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Study of Radionuclide Leaching from the Residues of K Basin Sludge Dissolution

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Abstract: The sludges remaining in the K Basins after removal of the spent N Reactor nuclear fuel will be conditioned for disposal. After conditioning, an acid-insoluble residue will remain that may require further leaching to properly condition it for disposal. This document presents a literature study to identify and recommend one or more chemical leaching treatments for laboratory testing, based on the likely compositions of the residues. The processes identified are a nitric acid – cerate leach, a silver-catalyzed persulfate leach, a nitric-hydrofluoric acid leach, an oxalic – citric acid reactor decontamination leach, a nitric – hydrochloric acid leach, a ammonium fluoride – nitrate leach, and a HEDPA – formaldehydesulfoxylate leach. All processes except the last two are recommended for testing in that order.
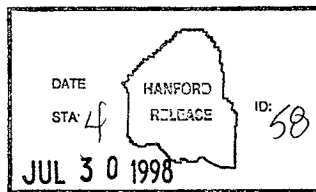
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1.0   INTRODUCTION

The sludges remaining in the KE and KW fuel storage basins after retrieval of the fuel are likely to contain hydrided uranium and uranium corrosion products, transuranic corrosion products, fission products, steel corrosion products, aluminum corrosion products, Zircaloy pieces, degraded concrete, sand, ion exchange beads and Zeolon ion exchange pellets (Makenas 1996, 1997, 1998). These sludges will have to be disposed of, but no one facility at Hanford will accept them *in toto* in their current state. The sludge conditioning project will develop the method(s) by which the sludges will be treated to separate the constituents and direct them towards the appropriate disposal facilities; either ERDF, for particulate, low-level radioactive, non-TRU wastes, or the TWRS tank 241-AW-105, for radioactive but otherwise non-PCB, non-reactive waste.

The challenge will be to successfully treat the various sludge components without adding any unacceptable ingredients to the treated product forms (Fowler 1995) (Corriveau 1996), and to treat the sludge safely and economically. Thus far, the general strategy (Pearce 1998) is to consolidate the sludges after fuel washing in the two basins into five streams. Each of these streams will be treated without further mixing. A testing strategy has been issued to develop the details of the general strategy (Flament 1998). Batches of sludge will be stripped of their ion exchange resins and then dissolved in nitric acid (de Lamartinie 1998). The filtered rinse of this dissolution will be treated to reduce its criticality hazard, then neutralized to be disposed of in tank 241-AW-105. It will contain no reactive solid components. On the other hand, the filtered particulate residue that didn't dissolve in nitric acid could possibly contain concentrations of fission and transuranic radionuclides (Carlson 1998) unacceptably high for burial in ERDF.

It is the purpose of this study to look at methods to remove the surviving radionuclides from the residues to the extent that the residue would meet ERDF criteria, and that the rinses would meet TWRS waste compatibility criteria, and recommend one or more methods to be tested for radionuclide leaching effectiveness.

2.0   BASES FOR ASSUMPTIONS AND CONDITIONS

A number of issues will affect our formulation of conditions to be met in this project. We begin by considering the restrictions that might be placed on disposing of the products of sludge leaching, namely the ERDF and TWRS acceptance criteria. Next, we examine what is presently known about the nitric acid dissolution performance and composition of residuals therefrom, to see what sort of additional decontamination

factors (DF) might need to be achieved. Then, we consider the plausible residue types that may be responsible for requiring additional DF's. Finally, we note the waste streams that will be processed and the probable residue types to be found in each.

2.1 DISPOSAL ACCEPTANCE CONSIDERATIONS

2.1.1 ERDF

ERDF has a list of limits on specific waste constituents, and a sum-of-fractions rule on wastes containing multiple listed constituents (Corriveau 1996). Of greatest importance to the present study are the non-TRU (<100nCi/g) limit on transuranics, the 32 Ci/m^3 limit on ^{137}Cs, and the 0.012 Ci/m^3 limit on ^{238}U and its daughters. Other limits are not likely to be exceeded by unleached residues.

2.1.2 TWRS Double Shell Tanks

TWRS defines the acceptability of wastes through a waste transfer compatibility program (Fowler 1995) that sets few clearly defined limits. Of that few, however, there are some of relevance to the present program because they make application of traditional iron-dissolution technology problematical. Chloride is limited in tank cars to 0.01M, while phosphate is generally limited to 0.1M. If the chelate content is equivalent to or greater than 10 g/L Total Organic Carbon (TOC), then the waste would be considered a chelate waste, which complicates (but does not rule out) the transfer to double shell tanks. Tank 241-AW-105 is not currently used as a complexant concentrate tank.

There appears to be no consideration in the TWRS waste compatibility program towards the effect of a waste constituent on future vitrification processes. Certainly the Disposal Program would appreciate not having to deal with more phosphate, sulfate or noble metals than are already present in the tanks, or with graphite. All these materials are detrimental to vitrification of tank wastes in glass.

An important point to keep in mind when judging the acceptability of leaching solutions for TWRS disposal is that the leaching solutions are approximately 15% of the total conditioning stream going to TWRS (de Lamartinie 1998), *i.e.*, there will be a 6.5-fold dilution of the

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leaching liquid waste stream that may allow some flexibility in selecting a leaching process.

