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J. C. Mailen[a]; J. T. Bell[a]
[a] Chemical Technology Division Oak Ridge National Laboratory Oak Ridge, Tennessee

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Potential for the Use of Hydrochloric Acid in Fission Reactor Fuel Recycle

J. C. MAILEN and J. T. BELL

CHEMICAL TECHNOLOGY DIVISION
OAK RIDGE NATIONAL LABORATORY
OAK RIDGE, TENNESSEE 37831

ABSTRACT

The chemistry and the effects of the use of hydrochloric acid (HCl) as the aqueous phase in fuel recycle are surveyed. Available data are sufficient to suggest that the separation of actinides and fission products in an HCl-trialkylamine system can be at least equal to that in the Purex process. Advantages of the HCl system are: simpler operation of the off-gas system, better separation of neptunium from uranium and plutonium, better control of the oxidation states of the dissolved species, and simpler recycle of the acid. A possible advantage is more complete dissolution of the fission products, leaving very little insoluble residue. Disadvantages include: the lack of development of methods for dissolution of oxide fuel in HCl, the sparsity of distribution data, the requirement for processing equipment constructed of tantalum, possible complications in the waste-handling system, and the dissolution of much of the cladding in the case of stainless-steel clad fuel. Systems using HCl are not attractive as replacements for Purex; however, there may be advantages to their use in some special applications.

INTRODUCTION

The Purex system for processing spent nuclear fuels uses a tributyl phosphate (TBP) extractant in a normal hydrocarbon diluent to extract uranium and plutonium from a nitric acid solution of spent

fuel. The chemical conditions of the original Purex system were partially dictated by the fact that stainless steel was the only commercially available, corrosion-resistant construction material. Other dissolvents, such as hydrochloric or sulfuric acids, were not considered since they are not compatible with stainless steel. Recently, tantalum equipment has become available that can be used in operations requiring hot HCl. Zircaloy or Hastelloy-C equipment can also be used in operations with HCl at ambient temperature (1). Since a major portion of the equipment expense in a fuel reprocessing facility results from the fabrication costs, rather than the cost of materials (the major cost of a reprocessing facility is for the concrete enclosure), the additional cost of using such corrosion-resistant equipment would be relatively small. Therefore, it may be useful to reexamine the possible use of aqueous systms other than those using nitric acid (HNO_3) for processing fuels. In fact, one flowsheet has already been proposed for reprocessing thorium-uranium fuels in an HCl system (2).

DISCUSSION

Dissolution

Fuel dissolution in HCl has not been extensively studied. Although metallic fuels would dissolve directly in HCl, there is concern about the hydrogen given off during dissolution. It would be prudent to dilute the hydrogen to below the explosive limit and to catalytically oxidize it immediately. The remainder of this discussion of dissolution will be concerned with reprocessing of the more prevalent oxide fuels.

Uranium oxide (UO_2) will not dissolve readily in HCl. Dissolution of UO_2 in HNO_3 and other dissolvents is favored by the oxidation of U(IV) to U(VI) or by the formation of strong complexes. Thus, the dissolution of oxide fuel in HCl will likely require the addition of oxidizing reagents. Scouting tests by the authors have shown that, in the absence of an oxidizing agent, dissolution of UO_2 in HCl does not occur at a significant rate. Sparging with air is not sufficient to enhance dissolution, but the addition of hydrogen peroxide H_2O_2 to hot 4 M HCl resulted in the immediate appearance of uranyl color. Dissolution ceased quickly, probably due to decomposition of the H_2O_2 in the hot HCl. The optimum dissolution conditions require a balance between the factors which increase the rate of dissolution (increased temperature, HCl concentration, and H_2O_2 concentration), and those which cause decomposition of H_2O_2 (increased temperature and HCl concentration). If peroxide cannot be used, because of safety considerations or its instability in the dissolvent (peroxide decomposition is catalyzed by the presence of iron), other oxidants would be necessary. High-fired plutonia dissolves slowly in concentrated, hot HCl, (3) but the dissolution rates in modest concentrations of HCl, and particularly as a minor component of a UO_2 matrix, are unknown. The effectiveness of HCl for the dissolution of the fission products is not known, but it can be estimated by examining the chemical forms of the fission products in the fuel (4) their dissolvability in HCl, and the solubilities of the chlorides formed (Table 1).

