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RADIOIODINE CONTROL DURING
REPROCESSING OF SHORT-COOLED
IRRADIATED NEPTUNIUM-237
ALUMINUM ALLOY
TARGET ELEMENTS
(A Preliminary Study)

W. O. Greenhalgh

April 1967

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Section 1. Introduction

A plutonium-238 production test is currently being carried out at Hanford. Target elements of neptunium-237 alloyed with aluminum are irradiated to produce the plutonium product. One aspect of the test is to demonstrate that short-cooled target elements can be chemically processed and the recovered neptunium recycled to the reactor within a specified time period. An important part of the chemical processing is control of short-lived radioisotopes, particularly radioiodine. This study was undertaken for the purpose of defining the extent of the radioiodine problem and developing methods for controlling or removing radioiodine where necessary.

Section 2. Summary and Conclusions

The chemistry of tracer radioiodine was studied in the following process steps: dissolution of the Np-Al target in nitric acid with mercury catalyst and subsequent feed stream manipulation and storage; extraction of neptunium and plutonium by 30% TBP-NPH (normal paraffin hydrocarbon); and absorption of neptunium and plutonium by Dowex 1 anion resin.

The following results were obtained:

1. Two per cent or less of the total radioiodine was evolved during dissolution tests using an aluminum metal stand-in.
2. Almost quantitative extraction of the feed stream radioiodine was exhibited by a 30% TBP-NPH batch contact.
3. Greater than 90% of the feed stream radioiodine was absorbed by anion resin during one short column pass.

Radioiodine accumulation and concentration during solvent extraction and resin absorption is hazardous and costly. Consequently, methods for removing radioiodine were investigated. Two methods (precipitation as mercuric iodide and extraction by phenyl mercuric acetate or phenyl mercuric nitrate in benzene) were found effective in removing feed stream radioiodine while only slightly affecting the neptunium content.

Section 3. Experimental Methods

The simulated target dissolutions were carried out in a half-liter Pyrex dissolver flask connected with a condenser-cold trap or a condenser-scrubber train.

The synthetic dissolver solutions were prepared by reacting iodine-131 spiked nitric acid-mercuric nitrate solution with aluminum metal. Baker & Adamson reagent or C. P. grade chemicals were made up to the following composition in a 100 ml liquid volume before dissolution:

- 0.15 mole aluminum metal
- 0.001 or 0.0005 mole mercuric nitrate
- 0.80 mole (for anion exchange feed) or
- 0.60 mole nitric acid (for solvent extraction feed)
- 1×10^{-6} mole potassium iodide
- $\sim 1 \times 10^7$ cpm iodine-131

The quantity of radioiodine evolved during a number of aluminum metal dissolutions was determined from the difference in iodine-131 concentrations before and after dissolution and by the amount of radioiodine that could be detected in the dissolver off-gas. The quantity of radioiodine removed by precipitation or ion exchange was determined by the difference in the feed stream iodine-131 concentration before and after the experiment. The quantity of radioiodine removed by solvent extraction was determined by the distribution ratio of iodine-131 between the two immiscible layers.

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The calculations in Section 5 are based on the conclusion that complex formation occurs between radioiodine and mercury(II) ion. This conclusion was reached after analysis of the following observations:

1. Addition of a small but observable quantity of elemental iodine ($> 10^{-5}$ moles) to a dissolver solution gave no colored solution; a colored solution would be expected if most of the iodine remained in the elemental form.
2. Addition of 0.001 m moles of silver ion per ml 0.01 M mercuric nitrate dissolver solution containing 0.001 F iodide ion gave neither a silver iodide nor silver iodate precipitate.
3. X-ray diffraction analysis of a red precipitate formed by the addition of potassium iodide to a dissolver solution showed the solid to be only mercuric iodide.
4. The mercuric iodide precipitate in No. 3 was completely redissolved by adding it to a dilute mercuric nitrate solution.

It was therefore concluded that complex formation had occurred and that so long as excess mercuric nitrate is present, radioiodine evolution from the aqueous system will not be a problem.

