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TRANSURANIUM QUARTERLY PROGRESS REPORT
FOR PERIOD ENDING NOVEMBER 30, 1963

OAK RIDGE NATIONAL LABORATORY

operated by

UNION CARBIDE CORPORATION

for the

U.S. ATOMIC ENERGY COMMISSION

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CHEMICAL TECHNOLOGY DIVISION
METALS AND CERAMICS DIVISION
PLANT AND EQUIPMENT DIVISION
REACTOR CHEMISTRY DIVISION
PLANT ENGINEERING, ORGDP

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PERIOD ENDING NOVEMBER 30, 1963

Compiled by
W. D. Burch

MAY 1964

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
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U. S. ATOMIC ENERGY COMMISSION

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1. ABSTRACT

All research, development, and design work directed toward the eventual operation of the Transuranium Processing Plant for the separation and recovery of gram quantities of the very heavy actinide elements is reported in this series of quarterly progress reports. Process development work is well advanced and will be culminated within six months through operation of a full-scale developmental and production facility, the Curium Recovery Facility. Design and procurement of processing equipment for the TRU Plant and construction of the facility by the lump-sum contractor is proceeding on schedule.

Process Development. -- The principal effect of alpha radiation in Tramex feed is a loss of acid at the rate of about 0.1 mole per liter per day at activity levels of 10 w/liter. Since Tramex process separations are greatly affected by small changes in acidity, methods for controlling acid losses caused by radiolysis are necessary. As previously reported, the addition of methanol to Tramex feed is effective in preventing acid loss. Further studies of this method for controlling acidity were made, and the effects of methanol on the Tramex extraction process were determined. As little as 2 vol % CH_3OH effectively stabilized for two weeks a 10 M LiCl --0.05 M HCl solution containing 10 w of alpha activity per liter. Phase separation is more rapid with methanol present, and the use of methanol in extraction tests caused no gross change in distribution coefficients, extraction kinetics, or fission product behavior. The use of methanol to inhibit acid loss in high-activity-level Tramex feed appears to be completely satisfactory and will be used in the Curium Recovery Facility.

Alamine 336 is now available from the manufacturer as the hydrochloride salt in diethylbenzene (DEB). Laboratory evaluation of this extractant indicates greater purity, with lower concentrations of primary and secondary amines than in the Alamine 336 previously received.

Numerous small-scale laboratory tests were made to study the problem of formation of unstable, nitrated, organic compounds resulting from the use of Alamine 336 $\cdot\text{HNO}_3$ --DEB in the Tramex process. Such hazards could exist

if solvent were heated in the presence of nitrate. It was possible to initiate two types of undesirable reactions. However, extreme conditions were necessary, such as evaporation to dryness, introduction of a continuous spark in the vapor phase, and excessive amounts of solvent. Results indicate that it is probably safe to adjust Tramex feed to 10 M LiCl by evaporation even if some solvent is present, but precautions should be taken to make certain that no solvent is present if LiNO_3 or $\text{Al}(\text{NO}_3)_3$ solutions must be heated.

In a successful test of a Talspeak process flowsheet, recovery of Cm^{242} was 99.9% complete, and a suitable decontamination factor (1000) from fission products was achieved. The process consisted in extraction of the lanthanides with 0.3 M di(2-ethylhexyl)phosphoric acid (HDEHP) in diisopropylbenzene (DIPB) from 1 M lactic acid, 0.1 M ammonium diethylenetriaminepentaacetate $[(\text{NH}_4)_5\text{DTPA}]$ at pH 3.0, followed by extraction of the Cm^{242} by 0.1 M HDEHP in Amsco 125-82 at pH 1.5, and stripping of the Cm^{242} with 1 M HNO_3 . Previous difficulties in extracting curium caused by slow extraction were overcome by pH adjustment, which also increased the Cm^{242} distribution coefficient. Comparative tests of extractants, diluents, and aqueous complexing agents showed advantages of HDEHP (extractant), DIPB (diluent), and DTPA (aqueous complexant) for the group separation step, and of n-dodecane or Amsco 125-82 as the diluent in the actinide recovery step.

Methods of separating americium and curium from each other were investigated at high alpha-activity levels in order to determine the feasibility of separating gram quantities of Am^{241} from Cm^{242} or Am^{243} from Cm^{244} . Satisfactory separations were made in small-scale tests at activity levels greater than 100 w/liter, both by precipitation and by ion exchange. Precipitation of pentavalent americium as KAmO_2CO_3 from potassium carbonate solution produced product which contained 94 to 98% of the americium and carried only 2.0 to 3.8% of the curium. Chromatographic elution of trivalent americium and curium from either anion or cation exchange resins containing alpha activity equal to 500-1000 watts per liter of resin was tested with solutions containing methanol to inhibit radiolytic gas formation. Product cuts were obtained which contained greater than 90% of

the curium with 1 to 3% americium, and 85 to 87% of the americium with 1 to 2% curium. Methanol effectively inhibited excessive radiolytic gas formation and made the chromatographic elution possible.

The transportation and storage of gram quantities of Cm^{242} may be simplified if curium can be dissolved in mercury. An attempt to form curium amalgam by electrolysis from an anhydrous solution of CmCl_3 and LiCl in methanol was partially successful. About 50% of the curium 242 was deposited in the mercury pool; however, additional electrolysis did not appreciably increase curium transfer to the mercury.

Acceptable prototype HFIR targets have been made with 20 to 200 μ particles of PuO_2 which were formed by a hydroxide precipitation method; however, PuO_2 preparation by a sol-gel process would make it easier to recycle fines. Preliminary indications are that a satisfactory sol-gel process for PuO_2 preparation can be developed. In small-scale laboratory tests, hard, glassy oxide with a density of $(10.3 \pm 1)\text{g/cc}$ was prepared by a sol-gel method. Large-particle-size AmO_2 was also successfully prepared by a hydroxide precipitation method, but attempts to prepare americium sols by methods used for making plutonium sol were not successful.

Curium Recovery Facility. -- The Curium Recovery Facility (CRF) will be used to test chemical processes at full activity levels for the Trans-uranium Element Program, to recover gram amounts of Am^{243} and Cm^{244} for use in research, and to purify gram amounts of Cm^{242} and Cm^{244} for use in thermoelectric converters. This facility is being installed in cells 3 and 4 of the High Level Chemical Development Facility, Building 4507. At the present time, construction of the cell-4 complex is nearly complete, and the cell-3 complex is in the design stage.

The CRF operating group has been organized, and cell-4 cold testing and equipment calibration are in progress. The last phase of the "cold" testing program will be a complete demonstration at flowsheet conditions but without curium or fission products. The present schedule calls for tracer tests of the process to start in February.

Experimental Engineering Studies. -- Testing of the phosphonate flowsheet (to be used ultimately for separation of americium and curium

from the heavier actinides) in 3/4-in.-diam pulse columns was continued using tantalum sieve plates. Flooding rates were measured at several temperature and pulse-frequency combinations at equal organic-aqueous flow rates; throughputs achieved were 18 ml/min (each phase) at 40 cpm, 25°C; 20 ml/min at 60 cpm, 25°C; and 24 to 24 ml/min at 60 cpm, 50°C. The extraction stage heights measured with europium tracer ranged from 7.6 to 10.3 in. and were almost independent of the variables tested. Air introduced at 1 ml/min to simulate the effects of radiolytic gas had no effects on stage heights.

The full-scale cell mockup was used in continuing demonstrations of design concepts with actual equipment models. The equipment transfer case now appears operable after several minor adjustments and alterations were made. The in-cell handling necessary for replacement of an alpha window was demonstrated by using a 300-lb steel plate to simulate the inch-thick window. The new Central Research Laboratory model F extended-reach manipulators performed very well. The mounting arrangement for cubicle service-line disconnects, which permits make-up of the service line joint as the equipment rack is positioned in the cubicle, was demonstrated using an alignment-jig mockup. A mockup of the sampler is ready for testing.

Target Fabrication Equipment Development. -- Development of the automatic programming equipment for controlling the end-closure welding using tungsten-inert gas techniques is progressing very satisfactorily. A few sample welds made in a vendor's plant by the electron-beam method were examined, and we conclude that this process gives excellent results. The two systems will be compared in order to weigh the merits of each before the equipment is constructed.

The four prototype capsules under irradiation in the Engineering Test Reactor since October continue to perform as desired. The first of these will be removed for examination in April.

The equipment for the glove-box line was assembled and will be installed in the alpha facility in Building 3019 during the next quarter. Some of the prototype equipment for the TRU Facility will be tested in this line during the fabrication of the next set of irradiation capsules.

Although the overall concept of the process for the TRU Facility has not changed, there have been significant changes in the location of cell equipment and in the concept of certain individual items. The status of each is described. It is estimated that the total project on target fabrication is 47.5% completed. The increased design effort in the past quarter enabled the effort to remain on the normal critical path schedule.

Design Studies. -- Construction of the Transuranium Processing Plant, which was started July 1, is continuing on schedule. Work was concentrated on those portions of the facility which are below grade, in both the building and the related outside services. The floors of the cells and all structural steel foundations have been poured in preparation for the start of steel erection.

Procurement of ORNL-furnished materials and equipment remains on a schedule that makes the items available to the contractor at the time stated in the contract specifications.

Design effort for the chemical process equipment was concentrated on the in-cubicle equipment racks and the inter-pit piping system. Of the nine equipment racks presently planned, one is being built and six are in the check-print stage of design. Drawings of the inter-pit piping plugs and the waste and off-gas headers in the tank pits have also been issued for check-print review. The bulk of the Zircaloy-2 plate, tubing, and bar stock required for fabrication of the process equipment was ordered; it will be delivered about the first of the year.

Conceptual designs for waste disposal and decontamination facilities were completed and approved. Plastic waste canisters, 8 in. high, will be removed from the transfer-area cubicle and accumulated in a "throw-away" concrete disposal carrier located in the limited-access area. Facilities in the decontamination room will include a glove box sufficiently large to handle equipment racks.

Complete drawing and specification lists, containing about 970 drawings, were prepared. Equipment design was 47% complete on November 22.

Design work was started for the cell-3 equipment in the Building 4507 Transuranium Development Facility. This cell will contain a dissolver and the equipment in support of the completed solvent extraction equipment in cell 4.

Corrosion Studies. -- Additional tests showed that ferric ions in the range of 5 to 100 ppm increases the corrosion rate of welded Zircaloy-2 in 6 M HCl by a factor of only 2 to 3 above the normal rate of 0.2 mils/month. Previous concern about greatly accelerated attack in the presence of iron was shown to be unfounded.

Analytical Methods Development. -- Development of a radiochemical method for the determination of Bk²⁴⁹ is in progress. The method, based on a two-stage liquid extraction, appears promising. Studies of the use of arsenazo III for the spectrophotometric determination of the rare earths, thorium and zirconium were expanded to include measurement in a number of acid environments of the decomposition rate of the reagent induced by absorbed gamma irradiation.

