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TRANSURANIUM QUARTERLY PROGRESS REPORT
FOR PERIOD ENDING AUGUST 31, 1962

OAK RIDGE NATIONAL LABORATORY

operated by

UNION CARBIDE CORPORATION

for the

U.S. ATOMIC ENERGY COMMISSION

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CHEMICAL TECHNOLOGY DIVISION
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ENGINEERING AND MECHANICAL DIVISION
REACTOR CHEMISTRY DIVISION
PLANT ENGINEERING, ORGDP

TRANSURANIUM QUARTERLY PROGRESS REPORT FOR PERIOD ENDING
AUGUST 31, 1962

Compiled by
D. E. Ferguson

DATE ISSUED

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TRANSURANIUM QUARTERLY PROGRESS REPORT FOR PERIOD ENDING
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1.0 ABSTRACT

The development of separation processes for the transuranium elements, process-equipment development, HFIR target-fabrication development, design of the TRU facility, design of development facilities, corrosion studies, analytical research and development, and the preparation of uranium-232 samples for research are reported here.

Process Development. Chemical processes and related equipment for the Transuranium Element Program are being developed. Progress is reported on the Tramex process, which is an amine solvent extraction from concentrated lithium chloride solutions to separate the transplutonium elements from corrosion and fission products. Another solvent extraction process for recovering americium and curium in nitrate wastes from plutonium processing is also reported.

The Tramex process was tested in laboratory scale runs with irradiated Am^{241} added at low activity levels, and recoveries of greater than 99.8% were obtained. The decontamination factor from mixed fission products was about 5000, which is more than adequate for the TRU program, but is not quite sufficient for preparation of Cm^{242} to be used in thermoelectric heat sources. The decontamination factor for ruthenium, which was the limiting contaminant, was about 1000. Preliminary tests with sodium nitrite added to the stripping solution to oxidize ruthenium to the tetravalent state, thus keeping it in the solvent, gave ruthenium decontamination factors of about 10^4 . This is just adequate for Cm^{242} heat source preparation. Development of methods for improving ruthenium decontamination will be continued.

Small-scale (1-2 ml) batch tests of the Tramex process at 10 w/liter, which is the proposed processing level, gave the same distribution coefficients noted in tracer-level experiments. Faster separation of the mixed phases noted at this radiation level is beneficial. Equipment has been fabricated for additional testing at full activity level.

Distribution coefficients determined for Tramex conditions show that Pu^{4+} , Sb^{3+} , Sn^{2+} , and Ba^{2+} will be separated from the transplutonium elements. The first three ions will be extracted but will remain in the solvent as the transplutonium elements are stripped into 4-10 M HCl. Although Ba^{2+} will follow the transplutonium elements during stripping, it can be prevented from entering the solvent in the first place if the AlCl_3 concentration in Tramex feed is less than a few hundredths molar.

The solvent extraction process being considered for Am-Cm recovery from plutonium process waste is an alternative to the anion exchange method that was discussed in the previous quarterly report. Like the anion exchange process, it is primarily useful

in separating americium and curium from aluminum and will also remove some fission products but not the rare earths. The extraction method was laboratory tested with simulated feed containing Cm^{242} tracer. The results were good with curium recovery of 99.8%. Though americium tracer was not used, it is probable that its recovery would have been at least as good as the curium recovery.

Equipment Development. Equipment development was continued on the evaluation and improvement of the contacting equipment used in the transuranium elements processing program. The Tramex flowsheet was used with nickel as a stand-in for americium and curium. Nickel was used because its extraction coefficient permits column operation at the same flow ratio as specified by the transuranic flowsheet. Small-scale (0.82- and 0.75-in.-ID) pulsed columns were used as the contacting equipment.

The results show that the extraction rate for nickel is adequate for conventional solvent extraction contactors. The stage-heights obtained with a pulse column, although three to four times higher than those for uranium extraction by TBP, are encouraging, considering the high viscosity of both the solvent and the aqueous phases. Sieve plates were superior to glass helical packing, based on efficiency and throughput.

HFIR Target Fabrication Process Development. Work was continued on the development of target-fabrication procedures, the development of equipment for target fabrication in the TRU Processing Facility, the development of plutonium-target-fabrication equipment, and the planning of procedures and methods to be used in the fabrication of HFIR target-irradiation prototypes. Welding studies led to the establishment of a technique that appears suitable for producing leak-tight end closures, with about 70% penetration. Pellet-fabrication studies set 50 mils as the minimum thickness of the aluminum-powder cap, and expansion measurements indicated that the expansion characteristics of reference pellets are comparable with those of type 1100 aluminum. The thermal conductivity of simulated target pellets was found to be a sensitive function of the actinide oxide loading, decreasing rapidly with increasing oxide content. Neutron activation tests showed that the reference pellets were permeable to fission gases.

Design, development, and fabrication of remote processing equipment is progressing on schedule. The pellet-fabrication press was completed and successfully operated in preliminary tests. Several items of similar equipment for the plutonium target-fabrication line are being built or procured.

Design Studies. Work was continued on the design of the process test facility, shielding studies, the design of chemical processing equipment, scheduling, and the TRU Facility. The design of the process test equipment is about 85% complete. The installation of the storage tanks for feed solutions in cell 4 pit is complete, and the shielding is complete. The cell walls proper were coated with epoxy glass-fiber lining in preparation for the installation of piping and other process equipment. The equipment-removal cubicle, makeup-area equipment, and some cell process equipment are being built.

Shielding studies were made in order to determine a suitable aggregate for use in the TRU shielding concrete. The shielding criteria are: It should attenuate the neutrons and gamma rays from a 3×10^{12} neutron/sec fission source such that the dose rate at the outside surface of the 54-in.-thick wall is less than 1 mrem/hr when the source is located 2 ft from the inside surface. Montana magnetite is preferred on the combined basis of shielding requirements and cost.

Work on the design of chemical-processing equipment was directed towards completing preliminary equipment and engineering flowsheets. In conjunction with this, materials of construction for all equipment items and piping systems were tentatively assigned. With respect to materials of construction for process equipment, material take-offs are being made so that an order may be placed by October 1 for all Hastelloy C and stainless steel required for the process equipment as part of the order for similar materials for the waste system and other building components. The choice of Hastelloy C was based on criteria imposed by corrosion rates and frequency of usage. The feed-adjustment tanks including the vessel head and all dip tubes must be of tantalum or be tantalum lined. Product storage tanks will probably be made of tantalum-lined Hastelloy C, with Zircaloy tops and dip tubes.

Materials Evaluation Studies. The objective of these studies is to determine radiation resistance of and ease of decontamination of protective coatings that will be used in certain parts of the TRU Facility. So far, the Amercoat 66 epoxy system is preferred. Though the decontamination factor is low (11), the radiation resistance, especially in deionized water, is higher than that for any other coating tested.

TRU Corrosion Studies. The study of corrosion in process solutions of proposed materials of construction for the TRU Processing Facility was continued. The corrosion resistance of welded specimens of Hastelloy B was superior to that of Hastelloy C in Tramex process solutions which contained either oxygen or hydrogen peroxide. The presence of ferric ion accelerated the corrosion of both Hastelloys from 5-10 mils/month to about 20 mils/month at 43°C. Niobium was found to be subject to hydrogen embrittlement in the various chloride-containing solutions. Both tantalum and Zircaloy-2 exhibited acceptable corrosion rates and resistance to hydrogen embrittlement under simulated process conditions.

Analytical Development. During this reporting period, alpha spectrometry was applied to the determination of Am^{241} in the presence of Cm^{242} . A 256-channel nuclear-data analyzer in conjunction with a silicon diode detector was used in conjunction with the work. After appropriate chemical treatment of the process solutions (Cm^{242} containing Am^{241} in the range 0.3 to 1.4%), the solutions were analyzed, and the uncertainty was $\pm 10\%$. Equipment is being readied for the absorption-spectrophotometric studies on the transplutonium elements.

Uranium-232 Preparation. Five samples of U^{232} were recovered and purified from two successive irradiations of 50 g of Pa^{231} . The recovery and purification methods are reported.

2.0 INTRODUCTION

Gram quantities of transuranium elements for research uses are to be produced in the High Flux Isotope Reactor, HFIR, and Transuranium Processing Facility, TRU, being built at Oak Ridge National Laboratory. Production of these elements in quantity will simplify research with them and make it possible to enlarge our knowledge of their chemistry, solid state physics, and metallurgy. Target material will be provided for production of still heavier and unknown elements.

Isotopes of curium, berkelium, californium, einsteinium, and fermium will result from irradiation in the HFIR of Pu^{242} and of a mixture of Am^{243} with Cm^{244} . These feed materials are being produced by long-term irradiation of 10-kg batches of Pu^{239} in a Savannah River reactor. The irradiated Pu^{239} will be processed at Savannah River to recover the Pu^{242} as decontaminated PuO_2 and the americium-curium as a solution containing about 3 kg of rare earth fission products. The rare earths will be removed from the americium-curium and the actinide oxides fabricated into HFIR targets in the TRU Facility. Also in the TRU Facility the irradiated HFIR targets will be processed to recover the heavy elements for research uses and the recovered curium isotopes will be refabricated into HFIR targets.

Progress reports are included on the development of separation processes for the transuranium elements, process equipment development, HFIR target fabrication development, design of the TRU Processing Facility, corrosion studies, analytical research and development, and the preparation of U^{232} samples. This work was performed by the Chemical Technology, Metals and Ceramics, Engineering and Mechanical, Analytical Chemistry, and Reactor Chemistry Divisions of Oak Ridge National Laboratory and the Design Group of the Oak Ridge Gaseous Diffusion Plant.

The previous report in this series is ORNL-3290.

3.0 PROCESS DEVELOPMENT

3.1 Chemical-Process Development

R. E. Leuze, R. D. Baybarz,
J. E. Bigelow, M. H. Lloyd

Methods for recovering transuranium elements from irradiated HFIR targets, recovering and purifying curium from irradiated Am^{241} , and recovering americium and curium from plutonium process waste are being developed. Actinide purification by the Tramex Process (tertiary amine extraction), fission product behavior, ruthenium-zirconium sorption on anion exchangers, hydrogen peroxide stability in HCl solutions, and recovery of americium-curium-rare earths from nitrate waste by tertiary amine extraction are reported below.

3.1.1 Tramex Process Evaluation

The data from laboratory-scale tests with spiked feeds show that the process is more than adequate for the Transuranium Element Program. However, additional removal of gamma active contaminants must be demonstrated for the preparation of Cm^{242} to be used in thermoelectric heat sources. The decontamination factor for ruthenium, which was the limiting contaminant, was about 1000. Preliminary tests with sodium nitrite added to the stripping solution to oxidize ruthenium to the tetravalent state, thus keeping it in the solvent, gave a ruthenium decontamination factor of about 10^4 which is just adequate. Additional studies will be made at higher activity levels.

A series of mixer-settler runs was made with irradiated Am^{241} at 0.2 w/liter to evaluate curium recovery and fission product decontamination. The process consisted in the use of 10.4 M LiCl-0.2 M AlCl_3 feed, 10.5 M LiCl scrub, 0.6 M Alamine 336 HCl-diethylbenzene (DEB) extractant at flow ratios of 1:1:2 for scrub, feed, and extractant.