These considerations are not completely certain, and in the final determination TWRS will have to help decide the acceptability of proposed process wastes for disposal.

2.2 DECONTAMINATION FACTORS REQUIRED

Recent testing results of nitric acid dissolution of a variety of KE basin sludges (Flament 1998a) provides data on actual residue compositions summarized in Table 1 below.

TABLE 1. DISSOLUTION TESTING RESULTS OF KE SLUDGE COMPOSITES AND RESIDUES										
Sludge and Residual	U (μg/g)	Fe (μg/g)	Ca (μg/g)	Al (μg/g)	Si (μg/g)	^{137}Cs (μCi/g)	^{241}Am (μCi/g)	^{239}Pu (μCi/g)	^{238}Pu (μCi/g)	TRU (nCi/g)
Canister Composite	4.22×10^5	7.42×10^3		1.14×10^4	4.69×10^3	498	59	74.4	61	1.94×10^5
Average Canister Composite Residuals	3.89×10^4	4.53×10^4		1.77×10^4	2.74×10^5	81.2	6.49	17.9	7.97	3.24×10^4
Maximum Canister Composite Residuals	5.36×10^4	1.81×10^5		2.49×10^4	3.56×10^5	155	9.27	26.9	10.6	4.68×10^4
Canister #96-08 Sludge	4.00×10^5	7.30×10^4		7.90×10^4		1180	77.3	17.4	93.5	1.88×10^5
Average Canister #96-08 Residual	2.10×10^4	3.14×10^4	1.93×10^4	4.44×10^4	3.09×10^5	72.4	5.05	5.86	0.36	1.13×10^4
Maximum Canister #96-08 Residuals	2.29×10^4	3.45×10^4	2.09×10^4	4.54×10^4	3.20×10^5	77.2	5.55	6.33	0.37	1.23×10^4
Floor/Weasel Pit Composite	5.41×10^4	2.99×10^5	4.66×10^4	1.13×10^4	6.06×10^4	825	55.3	14.1	14.3	8.36×10^4
Average Floor/Weasel Pit Residuals	2.12×10^3	6.66×10^4	1.85×10^4	5.82×10^3	3.07×10^5	21.0	0.311	0.832	0.864	2.01×10^3
Maximum Floor/Weasel Pit Residuals	2.42×10^3	2.12×10^5	2.26×10^4	7.58×10^3	3.55×10^5	64.5	0.592	2.99	3.03	6.61×10^3
Weasel Pit #T-20 Sludge	5.84×10^4	2.40×10^5		5.30×10^4	3.26×10^5	550	6.54	7.98	1.27	1.58×10^4
Average Weasel Pit #T-20 Residuals	4.60×10^3	1.57×10^5	6.10×10^3	1.77×10^4	2.56×10^5	45.2	0.254	0.393	0.061	7.07×10^2
Maximum Weasel Pit #T-20 Residuals	5.00×10^3	1.92×10^5	6.61×10^3	1.82×10^4	2.89×10^5	45.7	0.278	0.434	0.067	7.79×10^2

It is found in general that cesium, uranium, plutonium and americium remain above the individual ERDF disposal criteria. Not surprisingly, the residues derived from canister sludge contained the highest concentrations of radionuclides. An approximate, conservative calculation using the maximum values of these test results indicates that a leaching decontamination factor (DF) of 1360 would be needed to bring plutonium below ERDF limits. Americium would require a DF of 273, cesium a DF of 7, and uranium a DF of 2.2. A DF of 530 would be needed to bring the waste below TRU levels. However, the sum of fractions rule would require a uniform 2060 DF over all these nuclides to bring the worst residue

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as a whole into compliance with ERDF limits. A doubling of that factor to approximately 4000 would provide an allowance for process variation.

2.3 EXPECTED OR POSSIBLE RESIDUE TYPES

Several types of sludge components that are expected to be insoluble in the nitric acid leach will present varying problems in disposal of the residues. The processes to be selected will depend to a great extent on which of these materials actually contain the radionuclides.

2.3.1 Grafoil

The bits of grafoil to be found in KW sludges are expected to be removed in the ion exchange resin separation process and additionally are expected to be inert to actinide absorption. The treatment developed for removing actinides from the resin will be equally effective for washing Grafoil. Hence Grafoil is not considered further.

2.3.2 Organic Ion Exchange Resin

A small amount of ion exchange resin can be expected to escape the resin separation process and find its way to the acid-insoluble residue. Unfortunately, it is possible that only a small amount of resin could carry sufficient actinides into a residue to make it TRU waste. Thankfully, methods for treating that resin for ERDF storage are being considered in detail elsewhere (Dodd 1998). One of those methods may have to be considered when treating residue containing significant amounts of resin.