2. INTRODUCTION

Gram quantities of transuranium elements for research uses are to be produced in the High Flux Isotope Reactor (HFIR) and Transuranium Processing Plant (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 Pu²⁴² and of a mixture of Am²⁴³ with Cm²⁴⁴. These feed materials are being produced by long-term irradiation of 10-kg batches of Pu²³⁹ in a Savannah River reactor. The irradiated Pu²³⁹ was processed at Savannah River to recover the Pu²⁴² as decontaminated PuO₂ and the americium-curium as a solution containing about 3 kg of rare earth fission products. These feed materials have been shipped to ORNL. The rare earths will be removed from the americium-

curium and the actinide oxides fabricated into HFIR targets in TRU. Following irradiation in HFIR, scheduled to begin late in 1965, 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 design, development and fabrication, HFIR target fabrication development, construction of the TRU Processing Plant, corrosion studies and analytical research and development. This work was performed by the Chemical Technology, Metals and Ceramics, Plant and Equipment, Analytical Chemistry, and Reactor Chemistry Divisions of Oak Ridge National Laboratory, and the General Engineering Department of the Oak Ridge Gaseous Diffusion Plant.

Previous reports in this series are:

1. For period ending February 28, 1962 - ORNL-3290.
2. For period ending August 31, 1962 - ORNL-3375.
3. For period ending November 30, 1962 - ORNL-3408.
4. For period ending February 28, 1963 - ORNL-3482.
5. For period ending August 31, 1963 - ORNL-3558.

3. PROCESS DEVELOPMENT

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3.1 Tramex Process Development

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In the Tramex process, separation of actinide and lanthanide elements is achieved by extraction of the actinides into tertiary amines from concentrated lithium chloride solutions at low acidities. Process conditions have been determined, and extensive tests have been made in laboratory mixer-settler runs. Development studies reported here were directed toward (1) further improvement and testing of the Tramex process and included the investigation of methanol as an additive for controlling acid loss in Tramex feed by radiolysis, (2) evaluation of purified Alamine 336 supplied by the vendor, and (3) determination of hazards resulting from heating Tramex solvents, especially in the presence of nitrate solutions.

3.1.1 Acid Control in Tramex Feed

Since Tramex process separations are greatly affected by small changes in acidity, methods for controlling acid losses caused by radiolysis have been investigated. Although effective acid control has been demonstrated with several different reducing agents added to high-activity-level Tramex feed, methanol was the most satisfactory. As previously reported,¹ with no reducing agent in high-activity-level Tramex feed, the G value for H^+ destruction was about 1.4, but, with 10 vol % CH_3OH present in 10 M LiCl containing 18 w of Cm^{242} per liter, the solution was stable even after 25 days. During the entire 25 days, the acid concentration was 0.03 to 0.06 M, which is the proper range for good extraction. Additional tests with a few milliliters of solution at high activity levels have been made.

The use of methanol to inhibit acid loss by radiolysis appears to be completely satisfactory and does not produce any detrimental side effects in the extraction process. A 10 M LiCl--0.05 M HCl solution containing 10 w of alpha activity per liter, can be stabilized for two weeks by adding as little as 2 vol % CH_3OH . Without methanol, the solution will become acid deficient, and metal hydroxides will precipitate within 10 hr. The G value for methanol depletion is about 0.3. Phase separation is more rapid with methanol present, and the use of methanol in extraction tests caused no gross change in distribution coefficients, extraction kinetics, or fission product behavior.

Distribution coefficients were determined for americium and HCl between 0.6 M Alamine 336·HCl in diethylbenzene (DEB) and feed that consisted of 11 M LiCl or 10 M LiCl--0.15 M $AlCl_3$ and 2, 5, or 10 vol % CH_3OH . Free acid was varied from 0.02 to 0.30 M. No adverse effects due to the presence of as much as 10% CH_3OH were noted. Americium distribution coefficients were not changed by the addition of 2% CH_3OH , but increases of about 25% were obtained by the addition of either 5 or 10% CH_3OH . Hydrochloric acid distribution coefficients appear to be constant below feed

¹W. D. Burch et al., Transuranium Quart. Progr. Rept. Aug. 31, 1963, ORNL-3558, pp 18-21.

acid concentrations of 0.15 M. Acid distribution coefficients for 11 M LiCl feed at low acidities were about 5.0, 4.4, and 4.0 for 2, 5, and 10 vol % CH₃OH, respectively. Scouting experiments made with europium tracer indicate that europium behavior parallels that of americium.

The effect of methanol on curium distribution coefficients was investigated with a Cm²⁴² solution (equivalent to 10 w/liter) stabilized with 2% CH₃OH. Distribution coefficients were determined on feed adjusted to 10 M LiCl--0.03 M HCl--2 vol % CH₃OH by extracting portions of the feed solution with 0.6 M Alamine 336·HCl-DEB at periodic intervals for eight days. Curium distribution coefficients remained almost constant over the eight-day period, and no detrimental effects were noted.

The effect of methanol addition on curium and fission product behavior was investigated in a mixer-settler run of the Tramex process at tracer activity levels. The flowsheet consisted of extracting 10 M LiCl--0.03 M HCl--5% CH₃OH feed at 50°C with 2 volumes of 0.6 M Alamine 336·HCl in DEB and scrubbing the solvent with 1 volume of 11 M LiCl--0.02 M HCl. Curium extraction was greater than 99.9%, and ruthenium and zirconium extractions were 99 and 80%, respectively. The extracted curium was stripped into 8 M HCl, and the strip product was scrubbed with 0.1 M Alamine 336·HNO₂--0.5 M Alamine 336·HCl. Zirconium and ruthenium remained quantitatively in the organic phase. The overall curium recovery was 99.9%, and fission products were undetectable in the product, corresponding to a decontamination factor greater than 1000. Curium and fission product behaviors were typical of tests made in which methanol was not used, and no hydraulic or kinetic problems were encountered.

Although acid loss by radiolysis and the role that methanol plays in counteracting this loss are not completely understood, plans are to use methanol in Tramex processing. No more laboratory studies are planned at this time. The next tests will be made during process runs in the Curium Recovery Facility.

3.1.2 Evaluation of Purified Alamine 336

Alamine 336 is now available from the manufacturer as the hydrochloride salt dissolved in diethylbenzene. This extractant appears to contain fewer

impurities than the Alamine 336 that was previously supplied. Americium distribution coefficients were determined with two samples of this new material diluted to 0.6 M Alamine 336·HCl-DEB. Identical results were obtained with the two samples, and americium distribution coefficients were 20% higher than for the original material. This increase in distribution coefficients indicates greater purity with lower concentrations of primary and secondary amines.²

3.1.3 Tramex Safety

Numerous small-scale laboratory tests were made to determine the possibility of fires and explosions resulting from the use of Alamine 336·HNO₃-DEB in the Tramex process. Such risks would exist if the solvent were heated in the presence of other nitrates. This could occur either by the accidental carry-over of solvent with product curium, which will be converted from chloride to nitrate solutions, or by recycle of nitrate feed that had been contacted with the extractant. The overall results indicate that it is probably safe to adjust Tramex feed to 10 M LiCl by evaporation even if some solvent is present, but precautions should be taken to make certain that no solvent is present if LiNO₃ or Al(NO₃)₃ solutions must be heated.

It was possible to initiate two types of undesirable reactions; however, at least two of the following extreme conditions were required: evaporation to dryness, introduction of a continuous spark in the vapor phase, and the presence of excessive amounts of solvent. By use of a continuous spark in the vapor phase, it was possible in some cases to ignite the vapors; this effect appeared to be simple combustion. The second type of reaction occurred only when the contents of the reaction flask had been distilled nearly to dryness. This reaction could be described as a mild explosion, characterized by a flash of light and a rapid increase in pot temperature. Only two sets of conditions produced an audible detonation, and in no case was the explosion sufficiently violent to rupture the glass reaction flask.

²W. D. Burch et al., Transuranium Quart. Progr. Rept. Aug. 31, 1963, ORNL-3558, p 24.

In these experiments 3.2 to 50 vol % of 0.6 M Alamine 336·HNO₃-DEB in the presence of concentrated HNO₃ and other additives was distilled to dryness. Solvent volumes varied from 1 to 15 ml, while total volumes did not exceed 31. The reaction vessel consisted of a 100-ml, three-neck, round-bottom flask fitted with a thermocouple well, platinum electrodes through which a continuous spark could be introduced, and a sparge tube through which air or other gases could be introduced. Sparking was provided by a high-frequency Tesla coil, and heat was supplied by a heating mantle.

The conditions and results of typical experiments performed are listed in Table 1. No indications of fire or explosion could be produced by distilling a mixture of Alamine 336·HNO₃-DEB and concentrated HNO₃ to dryness. Equal organic and aqueous volumes, the use of fuming HNO₃, the addition of H₂O₂, continuous air sparging, extended refluxing before distillation, and a continuous spark at the end of the condenser all proved nonhazardous in repeated tests, even though pot temperatures as high as 416°C were reached. A black residue resulted from taking these mixtures to dryness, and the residue could be ignited with a flame but would not support combustion when the flame was removed.

A very mild explosion or flash occurred in this system when a continuous electric spark was introduced in the vapor-phase during distillation. This reaction occurred only after the solution had been evaporated nearly to dryness (147 to 150°C). A more violent reaction was produced without air sparging than when air sparging was used, and argon sparging was not effective in preventing the reaction. The addition of 11 M LiCl was effective in reducing this explosion hazard. With the addition of 11 M LiCl (16 volume %), mild flashes produced by continuous sparking did not occur until a pot temperature of 265°C was reached. The addition of 3 M AlCl₃ or 10 M LiCl--0.1 M AlCl₃ (16 volume %) completely eliminated any reaction even though the mixture was evaporated to pot temperatures as high as 375°C. This indicates that AlCl₃ is more effective than LiCl for suppressing the reaction.

The addition of either LiNO₃ or Al(NO₃)₃ definitely increases the risk of explosive when Alamine 336·HNO₃-DEB is distilled in the presence

Table 1. Summary of Laboratory Tests of Alamine 336-DEB Fire and Explosion Hazards

Initial Pot Mixture										
0.06 M Alamine 336 [•] HNO ₃ -DEB (vol %)	16 M HNO ₃ (vol %)	Other Additives (vol %)	Spark- ing	Sparg- ing	Vapor Ignition	Pot Temperature at Vapor Ignition (°C)	Explosion at Dryness	Severity of Explosion	Pot Temperature at Explosion (°C)	Maximum ^a Pot Temp. Reached (°C)
17	83	---	No	None	No	---	No	---	---	395
50	50	---	No	None	No	---	No	---	---	290
17	83	---	No	Air	No	---	No	---	---	380
4	96	---	Yes	Air	No	---	Yes	Flash	145	345
4	96	---	Yes	Argon	No	---	Yes	Flash	150	350
4	96	---	Yes	None	No	---	Yes	Audible ^b detonation	145	---
3	81	11 M LiCl, 16	Yes	Air	No	---	Yes	Slight flash	265	370
3	81	11 M LiCl, 16	Yes	Argon	No	---	Yes	Slight flash	260	340
3	81	3 M Al(Cl) ₃ , 16	Yes	Air	No	---	No	---	---	375
3	81	11 M LiCl 0.1 M Al(Cl) ₃ , 16	Yes	Air	No	---	No	---	---	375
3	81	8 M LiNO ₃ , 16	Yes	Argon	Yes	84	Yes	Audible ^b detonation	255	---
3	81	8 M LiNO ₃ , 16	No	Air	No	---	Yes	Audible ^b detonation	250	---
17	---	8 M LiNO ₃ , 83	No	Air	No	---	No	---	---	375
3	81	2.6 M Al(NO ₃) ₃ , 16	Yes	None	Yes	85	No	---	---	375

Table 1. Summary of Laboratory Tests of Alamine 336-DEB Fire and Explosion Hazards
(Continued)

Initial Pot Mixture						Pot Temperature at Vapor Ignition (°C)	Explosion at Dryness	Severity of Explosion	Pot Temperature at Explosion (°C)	Maximum ^a Pot Temp. Reached (°C)
0.06 M Alamine 336· HNO ₃ -DEB (vol %)	16 M HNO ₃ (vol %)	Additives (vol %)	Sparking	Sparging	Vapor Ignition					
3 Alamine only	81	8 M LiNO ₃ , 16	No	Air	No	---	Yes	Flash	275	375
3 DEB only	81	8 M LiNO ₅ , 16	No	Air	No	---	Yes	Flash	280	385
3 DEB only	81	2.6 M Al(NO ₃) ₃ , 16	Yes	None	Yes	70	No	---	---	380
3 DEB only	81	2.6 M Al(NO ₃) ₃ , 16	No	None	No	---	No	---	---	380

^aThe temperature noted is either the temperature reached immediately following the explosion at dryness, if such occurred, or the maximum temperature to which the pot was heated in the effort to create explosive conditions.

