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Consolidated Fuel Reprocessing Program Progress Report for Period April 1 to June 30, 1980

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**CONSOLIDATED FUEL REPROCESSING PROGRAM
PROGRESS REPORT
FOR PERIOD APRIL 1 TO JUNE 30, 1980**

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Foreword

The DOE has concentrated all U.S. research and development on fuel reprocessing into one major program – the Consolidated Fuel Reprocessing Program (CFRP) – under the management of the Oak Ridge National Laboratory and the Oak Ridge Operations Office.

This progress report is compiled from major contributions from three previous programs: (1) the Advanced Fuel Recycle Program at ORNL; (2) the Converter Fuel Reprocessing Program at Savannah River Laboratory; and (3) the reprocessing components of the HTGR Fuel Recycle Program, primarily at General Atomic and ORNL. The coverage is generally overview in nature. Experimental details and data have been limited (1) to make the report more concise and (2) to meet the requirements which would qualify the report for unrestricted distribution in the open literature.

1. Highlights

1.1 PROCESS RESEARCH AND DEVELOPMENT

B. L. Vondra

A study of the kinetics of Pu(IV) polymerization at 50°C has shown that at constant acid concentrations, the polymer growth rate has third-power dependence on the Pu(IV) concentration. This finding is in contrast to the generally reported linear relationship between growth rate and Pu(IV) concentration at 25°C.

Columns of pretreated silica gel have been tested in laboratory studies on solvent cleanup to demonstrate repeated loading, elution, and regeneration capabilities at 40°C.

The solvent extraction behavior of KTcO_4 in the $\text{H}_2\text{O}-\text{HNO}_3$ -tributyl phosphate (TBP)-dodecane system has been described for systems containing 0.1 to 4.0 M HNO_3 and 10 to 80% TBP at 25, 40, and 60°C.

The fourth SETF campaign was completed using fuel which had been irradiated to 2.05 TJ/kg (23 780 MWd/t) in the Dresden-1 BWR.

A report on the CONSEPT program has been submitted for publication. This computer program is capable of modeling real-time variations in flow rates and compositions for solvent extraction processes using proportional feedback or on-off controllers.

Chemical flowsheet conditions were modified for the co-denitration of U-Pu nitrate streams in rotary kilns. Resultant improvement in the ceramic properties of the product oxides has been partially demonstrated.

The multidetector x-ray fluorescence analyzer system has been installed and put into routine operation, and the operation of the prototype integrated remote pipettor system has been demonstrated.

1.2 ENGINEERING RESEARCH

W. S. Groenier

The newly installed oxygen supply subsystem and prototype off-gas filter for the full-scale experimental voloxidizer were tested in two operating campaigns, each of one week duration.

Gas-phase residence time distribution measurements are being obtained for voloxidation using helium as a tracer in a small rotary kiln. A helium leak detector coupled to a unit process controller provides automatic acquisition of helium concentration data.

Stress analyses were made for three candidate diaphragm designs for the voloxidizer rotating seal.

After 332 h of operation, selected alloy specimens in a rotary dissolver erosion-corrosion test continue to exhibit only very small changes in thickness and weight. The test is targeted to continue for 600 h.

Dissolver-digester tank studies have identified a pulse mode of agitation to be superior to continuous air-lift recirculation.

Basic hydraulic and extraction tests on the single-stage 12-cm centrifugal solvent extraction contactor have been completed at Argonne National Laboratory (ANL). The results of these tests have been provided to Savannah River Laboratory (SRL) where a multistage array is being designed for the Integrated Equipment Test (IET).

Initial tests of silver mordenite regeneration using a 4% hydrogen stream were only marginally successful in that it took more than 24 h to remove greater than 95% of the iodine which was previously sorbed from a CH_3I -air stream.

Tests of the selective absorption process for krypton removal in the combination column from a feed gas composed of air, krypton, and xenon have shown that, although the presence of xenon causes a displacement and flattening of the concentration peaks, adequate krypton withdrawal rates and product concentrations are obtainable.

1.3 ENGINEERING SYSTEMS

M. J. Feldman

A four-volume report, "Evaluation of Reprocessing Facility Configuration for a Nuclear Energy Center," has been completed. This study evaluates the costs of various facility configurations which could be used to

process a variety of fuels from different types of reactors. A computer program was developed to assist in this optimization.

The General Plant Facility contractor work on the Integrated Equipment Test (IET) facility has been completed, and the designs of the Remote Operating and Maintenance Demonstration (ROMD) and the Integrated Process Demonstration (IPD) portions of the IET are essentially complete.

A test station to evaluate the effectiveness of various television viewing methods for use with remote operations has been installed.

The conceptual design studies of the Hot Experimental Facility (HEF) are on schedule, and an outline of the conceptual design report (CDR) has been completed.

1.4 TECHNICAL SUPPORT

O. O. Yarbrow

The first of the four planned miniruns at the Barnwell Nuclear Fuel Plant (BNFP) was successfully conducted during May. Safeguards criteria for the conceptual design of the Hot Experimental Facility (HEF) have been finalized for submission to the Department of Energy (DOE), and the report, *Safeguards Design Philosophy for the Hot Experimental Facility*, has been published.

Interim validation calculations against the existing U+Pu critical data are 80% complete. Gamma transport calculations of the major sources in the HEF and two Monte Carlo calculations for investigating the shield design for the remote sampler electronics package were completed.

A nondestructive assay (NDA) demonstration experiment using Dresden I reactor spent fuel was com-

pleted. Preliminary results indicate agreement within 2% between the NDA and chemical assays.

The DOE has indicated informally that the Land Use Committee has approved the five primary sites identified in the NUS Corporation siting study for the proposed HEF as being compatible with the Land Use Plan for the Oak Ridge Reservation.

1.5 HTGR Fuel Reprocessing

B. L. Vondra

Two thousand unirradiated spherical fuel elements, manufactured in the Federal Republic of Germany (FRG), were successfully processed through the General Atomic (GA) head-end pilot plant as part of the US-FRG cooperative program for developing gas-cooled reactor spent fuel treatment technology.

More than 200 operating cycles have been completed in the pneumatic transport test loop charged with 115 kg (254 lb) of crushed FRG graphite spheres. The solids flow properties of FRG fuel were measured at several points in the process.

Kinetics studies on zirconium scrubbing from 30% TBP in normal paraffin hydrocarbon (NPH) indicate that neither uranium saturation nor acid concentration have significant effects on transfer rates. The presence of more than one zirconium species is suggested.

In studies of alternate solvents, TEHP and TBP demonstrated equivalent phase disengaging times under acidic conditions; however, the TEHP had a much longer disengaging time when contacted with alkaline solvent washing solutions.

2. Process Research and Development

B. L. Vondra

The process research and development activities provide processing parameters and recommend materials of construction for both the Integrated Equipment Test (IET) facility and the Hot Experimental Facility (HEF) design efforts; new process options (or steps) are also being explored. An emphasis is placed on solution properties in dissolution and solvent extraction steps, and attention is being given to several aspects of plutonium chemistry and solvent extraction technology. Hot-cell activities examine the voloxidation and dissolution properties of a variety of light-water reactor (LWR), fast breeder, and thorium fuels. Testing of partial partitioning in solvent extraction flowsheets is proceeding at ORNL in glove-box mixer-settler equipment and in the Solvent Extraction Test Facility (SETF), which has an irradiated fuel capacity in the kilogram-per-day range. Thorium fuels flowsheet testing at Savannah River Laboratory (SRL) is being carried out in mixer-settlers and in a new glove-box pulsed-column facility. The product conversion program is establishing methods for converting U-Pu nitrate solutions to fabricable mixed-oxide products. New analytical methods for process control are being developed and adapted where possible for in-line operation.

2.1 LABORATORY DEVELOPMENT (ORNL)

J. T. Bell

2.1.1 Pu(IV) Polymer Reaction in Aqueous Solutions

L. M. Toth, M. M. Osborne, and H. A. Friedman

Kinetics of Pu(IV) polymerization in solutions of 0.025 and 0.125 *M* Pu(IV) with make-up acidities of 0.13 to 0.23 *M* HNO₃, at 50°C, are essentially complete. Polymerization rates at these conditions were determined before 30% of the plutonium had polymerized. The mechanism includes an initial induction period which is a function of Pu(IV), acid concentrations, and probably temperature. Polymer growth occurs after the induction period and at a rate which has third power dependence on the Pu(IV) concentration at constant acid concentrations. This finding is in contrast to the generally reported linear relation between growth rate and Pu(IV) at 25°C.

2.1.2 Plutonium Reduction-Stripping Studies

M. H. Lloyd and S. E. North

A new reduction-stripping procedure was used to recover about 25 g of plutonium from aged waste

solutions of Pu(IV) in tributyl phosphate-normal dodecane (TBP-NDD) where much of the Pu(IV) was complexed with dibutyl phosphate and/or monobutyl phosphate. Plutonium was effectively stripped from the solutions by a single contact with 1/4 volume of 0.05 *M* ascorbic acid in 1 *M* HNO₃. This procedure and subsequent valence adjustments were significantly more simple than procedures using acidic ferrous sulfamate.

2.1.3 Plutonium Depolymerization Studies

O. K. Tallent, P. C. Arwood, and K. E. Dodson

In preliminary tests, the electrolytic depolymerization of Pu(IV) polymer has been effective for a variety of conditions. Both unaged and aged polymers, with and without uranyl ion, were depolymerized in solutions of 0.3 to 8.0 *M* HNO₃. The principal product was the Pu⁴⁺ ion although other intermediate ionic species were observed. The depolymerization rate was highly dependent on cell current and the surface area of the electrodes. The observed rate was essentially independent of temperature from 25 to 50°C. Based on this preliminary experience, the electrolytic depolymerization reaction is first-order with respect to Pu(IV) polymer concentrations.

2.1.4 Solvent Cleanup

O. K. Tallent, P. C. Arwood, and K. E. Dodson

In a series of solvent cleanup tests, columns of pretreated silica gel were operated to demonstrate repeated loading, elution, and regeneration capabilities at 40°C. The feed solvent contained $\sim 0.001 M$ U, $\sim 0.001 M$ DBP, and $< 0.01 M$ HNO_3 . The solvent cleanup efficiency was high and did not decrease significantly over three loading-regeneration cycles. However, there is some physical degeneration of the column with cycling which would limit the column life.