2.3.3 Zircaloy-2 Pieces

Most of the loose Zircaloy-2 cladding generated by the disintegration of N Reactor fuel is expected to be in pieces larger than 0.25 inch, and thus will not be a part of the sludges. However, a small amount of cladding particulate may find its way to sludge residues. It has been shown that N Reactor fuel hulls contain greater than TRU levels of infused actinides (Swanson 1985), which would make them unacceptable for disposal to ERDF. Even after two dissolutions with nitric acid, these hulls contained 420 nCi/g of transuranic alpha emitters, as well as 20 μCi/g ^{137}Cs and 1200 μg/g uranium. Continued treatments with

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boiling nitric acid did reduce these values somewhat, but not enough to make the hulls non-TRU. If Zircaloy pieces turn out to be the carriers of significant amounts of actinides or cesium in a residue, then they likely will have to be dissolved in something like a PUREX cladding dissolver solution, because they will equally likely be inert to leaching methods proposed for other residue constituents.

2.3.4 Active Siliceous Minerals

Recent testing at PNNL (Carlson 1998) (Flament 1998a) has shown that nitric acid - insoluble residues from a canister sludge composite are only about 1% of the total sludge, but that these residues require additional decontamination of the actinides U, Pu and Am. In addition, it was found that these residues consist mostly of amorphous silica with small amounts of the mineral goethite, or FeO(OH). Thus, it is important to consider whether the actinides could be associated with the silica. In alkali, it has been shown that plutonium (IV) can interact with silica and iron oxides, but not with aluminum oxides (Krot 1998). It is purely speculative to contemplate that this would also hold true in neutral solution, so we have less confidence this is actually occurring.

Another opportunity for actinide-silicon interactions is afforded by the composition of, and corrosion of, the metallic N Reactor Fuel. The fuel was made of uranium alloy 601 (Westinghouse 1988) (Weakley 1979), containing up to 125 ppm silicon among other constituents.

A third opportunity for interaction lies in the KE basin concrete surfaces. The friable nature of the first few millimeters of these surfaces indicates that some corrosion and release of solid, silicate-bearing concrete has taken place. The basin water had a neutral pH, and hence the actinide solubilities were very low (Thomason 1992) But it is still possible that actinides and active silica interacted in solution over two decades' time.

The concrete surfaces of KE basin are friable and have shown a strong affinity for radiocesium (Bechtold 1981). For this reason some concrete constituents along with absorbed cesium can be expected to be found in the residues. In addition, during past operations at KE basin at least one accidental release of 0.57 m^3 of Zeolon 900 ion exchange medium occurred (Hoefer 1997). This medium is strongly cesium specific.

If radionuclides are indeed mineralized with silica, then their release may require a reagent known to react with silica, such as hydrofluoric acid.

2.3.5 Iron Compounds

As mentioned above, recent testing has shown that a small amount of goethite, FeO(OH), survived the nitric acid dissolution step. There has been continuous corrosion of steel and reprecipitation of hydrous iron oxides in both KE and KW basin since their use as supplemental fuel storage pools began, and it is well known that iron hydroxide precipitation is a good carrier of plutonium. Furthermore, the N reactor fuel alloy contained up to 400 ppm iron. Hence, there was ample opportunity for actinides to interact with corroding/precipitating iron, and it is possible that actinides could be found in the iron phases of the residues. If so, then reagents that attack iron phases, specifically goethite, would be needed to free these actinides from the residues.

2.3.6 Actinide-Absorbing Minerals

It has been speculated that actinides can be incorporated in a variety of uranium-silicate minerals during oxidative aqueous corrosion of UO_2 nuclear fuel (Burns 1997). It has also been shown that certain clay minerals have an affinity for actinides (Eberly 1996). Because uranium oxide has been found in K basin sludges, and because the KE basin concrete walls were produced from local aggregates, and because soil and sand have blown into the basins over the years, we cannot rule out these possibilities as being present in general sludges, and perhaps a portion of them might survive the acid-dissolution to become part of the residue.

2.3.7 Refractory Actinide Phases

Technological experience says that difficult-to-dissolve plutonium must be in the form of a refractory oxide PuO_2. However, the radionuclides in K Basin sludges came from the low-temperature aqueous corrosion of the low-burnup metallic uranium N Reactor fuel, and for that reason we would ordinarily say that very little refractory plutonium oxide should be present (Hashke 1990) (Hashke 1996). However, we shouldn't rule out the possibility entirely, given that the oxidized plutonium has had two decades' time to age towards PuO_2.

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2.3.8 Inert Minerals and Organic Detritus

Most of the residue is expected to be made up of blow sand, filter sand and soil minerals such as muscovite or other micas that are acid insoluble. The history of K basin operation includes the intrusions of birds and insects into the buildings, leaving the possibility of an occasional feather in the basin. Digestions of K basin sludges in the past have revealed small amounts of survivable oily deposits in the digestion vessels. Materials like these are not expected to have a particular affinity for actinides or cesium, and therefore will not require direct chemical attack in order to rinse actinides away from them. They may, however, be an interference by consuming the reagents in useless side reactions.

2.4 WASTE STREAMS

Each of the five sludge streams should yield residues having unique proportions of the residue types discussed above. There may be an advantage to tailoring the leaching process to each of them as they are conditioned one by one. Their compositions are described here.