With eight scrub and eight extraction stages operating at about 60% efficiency, 99.9% of the curium was recovered, and the rare earth decontamination factor was about 5×10^3 . Ruthenium and zirconium were quantitatively extracted with the curium. In the curium strip with 8 M HCl, with an equal-volume side stream of 1% H_2O_2 and an equal volume of fresh organic scrub, most of the zirconium was kept in the organic phase (DF about 10^4). The ruthenium also remains mainly in the organic phase. The small amount stripped, less than 0.1%, was nonextractable with a fresh solvent scrub. Silver-111 is extracted from 11 M LiCl. During the curium strip, Ag^{111} is stripped by 8 M HCl but is extracted from 4 M HCl, causing it to reflux in the mixer-settler. Overall curium recovery was greater than 99.8% with a fission product gamma DF of about 10^3 .

A small-scale (1-2 ml) batch test of the Tramex process at 10 w/liter was made with 1 mg of irradiated Am^{241} to determine if high alpha radiation levels produce adverse effects. Distribution coefficients were the same as noted in tracer-level experiments with the only effect noted being faster phase separation. Based on the excellent behavior of the solvent extraction system at 10 w/liter, it appears that processing at 20 w/liter may be feasible.

3.1.2 Ruthenium Extraction Behavior

The behavior of ruthenium in the Tramex flowsheet is being investigated. For the Tramex process, the decontamination factor for ruthenium is about 1000. Addition of an oxidizing agent to the strip solution has increased this to 10^4 which is just adequate for purification of Cm^{242} to be used in heat sources. Ruthenium exists as Ru^{3+} and Ru^{4+} in HCl and LiCl solutions. Hydrogen peroxide will oxidize the Ru^{3+} to Ru^{4+} in HCl and LiCl. Tetravalent ruthenium is strongly extracted into 0.6 M Alamine 336 HCl-DEB from LiCl and HCl solutions while Ru^{3+} is weakly extracted. The Ru^{4+} that extracts is slowly reduced by the amine to the inextractable Ru^{3+} . During curium

stripping operations it is necessary to provide an oxidant to reoxidize the Ru^{3+} to the extractable Ru^{4+} , thus maintaining it in the organic phase. During mixer-settler operations a small portion (≤ 0.1) of the ruthenium is stripped with the curium and will not be re-extracted with fresh organic. The use of H_2O_2 in the HCl strip solution was used as an oxidant for Ru^{3+} ; however, recent information on H_2O_2 stability indicates that it is unsuitable for use at elevated temperatures.

In cold laboratory tests reduction of the ruthenium with aluminum metal in the presence of $\text{N}_2\text{H}_4 \cdot \text{HCl}$ produced a ruthenium species that had a distribution coefficient (DC) of less than 0.05. However, in mixer-settler tests with irradiated Am^{241} , in which the feed was treated in this manner, about 2% of the ruthenium was extracted along with the actinides.

3.1.3 Extraction Behavior of Plutonium, Antimony, Tin, and Barium

The extraction behavior of Pu^{4+} , Sb^{3+} , Sn^{2+} , and Ba^{2+} in the Tramex Process was investigated, and the results are given below. The process ensures the removal of these elements from the americium-curium products. The Sb^{3+} , Sn^{2+} , and Pu^{4+} are extracted and remain in the organic phase as the americium-curium are stripped by 4-10 M HCl. The Ba^{2+} follows americium and curium in the stripping, but it can be prevented from even entering the organic phase in the first place if the AlCl_3 concentration is less than a few hundredths molar.

Plutonium is strongly extracted from 11 M LiCl into 0.6 M Alamine 336 HCl-DEB with a DC of about 1400. The plutonium extraction in HCl solutions increased with HCl concentration to 7 N then decreased slightly (Fig. 3.1).

Trivalent Sb had a DC of 23 from 11 M LiCl into 0.6 M Alamine 336 HCl-DEB, and its extraction from HCl decreased with increasing acidity.

Divalent Sn had a DC of 11 from 11 M LiCl and a DC of 70 from 9 M LiCl-0.33 M AlCl_3 into 0.6 M Alamine 336 HCl-DEB. The presence of AlCl_3 greatly enhances the Sn^{2+} extraction. The Sn^{2+} extraction in HCl increases to a maximum at 5 N HCl then decreases with increasing acidity.

Barium extraction is enhanced by the presence of AlCl_3 in LiCl. The barium DC increased from 0.1 at 0.0025 M AlCl_3 to about 2 at 0.06 M AlCl_3 in 11 N LiCl vs 0.6 M Alamine 336 HCl-DEB. The extraction from HCl solutions increased with acidity.

3.1.4 Tramex Process Test Using Cm^{242}

Equipment for a small-scale test of the Tramex Process was installed in cell 2 of the analytical hot cells in Building 3019. The objective of the test is to determine the behavior of short-cooled fission products at full-activity level of 10 w/liter. Results will indicate the appropriate direction of further process development work.

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Fig. 3.1. Extractability of plutonium, tin, antimony, and barium.

in order to obtain the required separation of transuranium elements from fission products. This work will include a demonstration of the recovery and decontamination possible by this process. Work done so far and work planned are described below.

Approximately 100 mg Am^{241} were canned in aluminum and irradiated for 39 days at a perturbed neutron flux of 1.4×10^{14} . The Cm^{242} content, measured by a calorimeter, was 14.5 mg at the time of reactor discharge and was very near the calculated value. At the time of processing, 12.8 mg of Cm^{242} will remain. This will be made up into 200 ml of Tramex feed at a power density of 8 w/liter.

The solvent extraction portion of the test will consist of six batch-operated mixing chambers, each with its own stirrer. Calibrated solution-transfer chambers provide for interstage transfer of the settled phases, while similar devices meter the feeds. It is anticipated that a complete cycle of stirring (2 min), settling (3 to 5 min), and interstage transfer can be accomplished in 15 min. Vacuum for transfer will be provided by a water aspirator discharging first to a phase separator and then to hot drain. The 200 ml of hot feed will allow about 25 cycles of 8 ml each. The corresponding cyclic feed rates of scrub and organic solvent will be 5 and 10 ml, respectively. The entire operating period for the solvent extraction part of the process will be about 10 hr. Raffinate, waste solvent, and product will be sampled several times during the operation, and at least two samples will be taken from each stage.

3.1.5 Ruthenium-Zirconium Sorption on Anion Exchange Resin

Methods other than solvent extraction for removing ruthenium and zirconium from americium-curium solutions were investigated and are reported below. The ion exchange behavior of ruthenium and zirconium on Permutit SK and Dowex 1 resin was investigated as a possible second-cycle process for removing fission products.

The ruthenium-zirconium DF's are both about 1000 in this procedure for preparing a feed for the Tramex process in order to get an extra DF. However, this step may be unnecessary, and efforts will be confined to improving the Tramex process, itself.

The adsorption of Ru^{4+} and Zr^{4+} on Permutit SK and Dowex 1-8X from HCl solutions differed in some respects (Figs. 3.2 and 3.3). The sorption of zirconium was greater on Permutit SK than on Dowex 1, and zirconium was sorbed at low acid as well as high acid concentrations. Ruthenium sorption was greater on Dowex 1 than on Permutit SK but the effect of acidity on sorption was the same for both resins.

The DC's for ruthenium and zirconium in an LiCl-HCl system gives the possibility of using one feed condition to achieve high DC for both elements (Fig. 3.4). In 5 M LiCl (varying the acid from 1 to 4 N) vs Permutit SK, the ruthenium DC drops while the zirconium DC increases. At the following feed condition, 5 M LiCl-3.8 M HCl, the DC's are 90 for Ru^{4+} and Zr^{4+} . With this feed condition macro quantities of ruthenium and zirconium were loaded on a Permutit SK resin column to the extent of about 3 g per liter of resin, giving a DF of about 10^3 .

Fig. 3.2. Ruthenium and zirconium sorption on Permutit SK resin.

Fig. 3.3. Ruthenium and zirconium sorption on Dowex 1-8X.

Fig. 3.4. Ruthenium and zirconium sorption on Permutit SK from 5 M LiCl containing HCl.

Fig. 3.5. Effect of temperature on sorption of ruthenium and zirconium on Permutit SK from 5 M LiCl-3 M HCl.

The temperature had a marked effect on the DC for ruthenium and zirconium (Fig. 3.5). The DC's increased rapidly to about 40°C then increased slowly with temperature.

3.1.6 H₂O₂ Stability in HCl Solutions

The stability of H₂O₂ in 8 M HCl at 30 and 65°C was determined (Fig. 3.6).

Fig. 3.6. Stability of H₂O₂ in 8 M HCl.

The H₂O₂ has a T_{1/2} of about 5 min at 30°C and a T_{1/2} of about 1 min at 65°C. Since H₂O₂ decomposes rapidly in HCl solutions (O₂ and Cl₂ are produced), it cannot be relied upon to oxidize Ru³⁺ to Ru⁴⁺ during a stripping operation.

3.1.7 Americium-Curium Recovery

Americium and curium in plutonium process waste can be recovered by extracting the americium and curium and the rare earth fission products with 0.6 M Alamine 336 HNO₃ in DEB from 1 to 2 M Al(NO₃)₃ solutions, scrubbing with 8 M LiNO₃ and eluting with 3 M HCl to yield a nitrate-free solution that may be converted to 11 M LiCl for selective extraction of americium and curium by Alamine 336 HCl-DEB.

Distribution coefficients for curium from 3 to 8 M LiNO₃ vs 30% Alamine 336 HNO₃ in DEB were determined (Fig. 3.7), and are proportional to the 7th power of the LiNO₃ concentration. Al(NO₃)₃ and LiNO₃ were interchangeable with no difference in extractability. The distribution coefficients were proportional to the square of the Alamine 336 HNO₃ molarity in DEB (Fig. 3.8).

Fig. 3.7. Effect of LiNO_3 concentration on curium extraction by 30% Alamine 336 HNO_3 -DEB.

Fig. 3.8. Effect of amine concentration on curium extraction from 2 M $\text{Al}(\text{NO}_3)_3$.

A mixer-settler extraction of a curium-ruthenium solution in 1 M $\text{Al}(\text{NO}_3)_3$ was made. The feed was 1 M $\text{Al}(\text{NO}_3)_3$ and contained Cm^{242} and $\text{Ru}^{103-106}$. The scrub was 8 M LiNO_3 , and the extractant was 0.6 M Alamine 336 HNO_3 -DEB. Flow rates of 1:1:2 were used for scrub, feed, and extraction, respectively. A curium recovery of 99.8% was achieved with eight scrub and eight extraction mechanical stages operating at about 70% efficiency. The ruthenium split, with about 60% extraction. The pregnant organic was stripped with 1/2 volume of 3 M HCl , with a 1/2 volume organic scrub of 0.6 M Alamine 336 (neutral)-DEB. The overall curium recovery was 99.8%, with a ruthenium DF of 10^3 . The ruthenium remained in the organic phase during stripping. The curium product was 2.4 M HCl -less than 0.01 M NO_3^- , which is suitable for feed adjustment for the Tramex process.

3.2 Process-Equipment Development

P. A. Haas, A. D. Ryon

3.2.1 Solvent Extraction Contactor Development (F. L. Daley)

The data reported this quarter is part of the evaluation and improvement studies of the contacting equipment used in the transuranic element processing program. The flowsheet studied consists of the extraction of nickel (as a stand-in for americium and curium) from 11 M LiCl by 30% Alamine 336 in diethylbenzene (DEB) and subsequent stripping of the nickel into 2 M HCl . Nickel was used because its extraction coefficient permits column operation at the same flow ratio as used in the transuranic flowsheet. Small-scale (0.82- and 0.75-in.-ID) pulse columns were used as the contacting equipment.

