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An Economic Comparison of Proposed HF and Ce(IV)-Promoted Secondary Dissolution Processes for the Recovery of Plutonium from Spent Nuclear Fuel Residues

B. E. Lewis

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CONSOLIDATED FUEL REPROCESSING PROGRAM

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AN ECONOMIC COMPARISON OF PROPOSED HF AND Ce(IV)-PROMOTED SECONDARY DISSOLUTION PROCESSES FOR THE RECOVERY OF PLUTONIUM FROM SPENT NUCLEAR FUEL RESIDUES

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CONTENTS

ABSTRACT v
1. INTRODUCTION 1-1
2. PROCESS DESCRIPTIONS 2-1
2.1 Ce(IV)-Catalyzed Dissolutions 2-1
2.2 HNO_3-HF-Promoted Dissolution 2-3
3. PROCESS AND EQUIPMENT SIMILARITIES AND DIFFERENCES 3-1
3.1 Similarities 3-1
3.2 Significant Differences 3-3
3.2.1 Equipment 3-4
3.2.2 Process 3-4
3.2.3 Operator Time 3-5
4. RELATIVE COSTS 4-1
5. ANALYSIS OF COSTS 5-1
6. UNCERTAINTIES 6-1
7. CONCLUSIONS 7-1
8. ACKNOWLEDGMENTS 8-1
9. REFERENCES 9-1

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B. E. Lewis

ABSTRACT

Reprocessing of spent nuclear fuel from liquid-metal fast breeder reactors (LMFBRs) involves several steps beginning with the transport of spent fuel from the reactor to the reprocessing facility, where it is cleaned and disassembled to prepare it for chemical dissolution. Once the fuel is dissolved, the resulting solution is clarified to remove any undissolved fuel materials. The undissolved material collected in the clarification step is assayed to determine its plutonium content. If the plutonium content of the undissolved residue is greater than a preset acceptable loss limit, it may be advisable to provide a separate secondary dissolution process. Several different process options are available for the secondary dissolution step, the most viable of which involve the use of either HF or Ce(IV) as catalysts in conjunction with HNO_3 to promote the dissolution. System designs and operating procedures have been proposed for each process. The relevant capital and operating costs have been determined and compared for specified process and equipment designs for both the HF and Ce(IV) dissolution processes. It has been assumed that either process could be installed in existing cell space in a reprocessing plant such as the Breeder Reprocessing Engineering Test (BRET) facility, which will provide all necessary utilities and maintenance facilities. The cost of providing utilities, cell space, and remote maintenance has not been included in this analysis.

Based on the assumptions made in this study, the present worth cost of the HF process is ~34% more than the estimated 1984 Ce(IV) process cost of $584,000. Both processes have uncertainties that could influence their cost as well as their usefulness. The Ce(IV) process is largely unproven with irradiated fuels and would therefore require additional development. However, the potential benefits of the Ce(IV) process appear to warrant its continued study.

1. INTRODUCTION

The spent fuel from liquid-metal fast breeder reactors (LMFBRs) will contain valuable plutonia and urania, which must be recovered at a reprocessing facility. During reprocessing, the spent fuel elements undergo mechanical subdivision followed by dissolution in nitric acid. Plutonium dioxide that is not in solid solution with UO_2 dissolves at a very slow rate. However, most of the current LMFBR fuel is present as a homogeneous solid solution as a result of improved fabrication techniques and high burnup levels and temperatures in the reactor. But, in the cooler outer portions of the fuel rod, the initial nonhomogeneity of the fuel oxides remains and is believed to result in material that is difficult to dissolve. Also, at extremely high temperatures where significant centerline melting occurs, restructuring of the fuel can allow the migration and collection of some of the plutonium in plutonium-rich nodules. Residues that remain after dissolution are composed of slowly dissolving mixed oxides rich in PuO_2, and refractory fission product metals, such as Ru, Rh, Pd, Mo, and Tc. Based on past dissolution experiments, the estimated quantities of insolubles given in Table 1 are obtained.[1] These residues are collected in a disposable centrifuge bowl where they are assayed to determine their plutonium content. If the quantity of plutonium in the residue is significant, then it may be advisable to recover the heavy metals in a separate secondary dissolution process. Generally, the dissolution of these residues requires the presence of dissolution promoters, traditionally HF, to achieve reasonable dissolution rates. The HF is a very effective dissolution promoter but has certain drawbacks, some of which are (1) the dissolution rate decreases markedly with time, (2) the last fraction of PuO_2 in a given batch is difficult to dissolve, (3) new HF must be added periodically, (4) the dissolvent must be periodically replaced with fresh material, (5) prediction of the dissolution progress is difficult, (6) HF distributes throughout the reprocessing plant as a result of its volatility, (7) HF is very corrosive to process equipment, and (8) the ultimate disposal of Purex process wastes containing fluorides is extremely complicated.[2,3]

An alternative process for the dissolution of PuO_2 residues uses Ce(IV) in nitric acid as a dissolution promoter. Some of the advantages of using Ce(IV) rather than HF lie in the corrosion control and containability of process wastes. Small amounts of cerium will be present in the incoming fuel; therefore addition of more will not add any new compounds to the waste system. The corrosiveness of Ce(IV) on stainless steels is similar to HF requiring use of a titanium dissolver; however, corrosion beyond the dissolver is easily controlled by the reduction of cerium to the trivalent state. Also, cerium does not significantly distribute into the off-gas system due to its negligible volatility, making it easier to contain in the liquid phase and therefore simpler to dispose of in the waste system. Cerium(IV) is reduced to its trivalent state by reaction with Pu(IV). Since ruthenium also reduces cerium, its removal from the residues must be considered. During dissolution, cerium must be continuously oxidized to maintain the reaction.