2.4.1 KE Floor/Weasel Pit Sludge with Canister/Fuel Wash Sludge ≤ 250 μm

This stream is liable to contain any or all the residue types discussed above except grafoil, but it should consist mostly of inert minerals.

2.4.2 KE Canister/Fuel Wash Sludge > 250 μm, ≤ 0.25 inch

This stream may contain all residue types except grafoil, but should be more concentrated relative to the floor sludge in the refractory actinides (if they do exist here), other actinide minerals, and small pieces of Zircaloy.

2.4.3 KW Floor and Pit Sludge

Because KW canisters were sealed save for a gas-lock, and the basin walls were painted, the residue from this sludge

should contain mostly iron corrosion products, aluminum
corrosion products, inert minerals and detritus.

2.4.4 KW Canister/Fuel Wash Sludge > 500 μm, $\leq$ 0.25 inch

The residue from this sludge should be very concentrated in
the materials that contain actinides, mostly fuel corrosion
products. There will also be small amounts of iron
corrosion products from the rusted mild steel support shoes
of the fuel elements, and aluminum corrosion products from
the Mark I canister caps. The residue should be largely
free of inert minerals such as sand. Small pieces of
grafoil not removed by the resin strip process could be
present, as well as could Zircaloy pieces.

2.4.5 KW Settler Tank Sludge $\leq$ 500 μm

This sludge should come mostly from inside the KW canisters,
and therefore be very much like the wash sludge above,
except perhaps containing less Zircaloy and grafoil.

3.0 PROCESSING ALTERNATIVES IDENTIFIED

Based on the likely or possible residue types, there do exist laboratory
technologies to free the radionuclides from each of them with high
certainty. For the soil-like minerals containing iron and inert sand,
mixtures of nitric and hydrochloric acids with small amounts of
hydrofluoric acid are normally used to leach actinides away for analysis
(Catlow 1997) (Bechtold 1994) (Bock 1979). For refractory actinides
possibly embedded in siliceous or other minerals, there exist fusions
with alkali (Schulz 1991) (Makenas 1998) (Bock 1979) or other total
dissolution methods (Byers 1974) involving hydrofluorination, KF and
pyrosulfate fusions to completely dissolve minerals. For Zircaloy-2,
there is the PUREX decladding solvent, an aqueous $NH_4F-NH_4NO_3$ solution.

However, most of these technologies are viewed on a processing scale as
severe, corrosive to equipment, hazardous, time-consuming and producing
waste difficult to dispose of. It is preferred to find alternatives
that may accomplish the radionuclide dissolution by selective leaching
without resorting to complete consumption of the residues.

After sorting through many possibilities we find there are a number of
practical methods available whose effectiveness is promising, whose
effectiveness can be easily verified if necessary by simple laboratory
tests, and whose waste streams can be made acceptable to TWRS and ERDF.
However, they are not universal. Each practical alternative is proposed

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for a specific residue type, and it will have reduced effectiveness towards other waste types. The ultimate choices will depend on which waste type happens to be predominant.

These more practical alternatives for leaching the residues are summarized in Table 2, and they are individually discussed below. After a discussion of the alternatives will come a section on logical recommendations.

TABLE 2. RESIDUE LEACHING ALTERNATIVES BY RESIDUE TYPE								
Leaching Process	Residue Type							
	Refractory Actinides	Active Silicates	Other Actinide Minerals	FeO(OH)	Inert Minerals and Organics	Organic Ion Exchange Resin	Grafoil	Zircaloy
Ce(IV) 0.4M HNO_3 6M T =60-90°C t = 2 hr.	Effective Not Corrosive TWRS, ERDF compatible	Possibly Effective for PuO_2 in Silica	Possibly Effective	Not Effective	Effective as Rinse Only Organics will consume reagent.	Effective but Consumes Reagent	Effective as Rinse Only	Effective as Rinse Only Not Effective for Infused Nuclides
$K_2S_2O_8$ = 0.5M $AgNO_3$ = 0.2M HNO_3 = 3M T = ambient t = 2 hr.	Effective	Possibly Effective for PuO_2 in Silica	Possibly Effective	Only Slight Effectiveness through H_2SO_4 generation	Effective as Rinse Only	Not Effective	Effective as Rinse Only	Effective as Rinse Only Not Effective for Infused Nuclides
HNO_3 = 6M HF = 0.3M T = 60-90°C t = 2 hr.	Effective	Effective Corrosive to Equipment Requires Offgas Treatment	Effective	Not Effective	Effective as Rinse Only but Consumes Reagent	Not Effective	Effective as Rinse Only	Effective as Rinse Only Not Effective for Infused Nuclides
"CITROX": Oxalic Acid = 0.16M Citric Acid = 0.3M T = 80-100°C t = 2 hr.	Not Effective	Not Effective	Not Effective	Effective(?) May Require Waste Organic Destruction	Effective as Rinse Only	Partially Effective	Effective as Rinse Only	Effective as Rinse Only Not Effective for Infused Nuclides
HNO_3 = 5M HCl = 0.3M T = 60 - 100°C t = 2 hr.	Not Effective	Not Effective	Possibly Effective	Effective Waste Not Acceptable to TWRS without Treatment Corrosive to Equipment	Possibly Effective	Not Effective	Effective as Rinse Only	Effective as Rinse Only Not Effective for Infused Nuclides
NH_4F = 5.5M NH_4NO_3 = 0.5M T = Boiling t = 6 hr.	Not Effective	Not Effective	Not Effective	Not Effective	Effective as Rinse Only	Not Effective	Effective as Rinse Only	Effective Requires Waste F^- Neutralization, H_2, NH_3 offgas
HEDPA $NaSO_2CH_2OH$ Details not available at this time	Not Effective	Not Effective	Not Effective	Stated Effective in Secondary Reference Primary Reference and Details Unavailable	Effective as Rinse Only	Not Effective	Effective as Rinse Only	Effective as Rinse Only Not Effective for Infused Nuclides