^bBut not severe enough to break glass evaporation pot.

of HNO_3 . In the presence of 8 M LiNO_3 (16 vol %), an audible detonation occurred at 250°C , with sufficient force to blow all fittings out of the reaction flask. This explosion occurred with or without a continuous spark in the vapor phase, but, when the spark was used, multiple vapor-phase ignitions occurred, beginning at about 85°C . Multiple vapor-phase ignitions, at 85°C , were also promoted by $\text{Al}(\text{NO}_3)_3$, but there was no reaction upon heating to dryness in the absence of a spark.

When either Alamine 336 or DEB mixed with HNO_3 and LiNO_3 solution was distilled to dryness, a much less violent reaction occurred than when both Alamine 336 and DEB were present. Multiple vapor ignitions occurred with a continuous spark in the vapor phase in the case of DEB, but not when Alamine 336 was distilled.

Since methanol will be used in Tramex feed, the flammability of 2 to 10% CH_3OH in 11 M LiCl was investigated. These mixtures were distilled to dryness as in the above tests. Methanol vapors could not be ignited with a continuous spark in the vapor phase of the reaction flask either with or without an air sparge. However, a spark at the condenser outlet did ignite the methanol vapor. The vapor was ignited twice for brief intervals (2 to 3 sec) when the pot temperature reached 133°C .

3.2 TALSPEAK: Separation of Trivalent Actinides from Lanthanides by Organophosphorus Extraction from Aminopolyacetic Acid Solutions

Boyd Weaver

F. A. Kappelmann

R. D. Baybarz

Talspeak is a new method of separating the trivalent actinides from the lanthanides; it is being developed as a possible alternative to the Tramex process. The group-separation part of the Talspeak process consists of preferential extraction of the lanthanides and yttrium with a monoacidic organophosphate or phosphonate from an aqueous solution containing an aminopolyacetic acid (complexing agent). The preferred extractant is an aromatic hydrocarbon solution of di(2-ethylhexyl)phosphoric acid (HDEHP), and the preferred aqueous complexing agent is diethylenetriaminepentaacetic acid (DTPA) in a carboxylic acid medium. Satisfactory separations have also been made with 2-ethylhexylphenylphosphonic acid [$\text{HEH}(\phi\text{P})$]. The actinides are extracted from the aqueous raffinate with

a more highly concentrated solution of the extractant in an aliphatic diluent and are stripped from the extractant with dilute nitric acid.

This method is being developed for process use through the cooperative efforts of two groups. The first group has been engaged principally in studies of the extraction behavior of americium, curium, and fission products at tracer levels. The other group has been engaged principally in the testing of extraction flowsheets with multistage equipment at high levels of activity and in studies of the extraction of the heavier actinides, including californium and, occasionally, einsteinium and fermium. Results of the first study are being compiled into a topical report which will be issued later as ORNL-3559.

3.2.1 Kinetics: Rates of Extraction Vary with Conditions

In previously reported multistage tests of suggested flowsheets,³ excellent separations of the lanthanides from americium and curium were achieved, but extraction of the actinides from the raffinate was much poorer than predicted from earlier equilibrium extraction data. Subsequent tests confirmed the assumption that low rates of extraction were responsible for the ineffectiveness of the extraction. This is attributed to low rates of dissociation of the actinide-DTPA complexes. Mixing-chamber holdup times of about one minute in the mixer-settlers were insufficient to attain equilibrium.

In the recent tests, studies were made of the effects of extraction composition and concentration, diluent composition, and aqueous-phase composition on the rate of americium extraction. The tests consisted in determining apparent distribution coefficients in separatory-funnel "shake-outs," with contact times varying over the range of 30 sec to 60 min. In extractions by di(2-ethylhexyl)phosphoric acid (HDEHP) in diisopropylbenzene (DIPB) under conditions suitable for leaving americium in the aqueous phase while extracting the lanthanides, the small extraction of americium was apparently very rapid (Fig. 1). When extractability was

³W. D. Burch et al., Transuranium Quart. Progr. Rept. Aug. 31, 1963, ORNL-3558, pp 42-45.

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increased by increasing the concentration of HDEHP from 0.3 M to 1 M and/or changing the diluent to n-dodecane, the extraction rate became lower. Extractions by the stronger extractant 2-ethylhexylphenylphosphonic acid [HEH(Φ P)] were slow when the diluent was DIPB (Fig. 2), slower when it was n-dodecane, and still slower when the concentration of sodium diethylenetriaminepentaacetate (Na_5DTPA) was increased (Fig. 3). The time required to reach equilibrium was as long as 20 min in some cases. It should be noted that the increase in concentration of Na_5DTPA increased the pH of the aqueous phase, so that the rates of extraction shown in Fig. 3 may be influenced by the complexant concentration, the pH, or both.

Two methods were found for accelerating extraction, both involving increases in acidity of the aqueous phase. For example, in extractions by HEH(Φ P) in n-dodecane from solutions adjusted to 4 M in lactic acid, there were high coefficients and little, if any, delay in extraction (Fig. 4). However, a preferred method of increasing the acidity is by addition of nitric acid. When sufficient nitric acid was added to 1 M lactic acid, 0.1 M Na_5DTPA to reduce the pH to less than about 2.3, there was no observable delay in extraction by 1 M HDEHP in n-dodecane. The americium distribution coefficient was also increased to a maximum greater than 100 at pH about 1.5, (Fig. 5). Thus, it is possible and probably preferable to use the same reagent, HDEHP, in the Talspeak process for both the group separation and actinide recovery steps.

Temperature apparently had no effect on rates of extraction but did affect distribution coefficients. No differences in rates were observed in tests at 2°, 25°, and 50°C. Increasing the temperature from 25° to 50° decreased the distribution coefficient of europium by a factor of 3 but that of americium by a factor of only 2, thus decreasing the separation factor by one-third. Coefficients for both elements were 3 times as high as 2° as at 25°.

3.2.2 Successful Flowsheet Test

From batch test data obtained thus far, it is possible to propose several flowsheet arrangements of the Talspeak process that should give satisfactory operation. One of these was tested in multistage continuous

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Fig. 2. Increasing Extraction of Americium by HEH[ϕ P] in DIPB Decreases Rate of Extraction. Aqueous phase: 1 M lactic acid plus Na₅DTPA.

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Fig. 3. Increasing Extraction of Americium by HEH[ϕ P] in n-Dodecane Decreases Rate of Extraction.

Fig. 4. Increasing Concentration of Lactic Acid Increases Rate of Extraction of Americium by HEH[φP]. Aqueous phase: 4 M lactic acid, 0.05 M Na₅DTPA.

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Fig. 5. Extraction of Americium with HDEHP Is Maximum at pH about 1.5 and Nearly Independent of NaNO_3 Concentration. Extractant: 1 M HDEHP in n-dodecane. Aqueous phase: 1 M lactic acid, 0.09 M citric acid, 0.05 M Na_5DTPA , adjusted with HNO_3 .

countercurrent equipment, with excellent results. A schematic diagram of the experimental flowsheet is shown in Fig. 6. The feed material was a solution of irradiated americium containing curium and fission products. There were 16 mixer-settler stages in each of the three main steps: rare earth extraction, curium extraction, and curium stripping. The feed entered at the ninth stage in each case, and flowrates were adjusted to give a mixer-chamber holdup of about 1 min in each stage. Other operating conditions are shown in the figure. The circled numbers denote the relative flow volumes of feed, solvent, scrub, and strip solutions.

Results from the experiment were as follows: in the first cycle, 99.9% of the fission product rare earths were removed, with a Cm^{242} loss of less than 0.1%; in the second cycle, 99.96% of the Cm^{242} was extracted; and in the third cycle, 99.9% of the Cm^{242} was stripped. No fission products were detectable in the product, indicating a decontamination factor greater than 1000.

The small amount of Pu^{238} derived from decay of Cm^{242} remained in the raffinate when the americium and curium were extracted. Recovery of this plutonium would require an additional step.

3.2.3 Reagent Concentration Dependence Varies With Composition of Both Organic and Aqueous Phases

Extractions of lanthanides and trivalent actinides by monoacidic phosphates and phosphonates from aqueous solutions containing complexing agents were expected to be proportional to about the third power of the free reagent concentration, as they are in extractions from mineral acids. However, the reagent concentration power dependences in extractions of americium by HDEHP in DIPB varied between 2.6 and 2.9 (Fig. 7). Some extractions by $\text{HEH}(\Phi\text{P})$ had a much lower power dependence; for example, only 1.4 for extraction by $\text{HEH}(\Phi\text{P})$ in *n*-dodecane from 4 M lactic acid, 0.05 M Na_5DTPA (Fig. 8). In these cases, increasing the extractant concentration is not so effective in increasing distribution coefficients. It is evident that the experimentally determined reagent concentration dependence must be used in calculating the necessary phase ratios for extraction under a particular set of conditions.

Fig. 6. Experimental Talspeak Process Flowsheet.

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Fig. 7. Extractions by HDEHP in DIPB Are Proportional to Nearly the Third Power of the Extractant Concentration.

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Fig. 8. Some Extractions of Americium With HEH[ϕP] Have Very Low Reagent Concentration Dependencies.

3.2.4 Preferred Extractant Diluents: Diisopropylbenzene and *n*-Dodecane

Previous experiments have shown that aromatic diluents such as diisopropylbenzene (DIPB) are highly satisfactory for use in the actinide-lanthanide separation step of the Talspeak process, whereas aliphatic diluents such as *n*-dodecane are best in the actinide recovery step. Recently, additional extraction tests have been made with HEH(ϕ P) in a broad range of diluents, some of which would not be suitable for process use because of volatility or flammability. The results of the recent tests (Table 2) confirm those from previous experiments in showing the superiority of DIPB for actinide-lanthanide separation and the superiority of aliphatic diluents, such as Amsco 125-82 or *n*-dodecane, for actinide recovery.

3.2.5 Preferred Complexing Agent: Diethylenetriaminepentaacetic Acid

In further studies to determine the best conditions for separating the lanthanides from both the lighter and heavier elements of the actinide group, tests were made of the extraction of Ce, Eu, Am, and Cf by HEH(ϕ P) from solutions containing a variety of aminopolyacetic acids, including some different from those previously tested in extractions by HDEHP. The highest separation between the lanthanides and americium was obtained with DTPA (Table 3), but the separation factor between europium and californium was only 9. The complexant 1,2-cyclohexanediaminetetraacetic acid reversed the order of actinide extraction and was very unfavorable for separation of the lanthanides from americium.