2.1.5 Formation and Destruction of Hydrazoic Acid

C. D. Bopp, J. C. Mailen, and C. T. Thompson

The reaction of hydrazoic acid (HN_3) with nitrous acid (HNO_2) in 30% TBP—normal paraffin hydrocarbon solution was investigated to determine if HN_3 can be destroyed by HNO_2 in the organic phase. The rates of the HN_3 - HNO_2 reaction were first-order with respect to HN_3 and HNO_2 and showed about a second power dependence on the HNO_3 concentration in the organic phase.

For comparison, the same reaction was conducted with the TBP solution in contact with dilute nitric acid. The organic phase was well stirred, but the phases were not dispersed. The reaction rate under these conditions was about twice that observed for the organic phase alone. The increased rate presumably resulted from the partial transfer of reagents to the aqueous phase where instantaneous reaction would be expected.

2.1.6 Oxalate Destruction Using Hydrogen Peroxide

O. K. Tallent, P. C. Arwood, and J. C. Mailen

The destruction of oxalate by reaction with hydrogen peroxide in refluxing dilute acid has been studied for potential use in handling wastes from a hydrazine oxalate solvent scrubbing system. A summary report on these activities is in preparation. The rate for this reaction increases as about the 0.3 power of the hydrogen ion concentration between 0.04 and 0.55 $M H^+$ and suggests that the reactive oxalate form is the nonionized oxalic acid. The direct reaction of hydrazine oxalate with hydrogen peroxide in slightly acidic solutions was found too slow for practical application. However, the destruction of hydrazine with nitrous acid followed by destruction of the oxalate with hydrogen peroxide shows promise since both reactions are relatively fast.

2.1.7 Fission Product Chemistry

D. J. Pruett and W. B. Howerton

A manuscript describing the solvent extraction behavior of $KTcO_4$ in an H_2O - HNO_3 -TBP-dodecane

system has been completed and submitted for publication. Distribution coefficients were measured for systems containing 0.1 to 4.0 M aqueous nitric acid and 10 to 80% TBP in dodecane at 25, 40, and 60°C. Preliminary results show that the addition of uranium to the system increases the distribution of technetium to the organic phase.

2.1.8 Evaluation of Alternate Extractants

W. D. Arnold and F. I. Case

Measurements of the dependence of uranium extraction on extractant concentration were intended to lower concentrations of TBP, trihexyl phosphate (THP) and tri(2-ethylhexyl) phosphate (TEHP). Uranium extraction coefficients from 1 M HNO_3 increased as the second power of the extractant concentration between 0.04 M and 0.3 M extractant. Uranium extraction coefficients were higher for THP and TEHP than for TBP over a range of nitric acid concentrations and temperatures.

Phase separation from 3 M HNO_3 in static mixer tests was slightly slower with TEHP than with TBP. Aqueous continuous mixing gave more rapid separation, and the separated phases, particularly the organics, were less turbid. Phase separation after contact with sodium carbonate solvent wash solution was much slower for TEHP than TBP.

2.1.9 Spent Solvent Washing

D. W. Holladay and W. D. Bond

Efforts were continued on (1) evaluating aqueous solutions of hydrazine oxalate for scrubbing and (2) scoping tests of the use of electrolytic methods to convert hydrazine in the spent scrub solution to innocuous gaseous products. Simulated spent solvent was used that consisted of 30% TBP containing DBP, MBP, and HNO_3 .

Continuous tests in a single-stage mixer-settler showed that DBP was removed efficiently if the acidity of the scrub solution was maintained in the pH 7-8 range with additions of free hydrazine. Nitric acid removal was essentially quantitative.

In the electrolytic studies, satisfactory decomposition of hydrazine nitrate was obtained with reasonable cell efficiencies provided the anode and cathode current densities were in the proper range.

2.1.10 Nitric Acid and Water Recycle

W. D. Arnold and W. D. Bond

Initial experimental studies in this program are being made to determine the behavior of dissolved TBP in the low-level acid waste (LAW) evaporator of the HEF

reference flowsheet. This evaporator fractionates nitric acid solutions from throughout the reprocessing plant to $\sim 13\text{ M HNO}_3$ in the bottoms and 0.001 to 0.01 M HNO_3 in the overheads; these products are combined to prepare a concentration range of nitric acid solutions for recycle. Some of the solutions processed in the evaporator will contain dissolved TBP from previous contact with an organic phase. Our tests will include measurements of the TBP degradation rate, the degradation product distribution in the evaporator products, and the effect of the degradation products on performance when the nitric acid and water are recycled. A glass distillation system was built that will process 2 ml/min of 1.8 M HNO_3 feed. It performed satisfactorily in shakedown tests with water and with nitric acid feed.

2.2 SAVANNAH RIVER LABORATORY PROCESS DEVELOPMENT

J. D. Spencer and S. F. Peterson

2.2.1 Coprocessing to Breeder Concentrations

M. C. Thompson

Previous tests have demonstrated that U(IV), if present in sufficient concentration in the strip solution, can give product solutions in which 24 to 28% of the total heavy metal is plutonium. Losses to the uranium stream were high in most of these tests because the amount of U(IV) was insufficient. The problems were overcome by using more U(IV) and adding it to two stages of the mixer-settler instead of one as in the earlier tests.

In a test with irradiated Saxton mixed-oxide fuel solution, plutonium losses to the uranium product were 0.03% with U(IV) added at stages 10 and 15 and with a U(IV) to plutonium ratio of 2.5. The plutonium distribution coefficient was below 0.1 as expected for Pu(III) in all stages except stage 2. The low plutonium distribution coefficients show adequate reductant in most stages of the mixer-settler.

Two miniature mixer-settler tests were completed with simulated solutions in which U(IV) was introduced into stages 8 and 14 to determine the optimum points for introduction of U(IV). Mixer-settler operation was good throughout both tests. In these tests, the plutonium distribution coefficients were sufficiently low (<0.1) in all stages except stage 16. The distribution coefficients were lowest in stages 2–8. The plutonium concentration in the product, as a fraction of the total heavy metal concentration, was 26 times that of the feed. Losses of plutonium to the uranium stream were high in both tests (0.57 and 0.12). The results indicate that more of the U(IV) should be added at the stage close to the plutonium product exit with the remainder added to stage 8.

2.2.2 Tritium Removal Tests

J. B. Pickett

Tests of tritium removal, dissolution, and residue characterization continued in the SRL High-Level Caves with Elk River fuel initially 95.4% ThO_2 –4.6% UO_2 , irradiated to $\sim 12\,000\text{ MWd/t}$ and cooled ~ 12 years.

The effect of high roasting temperature on tritium evolution was studied on three samples of Elk River fuel from a homogeneous feed, which were heated in air at 600, 800, and 1000°C for 30 h. The tritium evolved during roasting was trapped on molecular sieves, and the residual tritium was determined after subsequent dissolution. Each of the samples was dissolved in refluxing Thorex dissolvent for 12 h; the percentage of dissolution was determined at 6, 9, and 12 h.

The tritium evolution data indicate that the goal decontamination (decontamination factor of 60) can be attained by heating the fuel at 1000°C for approximately 10 h. Gas evolution data from a test with agitation (at 600°C) indicate that after 15 to 18 h, tritium removal is improved. Pulverization of the thorium fuel to very small particle size ($<10\text{ }\mu$) will increase tritium evolution, but only 70 to 80% of the tritium is released at 600°C in 10 to 12 h of heating. Thus, a high roasting temperature is the key variable affecting tritium release from thorium-based fuels.

Calculations were completed for the fission product inventory (including tritium) as a function of burnup and cooling time for Elk River fuel. These calculations support measurements on this fuel in the High-Level Caves and correlate tritium content with the content of specific fission products. These results indicate that ^{137}Cs is an excellent monitor for estimating the tritium content because, to a first order, the ratio of tritium to ^{137}Cs is insensitive to burnup, flux level, flux shape, and fuel composition.

2.2.3 Pulsed Column Facility

D. J. Reif

The Pulsed Column Facility installation is complete, but use with radioactive materials has been delayed to allow correction of design deficiencies and to incorporate changes to improve overall operability. The need for these alterations was discovered during operator training and equipment checkout using the TBP solvent and nitric acid. Studies with radioactive materials are to begin during the fourth quarter of FY 1980. The cold tests showed that the column-to-column geometry was unsatisfactory for acceptable gravity flow of the organic from one column to the next. This was corrected by removing the air lift system, converting the head pots to reservoirs, and installing variable-speed metering pumps to feed the reservoir contents to the next desired process

element. Also, the facility was modified to provide easier and quicker adjustment or changeout of electropulse column anolyte. Corrective actions were taken to maintain safe operation of the evaporator and to prevent glove box activity from passing through cold feed lines.

2.2.4 On-line Instrumentation for the Pulsed Column Facility

J. R. Cadieux and S. F. Peterson

The detectors for on-line determination of stream density, alpha content, and gamma control in the Pulsed Column Facility were installed and checked for operability. An electrical noise interference from air pulse solenoids was found; steps to eliminate the interference are under investigation. The computer interfacing for all the detector systems is complete and operable.

2.2.5 Joint SRL-PNL Thorium Irradiation Program

J. B. Pickett, J. L. Steimke, and D. S. Cramer

A joint program between the Savannah River Laboratory (SRL) and Battelle-Pacific Northwest Laboratory (PNL) to prepare irradiated $\text{ThO}_2\text{-UO}_2$ reference case ceramic fuel pellets was completed. The test material is a proliferation-resistant composition of 80% ThO_2 - 20% UO_2 . Five types of ceramic pellets were prepared at PNL and loaded into 30-in.-long Zircaloy rodlets. The irradiated materials are to be used in head-end, dissolution, and extraction studies at SRL after characterization of irradiation performance by PNL. When the samples are received from PNL, they will be inventoried and stored at SRL for possible later irradiation.

2.3 HOT-CELL DEVELOPMENT (ORNL)

V. C. A. Vaughen

2.3.1 Flowsheet Parametrics for Mixed-Oxide Fuels

J. H. Goode, R. G. Stacy, J. R. Travis, and C. S. Webster

The Hanford Engineering Development Laboratory (HEDL) experiment P-15 involved the comparative evaluation of the irradiation performance of four batches of $(^{238}\text{U}, \text{Pu})\text{O}_2$ pellets prepared in different ways. The four types of fuel were irradiated to 1300 GJ/kg (15 000 MWd/t) in subassembly EBR-2.