TABLE 1
Forms and Solubilities of Fission Products

Element	Likely form	Dissolvability in HCl	Solubility of chloride
Se	selenide	yes	oxide soluble
Br	bromide	yes	soluble
Rb	oxide, iodide	yes	soluble
Sr	oxide	yes	soluble
Zr	oxide	probable	soluble
Nb	oxide	low	soluble
Mo	alloy[a], oxide	partial	soluble
Tc	alloy[a]	partial	soluble
Ru	alloy[a]	partial	soluble
Rh	alloy[a]	partial	soluble
Pd	alloy[a]	partial	soluble
Ag	metallic	yes	soluble as $AgCl_2^-$
Sb	metallic	yes	soluble
Te	telluride	yes	soluble as TeO_2
I	iodide	yes	soluble
Cs	uranate, iodide	yes	soluble
Ba	oxide	yes	soluble
Rare earths	oxide	yes	soluble

[a]Present as noble metal alloy.

As can be seen, most of the fission products will dissolve. Silver will form the soluble dichloride complex in concentrated chloride solution. Recent tests by the authors indicate that the noble metal alloy is not readily soluble. Interactions in solution may form insoluble compounds such as Cs_2PdCl_6 (5). Dissolution of most fission products in HCl is not greatly different from that in HNO_3.

The authors have recent research results which indicate that the stainless steel cladding would be completely dissolved in HCl, eliminating the need for subsequent handling. This would, however, then increase the concentrations of iron and other stainless steel components in the solvent extraction system. The complete dissolution of the stainless steel cladding would eliminate one waste stream but would add significantly to the high-level waste. Zircaloy cladding would not dissolve appreciably in HCl.

If the reactor fuel is dissolved in a mixture of HCl and H_2O_2 (or by the use of other oxidation agents), the oxidation state of the multivalent elements will be fixed, unless a special step is added to effect a change. Table 2 presents a listing of the various oxidation/reduction ionic couples and their oxidation potential in various media (6,7). Oxidation potentials for nonchloride solutions may not apply in the chloride system. It is known that oxidation by H_2O_2 yields a mixture of Pu(III) and Pu(IV) in 6 M HCl (8). If this is taken as the potential of the solution for solvent extraction, the uranium would be present as U(VI), neptunium as Np(V), iron as Fe(III), and iodine as

TABLE 2
Oxidation Potentials for Various Ionic Couples

Ionic couple	Potential (V)	Medium	Reference
Ru(2+)/(3+)	-0.084	1-6 M HCl	Weast (7)
Mo(0)/(6+)	0		Weast (7)
Np(3+)/(4+)	0.14	1 M HCl	Heslop (6)
Sb(0)/(3+)	0.212		Weast (7)
U(4+)/(6+)	0.334		Weast (7)
Nb(3+)/(5+)	0.344	2 M HCl	Weast (7)
Tc(0)/(7+)	0.47		Heslop (6)
I(1-)/(0)	0.53		Heslop (6)
Te(0)/(4+)	0.63	2.5 M HCl	Weast (7)
Sb(3+)/(5+)	0.64		Weast (7)
Se(0)/(4+)	0.74		Weast (7)
Np(4+)/(5+)	0.74	1 M HCl	Heslop (6)
Fe(2+)/(3+)	0.77		Heslop (6)
Ru(3+)/(4+)	0.858	2 M HCl	Weast (7)
Pu(3+)/(4+)	0.97	1 M HCl	Heslop (6)
Pu(4+)/(6+)	1.052	1 M HCl	Weast (7)
Cr(3+)/(6+)	1.1	2 M H_2SO_4	Weast (7)
Np(5+)/(6+)	1.14	1 M HCl	Heslop (6)
Se(4+)/(6+)	1.15		Weast (7)
I(0)/(5+)	1.19		Heslop (6)
Ce(3+)/(4+)	1.28	HCl	Heslop (6)
Pd(2+)/(4+)	1.29	Chloride	Weast (7)
I(0)/(7+)	1.38		Heslop (6)
I(0)/(1+)	1.45		Heslop (6)
Co(2+)/(3+)	1.84		Heslop (6)
Am(3+)/(4+)	2.4		Heslop (6)

