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January 1992 - December 1992

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Rockwell International
Rocketdyne Division

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**Technical Progress Report
Private Sector Initiatives Between
the United States and Japan**

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Rockwell International

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ACRONYMS

CRIEPI	Central Research Institute for the Electric Power Industry
DOE	Department of Energy
FY	fiscal year
HEPA	high-efficiency particulate air
ICP	inductive coupled plasma
JAPC	Japan Atomic Power Company
KHI	Kawasaki Heavy Industries
LMR	liquid metal reactor
MURR	Missouri University Research Reactor
P/T	partitioning/transmutation
SSFL	Santa Susana Field Laboratory
TRU	transuranic

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INTRODUCTION

The Private Sector Initiatives Program was started in mid-1987 under the cognizance of the Office of the Deputy Assistant Secretary for Reactor Systems, Development Technology (NE-50). The purpose of this program is to utilize industrial ties between U.S. Firms and their Japanese counterparts, to enhance U.S. access to advanced Japanese reactor programs, and ultimately, to provide a basis for U.S. vendor participation in advanced Japanese liquid metal reactor (LMR) designs. The program includes participation from the three U.S. vendors: General Electric Company, Rockwell International, and Westinghouse.

Rockwell activities on the Private Sector Initiatives Program were implemented through an existing relationship with Kawasaki Heavy Industries (KHI). Rockwell and KHI reviewed their respective LMR research needs to identify and screen suitable areas for collaborative efforts. These areas were determined to be of mutual benefit and were approved by the Department of Energy (DOE) and Japan Atomic Power Company (JAPC). Rockwell and KHI performed their respective portions of tasks within these selected areas and exchanged results. Agreed-upon exchange areas for the Rockwell/KHI private sector initiatives collaborative efforts are

- Plant-type evaluations
- Shrouded inducer pump
- Inherent safe core
- Passive shutdown heat removal system
- Booster tube steam generator
- Advanced piping
- Transuranic (TRU) partitioning
- Dry sodium removal.

Results of prior year activities are reported in References 1 through 5.

SUMMARY

This annual report for calendar year 1992 describes the efforts performed under the Private Sector Initiatives contract. The report also describes those efforts that have continued with private funding after being initiated under this contract.

One privately funded, continuing activity is described in Task 2. The task is the development of a pyrochemical process, called TRUMP-S (TRansUranic Management by Pyroprocessing Separation), for partitioning actinides from PUREX waste. This effort is funded by the Central Research Institute for the Electric Power industry, Kawasaki Heavy Industries (KHI), and Rockwell International. Progress made on TRUMP-S during 1992 involves interpretation of americium data, cathode and anode kinetics tests, separation tests, and methods to enhance the actinide analytical sensitivity.

TASK 2. TRU PARTITIONING

2.1 OBJECTIVE

The overall objective of the Rockwell International/Kawasaki Heavy Industries/Central Research Institute for the Electric Power Industry (Rockwell/KHI/CRIEPI) program is to develop a process to recover 99% of the remaining transuranic (TRU) materials (Np, Pu, Am, Cm) from PUREX waste to simplify nuclear waste management. The purpose of the program is to separate the high-level nuclear waste into a TRU-rich stream and a TRU-depleted stream to reduce waste management costs. The actinide product can then be transmuted into stable products or stored. This can markedly reduce the amount of actinides in the waste requiring long-term isolation or repository emplacement and can supply significant quantities of fissile and fertile materials for power production during transmutation. The process goals are to separate 99% of each actinide from the waste into an actinide-rich product which is at least 90% actinides.

2.2 ACCOMPLISHMENTS

2.2.1 Interpretation of Americium Data

Performing experiments with americium (Am) is complicated by the fact that americium exhibits both divalent and trivalent character and is sufficiently radioactive to exhibit autooxidation of the lower valent species, Am(II), to a higher valent species, Am(III). Autooxidation/autoreduction means that one or more radiolysis products are formed *in situ*, which can change the valence state of the americium even when no external electrochemical work is introduced into the system. This phenomena has been reported in the plutonium⁶⁻¹⁰ and americium¹¹ literature for aqueous solutions of their compounds and appears to carry over to molten salt systems of these materials also.