The voloxidation of the high-burnup fuel containing $(\text{U}, 20\% \text{Pu})\text{O}_2$ or $(\text{U}, 25\% \text{Pu})\text{O}_2$, manufactured at HEDL for the P-15 rods, was completed. More than 95% of the mixed oxide was released from the cladding during voloxidation of 1.3-cm-long pieces in air at 600°C. Tritium removal during voloxidation was essentially complete in all experiments. However, samples of

$(\text{U}, 25\% \text{Pu})\text{O}_2$ fuel had higher ^3H concentrations by a factor of 4 than were present in the 20% PuO_2 samples.

The voloxidation of low burnup (56 GJ/kg) mixed oxide containing 20 or 25% PuO_2 , manufactured at HEDL for the P-15 irradiation experiment, was completed. Pulverization of the 25% PuO_2 fuel was not as extensive as it was for the 20% PuO_2 fuel (39% of the product sized $<44 \mu\text{m}$ vs about 69% for the 20% PuO_2 fuel) but more tritium was evolved.

Dissolution parametrics

Baseline dissolution tests were conducted with unvoloxidized fuel samples from both the commercial vendor and HEDL. The fuel was refluxed for 6 h in sufficient 7 M HNO_3 to yield a final heavy metal concentration of 200 g/l. The residue was centrifuged, washed with 3 M HNO_3 , and leached for a second 6-h period in 200 g U/l - 3 M HNO_3 . A third leach, containing fluoride, was then used before the insoluble residue was finally fused in Na_2CO_3 for analysis.

Based on previous tests with unirradiated samples of this fuel, the irradiation improved the solubility of the unvoloxidized fuel. However, voloxidation increased the amount of dissolution residue and the amount of plutonium left undissolved after HNO_3 leaching. The fuel prepared by coprecipitation was more soluble than that prepared by mechanical blending ($\sim 0.1\%$ vs $\sim 2\%$ of the plutonium was insoluble in nitric acid). In summary, irradiation improved the solubility, whereas voloxidation decreased the solubility. These results are similar to those obtained previously with other fuels.

2.3.2 Fuel Procurement

J. H. Goode

About 1 kg of unirradiated $(\text{U}, \text{Pu})\text{C}$ pellets was received from the Westinghouse Advanced Reactors Division. The material will be used for flowsheet development studies as a supplement to hot-cell experiments with similar irradiated material. Schedules for transporting and processing Browns Ferry fuel for use at ORNL have been confirmed and should be received during the last quarter of FY 1980. Hanford Engineering Development Laboratory has released more irradiated Fast-Flux Test Facility vendor fuel for ORNL studies; these will also be available during the last quarter of FY 1980.

2.3.3 Code Development

A. D. Mitchell

A problem with the mass balances in the SEPHIS-MOD4 program was discovered for predictive cases where plutonium is being reduced, especially when large

amounts of plutonium are reduced during one time increment. The distribution coefficients, calculated before and after the reduction calculations, differ to such an extent that the program "loses" some of the solute in the organic phase.

A report on the CONSEPT program has been submitted for publication. This program is capable of modeling real-time variations in flow rates and compositions for solvent extraction processes using proportional feedback or on-off controllers.

Tri-(2-ethylhexyl) phosphate (TEHP) is being considered as an alternate extractant to tri(n-butyl) phosphate (TBP) for the Purex process. Limited distribution data have been obtained for the TEHP system. These data were correlated by the same method used previously in describing the correlations for TBP in the SEPHIS program. The correlations reproduce the initial data very well. These correlations, although preliminary, are available for use in the SEPHIS-MOD4 version available at ORNL, but the results of such calculations are recommended as rough estimations of actual performance.

2.3.4 Oxidation of Carbide Fuels

D. O. Campbell and W. L. Pattison

The second and third heat treatment tests with irradiated U-Pu carbide fuel were completed, and another is planned before termination of the heat treatment studies. The second test was conducted with CO₂ carried in flowing helium at 800°C for 24 h. Nearly all the fuel was converted to a granular powder which appeared to have been forcibly expelled from the cladding. The cladding bulged somewhat, with rings of greater diameter, possibly from expansion of the contained fuel during the oxidation.

The third test was carried out in two parts, the first of which was for 24 h in flowing helium at 800°C. At that point, the furnace was cooled, and the fuel was removed, photographed, and weighed. The fuel was unchanged in appearance and had lost 0.3% of its weight. However, fission product deposition caused major changes in the appearance of the quartz furnace tube. Extremely radioactive deposits were found in the hot zone in the vicinity of the fuel, the cooler zone at the downstream end of the furnace, and in the gas outlet tube.

The fuel was replaced in a new furnace tube and heated to 800°C for 6 h in CO₂-helium sweep gas. Following this, about half of the fuel had been expelled from the cladding as granular powder and half remained in the cladding. There was a weight gain of about 3%, corresponding to conversion of about half the fuel from carbide to oxide.

The bulk of the treated fuel dissolved readily in nitric acid but there have been relatively large amounts

of residue that dissolved very slowly. The residue appears to be greater in amount and different in behavior than in the case of oxide fuel, but no quantitative data are available yet. The reacted fuel consists of a wide range of particle sizes from 10 to 20 mesh down to -325 mesh. Tritium was effectively released from the reacted fuel.

With respect to reprocessing, two problems are apparent that are more severe than for oxide fuels, namely higher volatilization of fission products from the fuel and a higher residue fraction that resists dissolution in nitric acid.

2.3.5 Plutonium Flowsheet Tests

B. A. Hannaford and W. D. Bond

Efforts were made to determine the cause of disagreement between analytical measurements of uranium and plutonium in 30% TBP and the corresponding concentrations calculated from in-line density measurements during mixer-settler extraction experiments. The calculated uranium concentrations were positively biased with respect to analytical results by the Davies-Gray method. Postulated explanations such as error in the measuring temperature or entrainment of aqueous phase were ruled out by the magnitude of the error.

Carefully controlled experiments in which test solutions were prepared by a series of dilutions, and density measurements were made with static samples in a Mettler density meter again showed that uranium concentrations calculated from density measurements were biased on the high side for both aqueous and organic solutions. However, the bias was only about 20% of that observed in the tests with flowing solution. An experiment was initiated to determine if a static aqueous film could exist in the density tube under conditions of continuous organic flow.

A second miniature spectrophotometer was obtained for in-line use in the glove boxes. Measurements of absorbance at 416 nm indicated that they would cover the range of about 10 to 105 g/l of uranium in the aqueous phase, and that the presence of uranium will not interfere with Pu⁴⁺ determination. Output signal lines were connected to the Hewlett-Packard data acquisition system for continuous recording.

2.4 SOLVENT EXTRACTION TEST FACILITY

L. J. King, D. E. Benker, J. E. Bigelow, E. W. Brown, E. B. Cagle, F. R. Chattin, E. D. Collins, S. C. McGuire, D. B. Owsley, R. G. Ross, H. C. Savage, R. C. Shipwash, C. E. Waddell, and A. V. Wilder

The fourth Solvent Extraction Test Facility (SETF) campaign was completed in June 1980. The fuel processed during this campaign was irradiated to 2.05 TJ/kg (23 780 MWd/t) in the Dresden-1 boiling water

reactor (BWR) and discharged on September 1, 1975. Three batches of sheared fuel rods, each containing about 3 kg of heavy metal, were processed. The fuel was dissolved in nitric acid solutions by the same procedure used in previous SETF campaigns with pressurized water reactor (PWR) fuel from the H. B. Robinson-2 reactor. No difficulties were encountered.

During campaign 4, the dissolver solutions were clarified by filtration, using a new type of precoated filter and filtration technique. Results were much better than in previous runs; the filtrations were accomplished at the desired flow rates and the filtrates were lightly colored and free of visible solids. In the subsequent solvent extraction runs, only a relatively small amount of emulsion formed in the settlers near the feed stage.

In the solvent extraction system, results were about the same as were obtained earlier with the H. B. Robinson-2 PWR fuel. Uranium and plutonium losses to the raffinate (AW) stream were 0.04 and 0.002%. Decontamination factors were 3×10^4 for ^{106}Ru , 4×10^6 for ^{137}Cs , and $>2 \times 10^5$ for ^{144}Ce .

The plutonium was reductively stripped using hydroxylamine nitrate (HAN) during the first half of run 4-SX-1 and hydrazine-stabilized HAN during the second half. The plutonium was stripped effectively (99.95% recovery), even without the use of hydrazine. Part of the uranium was stripped along with the plutonium; the partitioning of the uranium was regulated by means of an organic backscrub stream. The plutonium concentration in the U-Pu product was a factor of about 20 higher than in the fuel solution.

During the first half of the second run (4-SX-2), the plutonium was stripped as Pu(IV) and during the latter half, a small amount of an organic soluble reductant was added via the organic backscrub stream to partially reduce the plutonium. The yields of stripped plutonium were 97.9 and 99.92%, and the concentration factors obtained were 17 and 14.

2.5 PRODUCT CONVERSION

R. E. Norman

2.5.1 Gel Sphere Conversion Development

Chemical flowsheet development

M. H. Lloyd, R. L. Fellows, J. L. Collins, D. H. Newman, S. E. Shell, and W. B. Stines

Flowsheet studies were expanded to a wider range of processing parameters during this quarter. Important parameters identified were the gelling agent-to-metal mole ratio, the sphere forming temperature, the nitrate-to-metal mole ratio of the feed, and the drying

procedure. These parameters define the crystalline nature of the product gel sphere which is important to further processing.

Pellet screening tests on six U-Pu gel sphere batches prepared last quarter gave pellet densities ranging from 72 to 90% of theoretical. A duplicate 50-g batch of one of these samples was prepared and transferred to PNL for evaluation.

Seven additional batches were prepared this quarter and are awaiting pellet screening tests. About 70 g of a duplicate of one of these batches has been transferred for evaluation.