I(0). Other oxidation states may be achieved by changing the oxidation potential of the solution; for example, if the fuel is dissolved in hydrochloric acid with Ce(IV) present, the potential would increase to ~1.28 V. Under these conditions, the neptunium and plutonium would be hexavalent, and the iodine would likely be pentavalent.

One possible problem with the use of hydrogen peroxide in dissolution is the formation of insoluble peroxides of uranium or plutonium. The equilibrium solubility for uranium peroxide is given by: (9)

$$K_U = [UO_2^{2+}][H_2O_2]/[H^+]^2 = 1.94 \times 10^{-3} \quad (1)$$

and that for plutonium peroxide is given by (10):

$$K_{Pu} = [Pu^{4+}][H_2O_2]^{1.5}/[H^+]^4 = 2.1 \times 10^{-5}. \quad (2)$$

TABLE 3
Solubilities of Uranium and Plutonium Peroxides

Molar concentrations		Molar solubilities	
H^+	H_2O_2	Uranium	Plutonium
1	0.1	1.94×10^{-2}	6.64×10^{-4}
1	1.0	1.94×10^{-3}	2.1×10^{-5}
2	0.1	7.76×10^{-2}	1.06×10^{-2}
2	1.0	7.76×10^{-3}	3.36×10^{-4}
3	0.1	1.75×10^{-1}	5.4×10^{-2}
3	1.0	1.75×10^{-2}	1.7×10^{-3}
4	0.1	3.1×10^{-1}	1.7×10^{-1}
4	1.0	3.1×10^{-2}	5.8×10^{-3}
5	0.1	4.85×10^{-1}	4.15×10^{-1}
5	1.0	4.85×10^{-2}	1.31×10^{-2}

These relationships were used to calculate the solubilities given in Table 3 for various HCl and H_2O_2 concentrations.

These data indicate that solubilities of the peroxides are sufficient for dissolution of fuel in >5 M HCl with reasonable levels of H_2O_2. The solubilities are not great enough to allow adding all the H_2O_2 initially; a continuous or periodic addition process would be necessary.

Off-Gas System

Operation of the dissolver off-gas system should be simpler in HCl than in the HNO_3 system. The only volatile elements to be expected are the rare gases, Kr and Xe, I_2, and a small amount of $^3H_2$. Volatilization of the iodine from HCl, assuming that the solution potential is proper for the elemental form to be stable, would be more difficult than from nitric acid, because of the complexing of iodine by chloride to yield the I_2Cl^- ion. The equilibrium constant for the formation of this ion is ~2.5; thus, only ~0.077 of the iodine would be present as free I_2 (11). If the distribution coefficient of free I_2 from HCl to the off-gas is 68 [the same as that from water to air (12)], then the overall distribution coefficient for all iodine species, including those which are nonvolatile, is ~5. This should be sufficient for relatively easy removal of the iodine. The absence of nitrogen oxides in the off-gas would allow the use of impregnated activated charcoal as an iodine trap, without the possibility of ignition (13) or explosion (13,14). The iodine could alternatively be adsorbed on lead-, potassium-, or cadmium-exchanged zeolites (15–19). These materials, while excellent adsorbents for iodine, are less effective in the presence of NO_x and moisture. Oxidation procedures for trapping iodine, such as the Iodox (20) or electrochemical methods (21), would operate better without the reductive load of the nitrogen oxides. It is obvious that the elimination of HNO_3 would greatly increase the options for iodine handling and should significantly simplify and improve the over-

all iodine retention system. If recovery of krypton and xenon is required, the absence of nitrogen oxides would also be beneficial (22). In the nitric acid system, recycle of the nitrogen oxides requires a reaction system for the recombination of the nitrogen oxides with oxygen and water (23); simple condensation serves for the recycle of HCl from the off-gas stream. For all these reasons, the off-gas treatment facility for an HCl system would be much less complicated and less expensive than that for an HNO_3 system.