When metal is present in Am molten salt solutions, there is also a corrosion mechanism that can reduce oxidized americium, Am(III), to the reduced species Am(II) by dissolving the metal ($\text{Am} + 2\text{Am}^{+3} = 3\text{Am}^{+2}$). Thus experiments with americium must be made rapidly enough to prevent these *in situ* valence changes that occur from interfering with interpretation of the results. To accomplish this, rapid electroanalytical techniques have been developed and used on this program¹².

Katz et al.¹¹ states that both autoreduction and autooxidation has been observed in Am solutions, is the first order in Am concentration and the rate varies from 4%/h to about 10%/h for various media, which is in good agreement with our value of 2.6%/h considering the different media (aqueous versus molten salt) used. Rabideau¹⁰ found autoreduction rates were about 0.02%/h for Pu-239, which is consistent with the 2.6%/h found for Am-239 considering the factor of 56 difference in the radiation levels in solutions of these two materials.

Leary and Mullins¹³⁻¹⁵ presented data to show that Am(II) was the predominantly stable oxidized Am species in the presence of metal. The Am/Am(II) standard potential and free energy of formation can be derived from the data of Alire¹⁶ and Leary and Mullins; from these data, the standard potentials were found to be -2.839 V and -2.877 V, respectively. Using a cyclic voltametric technique, Grimmer et al.¹² found the Am/Am(II) standard potential to be -2.86 V and that the

Am(III)/Am(II) redox potential was observed at a 0.48 V more positive potential. Grimmer's data show that the Am(0)/Am(III) couple is unstable and should not be directly observable due to the reduction of the Am(III) to Am(II) by the metal. Thus, both Leary¹⁵ and Grimmer¹² concluded that only Am(II) is the electrochemically stable species of Am in the presence of metal; the Am(III) reported by some molten salt investigators is presumably an autooxidation product.

Using an electrodeposition technique developed to measure the standard potentials of the other actinides and lanthanides, the standard Am/Am(II) potential was found to be -2.85 V. During these tests, a straight line with a slope of $n = 2$ for a potential versus $\log [Am]$ plot was only observed when a voltage was applied initially that could have reduced the Am(III) to Am(II). In the other tests a concave downward slope was observed, which should be expected if both Am(III) and Am(II) were present initially, and the Am(III) was reduced to Am(II) during the tests.

Grantham¹⁷ et. al. also fabricated an Am metal electrode from a piece of Am metal contained in a Ta wire cage. This electrode was dipped into molten LiCl-KCl eutectic containing Am chloride of various concentrations to obtain a potential versus $\log X [Am]$ diagram. The Am standard potential obtained from these measurements was somewhat less at -2.83 V but in good agreement with the data obtained in the electrodeposition test. The data used to obtain these standard potentials are given in Figure 1.

Considering the dual valency of americium and the autooxidation occurring in this system as well as the three different techniques we used to study Am, the spread in our data, -2.83 to -2.86V, is reasonable and is in good agreement with Alire¹⁶ and Leary.¹³⁻¹⁵ The activity coefficient of Am in cadmium solvent was found to be 10^{-4} , in good agreement with the other actinides.

2.2.2 Cathode and Anode Studies

The major purpose of the cathode and anode studies was to demonstrate that the metal and metal salt complexes formed in the cadmium and eutectic solvents, respectively do not impair the transfer of these materials from the anode to the cathode during processing.

Cathode studies utilize cyclic voltammetry and chronopotentiometry to study the reactions taking place at the cathode during deposition. To date, Ce, Mg, Nd, U, Np, and Am cathode deposition reactions have been studied and in all cases the reaction has been found to be rapid, reversible, and not influenced by any precursor reactions. Thus, the cathode reaction should depend on how fast the materials can be brought to the cathode surface by mixing and not by precursor chemical reactions in solution. Am cathode data from which this conclusion was obtained is shown in Figure 2.