Thus, as reported previously,⁴ the preferred method found thus far for separating the lanthanides from all the trivalent actinides is extraction by HDEHP in diisopropylbenzene from a solution of DTPA in lactic acid. This method gave separation factors of about 100 between europium and americium and about 20 between europium and californium.

3.2.6 Conclusions

Data from single-stage extraction experiments and a continuous multi-stage flowsheet test show that americium and curium can be separated

⁴W. D. Burch, et al., Transuranium Quart. Progr. Rept. Aug. 31, 1963, ORNL-3558, pp 42-45.

Table 2. Effect of Diluent on Separation Factors
between Europium and Americium

Extractant: HEH(ϕ P)

Diluent	Distribution Coefficients, E_a^0			Separation Factors, Eu/Am
	Eu	Ce	Am	
Benzene	1.3	1.4	0.016	80
Toluene	1.8	2.3	0.024	75
Xylene	2.6	3.8	0.029	91
Mesitylene	4.5	5.8	0.050	91
Diethylbenzene	5.9	8.4	0.062	95
Diisopropylbenzene	15.2	16.3	0.12	127
Triethylbenzene	10.3	14.3	0.096	107
Heptane	128	385	3.17	40
Cyclohexane	30	131	0.98	31
Decane	208	490	3.6	58
<u>n</u> -Dodecane	165	163	2.6	64
Amsco 125-82	190	243	3.6	52
Chloroform	0.12	0.24	0.0054	22
Carbon tetrachloride	0.16	0.12	0.06	2.6
<u>o</u> -Dichlorobenzene	0.94	0.82	0.014	67

Table 3. Diethylenetriaminepentaacetic Acid Is Preferred Complexing Agent for Separation of Lanthanides from Americium

Organic phase: 0.1 M HEH(Φ P) in DEB

Aqueous phase: 1 M lactic acid plus stated complexant

Complexing Acid			Distribution Coefficients, E_a^o				Separation Factors	
Name	Concentration (M)	pH	Eu	Ce	Am	Cf	RE/Am ^a	RE/Cf ^a
Iminodiacetic	0.1	2.5	1800	85	60	--	1.4	--
Hydroxyethyliminodiacetic	0.1	2.5	150	120	35	--	3.4	--
Ethyleneglycol bis(amino-ethylether)tetraacetic	0.05	2.5	920	14	34	277	<1	<1
Ethylenediamine di(o-hydroxy-phenylacetic)	0.01	2.4	1600	200	83	520	2.3	<1
Diaminoethylethertetraacetic	0.05	3.2	13	5.6	0.15	2.3	37	2.4
Hydroxyethylethylenediamine-triacetic	0.1	2.5	240	245	4	20	60	12
1,2-cyclohexanediaminetetraacetic	0.1	4.0	7.0	7.5	0.95	0.35	7	20
Diethylenetriaminepentaacetic	0.1	2.8	4.8	5.3	0.05	0.52	96	9.2
Triethylenetetraminehexaacetic	0.1	2.5	70	21	0.54	2.5	39	8.4

^aThe RE (rare earth) in each case is the less extractable of Eu and Ce.

effectively from the lanthanides and other fission products by the Talspeak process. Purification of californium by this process is less effective but feasible. Further work will include measurements of the separation of the actinides from contaminating elements other than fission products which are encountered in processing.

3.3 Americium-Curium Separations at High Alpha-Activity Levels by Precipitation and Ion Exchange

R. D. Baybarz

Methods of separating americium and curium from each other are being investigated at high alpha-activity levels in order to determine the feasibility of separating gram quantities of Am^{241} from Cm^{242} , or Am^{243} from Cm^{244} . Satisfactory separations have been made in small-scale tests at activity levels greater than 100 w/liter not only by precipitation of pentavalent americium as KAmO_2CO_3 from potassium carbonate solutions but also by chromatographic elution of trivalent americium and curium from ion exchange resins with solutions containing methanol to inhibit radiolytic gas formation.

3.3.1 Precipitation of KAmO_2CO_3

The precipitation of KAmO_2CO_3 from K_2CO_3 solutions was investigated at alpha-activity levels of 40 to 130 w/liter. It was found that americium can be satisfactorily oxidized at these activity levels. The precipitate contained 94 to 98% of the americium and carried from 2.0 to 3.3% of the curium (Table 4). Oxidation of americium with NaOCl proceeds rapidly at elevated temperature, and most of the americium precipitated within 15 min. Americium can also be oxidized to the pentavalent state with persulfate at temperatures of 80 to 90°C. A trace of silver was added to aid the oxidation; however, it does not seem to be required. In the presence of potassium, americium oxidation does not proceed beyond the 5+ state; but in sodium carbonate solutions some americium is oxidized to the soluble 6+ state. Curium in the filtrate precipitated after about 10 hr, presumably as a result of the complete depletion of acid due to radiolysis. The precipitated curium appeared to be the

Table 4. Americium-Curium Separation by KAmO_2CO_3 Precipitation

Conditions	Initial Conc. of Am^{241} (g/liter)	Initial Conc. of Cm^{242} (g/liter)	% Am pptd.	% Cm carried	Initial Alpha Activity Level in Feed (w/liter)
3 $\underline{\text{M}}$ K_2CO_3 --0.1 $\underline{\text{M}}$ NaOCl at 95°C for 1 hr	6.2	0.61	94	3.8	65
3 $\underline{\text{M}}$ K_2CO_3 --0.1 $\underline{\text{M}}$ NaOCl at 70°C for 2 hr	2.8	0.36	96	2.6	45
4 $\underline{\text{M}}$ K_2CO_3 --0.05 $\underline{\text{M}}$ $\text{K}_2\text{S}_2\text{O}_8$ at 90°C for 1 hr	1.3	0.31	98	2.0	40
4 $\underline{\text{M}}$ K_2CO_3 --0.05 $\underline{\text{M}}$ $\text{K}_2\text{S}_2\text{O}_8$, plus trace Ag, at 90°C for 3 hr	5.2	1.05	96	3.1	130

hydroxide since it was readily soluble in dilute acid, although the possibility of oxalate from the dimerization of the CO_2^- radical to form $\text{C}_2\text{O}_4^{2-}$ could not be ruled out.

3.3.2 Ion Exchange Separation

Since the addition of methanol to Tramex feeds was effective in greatly decreasing the radiolytic gas production, the procedure was applied to ion exchange separations of americium and curium. As a result, good americium and curium separations at high activity levels are possible.

Americium and curium were loaded on 10 ml of Dowex 1-8X (250 to 270 mesh) resin from 8 M LiNO_3 --20 vol % CH_3OH at an activity level of 150 w/liter. The methanol greatly inhibited the gassing of the solution, although it was not completely eliminated. Curium loaded on the column in a band at an activity level of 500 to 1000 w/liter. Although the resin became discolored within a few minutes, there was little gassing. Americium and curium were eluted with 3.6 M LiNO_3 --20 vol % CH_3OH , giving one product containing 94% of the curium with 3% of the americium, and another product containing 87% of the americium with 0.8% of the curium. Without the methanol to inhibit radiolytic gas formation, the column would have been inoperable because of large pockets of radiolytic gas.

Methanol was also used to decrease the radiolytic production of gas during americium-curium separation by elution from cation resin with 0.5 M NH_4 α -hydroxyisobutyrate--20 vol % CH_3OH at pH 4.1. At activity levels of 500 to 1000 watts per liter of resin, the column operated satisfactorily, with no disruption of the bands. One product contained 90% of the curium with 1% of americium, and the other contained 85% of the americium with 2% of the curium.

The use of methanol to inhibit radiolytic gas formation may make it possible to separate einsteinium and fermium from gram quantities of californium by chromatographic elution from cation exchange resin with α -hydroxyisobutyrate. This is of great importance since no other satisfactory method is available for this separation. The Cm^{242} alpha activity levels in the above mentioned laboratory runs correspond to a Cf^{252} loading of 1.5-2.0 g per liter of resin. However, larger-scale tests at the same activity levels are needed to prove the usefulness of this method.

3.4 Preparation of Curium Amalgam

R. D. Baybarz

The transportation and storage of gram quantities of Cm^{242} may be simplified if curium can be dissolved in mercury, since radiolytic gas formation would not be a problem, and the high thermal conductivity of mercury will facilitate heat removal. An attempt to form curium amalgam by electrolysis from an anhydrous solution of CmCl_3 and LiCl in methanol was partially successful. About 50% of the Cm^{242} was deposited in the mercury pool by the method of Onstott⁵ for electrolyzing lanthanides into a mercury cathode.

Curium was electrolyzed at a current density of 0.16 ma/cm^2 from 25 ml of CH_3OH that was $5 \times 10^{-3} \text{ M}$ in CmCl_3 , 0.1 M in LiCl , and about 0.1% H_2O . The anode was 5 cm^2 of platinum, and the cathode was a pool of mercury. Electrical connection to the cathode was made with a platinum wire sealed in glass. The methanol solution was refluxed during electrolysis to prevent concentration gradients. Analysis of the methanol solution indicated that 54% of the curium was transferred to the mercury within 30 min. Additional electrolysis did not appreciably increase the amount of curium deposited in the mercury. At the end of 4 hr, the curium content in the mercury was only 58%. The activity level in the starting feed was 30 w/liter, and, after electrolysis, the activity level in the mercury was 17 w/liter.

3.5 Preparation of Oxides for High-Flux Isotope Reactor (HFIR) Targets

M. H. Lloyd

HFIR targets made from pressed pellets of aluminum powder and actinide oxide must have a continuous aluminum phase for satisfactory heat transfer during irradiation. Acceptable prototype targets have been made with PuO_2 particles between 20 and 200 μ in diameter; however, with smaller particles, the oxide phase becomes continuous. This oxide was prepared by precipitating $\text{Pu}(\text{OH})_4$, washing, drying, calcining to the oxide at 1150°C , and

⁵E. I. Onstott, J. Inorg. Chem. 2, 967 (1963).

grinding. About 25% of the oxide was less than 20 μ in diameter and could not be used. Recycle is a problem since either dried hydroxide or high-fired oxide is difficult to dissolve, even in boiling concentrated nitric acid containing HF catalyst.

The preparation of PuO_2 by a sol-gel method would simplify the recycle problem because the dried gel can be sized before firing at 1150°C and because the fines generated by grinding the gel are readily dispersible in water. Preliminary indications are that a satisfactory sol-gel process for the preparation of dense PuO_2 can be developed. Large-particle-size AmO_2 was also successfully prepared by the hydroxide precipitation method, but attempts to prepare an americium sol by the methods used for plutonium sol preparation were not successful.

