered

No Treatment	Pretreatment	Intermediate Treatment	Immobilization
No Treatment	Segregation	Decontamination	Melting
Assay	Size Reduction	Incineration	Encapsulation
		Slagging Pyrolysis	Compaction
		Precipitation	Cementing
		Ion Exchange	Vitrification
		Calcination	Hot Pressing
		Combine with HLW	
		Dilute to LLW	

Under the category of no treatment, assay is considered to be necessary for any treatment option, even no treatment. Pretreatments may be required for some of the treatment processes. Intermediate treatments can frequently reduce volume but may not improve on the characteristics of the final waste forms, although some of these may constitute the final treatment (e.g., slagging pyrolysis). Immobilization processing will improve the final waste form in all cases.

With these potential processes identified, and after review of their applicability and limitations, the combinations of treatment processes were selected to process each waste type in each strategy option. The processes selected are given in Table IV.

The best treatment method to reduce treatment costs (but not necessarily total costs) is to not treat the wastes. Option 1 is the no treatment strategy where all wastes are packaged as generated.

The treatment method that was viewed as the simplest, but yet gave major volume reduction, was compaction. Option 2 is the minimum treatment strategy. All of the wastes are compacted in a 1000 psi compactor, except for the fluorinator solids, which are shipped without treatment. Size reduction may be necessary for some of the wastes to allow them to fit into the 160-gallon compaction canisters.

Cementation was selected for Option 3 where the objective is to produce one waste form type. For this process the hulls and hardware are mixed with cement in a mixer and then poured into 160-gallon canisters. Cement is simply cast over the failed equipment, but the other wastes are shredded where necessary and then placed into the drum or canister with cement and water. The cement is mixed in the drum or canister and allowed to set up.

In Option 4 the volumes of the wastes are reduced by melting. All of the metallic wastes are melted together, which results in an alloy of lower melting temperature than the "original" alloys. The vacuum induction melting method was selected for melting. In this process the metals are melted in a large crucible and then cast into the final thick-walled canisters. The filters are shredded and incinerated with other combustible materials to remove the glues and oxidize the aluminum. (Note, subsequent tests indicated that aluminum oxidation by this technique may not be feasible.) The remaining metals are added to the metallic wastes above. The general process trash is sorted into metals, plastics, rubber, and cellulose. The metals are combined with those above, the cellulose is incinerated, and the plastics and chloride-containing rubbers are hot pressed. The ash and incinerator scrubber residues are melted as a separate batch in the melter. Fluorinator solids are also melted.

Option 5 is the most complex treatment strategy. The hulls are separated from the hardware and cryogenically cracked to form quarter sections that allow access of the decontamination medium. After treatment in a centrifugal barrel finisher, the hulls can be classed as LLW. Most failed equipment is decontaminated. The filters are shredded and incinerated. The TRU fluorinator solids are blended with LLW fluorinator solids to become LLW. The general process trash is sorted. All the metals, other than hulls, are decontaminated where possible. The plastic and rubber are also decontaminated with the metals. The cellulose is incinerated with the filters. Ash and scrubber residues and decontamination sludges are added to the HLW vitrification process assumed to be present at the reprocessing plant. The secondary waste sludge from decontamination of the hulls turns out to be a significant stream. Therefore a Suboption B was also included, where the hull decontamination sludge was treated separately by a hot pressing process. Residual materials that could not be decontaminated are compacted.

The sixth treatment option produces waste forms without any combustible content. The metallic wastes in Option 6 are melted as in Option 4. All filters and trash are shredded and incinerated. The ash, residues and scrubber solids are mixed with cement by an in-drum process. Fluorinator solids are cemented with sufficient cement to immobilize them and to convert them to LLW (about a 30% loading in the cement).