Ideally, any secondary dissolution system should have the following characteristics: (1) it must not introduce materials into the process that interfere with downstream process steps, (2) it must not produce waste products that are difficult to handle, (3) it must be compatible with

Table 1. Estimated characteristics of BRET dissolver residues from reprocessing one MTHM[a] of Fast Flux Test Facility[b] fuel

Volume: 2.4 L; particle size: 1 to 20 μm;
particle density: 9 g/mL; bulk density: 5 g/mL

Constituent	Percent of total in fuel	Mass (g)	Activity (Ci) at 180 d[c]	Activity (Ci) at 1 year[c]	Thermal power (W) at 180 d[c]	Thermal power (W) at 1 year[c]
Alloy						
Mo	28.0	1.5×10^3	~0	~0	~0	~0
Tc	27.6	4.0×10^2	6.8	6.8	3.4×10^{-3}	3.4×10^{-3}
Ru	42.8	2.4×10^3	9.1×10^5	5.5×10^5	4.9×10^2	4.9×10
Rh	48.2	8.1×10^2	1.0×10^6	6.2×10^5	8.4×10^3	5.9×10^3
Pd	20.5	7.3×10^2	8.2×10^{-2}	8.2×10^{-2}	4.9×10^{-6}	4.9×10^{-6}
Ag	38.0	1.6×10^2	2.0×10^3	1.2×10^3	3.3×10	2.0×10
Te	7.5	8.0×10	5.3×10^3	2.0×10^3	5.9	1.8
Subtotal		6.1×10^3	1.9×10^6	1.2×10^6	9.0×10^3	6.0×10^3
Actinides						
U	0.21	1.5×10^3	1.7×10^{-2}	1.6×10^{-2}	4.5×10^{-5}	4.5×10^{-5}
Pu	0.65	1.6×10^3	2.2×10^3	2.1×10^3	5.2	5.3
Subtotal		3.1×10^3	2.2×10^3	2.1×10^3	5.2	5.3
Other[d]						
Cr	0.041	2.6×10^2	3.0×10^{-1}	2.9×10^{-3}	6.4×10^{-5}	6.3×10^{-7}
Fe	0.041	9.9×10^2	4.2	3.6	6.8×10^{-4}	1.5×10^{-4}
Ni	0.041	2.9×10^2	2.1×10^{-1}	2.1×10^{-1}	8.4×10^{-5}	8.3×10^{-5}
Inerts		1.5×10^3	0	0	0	0
Subtotal		2.8×10^3	4.7	3.8	8.3×10^{-4}	2.3×10^{-4}
TOTAL		1.2×10^4	1.9×10^6	1.2×10^6	9.0×10^3	6.0×10^3

[a]Metric tonne of initial heavy metal.

[b]Based on FFTF fuel irradiated to 60,000 MWd/MgHM at 150 MW/MgHM. These solids are collected in centrifuge bowls 5.7 cm in diam by 25.4 cm long. Each bowl has a nominal capacity of 0.5 kg or the residue from about 16 h of BRET operation.

[c]Time of fuel reprocessing following discharge from the reactor.

[d]Stainless steel components are present as metallic fines, and inerts are composed of dirt and silica.

existing materials and processes, and (4) it must be a reliable and maintainable process. Neither of the presently considered secondary dissolution processes completely meets all the above criteria. However, it is assumed that each process is technically feasible.

In this report, cost comparisons between the two processes are made based on both equipment and operating costs. To simplify the comparison, it is assumed that certain costs such as those for utilities, basic services, and cell space would be the same for each process, and therefore these costs are not included in the economic comparison. This is a reasonable simplification since the secondary dissolution process, if necessary, is anticipated to be installed in an existing facility having the necessary utilities in place.

In developing the cost estimates, use was made of information derived from the conceptual design and cost estimates for the Breeder Reprocessing Engineering Test (BRET) facility proposed for installation in the Fuels and Materials Examination Facility (FMEF) at the Hanford Engineering Development Laboratory (HEDL) at Richland, Washington. [The BRET project was recently deferred by the Department of Energy (DOE), and activities are being phased down.]

2. PROCESS DESCRIPTIONS

Each dissolution process has specific requirements that contribute to its cost. Following are descriptions of each process to facilitate comparisons of pertinent cost data.

2.1 Ce(IV)-CATALYZED DISSOLUTIONS

In the Ce(IV) dissolution process, the concentration of Ce(IV) relative to Ce(III) must be maintained at a sufficient level to be an effective PuO_2 dissolution promoter. The optimal mixture of components for this process consists of 4 *M* HNO_3 and 0.1 *M* Ce(IV) with a Ce(IV) to Ce(III) ratio >1. In order for Ce(IV) to be an effective PuO_2 dissolution promoter, ruthenium must be removed from fuel residues since it is preferentially oxidized resulting in the reduction of Ce(IV) to Ce(III) as occurs in the dissolution of PuO_2. It has been shown that ruthenium can be volatilized from a solution by oxidation to RuO_4 in the presence of a strong oxidant such as cerium.[4,5] The RuO_4 is reduced to nonvolatile RuO_2 upon contact with stainless steel.[6] Therefore, volatilized ruthenium can most probably be removed from the off-gas stream by reaction with a stainless steel mesh trap. While this combined technology has not been adequately demonstrated in controlled experiments, it is proposed here as a means of implementing the Ce(IV) dissolution process.

In order to minimize the amount of cerium used per unit of PuO_2 dissolved, it is necessary to regenerate the Ce(IV) that is reduced to Ce(III) in the dissolution and ruthenium volatilization processes. Ozone can be used to reoxidize Ce(III); however, this could potentially lead to problems in trapping ruthenium if excess ozone were present in the off-gas. Electrolytic oxidation represents an alternate means of regenerating Ce(IV). Relatively low current densities ~0.4 $A/(dm)^2$ have been successfully used with platinum screen electrodes in the regeneration process. An electrolytic cell is assumed to be an integral part of the secondary dissolver vessel. A sparged, shrouded/vented cathode, opposite an unshrouded anode, will be used to prevent the HNO_2 produced at the cathode from reducing the Ce(IV) produced at the anode. The cathode reactions for the formation of HNO_2 in the electrolysis of HNO_3 are as follows:

$$H^+ + e \rightleftharpoons (H) , \tag{1}$$

$$HNO_3 + 2(H) \rightleftharpoons HNO_2 + H_2O , \tag{2}$$

$$2(H) \rightleftharpoons H_2 , \tag{3}$$

where (H) denotes atomic hydrogen sorbed by the platinum cathode. The occurrence of secondary reactions, either (2) or (3), depends on the conditions of the electrolysis. It is important to

use low current densities to avoid production of large quantities of hydrogen. The Ce(IV) reaction with plutonium can be represented as

$$2Ce^{4+} + Pu^{4+} + 2H_2O = PuO_2^{2+} + 2Ce^{3+} + 4H^+ . \quad (4)$$

The Ce(III) is then reoxidized at the anode according to

$$Ce^{3+} \rightleftharpoons Ce^{4+} + e^- . \quad (5)$$

Once the dissolution process is complete, excess Ce(IV) must be reduced to Ce(III) to minimize corrosion in the downstream stainless steel equipment. A convenient means of reducing the cerium uses an NO or NO_2 sparge at the completion of dissolution.[7]