3.5.1 PuO_2 Sol Preparation

Apparent plutonium sols were prepared by digesting freshly precipitated and washed $\text{Pu}(\text{OH})_4$ at 60°C for 8 hr at a pH of 1.5. The highest pH at which complete dispersion occurred, regardless of digestion time or temperature, was 1.5. This pH may be a function of batch size and plutonium concentration, as well as of hydrogen ion concentration. Solids produced upon drying such sols at 120°C were readily dispersible in water; however, they are soft and friable and appear to be quite porous. When this gel was redispersed in water, the pH of the sol was 1.5. Calcination of the gel produced a porous friable PuO_2 product which does not appear to be suitable for HFIR target preparation. However, greatly improved product was obtained by heating the dried gel at 160°C for 2 hr, redispersing as a sol in water, drying to the gel, and calcining. The pH of the sol formed by resuspending the original gel after heating at 160°C for 2 hr was 1.8. Calcination of this gel to the oxide at 1150°C produced a hard glassy product with a density of 10.38 g/cc ($\pm 10\%$). Such material should be suitable for HFIR target preparation.

An attempt was made to determine the highest temperature at which a dried gel could be heated and still be redispersed in water; however, it appears that both temperature and heating time are critical. A dried gel

heated at 220°C for 1.5 hr was dispersible in water, but a subsequent batch heated at 220°C for 3 hr could not be redispersed.

3.5.2 AmO₂ Preparation

Americium oxide (AmO₂) of suitable particle size was successfully prepared by the hydroxide precipitation method used to prepare PuO₂. In this method, americium was precipitated from nitric acid solution with an excess of NH₄OH. The precipitate was very fine, but it could be successfully filtered with a medium-porosity glass frit. Following washing, the hydroxide was allowed to air dry for several days. Small, hard particles formed during the drying process. After calcination at 600°C to the oxide, nearly all particles were between 20 and 200 μ in diameter. It is not known at this time whether the formation of particles of the desired size without grinding is typical of this preparation or simply fortuitous. The density of this product was 10.0 g/cc (±10%). Attempts to disperse americium hydroxide as a sol by the procedures outlined for plutonium sol preparation were not successful. Other possible methods of sol preparation have not yet been investigated.

4. CURIUM RECOVERY FACILITY

V. C. A. Vaughen	R. L. Jolley
J. E. Bigelow	W. T. McDuffee
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The Curium Recovery Facility (CRF) will be used to test chemical processes (primarily TRAMEX) at full activity levels for the Transuranium program, to recover gram amounts of Am²⁴³ and Cm²⁴⁴ for use in research, and to purify gram amounts of Cm²⁴² and Cm²⁴⁴ for use in thermoelectric generators. This facility is being installed in cells 3 and 4 of the High Level Chemical Development Facility, Building 4507. At present, construction of the cell-4 complex is nearly complete. The cell-3 complex is in the design stage.

Curium Recovery Facility operations are to be divided between cells 3 and 4 as follows. Cell 4 provides one cycle of solvent extraction with the capability of converting feeds from nitrate solution to chloride

solutions, separating actinides and lanthanides by the Tramex process, and separating americium and curium from californium and the higher actinides. Cell 4 also provides equipment for converting the product from chloride solution to nitrate solutions and for sample removal. Equipment in cell 3 will provide for loading irradiated targets from transport carriers, dissolving targets in acid or caustic, removing product for shipment, and making special separations such as americium from curium. The processing will be done remotely in the shielded cells. The operating panel is located in the front of the cell windows on the ground level; cold-solution makeup and additions will be made at the second-level balcony in back of the cell block; equipment, samples, and other materials will enter and leave through glove boxes located on top of the cells. An underground tank outside of Building 4507 will be used to store a nitric acid feed solution containing Am^{243} , Cm^{244} , and rare earth fission products.

During the last quarter, a major effort in the process development program was the "cold" testing of the Curium Recovery Facility. The CRF operating group, organized September 1, 1963, consists of a supervisor, a data analyst, three operating shifts of one shift supervisor and two technicians each, and an alternate shift supervisor. Formal orientation, completed the first week, included a discussion of the program objectives, chemical and equipment flowsheets, and safety. Other major phases of the cold testing that are now in progress are: comparing locations of components as actually installed with locations as shown on the equipment flowsheets, cleaning of all lines and equipment by flushing with water, calibrating tanks and pumps, and determining the operability of every piece of equipment. Operating procedures are also being written. The last phase of the cold testing program will be a complete demonstration at flowsheet conditions but without curium or fission products.

Checkout of most of the equipment in cell 4, the operating area, and the makeup area is complete except for the mixer-settler rack. This rack is being modified, and the mixing impellers are being fabricated. Equipment in the roof area for sample removal and equipment removal and installation must still be tested.

During the cold testing, repairs and changes are made to the equipment as required. Most of these have been relatively minor. However, extensive repairs of the polyethylene lines, especially where the lines have been welded to the disconnects, have been necessary. There have been some difficulties with several pumps, and at present it is not possible to maintain a uniform flow rate for solutions pumped from the makeup area into cell 4. This appears to be the result of air blocking and can probably be corrected with surge pots and spring-loaded check valves in the transfer lines.

The present schedule calls for tracer tests of the flowsheets to start in February.

5. EXPERIMENTAL ENGINEERING STUDIES

P. A. Haas A. D. Ryon

Experimental studies are being conducted to develop equipment and procedures and to obtain design information for the TRU processing facility cells. This includes testing of the chemical-processing-cell equipment and of the transfer, shielding, and manipulator concepts of all the cells. The principal studies in progress include pulsed columns, equipment transfer, pumps, disconnects, and cell lighting and window maintenance.

5.1 Pulsed Column Studies

F. L. Daley

The phosphonate process, for the separation of the transplutonium elements into an americium-curium fraction and a transcurium fraction, is being tested in the 3/4-in.-ID glass pulsed-column complex. The column has an active length of 48 in. and contains tantalum sieve plates (1/32-in. holes, 5% free area, spaced 1/4-in. apart). Europium tracer, a standin for californium, is extracted from 1.3 M HCl by 1 M 2EH(ϕ P)A in diethylbenzene (DEB) and stripped into 3 M HCl, at flow ratios of 2:2:1 (feed:organic:strip). Physical properties of the solutions are given in Table 5. The pulse amplitude is set at 0.2 in. for all tests.

Table 5. Physical Properties of Phosphonate Solutions at 25°C

	Density (g/cc)	Viscosity (centipoise)	Surface Tension (dynes/cm)	Interfacial Tension (dynes/cm)
1 <u>M</u> 2EH(ϕ P)A in DEB	0.917	1.80	30.9	
1.3 <u>M</u> HCl	1.020	1.02	49.8	18.4
3.0 <u>M</u> HCl	1.052	1.09	51.9	18.4

Table 6. Run Data

Temp. (°C)	Pulse Frequency (cpm)		Flow Rates (ml/min) Feed-Organic-Strip	Stage Height (in.)	
	Extract.	Strip		Extract.	Strip
25	40	40	10 - 10 - 5	< 10.0	12.1
25	45	40	20 - 20 - 10	8.3	16.0
25	45	-	20 - 20	10.3	-
25 ^a	60	-	10 - 10	10.0	-
25 ^a	60	-	10 - 10	10.2	-
50	40	40	20 - 20 - 10	7.6	9.6
50	40	40	20 - 20 - 10	9.0	11.7

^aAir introduced into column during run.

Flooding studies on the phosphonate process, at 25°C, indicate a slight dependence on pulse frequency. The throughput is 18 ml/min for each phase at 40 cpm, and 20 to 20 ml/min at 60 cpm. Increasing the column temperature to 50°C at 60 cpm increases the throughput to 24 ml/min (about 17%) but has a lesser effect at 40 cpm - an increase of only 10% to 20 ml/min.

Batch phase-breaking tests for the phosphonate system show the relatively easier problem with phase separation. At an aqueous/organic ratio of 1, and with the aqueous phase continuous at 25°C, a break time of 40 sec was observed, compared with 150 sec for the Tramex system.

Extraction stage heights measured with europium tracer (Table 6) showed almost no dependence on the variables tested, which were pulse frequencies of 40 and 60 cpm, throughputs of 10-10-5 and 20-20-10 ml/min, and temperatures of 25 and 50°C. The stage heights measured ranged from 7.6 to 10.3 in. Stripping-stage heights increased from 12 to 16 in. at 25°C as the throughput was doubled, but decreased to 10 in. at 50°C at the highest throughput.

Air, at 1 ml/min, introduced into the bottom of the column during operation to simulate the effect of release of radiolytic gas (calculated rate, 0.5 ml/min from Cm^{242} at 10 w/liter) had no effect on the extraction stage height (10 in.).

5.2 Cell Mockup Studies

T. S. Mackey

Test operation and adjustment of the equipment transfer case, which provides complete containment during equipment transfers into and out of the cell, are nearing completion. The adjustments include alignment and pinning of the door-guide plates, proper shimming of the door brackets, remachining galled surfaces in the hoist, the installation of a two-lead worm gear in the drive section of the hoist, the addition of positioning screws for the hoist drum, and adjustments of the transfer case door-lock drives and cam plate. After proper door position was obtained, a leaktight seal against 1-1/2 in. of water was accomplished. The final

model will use a 1/32-in. metal bead on the seal surface to improve the seal on the flat rubber gaskets. The 1/16-in. solid-rubber window gaskets were replaced with 1/8-in. sponge-rubber gaskets to reduce leakage; following replacement, a leak rate of 0.8 liter of air per minute at 0.3 in. of water pressure was measured.

The equipment-transfer-case hoist functioned satisfactorily after the above changes were made. Equipment racks with a load of about 400 lb were drawn into position in the transfer case and then reinstalled successfully. The right-side equipment rack was removed and installed in the back rack position, and vice versa, demonstrating the feasibility of the transfer system. Some difficulty in starting the equipment rack into the transfer case was noted, and the design was changed to provide a guide with additional flare to permit easier entrance. An idler pulley will be installed on the hoist mechanism to increase the cable length leading to the level-wind mechanism that places the cable on the hoist drum.

Preliminary tests to demonstrate the feasibility of removing the alpha-seal window into the cubicle were completed. A heavy steel plate weighing 300 lb, which simulated the window, was handled with the transfer-case hoist and the cell manipulators. The plate was moved from its rest position with a small block and tackle, operated by the manipulators, to a new position for disassembly and then to another for bagging. Refinements were made in the canvas bag design and the mechanism for moving the window from its original position. The tests will be completed with a window mockup furnished by the window manufacturer.

The new Central Research Laboratory model F, heavy duty, extended-reach manipulators performed very well. The locking feature proved very helpful in handling the transfer of equipment racks. This feature enables one operator to perform the work that would otherwise require two.

The feasibility of making up all the service-line disconnects simultaneously as the equipment rack is positioned on the tracks was demonstrated by using an alignment-jig mockup installed on the back equipment rack track. The male disconnects with a short length of 1/2 in. tubing

and a 90° bend were clamped into a split manifold bolted to the equipment rack. The disconnects were then aligned to the jig. Perfect registration of the disconnects with the clamps on the side rack was made when the rack was moved from the jig to the side rack.

The feasibility of installing and removing jumper lines from the hot disconnect well to the equipment rack in the presently proposed configurations was demonstrated. A mockup of the tube bundle with the hot disconnect well was completed and installed in the mockup. A mockup of the TRU sampler was completed and is ready for testing.

6. DEVELOPMENT OF A FABRICATION PROCESS AND REMOTE FABRICATION EQUIPMENT FOR HIGH FLUX ISOTOPE REACTOR TARGET ELEMENTS

D. A. Douglas, Jr.

6.1 Laboratory Research and Development: Welding of Target End Closures

C. H. Wodtke

Work in the Metals and Ceramics Division falls under four broad categories: (1) development of necessary fabrication processes to produce an HFIR target rod, (2) irradiation of targets to test the adequacy of the design and the fabrication processes, (3) design of the required fabrication equipment and operation of a glove-box production line to make plutonium-bearing targets for the initial reactor loadings, and (4) the design of equipment for the remote fabrication of targets in three cubicles of the TRU facility. Progress in these areas for the last quarter is reported below.