Process and equipment development

A. D. Ryon, R. D. Spence, P. A. Haas, V. L. Fowler, and K. E. Sullivan

Equipment development was continued for chemical flowsheet studies and for design of an engineering facility for producing Pu-U oxide spheres. The use of a high viscosity gelation medium has required considerable innovation and development to obtain good control of drop size and to provide adequate residence time for gelation. The use of a surfactant reduced sphere contact on the surfaces of the equipment, but it complicates the flow through the aging section. A new forming nozzle shows promise of being superior to those previously used.

2.5.2 Direct Denitration Conversion Development

Lab-scale studies

J. C. Mailen, O. K. Tallent, D. J. Pruett, W. B. Stines, D. W. Lee, and D. R. McTaggart

Differential scanning calorimetry thermograms at various heating rates between ambient conditions and 370°C have been used to study the rate of decomposition of uranyl nitrate hexahydrate. The shifts of five endotherm peaks with heating rate were used to determine activation energies for dehydration and denitrations. Uranyl nitrate solids, prepared by evaporation of a uranyl nitrate-nitric acid solution to dryness at room temperature, were studied by thermogravimetric apparatus. The thermogram appeared very similar to previous thermograms for uranyl nitrate hexahydrate.

A reduced-scale rotary calciner has been designed and constructed. This unit will be tested for continuous thermal denitration of U-Th solutions, and when operational, will be installed in a glove box for U-Pu tests.

Process and equipment development

P. A. Haas, R. D. Arthur, and D. Peishel

Development studies on the use of rotary kilns for co-denitration are continuing. Thirty-one runs, including

twenty-six with depleted uranium, were made in an 8-cm-ID and 90-cm-long rotary kiln test unit.

A chemical flowsheet, modified to improve the ceramic properties of the product oxides, has been partially demonstrated. The principal criterion is the density of UO_2 pellets when the product oxides are fabricated into pellets by a standard procedure. The usual denitration conditions for UO_3 preparation in the rotary kiln give sintered densities of 68 to 70% of theoretical while the modified kiln conditions consistently give sintered densities which are 92 or 93% of theoretical. Large improvements in pellet densities at the modified conditions are evident with $(\text{U,Th})\text{O}_x$ prepared by batch denitration.

Steady-state discharge of UO_3 has been demonstrated including one test with 8.5 h of uranium feed. Three chemical or equipment flowsheet conditions tend to result in accumulation of solids on the kiln wall and should be avoided. For the preferred flowsheet conditions, the product UO_3 shows the same iron contents (90 to 150 ppm) as the feed $\text{UO}_2(\text{NO}_3)_2$ solution, indicating that stainless-steel is an acceptable material of construction for the kiln.

Material balance results for batch denitrations were good and did not reveal any complications for waste handling. The continuous rotary kiln is operated with countercurrent flow of the exit gases and feed solution in a packed scrubber. Uranium losses were low and other measurements of condensate and noncondensed compositions and rates are in progress.

2.5.3 Laboratory Product Conversion Studies — Peroxide Coprecipitation

J. C. Mailen, D. R. McTaggart, and W. B. Stines

Pellet screening studies with the single batch of UO_2 -30% PuO_2 powder prepared by peroxide coprecipitation were continued. This batch of material was reground, pressed, and sintered giving a geometric pellet density still only 70% of theoretical density, indicating that the additional milling was ineffective in improving the sintered density. No further work in this area is currently planned.

2.5.4 Building 3019 Facility Modifications

F. E. Harrington, J. R. Parrott,
D. F. Williams, and F. L. Daley

Facilities will be developed in Building 3019 to prepare kilogram quantities of mixed heavy metal (U, Pu) oxides by the gel sphere and thermal denitration routes.

Currently, laboratory space is being cleaned for the installation of the gel sphere development line. The

glove boxes have been designed and the purchase requisition has been sent to vendors. Detailed design has begun for one of the required six experimental systems. Specifications are nearing completion for auxiliary equipment to support the experimental systems.

Detailed design has begun for the High Alpha Off-Gas System for Laboratories 208 through 211.

2.5.5 Product Conversion Development at SRL

J. D. Spencer, M. K. Jones, and O. M. Ebra-Lima

The Fuel Cycle Technology Division at SRL has designed and is operating a Finishing Process Development Facility (FPDF) to study an ammonium diuranate slurry calcination process in equipment with a capacity of 100 kg U/d.

Off-gas studies

W. J. Jenkins and D. U. Cook

A series of process tests was completed during which calciner off-gas was analyzed for NO , NO_2 , and NH_3 . The purpose of the tests was to determine the mode of ammonium nitrate disappearance in the fluidized bed calciner. Preliminary results indicate that a fraction of the ammonium nitrate fed to the calciner vaporizes as NH_3 and (presumably) HNO_3 when an excess of NH_4OH is present in the calciner feed solution. Apparently, no ammonium nitrate vaporizes without excess ammonia in the feed.

2.6 ANALYTICAL CHEMISTRY DEVELOPMENT (ORNL)

D. A. Costanzo

2.6.1 Chromatography

J. M. Keller

A Dionex model 10 ion chromatograph has been modified to provide the capability for anion and cation analysis of low-activity samples. The modifications were such that all components of the instrument which come in contact with the sample may be contained within a hood or glove box. The columns, injector, and detector cell were separated from the console and enclosed in a stainless-steel cabinet.

The suppressor column used with ion chromatography to suppress the background conductivity has several disadvantages involving instrument operation and analytical complications. A bipolar-pulse technique for measuring conductivity readily lends itself to electronic nulling of the background current. Electronic design changes of the bipolar-pulse conductivity detector to improve thermal stability, noise suppression, and current

output have been completed. Assembly of the prototype is approximately 50% complete.

2.6.2 X-Ray Fluorescence (XRF) Systems Development

J. H. Stewart, Jr.

A multidetector x-ray fluorescence system has been installed and is performing according to specifications. The system was designed by United Scientific Corporation to measure the fissile elements (uranium, plutonium, thorium) and associated trace impurities in aqueous or organic materials using energy dispersive analysis. The system consists of a computer (Tracor Northern TN-4000) which controls a Si(Li) detector with pulsed-Mo-target x-ray tube for analysis of non-radioactive materials. A planar germanium detector with isotopic sources (^{241}Am , ^{109}Cd , and ^{57}Co) and Si(Li) detector and x-ray tube have been installed in a glove box for analysis of radioactive materials. The detectors and associated automatic sample changers can be operated singly or simultaneously by the system console. This is believed to be the only system in the U. S. with these capabilities.

A helium flow valve assembly was received and installed. With this assembly, analyses in an inert atmosphere and at normal pressures can be performed. The lower atomic-numbered elements (P, S, Cl, K) can be analyzed in liquid samples, a capability unattainable in the usual vacuum inert gas chamber.

The analysis of 5- μg total titanium or zirconium has been demonstrated by precipitation of the elements from solution, followed by concentration onto a millipore filter disk.

2.6.3 Nuclear and Radiochemical Methods Development

L. C. Bate, J. F. Emery, and J. R. Stokely

Previous studies have shown that samples containing iodine as iodide are unstable, and that losses of iodine to the walls of the sample container could affect analytical results. Additional studies show that if iodine is present as iodate in the sample solution, no losses are evident. The valence state of iodine in reactor fuel samples received for ^{129}I analysis is being established to determine if corrective measures are needed to prevent sample deterioration.

Work to improve analysis methodology for $^{14}\text{CO}_2$ in gas bulbs has continued. Recovery yields of ^{14}C from the gas bulbs was increased from approximately 20 to 80–85% by procedural modification. Additional efforts will be made to further increase the recovery as soon as a new gas standard is received.

A Packard model 2405 liquid scintillation counter is used to provide a large number of routine analyses for tritium, ^{14}C , and ^{99}Tc . A system of FORTRAN computer programs has been developed to allow interfacing of the liquid scintillation counter to an existing Nuclear Data 6620 data acquisition system. This system accepts counting results, performs calculations, and prepares the data for direct entry into a management system. At the present time, data transfer between the systems is by paper tape, but plans are being made to provide a direct, hard-wire link between the systems.

2.6.4 Spark-Source Mass Spectrometry Development

R. J. Warmack and J. A. Carter

Cold testing of the shielded spark-source mass spectrometer has begun. The apparatus was installed and aligned following the completion of building modifications. Power, vacuum, pneumatic, water, and liquid nitrogen lines were installed and tested. A prototype device for sample electrode handling was designed and tested.

A method for determining fission product elements in dissolver solutions was completed. Using a synthetic waste solution containing Eu, Sm, Nd, Pr, Ce, La, Ba, Cs, Te, Pd, Rh, Ru, Mo, Zr, Y, Sr, and Rb at undiluted levels of 39 to 812 ppm (wt), analyses showed an average precision of 16.5% (standard deviation) and an average bias of 8.5% in eight determinations.

2.6.5 Hot-Cell Analytical Instrumentation Development

L. N. Klatt and D. E. Goeringer

Operation of the prototype integrated remote pipettor system has been demonstrated. An interface circuit board was designed and fabricated for compatibility with the selected microcomputer controller. The mechanical, electronic, and software modules have been assembled into an operating unit.

The design of the final packaging for the pipettor system has begun, and preliminary development of the mechanical and electronic modules for the remote titrator has been initiated.

2.7 ANALYTICAL CHEMISTRY DEVELOPMENT (SRL)

J. D. Spencer, M. L. Hyder, and T. J. Anderson

Activities in this CFRP work task were terminated at the end of the second quarter of FY 1980 due to reallocation of funding.

2.8 MATERIALS COMPATIBILITY

J. C. Griess

The experimental work for this task is conducted at Battelle Columbus Laboratory under subcontract to ORNL.

2.8.1 Experimental Corrosion Studies at Battelle-Columbus Laboratories

J. A. Beavers, R. R. White, and W. E. Berry

Off-gas corrosion studies

Studies with iron- and nickel-base alloys in air streams that contained 1.0×10^{-5} g/ml water and variable concentrations of iodine showed that both the overall corrosion and pit depth increased as the iodine concentration increased. Rates were only slightly higher at 40°C than at 25°C. Preliminary tests with a high silicon stainless steel (4 Si, 15 Ni, 18 Cr, 0.02 C, and designated EAS-2 Si) in the above environments showed that pitting was less severe than with the other austenitic stainless steels.