Figure 1. Projected and Selected AmCl_x Potentials

Figure 2. Am Peak Cathode Current vs (Scan Rate)^{1/2}

The anode studies use small amplitude current-overpotential measurements to study the chemistry of the anode oxidation reaction. Linear current overpotential relationships indicate that simple Butler-Volmer type kinetics are occurring at the anode. The kinetic constant, the exchange current density, is a fundamental electrochemical property that can be obtained from the slope of the current-overpotential plots. To date, the oxidation of Gd, Nd, La, Y, Np, and Pu from Cd have been studied. In all cases, the current-overpotential plots for these materials have been linear, indicating a simple electron transfer oxidation. The calculated exchange current densities indicate that the anode oxidation reaction from Cd is rapid and does not depend on any precursor chemical reactions in the Cd or salt solutions. The exchange current densities of Gd, La, Y, and Np are given in Table 1.

Table 1. Exchange Current Densities

X in Salt	X in Cd	i_0 (mA/cm ²)	i_0 Scaled to $X_{MCl_3} = 0.0015$ $X_{M/Cd} = 0.005$
Gd - 0.0017	0.024	50	9.5
La - 0.0012	0.00075 (satd)	4.7	39
Y - 0.0022	0.079 (satd)	0.9	0.4
Np - 0.0015	0.0045	2.7	3

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The corrosion studies were made to determine the rate at which the corrosion reaction would redissolve the electrodeposited metal. The corrosion rate of Gd and Y have been studied under TRUMP-S conditions. These studies have shown that the Gd and Y electrodeposits are stable for at least 100 min, which provides adequate time to separate these deposits from the system without significant corrosion occurring. The Gd and Y corrosion data are given in Figure 3.

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Figure 3. Corrosion of Gd and Y at 500 C

2.2.3 Distribution Measurements

The salt to cadmium distribution coefficient data for the U-Nd, Np-Nd, and Pu-Nd systems are given in Figures 4 through 6. These data are in good agreement with data obtained at CRIEPI. The logarithm of distribution plots show a linear relationship over 1-2 orders of magnitude. The distribution coefficients can be developed from the slopes of the lines in these plots and are 43, 18, and 21 for the U, Np, and Pu in Nd containing solvents, respectively.

2.2.4 Separation Studies

2.2.4.1 Rare Earth Separation Tests

Five rare earth separation tests have been completed: three two-element tests, one four-element test, and one five-element test. All the tests contained La and either Nd or Gd to obtain the widest potential difference available for the rare earths being studied on the TRUMP-S program. Intermediate potential elements Pr and/or Ce were present in the four- and five-element tests.

The first test was run at constant current while the last three tests were run under potential control. At equal concentrations, the best separation obtained between Nd and La was 4:1 under constant current control while the best separation under potential control was 10:1. At constant potential, the best separation of Gd and La was 72:1 when they were present at equal concentrations; the separation obtained when Gd was present at 10% the concentration of La was 3:1.

These data show that potential control enhances separation significantly. The most unexpected result was that maximum separation of Gd, Ce, and Nd from La occurred at a potential about 40 mV less negative than the potential of the most negative element, whereas one might expect maximum separation at a potential just positive of the element with the most negative potential. These data suggest that the rapid deposition of one element disrupts the normal exchange current density of the other elements at the electrode. This disruption depresses the activity of the other elements on the electrode so that a significant net deposition results in the other elements that can deposit. Another unexpected observation was that appreciable current was passed at 50 mV above the potential of the least negative element present. This current flow might be related to divalent ions present in these systems as shown by Bronstein¹⁸.

2.2.4.2 Actinide/Rare Earth Separation Tests

Three uranium separation tests from LiCl-KCl eutectic melt containing GdCl₃ and NdCl₃ have been completed. The test results showed (1) over 99% of the uranium removed from the salt, and (2) excellent actinide/rare earth separation. The overpotential versus current data were obtained at four different UCl₃ concentrations.

Figure 4. U-Nd Distribution Data

Figure 5 Np-Nd Distribution Data

Figure 6. Pu-Nd Distribution Data

Initially in these tests, it appeared that only a little over 90% of the uranium could be removed from gadolinium and neodymium solutions, whereas theoretically it would appear that 99.9999+% should be removed. The problem was that neodymium was interfering with the uranium analysis – 1 ppm neodymium was responding as 0.116 ppm of uranium during analysis. After the correction for the amount of Nd present was made, it was established that no uranium was detectable. Thus, greater than 99.4% of the uranium was removed.