The major radioiodine control problem occurs during isolation and purification of the neptunium and plutonium products. Radioiodine accumulates and concentrates in either 30% TBP-NPH or Dowex 1 anion resin as shown in Tables III and V. Radioiodine extraction or absorption from a typical feed by TBP-NPH or Dowex resin is greater than 90%, while subsequent removal by washing is only 10 to 15% effective. To eliminate this radiation hazard, methods to remove the contamination from the feed stream prior to solvent extraction or anion exchange were devised.

Data on feed stream radioiodine removal are shown in Tables II and IV. Table II contains data on radioiodine removal by mercuric iodide precipitation carried out by adding a stoichiometric quantity of non-radioactive potassium iodide. Radioiodine removal by this method should be 98% effective for a 0.005 F precipitate

according to mercuric iodide solubility values obtained from Seidel⁽¹⁾. The removal data show that this is indeed possible if sufficient precipitation time is used; for reprocessing purposes 30 minutes should be adequate if stirring of the mixture is possible. Table II data also show that multiple precipitations are effective, and that neptunium losses should be about 0.3% per strike.

Table IV contains data on a solvent extraction method for removing feed stream radioiodine. This method, developed for chloride removal by Schulz⁽²⁾, uses a benzene solution of phenyl mercuric nitrate or phenyl mercuric acetate for the extractant. Batch contacts of the 0.005 M benzene extractant with synthetic feed consistently showed removal of 75% of the radioiodine. The corresponding neptunium loss during contact is about 0.3%.

Although two methods for radioiodine removal have been demonstrated, some contamination of anion resin and extraction solvent will undoubtedly occur. A method for removing radioiodine contamination from 30% TBP-NPH was developed. Cleanup of the TBP solvent is possible by passing it through a column of silver nitrate crystals. Table V shows that at least 70% of the contamination can be removed in one pass and with multiple passes, a decontamination factor of 2000 or greater is possible. One drawback, however, may be loss of silver nitrate due to solubility; measurement of the silver nitrate solubility in Hanford Purex solvent gave 1.5 g/l.

Section 5. Calculation of Iodine Containing Species in a HNO_3 - 0.005 M $\text{Hg}(\text{NO}_3)_2$ Dissolver Solution

In the following section, approximate expressions for the concentration of the species $\text{I}_2(\text{aq})$, I^- , HgI^+ , and $\text{HgI}_2(\text{aq})$ in a HNO_3 - 0.005 M $\text{Hg}(\text{NO}_3)_2$ dissolver

- (1) Seidel, Atherton, revised by Linke, William F., "Solubilities Inorganic and Metal-Organic Compounds", Vol. 1, 4th ed. O. Van Nostrand Company, Inc., N. Y., N. Y., 1958, p. 1232.
- (2) Schulz, W. W., "Removal of Chloride from Aqueous Solutions", U. S. Patent 2,898,203.

solution are derived and the values calculated. The expressions are derived from mercuric iodide equilibrium formation constants published by Hansen, Izatt, and Christensen⁽³⁾, and half-cell potentials compiled by Latimer⁽⁴⁾. The formation constants (K_1 and K_2) for HgI^+ and $\text{HgI}_2(\text{aq})$ from Hansen and coworkers are:

Since K_1 and K_2 are thermodynamic constants, an expression for the species in activity terms (A) or in concentration terms (C) can be written

$$(1) \quad \frac{A_{\text{HgI}^+}}{A_{\text{Hg}^{++}} A_{\text{I}^-}} = 7.3 \times 10^{12} \quad \text{or} \quad 7.3 \times 10^{12} = \frac{C_{\text{HgI}^+} \gamma_{\text{HgI}^+}}{(C_{\text{Hg}^{++}} \gamma_{\text{Hg}^{++}})(C_{\text{I}^-} \gamma_{\text{I}^-})}$$

where " γ " represents the activity coefficient of the ions. The determination of the activity coefficients for the various species in a strong nitric acid-aluminum nitrate solution would be difficult and probably unnecessary for the purposes of these calculations. Use of concentrations as approximate activities should suffice. Accordingly, an equilibrium expression in terms of concentration is written

$$(2) \quad \frac{C_{\text{HgI}^+}}{(C_{\text{Hg}^{++}})(C_{\text{I}^-})} \cong 7.3 \times 10^{12}$$

or in terms of the iodide concentration

$$(3) \quad C_{\text{I}^-} \cong \frac{C_{\text{HgI}^+}}{(7.3 \times 10^{12})(C_{\text{Hg}^{++}})}$$