The results show that the rate of extraction of nickel is adequate for conventional solvent extraction contactors. The stage heights obtained with a pulse column, although 3 to 4 times higher than those for uranium extraction by TBP, are encouraging, considering the high viscosity of both the solvent and aqueous phases. Sieve plates were superior to glass helical packing, based on efficiency and throughput.

Batch Rate Tests

The rate constants for nickel extraction and stripping are adequate for conventional contactor operations. Batch rate tests were made in order to determine the rate constant (k_a) of the extraction and stripping cycle. A 6-in.-diam, 12-in.-high, baffled tank mixer equipped with a 3-in.-diam 6-blade turbine was used. The power input to the mixer was held at 2.5 hp per 1000 gal. Solution temperature was controlled at 25°C for these tests. An aqueous-continuous and an organic-continuous batch extraction-rate test at a phase ratio (aqueous/organic) of 1:1 and an organic-continuous batch-stripping test at an A/O of 1:3 were made. Samples were taken at timed intervals during the tests.

Table 3.1 Batch Rate Data for Extraction and Stripping Cycle

Extraction Cycle											
Aqueous Feed: 11 M LiCl ~5 Ni g/liter Organic Feed: 30% Alamine 336 in DEB Phase Ratio (A/O): 1:1 Temperature: 25°C											
Continuous Phase	Nickel Concentration in Aqueous Phase (g/liter)										
	0 sec	5	10	20	30	45	60	120	240	480	960
Aqueous	5.51	2.78	2.46	2.15	1.98	1.77	1.61	1.38	1.22	1.21	1.19
Organic	5.51	2.46	2.00	1.71	1.52	1.42	1.31	1.25	1.20	1.18	1.19
Stripping Cycle											
Aqueous Feed: 2 M HCl Organic Feed: 30% Alamine 336 in DEB ~4 g/liter Ni Phase Ratio (A/O): 1:3											
Continuous Phase	Nickel Concentration in Organic Phase (g/liter)										
	0 sec	5	10	20	30	45	60	120	240	480	960
Organic	4.38	0.41	0.10	0.009	0.005	0.002	0.003	0.003	0.004	0.002	0.001

The rate data (Table 3.1) for the batch tests were plotted (Fig. 3.9) and the rate constants (k_a) calculated (ORNL-2951). Organic-continuous operation of the extraction cycle gave a k_a of 1.26 min^{-1} , while aqueous-continuous operation gave a k_a of 0.79 min^{-1} . Organic-continuous operation of the stripping cycle gave a k_a of 13.8 min^{-1} . The rate constants for the extraction of nickel from 11 M LiCl by 30% Alamine 336 in DEB are of the same order of magnitude as that found for the Dapex Process (ORNL-2951) (extraction of uranium from a sulphuric acid solution by di-2-ethylhexyl phosphoric acid, $k_a = 0.54 \text{ min}^{-1}$) but much less than that found for the extraction of uranium from a nitrate solution by TBP ($k_a = 17.3 \text{ min}^{-1}$).

Continuous Runs

Two small, glass pulse columns (0.82-in. ID by 30 in. long, and 0.75-in. ID by 24 in. long) were erected and tested to determine stage heights for the extraction and stripping cycles. The column internals were: stainless steel sieve plates; 1/32 in. holes, 23% free area, spaced 1/4 in. apart; one hole nozzle plate; and 1/8-in. glass helices. Micro-bellows pumps, manufactured by Research Appliance Co., equipped with calibration tubes, were used as feed pumps. The columns were pulsed by either a Milton-Roy positive displacement pump or by a micro-bellows pump with the check valves removed. The aqueous-organic interface in the column was controlled by use of an external gravity leg on the aqueous discharge stream. All tests were conducted with the solution throughput at approximately 80% of flooding. The column end streams were sampled periodically during each run for analysis. All tests were conducted at about 25°C. Table 3.2 gives the physical properties of the solutions.

Table 3.2 Physical Properties

Solution	Specific Gravity (g/cc at 25°C)	Viscosity (centipoise at 30°C)
30% Alamine 336 in DEB	0.871	6.05
11 M LiCl	1.242	5.96
2 M HCl	1.017	0.873
30% TBP in Amsco	0.814	1.63
0.5 M HNO_3 24 g U/liter	1.020	1.03

The high viscosity of the 30% Alamine 336 in DEB-11 M LiCl solutions tends to make pulse-column operation difficult. As a comparison, a Purex flowsheet (solutions of more favorable physical properties) was tested in the 0.82-in.-ID pulse column, packed with sieve plates. An HETS, calculated by use of a McCabe-Thiele plot (Table 3.3), of 5.3 in. was realized. The performance of the column was similar to that reported, HETS of about 2 in., by Hanford¹ in a small pulse column of essentially the same design.

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Fig. 3.9. Batch rate curves for transfer of nickel.

Table 3.3. Purex Run Data

Aqueous: 0.5 M HNO_3 , 23.3 g U/liter, flow 20 ml/min
 Organic: 30% TBP in Amsco, flow 20 ml/min
 Pulser: micro-bellows, frequency 40 cpm, amplitude 0.20 in.

Flowing-Stream Samples		
Time (min)	Uranium Concentration (g/liter)	
	Raffinate	Extract
45	0.142	24.9
60	0.123	24.9
75	0.120	24.2
90	0.099	24.3
105	0.098	24.2
120	0.103	25.0

Equilibrium Samples		
Aqueous	Organic	$\frac{D}{A}$
6.41	17.1	2.7
1.24	3.81	3.1
0.009	0.039	4.3

Effect of Pulse Frequency. Three nickel extraction runs were made by using the 30% Alamine 336 in DEB-11 M LiCl flowsheet in order to study the effect of pulse frequency on the HETS. The 0.82-in.-ID by 30-in.-long pulse column packed with sieve plates and pulsed by a micro-bellows pulser was used for these tests. The aqueous-organic interface was held at the top of the column. Pulse amplitude was held constant at 0.2 in. The data presented in Tables 3.4 and 3.5 show that as pulse frequency was increased, the column HETS decreased, from 20 in. at 40 cpm to 10 in. at 130 cpm to 6.6 in. at 200 cpm.

Effect of Pulser Wave Type. The micro-bellows pulser has a stroke adjustment which stops the bellows movement during part of the cam rotation. At the stroke required for a 0.2-in. amplitude, the bellows is in contact with the cam for approximately 1/3 of a revolution, which results in a truncated sine-wave pulse. A Milton-Roy piston pump, with the check valves removed, was installed in order to compare the effect on HETS of the sinusoidal pulse wave produced by the Milton-Roy pulser with the truncated wave produced by the micro-bellows pulser. The column was operated aqueous-continuous (top interface). No effect on HETS was noted at low pulse frequency (40 cpm), but as the pulse frequency was increased to 200 cpm, the micro-bellows pulser gave a shorter HETS by almost a factor of two (Table 3.6). This is probably due to finer disper-

Table 3.4. Nickel Extraction and Strip Data for Pulse Columns

Run No.	Solutions		Type of Column and Packing	Pulser			Inter-face	Flow Rate (ml/min)		Nickel Feed Conc. (g/liter)	Analysis of End Streams, Ni Conc. (g/liter)		HETS (in.) ^b
	Aq	Org		Type	Frequency (cpm)	Amplitude (in.)		Aq	Org		Aq ^a	Org ^a	
4	11 M LiCl	30% Alamine 336 in DEB	0.82-in. sieve plate	Micro-bellows	40	0.2	Top	15	15	3.49	0.561	2.79	20
5	11 M LiCl	30% Alamine 336 in DEB	0.82-in. sieve plate	Micro-bellows	130	0.2	Top	5	5	4.85	0.236	5.00	10.1
6	11 M LiCl	30% Alamine 336 in DEB	0.82-in. sieve plate	Micro-bellows	200	0.2	Top	5	5	4.85	0.008	5.36	6.6
10	11 M LiCl	30% Alamine 336 in DEB	0.82-in. sieve plate	Milton-Roy	40	0.2	Top	12	13	4.05	0.747	3.23	22
11	11 M LiCl	30% Alamine 336 in DEB	0.82-in. sieve plate	Milton-Roy	130	0.2	Top	5	5	4.05	0.733	3.60	22
14	11 M LiCl	30% Alamine 336 in DEB	0.82-in. sieve plate	Milton-Roy	130	0.1	Bottom	5	5	3.55	0.504	4.50	16
16	11 M LiCl	30% Alamine 336 in DEB	0.82-in. sieve plate	Milton-Roy	200	0.2	Top	5	5	4.43	0.350	4.00	12
17	11 M LiCl	30% Alamine 336 in DEB	0.75-in. 1-hole nozzle plate	Milton-Roy	30	0.1	Top	5	5	3.94	0.866	2.80	26
20	11 M LiCl	30% Alamine 336 in DEB	0.75-in. 1/8-in. helices	Milton-Roy	60	0.1	Top	5	5	3.80	2.74	1.00	>>24
12	2.0 M HCl	30% Alamine 336 in DEB	0.82-in. 1D sieve plate	Milton-Roy	60	0.2	Bottom	5	20	3.06	8.65	0.005	

^a Average of last three end-stream samples.^b Calculated by use of a McCabe-Thiele plot.

Table 3.5. Equilibrium Data for Nickel, 11 M LiCl vs 30% Alamine 336 in DEB

Run No.	Ni Concentration (g/liter)		D_A^O	Run No.	Ni Concentration (g/liter)		D_A^O
	Aqueous	Organic			Aqueous	Organic	
4	0.069	0.443	6.42	16	0.250	1.11	4.44
	0.774	2.72	3.51		1.12	3.10	2.77
	1.89	3.90	2.06		2.07	4.61	2.23
5 and 6	0.031	0.253	8.16	17	0.138	0.637	4.62
	1.27	3.67	2.89		1.00	2.96	2.96
	4.01	5.63	1.40		2.37	4.41	1.86
	4.37	5.23	1.20	20	0.581	2.22	3.82
10 and 11	0.474	1.92	4.05		0.936	3.18	3.40
	1.99	5.23	2.65		1.25	3.55 ^a	2.84
	2.24	5.15	2.30	12	2.84	0.001	0.0005
14	0.949	3.38	3.56		12.7	0.002	0.0002
	2.28	4.80	2.11		14.5	0.001	0.0001

^aCalculated by material balance.

Table 3.6. Effect of Pulse-Wave Type on HETS

Pulse Frequency (cpm)	Pulse Amplitude (in.)	Type of Pulser			
		Micro-bellows		Milton-Roy	
		Run No.	HETS (in.)	Run No.	HETS (in.)
40	0.2	4	20	10	22
130	0.2	5	10	11	22
200	0.2	6	6.6	16	12

sion developed by the bellows pulser. Tables 3.4 and 3.5 contain the data for these tests.

Effect of Column Interface Position. One nickel extraction run (run 14) was made in the 0.82-in.-ID column, packed with sieve plates, with the organic-aqueous interface held at the bottom of the column. When the Milton-Roy pulser was set at a frequency of 130 cpm, amplitude 0.1 in., an HETS of 16 in. was realized (Tables 3.4 and 3.5). Comparison of the bottom-interface run with a similar top-interface run (run 11, frequency 130, amplitude 0.2 in., HETS 22 in.) shows that the bottom interface run gave more efficient extraction.