A flow diagram for the Ce(IV) process is shown in Fig. 1. A makeup tank will be located outside the process cell for mixing and reacting Gd_2O_3 (neutron poison), HNO_3, H_2O, and CeO_2 in the required amounts. The makeup feed solution will then be pumped in-cell to the secondary dissolver vessel containing the spent fuel residues. The solution in the secondary dissolver will be sparged and heated. An electrolytic cell located inside the dissolver vessel will

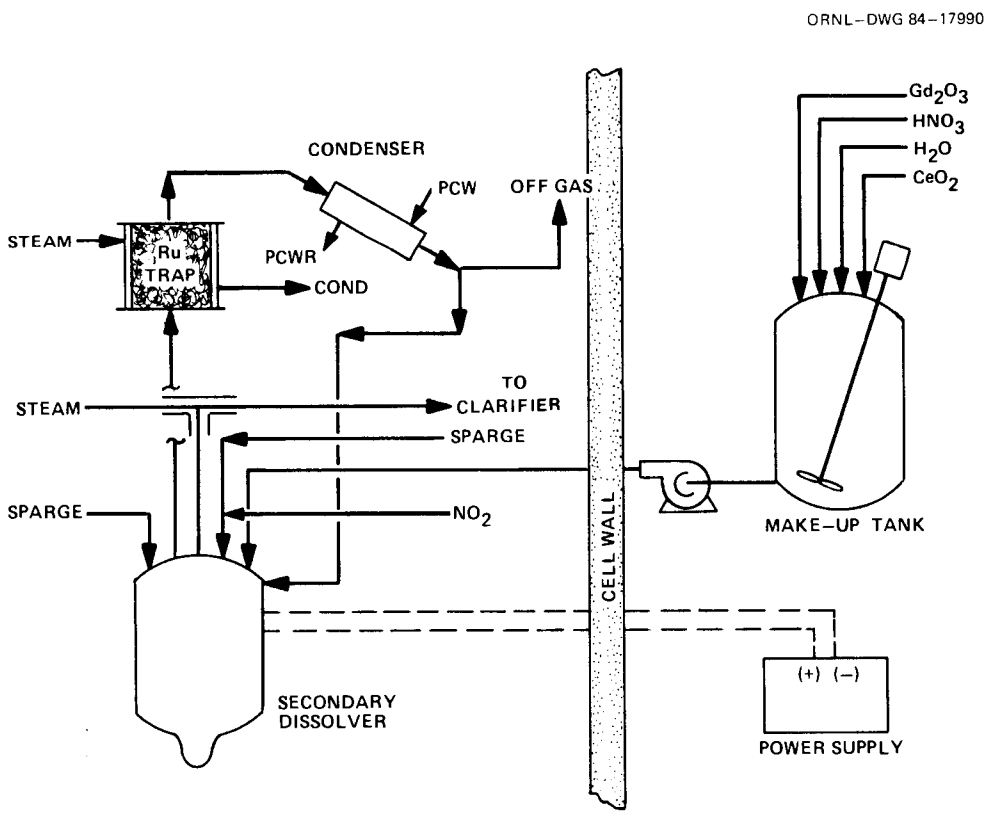

Fig. 1. Proposed flow diagram for the Ce(IV)-promoted secondary dissolution process.

continuously oxidize cerium. The power supply for the electrolytic cell will be located outside the process cell. Off-gas from the dissolver will pass through a heated ruthenium trap prior to entering a downdraft condenser. Condensate from the condenser will be recycled back to the dissolver, and the noncondensables will be sent to the facility off-gas system.

At the end of the dissolution, the dissolver solution will be cooled and sparged with NO_2 to reduce cerium to the trivalent, noncorrosive state. The dissolver solution will then be transferred to the primary clarifier in the main process.

2.2 HNO_3-HF–PROMOTED DISSOLUTION

Most often the dissolution of refractory PuO_2 from dissolver residues has been accomplished by boiling the residue in 8 *M* HNO_3 with 0.05 *M* HF.[8] Use of fluorides in process equipment presents severe corrosion problems for most materials of construction. Only tantalum appears to have sufficient corrosion resistance (corrosion rate ~1 mil/year) to withstand the HF process conditions.[9] Complexing fluoride with such reagents as aluminum nitrate reduces corrosion but also decreases the effectiveness of the PuO_2 dissolvent. Therefore, complexing will be done only after the dissolution reactions are complete. Complexing not only decreases fluoride corrosion but also aids in plutonium recovery in solvent extraction by breaking the fluoride complex of plutonium.[3] At a low nitric acid concentration (~4 *M*), fluoride is effectively complexed with a mixture of $Al(NO_3)_3 \cdot Ca(NO_3)_2$. The resulting solution allows the separation of HF in the acid recovery process with a decontamination factor of ~500 when a 3.3-to-1 mixture of $Ca(NO_3)_2$ to $Al(NO_3)_3$ is used as the complexing agent.[10]

Like the Ce(IV) process, the HF dissolution benefits from continual oxidation. The proposed rate-controlling step in the dissolution of fluorinated PuO_2 reaction sites involves the formation of a PuF_2^{2+} complex by the following reaction:

$$\begin{array}{l} \quad\;\; | \;\; / \; F \\ Pu \\ \quad\;\; | \;\; \backslash \; O \end{array} + F^- + 2H^+ \leftrightharpoons PuF_2^{2+} + H_2O + e^- . \qquad (6)$$