By the same type of reasoning the approximate equilibrium expression for the concentration of $\text{HgI}_2(\text{aq})$ can be written

$$(4) \quad C_{\text{HgI}_2(\text{aq})} \cong (8.9 \times 10^{10})(C_{\text{HgI}^+})(C_{\text{I}^-})$$

Now using half-cell reactions one can write the probable reaction for the oxidation of iodide ion to elemental iodine as shown where " E° " is the standard

(3) Hansen, Lee D., Izatt, Reed M., and Christensen, James J., Inorg. Chem., 2, 1243 (1963).

(4) Latimer, W. M., "Oxidation Potentials", 2nd ed., Prentice-Hall Inc., Englewood Cliffs, N. J., 1952.

potential.

Next using the thermodynamic expression relating standard cell potential to standard free energy and hence to the thermodynamic equilibrium constant, one can calculate the constant (K).

$$- n F E^\circ = \Delta F^\circ = -RT \ln K$$

$$(6) \quad \log K = \frac{n F E^\circ}{2.3 RT}$$

at room temperature:

$$(7) \quad \log K = \frac{(2 \text{ eq/mole})(23.060 \text{ cal/volt eq})(+0.40 \text{ volts})}{(2.3)(1.986 \text{ cal/mole deg})(298 \text{ deg})} = 13.6$$

$$(8) \quad K = 4 \times 10^{13}$$

With the equilibrium constant "K" and by the same reasoning used before, an approximate expression for the concentration of elemental iodine can be written

$$(9) \quad \frac{(A_{\text{I}_{2(\text{aq})}})(A_{\text{HNO}_2})(A_{\text{H}_2\text{O}})}{(A_{\text{H}^+})^3(A_{\text{NO}_3^-})(A_{\text{I}^-})^2} = K$$

$$(10) \quad \frac{(C_{\text{I}_{2(\text{aq})}})(C_{\text{HNO}_2})}{(C_{\text{H}^+})^3(C_{\text{NO}_3^-})(C_{\text{I}^-})^2} \approx 4 \times 10^{13};$$

$$(11) \quad C_{\text{I}_{2(\text{aq})}} \approx \frac{(4 \times 10^{13})(C_{\text{H}^+})^3(C_{\text{NO}_3^-})(C_{\text{I}^-})^2}{C_{\text{HNO}_2}}$$

or substituting the expression of equation 3 for C_{I^-}

$$(12) \quad C_{\text{I}_{2(\text{aq})}} \approx \frac{(4 \times 10^{13})(C_{\text{H}^+})^3(C_{\text{NO}_3^-})(C_{\text{HgI}^+})^2}{(C_{\text{HNO}_2})(C_{\text{Hg}^{++}})^2(7.3 \times 10^{12})^2}$$

Expressions for the species I^- , I_2 , and $HgI_2(aq)$ have been derived, but the concentration of HgI^+ is needed before calculations can be made. The anticipated formal concentration of iodine in a typical dissolver solution is about 1×10^{-5} M, which should be approximately the concentration of the HgI^+ species since the mercury(II) ion content is 500 times greater than the iodine. Using this estimated value for HgI^+ concentration, a value of $\geq 1 \times 10^{-3}$ M for C_{HNO_2} (expected by the radiation degradation of nitric acid), a value of 3M for C_H^+ , and 5M for $C_{NO_3^-}$ the concentrations of I^- , $I_2(aq)$, and $HgI_2(aq)$ are calculated.

$$(13) \quad C_{I_2(aq)} \cong \frac{4 \times 10^{13} (3)^3 (5) (1 \times 10^{-5})^2}{(1 \times 10^{-3}) (5 \times 10^{-3})^2 (7.3 \times 10^{12})^2} = 3 \times 10^{-13} \underline{M}$$

$$(14) \quad C_{I^-} \cong \frac{1 \times 10^{-5}}{(5 \times 10^{-3}) (7 \times 10^{12})} \cong 3 \times 10^{-16} \underline{M}$$

$$(15) \quad C_{HgI_2(aq)} \cong (8.9 \times 10^{10}) (1 \times 10^{-5}) (3 \times 10^{-16}) \cong 3 \times 10^{-10} \underline{M}$$

$$(16) \quad C_{HgI^+} \cong 1 \times 10^{-5} - C_{I_2(aq)} - C_{I^-} - C_{HgI_2(aq)} \cong 1 \times 10^{-5} \underline{M}$$