Nozzle-Plate Test. A glass column (24 in. long, 0.75-in. ID) packed with nozzle plates (one 1/16-in.-ID nozzle protruding 0.1 in. above and below the plate, 2% free area, spaced 0.5 in. apart) was tested in run 17 with the nickel extraction flowsheet (Tables 3.4 and 3.5). The HETS was 26 in., using the Milton-Roy piston pump at a frequency of 30 cpm and an amplitude of 0.1 in.

Packed Column Test. A 0.75-in.-ID by 24-in.-long glass column, packed with 1/8-in. glass helices was tested (run 20) with the nickel extraction flowsheet (Tables 3.4 and 3.5). With the Milton-Roy pulser at a frequency of 60 cpm and an amplitude of 0.1 in., less than one stage of extraction was realized—calculated HETS of 80 in.

Stripping Test. One run (run 12) was made in the 0.82-in.-ID column packed with sieve plates to test the stripping of nickel from 30% Alamine 336 in DEB by 2 M HCl. Due to the extremely low distribution coefficient (D_A^O) of 0.001 (Tables 3.4 and 3.5), the nickel concentration of the stripped organic was too low for analysis and HETS calculations. One theoretical stage in the column is adequate.

3.2.2 TRU Mockup (T. S. Mackey)

The initial installation of mockup of the TRU Processing Facility in Building 9204-1, Y-12 Plant, was completed. The following items will be investigated in the order listed:

1. Evaluation of the manipulator booting seals.
2. General operational features such as manipulator coverage, viewing areas, etc.
3. Automatic port-closure evaluation.
4. Intercell conveyor testing.
5. Intercell pass-through evaluation.
6. Equipment-rack handling.
7. Lighting-equipment maintenance.

4.0 HFIR TARGET-FABRICATION-PROCESS DEVELOPMENT

W. C. Thurber

4.1 Target-Fabrication Development

4.1.1 Target-Rod Welding Studies (R. G. Gilliland)

Investigations are continuing on the development of end-closure welding procedures for remotely sealing the HFIR target rods. The objective is to effect high-integrity closures in 3/8-in.-OD by 0.060-in.-wall, type X-8001 aluminum tubing without filler-metal additions.

The problems are primarily those of nonfusion in the weld area and weld-metal blowout of the final closure weld in the target rod. Nonfusion is believed to be associated with the tenacious surface oxide on the aluminum. Weld-metal blowout in the final target rod weld is believed to be a result of the buildup of gas pressure inside of the element from the rapid temperature rise during welding. Blowout then occurs through the molten pool. Figure 4.1 is an illustration of the effects of gas-

Fig. 4.1. Fusion weld in X-8001 aluminum showing giant pore created by gas-pressure buildup during welding. Etchant: 1% HF-1% HNO₃-1% HCl-bal H₂O. 30X.

Fig. 4.2. Typical fusion welds in X-8001 aluminum, made by using the (a) butt-, (b) plug-, and (c) edge-joint designs. Etchant: 1% HF-1% HNO_3 -1% HCl-bal H_2O . 30X. Reduced 20%.

pressure buildup. It should be noted that this difficulty does not occur in making the initial closure weld.

As a result of these problems, together with an irregular weld-bead contour, a study of the influence of joint design was initiated. Welds were made by using several basic end-cap joint designs: butt, plug, and edge. Illustrations of welds made with these designs are shown in Fig. 4.2a, b, and c. These are typical welds made in open-ended tubes with air-cooled copper chill-rings placed on both sides of the joint.

When the second and final closure weld is made, only the plug-type welds can be made with a moderately low rate of rejection (25%). Weld-metal blowout is a greater problem with the other joint designs. An improvement in joining consistency was accomplished when the welding conditions were adjusted to reduce the heat input to the weld, but weld penetration was decreased.

Currently, the best results are attained when the final weld in the target rod is made such that there is approximately 70% penetration. The photograph of Fig. 4.3 illustrates such a weld. The welding condition required for producing welds of this

Fig. 4.3. Typical final closure weld in X-8001 aluminum. Etchant: 2% HF, H₂O. 30X.

configuration is a welding current of 38 to 40 amps at a speed of 9 to 10 rpm. Using these conditions, dummy elements are being fabricated to aid in the investigations of tube collapse and coolant flow.

Studies are continuing in order to develop improved techniques for welding these target rods. Included in the work are technique refinements such as fit-up, joint chilling, etc., and new apparatus development that provides very rapid travel speeds, with improved chilling.

4.1.2 Pellet Fabrication and Properties (D. M. Hewette II)

Pellet Fabrication. To produce pellets that have contamination-free surfaces, the aluminum-target oxide mix is pressed in a thin-walled aluminum tube capped with pure aluminum powder on each end. Original specifications² called for a 25-mil-thick cap. Difficulty has been encountered in obtaining sound end caps of this thickness even when the samples are manually prepared in a "cold" laboratory under ideal conditions. The problem involves obtaining an even distribution of cap powder covering the oxide mix.

To further examine this problem, an experiment was performed in which two groups of 30 pellets were pressed, one with 25-mil caps and the other with 50-mil caps. The 25-mil cap pellets were prepared by carefully tamping the oxide-aluminum mix into the die assembly prior to loading the top-cap powder. The 50-mil cap pellets were prepared by simply pouring the oxide-aluminum mix into the die assembly, gently tapping the die to level the mix somewhat, and pouring the cap powder on top of the mix. Subsequent to pressing, the pellets were radiographed to determine the integrity of the end caps. A pellet was rejected if the aluminum-oxide mix projected through the cap to the surface. In the group with 25-mil caps, 15 out of 30 pellets were rejected, while in the 50-mil group only 1 out of 30 was defective. The use of a 50-mil end cap rather than a 25 appears advantageous for confinement of the contamination.

Pellet Resistance to Thermal Shock. Six target pellets of the original design concept were thermally cycled between room temperature and 600°C six times and between room temperature and 321°C an additional 46 times. No pellet degradation or dimensional changes were noted.

Thermal Expansion of Target Pellets. Dilatometric measurements have been made on HFIR target pellets of the present design concept. Linear thermal-expansion measurements were made between 25 and 400°C in vacuum. A dilatometer was used in which the expansion of the sample is not measured directly, but is determined by the differential expansion between the material being tested and the materials of construction in the dilatometer.³ Since the calibration of the dilatometer is based on a 1-in.-long sample, two target pellets were stacked to yield a suitable sample.

Average coefficients of expansion were calculated for two sets of specimens and are listed in Table 4.1 along with values for type 1100 wrought aluminum. There

Table 4.1. Dilatometric Measurements on HFIR Target Pellets^a

Preparation: Pressed at 30 tsi to produce pellets having a core of approximately 90% theoretical density

Temperature (°C)	Average Coefficient of Linear Expansion (in./in./°C × 10 ⁶)		
	Samples 7 and 14	Samples 29 and 30	Wrought Type 1100 Aluminum
25-100	16.0	17.0	24.0
25-200	22.5	22.7	24.9
25-300	24.6	24.4	25.8
25-400	24.5	25.2	26.9

^aMaterial: Al-11 vol % Gd₂O₃ pressed in type 1100 aluminum tube and covered top and bottom with type 1100 aluminum cap powder.

is no real difference in thermal expansion behavior between the two sets of target pellets. Comparing the results obtained on the target pellets with those obtained on wrought aluminum, there is fairly close agreement for all temperature ranges except for the range 25 to 100°C. No permanent axial or radial expansion was noted with these composite pellets. Accordingly, it is concluded that the thermal expansion characteristics of the target pellets will be essentially the same as that for type 1100 aluminum.

4.1.3 Thermal Conductivity of Cold-Pressed Al-Gd₂O₃ Pellets (D. L. McElroy, W. Fulkerson, T. Kollie)

The thermal conductivity of cold-pressed pellets of aluminum and 0, 10, 15, 20, and 25 vol % Gd₂O₃ (as a stand-in for the actinide oxides) was measured at 75°C in the thermal comparator apparatus. The thermal conductivity values decreased rapidly with increasing Gd₂O₃ as noted below:

Oxide Concentration (vol %)	Thermal Conductivity (w/m °C)
0	146
10	25.5
15	15.3
20	8.45
25	8.45

Analysis of the data by the Raleigh-Maxwell equation for two-phase structures verified the observation (by microscopy) that the aluminum was the discontinuous phase

and the gadolinia was the continuous one. Expected axial temperatures were calculated for the target rods, assuming an aluminum metal surface temperature of 155°C, radial heat flow, a pellet thermal conductivity independent of temperature, and a power level of 29.5 kw per target rod. The present reference pellet loading of 11.8 vol % PuO_2 fixes the centerline temperature at 485°C, and melting of the aluminum particles is predicted for pellets containing about 17 vol % oxide.

Subsequently, tests at 40 and 125°C were completed, and the thermal conductivity values obtained are within $\pm 10\%$ of those at 75°C, indicating only a modest temperature dependence for these pellets. Pellets containing a mixture of aluminum and UO_2 were recently received for testing, and metallography established the continuous phase to be aluminum. Conductivity measurements are not yet complete.

4.1.4 Fission-Gas Release from Target Pellets (J. L. Scott, R. E. Adams)

It has been estimated that 215 cc (STP) of gaseous fission products will be generated during the irradiation of a first-cycle target rod. One cause for concern is uncertainty about the mobility of these fission gases within the rods. Are they retained in the lattice of the individual pellets, and, if not, is there a path by which they can migrate to the end plenum that is provided for accumulation?

To answer this question, a series of tests were conducted with pellets containing normal enrichment ADU-route UO_2 as a stand-in for PuO_2 . The pellets were made by pressing an 18 vol % UO_2 -Al powder mixture in a type 1100 aluminum tube, with an aluminum-powder cover approximately 25-mils thick at the top and bottom. The final pellet size was 0.250-in. in diameter and 0.500 in. long.

Four pellets were irradiated for different lengths of time in the ORNL Graphite Reactor at ambient temperatures. The irradiation was conducted in aluminum capsules that had been welded shut in helium and then leak tested to establish weld integrity. After a one-week decay period, the capsules were punctured, and the gases that were released during irradiation were drawn into activated charcoal traps at 74°K. The bulbs were then analyzed quantitatively for Xe^{133} . Subsequently, the target pellets were given heat treatments at about 200, 400, and 600°C in vacuum in order to determine the fractional release of xenon as a function of temperature. Finally, the pellets were heated to about 1000°C, which is well above the melting point of aluminum (660°C), in an effort to drive off the remainder of the Xe^{133} .

The results of the four tests are given in Table 4.2. In all tests there was a significant release of Xe^{133} from the sealed capsules upon puncture. This release indicates that the pressed aluminum end plugs are porous to fission gases. It was also found that the fractional release on puncture decreased with increasing exposure over the range examined. The reason for this anomaly is not known. The fractional release on postirradiation heating was small, relative to the amount of xenon released during irradiation, until the pellets reached the temperature at which the UO_2 -Al reaction occurs in the absence of radiation (about 600°C). When the reaction occurs,

Table 4.2. Xenon¹³³ Release from HFIR Target Pellets

Specimen	6	5	9	10
Exposure (total fissions x 10 ¹⁵)	0.018	0.15	1.45	1.35
Fractional release ^{a,b} on puncture	5.0	4.7	3.8	3.8
200°C	ND ^c	ND ^c	0.6	0.6
400°C	ND ^c	ND ^c	0.25	0.1
600°C	ND ^c	ND ^c	8.5	6.1
1000°C	<u>83.2</u>	<u>99.2</u>	<u>71.7</u>	<u>93.4</u>
Total release ^a	88.2	103.7	84.8	104.0

^aCalculated on the basis of burnup.