3.1   REFRACTORY ACTINIDE DISSOLUTION BY CERATE ALTERNATIVE

3.1.1 Intended Application

This process is considered principally for dissolving refractory actinide oxide compounds that are not mineralized with other elements. Therefore it should be best when residues of that type predominate.

3.1.2 Effectiveness For Other Residue Forms

The process is also very similar to the preferred process for removing actinides from the mixed-bed organic ion exchange resin (Dodd 1998), except of course in that process the cerate is used to displace the actinide from the anion resin, not to dissolve PuO_2. If enough cerate is present, it can be expected to accomplish both tasks.

There is an implicit suggestion in the literature that cerate has some effectiveness towards actinides mineralized with silica, and also an inference that it may be effective against iron phases, because it has been found to successfully decontaminate stainless steel surfaces when applied electrochemically (Nikitina 1997a). The process is not expected to be effective towards other waste types except through its rinsing action.. It will tend to be consumed by the presence of organics, but shouldn't be consumed by other expected residue forms such as sand.

3.1.3 Suggested Process Parameters

Cerate (from $(NH_4)_2Ce(NO_3)_6$): 0.4M

HNO_3: 6M

Quantity: 2:1 Liquid to Residue by Volume

Agitate to suspend residue

Temperature: 60 - 90 °C

Time: 2 hr.

Wash with same, rinse with water

Rinses to Neutralization

3.1.4 Discussion

Cerate ion is a convenient oxidizing agent in analytical
chemistry, where one of its principal uses is in determining
organic compounds by stoichiometrically oxidizing them at
temperatures above 50°C (Kilbourn 1992) (Smith 1964). Its
use in nuclear technology is largely through its ability to
dissolve large amounts of refractory PuO_2 when applied
electrochemically (Bray 1982) (Nikitina 1997a), where
efficiencies of dissolution of large amounts of refractory
PuO_2 of 98% have been achieved. The cerate tends to
defunctionalize anion exchange resin, and, as an anionic
complex analogous to the plutonium, it displaces actinide
anions from the same resins. It appears practical to use an
excess of reagent for trace amounts of actinides instead of
using an electrochemical cell.

3.2   SILVER-CATALYZED PERSULFATE OXIDATION OF REFRACTORY ACTINIDES
ALTERNATIVE

3.2.1 Intended Application

This process has been developed for dissolving refractory
actinide oxides that are not mineralized with other elements
(Fisher 1991) and hence is best applied when this type of
residue predominates.

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3.2.2 Effectiveness For Other Residue Forms

This process may have some effectiveness towards actinides mineralized with silica, because it has been shown effective in recovering Pu from crucible slag, ashes and soils (Fisher, 1991) (Nikitina 1997a). But, it is not expected to be effective towards other waste types except through its rinsing action, though the sulfuric acid byproduct may be beneficial in dissolving iron oxide phases. The $S_2O_8^{2-}$ reagent will tend to be consumed by the presence of organics at low temperature to a lesser extent than cerate. but it shouldn't be consumed by other expected residue forms such as sand.

3.2.3 Suggested Process Parameters

Persulfate (from $K_2S_2O_8$): 0.5M

Silver (from $AgNO_3$): 0.2M

HNO_3: 3M

Quantity: 2:1 Liquid to Residue by Volume

Agitate to suspend residue

Temperature: 25 - 40 °C

Time: 2 hr.

Wash with Same, Rinse with Water;

Rinses to Neutralization

3.2.4 Discussion

Silver-catalyzed oxidation is normally applied electrochemically in nuclear technology to dissolve refractory PuO_2 (Nikitina 1997) (Nikitina 1997a). It has also been studied electrochemically as an organic destruction agent (Smith 1997). It is a faster method at low temperatures than cerium (IV)-mediated electrochemical oxidation. It has been shown at Hanford that the electrochemical cell can be replaced by excess persulfate (Fisher 1991) to effectively dissolve large amounts of PuO_2, or to recover PuO_2 from scrap, soil or ash. Efficiencies of 89 to 100 percent have been seen for single contacts. A multiple contact experiment against fine soil has showm *ca.*

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80% recovery per contact. This has also been demonstrated electrochemically in France (Nikitina 1997a), where the high silica content of the ash lends support to the idea that this process may help remove Pu from within a silica matrix. At higher temperatures, the reagents tend to be consumed by the oxidation of water. Hot, silver-catalyzed persulfate oxidation is one of two methods used at the 222-S labs for determination of total organic carbon (Schroeder 1997), however as an analytical method it gives poor recoveries for non-functionalized organics, and it has been observed to be ineffective at dissolving ion exchange resin beads.