The conclusion from these calculations is that radioiodine evolution during normal feed stream manipulations or storage would be nil due to the small concentration of elemental iodine. Iodine present as iodate ion is not considered here but calculations indicate that its concentration would be smaller still than the above species.

Dissolution Data

Table I		I^{131} Activity cpm/4 ml	% I^{131} Evolved
No.	Sample Description		
1	Initial 0.01M $Hg(NO_3)_2$ makeup sol'n	676,736	
	Solution after Al dissolution	662,072	2 by difference
	Off-gas dry ice trap	3,750	0.6 by trapping I^{131} in off-gas
2	Initial 0.005M $Hg(NO_3)_2$ makeup sol'n	25,392	
	Solution after Al dissolution	24,825	2
3	Initial 0.005M $Hg(NO_3)_2$ makeup sol'n	58,610	
	Solution after Al dissolution	58,768	0
4	Initial 0.005M $Hg(NO_3)_2$ makeup sol'n	215,450	
	Solution after Al dissolution	216,795	0 by difference
	100 ml 2.5M NaOH scrub sol'n	39	0.5 by trapping I^{131} in off-gas

Precipitation Data

Table II		I^{131} Activity cpm/4 ml	% I^{131} Removed
No.	Sample Description		
1	a. 0.01M $Hg(NO_3)_2$ Dissolver Sol'n	238,430	
	b. Filtrate after HgI_2 precipitation	4,668	98
2	a. 0.01M $Hg(NO_3)_2$ Dissolver Sol'n	9,301	
	b. Filtrate after HgI_2 precipitation	327	97
	c. Filtrate after HgI_2 precipitation and iodine titration	92	99
	d. Filtrate after HgI_2 and 0.001M AgI precipitation	75	99.2
3	a. 0.005M $Hg(NO_3)_2$ Dissolver Sol'n	59,967	
	b. Filtrate after HgI_2 precipitation from a 0.3M N_2H_4 solution	1,259	98
	c. Filtrate of HgI_2 and 0.001M HgI precipitation	1,176	98
4	a. 0.005M $Hg(NO_3)_2$ Dissolver sol'n	444,852	
	b. 3 rapid HgI_2 precipitation	701	99.25
	c. 1 equilibrium HgI_2 precipitation	53	99.987 Total or 92% removal of iodine from 4b.
5	Made 0.005M $Hg(NO_3)_2$ dissolver solution 0.001M in KI and 0.001M in $AgNO_3$ - no precipitation occurred.		
		Np^{239} Activity	% Np^{239} Removed
6	a. 0.005M $Hg(NO_3)_2$ Dissolver sol'n with Np^{239} Tracer	8,484	
	b. Filtrate after HgI_2 precipitation	8,454	0.3

Solvent Extraction Data

Table III

No.	Sample Description	Activity I^{131} cpm/4 ml	% I^{131} in Extractant	Distribution Ratio (E_d)
30% TBP-NPH Extraction of Radioiodine				
1.	0.01M $(HgNO_3)_2$ dissolver sol'n organic (30% TBP-NPH)	21,327 1,540,298	99	
2.	0.01M $Hg(NO_3)_2$ dissolver sol'n organic (30% TBP-NPH)	25,084 639,445	96	
3.	0.01M $Hg(NO_3)_2$ dissolver sol'n organic (30% TBP-NPH)	26,073 777,763	97	
4.	0.01M $Hg(NO_3)_2$ dissolver sol'n organic (30% TBP-NPH)	373 8,936	96	
Avg.			97 ± 1	33
5.	0.005M $Hg(NO_3)_2$ dissolver sol'n organic (30% TBP-NPH)	2,764 52,924	95	
6.	0.1M NaOH strip I^{131} in organic (30% TBP-NPH)	12,059 1,398,776	99	117
7.	distilled water strip I^{131} in organic (30% TBP-NPH)	52,141 1,348,790	96	26
8.	10% Na_2CO_3 strip I^{131} in organic (30% TBP-NPH)	61,906 316,840	81	5
9.	0.01M $Hg(NO_3)_2$ strip I^{131} in organic (30% TBP-NPH)	89,537 792,525	89	9
10.	0.01M $AgNO_3$ strip I^{131} in organic (30% TBP-NPH)	77,465 696,935	89	9
11.	0.01M $Hg(NO_3)_2$ dissolver sol'n organic (100% NPH)	1,081,119 30,135	3	0.03