^bSamples were held at each temperature about 24 hr, except for 1000°C treatment.

^cND = not determined.

the amount of xenon released appears to be related to the degree of completion of the reaction. Upon melting, essentially all the xenon escaped. The variations in the reported values of total release, which were calculated on the basis of burnup, are probably due to errors in the measurement of the integrated flux.

The principal fact demonstrated by these tests is that the aluminum end-plugs are permeable to xenon. In all the tests, the burnup was negligible and heat treatments were conducted without fissioning. These conditions were quite different from those which will exist in HFIR target pellets after prolonged irradiation. The behavior of fission gases under such conditions can be determined only by testing prototype pellets in-pile to burnups similar to those in HFIR. Such experiments are planned. (See Sect 4.4).

4.1.5 Materials Procurement (S. A. Rabin)

Finned Tubing. About 100 ft of X-8001 aluminum finned tubing was purchased from Precision Extrusions, Inc. (Bensenville, Illinois). Owing to the stringent dimensional tolerances, the material was furnished on a best-efforts basis. The temper was as-fabricated (X-8001-F), the tubing having been hot extruded at 900°F followed by a cold-stretch straightening operation. The latter step results in a slight amount of residual cold work.

Chemical analyses of the finned tubing showed it to be within the prescribed limits. Dimensional inspection indicated that the outside diameter, inside diameter, and fin-to-fin tip tolerances were met; however, the wall is eccentric and the fins

are, consequently, slightly spiralled. The maximum bow over a 3-ft length also is greater than the specified 0.010-in. per foot of length. Further dimensional measurements are in progress.

Because of the heavy wall and continuous longitudinal fins on the as-received tubing, ultrasonic and eddy-current nondestructive tests are not applicable without further development. Ultrasonic inspection of a single tube from which the fins had been stripped showed no discontinuities as great as indications from a 5% notch reference standard. With a 0.003-in. notch fluid-penetrant inspection of the same material showed no imperfections. The tube was then cut in half along its longitudinal axis and penetrant inspected. No defects greater than 0.003-in. in depth were found. Radiographic inspection of a piece of machined tubing of the irradiation-experiment configuration indicated a circumferential crack-type discontinuity at one location. Linear indications were found with visual and fluid-penetrant examination at the base of the fins on the nonmachined surface, but no flaws were found where the tubing had been machined below the root of the fins.

Hexagonal Tubing. Slightly over 100 ft of X-8001-F hexagonal tubing was obtained from the Aluminum Company of America. The material thus far inspected is within tolerance across the flats but exceeds the 0.005-in.-per-foot-of-length straightness specification. It was also observed that the tubing is slightly rounded at the corners, but this could presumably be rectified by modified die design. Results of chemical analyses were similar to those for the finned tubing. Nondestructive testing of the hexagonal tubing was confined to visual, fluid-penetrant, and radiographic inspections. No discontinuities have been found. Further dimensional and nondestructive examinations are in progress.

4.1.6 Pellet and Target-Rod Decontamination (V. H. Kiplinger, M. K. Preston)

Tests to evaluate various methods for remote decontamination of both pellets and target rods have continued. Two steam cleaning units were tested and vapor-blast tests were also performed, but both of these cleaning methods were less effective than ultrasonic cleaning. Dry abrasive cleaners were not tested because of space limitations in the cubicle, excessive air consumption, and an apparent disposal problem involving quantities of grit that would become contaminated.

The first steam cleaning unit tested operated at 60 psig steam pressure. The cleaning solution was siphoned into the stream by jet action. The second unit evaluated, an injector pump, used the same steam pressure but operated at much greater nozzle pressure than the first (240 psig). In neither case, however, was decontamination of a mockup target rod satisfactory. The target rod was divided into 6-in. sections having various degrees of surface finish, from 20 to 120 μ in., and UF_4 powder was rubbed into the rod to provide a source of alpha contamination. The amount of surface roughness within this range did not affect the final cleanness appreciably unless the roughness was caused by a peening action after the surface was contaminated. For each degree of finish, a small but significant alpha count was usually

obtained after thoroughly steaming the rod. The high-velocity steam jet was somewhat less effective in cleaning than the low-pressure jet, but this difference was attributed mostly to the cleaning solutions used. Decon 4513 and 4306-C solutions were used with the high-velocity jet; whereas, trisodium phosphate was used with the 60-psig nozzle.

Decontamination tests using vapor-blast equipment were limited because it was decided that too much air would be required to operate such an installation inside a cubicle. Liquid sealing and recirculation, indexing the target rod and gun nozzle, and preventing the plugging and erosion of drain lines with abrasive grit disposal were other factors that discouraged decontamination by vapor blasting. However, four tests indicated that 0.0002 in. of metal could be effectively removed in about 1 min by using 3 gal of 400-mesh alumina grit and 7.5 gal of water. Other types of grit were not evaluated, although it was realized that alumina grit was not easily dissolved after contamination.

A series of ultrasonic cleaning tests were performed to determine the relative merits of various cleaning solutions on type 1100 aluminum rods and pellets. A 1-kw, 20-kc ultrasonic cleaner was used.

In the first test, aluminum rod sections that were still contaminated with UF_4 after steam cleaning were then soaked and ultrasonically cleaned in Decon 4306-C solution at 155°F (Table 4.3). The purpose of the test was to determine the effect of

Table 4.3. Aluminum-Rod Decontamination Test

Zone No.	Surface Finish (μin.)	Residual After Steam Cleaning (α cts/min)	Soaking, Cleaning Time (min)	Average Residual Content after Soaking (α cts/min)	Ultrasonic Cleaning Time (min)	Average Residual Counts after Ultrasonic Cleaning (+10 α cts/min)
1	25	75	60	18	60	8
2	30	83	60	11	30	1
3	35	77			60	0
4	33	127			30	1
5	50	34			5	2
6	50	148			15	-10
7	80	156			15	-10
8	115	118			5	2

surface finish on decontamination efficiency. It was found that soaking did not remove all the contamination from the rods but that subsequent ultrasonic cleaning was effective in reducing the contamination to zero for each degree of surface finish. Furthermore, a 5-min cleaning period was as effective as a 1-hr cleaning period, within the limits of accuracy of the measuring equipment.

Next, it was decided to check the effectiveness of an ultrasonically agitated bath of Decon 4306-C in removing the stearic acid die lubricant. After rubbing the 3/8-in. rods with UF_4 and taking initial alpha readings, a saturated solution of stearic acid in acetone was painted over the contaminated areas. At this point, the total initial alpha count was approximately 1000 counts/min. Rod zones 1, 3, 5, and 7 were then ultrasonically cleaned in the standard solution of Decon 4306-C (12 oz per gal of water) for 1 min, leaving a residual activity of 131 counts/min. Zones 2, 4, 6, and 8 were similarly cleaned for 5 min, leaving a residual count of 119 counts/min, indicating that there was incomplete cleaning in both cases.

As a continuation of the stearic acid and contamination removal tests, the 54 small pellets were recontaminated in the three groups of 18 each and then coated with stearic acid. The initial alpha readings were: Group I, 130 counts/min; group II, 117 counts/min; and group III, 190 counts/min.

Group I was ultrasonically cleaned for 1 min at 155°F in a solution of 15 g of NaOH to 280 cc of water. There was a rather violent boiling action, and 0.001 in. of metal was removed from the rod diameters in 1 min. No measurable alpha contamination remained after cleaning.

Group II was ultrasonically cleaned similarly in a solution of 15 g of trisodium phosphate (Na_3PO_4) in 280 cc of water. Again, no residual alpha contamination was detectable. While the pellets were visibly brightened, no measurable (less than 0.0005 in.) metal was removed. A slight boiling action was noted.

Group III was ultrasonically cleaned for 1 min at 155°F in a Decon 4305-C solution (6 oz of powder per gal of water). A residual alpha reading of 23 counts/min was observed, indicating incomplete cleaning. However, this result was considerably better than that from a preliminary test using the more concentrated Decon solution, which left about 120 counts/min.

Recently a group of uncontaminated reference target pellets were subjected to an ultrasonic-cleaning evaluation. The cylindrical surface of these pellets contained accordion-like folds apparently filled with metallic powder and stearic acid die lubricant.

Ultrasonic cleaning times of 1/2, 1, and 5 min were used with solutions of 15 g of Na_3PO_4 in 280 cc water and 1.5 g of NaOH in 280 cc water. Microscopy indicated that the 1/2-min period was insufficient to allow the cleaning solutions to penetrate the deepest folds, while both the 1- and 5-min periods appeared to be satisfactory.

A soaking period of up to 30 min in these solutions without ultrasonic vibration did not remove the deeply embedded surface contaminants.

In both the static soaking and ultrasonic cleaning, particles were lost from around the perimeter of the green-pressed aluminum cap. Also, a greater reaction rate was noted in this area. This problem is not yet resolved.

4.2 Development of Equipment for Fabrication Cubicles in TRU Processing Facility

M. K. Preston, A. L. Lotts, R. I. Deaderick

The basic criteria established for the target-rod fabrication process as outlined in the previous quarterly progress report⁴ have not changed. Status of equipment development for the fabrication cubicles in the TRU Processing Facility is discussed in following sections.

4.2.1 Cubicle 3

The process flowsheet and equipment layout for cubicle 3 remain as outlined previously. Conceptual designs on nine pieces of equipment for the cubicle were approved, and detailed design is under way or has been completed. Design approval was given on four completed designs, and fabrication work orders or purchase requisitions were issued.

The following is the equipment-design status for cubicle 3.

Actinide Calciner. The actinide calciner has not yet been conceptually designed because research has not progressed to a point where the criteria can be established.

Batch Scale. Design will be started after the pellet load scale design is completed and operable.

Blender Dispenser. A blending and metering device was conceptually designed, and studies of container configuration are continuing. The container will probably be a small bar mill or an "x" blender incorporating balls to break up agglomerates.

Pellet-Load Scale. Preliminary design and machine work incident to the testing and evaluation of the torsion balance for the weighing of pellet loading are in progress. A transducer is affixed to the scale in lieu of the weighing pan, and the motion of this unit is displayed on a differential transformer indicator. Testing of the equipment indicates that the desired accuracy of $\pm 1\%$ for a pellet loading of 0.5 to 0.75 g can be obtained.

Cap Powder Loader. A new conceptual design was completed and approved. Detailed design has started.

Die-Assembly Feeding Station. The conceptual design and final design are complete and approved, and the fabrication work order was written.

Pellet Press (see Fig. 4.4): Fabrication of the pellet press is complete and installation of piping and instrumentation is under way. Preliminary operation of the press using plant air instead of high-pressure water indicates satisfactory operation.

Pellet Ultrasonic Cleaner and Dryer. This equipment consists of four basic designs: (1) the ultrasonic cleaner and rotary vibratory feeder, (2) ultrasonic cleaner auxiliary enclosure, (3) pellet dryer and rotary vibratory feeder, and (4) pellet dryer auxiliary enclosure. Conceptual design has been approved. Design details are complete and are being checked for correctness. The ultrasonic cleaning container and rotary vibratory feeders are being procured from outside vendors.