Solvent Extraction

The use of a chloride system in tantalum equipment leads to the possibility of significant changes in the solvent extraction system. Certainly, the diluent could be changed to a chlorinated hydrocarbon or a Freon-type compound, since corrosion of process equipment by chloride would not be a problem. Aromatic diluents could be considered in the absence of nitrating agents. The use of a nonhydrocarbon diluent would eliminate the possibility of fires caused by the diluent and would allow use of a much higher temperature in the solvent extraction system, when desired. The availability of a wider range of operating temperatures also increases the potential for making separations by temperature variation.

A complete discussion of the possible solvent extraction flowsheets in a chloride separation system is beyond the scope of this paper; we will briefly examine the use of tertiary amines in aromatic diluents as an example. The distribution data for this system (from three sources) are summarized in Table 4. For this discussion, it is assumed that the oxidation potential of the dissolver solution is such that the uranium is hexavalent, the plutonium is largely tetravalent, the neptunium is pentavalent, the americium is pentavalent, and the iron is trivalent. This condition could prevail if the dissolution were accomplished in HCl with H_2O_2 or if the dissolver solution's redox potential has been otherwise adjusted to a similar value. Figure 1 shows the distribution coefficients of U(VI) and Pu(IV) between HCl and 1 vol % trioctylamine (24). From these data, it is obvious that both

TABLE 4
Available Distribution Data for HCl-Tertiary Amine System

Extractant	Diluent	Elements	Reference
Adogen 354[a]	Aromatic[b]	U, Ru, Zr, Mo, Ce, Nb, Fe	Thomas (1)
Alamine 336[a]	Diethylbenzene	Fission and corrosion products plus U	Seeley (25)
Tri-n-octyl-amine	Xylene	U, Pu, Np	Keder (24)

[a]Mixture of straight-chain tertiary amines; mainly trioctyl- and tridecyl- amines.

[b]Mixture; primarily of methyl-, ethyl-, diethyl- and trimethylbenzenes.

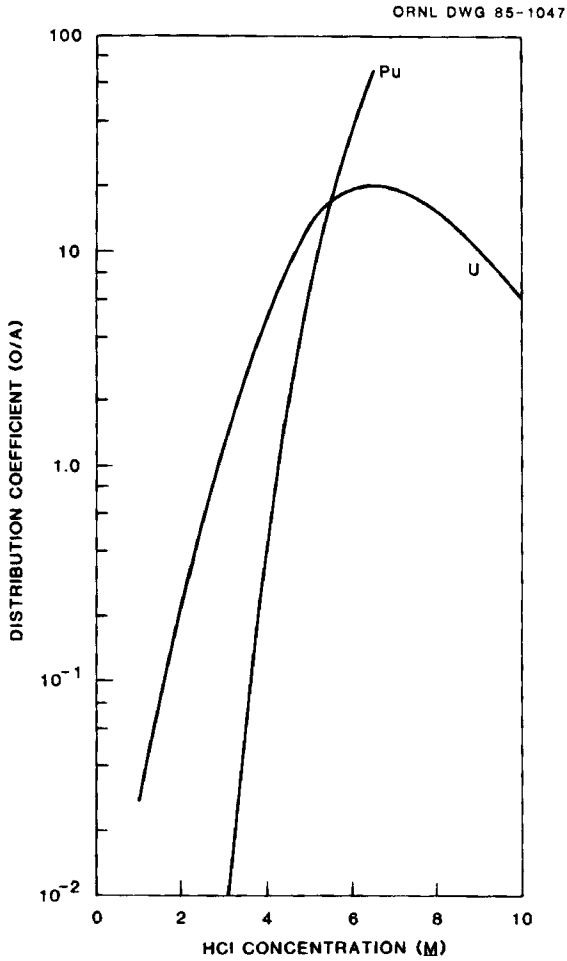

Fig. 1. Distribution coefficients of uranium and plutonium from hydrochloric acid to 1% trioctylamine in xylene [Keden (24)].