It is expected that the PuF_2^{2+} complex will dissociate in solution, and the electron produced in reaction (6) will absorb or react with the remaining PuO_2.[2] The Pu(IV) ion strongly complexes one fluoride and makes it unavailable for dissolution of additional PuO_2. Common practice has been to add additional HF periodically to maintain a useful concentration of free HF. An attractive alternative is to oxidize the Pu(IV) to Pu(VI), which does not form strong complexes with fluoride; this increases the free fluoride while minimizing the fluoride that must be dealt with in later process steps. The Pu(IV) can be oxidized by the addition of Ag_2O_2; however, a more convenient method utilizes electrolysis. Laboratory-scale experiments have been conducted where an applied potential of 2.0 V and current flow of 200 mA were successfully used to dissolve 0.5 to 1.0 g of PuO_2 microspheres in ~30 to 40 mL of dissolvent. In these experiments, the cathode was shielded from the anode and vented separately.[2] While

scale-up of the electrolytic regeneration process has not been demonstrated, it is assumed to be feasible for the purposes of this report. As in the Ce(IV) process, it is assumed that low current densities are applied to platinum screen electrodes in the electrolytic cell assembly to minimize the production of hydrogen as described in reactions (1), (2), and (3).

Evidence exists to suggest that Pu(VI) may be corrosive to stainless steel vessels.[11] If this is the case, then a reduction step is needed to ensure that Pu(VI) is not present in the final product from the HF dissolution process. Since the evidence in this area is not conclusive, a final reduction step has not been included in the HF process for this analysis.

To further ensure fluoride containment, a fluoride trap will be located in the off-gas line from the process. An alumina solid adsorbent will be used to remove HF vapors quantitatively from the off-gas of the secondary dissolver before it is sent to the facility off-gas system.[10] A similar trap will also be required in the facility acid recycle system. The HF traps will be heated to eliminate problems with condensation. It is assumed that the 10-L traps will be replaced approximately every 30 d.

A flow diagram for the Ce(IV) process is shown in Fig. 1. A makeup tank will be located outside the process cell for mixing and reacting Gd_2O_3 (neutron poison), HNO_3, H_2O, and CeO_2 in the required amounts. The makeup feed solution will then be pumped in-cell to the secondary dissolver vessel containing the spent fuel residues. The solution in the secondary dissolver will be sparged and heated. An electrolytic cell located inside the dissolver vessel will

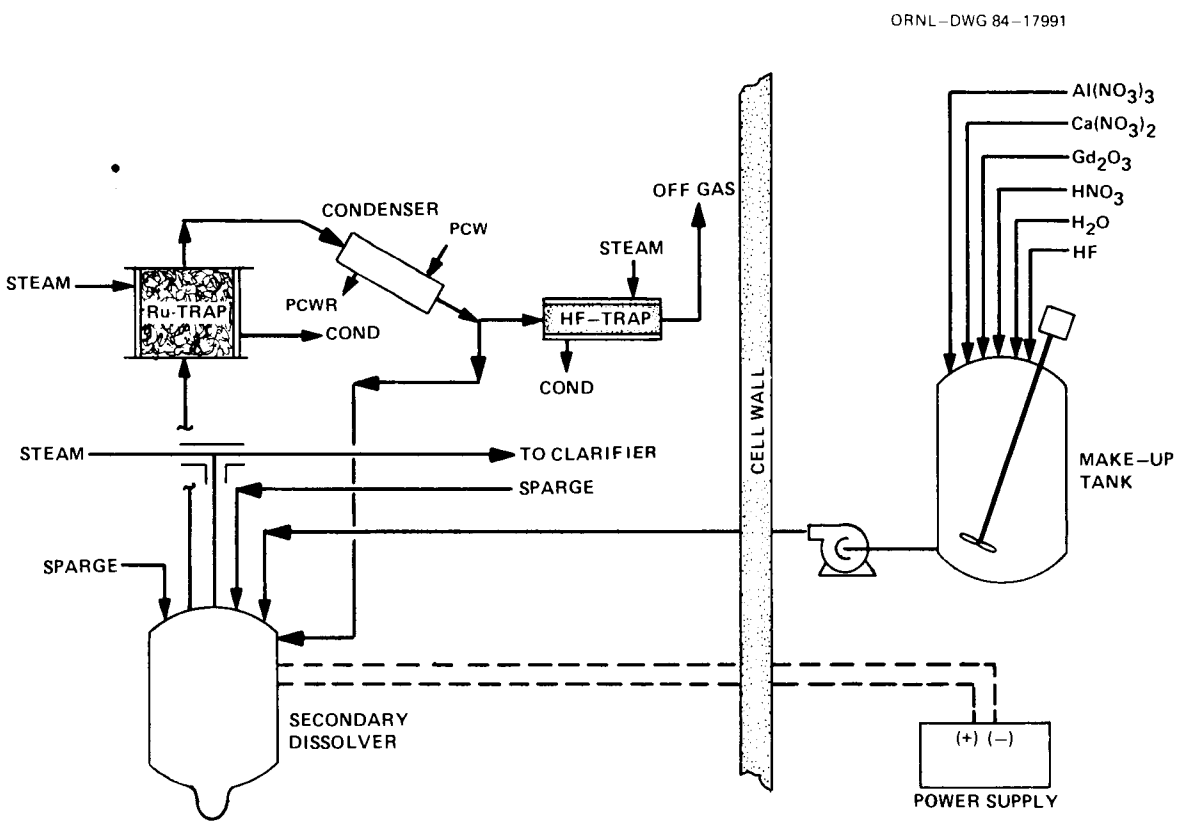

Fig. 2. Proposed flow diagram for the HF-promoted secondary dissolution process.

secondary dissolver vessel where it will be heated and sparged to effect better contact with the spent fuel residues. An electrolytic cell located inside the dissolver vessel will continually oxidize plutonium to maintain the free fluoride concentration at an acceptable level. The power supply for the electrolytic cell will be located outside the process cell. Off-gas from the dissolver will pass through a heated ruthenium trap prior to entering a downdraft condenser. Condensate from the condenser will be recycled back to the dissolver, and the noncondensables will be sent to an HF trap before going to the facility off-gas system.