Transfer Arm III. The conceptual design for this transfer has changed since the last report. It now has horizontal rotary motion only and is controlled by positioning cams located on the equipment to which the transfer is being made. The conceptual design was approved and detailed design has started.

Miscellaneous Containers. Containers for transporting punch and die assemblies, top punches, and their feed magazines have been designed and approved. These containers will also be used to collect and dispose of used equipment. The pellet transfer container, which has also been approved, is to collect fabricated pellets at the press for transporting them to the ultrasonic cleaner.

4.2.2 Cubicle 2

The process flowsheet and equipment layout for cubicle 2 have not changed. Conceptual designs on two additional pieces of equipment for cubicle 2 were approved. Detailed design is complete and approved on these items, and one concept previously approved is ready for final design approval. The following is the equipment-design status for cubicle 2.

Pellet Inspection and Loading Equipment. The pellet inspection and loading equipment consists of the pellet-diameter gage, stacked-pellet-length gage, two pellet feeders, and target-tube loader---all in an auxiliary enclosure. Detailed design is complete and the drawings were checked for correctness.

End-Cap Welding Machine. This equipment will consist of a tube-end cleaner, helium-filling system, end-cap placement device, and the end-cap welding head. Preliminary studies were started; however, conceptual design is being delayed until a suitable and reliable welding procedure can be established.

Helium-Leak-Test System. A commercial leak detector was purchased and is being modified and tested in order to simulate the long suction and discharge lines and to test the absolute filters that must be used. Recent tests indicate that the

Fig. 4.4. Pellet press - Transuranium Processing Plant.

absolute filters proposed for the facility will not retain or impede the flow of helium from a standard leak. The leak-test-chamber design is completed and approved, and fabrication has started.

Hydrostatic Collapse Equipment. The pressure chamber was designed and approved. However, recent investigations indicate that the proposed collapse pressure of 15,000 psi may have to be increased appreciably to ensure contact between the target tube and pellets. On this basis, the fabrication is being delayed until more information is available.

Target Ultrasonic Cleaner. This equipment will consist of an ultrasonic cleaning tank and transfer mechanism for removing the target from the tank to another transfer device for passing the target through the wall to cubicle 1---all of which is to be enclosed in an auxiliary housing. Conceptual design is just starting, and vendors were contacted for information on the ultrasonic cleaner.

Target-Transfer Arm. Conceptual design of the target-transfer arm awaits the establishment of more information on the process equipment in the cell and a firm decision on the configuration of the target rod itself. The transfer arm, however, will have rotary, horizontal, and vertical motions, powered by electric motors and positioned by positive steps for the various positions.

4.2.3 Cubicle 1

The process flowsheet and equipment layout for cubicle 1 have not changed. Equipment-concept design status has not changed appreciably since the previous quarterly progress report because the target rod design has not been firmly established.

4.3 Plutonium Target Fabrication Equipment Development

A. L. Lotts

During this reporting period, there was little change in the plans for the plutonium target fabrication, which were discussed in detail in the previous quarterly report.⁵ However, the schedule has changed: Instead of the operational date of January 1963, "hot" operation of the equipment is not now required until July 1963 because of changes in the HFIR. The equipment for the plutonium target line is in various stages of design and procurement; none has been placed in operation or tested. The following is a summary of the status of the equipment.

4.3.1 Glove Boxes

The design of the standard glove boxes was revised to provide 12-in.-diam bag-out ports in both ends of each box, a seal door to cover each bag-out port, and certain minor modifications to facilitate mounting of equipment and enable handling

of the 35-in. target elements. Seven of these boxes, two 36 in. long and five 72 in. long—all made primarily of carbon steel and glass and painted with Amercoat No. 74—are being procured.

4.3.2 Powder-Preparation Equipment

The PuO_2 to be supplied to the process will probably be -325-mesh material. However, to ensure that all material is -325 mesh, a ball mill and screen will be used when necessary. A Fisher 8-oz "Minimill" was purchased for use in ball milling small volumes of PuO_2 . Other items that have been purchased are a torsion balance for weighing in the boxes and a Gramatic balance for weighing that is to be done outside the boxes. A suitable commercial blender has not been found.

4.3.3 Pellet-Preparation Equipment

A 10-ton press for making pellets is being assembled in ORNL shops. A furnace for dewaxing the pellets, an ultrasonic cleaning unit for decontaminating them, and a torsion balance for checking pellet weight were obtained.

4.3.4 Tube-Closure Equipment

An apparatus for evacuating and helium backfilling the tube, placing the tube end cap, and welding the cap is now being assembled. A 60-in. lathe bed will be used to hold the assembled element in the horizontal position for automatic welding of the end-cap joint. The mechanical components and control circuit for the device were built, and a direct-current welding machine was obtained.

4.3.5 Hydrostatic-Collapse Equipment

The design of the hydrostatic-collapse equipment for the TRU target-fabrication line was modified to fit glove-box operation. However, the approval of the design for construction has not been given because of incomplete data on collapse pressures required.

4.3.6 Helium-Leak-Test Equipment

The design of the leak-test chamber for TRU was modified to suit glove-box operation. Construction has not started. A leak detector equipped with an automatic test-port station was received.

4.4 Fabrication of HFIR Target Irradiation Prototype

A. L. Lotts, D. M. Hewette II, R. G. Gilliland

Considerable attention was given during this quarter to planning procedures and techniques to be used in the fabrication of HFIR target irradiation prototypes. By use

of existing equipment where possible, four prototype HFIR target rods are to be fabricated, using Pu^{239} as a substitute for Pu^{242} , and other actinide elements and simulate the fissioning that will occur in the actual targets. Three of these rods will be irradiated in the Materials Testing Reactor (MTR) to validate the design of the HFIR target element from the standpoint of resistance to irradiation. One prototype rod will be retained as a control for the experiment. It differs from the HFIR target element in one principal respect: It is 24 1/2 in. long and contains a fissionable loading approximately 14 in. long, while the reference HFIR target element is 35 in. long and contains a loading 20 in. long.

The prototypes will be fabricated by using the methods proposed for the HFIR target elements. All items of equipment required (except an end-cap welding machine and a helium-leak-test system, which are now being assembled) are on hand.

5.0 DESIGN STUDIES

W. E. Unger

5.1 Design of Process Test Facility

F. L. Peishel, W. R. Whitson, T. Shapiro, B. J. Crump.

The process-test facility will be located in cells 3 and 4 of Building 4507. Cell 4 will contain solvent extraction process equipment. The head-end equipment, consisting of a dissolver and the accompanying gas-handling equipment will be located in cell 3.

The design of the cell process equipment is about 85% complete. The installation of the storage tanks for feed solutions in cell 4 pit is complete, and the shielding is in place. The cell walls proper were coated with epoxy glass-fiber lining in preparation for the installation of piping and other process equipment. The equipment-removal cubicle, makeup-area equipment, and some cell process equipment are being built. The equipment for the solvent extraction portion of the flowsheets will be arranged in cell 4, as shown in ORNL-3290, p 70 and 72.

Two 16-stage mixer settlers, designated MS-442 and MS-446, are to be provided to replace the previous 16-8 combination in order to improve product decontamination and permit a greater margin of variation in process conditions.

The tankage will be either glass or tantalum-lined Hastelloy C with Zircaloy-2 tops. Where boiling solutions or solutions containing mixtures of nitric and hydrochloric acids are involved, the tank tops will be coated with tantalum to improve corrosion resistance. All coils and dip lines are also made of tantalum tubing.

A typical arrangement of a cell tank is shown in ORNL-3290, p 73. This drawing also shows the disconnect array on the top for allowing the removal of each of the tanks by removing the jumpers between the tanks and the tubing and disconnect rack in the cell.

Pumps, mixer settlers, and miscellaneous supporting equipment for the solvent extraction process will be located on removable racks above the tankage. Piping from the operating and makeup areas will enter the cell through existing sleeves terminating inside new bulkhead boxes. The tubing will penetrate these boxes through fittings that will maintain an alpha seal with the cell. The large equipment in the cell will require the use of an overhead bridge crane and jib crane (as shown in ORNL-3290, p 72) in order to move the equipment under the 19- x 38-in. opening. The equipment or rack is then transferred to the hoist located in the large equipment removal box located in the penthouse of Building 4507.

The equipment removal box, as shown in ORNL-3290, p 72, is equipped with a hoist and work table for removal and maintenance of contaminated cell equipment. It is also equipped to bag large or small equipment in or out through three openings.

A design concept for removing beta-gamma contaminated items into a shielded carrier will be developed and the equipment built when required for contaminated-equipment handling.

Booting for the extended-reach, heavy-duty manipulators is under design to provide a positive seal at the cell wall; it will be capable of easy removal without jeopardy to the alpha-tight integrity of the shield wall.

A condensation of the detailed critical path schedule for cell 4 of Building 4507 is shown in Fig. 5.1. Cold startup is scheduled for November 1962, and initial hot operation should begin about a month later. Delivery dates of the tantalum, Hastelloy, and zirconium may affect exact compliance with the schedule.

5.2 Shielding

J. P. Nichols, T. E. Northup, E. D. Arnold

Shielding and economic studies were made in order to determine suitable combinations of aggregate for use in the TRU shielding concrete. The concrete should have minimum cost and attenuate the neutrons and gamma rays from a 3×10^{12} -neutron/sec fission source such that the dose rate at the outside surface of the 54-in.-thick concrete cell wall is less than 1 mrem/hr when the source is located 2 ft from the inside surface.

In earlier studies it was determined that a concrete of water content of 12 lb/ft³, an iron content of 100 lb/ft³, and total density of about 220 lb/ft³, equivalent to that developed by HAPO by using Montana magnetite ores, satisfies the dose-rate criterion. The Montana ore is primarily magnetite but contains some limonite, which contributes

Fig. 5.1. Cell 4, Building 4507, condensed critical path schedule.

chemically combined water. Concrete made from this ore contains the desired water content of 12 lb/ft³ at 70 to 80°F (the anticipated operating temperature of the bulk of the TRU concrete shield) and retains 9 lb of water per cubic foot after a prolonged exposure at 100°C. Other components of Montana magnetite concrete, in addition to water, in pounds per cubic foot are: Fe, 122; O, 71; Ca, 7.8; Si, 5.7; Al, 1.3; Mg, 0.25; and S, 0.19.

Renupak shielding calculations (Fig. 5.2) were made in order to compare Montana magnetite concrete with possible alternates that can be made from selected high-grade aggregates. The alternates consisted of combinations of heavy but nonhydrogeneous iron ore with serpentine, which has high water retention. Concrete made from a mixture of Tennessee ferrophosphorus and Maryland serpentine was determined to have satisfactory attenuation properties, equivalent to those of Montana magnetite that contain 12 lb of water per cubic foot, while a concrete made from a mixture of Montana magnetite and Georgia serpentine was determined to be unsuitable.

The choice between use of Montana magnetite or Tennessee ferrophos--Maryland serpentine aggregates for the TRU concrete will depend on cost. Total aggregate costs for the TRU Facility is about \$89,700 if Montana magnetite is used and may vary from \$55,000 to \$132,000 if Tennessee ferrophos--Georgia serpentine is used. Studies have been initiated by the AEC to determine better cost information on the ferrophos-serpentine concrete.