elements can be extracted from ~5 M HCl with distribution coefficients of ~20. At lower HCl concentrations, the plutonium is much less extractable than the uranium. Thus, stripping with ~3 M HCl will remove the plutonium (distribution coefficient of ~0.01) while the uranium will largely be retained (distribution coefficient of ~1.5); this is a separation factor of ~150. In order to place the literature data in perspective, it is necessary to determine the effects of HCl concentration on the extraction of U, Pu, and Np species. These results are given as power effects (the slope of the distribution vs HCl concentration on a log-log plot) in Table 5 for the various references.

Figure 2 shows the uranium data from three literature sources (1, 24, 25), with Keder's line (24) being estimated from a single point at 0.1 M and the experimentally determined effect of HCl concentration on the extraction. The plutonium line is also from Keder (24) and is estimated from a single point at 0.1 M amine concentration plus the experimentally determined effect of HCl concentration on the extraction. From this plot, the extraction of uranium and plutonium from ~5 M HCl into 0.1 M amine should give a plutonium distribution coefficient of ~100 and a uranium distribution coefficient of ~5 to 100. Since Keder's data indicate that the effect of amine concentration is the same for both uranium and plutonium, we would expect nearly the same separation factors at different amine concentrations. Stripping with 3 M HCl would give a plutonium distribution coefficient of ~0.2 and a uranium distribution coefficient between 1.2 and 12, with separation factors of 6 to 60. Again, the fact that Keder's data give a separation factor of ~150 indicates that the larger separation factor is likely. Based on these literature data, the separation of uranium and plutonium in this system should be simple; both have large distribution coefficients, requiring few extraction stages. The plutonium can be selectively stripped, leaving the uranium in the organic phase.

TABLE 5
Power Effect of HCl Concentration on Extraction of Actinides by Trialkylamines

Species	Power Effect, (Reference No.)		
	Thomas (1)	Seeley (25)	Keder (24)
U(VI)	~2	~4	~ 4[a]
Pu(IV)			~11
Pu(VI)			~ 5[a]
Np(IV)			~12
Np(VI)			~ 4[a]

[a]U(VI) has a maximum distribution from ~7 M HCl (24); a maximum was not shown by data from the other references.