3.3   REFRACTORY ACTINIDES, SILICA-MINERALIZED ACTINIDES AND CESIUM-MINERALIZED ACTINIDES DISSOLUTION WITH NITRIC-HYDROFLUORIC ACIDS ALTERNATIVE

3.3.1 Intended Application

This process is intended primarily for actinides and cesium contained within siliceous minerals, where the hydrofluoric acid can volatize the silicate or aluminosilicate minerals, leaving non-volatile radionuclides in solution. Because of the volatilization, the SiF_4 offgas will have to be sparged through a dilute alkali solution and defluoridized by sequestration of fluoride with aluminum nitrate. The reagents are corrosive to equipment, and neutralization of the rinses will also have to include defluoridization with aluminum nitrate. A large amount of inert siliceous material in the residue may consume the HF without producing the desired decontamination, thus requiring replenishment of HF during the process.

3.3.2 Effectiveness for Other Residue Forms

The process is also traditionally used for dissolving refractory plutonium oxide (Nikitina 1997). In fact, a similar process is the recommended process for the AIDA/MOX 1 program in France (Bourges 1997). There is chemical inference that the process might work to free nuclides from iron minerals based on successful use of similar formulations as reactor piping decontamination agents (Schaller 1984) (Mertz 1996).

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3.3.3 Suggested Process Parameters

HNO_3: 6M

HF: 0.3M

Quantity: 2:1 Liquid:Residue by Volume

Agitate to Suspend Residue

Temperature: 60 - 90 °C

Time: 2 hr.

Sparge offgas through dilute alkali

Wash with Same

Rinse with Water

Rinses Combined with Sparge, then to Aluminum Fluoride
Precipitation and Neutralization

3.3.4 Discussion

This process is based on a variety of traditional methods
for dissolving refractory plutonium dioxide (Nikitina 1997),
or for liberating radionuclides such as ^{137}Cs from siliceous
materials. The silica is destroyed and silicon tends to
volatize as SiF_4, while the actinides remain non-volatile.

The reagent concentration choices suggested here are a
compromise between effectiveness and corrosiveness towards
the equipment. Traditional applications have varied widely
in compositions. For example, to dissolve plutonium oxide
or recover plutonium from scrap at PFP, the nitric acid
concentration was normally 12 M and the temperature was at
the solution boiling point. Efficiencies of dissolution
vary, depending on the pretreatment of the substrate and the
concentration of reagents. In a most difficult case,
without other additives such as H_2SO_4, AgO or Ce(IV), a
boiling 12M HNO_3, 0.1MHF mixture could achieve 50-54.2%
dissolution of >1500°C-calcined PuO_2. If large amounts of
inert siliceous materials are present in the residue, it may
be necessary to replenish HF during the leaching process.

Nitric - hydrofluoric acids have been used as reactor piping
or stainless steel decontamination reagents in the
concentration range 0.8M nitric acid, 0.25M hydrofluoric

acid (Schaller 1984). However, it was noted that radionuclides in iron oxide deposits are dislodged and made available for filtration to a greater extent than they are actually dissolved by these reagents. Here, we desire the radionuclides to be dissolved so that they may pass through the filter used to isolate the clean residue. Therefore the method may not be sufficiently effective against relatively large amounts of iron mineral deposits.

There is operational experience at PFP with the precipitation of silicates by sparging SiF_4 through dilute alkali that shows the precipitation to be slow, and at times the precipitate can subsequently deposit on the equipment, clog small passages and otherwise be an annoyance (Barney 1998). The sparge should be arranged to minimize that annoyance.

3.4 IRON DISSOLUTION BY CITROX ALTERNATIVE

3.4.1 Intended Application

This assumes that dissolution of iron minerals will free the radionuclides from the residue. For a less corrosive form of iron oxide dissolution than the use of HCl, this is a commercially available reactor decontamination technology that is compatible with TWRS acceptance -- a variation of the CITROX process (Serne 1996). Citric and oxalic acids totaling 6% of both reagents are stirred with the residue. The oxalic acid dissolves rust while the citric acid helps keep the iron in solution. The process may not be completely effective if refractory iron minerals such as goethite prove resistant to it. The neutralized rinses may be classified as complexant waste by TWRS acceptance criteria, which may complicate disposal to TWRS without an organic destruction treatment process.

3.4.2 Effectiveness for Other Residue Forms

The oxalic acid will also dissolve aluminum hydroxide. At the concentrations suggested, any liberated Pu(IV) should be complexed and held in solution by the oxalate, and not absorb onto anion exchange resin beads. The process is not expected to be effective against any other of the significant residue forms except as a rinse.

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3.4.3 Suggested Process Parameters

Oxalic Acid: 0.3M

Citric Acid: 0.16M

Quantity: 2:1 Liquid:Residue by Volume

Agitate to Suspend Residue

Temperature: 85 - 100°C

Time: 2 hr.