At the end of the dissolution, the dissolver solution will be cooled and complexed with a $Al(NO_3)_3$–$Ca(NO_3)_2$ solution from the makeup tank. The complexed dissolver solution will then be transferred to the primary clarifier in the main process.

3. PROCESS AND EQUIPMENT SIMILARITIES AND DIFFERENCES

3.1 SIMILARITIES

Each process under consideration here will be required to handle identical forms of insoluble material. Insolubles will be contained in disposable centrifuge bowls ~2.25 in. diam and 12 in. long with small openings at each end. The dissolver vessel will be sized to hold two solid canisters and will provide for fluid recirculation and sparging for each canister. Basic dissolver design for each process is assumed to be the same. The dissolver will contain ~80 L of solution during dissolution and have a surge capacity of ~200 L.

Each process requires continual oxidation to sustain the dissolution reactions at reasonable levels. The assumed means of oxidation uses an electrolytic cell assembly with platinum-screen electrodes. A somewhat less expensive alternative electrode of platinum-plated titanium is available but was not used in this analysis. The cathode of each cell will be sparged and vented to the off-gas system. Each electrolytic cell assembly will be located in the dissolver vessel and have a separate remote dc power supply. Both processes also require a makeup tank, transfer pump, ruthenium trap, and an overhead condenser. Generally, piping is assumed to be either 3/8-in. or 1/2-in. sch 40 of the appropriate material. There are very few differences in equipment between the two processes other than materials of construction. Table 2 lists the equipment necessary for both processes and indicates whether or not each piece will

Table 2. Summary of major equipment components

Description	Required in process		Included in cost analysis
	Ce(IV)	HF	
200-L jacketed dissolver vessel	Yes[a]	Yes[b]	Yes
Electrolytic cell power supply, cathode sparge	Yes	Yes	No
150-L makeup tank	Yes[a]	Yes[b]	Yes
Centrifugal transfer pump	Yes[c]	Yes[c]	No
10-L ruthenium trap, jacketed with remote connectors	Yes[a]	Yes[a]	No
Downdraft condenser	Yes[d]	Yes[d]	No
10-L HF trap, jacketed with remote connectors	No	Yes[a]	Yes
Steam jet eductor	Yes[d]	Yes[d]	No

[a]Titanium construction.
[b]Tantalum construction.
[c]Teflon-wetted parts.
[d]Stainless steel construction.

be included in the cost analysis. Table 3 is a survey of chemical feed stocks for each process that will be included in the cost analysis.

Table 3. Chemical feed stocks

Feed stock	Required in process		Included in cost analysis
	Ce(IV)	HF	
HNO_3	Yes	Yes	No[a]
H_2O	Yes	Yes	No[a]
Gd_2O_3	Yes	Yes	No
HF	No	Yes	Yes
CeO_2	Yes	No	Yes
$Al(NO_3)_3 \cdot 9H_2O$	No	Yes	Yes
$Ca(NO_3)_2$	No	Yes	Yes
NO_2	Yes	No	Yes
Air	Yes	Yes	No
Alumina	No	Yes	Yes

[a]Not costed; source will be facility recycle system.

Basic services, utilities, maintenance, piping connectors, and cell space will not be costed in this analysis since both processes have similar requirements. Process control will be accomplished using sensors and actuators operated remotely by a central process computer. Only the costs of those sensors and actuators unique to each process will be included in the comparison; central computer costs will not be considered. A summary of the instrumentation requirements for each process and identification of those to be included in the cost comparison are given in Table 4. Each process will be operated at ~100°C with continuous agitation using gas-lift recirculation and air sparges. The overhead condensers for each process will be operated in a downdraft manner at ~20 to 25°C so that most of the condensables are recycled to the dissolver.

Table 4. Survey of instrumentation included in cost analysis

Instrument measurement or function	Required in process		Included in cost analysis
	Ce(IV)	HF	
Dissolver temperature control	Yes	Yes	No
Ruthenium trap temperature control and pressure drop	Yes	Yes	No
Steam jet controls	Yes	Yes	No
Transfer pump control/monitor	Yes	Yes	No
NO_2 sparge control	Yes	No	Yes
Gadolinium concentration monitors	Yes	Yes	No
Air sparge controls	Yes	Yes	No
Condenser temperature controls	Yes	Yes	No
Makeup tank agitator control	Yes	Yes	No
Makeup tank level/density measurement	Yes	Yes	No
Dissolver level/density measurement	Yes	Yes	No
HF trap temperature control and pressure drop	No	Yes	Yes

3.2 SIGNIFICANT DIFFERENCES

Only those differences in equipment and process operation that result in significant cost disparities will be included in the primary analysis. These differences, from Tables 2 through 4, are summarized in Table 5.

Table 5. Summary of significant process and equipment differences

Item	Process	
	Ce(IV)	HF
Process equipment		HF traps
Construction material:		
Dissolver vessel	Titanium	Tantalum
Makeup tank	Titanium	Tantalum
Piping	Titanium	Tantalum
Control loops	NO_2	HF traps ΔP HF trap temperature
Chemical feed stocks	CeO_2 NO_2	Alumina $Al(NO_3)_3$–$Ca(NO_3)_2$ HF

3.2.1 Equipment

The HF process requires two more pieces of equipment than the Ce(IV) process. The additional equipment is primarily necessary to ensure more complete isolation of HF from the off-gas system. Other differences relate to materials of construction and the number of process control loops. While both processes are highly corrosive, the HF process requires tantalum vessels and piping whereas the Ce(IV) process can be operated in titanium equipment.[4] The HF process requires instrumentation to monitor and control the performance of the HF traps. Pressure drops across the traps will be measured to determine when changeout is necessary, although for the cost analysis it is assumed that the traps will be replaced every 30 d. The HF traps also require temperature control loops to maintain the temperature at about the dew point of the entering gas stream. The Ce(IV) process will require one additional flow control loop for the NO_2 sparge used to reduce Ce(IV) to Ce(III) for corrosion control.