Based on the treatments schemes for each of the 6 strategies discussed above, the final packaged wastes volumes are shown in Table V. Option 5 has the smallest volume of waste that would go to the repository and the largest volume of LLW. Option 4 has the smallest total volume of all categories of waste. The volume numbers are an indication of the disposal

TABLE IV

Description of Each Treatment Option by Waste Type

Option	Waste Type					Remarks
	Hardware and Hulls	Failed Equipment	Filters	Fluorinator Solids	General Process Trash	
1 No treatment	Package as generated	Package as generated	Package as generated	Package as generated	Package as generated	Package as generated
Container size, gal	TRUW: 600	TRUW: 55, 600	TRUW: 55, 80, 600	TRUW: 55	TRUW: 55, 600	
2 Minimum treatment	RH compact in CH or RH batches	Size reduce as required and RH compact in CH or RH batches	Size reduce as required and RH compact in CH or RH batches	Package as generated	Size reduce as required and RH compact in CH or RH batches	One compactor will compact batches of CH or RH waste
Container size, gal	TRUW: 160	TRUW: 160	TRUW: 160	TRUW: 55	TRUW: 160	
3 Minimum number of processes and products	Premix with cement and package in original container	Size reduce as required. Pour cement over failed equipment in original container.	Shred in either CH or RH shredder. Cement in CH or RH batches in RH in-drum mixer.	Use to replace aggregate for cementing of shredded filters and GPT waste.	Shred in either CH or RH shredder. Cement in CH or RH batches in RH in-drum mixer.	Aluminum in the filters is a potential problem. May want all RH waste in 600 gal cans. Cementing could transform TRUW to LLW.
Container size, gal	TRUW: 600	TRUW: 55, 600	TRUW: 55	TRUW: 55		
4 Maximum volume reduction without decontamination	Size reduce as required and melt in RH batches in melter	Size reduce as required and then melt in either CH or RH batches in melter	Shred filters in CH or RH shredder. Incinerate shredded filters. Collect metals and melt in CH or RH batches. Melt ash and media in CH or RH batches.	Melt in RH batches in melter	Sort. Melt metals in CH or RH metal batches in melter. RH hot press plastic and rubber in CH or RH batches. Burn cellulose. Melt ash and scrubbing solution residues in either CH or RH batches.	
Container size, gal	TRUW: 160	TRUW: 160	TRUW: 160	TRUW: 160	TRUW: 160	
5 Maximum volume reduction with decontamination	Cryogenic cracking of hulls. Centrifugal decontaminate hulls to LLW. Decontaminate hardware to LLW by vibratory finishing Option A: Decontamination solution is vitrified with HLW glass Option B: Decontamination solution is dried and then hot pressed	Sort. Size reduce as required. Compact non-decontaminables. Vibratory finish decontaminable failed equipment to LLW. Decontamination solution is vitrified with HLW glass.	Shred in either CH or RH shredder. Incinerate in RH incinerator. Separate metals from ash and media. Vibratory finish metals to LLW. Vitrify ash, residues, media and decontamination solids with HLW glass.	Blend with other fluorinator solids as LLW in RH blender	Sort and size reduce as necessary. Decontaminate portion of the metals and all of the plastic and rubber to LLW. Shred and burn cellulose. Decontaminate scrubbing solution to LLW and cement decontamination solution to TRUW. Vitrify ash, scrubber and decontamination solids with HLW glass. Compact non-decontaminable metals with failed equipment.	Chlorine and organics in HLW are potential process problem. Possible pre-oxidation step needed for Zr fines.
Container size, gal	LLW: 160 HLW Suboption A: 85 HLW Suboption B: 53	TRUW: 160 LLW: 160 HLW Suboption A: 85 HLW Suboption B: 53	LLW: 160 HLW Suboption A: 85 HLW Suboption B: 53	LLW: 160	TRUW: 55, 160 LLW: 160 HLW Suboption A: 85 HLW Suboption B: 53	
6 Noncombustible waste forms	Same as Option 4	Same as Option 4	Shred in either CH or RH shredder. Burn. Cement ash, media, concentrated scrubbing solution and metals in HR in-drum mixer.	Mix with sufficient cement in in-drum mixer for LLW	Treat noncombustibles as failed equipment. Burn all combustibles. Cement ash and scrubber solution.	Option objective: no combustibles to the repository
Container size, gal	TRU: 160	TRU: 160	TRU: 55	LLW: 160	TRU: 55, 160	

and transportation costs that will follow. The volume reduction achieved through Options 4 and 6 for all waste categories is more than a factor of 10. The maximum volume reduction factor for repository-bound waste is in Option 5, and is greater than 40.