Tests in HNO₃

During 500 h in boiling 70% HNO₃, EAS-2 Si corroded about a factor of 10 less than type 304L stain-

less steel; however, the rate (625 μm/year) was still too high for consideration in the high-temperature part of the Iodox process. In 90% HNO₃ at 40°C, corrosion rates were only 2 and 6 μm/year for unwelded and welded specimens, respectively; therefore, the alloy may be useful for acid storage tanks and for the absorption column in the Iodox system.

The presence of ruthenium in boiling 6 to 12 M HNO₃ greatly increased the rate of oxidation of Cr(III) to Cr(VI), which in turn increases corrosion of iron- and nickel-base alloys.

To investigate corrosion that may occur in mechanical connectors, crevice tests with type 304L stainless steel were conducted in boiling 6 to 9 M HNO₃. Cr(VI) had no detectable effect either on freely exposed surfaces or in crevices. In 9 M HNO₃, weight losses were three times greater in the presence of Cr(VI) than in its absence, and significant end grain attack occurred, particularly when the end grains were in a crevice. Slight intergranular attack was also noted on all surfaces exposed in the latter solution.

3. Engineering Research

W. S. Groenier

The various engineering research studies summarized here represent the first phase of the equipment development program and provide a basis for further design and development of specific engineering test components. Emphasis is placed on the identification, study, selection, development, and characterization of unit processes through the application of basic or fundamental experimental engineering techniques.

Studies of the voloxidation, dissolution, feed preparation, solvent extraction, and off-gas treatment processes are in progress, and development efforts are described. Work directed toward voloxidizer and solvent extraction equipment development is also reported.

3.1 VOLOXIDATION

M. E. Whatley

The voloxidation process is being developed as a head-end method for removing tritium from spent uranium reactor fuel prior to aqueous processing. Based on experimental work, it appears that this objective can be met by reacting the oxide fuel with oxygen or air in a temperature range of 720 to 920 K. The release of tritium and, to a lesser extent, some of the other fission products occurs when UO_2 is restructured to U_3O_8 during oxidation. The early removal of tritium from the fuel into a relatively small volume is desirable to avoid mixing the tritium as water with aqueous streams in subsequent process steps. A secondary benefit of voloxidation is the oxidation of residual sodium in the fuel prior to dissolution.

An investigation of the applicability of rotary kilns to the voloxidation process has been continued by performing heat and mass transport tests using a small rotary kiln, continuing a kiln-seal evaluation study, and continuing an evaluation of molecular sieves for tritium retention. Tests of a 0.5-t/d voloxidizer system are also described; this major effort features the test and evaluation of commercial components.

3.1.1 Full-Scale Experimental Voloxidizer (FSEV)

T. D. Welch, B. B. Spencer, and M. E. Whatley

An experimental voloxidation system scaled to 0.5-t/d has been provided to study (in nonradioactive

experiments) certain important process parameters and the applicability of some full-scale standard commercial equipment components, to extend and verify basic data on heat and mass transport in the voloxidation system, and to address the problems of process control.

Operations

The replacement drum received in April was found to have major deviations in the internal flights and welding; therefore, the drum was rejected and returned to the vendor.

The oxygen supply subsystem and the prototype off-gas filter were recently installed. Two week-long runs were carried out to test the new installations and to collect data on gas residence time, oxygen control, solids behavior, and the kiln heat balance.

Instrumentation

W. F. Johnson, S. S. Gould, and J. K. Yarbrough

The ACCOL "F" software package for the FSEV unit process controller was upgraded to effect a savings in overall system time and a decrease in input/output wait time. The savings in time permitted faster module update rates, which gave better control of the FSEV. Installation of the color CRT operator interface display package is complete.

A preliminary oxygen supply and control system was installed and tested. The control loop consisted of a single PID controller and an oxygen analyzer at the product exit end of the kiln. A D/P cell was also installed in the oxygen supply line to provide flow data.

The drum temperature telemetry system was installed and calibrated, and the functional operability of the system was demonstrated. Slight modifications have been made to improve the effectiveness of the calibration procedures and the reliability of data transmission.

Data analysis

During FSEV runs 10 and 11, the data were recorded on certified data cassettes using the Techtran recorder. The data have been corrected (i.e., the run times and accumulated variables after a reset) using a computer code and then transferring to standard-label nine-track tapes on the IBM system. Microfiche copies of the data have been made and placed in the FSEV file with the data cassettes.

Another computer program (CRUNCH) has been written and debugged to retrieve, manipulate, correlate, and display (in tabular or graphical form) any set of up to eight variables from the nine-track tapes by a simple formatted input statement.

3.1.2 Rotary-Kiln Heat and Mass Transport Studies

B. B. Spencer and M. E. Whatley

A helium leak detector was evaluated for use as an on-line gas analyzer for gas-phase residence time distribution (RTD) measurements in a rotary kiln. This device was selected because of the simplicity and convenience of using a small amount of helium as a tracer in these experiments. The lag time associated with the readings due to the sample line and response time of the instrument was measured experimentally. The device proved to be satisfactory for RTD measurements.

A total of 102 gas-phase RTD measurements were performed. Each run was made isothermally at room temperature. The varied parameters were material inventory, rotational speed, and gas velocity. The transcription of numerical data from the strip-chart records to the computer system is well underway, and an analysis program has been written. The program computes and/or tabulates the mean time, the variance, the skewness factor, the dispersion numbers for both the closed- and open-vessel models, and operating parameters. The reduced data are being examined.

Instrumentation

W. F. Johnson, S. S. Gould, and J. K. Yarbrough

Hardware and software have recently been implemented on the Bristol UCS-3000 unit process controller to allow the automatic acquisition of helium concentration data. Software and operational procedures have been written. The system will allow data recording to cassette tape and hard copy and eliminate the time-

consuming process of taking numerical data from strip-chart records in subsequent experiments.

3.1.3 Tritium Retention from Voloxidizer Process Off-Gas

W. D. Holland

This activity is being performed under subcontract at Tennessee Technological University to determine the effect of iodine on the operation of a water vapor removal system for the treatment of voloxidizer off-gases. Tritiated water vapor is to be removed by a packed-bed adsorber using molecular sieves. Current efforts involve the development of a mathematical model to describe bed performance, the development of measurement techniques for iodine and water vapor analysis, and an investigation of the effect of the binder material used in pelletized molecular sieves on iodine adsorption.

The basis for the mathematical model is a computer solution developed by O. A. Meyer.¹ Initial tests using data obtained from experimental drying runs will begin when the program is operating.

The assembly of an apparatus to model a cartridge-type adsorption system has been completed. Experiments using a column-mounted electrode to determine the extent of water loading in a packed drying bed have also been completed. This device, which measures the change in bed capacitance caused by water loading, shows promise as a means of detecting water breakthrough.

Efforts to adapt a commercially available iodine analyzer (Mast model 724-6) to measure the relatively high iodine concentrations used in this study have been only moderately successful. The modified apparatus has favorable accuracy and response time characteristics, but overloads quickly at high iodine concentrations.

3.1.4 Rotary-Seal Tests

J. G. Morgan

In the conceptual design of the prototypical voloxidizer, isolation from the cell atmosphere is achieved by means of inlet and discharge seals. A spring-loaded graphite ring bears against the rotating kiln flange to provide a sealing surface. A corrugated stainless steel diaphragm accommodates movement of the seal as the kiln expands or runs eccentric. The present test of the seal is being conducted in a dusty atmosphere at a temperature of 366 K. The seal has successfully accumulated 1200 h of run time. Ring wear is measured periodically during this test.

Stress analyses were carried out on alternate candidate diaphragm designs. These included thin (0.0254 cm), thick (0.0762 cm), and fully corrugated geometries.

3.1.5 Rotary-Kiln Corrosion Test

W. D. Holland and M. E. Whatley

Two identical drums (45.7-cm-diam X 50.8-cm-long) fabricated from type 316 stainless steel and Incoloy 800H are being used to evaluate materials for rotary-kiln service. Modifications to experimental equipment which will allow test surfaces to be exposed to iodine-containing moist air have been completed.

3.2 DISSOLUTION

J. Q. Kirkman

The objective of the dissolution task is to ensure that LMFBR fuels can be dissolved in nitric acid to give high metal recoveries. Criticality control and off-gas considerations require dissolver design and operation to be performed within rather narrow limits. Continuous dissolving methods, which appear to offer superior solutions to these and other problems, are being emphasized. The evaluation of compartmented rotary dissolvers has continued.

Work has also progressed on an advanced dissolver design concept and on the development of a method for providing solids agitation in a digester tank.

3.2.1 Rotary Dissolver Erosion-Corrosion Test

W. E. Eldridge

Erosion-corrosion testing of five candidate rotary dissolver construction materials has continued with the completion of two 100-h runs, bringing total specimen exposure time to 332 h. As in previous runs, simulated sheared stainless steel feed material was added to the 1.22-m-diam rotary dissolver, and material specimens of 305-L, 316-L, E-Brite 26-1, Incoloy 825, and Titanium 75A were bolted into the wall of the dissolver using insulators to prevent galvanic corrosion. An 8 M HNO₃ solution was circulated through the system at temperatures ranging from 363 to 373 K.

The exposed material specimens were inspected; weight change results are summarized in Table 3.1. An evaluation of the results of the recent two runs indicates that the combined effects of impaction and abrasion do not appear to significantly accelerate material corrosion.

A final erosion-corrosion run which will provide a total of 600 h of specimen exposure is in progress. New simulated feed material is charged every 100 h.

3.2.2 Advanced Rotary Dissolver Development

W. E. Eldridge

Tests of the advanced rotary dissolver model have demonstrated that the lifting flights in the conic section promote backmixing of solids. A replacement conic section without lifting flights has been designed and is being

Table 3.1. Weight change of specimens from rotary dissolver erosion-corrosion test

Alloy	Designation	Weight loss (mg/cm ²) at			
		32 h	132 h	232 h	332 h
Incoloy 825	A ^a	3.27	5.90	7.17	8.27
Incoloy 825	B ^b	3.36	5.96	7.33	8.28
Titanium 75A	A	0.66	0.85	1.26	1.29
Titanium 75A	B	0.62	0.84	0.92	1.10
304L SS	A	0.37	1.20	1.84	2.39
304L SS	B	0.34	1.28	2.06	2.68
316L SS	A	0.43	1.66	2.61	3.34
316L SS	B	0.60	1.83	2.79	3.70
E-Brite 26-1	A	0.98	2.17	3.45	3.82
E-Brite 26-1	B	0.98	2.24	3.28	4.26

^aZone of no impaction and abrasion.