5.3 Chemical-Processing-Equipment Design

W. D. Burch, O. O. Yarbrow, B. J. Crump

Design efforts were directed towards completing the preliminary equipment and engineering flowsheets. In conjunction with this, materials of construction for all equipment items and piping systems were tentatively assigned. The ORGDP Engineering Department will assist in the detailed design of equipment in order to complement the manpower available at ORNL.

5.3.1 Equipment Flowsheets

The final two (of the set of five) equipment flowsheets, showing miscellaneous storage, waste, and rework tanks in cells 1 through 5, were completed. Minor revisions to the main-line processing equipment flowsheets were made to bring them up to date. These revisions included (1) some relocation of tanks to provide more space for expansion in the tank pits directly behind the chemical processing cubicles and (2) a switch of the solvent extraction contactors from mixer settlers to pulse columns. The decision to switch to pulse columns is still subject to confirmation of preliminary stage height data that indicated that pulse columns were feasible.

5.3.2 Engineering Flowsheets

The first draft of ten engineering flowsheets, of the total of 13 or 14 required,

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Fig. 5.2. Effect of type of concrete and concrete shield thickness on the dose rates from a point isotropic fission source.

is completed. Space limitations dictated that cubicles and tank pits be drawn on different sheets. Connecting lines on all flowsheets are "match-lined" in both horizontal directions and in the vertical plane where cubicle lines drain to the corresponding tank pit.

5.3.3 Materials of Construction for Process Equipment

Included in the line-numbering system on the engineering flowsheets is the size and material of construction of each line. Material take-offs are being made so that an order may be placed by October 1 for all Hastelloy C and stainless steel required for the process equipment as part of the order for similar materials for the waste system and other building components. In many cases, the choice of material for various pieces of equipment is somewhat arbitrary and subject to change. The flowsheets at present assume that Hastelloy C is acceptable wherever corrosion rates will be less than 3 mils/month for service in the separations systems and less than 1 mil/month in storage tanks. Higher corrosion rates may be tolerated where only infrequent usage is contemplated. The problem of corrosion product accumulation in product streams has not been examined in detail and may place more severe limitations on corrosion rates than those stated above, since the nickel contributed from the corrosion of Hastelloy C is not readily separable from the curium product.

Feed-adjustment tanks, including the vessel head and all dip tubes must be tantalum or tantalum lined. Product storage tanks will probably be tantalum-lined Hastelloy C vessels with Zircaloy tops and dip tubes. Zircaloy may be desirable for many applications where corrosion rates on Hastelloy C are borderline, since, in general, the corrosion rate of Zircaloy is a tenth of that for Hastelloy C. Base metal prices of Zircaloy are four times higher, but fabricated equipment costs are probably comparable. Fabrication problems and costs for both materials are under investigation. The requirement for the special heat treatment of Hastelloy C after welding places severe limitations on fabrication techniques.

5.3.4 Detailed Equipment Design

Engineering personnel from ORGDP will assist in the detailed design of the chemical processing equipment. A group of two or three engineers and about six or eight draftsmen will be required for 18 months. The exact number assigned to the job will be governed by the pace requirements defined by the Critical Path Schedule (CPS). Detailed estimates of the individual design activities on the CPS arrow diagrams indicate that 250 drawings, requiring about 12 man-years of drafting time and 6 man-years of engineering time, will be necessary to complete the processing equipment design.

The ORGDP design effort will start in September. At that time all engineering flowsheets will be completed, and design criteria for the components to be designed initially will have been prepared.

5.4 Scheduling

B. N. Robards

Catalytic Construction Company, the architect engineer, and ORNL developed arrow diagrams showing their respective responsibilities independently of each other, and the diagrams were combined into two master diagrams that reflect cost-plus-fixed-fee (CPFF) and lump-sum (LS) approaches to the construction of the facility.

The ORNL participation in the overall project was subdivided into the following phases: analytical chemistry equipment, chemical process equipment, instrumentation, mechanical equipment, metallurgy equipment, and equipment installation. Diagrams reflecting the sequence and duration of the activities describing the above phases were completed. These were combined by Catalytic Construction Company with the construction diagrams to make the master diagrams.

Figure 5.3 illustrates the relationships of the various phases to the overall project. Top and bottom dates are for CPFF and LS approaches, respectively. These dates, obtained from inspection of the combined diagrams, will be verified shortly by the computer.

After the addition of numbers and manpower requirements, the ORNL diagrams were individually run through the computer. These separate runs were then analyzed to identify the critical path through each of the diagrams and to determine manpower requirements.

Manpower leveling of the various ORNL phases was initiated. Manpower leveling is a schedule to make the manpower requirements as nearly constant as practical without jeopardizing the final completion date.

5.5 TRU Facility

B. F. Bottenfield, F. L. Hannon, G. B. Berry, R. A. Schmidt

The Title I Engineering Report for the design of the TRU Facility was issued in final form on July 16, 1962, and consisted of 112 A-E drawings, seven ORNL drawings and appropriate descriptions, criteria, equipment lists, and specifications. The cost estimate totaled \$8.7 million, of which about \$7,580,000 was for direct and indirect charges and about \$1,120,000 (15%) was contingency. The contract portion (building) of the estimate totaled about \$4,010,000, and the UCNC participation amounts to about \$3,950,000, both of which estimates include engineering and contingency.

Title II design is progressing on schedule and is above 25% complete. Seventeen Title II flowsheets and electrical drawings have been received. Completion of Title II engineering is scheduled for late December 1962.

Fig. 5.3. Schematic critical path schedule - TRU Facility.

6.0 MATERIALS EVALUATION STUDIES

G. A. West, A. B. Meservey

6.1 Radiation Damage to Protective Coatings

Radiation-damage tests are being made in order to determine the usable life of various protective coatings, plastic flooring materials and other materials of construction exposed to radiation in potential TRU-facility applications. Many coatings are exhibiting surprising tolerance to gamma radiation, and it is not yet possible to select the most outstanding types. Application methods and wear tests are also being conducted. Amerplate, a vinyl flooring material (plasticized vinyl sheeting) appears unchanged after more than one month's exposure to heavy traffic and janitorial cleaning.

Twelve epoxies, five phenolics, two polyesters, 13 vinyls, and 12 miscellaneous coatings and materials were exposed to a gamma flux of about 1×10^6 rads/hr in both air and deionized water in the ORNL Co⁶⁰ source at 40-50°C. They are listed below. Based on visual observations, the following coatings were superior and had about equal resistance to a gamma exposure of 2.0×10^9 rads in environments of air and deionized water. Tests will continue until all specimens fail.

Amercoat No. 74 surfacer with Fiberglas
and No. 66 seal coat, epoxy

Wisconsin Protective Coating Corporation

No. 7100, phenolic
No. 7122, phenolic
No. 7155, phenolic

Pittsburg Plate Glass Company

No. UL9647, epoxy
No. 12168, 100% solids, epoxy

Sika Chemical Company

Colma Protective Coating, epoxy
Colma Surface-Kote, epoxy

Carboline Company

No. 300, phenolic
No. 305, phenolic

Three vinyl coatings were surprisingly resistant to gamma exposures greater than 1×10^9 rads in air: Prufcoat P62-HSV, Prufcoat P50-HSV, and Amercoat 86-99.

6.2 Protective Coatings and Their Relative Ease of Decontamination

Thirty-one protective coatings on steel and concrete were tested for their tendency to fix surface contamination. The coatings were contacted with a solution of mixed fission products, pH 1 to 2, giving initial beta-gamma readings of about 3 r/hr at 2 in. after drying in air for 20 hr. Each specimen was then scrubbed with 25 strokes of a medium-bristle brush, using a mild detergent consisting of a 0.1% solution of "Tergitol" in H₂O, at room temperature. This in effect measures the amount of activity that is fixed on the surface of the coating and that requires a more stringent treatment for removal. The results of these tests are given below (Table 6.1) in terms of the decontamination factor obtained by the light scrubbing.

The Amercoat 66 epoxy system, not shown on the table, seems to be the most acceptable coating, so far. Though the DF is low (11), the radiation resistance is very good. It is this radiation resistance, especially in deionized water, that makes the coating preferable. U. S. Stoneware's R.O. 221 Primer-567-170 finish, which had a DF of more than 1000, can be ruled out on the basis of its poor radiation resistance in deionized water.

Table 6.1 List of Protective Coatings and Their Relative Ease of Decontamination

Coating	Decontamination Factor
Amercoat 74 primer - No. 74B surfacer	5
Amercoat 74B surfacer - No. 74 seal-white	11
Amercoat 1680 over Demitcote No. 3	2
Amercoat 86 primer, No. 99 white	65 ^a
Amercoat 74B surfacer, No. 74 seal gray	28
Amercoat 74 surfacer -Fiberglas No. 66 seal coat	11
Plasite 9009	10
Plasite 7155	4
Plasite 7122	30 ^a
Plasite 7100	3
Prufcoat P-50, H.S.V. finish	350 ^a
Prufcoat P-62, H.S.V. finish	400 ^a
Prufcoat Epoxy modified polyester	250 ^a
Pittsburg Plate Glass VC 9647 under UL8134 vinyl	100 ^a
Pittsburg Plate Glass 100% solids epoxy-VC-12168	100 ^a
Pittsburg Plate Glass VC-9647 high solids epoxy	12
Colma protective coating - 1 dip	9
Colma Surface Kote - 2 dips	25

Table 6.1 (continued)

Coating	Decontamination Factor
Carboline 1C Phenoline 300 finish	5
Carboline 3C Phenoline 368 finish	9
Carboline 2C Phenoline 305 finish	3
Carboline 3C Phenoline 2351-53	2
U. S. Stoneware, R.O. 221 Primer-TP-216 finish	>1000 ^a
U. S. Stoneware, R.O. 221 Primer-567-170 finish	100 ^a
U. S. Stoneware, No. 7	8
U. S. Stoneware, Tygorust Primer, 5B-312 finish	50 ^a
U. S. Stoneware, Tygorust Primer, AV-461 top	80 ^a
Wilbur and Williams No. 1712 primer-liquid glass finish	170
Wilbur and Williams - Epoxy polyimide under "Tile Coat"	10

^a Average of two tests.

7.0 TRU CORROSION STUDIES

J. L. English, J. C. Griess

Pertinent corrosion data for a number of proposed materials of construction for components in the transuranium separation facility are presented. The materials include Hastelloys B and C, niobium, tantalum, titanium, and Zircaloy-2.

Hastelloys B and C. Corrosion tests with welded specimens of Hastelloys B and C were completed in (1) oxygenated 3 M HCl at 43°C, (2) oxygenated 10 M LiCl-0.1 M HCl at 130°C, and (3) hydrogen peroxide-containing 10 M HCl-0.0023 M LaCl₃ at 35°C. Final penetration rates are summarized in Table 7.1. All solutions were replaced at the end of 100-hr intervals. With the exception of the latter environment, Hastelloy B exhibited resistance superior to that of Hastelloy C. Rates for the former ranged from 1.8 to 5.7 mils/month in all three environments. Hastelloy C was subject to heavy attack in the first two environments, with rates varying from 5.0 to 40.2 mils/month. In the 10 M HCl-0.0023 M LaCl₃-H₂O₂ environment, the alloy was equally resistant as Hastelloy B. In the three environments, all Hastelloy C specimens were susceptible to preferential attack in heat-affected zones adjacent to welds, with observed penetration ranging up to 30 mils. The use of a postweld heat treatment at 1227°C (2240°F) eliminated the heat-affected-zone attack without affecting the general corrosion resistance of the alloy. The only environment that produced heat-affected-zone attack in Hastelloy B was the boiling lithium chloride-hydrochloric acid solution. After 1000 hr, separation of the 1/8-in.-thick, solution-exposed specimen occurred at a weld-metal interface.