Work is continuing in an effort to develop procedures and set up automated tungsten-arc welding equipment for producing end-closure welds in the aluminum targets. A high-speed-of-response Sanborn recorder and a precise program control unit for the welding machine were obtained, and work is in progress on hookup and operational checks of these units. Indications are that the welding-machine-program control will prove quite satisfactory for automatic control of the many steps comprising the welding cycle.

Samples have been machined, and test welds will be made to determine the consistency of weld quality and the mechanical properties of the most promising joint configurations previously reported.⁶

Electron-beam welding also is being explored as an alternative method for making target closures. The process is attractive for several reasons: (1) The weld-metal-blowout problem should be eliminated since the weld is made in a vacuum. (2) The high depth-to-width ratio characteristic of electron-beam welds increases the probability of routinely attaining satisfactory weld penetration. (3) The high concentration of heat produced by the electron beam minimizes heating of the adjacent target wall. One major disadvantage arises because of the vacuum welding; the subsequent helium leak check to ensure weld integrity is no longer feasible. Because the preliminary test welds described below were of very good quality, it is believed that the electron-beam welding method warrants further concentrated work in this investigation.

Arrangements were made to have test welds made in the laboratory of Sciaky Brothers, Inc., Chicago, Illinois, since suitable electron-beam welding equipment is not available here.

Joints of three configurations were prepared from type X-8001 aluminum and then welded. These were: (1) butt joint, with alignment sleeve, for complete penetration butt weld; (2) lap joint for square-groove weld; and (3) lap joint for fillet weld.

Typical butt joints of type (1) are shown in Fig. 9, before assembly and after welding. Contour and cleanness of the weld surface are very good. Plug shape and the alignment sleeve are shown in the longitudinal sections in Fig. 10a. Figure 10b shows that the weld is sound, has good contour on the root side, and that the objective of complete penetration through the tube wall was achieved.

Typical lap joints for square-groove welds (type 2) are shown in Fig. 11, before assembly and after welding. Again, the surface conditions

⁶W. D. Burch et al., Transuranium Quart. Progr. Rept. Aug. 31, 1963, ORNL-3558, pp 63-66.

Fig. 9. Typical Butt Joints of Type 1 Before Assembly and After Welding. The welds have good contour and are very clean.

Fig. 10. Cross Sections of Type-1 Weld, Showing:
(a) Plug Shape and Alignment Sleeve, and
(b) Enlarged View of Weld Area, Showing Complete Penetration.

Fig. 11. Typical Lap Joints for Square Groove Welds (Type 2).
A good surface condition is again evident.

are very good. The cross section shown in Fig. 12 shows very good weld contour and penetration. Although small porosity is present, the incomplete fusion on the root so frequently found in tungsten-arc welds is absent.

A section of a typical fillet welded lap joint (type 3) is shown in Fig. 13. It also is free of root defects such as incomplete fusion.

6.2 Irradiation of Target Prototypes

6.2.1 Fabrication of Irradiation Prototypes (J. D. Sease and D. M. Hewette)

A report⁷ describing the fabrication procedures and preirradiation data of the HFIR target prototype elements currently being irradiated in the Engineering Test Reactor (ETR) is in press.

The prototype elements differ from the proposed HFIR targets in length and amount of fissionable material loading. The prototype is 24.5 in. long and contains an active fuel length of 14 1/4 in., while the reference HFIR target element is 35 in. long and contains a fuel column 20 in. long. The 10 g of Pu^{239} loaded in each element corresponds to a 14% loading of PuO_2 .

For the irradiation experiment, six elements were fabricated; four are being irradiated in the ETR. The procedure employed to make the prototype elements was identical to the proposed HFIR target fabrication procedure and consists of (1) pressing of aluminum-jacketed PuO_2 -Al dispersion pellets, (2) loading the pellets into the fuel rod, (3) end-cap welding, (4) hydrostatic collapse, and (5) attachment of hex can.

6.2.2 Status of the Irradiation Experiment (S. A. Rabin, S. D. Clinton, and J. W. Ullmann)

The four HFIR target prototype irradiation rods⁸ were inserted in the ETR on October 28, 1963, and have performed satisfactorily to date. It

⁷J. D. Sease and D. M. Hewette II, Fabrication and Preirradiation Data for HFIR Prototype Target Rods, (to be published).

⁸W. D. Burch et al., Transuranium Quart. Progr. Rept. Aug. 31, 1963, ORNL-3558, pp 64-71.

Fig. 12. Cross Section of Type-2 Weld, Showing Good Weld Contour and Penetration. Some very slight porosity is evident in the weld.

Fig. 13. Cross Section of Fillet-Welded Lap Joint (Type 3). The weld is sound and free of root defects.

had been intended to irradiate the rods in the Materials Testing Reactor because it has a higher load factor than the ETR, but, a scheduling problem was encountered, and the capsules were then placed in the ETR. A compensating factor is that the higher coolant pressure in the ETR allows for a higher surface temperature and, therefore, provides a higher safety factor; in addition, the higher pressure is more comparable to HFIR conditions.

Two of the capsules are operating at an advertised maximum, unperturbed thermal-neutron flux of 1.8×10^{14} neutrons/cm²-sec, and the other two are operating at 1.6×10^{14} . It is planned to remove one of the capsules during the April 13, 1964, shutdown. It is estimated that this capsule will have achieved a total integrated flux of 1.2×10^{21} neutrons/cm² and a burnup of about 40% of the plutonium atoms. The postirradiation examination is scheduled for June 1964.

6.3 Development of Equipment for Fabricating Plutonium Targets

J. D. Sease and A. L. Lotts

Testing of the plutonium target fabrication equipment was completed, and the equipment is almost ready for operation. Metallographic equipment is being procured for incorporation as an integral part of the plutonium target fabrication line. The following is a summary of the status of the major components of the process equipment.

6.3.1 Glove Boxes

The glove boxes necessary to house the fabrication equipment are ready for operation. A 1-in.-thick steel plate was placed in the bottom of the weighing box to minimize vibration.

6.3.2 Powder-Preparation Equipment

The blender-dispenser and the prototype pellet-loading scale developed for the TRU facility (Section 6.4) have been incorporated in the target fabrication line. The use of this equipment in plutonium target fabrication should be a good demonstration of its reliability. Slight modification of the controls are being made so that the blender-dispenser and prototype scale will be more amenable to glove-box operation.

6.3.3 Pellet-Preparation Equipment

A bypass pressure-regulating valve has been installed on the hydraulic pump used with the pellet press. This bypass valve allows accurate control and regulation of the pellet pressing pressure.

6.3.4 Tube-Closure Equipment

Efforts have been concentrated on refining the equipment and procedures in order to increase the reliability of the welding procedure. Equipment refinement includes a torch positioner that allows positive torch adjustment in three planes. A collet "chill" block, which also serves as the primary rod-rotating mechanism, has been developed. With the collet chill block, the total indicated "runout" of the rod during rotation is less than 0.002 in. To obtain better rod-rotating-speed control, a variable-speed direct-current motor has been incorporated as the rod rotating mechanism.

The excellent closure welds obtained on the prototype irradiation specimens that were welded with a reduced pressure inside the tube⁹ led to an investigation of the effect of a reduced pressure inside the tube on the first closure weld. Welds were made on 15 tubes after "pulling" a static vacuum on the tube after the swaging operation. It was noted that a vacuum of about 10^{-3} to 10^{-4} torr could be easily obtained, and no loss in vacuum was seen, as measured with a mercury manometer. Radiography of welds made by this technique revealed almost no porosity in all cases. The welding procedure now produces welds, about 90% of which are acceptable, and which have the characteristics of very slight porosity, leak-tightness, and good penetration.

6.3.5 Hydrostatic Collapse

No change was made in the hydrostatic collapse apparatus since successful operation had been previously demonstrated during fabrication of the irradiation prototype elements.

⁹W. D. Burch et al., Transuranium Quart. Progr. Rept. Aug. 31, 1963, ORNL-3558, p 71.

6.3.6 Helium Leak-Test Equipment

An automatic filler for the liquid-nitrogen cold trap on the helium leak detector was obtained. The automatic filler will allow continuous operation of the leak-detection system.

6.3.7 Metallographic Equipment

Metallographic equipment is being obtained to characterize the microstructures of the as-fabricated plutonium target pellets. The PuO_2 composition and distribution, which can be correlated to thermal conductivity, can be readily determined by metallography.

This equipment will be housed in two alpha-tight glove boxes: one, a standard 6-ft box; the other a specially designed examination box that will house a small bench metallograph. The sample-preparation equipment, which will be contained in the 6-ft box, will consist of a small cutoff saw, speed lapping machine, and a vibratory polisher. The metallograph will be equipped with a 35-mm camera. Currently, only the metallograph has been obtained.

6.4 Development of Equipment for Fabricating Targets in the TRU Facility

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

The basic criteria established for the target-element fabrication process have changed little from that outlined in the first quarterly report.¹⁰ The status of this work is shown in Table 7. Some equipment concepts, however, have changed and equipment has been added, deleted, or relocated as indicated by fabrication process development.

¹⁰ D. E. Ferguson, et al., Transuranium Quart. Progr. Rept. Feb. 28, 1962, ORNL-3290, p 36.

Table 7. Degree of Completion (%) of the Design and Fabrication of Equipment

Cubicle No.	Conceptual Design	Detailed Design	Equipment Fabrication
3	88	68	48
2	74	50	30
1	45	15	0
Overall percentage	69	45	26
Project percentage, 47.5			

6.4.1 Cubicle 3

The fabrication flow diagram for cubicle 3 has not changed, but equipment is being added to improve the powder preparation and pelletizing operations. The equipment locations were changed slightly to provide wall mounting of the die and batch scales. The ultrasonic cleaner for the pellets will be relocated in the left-rear corner, and a more compact thermal dryer is being designed.

Actinide Calciner. -- A round, top-loading muffle furnace was ordered. The unit is about 15 in. in diameter and 19 in. high and has a cavity 5 in. in diameter and 6 in. deep. The furnace (which is rated at 1.4 kw) is capable of reaching a temperature of 1850°F.

Aluminum-Powder-Addition Station. -- A conceptual design was approved for the aluminum-powder-addition station, and detailed design is under way. This device will consist of a vibrating mount to accept the blender container, and a glass separatory funnel with an enlarged opening in the stop cock. The funnel will be charged with the required weight of aluminum powder outside the cell and then transported, via the intercell conveyor, to cell 3.

Blender-Dispenser. -- Fabrication and bench testing of the blender-dispenser is complete (Fig. 14). This unit was bench tested jointly with the prototype die scale and will be discussed under the section on the die scale.

Fig. 14. Blender-Dispenser and Die Scale Bench Test, Transuranium Processing Plant.

Blender-Dispenser Elevating Mechanism. -- Detailed design of the blender-dispenser elevating mechanism is complete and ready for final approval. This device is required to support and properly position the elevation of the blender container discharge nozzle over the die scale platform, and to raise the blender-dispenser sufficiently to attach the container.