^bImpaction and abrasion zone.

fabricated. The new section will not alter the spiral screw baffle size.

3.2.3 Solids Suspension Tests

B. E. Lewis

A digester tank test stand has been fabricated to study various means of suspending solids in a simulated dissolver solution. Suspension will be accomplished by either air pulsing, air sparging, or air-lift circulation at a controlled flow rate.

Hardware and software modifications to the digester system have been made to allow slow venting during the pulse mode of agitation. Based on preliminary tests, the pulse mode of agitation appears to be superior to the continuous air-lift recirculation method. New mounting brackets for the particle concentration sensors were installed to eliminate some of the instabilities in the sensor response. Sensor characterization tests have shown that small air bubbles have little effect on performance; however, large bubbles produce noticeable fluctuations. New dual-pen recorders, which were installed to monitor the output from the concentration sensors, revealed instabilities in the zero setting. Work is in progress to determine the cause of this problem.

Preliminary experiments to estimate the quantity of solids expected to collect on the conic walls of a stainless steel Pachucca-type digester, of similar design to the Plexiglas tank under study, indicated that only about 10 g of material (with a density similar to tungsten in the size range of 5 to 8 μ m) would be expected to collect on the 1.05 to 1.40 rad (60 to 80°) inclined walls.

General designs for Pachucca-type digester tanks (0.5-t/d capacity) have been prepared for input into the general space requirements for the IET dissolver system.

3.2.4 Concentration Profile Studies

B. E. Lewis

Modifications to the unsteady-state dissolver material balance code were made to upgrade the code to current design specifications. As a result of several modifications, an unexplained peak has appeared in the predicted concentration profiles. Program debugging is continuing.

3.3 FEED PREPARATION

W. S. Groenier

The aqueous feed discharged from the primary dissolver system will contain solids (undissolved fission products, corrosion products, etc.) and will require clarification prior to solvent extraction. Preparation of the feed for solvent extraction will also include adjustment of the plutonium valence and the nitric acid concentration, as well as treatment to remove iodine. This task also encompasses the development of a variety of other processes associated with the liquid portion of fuel reprocessing, such as secondary dissolution methods (with more corrosive reagents and for fuel constituents not soluble in the primary dissolver), accountability systems, and fluid transfer and metering systems.

Several new activities have begun. These include sensor developments for solids, iodine, and fluoride ions in liquid streams and tankage, and a test of a modified nitric acid dehydration system having application to the Iodex off-gas treatment process.

3.3.1 Nitric Acid Dehydration Test

R. M. Counce

The study of the production of hyperazeotropic nitric acid (80 to 90 mass %) from subazeotropic nitric acid (<68.5 mass %) by an extractive distillation process using magnesium nitrate as the solvent has continued. The previous column concept was modified to achieve greater reliability and controllability, thus being more amenable to remotized operation.

Tests have confirmed the viability of the new design. Product nitric acid concentrations as high as 89 mass % have been obtained from stock feed streams of 68 to 70 mass %. Operational efficiencies are being obtained over a range of operating parameters. The patentability of the new concept is being investigated. An internal report describing the details of preliminary experiments has been written.

3.3.2 Monitor Development

R. L. Anderson, C. Feldman, W. W. Johnston, Jr.,
and H. H. Ross

The absorptiometric detection of I_2 vapor in the presence of NO_2 over a dissolver solution is being

studied. The broad NO_2 band, which overlaps the I_2 band considerably, creates a problem in obtaining reliable iodine values. It is proposed that NO_2 interference be eliminated by heating the gas to a temperature sufficient to dissociate the NO_2 into NO and O_2 , but insufficient to dissociate I_2 (i.e., in the 450 to 650°C range). Several designs of furnaces and gas absorption cells have been built to measure the optical properties of I_2 and NO_2 in this temperature range. Earlier work done at lower temperatures showed that the absorption spectrum of I_2 vapor over a 100-ppm solution of iodine in 3.5 M HNO_3 is easily detected when both solution and vapor are at 22°C. This spectrum should be even more intense at process operating temperatures (58 and 80°C), since the vapor pressure of I_2 will then be higher. In a closed system, the dissociation of NO_2 at 500°C was easily detectable. Using newer gas cells and furnaces, the behavior of these substances will now be investigated in an open system, where changes in absorption caused by changes in the total quantity of iodine can be differentiated from those caused by dissociation of the iodine.

The small-angle, laser-scattering, suspended-solids monitor manufactured by Leeds and Northrup was installed in a small flow loop; testing has begun. Ultrasonic monitors for both clarified-feed and slurry mixtures have been demonstrated by the vendor. Cost estimates for ultrasonic acoustic lens particle and slurry sensors were received from Micro Pure Systems.

3.4 SOLVENT EXTRACTION

M. E. Whatley

The development of solvent extraction techniques for the Purex process recovery of advanced fuels is continuing at several sites. The primary objective of a program at Argonne National Laboratory (ANL) is to design, fabricate, and test an annular centrifugal contactor of critically safe dimension to reprocess advanced fuels at a rate of about 0.5 t/d. The program also includes a supporting laboratory effort and will provide the basis for a subsequent effort at Savannah River Laboratory (SRL) to provide a multistaged system.

At ORNL, efforts are underway to improve and apply the SEPHIS and MATEX computer codes for flowsheet engineering studies.

Additional work is in progress at the General Atomic Company (GA) to perform contactor improvement studies, test flowsheets, and test instrument application concepts.

3.4.1 Centrifugal Contactor Development

R. A. Leonard, R. H. Pelto, and A. A. Zeigler (ANL)

Two different centrifugal contactors are being studied. An annular contactor with a 12-cm-diam rotor

is being investigated as a basis for the design of a multi-stage contactor array for use in the 0.5-t/d Integrated Equipment Test (IET) facility. The design and construction of that assembly (based on the results of ANL studies) is expected to take place at SRL.

A second contactor with a 4-cm-diam rotor is being tested to extend performance studies during extraction, to validate scaling correlations, and to ultimately provide an abbreviated multistage array for studying coupling problems.

Studies of the 12-cm contactor

Basic hydraulic and extraction tests on the single-stage 12-cm contactor have been completed and results have been provided to SRL. The hydraulic measurements were extended to include liquid level in the mixing zone. In general, the results were in good agreement with previously developed theories and correlations. Extraction efficiencies were measured as a function of throughput and level of mixing intensity.

Tests of the 4-cm contactor

Hydraulic performance tests have been completed on the single-stage annular centrifugal contactor equipped with a 4-cm-diam rotor. Performance is good at all organic-to-aqueous (O:A) ratios from 0.1 to 10. In no case was the liquid level in the mixing zone excessive.

A 4-stage contactor module, using 4-cm-diam rotors, has been designed; construction of the first module has begun.

3.4.2 Flowsheet Engineering Studies

R. T. Jubin and M. E. Whatley

Modifications to the MATEX code continued in an attempt to eliminate the oscillatory behavior noted in a few cases characterized by high internal reflux of the heavy metals. It is believed that this is a result of the method used to solve the system of equations. Several additional sophisticated matrix solvers other than Gaussian elimination are also being investigated.

Work is underway to model the distribution coefficients of the thorium, uranium, plutonium, nitric acid, and TBP system. Computerized data will be examined for consistency.

3.4.3 Alternate Contactor Studies

D. R. Engler and G. E. Benedict
(General Atomic Company)

Work is in progress at the General Atomic Company to develop alternate solvent extraction equipment and operational procedures. This work includes pulsed-

column and centrifugal contactor development as well as scrub and stripping column studies.

The major accomplishments were:

1. A rough draft report has been prepared, which summarizes work done to increase the operating capacity and/or efficiency of the organic-continuous pulsed columns using alternative pulse conditions and cartridge plate configurations.
2. An interim development report on Robatel centrifugal contactor studies has been prepared.
3. Rosette plates have shown greater capacity than standard nozzle plates in scrub column tests; however, rosette plates do not recover from flooding any better than nozzle plates and have poorer dispersion at lower pulse frequencies.
4. The annular stripping column performed smoothly in the stripping mode and quickly recovers from cyclic flood without large color line excursions typical of cylindrical stripping columns.

3.4.4 Flowsheet and Modeling Tests

D. R. Engler, G. E. Benedict, and
D. E. Swieda (General Atomic Company)

Activities during this period have focused on a continuation of the uranium-thorium profile tests and refinements in the process analysis and control in the solvent extraction plant.

Uranium-thorium profile test runs to investigate the effect of the aqueous-to-organic flow ratio on the operating line location have been completed. Data from recent runs are being reviewed and reanalyzed where necessary. Two procedures, X-ray fluorescence and activation analysis, are being studied as a means for analyzing uranium concentrations of >1 and <1 g/g respectively.

A Mettler DMAII remote density cell will be tested in the solvent extraction plant as a method to continuously monitor a flowing side stream from a phase-splitting sampler. The density measurements will be correlated with uranium loading. Additional control loops have been added to the solvent extraction system in order to control more precisely flow and stream-proportioning.

3.5 OFF-GAS PROCESSING

M. J. Stephenson

This task consists of the development of various processes for the treatment of reprocessing plant off-gases to ensure that environmental goals are met. Work includes the continued development of the noble gas and ^{14}C selective absorption process, the formulation

of plant engineering design criteria, a study of the chemical effects of impurities in the fluorocarbon solvent, and an extended investigation of the distribution coefficients of the noble gases in the fluorocarbon solvent. The use of solid sorbents for various off-gas purification applications is also being evaluated. Tests of advanced sorbent materials are underway at ORNL, and work on an integrated HTGR off-gas treatment system is proceeding at General Atomic Company. Savannah River Laboratory is beginning a new effort that will lead to an off-gas processing demonstration using a sidestream of a radioactive off-gas stream at the Savannah River Plant.

3.5.1 Fluorocarbon Absorption Process Development

**D. K. Little, M. J. Stephenson, and
B. E. Kanak (ORGDP)**

The full definition and optimization of the combination column for the selective absorption of krypton from a gas stream requires individual and simultaneous testing with various feed gas compositions. Two major feed gas components (in addition to krypton) are xenon and carbon dioxide. These components are being used, both singularly and simultaneously, to measure the effect on absorber performance for krypton and product withdrawal capability.