Based on the separations performed to date and the measured standard potentials of the actinides and Gd (the rare earth with the potential closest to the actinides), the actinide/rare earth separation could be as high as $10^{11}:1$, $10^7:1$, $10^5:1$, and $10^3:1$ for U, Np, Pu, and Am, respectively. These separations, if realizable in actual practice, would exceed the program goal of separating at least 99% of each actinide from PUREX residue.

2.2.5 Enhancement of Analytical Sensitivity

During Stage 3 of the TRUMP-S development studies, it is planned to demonstrate the two pyropartitioning steps (pyrochemical reduction, and electrorefining). To demonstrate 99.9% removal of actinides from molten salt and molten cadmium processing solvents, it is necessary to enhance the detectability of actinides. In Stage 3, we propose to start with concentrations found in typical PUREX waste where the starting concentrations vary by two orders of magnitude. Since it is required that we demonstrate 99% removal of each actinide, it will be necessary to increase the actinide detection limit of the analytical methods used by approximately two orders of magnitude.

2.2.5.1 ICP Detection Limits

The expected concentration in parts per million (ppm or micrograms per gram) of actinides in the process solvent salt or cadmium metal are given in Table 2 along with the projected ICP sensitivity and radioactive counting rates. The ICP samples assume 200 mg of sample have been dissolved in 20 grams of aqueous acid solution (i.e., diluted 1:100). Therefore, the actinide concentration in the sample would have to be two orders of magnitude greater than the ICP detection limit to be seen in the ICP spectra.

2.2.5.2 Radioactive Counting Detection Limits

The expected counting rates of the radioactive species activated in the Missouri University Research Reactor (MURR) are given in the last four columns in Table 2. Different counting techniques are planned for different actinides, and the detection limit may have to be determined by experiment. The counting technique and a preliminary estimate of the detection limit for the four actinides are given below.

Am will be determined by counting the intrinsic gamma activity due to Am-241 decay. Np will be activated prior to pyropartitioning and the resultant Np-238 decay gamma will be counted. The expected counting rates for Am and Np assuming a 30-min activation period in the neutron flux of the reactor for Np and a counting efficiency of 1% for the selected gamma ray are given in Table 2. The expected counting rates shown for Am and Np indicate that the radioactive counting techniques

Table 2. Expected Actinide Concentrations with ICP and Radioactive Counting Detection Sensitivities

Designated Sample No.	Sample Description			Expected Actinide Concentration (ppm) ^a				Concentration for ICP Detection (ppm) ^b				Expected Counting Rate (cts/min/ppm) ^c			
	Step	Solvent	Process	U	Np	Pu	Am	U	Np	Pu	Am	U ^d	Np ^e	Pu ^f	Am ^g
7XX-4AXX	4	Salt	Red. Less Act. M.	78,000	11,900	900	9,700	20	30	20	2	8.6	1.2E6	8E8	9.3E3
7XX-4BXX	4	Cd	Red. Less Act. M.	8	1.2	0.09	1.0	20	30	20	2	8.6	1.2E6	8E8	9.3E3
7XX-4CXX	4	Salt	Red. Act. & R.E.	50	6.0	0.5	5.0	20	30	20	2	8.6	1.2E6	8E8	9.3E3
7XX-4DXX	4	Cd	Red. Act. & R.E.	8,000	900	90	800	20	30	20	2	8.6	1.2E6	8E8	9.3E3
7XX-5AXX	5	Salt	After Elec. Ref.	15	1.8	0.2	1.5	20	30	20	2	8.6	1.2E6	8E8	9.3E3
7XX-5BXX	5	Cd	After Elec. Ref.	7	0.8	0.07	0.8	20	30	20	2	8.6	1.2E6	8E8	9.3E3
7XX-5CXX	5	Cath	Last Elec. Dep	8	1	0.1	1	20	30	20	2	8.6	1.2E6	8E8	9.3E3

^aAssumes 99.9% actinides present for 7XX-4AXX and 7XX-4DXX samples and 0.1% actinides present for 7XX-4BXX, 7XX-4CXX, 7XX-5AXX, and 7XX-5BXX; assume only 0.1% of Pu and Am present and 0.01% U and Np present in last electrodeposit where actinide free salt comprises 90% of sample.