Pellet-Load Scale. -- Fabrication and bench testing of the prototype pellet-load scale is complete (Fig.14). Performance of the prototype unit and the blender-dispenser was satisfactory. Blending of coarse and fine standin oxides with aluminum was effected with the efficiencies shown in Table 8. The load scale performed the weighing of analytical balance counter weights to within ± 0.005 g and controlled the weights of dispensed powders to within ± 0.015 g. Some improvements and changes to the combined system will be incorporated in the advanced design of the die scale and the subsequent blender-dispenser. The prototype units are now being installed in the Pu^{242} glove-box line to further test and evaluate the proposed designs. The detailed design of the advanced die scale, which was started, will incorporate features for the remote adjustment and replacement of components.

Table 8. Efficiency of Remote Blender as a Function of Particle Size and Blending Time

Blending Time (hr)	Max and Min Variation from Average Composition in wt %	
	14 vol % (32.5 wt %) CeO_2 (6 μ) plus type 1100 Al (-325 mesh)	14 vol % (33.85 wt %) Lindsay Mix Oxide, -325 mesh plus type 1100 Al (-100 mesh)
0.5	+0.86, -0.61	+8.71, -4.83
1.0	+0.98, -0.61	+3.91, -3.99

Batch Scale. -- Conceptual design of the batch scale was begun. This unit will be similar to the die scale in operation except that it will have a higher capacity to accept the tare weight of the blender container.

Die and Batch-Scale Support. -- Conceptual design of the support for the die and batch scale was approved and detailed design started.

Cap-Powder Station. -- Bench testing of the cap-powder-addition station revealed that dispensing of cover powder with the unit was not reliable. Some redesign and modification did not improve its performance. Conceptual studies for a better device are now under way. The latest idea is to have a preloaded magazine with containers having the required volume or weight of cap powder. These will be designed to dump the required powder into the dies.

Die-Assembly Feeder. -- Bench testing of the die-assembly feeder (Fig. 15) is complete. The hydraulic cylinder used to position the carriage will be changed to a motor drive to reduce the amount of piping and controls required. Redesign of the magazines will be started after mockup tests are under way.

Die-Holder Station. -- Detailed design of the die-holder station is complete.

Pellet Press. -- Bench testing of the pellet press (Fig. 16) is complete. This unit has been extensively tested, operated, and improved to the point now that damage to fabricated pellets or to components of the press has been greatly reduced. About 800 pellets have been produced during bench operation of the press. This unit will be installed in the mockup soon.

Transfer Arm. -- Bench testing of the transfer arm (Fig. 17) is complete, and the unit will be installed in the mockup for addition test and evaluation.

Equipment Mounting Base. -- Fabrication of the equipment-mounting base (Fig. 18) is complete. This item is now being installed in the cubicle 3 mockup.

Pellet Ultrasonic Cleaner and Thermal Dryer. -- Redesign of the pellet ultrasonic cleaner is being considered, and the pellet thermal dryer is now being redesigned. Bench testing of the prototype ultrasonic cleaner components indicated that these units were too large and too

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Fig. 15. Die Assembly Feeding Station Bench Test, Transuranium Processing Plant.

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Fig. 16. Pellet Press Bench Test, Transuranium Processing Plant.

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Fig. 17. Blender-Dispenser and Transfer Arm Bench Test, Transuranium Processing Plant.

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Fig. 18. Cubicle 3 Equipment Mounting Base, Transuranium Processing Plant.

heavy. A new concept for the thermal dryer is being evolved, and the ultrasonic cleaner is being relocated to the left-rear corner of the cubicle.

Cell Piping. -- Conceptual design and layout of the cubicle-3 piping and both supply and drains were started.

Electrical Design. -- Electrical design for cell 3 has been limited to wiring and controls for bench testing of separate pieces of equipment. The control module for the pellet press is complete and underwent testing during the bench test of the press. Some work was started on the control of the transfer arm to ensure that it traverses and stops at the required stations and that proper interlocks are provided.

Cubicle Crane. -- Detailed design of a crane that is to be employed in each of the metallurgical cubicles is under way. The bridge crane is completely motorized, and provisions have been made for remote replacement of motors and sensitive mechanical components.

6.4.2 Cubicle 2

The fabrication flow diagram for cubicle 2 changed as a result of moving the hydrostatic collapse equipment to cubicle 1.

Pellet Inspection and Loading Equipment. Fabrication of the pellet inspection and loading equipment (Fig. 19) is complete, and bench testing is under way. This equipment consists of six basic components: two pellet feeders, a pellet-diameter gage, a pellet-stacked-length track, the pellet-loading equipment, and the auxiliary enclosure that contains and positions these components. Redesign of the pellet-diameter gage was started. The idea of using vibration to move the pellets through the gage with a clearance of 0.003 in. did not work. The possibility of using an air gage is being studied. Basic operation of the complete system appears to be satisfactory.

End-Cap Welding Equipment. -- Conceptual design of the end-cap welding equipment was begun. This equipment is to perform the functions of top-void measurement, tube-liner installation, evacuation and backfilling with helium, end-cap insertion and roll swaging to effect a mechanical

Fig. 19. Pellet Inspection and Target Loading Equipment Bench Test,
Transuranium Processing Plant.

seal, wire brushing of the weld area, and the closure welding by tungsten inert-gas equipment.

Helium Leak-Test System. -- Bench testing of the prototype helium leak-test system, which was completed, showed that the required sensitivity of 1.0×10^{-8} standard cc/sec can be met. Therefore, final design of the helium leak-test system is now being accomplished. Detailed design of the control console is nearly complete. Presently, the design of the leak-test chamber is being modified to incorporate absolute filters to the evacuation lines and improvements to the chamber seal plate and top-cap actuator.

Target Ultrasonic Cleaner and Tube Transfer. -- The concept developed for cleaning the target had been approved, but additional information indicated that the concept should be further advanced to include immersible transducers. This also changes the concepts for the target enclosure and the mechanism for transfer of rods to cubicle 1.

The design being developed is as follows: the cleaning container will have two compartments: one filled with water will house the transducer. The other compartment, which will be filled with cleaning solution and water, will contain the target rod. The entire chamber can be revolved so that it is aligned with the target transfer tube. This new concept actually combines some operations and should reduce the amount of handling of the completed rod. In addition, the amount of solution to be sent to storage for analysis will be reduced.

Target-Transfer Arm. -- Design of the target-transfer arm will not be started until the design of all process equipment in cubicle 2 is firm.

Equipment for Transfer of Targets from Cubicle 2 to Cubicle 1. -- The concept developed for transferring the target through the wall between cubicles 1 and 2 was approved. Detailed design was started, but will not be completed until the new concepts for the target ultrasonic cleaner have been approved.

6.4.3 Cubicle 1

The fabrication flow diagram for cubicle 1 was changed in that hydrostatic collapse equipment and an additional helium leak-test chamber are to be included in the cell equipment.

Target-Rod Receiver. -- Detailed design of the target-rod receiver is to be continued and combined with the design of the surface-smear equipment, the conceptual design of which is complete.

Target-Rod X-Ray Unit. -- Specifications are being written so that an x-ray unit can be purchased. After receipt of the equipment, supports and methods for remotely removing and replacing the x-ray head must be devised.

Target-Rod and X-Ray-Film Holder. -- The conceptual design previously made for a target-rod and x-ray-film holder would not meet the new requirements established for inspection of the weld closure. In addition, it was deemed desirable that the entire rod length be x-rayed, so a new concept was devised. The concept provides for the positioning of a shielded drum-type film holder (into which the target rod is inserted), which will be indexed in increments of about 26° for exposing a new film surface. Vertical positioning of the rod is provided by the transfer arm, which will raise the rod in specified steps for x-ray exposure. The film holder has lead shielding to protect the film on the back side of the drum during x-ray operation. The holder is placed in an additional shielded container for transport via the intercell conveyor for processing. Shielding during this transport is required because it passes under the chemical cells, which will have a high radiation background.

Target Dimensional Inspection. -- A conceptual design for the target dimensional inspection was completed.

Hydrostatic-Collapse Equipment. -- Detailed design for the hydrostatic-collapse equipment is complete and approved. The device has not been fabricated because the testing of the design is being conducted on a similar unit fabricated for the plutonium target fabrication equipment. A high-pressure booster pump of special design to be placed in the cell adjacent to the collapse chamber was conceived. This pump would eliminate the need of pumping 20,000-psi water from the chemical makeup area to the cell. The booster pump is capable of having its components removed and replaced remotely and capable of transport via the intercell conveyor.

Hex-Can Installation Fixture. -- Two conceptual designs have been developed for a hex-can installation fixture. The first was made obsolete by a change of the target rod design. The latter concept was approved and detailed design was started.

Target Ultrasonic Cleaner. -- The design developed for the cubicle 2 target ultrasonic cleaner is to be utilized for cubicle 1. It will be basically the same except that it will be a permanently fixed two-compartment tank with immersible transducers.

Helium Leak-Test Unit. -- The leak-test chamber used in cubicle 1 will be identical in design to the one in cubicle 2. The entire leak-test system, including both chambers, will be operated from one control console.

Target-Transfer Arm. -- Conceptual design of the target-transfer arm was started. This equipment will have rotary and vertical motions with position indication and control of both motions.

Equipment-Mounting Base. -- Conceptual layout of the equipment-mounting base was started. This will be similar in design to the base shown in Fig. 18, the cubicle-3 equipment-mounting base.

7. TRANSURANIUM PROCESSING PLANT DESIGN

B. F. Bottenfield

W. D. Burch

7.1 Hot Development Facility, Cells 3 and 4, Building 4507

F. L. Peishel

Cells 3 and 4 in Building 4507 are being equipped to test transuranium process chemistry at full-scale activity levels and, in addition, to recover multigram quantities of curium-242 and -244 from irradiated sources. The installation of all equipment for feed adjustment and one cycle of solvent extraction has been completed in cell 4. A dissolver, with its related off-gas clean-up equipment and solution filter system, is being designed for installation in cell 3. This cell will also have a work area for laboratory-type operations.

The design of all in-cell equipment items is complete, and prints have been issued for comment. The pump and filter are identical to the corresponding TRU components.¹¹ The design of the structural and piping portions of the cell is proceeding rapidly.

A flowsheet of the equipment to be installed in the cell is shown in Fig. 20, and a preliminary layout of the arrangement of this equipment in the cell in Fig. 21. The unused area of the cell is reserved for future experimental apparatus as needed for supporting the cell-4 operations or for equipment to be used in further purification of product streams.

Fabrication of the pump is complete. Components of the glove-box station through which product solutions will be transferred are under construction, about 75% complete. The fabrication of the dissolver and filter has begun.

7.2 Chemical Processing Equipment

W. D. Burch

J. M. Turley

O. O. Yarbrow

7.2.1 Equipment Design

Detailed design of chemical processing equipment by the Engineering Department of the Oak Ridge Gaseous Diffusion Plant (ORGDP) is about 41% complete. The equipment rack containing the first-cycle solvent extraction system was designed and is being built. Design of six of the remaining eight equipment racks is nearing completion, with fabrication scheduled to begin early in 1964. Design of the second-cycle solvent extraction rack, which is almost identical to the first-cycle rack, will be started after mockup demonstration of the first-cycle rack is complete. The procedure for preparing oxide, with the desired particle-size distribution for the fabrication of pellets, has not been sufficiently developed to permit design of this system to be started. Designing of this equipment will be delayed about three months.