Earlier work featured the absorption of a single component (krypton) using dichlorodifluoromethane (R-12). A maximum product concentration of 32% krypton on an R-12-free basis was obtained from a 13-ppm feed gas concentration; overall removal of krypton was as high as 95%.

In tests using xenon, the concentration peak for xenon was 5 to 10 cm lower than that for krypton under similar conditions. Tests using both xenon and krypton showed concentration peaks up to 0.3 m higher than normal; peak smearing and flattening was evident. Absorber performance in removing krypton, however, was not significantly affected by this phenomenon, and actually may have been enhanced as a result of the co-absorbing flux of xenon. In one test, 96% of the krypton was removed from a feed gas containing 13 ppm krypton with a product concentration of 78% xenon and 7% krypton on an R-12-free basis. Carbon dioxide is being evaluated with krypton and with both krypton and xenon. As CO_2 is even more soluble than xenon in R-12, it is not surprising that initial results have indicated similar performance to the xenon tests.

3.5.2 Off-Gas Processing Demonstration

**J. D. Spencer, S. F. Peterson, A. J. Hill, Jr., and
M. C. Thompson (Savannah River Laboratory)**

A program document for the Integrated Hot Off-Gas Facility (IHOGF) has been prepared. Also, a draft

of a fieldwork package proposal/agreement was completed, approved, and submitted to the DOE for FY-81 budget review meetings.

Comments from a recent IHOGF meeting at ORNL have resulted in some changes in the conceptual process design. Calculations show that water from the reaction of barium hydroxide octahydrate with carbon-14 dioxide will add less than 10% more water to the system; therefore, the dilution of the tritium product is minimal. Barium hydroxide octahydrate instead of molecular sieves will be used in-line for ^{14}C collection. Tritium will be removed after ^{14}C .

Work on the Technical Data Summary (TDS) is ~60% complete. Detailed flowsheets were prepared, and calculation methods for sizing absorption columns were studied. Work is in progress on the process description and calculation of material balances. Sections on the criteria and selection of processes have also been revised.

3.5.3 Solid Sorbent Tests

R. T. Jubin

Within the HEF reference flowsheet, there are several places where solid sorbents can potentially be used to treat off-gases. Modern sorbents, such as metal substituted mordenites and silicic acid, are to be evaluated for those applications.

Based on results from the factorial designed experiments evaluating silver mordenite (AgZ), several additional tests were conducted to study the effects of temperature and humidity on CH_3I removal from an air stream. Increasing the bed temperature to above 423 K produced no increase in filter loading. An increase was previously noted when the bed temperature was increased from 373 to 423 K. Pretreating the filter with moist air instead of dry AgZ allowed a doubling of the CH_3I loading.

Early tests of silver mordenite were conducted by monitoring only the outside bed temperature. Recent tests, conducted to determine actual bed temperatures, indicated that outside temperatures were 30 to 50 K higher than the temperature of the first bed segment. Modifications to the system are being made to allow operation at a uniform temperature across the filter segments. A new main filter heater and stainless steel housing have been fabricated for this purpose.

Work was begun to regenerate the loaded AgZ material. During the first test, a dilute stream (4%) of H_2 at a temperature of 773 K removed ~4% of the CH_3I in 6 h. A thermodynamic analysis of the hydrogen regeneration process indicates a faster rate than this. Somewhat more satisfactory results have been reported for the regeneration of AgZ (loaded with elemental iodine) using 100% H_2 .² The possibility of

installing a pure H_2 system has been examined, and work is underway to prepare laboratory space for 100% H_2 regeneration tests. Longer regeneration tests (>24 h) show $>95\%$ of the loaded iodine can be removed using a dilute H_2 stream.

3.5.4 Integrated HTGR Off-Gas Treatment System

R. D. Zimmerman, U. S. Park, P. M. Hirsch,
P. W. Newman, and I. G. Khamis (GA)

The burner off-gas (BOG) component test has been completed except for the iodine absorber tests. These tests are being deferred until after the dissolver off-gas (DOG) component test to minimize corrosion by iodine in the system. All units tested (i.e., SO_2 adsorber, CO/HT oxidizer, and HTO adsorber) performed satisfactorily. In most test runs, the units showed higher decontamination factors (DFs) and conversion factors (CFs) than the design values. The DOG component test began with the NO_x converter test. Other tests to follow include those with the DOG moisture adsorber, radon decay bed, Kr/CO_2 adsorber, and I_2 adsorber.

Test results thus far verify the designs of system components. However, substantial difficulty with analytical instruments, especially the gas analyzers, delayed progress. The process control instrumentation necessary in a commercial-scale plant must receive more attention. Redundant analyzers should be installed to avoid a costly plant shutdown caused by faulty instruments.

The CVC* mass spectrometer for determining the amounts of krypton and other trace gases is undergoing assembly checkout. Krypton and mercury vapor have generated signals. Digital units are being checked and fine tuned for better resolution.

3.5.5 Freon Solvent Behavior Studies

V. Van Brunt (University of South Carolina)

The experimental portion of the activity for determining the solubilities of major off-gas components in R-12 includes the development of experimental procedures, the calibration of experimental equipment components over the desired temperature and pressure range, experimental operations for vapor pressure determinations, and operation for solubility determinations.

A second aspect of this activity concerns the refinement of thermodynamic consistency test procedures.

*Trade name of CVC Products, Inc.

REFERENCES

1. O. A. Meyer, *Dynamics of Nonisothermal Fixed Bed Adsorption*, Ph.D thesis, State University of New York at Buffalo (1966).
2. C. M. Slansky, B. D. Musgrave, B. R. Dickey, K. L. Rohde, and R. C. Shank, *Technical Division Quarterly Progress Report October-December 31, 1976*, ICP-1111 (March 1977).

4. Engineering Systems

M. J. Feldman

The scope of the work performed in the area of Engineering Systems spans the development of prototype equipment and the conceptual design of the Hot Experimental Facility (HEF). The design of prototype equipment is based on other research and development efforts, primarily those of the Process Research and Development and the Engineering Research sections of the CFRP. These efforts include the development of individual equipment items and systems and the interaction between various equipment items in series. The Integrated Equipment Test (IET) facility provides for the synergistic testing of equipment and also the testing of the remote operation capabilities and characteristics of these components and systems.

The conceptual design of equipment and systems to be incorporated into the HEF will be derived from these development efforts.

4.1 ENGINEERING ANALYSIS

The Engineering Analysis task performs studies and analyses in broad areas that overlap individual tasks and provide background and recommendations for decisions pertaining to programmatic guidance and directions.

4.1.1 Study of Reprocessing Facilities at a Nuclear Energy Center

M. J. Haire

A principal part of the subcontract with Exxon Nuclear Company was completed with the release of a four-volume report on "Evaluation of Reprocessing Facility Configuration for a Nuclear Energy Center." In this study, a central reprocessing complex within the center is envisioned for processing fuels from a variety of reactor types. The reprocessing requirements for reactors within the center and off-site satellite reactors generally increase and change with time. This study evaluated the optimal scenario, based on minimizing costs, for developing this reprocessing complex. Flow-sheets and equipment requirements were developed for eight different fuel types, from three reactor types, using either a uranium-plutonium or thorium-based fuel cycle. This information was incorporated into a computer program (REPRO) for estimating capital and operating costs. Concepts that proved to have a significant impact on costs include (1) the use of common facilities

(e.g., receiving stations and storage facilities) in conjunction with individual process buildings that are dedicated to a specific fuel type; (2) the provision of parallel processing lines within a process building when processing dissimilar fuel types; and (3) the processing of all available fuels in a single process line. Thus, the optimal reprocessing facility configuration was found to evolve with time. For those conditions where deployment of breeder reactors occurs at a slow rate, an initial flexible facility capable of reprocessing multiple fuel types is optimal. As the fuel load to the reprocessing facility grows, parallel lines to process the disparate fuel types become desirable. Finally, separate plants, each dedicated to processing specific fuel types, are most economical for large numbers of deployed reactors. Technologies requiring research and development prior to large-scale deployment of reprocessing plants are also discussed.

4.1.2 Tritium Confinement Study

W. R. Grimes and D. C. Hampson

This study, which is managed by ORNL with input from Exxon Nuclear Company at Richland, Washington and the Savannah River Laboratory, involves an in-depth examination of options for the reduction of tritium releases from future fuel reprocessing plants. The study will assess separate plants (each of 5 t/d capacity) for breeder and for light-water reactor (LWR) fuel. Options

for both the recovery of tritium and its management or disposal will be assessed. The base case plant for this study is assumed to use minimal water recycle and complete discharge of tritiated water to the atmosphere. The assessment will consider the incremental complexity of plant operation, incremental costs of the retention and disposal options, and incremental radiation risks to plant employees. It will also include an evaluation of the benefits of reduced exposure to the general public. The study goal is a final document — to be completed by the end of 1980 — that compares the cost, complexity, impacts, and state of development of the several options for both fuel types, and recommends priorities for development of the system(s) that appears optimal.

4.1.3 Thermal Analyses of Irradiated Fuel Subassemblies

M. J. Haire and C. A. Rhodes

Efforts have centered on documenting experimental and theoretical studies of thermal analyses during fuel handling and the shearing process.

4.1.4 Evaluation of Operating History of the Nuclear Fuel Services Plant at West Valley

D. C. Hampson

A short-term subcontract has been signed with E. Johnson Associates for the collection and analysis of operating data that was generated during the fuel reprocessing operations of Nuclear Fuel Services (NFS) at West Valley, New York (1966–1971). At present, much of the data is scattered at various organizations that were responsible for monitoring these operations. These organizations include the state of New York, the Nuclear Regulatory Commission, and the Department of Energy.

4.2 COMPONENT DEVELOPMENT

E. D. North

Component Development work is a follow-on to the Engineering Research effort. The Component Development mission is to design, fabricate, and demonstrate the performance of pilot-plant process equipment capable of remote operation and maintenance.

The dissolution and solvent extraction tasks are currently active. It is anticipated that feed preparation and product conversion tasks will be activated in the near future.