3.2.2 Process

Besides the obvious differences of using HF instead of Ce(IV), the HF process will use alumina as a solid adsorbent to remove fluoride from the off-gas stream. Complexing for corrosion control in the HF process will be done with a combination of $Al(NO_3)_3$ and $Ca(NO_3)_2$, whereas the Ce(IV) process will use an NO_2 sparge near the end of the dissolution period to reduce the corrosive Ce(IV) to relatively innocuous Ce(III). Table 6 gives a summary of the chemical requirements, assuming pure feed stocks, for each process. Although both HNO_3 and H_2O are used in each process in different amounts, they were not included in the cost comparison since they are assumed to be provided free of charge from the facility recycle system.

Table 6. Chemical requirements summary

Requirement	Quantity (kg/month)		Included in cost analysis
	Ce(IV)	HF	
HNO_3	40.3	80.7	No[a]
H_2O	127.8	106.4	No[a]
HF		0.16	Yes
CeO_2	2.8		Yes
NO_2	2.9		Yes
$Al(NO_3)_3 \cdot 9H_2O$		60.0	Yes
$Ca(NO_3)_2 \cdot 1.3H_2O$		86.6	Yes
Alumina		70.0	Yes
Stainless steel mesh	20.0	20.0	No
Air	3600.0	3600.0	No
Gd_2O_3	3.7	3.7	No

[a]From facility recycle system.

3.2.3 Operator Time

The operation of both the Ce(IV) and HF processes is very similar and may be operated primarily under computer control. The only operations, other than maintenance, requiring operator intervention are changeout of the traps in the off-gas train, changeout of residue containers in the dissolver vessel, and chemical feed stock replacement. Both processes require the periodic replacement of a ruthenium trap and residue containers, but only the HF process has two replaceable fluoride traps. It is estimated that the fluoride traps in the dissolver system and recycle system will be replaced every 30 d, and each will require ~2 h of operator time. Operations common to both processes have not been included in this analysis since they do not add to the cost difference between the two processes. It is assumed that each material handled in each process will be fed from the shipping containers (i.e., bottles or bags) located in a chemical makeup area. Each chemical will require an estimated 2 h/month of operator time for delivery and storage. It is also assumed that the dissolver solution will be replaced approximately every 2 weeks requiring ~3 h of operator time per chemical handled. A summary of the operator time required for each process operation is given in Table 7. The net difference in operator time requirements between the two secondary dissolution processes is an additional 14 h/month for the HF process. This difference results largely from the time required to change out the fluoride traps and for complexing operations in the HF process.

Table 7. Monthly operator time requirement summary

Process operation	Hours required		Frequency (No./month)
	Ce(IV)	HF	
Bowl changeout	16	16	8
CeO_2 dissolution	6		2
HF makeup		6	2
Gd_2O_3 dissolution	6	6	2
HNO_3, H_2O addition/sample	4	4	2
$Ca(NO_3)_2/Al(NO_3)_3$ complexing		6	2
NO_2 sparge	2		2
Ru trap changeout	4	4	1
HF trap changeout		8	2
Maintenance	8	8	2
CeO_2 delivery and storage	2		1
Gd_2O_3 delivery and storage	2	2	1
$Ca(NO_3)_2$ delivery and storage		2	1
$Al(NO_3)_3$ delivery and storage		2	1
NO_2 delivery and storage	2		1
Alumina delivery and storage		2	1
TOTAL	52	66	

4. RELATIVE COSTS

Cost data for this analysis have been collected from a variety of sources. The major differences in costs occur as a result of different construction material requirements. Only the items unique to one process or the other were costed in the primary analysis. The estimated equipment costs for the pertinent items from Table 2 are given in Table 8. The cost of the 200-L jacketed dissolver vessel was estimated from the cost of a 100-L vessel[12] using the six-tenths-factor rule[13] where

$$\text{Cost of equipment A} = \text{cost of equipment B} \left(\frac{\text{capacity of A}}{\text{capacity of B}}\right)^{0.6} . \tag{7}$$

Tantalum-lined vessels with 0.030-in. thick liners were assumed for the HF process. The cost of the HF traps was based on the estimated cost of a ruthenium trap from a recent cost estimate for the BRET facility by the Union Carbide Corporation Nuclear Division (UCCND).[14] The proposed HF trap is nearly identical to a ruthenium trap costed by UCCND. The UCCND ruthenium trap cost estimate was not adjusted for cost indexes since little time has elapsed since the original estimate.

Table 8. Cost data for equipment items unique to each process

Equipment	Cost ($)		Reference
	Ce(IV)	HF	
200-L jacketed dissolver vessel	6063	15,157	12
150-L makeup tank	3000	12,000	12
10-L jacketed HF trap with connectors (two required)		2668	14
TOTAL (to nearest 100)	9100	29,800	

Piping costs for the Ce(IV) and HF processes were based on the estimated run lengths of 3/8- and 1/2-in. pipe given in Tables 9 and 10 respectively. Only the cost of piping runs or materials unique to one or the other of the process was included in the primary cost analysis. In the HF system, tantalum was used only where solutions containing 0.05 *M* HF were in contact with the piping. All other piping was either titanium or stainless steel.