Solvent Extraction Data

Table IV			Phenylmercuric Acetate Conc. in Benzene	Phenylmercuric Nitrate Conc. in Benzene	Activity I ¹³¹ cpm/4 ml	% I ¹³¹ Extracted	Distri- bution Ratio (E _a)
No.	Sample Description						
Phenylmercuric nitrate and phenylmercuric acetate benzene extraction of radioiodine from 0.005M Hg(NO ₃) ₂ dissolver solutions.							
13.	aqueous with 5% acetic acid	-----	-----	-----	82,488		
	organic	0.005M	-----	-----	114,399	58	
14.	aqueous	-----	-----	-----	36,808		
	organic	0.005M	-----	-----	123,317	77	
15.	aqueous	-----	-----	-----	14,035		
	organic	0.005M	-----	-----	44,803	76	
16.	aqueous	-----	-----	-----	13,493		
	organic	0.005M	-----	-----	39,232	74	
Avg. omitting #13		0.005M				76 ± 1	3
17.	aqueous	-----	-----	-----	10,831		
	organic	saturated (<u>< 0.01M</u>)	-----	-----	43,355	80	
18.	aqueous	-----	-----	-----	9,982		
	organic	saturated	-----	-----	43,577	81	
Avg.						81	4
19.	aqueous	-----	-----	-----	55,817		
	organic	-----	0.005M	-----	141,012	72	
20.	aqueous	-----	-----	-----	44,578		
	organic	-----	0.005M	-----	145,949	77	
21.	aqueous	-----	-----	-----	14,314		
	organic	-----	0.005M	-----	42,314	74	
Avg.		-----	0.005M	-----		74 ± 2	3
22.	aqueous	-----	-----	-----	14,207		
	organic	-----	saturated (<u>< 0.01M</u>)	-----	43,768	75	
23.	aqueous	-----	-----	-----	10,518		
	organic	-----	saturated (<u>< 0.01M</u>)	-----	42,654	77	
Avg.						76	3
24.	aqueous	-----	-----	-----	3,863		
	1st organic contact	-----	0.005M	-----	41,131	75	
	2nd organic contact	-----	0.005M	-----	9,704	72	of I ¹³¹ remains
25.	aqueous	-----	-----	-----	4,417		of 1st contact
	1st organic contact	-----	0.005M	-----	35,712	71	
	2nd organic contact	-----	0.005M	-----	10,606	71	of I ¹³¹ remains
26.	aqueous	-----	-----	-----	701,295		of 1st contact
	organic	0	0	-----	115,000	14	
					Activity Np ²³⁹	% Np ²³⁹ Extracted	
27.	aqueous with Np ²³⁹ tracer				707,086		
	organic		.005M		2,350	0.3	

Miscellaneous Data

Table V

No.	Sample Description	I^{131}	
		Activity cpm/4 ml	% I^{131} Removed
1.	0.005M dissolver solution	91,912	
2.*	Dissolver solution #1 after pass through Dowex 1-2.8x resin	4,680	93
3.	1 vol 6M HNO_3 wash of resin column		15
4.	1 vol 0.3M HNO_3 wash of resin column		14
5.	a. 4 ml 30% TBP-NPH containing I^{131}	49,814	
	b.* Same solvent after 1 pass through column of silver nitrate	13,093	74
6.	a. 4 ml 30% TBP-NPH with I^{131}	410,966	
	b. Same solvent after equilibrium with $AgNO_3$ crystals	143	99.965

*Note: Column used is 9 cm high by 8 mm ID and the flow was ~ 3 ml/min

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