^bConcentration in sample before dilution for ICP analysis.

^cE = exponential to base 10 (i.e., 9.3E3 = 9,300, etc.)

^dU-237 count rate = 10^{-3} cts/min/ppb if σ (n, 2n) for U-238 = 1 millibarn; if Sr-89 U-235 fission product counted (U-235/Pu-239 fissions=3) ct. rate = 1.2E3

^eAssuming 30 minute activation before start of electrochemical test and counted 1 week after activation

^fAssuming Ru-105 fission product counted 1 hr after activated (Pu-239/U-235 fission yield ratio=3)

^gNatural disintegration rate of Am-241 counting Np-237 daughter gamma

of these samples should enhance the analytical sensitivity three orders of magnitude to the part per billion (ppb) or nanogram of actinide per gram of sample range.

Uranium (U) detection will be the most difficult to increase by three orders of magnitude. The most sensitive counting technique for U may be activation with epithermal neutrons for 5 sec and counting within 10 min of activation in the reactor. Using epithermal neutron activation enhances the uranium cross section two to three orders of magnitude greater than the cross section of Cl or K. Compton scattering of the high-energy gammas from Cl-38 and K-42, which enhances the U-239 counting background will be the most difficult problem to overcome. A test was conducted for uranium to determine a preliminary U detection limit. Using epithermal neutrons to activate U-238, the counting rate from the U-239 gamma after a 5-sec activation period was sufficient to discern about 1 ppm of uranium in a typical TRUMP-S mixture.

Using a high energy, short half-life fission product probably will provide the greatest sensitivity of detection for Pu. Assuming the Ru-105 fission product is used, the detection limit for plutonium should be less than 1 ppb.

The expected counting rates of the radioactive species activated in the MURR are given in the last four columns in Table 2. It appears that the detection sensitivity will be sufficient to determine the quantity of actinide remaining after 99% of each actinide has been removed.

2.2.6 Computer Modeling of the Process

Two computer programs to model the TRUMP-S separation are being developed. Dr. Retzliff's model (being written at MURR) attempts to rigorously model the difficult problem of diffusion through the salt solvent and the boundary layer that exists during deposition. Dr. Retzliff has been able to rigorously solve many of the equations required, but his model is not sufficiently developed to predict separations yet.

The Roy/Grimmett code uses fundamental electrochemical equations including both thermodynamic and kinetic terms to predict separations in a real-time, voltage-controlled process. Using an inert anode, the separation efficiency is given in Figure 7. An indication of the actinide deposition rates in a system using an inert anode and cathode is shown in Figure 8. The treatment of the current-overpotential equation used in this computer model matches laboratory experiments well. Constants in the model to predict the overall rate and limiting current have been chosen to match experiment.

The simulation calculations shown in this report indicate that the TRUMP-S goal of obtaining a metal product containing less than 10% rare earths along with removing 99% of the actinides from the process salt can be met in a narrow potential window located at approximately -1.94 V referenced to the silver chloride electrode. The approximately 15 mV width of this window is smaller than expected due to the inclusion of americium as a divalent ion in the salt.

Figure 7. Removal Efficiencies for "-1.94-V Test"

Figure 8. Salt Composition as a Function of Time

The slower the process approaches the limiting potential (-1.94 V in the " -1.94 V Test"), the better the product composition. In the simulation, the limiting potential of -1.94 V was approached at a constant current of 2 mA/cm^2 . Approaching it at lower current density would take more time, but produce a better product. This concept is limited by the equilibrium separation values. If the limiting potential is approached infinitely slowly, it would essentially be in equilibrium, producing the best possible separation with the given starting conditions.

2.3 PROGRAM SUMMARY

Basically the test results have not changed the process as initially outlined. The major effect of the test results to date on expected separation efficiency and product purity has been the fact that Am is divalent in equilibrium with metal while the other actinides and rare earths are predominantly trivalent. This will make Am more difficult to separate but the process should still accomplish process goals. The fact that rare earth potentials are more negative than actinide potentials at the anode may restrict the rate at which the process can be operated unless some method can be found to reduce the impact of this anodic trait on processing rates.

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