Installation costs for the equipment listed in Table 8 were based on a percentage of the purchase price. The percentage used for the installation costs for jacketed vessels was based on a combination of the low-end exchanger percentage and high-end metal tank percentage given by Peters and Timmerhaus.[13] Labor for installation of piping and connectors has been estimated to be ~3.8 times the material costs, based on the UCCND estimate for the BRET

Table 9. Material costs for piping for the Ce(IV) process

Run description		Nominal pipe size (in.)	Run length (ft)	Material	Cost ($)		Unique for Ce(IV) process	Reference
From	To				Per foot	Total		
Recycle H_2O header	Makeup tank	1/2	20	SS[a]	3.92	78	No	14
Recycle HNO_3 header	Makeup tank	1/2	20	SS	3.92	78	No	14
Makeup tank	Dissolver	3/8	60	Ti	20.25	1215	Yes	15
Sparger	Dissolver	3/8	16	Ti	20.25	324	Yes	15
Air header	Sparger	1/2	20	SS	3.92	78	No	15
Sparger	Electrode	3/8	5	Ti	20.25	101	Yes	15
Dissolver	Eductor	3/8	7	Ti	20.25	142	Yes	15
Makeup area	Dissolver	1/2	60	SS	3.92	235	Yes	14
Dissolver	Ru trap	3/8	5	Ti	20.25	101	No	15
Ru trap	Condenser	3/8	5	Ti	20.25	101	No	15
Condenser	Dissolver	3/8	15	Ti	20.25	304	No	15
Eductor	Clarifier	1/2	15	SS	3.92	59	No	14
Condenser	Off-gas	1/2	20	SS	3.92	78	No	14
TOTAL for items unique to Ce(IV) process (to nearest 100)						2000		

[a]300–series stainless steel

Table 10. Material costs for piping for the HF process

Run description		Nominal pipe size (in.)	Run length (ft)	Material	Cost ($)		Unique for HF process	Reference
From	To				Per foot	Total		
Recycle H_2O header	Makeup tank	1/2	20	SS[a]	3.92	78	No	14
Recycle HNO_3 header	Makeup tank	1/2	20	SS	3.92	78	No	14
Makeup tank	Dissolver	3/8	60	Ta	395.4	23,724	Yes	16
Sparger	Dissolver	3/8	16	Ta	395.4	6326	Yes	16
Air header	Sparger	1/2	20	SS	3.92	78	No	14
Sparger	Electrode	3/8	5	Ta	395.4	1977	Yes	16
Dissolver	Eductor	3/8	7	Ta	395.4	2768	Yes	16
Dissolver	Ru trap	3/8	5	Ti	20.25	101	No	15
Ru trap	Condenser	3/8	5	Ti	20.25	101	No	15
Condenser	Dissolver	3/8	15	Ti	20.25	304	No	15
Condenser	HF trap	3/8	3	Ti	20.25	61	Yes	15
Eductor	Clarifier	1/2	15	SS	3.92	59	No	14
HF trap	Off-gas	1/2	20	SS	3.92	78	No	14
TOTAL for items unique to Ce(IV) process (to nearest 100)						34,900		

[a]300–series stainless steel

reprocessing facility.[14] Table 11 gives a summary of the pertinent installation costs for the process equipment and piping. In the HF process, installation costs were based on titanium pipe prices plus an estimated 20% to account for handling heavier materials. This is believed to give a more accurate estimate of the installation costs for the HF process since most of the installation requirements for the two processes are similar.

Table 11. Installation costs in 1984 dollars for items unique to each system

Item	Percent of item cost	Installation cost ($) Ce(IV)	Installation cost ($) HF
200-L jacketed dissolver vessel	50	3000	4200
150-L makeup tank	40	1200	2100
10-L jacketed HF trap with connectors (two required)	50		1300
Piping	380	7700	9800
TOTAL		11,900	17,400

Chemical feed stock costs were based on the cost data given in Table 12. Table 13 summarizes the total monthly costs for chemical feed stocks. No costs were included for HNO_3 and H_2O because they were assumed to be supplied by the facility water and acid recycle systems free of charge.

Operator time costs are based on an estimated $35/man hour charge. This hourly rate includes all applicable overhead and is based on approximate wage and overhead rates for

Table 12. Chemical feed stock cost data

Feed stock	Cost ($)	Pure component unit cost ($/kg)	Reference
HF—in 10-lb polyethylene bottle, 48%	14	6.42	17
CeO_2—crystals in 1-lb bottle, 90%	24.27	59.34	17
Alumina—activated 4- to 8-mesh absorbent in 400-lb containers	408	1.02	17
$Al(NO_3)_3 \cdot 9H_2O$—crystals in 100-lb bag, commercial purity	351.45	7.73	17
$Ca(NO_3)_2$—crystals in 100-lb bag, 88%	22.87	0.57	17
NO_2—gas in 150-lb cylinders, 300-lb gross weight	1070	15.69	18

Table 13. Monthly costs for chemical feed stocks unique to each process

Feed stock	Cost ($/month)	
	Ce(IV)	HF
HF		1.03
CeO_2	166.15	
Alumina		71.40
$Al(NO_3)_3 \cdot 9H_2O$		463.80
$Ca(NO_3)_2$		49.36
NO_2	45.50	
TOTAL	211.65	585.59

union labor at the Oak Ridge National Laboratory. Using the operator time data in Table 7, the monthly costs for labor to operate the two processes are $1820 for the Ce(IV) process and $2310 for the HF process. Additional cost of operation of the HF process results from handling more material in the complexing and HF trap replacement operations.

The significant differences in instrumentation and control requirements for the two processes are summarized in Table 5. The costs of the additional instrumentation plus installation for each process is given in Table 14. Instrumentation costs were based on similar instrument costs from the UCCND cost estimate previously mentioned. These costs include instrument costs as well as labor costs for hookup to the central computer using the $35/h labor rate previously discussed.

Table 14. Instrumentation costs for measurement requirements not common to both processes

Measurement	Ce(IV)		HF	
	Quantity	Cost ($)	Quantity	Cost ($)
NO_2 gas flow control	1	4300		
HF trap ΔP			2	7400
HF trap temperature			2	13,700
TOTAL		4300		21,100

Depreciation costs given in Table 15 were based on the installed cost of piping, equipment, and instrumentation. Straight line depreciation over a 20-year useful life with a zero salvage value has been assumed.

In order to establish a basis from which to assess the significance of the relative costs determined in this work, it is necessary to estimate an approximate total system cost for one of the processes. The total estimated capital and operating costs for the Ce(IV) process are given

Table 15. Depreciation costs

Description	Ce(IV) Cost ($)	Ce(IV) Depreciation ($/year)	HF Cost ($)	HF Depreciation ($/year)
Equipment	9100	455	29,800	1490
Piping	2000	100	34,900	1745
Instrumentation	4300	215	21,100	1055
Installation	11,900	595	17,400	870
TOTAL (to nearest 100)		1400		5200

in Table 16 and 17. The costs in Tables 16 and 17 were based on a combination of similar equipment costs from the recent UCCND BRET process cost estimate and approximations. The only new element of cost not previously discussed is the connector cost in Table 16, which was taken from the UCCND BRET process cost estimate as a factor of 22.3 times the piping cost.