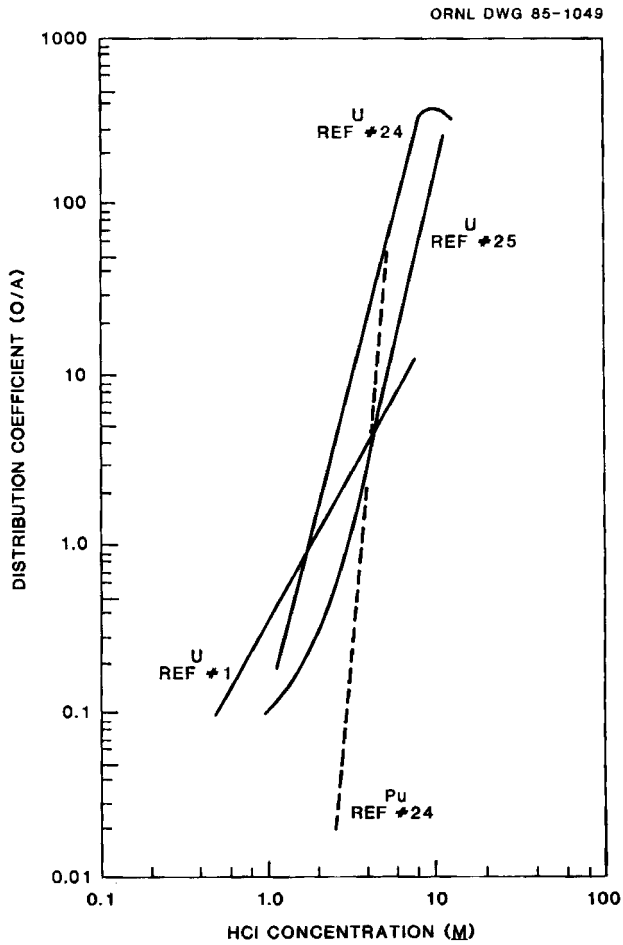

Fig. 2. Distribution coefficients of uranium and plutonium from hydrochloric acid to 0.1 M trialkylamine in aromatic diluent. Plutonium and uranium lines from Keder (24) are estimated from single data points at 4 M HCl.

Figure 3 illustrates data on the distribution coefficients of the most important extractable fission products and iron vs the HCl concentration. Zirconium, chromium, and the rare earths have insignificant distribution coefficients (25). If the initial extraction is from 5 M HCl, only Pd, Tc, and Fe will have distribution coefficients >1. If a stainless-steel-clad fuel is being processed, the mole ratio of iron to actinides could be as high as 0.18. The separation factors for separating the actinides (U and Pu) and the fission products Ru, Mo, and Nb should be ~200; thus, it should be possible to operate the extraction system to give a good separation. If the plutonium is stripped with 3 M HCl, a clean separation from Fe, Pd, and Tc should be obtained. The organic stream would then contain U, Pd, Tc, and Fe (if stainless-steel-clad fuel is processed). If the uranium is subsequently stripped with ~1 M HCl (uranium distribution coefficient of 0.1 to 0.3), the uranium could be selectively separated from the iron (distribution coefficient of ~6), the palladium (distribution coefficient of ~50), and the technetium (distribution coefficient of ~300). If a high recovery level of uranium and complete separation from iron are required, it may be desirable to reductively strip the iron from the solvent.

Solvent Cleanup

Solvent degradation should be much lower in an HCl system, since a significant portion of the solvent degradation in the Purex system is due to attack on the TBP and the diluent by nitric and nitrous acids. However, irradiation of HCl will generate chlorine free radicals, and these are expected to slowly convert the aromatic diluents, if used, to chlorinated cyclic compounds. Studies in this area would be useful, if such a process is contemplated. The degradation products of the amine system are less a problem in the formation of interfacial cruds and retained cations than are those from TBP, since the dialkylamines are poorer complexing agents than the trialkylamines (25).

One necessary cleanup operation in the HCl system will be the removal of the retained fission and corrosion products. Complete removal of palladium and technetium may require a reductive stripping or the use of a complexing agent. The iron can be removed by any of several methods, including a low-acid strip, reductive strip, or a strip with a complexing agent.

Other Approaches to Uranium/Plutonium Partitioning

It is possible that the separation of uranium and plutonium could be achieved by variations in acid concentration and temperature, since the temperature range of operation could be quite large for chlorinated diluents.

The separation of uranium and plutonium by valence adjustment combined with extraction or stripping operations should be easier in a chloride system. Plutonium(III) is stable in HCl without holding reductants, as contrasted with the HNO_3 system, where hydrazine is commonly added to destroy nitrite which acts as a catalyst to the HNO_3 oxidation of Pu(III) to Pu(IV). Electrolytic reduction should also be more straightforward in the HCl system than the nitrate system, without the