Wash with Same

Rinse with Water

Rinses to Neutralization

3.4.4 Discussion

This would be the least corrosive towards process equipment
of the iron-attacking processes considered. In normal
commercial application, CITROX is applied in very dilute
solution, being continuously circulated through a strong-
acid cation resin in the acid form to capture dissolved
metal and regenerate the reagent. Here, we would substitute
an excess of stirred reagents for the resin, and use enough
solution to cover and suspend the residue. The efficiency
of the process for iron-dissolution is hard to evaluate,
because dislodged minerals available for filtration also
result in decontamination. However, it can be said that the
dilute reagents used in commercial practice transport
several times their molar quantities of metals to the ion
exchange resins. After filtering, washing and water rinsing
the residue, the rinses are collected for neutralization.
When combined with the nitric acid dissolution rinse, the
result may become a complexant waste as defined by TWRS
($\geq$ 10 g/L TOC when evaporated to first precipitation). AW-
105 is not currently classified as a complexant concentrate
(CC) waste tank, and therefore would not accept CC waste.
If necessary to meet TWRS acceptance, the citric and oxalic
acids could be destroyed by hydrogen peroxide in strong
nitric acid solution before neutralization. The CITROX
process has additional appeal because there is local
commercial support for both this process (Serne 1996)
(Newton 1998) and support for general process application to
Hanford fuel storage basin desludging (Schneidmiller 1998).

However, there is some question whether the goethite will dissolve quickly enough, to be answered by simple lab testing for practical process operation.

3.5 DISSOLUTION OF IRON MINERALS AND TRACE AMOUNTS OF NON-REFRACTORY ACTINIDES ALTERNATIVE

3.5.1 Intended Application

This treatment assumes the radionuclides are locked in iron-based minerals. This method provides a large amount of acidic chloride to effectively complex iron. Major disadvantages are that this reagent is extremely corrosive for many metals of construction, and the chloride concentration would be unacceptable to TWRS without 30-fold dilution or a chloride-removal process.

3.5.2 Effectiveness for Other Residue Forms

This is not expected to be effective against any other of the significant residue forms except as a rinse.

3.5.3 Suggested Process Parameters

HNO_3: 5M

HCl: 0.3M

Quantity: 2:1 Liquid:Residue by Volume

Agitate to Suspend Residue

Temperature: 60°C – boiling

Time: 2 hr.

Wash with Same

Rinse with Water

Rinses to Chloride Removal and Neutralization

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3.5.4 Discussion

For iron minerals, reagents utilizing multi-molar concentrations of HCl are expected to provide analytically complete dissolution efficiency (Bock 1979). The method for dissolving non-refractory, loosely adherent radionuclides from KE basin sludge samples containing also iron-based minerals consisted of boiling 5M HNO_3 - 5M HCl - 0.03M HF (Bechtold 1994). Efficiencies varied among samples treated. For plutonium, dissolution efficiencies were from >56% to >99%, while for iron, they were >99.4%. As used to dissolve similar actinides from soil samples in radiochemical laboratory practice, the nitric and hydrochloric acid concentrations are mixed as concentrated reagents (Catlow 1997). Those mixtures and the suggested process all approximate aqua regia, and therefore can be extremely corrosive even to noble metals. Therefore the concentrations actually suggested are a compromise between effectiveness against the small amounts of iron mineral to be dissolved and corrosiveness towards the equipment. The need for chloride removal from the rinses may be met by a cerate-mediated electrochemical oxidation of chloride to Cl_2 gas (Koehly 1993).

Overall, this alternative is not attractive, because the extent of compromise away from the usual practice for dissolving iron is great, while the corrosiveness is still problematical.

3.6 DISSOLUTION OF ZIRCALOY PIECES BY ZIRFLEX CLADDING DISSOLVER SOLUTION ALTERNATIVE

3.6.1 Intended Application

If the residue includes Zircaloy particles or pieces which themselves contain infused radionuclides that drive the overall residue above ERDF acceptance limits, then the PUREX cladding dissolution process can dissolve the Zircaloy and free the radionuclides. The Zirflex process normally uses at least 0.4 M nitrate ion or 90 mg/L ferric ion to minimize hydrogen evolution, and perhaps an air or steam sparge to remove generated ammonia and maintain desired pH. Otherwise it is straightforward (Westinghouse 1989) and shouldn't require verification testing. The rinses may be defluoridized with aluminum nitrate before neutralization.

3.6.2 Effectiveness for Other Residue Forms

This process is not expected to be effective against any other of the significant residue waste forms except as a rinse.

3.6.3 Suggested Process Parameters

NH_4F: 5.3M

NH_4NO_3: 0.5M

Quantity: 2:1 Liquid:Solid by Volume or sufficient liquid to assure 6:1 F^-:Zr, whichever is greater

Agitate to Suspend Solids

Steam Sparge/Offgas Hydrogen and Ammonia

Time: 6 hr.