Table 16. Approximate total capital cost for the Ce(IV) process

Description	Cost ($)
Equipment	46,800
Instrumentation plus installation	82,900
Piping	2900
Connectors	64,500
Installation	24,200
TOTAL	221,300

Table 17. Approximate total operating costs for the Ce(IV) process

Description	Cost ($/year)
Feed stock	9700
Operator time	21,800
Depreciation	11,100
TOTAL	42,600

5. ANALYSIS OF COSTS

From a preliminary review of the cost data, it is readily apparent that the HF process is more expensive than the Ce(IV) process. Most of the difference in costs between the two processes is a result of the use of more expensive tantalum in the HF process. Normally, one would suggest that a less expensive construction material with a higher corrosion rate be used in the HF process, allowing for more frequent replacement of the equipment. However, due to the nature of the process and the expense and difficulty of replacing failed equipment in a radioactive environment, the replacement analysis was not pursued.

Tables 18 and 19 are the respective summaries of the pertinent capital and operating cost data from Sect. 4. These tables do not give the total costs of the two processes, but they do show that the HF process costs $76,000 more to install and $14,200 more per year to operate than the Ce(IV) process. The capital and operating cost differences between the HF and Ce(IV) processes are significant assuming a 20 to 25% uncertainty (as in the UCCND BRET process cost estimate) for the total system costs given in Tables 16 and 17 for the Ce(IV) process. Over the 20-year life of the plant, assuming a zero salvage value and a 10% interest rate, the present worth cost of the added capital and operating expense of the HF process is

Table 18. Total capital costs in 1984 dollars for items unique to each system

Description	Cost ($)	
	Ce(IV)	HF
Process equipment	9100	29,800
Process equipment installation	4200	7700
Piping	2000	34,900
Piping installation	7700	9800
Instrumentation plus installation	4300	21,100
TOTAL	27,300	103,300

Table 19. Total operating costs in 1984 dollars for items unique to each process

Description	Cost ($/year)	
	Ce(IV)	HF
Chemical feed stock	2500	7000
Labor	21,800	27,700
Depreciation	1400	5200
TOTAL	25,700	39,900

$196,600. The estimated total present worth cost of the Ce(IV) process from Tables 16 and 17 is $584,400. The added expense of the HF process is ~34% of the total present worth cost of the Ce(IV) process, a significant amount that could be used for additional development work on the Ce(IV) process. Table 20 summarizes these results.

Table 20. Summary of cost data

(Values are in 1984 dollars.)

Description	Ce(IV)	HF	Difference
Unique operating costs (per year)	25,700	39,900	14,200
Unique capital equipment costs	27,300	103,300	76,000
Total operating costs (per year)	42,600	56,800	14,200
Total capital equipment costs	221,300	297,300	76,000
Total present worth cost	584,400	781,000	196,600

6. UNCERTAINTIES

Cost reductions can be attained for both processes by use of tubing instead of pipe. Use of tubing in the HF process would result in a potentially greater cost savings than in the Ce(IV) process since the tantalum pipe specified for the HF process costs ~20 times more than the titanium pipe in the Ce(IV) process.

The corrosion rate of tantalum in HF solutions may be higher than originally anticipated. The previously reported 1 mil/year corrosion rate of tantalum was for exposure in a solution containing significant amounts of uranyl nitrate. Secondary dissolver solutions will contain only small amounts of uranium and plutonium nitrate. The absence of large amounts of heavy metal nitrates may lead to much higher corrosion rates.[19] Additional experiments are needed, using prototypic fuel solutions, to verify the suitability of tantalum as a material of construction.

This analysis has assumed that the technical feasibility of both processes is about equal. The Ce(IV) process, however, has only been partially successful in the dissolution of irradiated fuel.[20] The HF process, on the other hand, is the accepted standard means of dissolving plutonium on a laboratory scale. It has been hypothesized that the presence of ruthenium in irradiated fuels neutralizes the effectiveness of Ce(IV) as a dissolution promoter. This hypothesis has been accepted in this analysis, but it was further assumed that ruthenium could be volatilized from the residue and removed from the off-gas by an irreversible reaction with a stainless steel mesh trap. These processes must be demonstrated with irradiated fuels before the Ce(IV) process can be adopted.

The cost data in this analysis have primarily been based on actual vendor cost estimates for equipment, piping, instrumentation, and chemicals. Installation costs were based on material costs. A more accurate estimate of installation costs could be obtained from a detailed manpower estimate with the appropriate labor rate. However, for a preliminary cost comparison as done here, the fraction of material costs method of determining installation costs is probably sufficient.

Containment and complexing of HF in the HF process is essential for the protection of downstream process equipment. Evaporation of HF from the dissolver vessel is a potential source of release to the off-gas system. The HF traps specified in the process have been proven only on a laboratory scale. These traps should be scaled up and demonstrated prior to their use in the process. No costs for downstream process equipment corrosion due to HF attack have been included in this analysis. These costs could be significant.

The size of the dissolver vessel was assumed to be the same for each process. However, the vessel used in the Ce(IV) process could be about half the size of the one in the HF process. This would increase the size of the cost difference between the two processes.

The cost of including a reduction step for plutonium in the HF process was not included in this analysis since it is not clear that this step is essential. If the step were included, it would serve to increase the cost difference between the two processes, making the Ce(IV) process even more economically attractive.

7. CONCLUSIONS

Based on the assumptions made in this study, the present-worth cost of the HF process is ~34% more than the estimated 1984 Ce(IV) process cost of ~$584,400. Both processes have uncertainties that could influence their cost as well as their usefulness. The Ce(IV) process is largely unproven with irradiated fuels and would therefore require additional development. However, the potential benefits of the Ce(IV) process appear to warrant its continued study.

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