TABLE V

Volumes of Treated TRUW from Reprocessing Plants for the Six TRUW Treatment Options, m^3/1000 MTU

Option	Remote-Handled TRUW	Contact-Handled TRUW	Increase in		Total TRUW and HLW
			HLW	LLW	
1	653	320	-	-	963
2	200	37	-	-	237
3	573	59	-	-	632
4	73	5	-	-	78
5A	9	3	53	353	65
5B	15	3	3	353	21
6	73	19	-	38	92

Costs

Life-cycle costs were developed for the total waste management system for each of the treatment options for wastes from 70,000 MTU of spent fuel. These costs include treatment costs, transportation costs and disposal costs. Where incremental costs for treatment, transportation and disposal for high-level wastes occur (Option 5), and low-level wastes (Options 4, 5, and 6), these costs are added to those for the TRUW activities. Costs are given in 1985 dollars.

Treatment costs include capital costs for the commercial treatment facility (as additions to the reprocessing plant and including amortization of the facility costs) including canister costs. These costs are based on those in a prior study[3] with modifications for differences in this study. Transportation costs are based on commercial transport used in the McKee study, with updated unit costs.(4) Repository disposal costs were estimated using the RECON repository cost model.[5] This model calculates the life-cycle construction, operating and decommissioning costs of geologic repositories (exclusive of siting and site confirmation and development costs). The repository concept basis used is a conceptual design of a repository in basalt with a total capacity of HLW from 70,000 MTU.[6] It was assumed that RH-TRUW would be emplaced in repository boreholes as done for HLW, and CH-TRUW would be emplaced in the access tunnels to the HLW boreholes. LLW disposal costs were based on the disposal costs at the commercial burial ground near Barnwell, South Carolina.[7]

The life-cycle costs for the 6 strategies are given in Table VI. The capital cost is higher than operating costs for the treatment costs in all options. The total TRUW-related treatment costs increase from \$1.3 billion for no treatment (Option 1) to \$3.3 billion for Option 5.

Transportation costs vary widely, from a maximum of \$1.5 billion for no treatment to about \$0.1 billion for Options 4, 5 and 6. The effects of final waste volumes on transportation costs are readily seen here. In all but Options 5A and 5B, the savings in transportation costs alone provide economic justification for additional treatment (and volume reduction) costs. It should be also be pointed out that for the Suboptions 5A and 5B, the transportation costs for the incremental LLW are greater than for the TRUW, and in Suboption 5A the transportation cost for the incremental HLW is about 80% of the total transportation costs.

Disposal costs also decrease significantly with volume reduction, although they are somewhat less

TABLE VI

Costs for Treatment Transportation and Disposal of TRUW and Incremental HLW and LLW from 70,000 MTU of Reprocessed Spent Fuel (undiscounted 1985 \$ billions)

Option	Treatment			Transportation	Disposal(c)	Total	Cost Reduction Relative to Option 1
	Capital	Operating	Total				
1	0.83	0.49	1.32	1.51	2.40	5.23	--
2	1.23	0.34	1.57	0.34	1.72	3.63	1.60
3	1.34	0.30	1.64	1.32	2.13	5.09	0.14
4	1.86	0.51	2.37	0.13	1.03	3.53	1.70
5A(a,b)	2.51	0.74	3.25	0.30	1.40	4.95	0.28
5B(a,b)	2.51	0.73	3.24	0.07	1.34	4.65	0.58
6(a)	1.88	0.47	2.35	0.12	1.04	3.51	1.72

(a) Includes incremental costs for secondary HLW management
(b) Includes incremental costs for secondary LLW management
(c) Includes correction for change in HLW disposal costs at the repository

sensitive than are the transportation costs. The reduced sensitivity of disposal costs to waste volume is because of the fixed costs of the repository, and the proportional share of the repository costs increase for decreasing volumes of TRUW relative to HLW. Also, in Options 5A and 5B, high surcharges for the high-activity LLW results in LLW disposal costs that are highly significant.

A comparison of the total life-cycle costs for the treatment, transportation, and disposal of TRUW from 70,000 MTU of reprocessed spent fuel is also shown in Table VI. As can be noted, Options 2, 4, and 6 have the lowest total cost and all are about the same. The most expensive option is 1, no treatment. Compared to Option 1, the potential savings from treatment of the TRUW is up to $1.7 billion for the options studied. This indicates that there is major economic incentive to implement additional treatment methods. The investment costs will be small compared to the potential pay off.