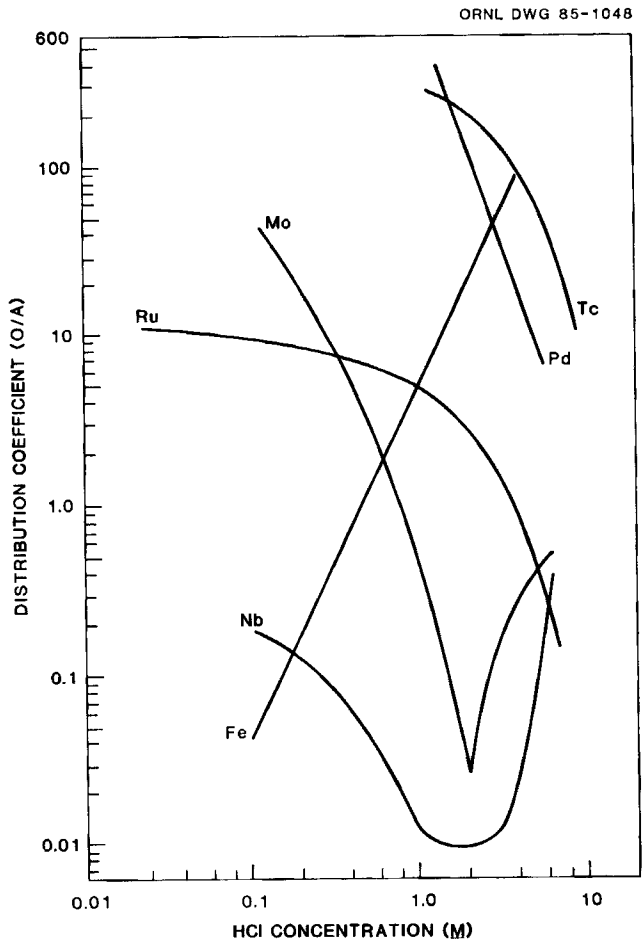

Fig. 3. Distribution coefficients of fission products and iron from hydrochloric acid to 0.1 M trialkylamine in aromatic diluent. Ru, Mo, Nb, and Fe data from Thomas (1); Pd and Tc data from Seeley (25).

necessity of hydrazine addition. Photolytic reductions, such as the formation of U(IV), should be more efficient in HCl systems, since the absorbance of other components of the solution will be reduced, and reverse reactions should be less important.

Waste Handling

Recovery of the HCl from the waste would be by distillation. No decomposition of HCl would occur, whereas with HNO_3 the nitrogen oxides must be reformed into HNO_3 to obtain complete recycle. The recovered, azeotropic HCl would be ~6 M; concentrations higher than this should be avoided in the process to simplify the acid recycle.

Denitration of the waste would also not be required with an HCl system. Solubilities should be as high or higher than those in a nitrate system, allowing waste concentration to proceed normally. Waste calcination would require equipment resistant to chloride attack, and the off-gas systems should be designed to cope with wet HCl and, for glass waste forms, $SiCl_4$. Pyrohydrolysis of the waste to convert chlorides to oxides might be necessary. Again, an additional cost for processing equipment materials resistant to chloride attack would be required.

Materials Options

It would be advantageous if some parts of a reprocessing plant using HCl could be made of polymeric materials. Polyvinyl chloride and Kynar materials offer some excellent advantages, especially with lower radiation and temperature levels. Such materials are currently used in commercial processing of nonnuclear materials in chloride systems.

CONCLUSIONS

Fuel reprocessing using an HCl system with a tertiary amine as the extractant appears chemically feasible. The advantages of this method are: (1) good separation of uranium and plutonium from fission products and neptunium, (2) separation of uranium from plutonium without use of reductive stripping, (3) improved off-gas handling, and (4) ease of recycle of the HCl. Disadvantages of the HCl system include: (1) the requirement for tantalum processing equipment, (2) undeveloped dissolution methods for oxide fuels, (3) significant dissolution of the stainless steel cladding, and (4) possible complications in waste handling.

The primary question is whether a chemically feasible, apparently advantageous, HCl solvent extraction process should, or can, be developed to compete with the relatively mature Purex process. The Purex process is an accepted, proven method that is being commercialized in several countries and considerable development work on fuel dissolution and solvent extraction distribution coefficients would be required before an HCl process could be competitive. It seems unlikely that funds for the required R&D will be available in the foreseeable future. Even if no technical impediments are found in the HCl scheme compared with the Purex system, many man-years of effort would be required to develop the new process to the current maturity of Purex. Hence, although some effort to establish a better data base for an HCl process and increase the general knowledge of fuel reprocessing are justified, it is probably impractical to pursue this new process.

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