Table 7.1. Corrosion of Welded Hastelloys B and C by Simulated TRU Environments

Environment	Alloy Type	Temp (°C)	Test Time (hr)	Specimen Location	Penetration Rate (mils/month)
3 M HCl- O ₂	Hastelloy B	43	1000	Solution	5.1
				Vapor	4.3
	Hastelloy C	43	400	Solution	19.4
				Vapor	5.0
10 M LiCl-0.1 M HCl-O ₂	Hastelloy B	130	1000	Solution	1.8
				Vapor	2.3
	Hastelloy C	130	1000	Solution	22.0
				Vapor	40.2
10 M HCl- 0.0023 M LaCl ₃ - H ₂ O ₂	Hastelloy B	35	1000	Solution	5.7
				Vapor	4.9
	Hastelloy C	35	1000	Solution	5.9
				Vapor	5.4

Table 7.2: Effect of Ferric Ion on Corrosion of Welded Hastelloys B and C by Oxygenated 6 M HCl Solution at 43°C

Alloy Type	Test Time (hr)	Specimen Location	Penetration Rate (mils/month)	
			No Added Fe(III)	200 ppm of Added Fe(III)
Hastelloy B	300	Solution	5.3	17.5
		Vapor	6.8	7.2
Hastelloy C	400	Solution	10.2	22.9
		Vapor	6.5	8.5

The possibility of an acceleration in attack rates on Hastelloys B and C due to buildup of corrosion-product iron in a simulated process solution was investigated. An oxygenated 6 M HCl solution at 43°C was selected for the tests on welded specimens of the two alloys that were exposed to the solution and the vapor. A concentration of 200 ppm of ferric ions (added as ferric chloride) was selected. Test solutions were replaced at 100-hr intervals. Penetration rates for comparable time periods in tests with and without added ferric ions are given in Table 7.2. Weight losses and approximate metal-thickness losses are shown in Figs. 7.1 and 7.2. Solution-exposed Hastelloy B underwent an increase in attack rate from 5.3 mils/month in the iron-free solution to 17.5 in the iron-containing solution, during a 300-hr period. In the vapor-phase tests, rates were nearly identical in both tests. Attack was uniform on all specimens. After 400 hr, the solution-exposed Hastelloy C specimen in the iron-free solution was 10.2 mils/month, compared with a rate of 22.9 in the solution containing the added 200 ppm of ferric ions. There was no significant difference between the vapor-phase rates in the two tests. All specimens were attacked preferentially in heat-affected zones.

Niobium. A number of tests were carried out to determine the sensitivity of niobium to hydrogen embrittlement in various chloride-containing solutions. In one case, a solution-exposed specimen cracked during bending after 1000 hr at 105°C in oxygenated 6 M HCl solution. The hydrogen content of the specimen was 530 ppm, compared with 4 ppm in the unexposed material. The corrosion rate on the specimen was 0.7 mils/month. No fracture occurred during bending in a specimen that was exposed for 1000 hr at 130°C in oxygenated 10 M LiCl-0.5 M HCl solution, although the hydrogen content was 35 ppm. In simulated dissolution tests with niobium coupled to aluminum and exposed in oxygenated 6 M HCl at 105°C, considerable hydrogen pickup was encountered in the former, 3500 ppm after 6.5 hr of contact time. The use of platinum tackwelded to the niobium in a similar test failed to prevent embrittlement of the niobium although some reduction in the amount of hydrogen pickup was obtained.

Tantalum. Tantalum was completely resistant to attack in all environments tested. The maximum rate observed, slightly less than 0.1 mil/month, was encountered in a hydrogen peroxide-containing 10 M LiCl-0.1 M HCl solution at 130°C. Some evidence of hydrogen pickup was observed in simulated dissolution tests in hot 6 M HCl solution with tantalum in contact with aluminum. After contact periods of 1.5 and 7 hr, the hydrogen content in tantalum specimens was 54 and 64 ppm, respectively, compared with 1 to 2 ppm in unexposed material. Neither test specimen exhibited cracking during a bend test.

Titanium. Previous tests with titanium exposed in hot hydrochloric acid solutions resulted in catastrophic attack. The addition of a strong oxidant to the solution (such as nitric acid) produced considerable improvement in resistance. For example, after 96 hr at 105°C in 6 M HCl-10 M HNO₃ solution, the attack rate on an immersed specimen was 3 mils/month. The vapor-exposed specimen corroded at a rate less than 0.1 mil/month.

Fig. 7.1. Solution- and vapor-phase corrosion of welded Hastelloy B in oxygenated 6 M HCl at 43°C, as influenced by the presence of ferric ion.

Fig. 7.2. Solution- and vapor-phase corrosion of welded Hastelloy C in oxygenated 6 M HCl at 43°C, as influenced by the presence of ferric ion.

Other tests were run at 105°C in 6 M HCl solution to which hydrogen peroxide was added. The average concentration of the latter was less than 0.001 M. Corrosion attack on the titanium was extremely severe in all cases; immersed specimens underwent complete dissolution in a short time.

Zircaloy-2. Extended tests (700 to 1000 hr duration) were carried out with welded Zircaloy-2 to determine the effect of oxidant species on corrosion behavior in 10 M LiCl-0.1 M HCl solution at 130°C. In one test, no oxidant was added; a second test was oxygen aerated; and, hydrogen peroxide (about 0.001 M) was present in a third test. Solution and vapor rates were 0.3 mil/month or less in all environments with the higher rates occurring in the no-added-oxidant test. Attack on all specimens was uniform.

The susceptibility of Zircaloy-2 to hydrogen pickup was negligible after 7 hr of contact time with aluminum in 6 M HCl solution at 105°C.

8.0 ANALYTICAL DEVELOPMENT

F. L. Moore, D. A. Costanzo

8.1 Radiochemical Determination of Am^{241}

During the period just ended, considerable effort was directed to the study and application of alpha spectrometry for the determination of Am^{241} in the presence of Cm^{242} . A 256-channel nuclear-data analyzer in conjunction with a silicon diode detector was used for this study. Suitable standards and synthetic mixtures were prepared and various samples of process solutions tested. Because Pu^{238} and Am^{241} are not sufficiently resolved by alpha spectrometry, the Pu^{238} was removed by the standard TTA method (Anal. Chem. 29, 1767 (1957)). This is a useful step inasmuch as the plutonium content of these solutions is of interest also. The plutonium-free solution was then analyzed by alpha spectrometry. Process solutions of Cm^{242} containing Am^{241} in the range 0.3 to 1.4% (as determined by alpha radioactivity) have been analyzed. The uncertainty was $\pm 10\%$.

8.2 Absorption Spectrophotometric Studies

A Lucite glove box was fabricated to contain the end compartments of the Cary model 14-PM spectrophotometer that is to be used to resolve the complex spectra of the actinide elements. It is planned to use this installation for the spectrophotometric determination of americium in americium-curium product, the determination of oxidation states of americium in fluoride solutions and the determination of chloro-complexes of americium in LiCl solutions.

Micro absorption cells (total capacity, 100 to 1000 microliters) were designed to permit measurements on solutions containing beta-, gamma-, or neutron-emitting materials which in macro amounts would require experiments to be performed in the hot-cell facility due to their high specific activity.

Auxiliary equipment and accessories (to be obtained) will permit spectral measurements at controlled temperatures, 10 to 100°C, and at preset intervals over a 24-hr arranged program for kinetic studies. The wavelength range is 1860 to 26500 Å.

9.0 URANIUM²³² PREPARATION

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Five samples of U²³² were recovered and purified from two successive irradiations of 50 g of Pa²³¹. From the first short (10 hr) irradiations in the ORR 32.9 mg of U²³² was recovered, containing 127 to 311 ppm of U²³³ (Table 8.1), and from the second, longer (15.2 days) irradiation 1.04 g of U²³² was isolated, containing 0.72% U²³³. After the decay of the Pa²³³, the Pa²³¹ will be decontaminated and converted to Pa₂O₅ for return to the United Kingdom Atomic Energy Authority.

Table 8.1. Summary of U²³² Samples^a

Product	Pa ²³¹ Irradi- ated	Average Perturbed Flux (nvt)	Decay Period	Amount of U ²³² Predicted	Amount of U ²³² Recovered	Amount of U ²³³ Predicted	Amount of U ²³³ , by Mass. Anal.
U-1	7.35 g	4.7x10 ¹⁸	0-43 hr	4.81 mg	4.29 mg	223 ppm	206 ppm
U-2			43-91 hr	1.62 mg	1.51 mg	354 ppm	311 ppm
U-3	40.6 g	4.1x10 ¹⁸	0-43 hr	23.1 mg	21.6 mg	181 ppm	127 ppm
U-4			43-77 hr	6.17 mg	5.49 mg	290 ppm	204 ppm
U-5	45 g	1.25x10 ²⁰	8.2 day	1.12 g	1.04 g	1.75%	0.72%

^aAll samples were analyzed on the mass spectrometer for mass 231 (Pa). No detectable amount was found in any sample except U-4. Analysis of U-4 by radiochemical methods gave a value of less than 5% Pa²³¹.

The dissolution of the irradiated targets and primary isolation of the uranium was carried out according to the procedure previously described.⁶ The final purification of the uranium was achieved by adjusting the solution to 6 N HCl-0.4 N HF and then passing it through a small column of Dowex 1-X4. The uranium was eluted with dilute

HCl and evaporated almost to dryness and taken up in 6 N HNO₃. This nitrate solution was extracted with 20% TBP in diethylbenzene and stripped with water to obtain the final U²³² product as a nitrate solution. Considerable decontamination of the uranium was obtained in this solvent extraction cycle. The 1-g product, after elution from the resin column the second time, read greater than 100 r/hr at 4 in. The final solution, in the same volume of solution (600 ml), read 2 r/hr at 2 in.

Difficulty was encountered in the preparation of Pa₂O₅ for the second irradiation. The protactinium intermediate product was precipitated with NH₄OH, filtered, and ignited to 700°C. After approximately 10% of the oxide had been prepared, it was found that the gross volume of the powder was too large to fabricate again into cermet pellets similar to the first irradiation. Substitution of a urea precipitation for the NH₄OH yielded no better product, though ignition to 1000°C gave a somewhat more dense powder. Chemical analysis of the powder revealed that it contained approximately 13 wt % aluminum. By igniting to 1000°C and by using a smaller amount of aluminum metal in the pellets, the Pa₂O₅ was encapsulated for the second irradiation without removal of the Al₂O₃. The protactinium intermediate product from the second irradiation is being stored to allow the residual Pa²³³ to decay before any further processing.

The hot-cell equipment proved satisfactory for all three runs except for the heavy-duty model 8 manipulators, which required almost continuous maintenance. It was suspected that the polyethylene dissolver vessel may have received a damaging amount of radiation from the first two runs, and it was removed and replaced before the third run. This was done in the cell, manually, after all solutions had been removed. Radiation surveys made at this time indicated that no significant amount of protactinium remained in the cell either in the equipment or as surface contamination.

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