Processing and Waste Forms Considerations

The strategy analysis in this study also took into consideration the requirements for waste forms and canisters and waste processing characteristics. The waste form considerations include recognition that the requirements being developed include those for defense TRUW to be sent to the Waste Isolation Pilot Plant (WIPP), the Class C LLW requirements in 10 CFR 61, and the HLW and TRUW requirements in 10 CFR 60 for geologic disposal. Since the detailed disposal method for TRUW has not yet been established and detailed characterization data for the respective waste forms have not been obtained, the acceptability of waste forms cannot be fully judged. However, it seems likely that waste forms will be required to keep leach rates low in ground water, to provide immobilization of particulates, to have high chemical durability, and to contain no pyrophoric and combustible materials. In comparing the expected waste form characteristics in the treatment options with these potential requirements, Option 6 was judged to be the most likely to be acceptable under the anticipated requirements. Options 3, 4, and 5 were considered likely to be generally acceptable. However, there are concerns about the particulate, potentially pyrophoric, and combustible materials present in Options 1 and 2.

Processing characteristics such as operational safety, process complexity, technology status, and process flexibility were evaluated qualitatively by the authors. The simpler treatment options (Options 1 and 2) possess the more favorable processing characteristics. Options 4 and 5 were judged to have the least favorable processing characteristics of the six options, and Options 3 and 6 were judged to be intermediate.

The options were each ranked 1 (best) through 6 relative to waste form characteristics, processing characteristics and economics as given in Table VII, and based on the preceding discussions.

This simple comparison provides some valuable insights. The ranking indicates that Option 6 is the most favorable and Options 1 and 5 are the least favorable. Although waste form requirements may not be known currently, they may well provide "go/no-go" bases for evaluating the waste forms for any strategy. In that case, the options with the poorer waste form ranking (higher numbers) could well be eliminated, and the better waste forms would have a better chance of meeting the requirements. The more extensive treatment strategies (Options 4, 5, and 6) are ranked the most desirable in the waste form category, with the ranking for Option 6 as the most favorable. Option 6 also presents the most favorable system economics and has the most favorable processing characteristics of the more extensive treatment options (Options 4, 5, and 6). Option 5 appears to be the least favorable of the more extensive treatment options.

Based on these evaluations, it appears that Option 6 potentially may have the most favorable characteristics of all the options studied. Option 4 appears to have the next most favorable characteristics of the more extensive treatment options, and ranks least favorably only in processing characteristics. It should be noted that in the overall rankings, processing would have to be given greater weight than the combined categories of system economics and

TABLE VII

Ranking of the Selected TRUW Treatment Options

Option	System Economics [a]	Waste Form Characteristics [a]	Processing Characteristics [a]	Approximate Overall Ranking [b]
1	6	6	1	13
2	3	5	2	10
3	5	4	3	12
4	2	2	5	9
5	4	3	6	13
6	1	1	4	6

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[a] Ranking of from 1 (most favorable) to 6 (least favorable of the group).

[b] Approximate overall ranking is by addition of the prior three values for each option, with the lower numbers being the most favorable.

waste form characteristics to change the overall rankings. Thus, the rankings shown are believed to be reasonably appropriate.

Implementation of Options 1, 2, and 3 would require little or no R&D, but these options rank relatively poorly. Option 6 appears to offer significant potential advantages over the other options, and R&D for this option appears to be warranted in a fuel cycle with reprocessing. Next in line for potential improvements in the waste management system is Option 4.

Other options could be defined for further evaluation based on combining of favorable characteristics of specific treatments from the six options presented here. For example, a simplified decontamination option that would decontaminate the hulls and leave the remaining wastes to be treated by other methods could be examined. Hulls are of special interest because they are the largest volume stream and have high radiation levels.

Conclusions

The primary conclusion from the study is that extensive treatment of TRUW is warranted from both cost and waste form characteristics considerations, and that the characteristics of most of the processing systems studied appear to be acceptable.

The key treatment methods that appear promising are those that reduce volume or improve the quality of the waste form. These methods generally include melting, incineration, and vitrification. Simple compaction appears favorable if the waste form is determined to be suitable. The bulk of the transuranic and high-activity wastes from a central spent fuel consolidation facility, such as a repository or MRS facility, are similar to those considered here. A similar evaluation is nearing completion on these latter wastes, and early results indicate similar incentives for their treatment.

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