Temperature: Boiling

Rinse with boiling water before cooling

Rinses to Defluoridization and Neutralization

3.6.4 Discussion

With the depletion of ammonium ion as volatile ammonia while reaction progresses, a desirable free fluoride ion concentration of 6M provides an 80 mil/hr dissolution rate, which over 6 hours should dissolve the cladding pieces and their infused actinides with 100% efficiency. The assurance of 6:1 fluoride:zirconium and the rinse step are meant to prevent precipitation of zirconium before filtration. The hydrogen and ammonia offgases can be an explosion hazard if they are not safely handled Verification testing is probably unnecessary for such a well-established treatment. Only production testing of fluoride addition may be necessary to counter any possible fluoride consumption by sand or soil in the residue.

3.7 DISSOLUTION OF GOETHITE BY HEDPA AND REDUCING AGENT ALTERNATIVE

3.7.1 Intended Application

This process is explicitly called out in a secondary reference (Schulz 1991) as being effective in dissolving goethite. The HEDPA in the form of an alkylphosphonic acid mixture is commercially available under the trade name IONQUEST 201. Together with the reducing agent sodium formaldehydesulfoxylate it dissolves goethite, and is later easily destroyed, leaving phosphate, sulfate and carbon dioxide. The sulfate and phosphate are not desirable in vitrification feeds, though small quantities may not be unacceptably offensive.

3.7.2 Effectiveness for Other Residue Forms

The HEDPA process is not expected to be effective against any other of the significant residue forms.

3.7.3 Suggested Process Steps

The details of the process are not known at this time.

3.7.4 Discussion

The attraction of this process is the claim of its direct effectiveness against goethite, an identified mineral in the canister sludge residuals that themselves are above TRU levels. Also attractive is the ability to destroy the reagents by digestion in HNO_3 or $HNO_3 - H_2O_2$ solutions. This destruction could be accomplished by adding the rinses to the nitric acid dissolution rinse and digesting before neutralization.

This process will not be recommended, however, because exact details have not been found. It will remain a possibility to be investigated further only if it is determined that complete dissolution of goethite is necessary and is unachievable by any other recommended process.

4.0 PROCESS SELECTION and IMPLEMENTATION

4.1 Speciation

The first matter in selecting the processes will be to speciate the actinides and cesium in the residues, in order to justify or rule out the need for any of the proposed process steps.

4.2 Testing and Optimization

The proposed process conditions should be tested for efficacy, with the exception of the Zirflex process, and the suggested process parameters optimized. There is also a little flexibility among the processes for combining them. For example, there may be an acceptable combination of HNO_3 - HCl - HF not considered above that might prove to be optimally effective as a single process. However, the flexibility for combining alternatives is limited in general. Consideration should be made to selecting lined vessels and equipment for the processes to eliminate corrosion problems, especially for chloride or fluoride-bearing reagents.

4.3 Prioritization

The following order of priorities is followed for selecting a process or processes:

4.3.1 Known effectiveness in releasing the TRU content from the most likely residue type.

4.3.2 Compatibility with TWRS acceptance

4.3.3 Compatibility with ERDF acceptance

4.3.4 Operational Complexity:

      4.3.4.1   Corrosivity requiring special equipment

      4.3.4.2   Requirements for secondary waste management

5.0 PROCESS RECOMMENDATIONS

The bases for recommending leaching processes are likely to change rapidly as more is learned about the nature of the residuals from canister sludge dissolution tests. At this time we expect the largest volume of sludge will contain a major proportion of inert minerals such as sand and other soil components, and a smaller proportion of fuel

corrosion products. On that basis, the following order of choices for testing is presented:

5.1 The <u>Cerate process</u> is the first choice due to its broad effectiveness for actinides in refractory oxide form, and in organic ion exchange resin beads, as well as less certain effectiveness for actinides in active siliceous materials. It should not be consumed uselessly by inert minerals except organic detritus. The process will not require complex secondary waste treatment.

5.2 The <u>Persulfate-Silver process</u> is the second choice due to the same general effectiveness as the cerate process except for its lack of effectiveness against the ion exchange resin beads. It also will not be consumed uselessly by inert minerals or organic detritus. Neutralization of rinses may need to be preceded by a reductant step to destroy excess persulfate.

5.3 The <u>Nitric-Hydrofluoric Acid process</u> is the third choice. It is traditionally effective against refractory actinides and the major portion of problematical residues, i.e., siliceous materials. It also has been successfully used to decontaminate ferrous metals. The HF may need to be replenished during processing due to consumption by inert minerals, and offgas treatment is required. It is also more corrosive than the previous choices.

5.4 A variation of the <u>CITROX</u> process is next suggested, if the other processes don't work, and/or the actinides are found to be residing in iron-bearing or aluminum-bearing minerals. It is the least corrosive against ferrous metals of the processes used to dissolve iron minerals.

5.5 The <u>Nitric-Hydrochloric</u> Acid process is the last general process to recommend, because of its limited generality of effectiveness, the extent of compromise to reduce its extreme corrosivity, and the need to somehow remove the waste chloride.

5.6 The <u>Zirflex</u> process is recommended only for the special case when Zircaloy particles are found to be the carrier of TRU level actinides in the residue. It requires some sparging and concentration control during processing, as well as flammable offgas treatment.

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