¹¹W. D. Burch et al., Transuranium Quart. Progr. Rept. Aug. 31, 1963, ORNL-3558, pp 88, 92, 93.

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Fig. 20. Cell 3, Building 4507, Equipment Flowsheet.

Fig. 21. Plan Layout of Cell 3.

7.2.2 Intercell Piping

Piping connections between cells will be routed through intercell piping plugs (Fig. 22) set into 1- by 5-ft openings in the cell partition walls. Jumper lines will run from disconnects mounted on these plugs, either to the plug on the opposite cell wall or to other locations in the same pit. A metal frame will be permanently mounted in the opening in the partition wall and will act to properly locate the removable piping plug. The off-gas header will be installed and sealed in a second smaller opening in the upper portion of this frame. The removable plug will be gasketed to the frame on one side. On the opposite side, the crack between the frame and plug will be caulked. The process jumper lines, including the waste header in any one pit, can be remotely replaced without disturbing either the piping plugs or the piping in the adjoining pits.¹² Individual sections of the off-gas header can be replaced, but it is necessary to disconnect and back off all sections from the end of the cell bank to the particular section to be removed.

7.2.3 Fabrication and Procurement

Fabrication of the rack structure for the first-cycle solvent extraction system was completed, and components are being fabricated and mounted on the rack. Machining and assembly of the pulsed-column flow restrictors, and three pulsers and two pumps were completed. Welders were qualified for Zircaloy-2 welding, in preparation for fabrication of the pulsed columns, holdup tanks, and piping.

Work orders were written for the fabrication of the four 12-unit samplers that mount on the back equipment racks in cells 4 through 7, and for the 5785 disconnect ferrules required for the fabrication of process equipment and piping.

Acceptable bids for all Zircaloy-2 tubing, bar, and plate, exclusive of material for product storage tanks, were received and the orders placed. The orders totaled \$90,900, about 20% less than estimates based on previous purchases.

¹²W. D. Burch et al., Transuranium Quart. Progr. Rept. Aug. 31, 1963, ORNL-3558, pp 95-99.

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Fig. 22. Intercell Piping System Elevation.

7.3 Design and Construction of the TRU Processing Plant

B. F. Bottenfield

G. B. Berry

B. B. Klima

F. L. Hannon

7.3.1 Facility Construction

Construction of the Transuranium Processing Plant by Blount Brothers is on schedule, based on their critical-path analysis. All grade beams have been poured, including the support walls under the loading platforms; the pipe-tunnel floor, cell and vessel off-gas filter-pit floors, and all cell and tank-pit floors have been poured. Installation of the 30-in. stainless steel cell-ventilation duct and other connections beneath the pipe-tunnel floor are complete. The cooling water booster pump pit, numerous valve boxes, diversion pits, electrical manholes and telephone handholes have been completed; installation of service piping is progressing; the low level waste pits have been excavated, and the related piping is being installed. Structural steel has been delivered to the site for installation.

7.3.2 Procurement

All procurement of ORNL-furnished items for the lump-sum contractor is on schedule, although some promised delivery dates have slipped drastically. Most of the basic material (pipe, pipe fittings, sheet, plate, bar stock, special fittings, etc.) in both stainless steel and Hastelloy C have been delivered. Fabrication of viewing-window sleeves, instruments, valves, and Hastelloy C cell and cubicle floor pans and piping assemblies is being conducted on a schedule that will result in their delivery to the contractor as set forth in the contract specifications.

7.3.3 Mechanical Equipment Design

Design modifications for the intercell conveyor components have been completed, and drafting is nearly 80% complete. After the conveyor-control component modifications are made in the shop, this equipment will be tested in the mockup.

Drawings of all cubicle-cooling equipment are almost 80% complete. A full-scale mockup of the cooling equipment will be built and tested.

The conceptual design of the glove-box station to be used in the de-contamination room was approved, and cost estimates for fabrication and design of this equipment are being made. The conceptual design of the waste disposal station to be located in the limited-access area is being revised to reduce the size of the platform. This station consists of a throw-away type of waste carrier which will hold up to 15 or 20 plastic waste canisters and an attached containment box which permits semidirect bagging operations to be performed. Equipment racks may be thrown away, using a similar carrier with a modified transition section which connects to the containment box.

7.4 Critical Path Scheduling

C. H. LaMaster

The total ORNL design effort, consisting of the design of chemical, mechanical, and target-fabrication equipment was on schedule as of the November 22, 1963, progress report period. The total estimated manpower required for this phase of the project, to extend to May 1, 1965, is now 12,865 man-days, of which 6435 had been expended as of November 22, 1963. Estimated completion as of this period, based upon a scheduled probable curve, was 47.6%.

Approximately 24 engineering personnel from the ORNL Plant and Equipment Division and the ORGDP Engineering Department are devoting full time to design. It has been estimated that 970 engineering drawings will be required. At the request of the Atomic Energy Commission, a complete drawing and specification list was compiled. A breakdown of subgroups of the number of drawings required is as follows: chemical process, 470; mechanical, 171; target fabrication, 251; and instrumentation and controls, 75.

A leveled critical path schedule for shop fabrication of chemical processing equipment and assemblies was completed November 29, 1963. The schedule comprises 6910 man-days of shop fabrication, including 1500 man-days for miscellaneous developmental items. The late-finish date for the fabrication work is June 2, 1965.

8. CORROSION STUDIES

J. L. English J. C. Griess

Laboratory tests were continued to determine the effect of ferric ions on the corrosion of welded Zircaloy-2 by boiling and oxygenated 6 M HCl solution. A preliminary test indicated that 50 ppm of Fe^{3+} in the acid increased the Zircaloy-2 corrosion rate to 2 mils/month, compared with 0.2 in the absence of iron. The present tests were conducted in boiling and oxygenated 6 M HCl solutions containing Fe^{3+} concentrations of 5, 10, 15, 25 and 100 ppm (added as ferric chloride). Test solutions were replaced at 100-hr intervals. Table 9 summarizes the results.

Table 9. Effect of Ferric Ions on the Corrosion of Welded Zircaloy-2 by Boiling and Oxygenated 6 M HCl Solution

Ferric Ion Concentration ^a (ppm)	Test Time (hr)	Average Corrosion Rate (mil/month)
0	1000	0.2
5	800	0.5
10	800	0.7
15	800	0.4
25	800	0.5
100	800	0.5

^aAdded as chloride.

All concentrations of ferric ions produced a very mild acceleration in attack rates, but in no case was a rate as high as that in the preliminary test. Rates varied between 0.4 and 0.7 mil/month, which represented a two- to threefold increase over the rate obtained in the absence of ferric ions. These data indicate that concentrations of ferric ion up to 100 ppm in the 6 M HCl will not cause excessive corrosion rates. Analyses of the specimens to determine the extent of hydrogen pickup by the Zircaloy-2 have not been completed.

9. DEVELOPMENT OF ANALYTICAL METHODS

L. T. Corbin

J. C. White

9.1 Radiochemical Determination of Berkelium-249

F. L. Moore

W. T. Mullins

Work was started on the development of an analytical radiochemical method for the determination of Bk^{249} . A two-step liquid-liquid extraction is under study. A preliminary extraction of Bk^{4+} and Ce^{4+} is first made from 10 M HNO_3 with di-(2 ethylhexyl)phosphoric acid. The Bk^{249} is then separated, as Bk^{3+} , from Ce^{4+} by extraction with a tertiary amine, Alamine 336S (dissolved in xylene) from hydrochloric acid solution. The preliminary extraction effects a 98% recovery of berkelium and cerium, with a decontamination factor of greater than 10^6 from mixed fission product solution. Current efforts are being devoted to improvement and refinement of the amine extraction step of Bk^{3+} and evaluation of counting methods for the final measurement of Bk^{249} .

9.2 Evaluation of Arsenazo III as a Reagent for Use in the Transuranium Program

G. L. Kochanny, Jr.

In a previous report,¹³ results of investigation of arsenazo III [1,8-dihydroxyl-3,6-disulfo-3,7-bis(2' arsonophenyl-azo)naphthalene] as a reagent for the spectrophotometric determination of the rare earths, thorium, and zirconium were given. During this period the stability of the reagent to gamma radiation was studied. Experiments were completed on the radiolytic decomposition of arsenazo III in various acids and distilled water.

For 1×10^{-4} M and 2.5×10^{-5} M solutions of the reagent in 9 N HCl , the average rates of decomposition of arsenazo III are 2×10^{-6} and 4×10^{-6} mole liter⁻¹ kilorad⁻¹ of absorbed gamma radiation, respectively. For a 1×10^{-4} M solution in 1 N HCl , the average rate of decomposition

¹³W. D. Burch, et al., Transuranium Quart. Progr. Rept. Aug. 31, 1963, ORNL-3558, p 115.

is 9×10^{-7} mole liter⁻¹ kilorad⁻¹. In 9 N sulfuric and in 9 N phosphoric acids, the initial rate of decomposition is the same as that in 9 N HCl, but it becomes slower as the reaction proceeds. In distilled water, the average rate of decomposition is about 5×10^{-7} mole liter⁻¹ kilorad⁻¹. The initial and final rates of decomposition of arsenazo III in acetic acid solutions are fairly constant at 9×10^{-8} and 2×10^{-6} mole liter⁻¹ kilorad⁻¹, respectively. At a specific absorbed dose, the initial rate of decomposition changes sharply into the final rate of decomposition. The absorbed dose at which this phenomenon occurs is dependent on the acetic acid concentration. Values of these transition points are: 1 N acetic acid, 70 kilorads; 2 N acetic acid, 73 kilorads; 4 N acetic acid, 72 kilorads; 9 N acetic acid, 100 kilorads; 6 N acetic acid, >180 kilorads.

Attempts were made to identify the decomposition products by means of infrared spectra measurements. Freeze drying was used to crystallize the products from solution. The solids were then pressed into disks after mixing with KBr. Results have been inconclusive thus far; no definite evidence of differences in the spectra of irradiated and unirradiated arsenazo III solutions was observed. Attention has now been turned to isolating and identifying gaseous products from the radiolytic decomposition.

9.3 Evaluation of Methods

H. C. Meyer

The review and evaluation of methods for fission product determination continued during this quarter. The determination of zirconium by TTA extraction¹⁴ has been accepted as a standard radiochemical method for the TRU program. On seven Tramex type solutions, the results of extraction indicate that zirconium recovery is $98 \pm 1\%$ with a decontamination of $>5 \times 10^5$ from α activity. The determination of cerium by extraction with di-2-ethylhexylphosphoric acid in n-heptane¹⁵ on the same seven samples

¹⁴F. L. Moore, Anal. Chem. 28, 997 (1956).

¹⁵J. J. McCown and R. P. Larsen, Anal. Chem. 32, 597 (1960).

gave a decontamination of $>10^4$ from alpha activity. With a precipitation step preceding the extraction, the decontamination rose to $>10^5$, with a carrier recovery of $80 \pm 4\%$. The cerium recovery in both cases agreed and were $96 \pm 2\%$.

At the request of R. D. Baybarz, a computer program was compiled for the determination of stability constants of metal complexes. As compiled, the program is limited to potentiometric titrations of the one electron-releasing and one anionic group (e.g., glycine) but can be changed to handle two electron-releasing groups (e.g., ethylenediamine), or two anionic groups (e.g., oxalic acid) if the need arises.

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