4.2.1 Dissolution

J. Q. Kirkman and F. E. Weber

The Component Development effort in the dissolution area is directed toward designing, constructing,

and testing a 0.5 t/d pilot-plant-scale dissolver system. The system consists of a remotely maintainable rotary dissolver integrated with the support equipment necessary to provide a functional continuous dissolution system. In this concept, the fluids flow in one direction through a horizontal rotating drum while the solids travel in the opposite direction.

Prototypical rotary dissolver

The rotary dissolver was operated extensively, accumulating an additional 325 h from all testing. The total accumulated operating time on the machine since installation is now 1360 h.

A majority of the dissolver operating time was spent performing tests investigating the solids handling characteristics of the dissolver drum. In particular, the backmixing of hulls through the interstage liquid transfer ports and the backscatter of solids through the solids transfer chutes were investigated. Both of these mechanisms promote solids accumulation within the drum causing higher than expected solids inventories.

The following conclusions can be drawn from the tests:

1. Preferential backmixing of small solids, such as fuel tubing and fuel spacer wires, through the interstage liquid transfer ports is a major contributor to drum inventories being higher than design expectations. The two or three transfer ports nearest the agitation baffle in a stage account for about one-half of the small solids backmixing.
2. Some backscatter of all solids sizes (wires, hulls, and shroud) is occurring through the solids transfer ducts between all stages. Although this backscatter is usually much less than that occurring through the liquid ports, there may be certain mass loadings within individual stages where the transfer duct mechanism is a significant factor. Congestion problems in the feed stage have been attributed to backscatter through the transfer duct from the first dissolving stage.
3. The incomplete transfer of solids from stage to stage occurs at very infrequent intervals and does not appear to be a significant factor in creating high drum inventories.

Investigations into methods for eliminating solids backmixing through the liquid transfer ports and measurements of the transport behavior of fuel pin spacer wires will be studied in future tests.

Dissolver support systems and process equipment

Support systems integrated with the rotary dissolver include feed, product collection, product clarification, NO_x scrubbing, and digital control systems. These

components performed satisfactorily without failure during the several hundred hours of accumulated dissolver testing.

Dissolver instrumentation

D. M. Fleischer and W. F. Johnson

Instrumentation activities in the dissolver system have focused on upgrading hardware, developing system software, and planning for the IET dissolver system instrumentation package.

Instrumentation hardware additions and improvements have included the following:

1. A new tank instrumentation module containing level and density bubble tubes in one removable package has been installed and tested. The performance of the module is an improvement over the previous system primarily because the ability to remove the assembly allows precise measurement of bubble tube separations. Improving the accuracy of the density reading by carefully controlling the air purge rates and individualizing the signal processing technique is presently being investigated.
2. Current-limiting electrical components were installed in the power supply to the dissolver motor controller to prevent repeated failures due to power transients in the supply line.
3. Installation of a new memory control board and correction of low voltage on a power supply unit have eliminated the intermittent failures experienced by the Bristol process controller.
4. Refinements to the solids feed control logic have been made to increase the accuracy of the hulls and shroud feed rates. Data on the weight of feed in the feed hoppers is automatically recorded. A real-time computer program, installed on the CODEDACS system, has been developed to calculate the solids inventory in the drum and average solids feed rates to the drum.
5. The quality of dissolver tank density signals has been significantly improved by filtering the input to the Bristol to remove distortions.

Planning for installation of the dissolver system in the IET has begun. A cost estimate for hardware and software necessary to integrate the dissolver system into the IET control scheme has been made. Drawings showing the control and monitoring signals to be sampled have been prepared to define the control system.

4.2.2 Solvent Extraction

J. D. Spencer and M. E. Hodges
(Savannah River Laboratory)

The 0.5 t/d contactor has been fabricated. The drive spindle is scheduled for delivery in July at which time

balancing tests can begin. The construction of the test facility is about one month behind schedule. Since the drive spindle is not available, the overall schedule has not yet been affected.

4.2.3 Feed Clarification

J. D. Spencer, S. F. Peterson, and G. C. Rodriques
(Savannah River Laboratory)

Iron oxide (Fe_2O_3) and carbon lampblack have been selected as cold stand-ins for undissolved solids. Preliminary tests showed that the iron oxide agglomerates slightly in 3 M nitric acid causing difficulty in clarification. The addition of a small amount (0.8%) of Dicalite filter aid improves clarification in a bomb type filter.

Variable speed De Laval Gyro-Tester centrifuges have been ordered for the centrifuge tests.

4.3 SYSTEMS DEVELOPMENT

N. R. Grant

The Systems Development work is directed toward the development of certain major mechanical components and systems required for fuel reprocessing. The goal of Systems Development is to design, fabricate, and operate pilot-scale, prototypic, remotely operable equipment.

The tasks assigned to this area are the disassembly and cutting systems, voloxidation equipment, the IET facility, receiving and storage systems, special remote systems, remote control engineering systems, waste handling systems, and procurement of depleted uranium fuel for use in equipment and process development.

4.3.1 Disassembly and Cutting

S. A. Meacham, E. C. Bradley, D. K. Lorenzo,
B. S. Weil, and W. F. Johnson

The disassembly and fuel cutting efforts are responsible for the mechanical preparation of the fuel for the succeeding steps of voloxidation and dissolution. To accomplish this objective, the task will develop equipment that will remove the undesired components, such as inlet and outlet nozzles, from the fuel assembly and cut the remaining portion of the assembly into short pieces. The sheared product will expose the contained fuel for subsequent oxidation (in the voloxidizer) and leaching (in the dissolver). The goal is to produce the necessary design, equipment, and data required for the successful operation of prototype units in the IET and similar equipment in the HEF.

Disassembly systems

Disassembly system (E. C. Bradley and R. E. Eversole). The final report on the disassembly systems integration study was received from Sundstrand Energy

Systems. An evaluation of the various disassembly scenarios (including the work performed by Sundstrand) is being prepared.

An assessment of the impact of handling the conceptual design study (CDS) and prototype large breeder reactor (PLBR) fuels in the disassembly system has been completed. Based on the current configuration of CDS fuels, there are no major changes required of disassembly system equipment to accommodate that fuel. However, because of the lower gas plenum in the current PLBR fuel designs, the handling requirements for that fuel have major impacts on the disassembly system equipment as well as the operating scenario. The disassembly system equipment can handle CDS, Clinch River Breeder Reactor, Fast-Flux Test Facility, and light-water reactor fuels without major equipment changes.

Plasma torch development (D. K. Lorenzo). All cutting tests with this equipment were completed and the plasma torch equipment was placed on standby status. A draft report on the plasma torch experimental program was completed.

Laser cutting development (B. S. Weil). The extension of the subcontract to develop design base information for a laser cutting system has proceeded on schedule. Two visits to United Technologies Research Center were made this quarter to observe cutting tests and provide contract guidance for the studies in progress. A study of the advantages of an active (motor driven) and passive (gravity driven) follower mechanism was completed. Both follower systems performed acceptably, but the passive follower is preferred because of its overall simplicity. The parametric tests to determine the optimum cutting conditions for cropping circumferential and kerfing (scoring) type cuts were completed. Efforts to disassemble full-scale dummy fuel assemblies were initiated. A study of the requirements and techniques for remotely aligning a laser beam in a hot cell is complete. The study of potential methods for determining the success of laser cutting on a fuel assembly has made limited progress.

Gas cooling/recirculation system (E. C. Bradley, R. E. Eversole, and D. K. Lorenzo). The gas cooling/recirculation system (GCRS) provides cooling for the fuel and containment for fission gases and particulates during all modes of shear and disassembly systems operation. The proposed HEF off-gas requirements result in a low off-gas flow rate which places severe sealing and operating requirements on the head-end equipment. Solutions to this problem include (1) increasing the off-gas flow capability, (2) relaxation of containment requirements for fission gas and

particulates, and (3) developing seals to minimize leakage to the shear and disassembly systems.

Shear systems

Remote shear (D. K. Lorenzo, D. C. Watkin, and J. H. Evans). The remote shear system advanced conceptual reference design has been completed. The bid package for detailed design and fabrication has been sent to potential vendors.

Remote shear model (D. C. Watkin). The one-fourth-scale model of the remote shear has been completed and a video and audio script has been prepared for videotaping. Still photographs have been made of the model and its components. This activity has been completed.

Prototype shear tooling (B. S. Weil). A prototypic set of shear tooling (compactor, fixed blade, and moving blade) has been fabricated. Dimension inspection, epoxy duplicates, and x rays have been made to document the as-built condition prior to initiating long-term tests.

Hydraulic fluid studies (E. C. Bradley). The stainless steel sample containers for the water-based hydraulic fluid irradiation tests were received and modified for irradiation in the ORNL cobalt garden. Fresh samples of the high water-based hydraulic fluid were received from both suppliers and loaded into the containers. The sample irradiation phase of the experiments was initiated at ORNL. Arrangements are underway to irradiate several samples at Argonne National Laboratory for the higher irradiation rates.

Remote shear instrumentation and control (W. F. Johnson, L. E. Ottinger, and J. K. Yarborough). The primary effort has been the preparation of the instrumentation and control input to the remote shear bid package. Three major items completed were: a system design report, an instrumentation and control system specification for the overall automatic control system, and a specification package, including drawings for the manual control station to be used by the seller for checkout and testing of the remote shear system.

4.3.2 Prototype Voloxidizer

J. E. Van Cleve, Jr.

The drawings for the advanced conceptual design were completed, the system design requirements document was drafted, and a cost estimate is being prepared based on this information.

4.3.3 Integrated Equipment Test (IET) Facility

J. G. Stradley, H. W. Harvey, and E. L. Nicholson

The IET task consists of the design and construction of modifications and additions to an existing area

at ORNL (7600 area) to provide an experimental engineering laboratory where systems testing on process equipment can be studied and evaluated and where remote handling and maintenance procedures can be developed.

The project is comprised of three basic parts. The first of these establishes an area where specific process equipment pieces that have evolved from development efforts can be joined together with supporting process equipment and operated as an integrated system. This portion of the IET facility is designated as the Integrated Process Demonstration (IPD). The second part consists of the construction of special facilities where testing of the remote operation and maintenance capabilities of process equipment can be performed. This area is named the Remote Operation and Maintenance Demonstration (ROMD) and consists of a cell mock-up area complete with simulated shielding, viewing systems, manipulators, cranes, and transfer devices. The third part is the General Plant Facility (GPF), which is comprised of structural modifications, utility system expansions, and construction of auxiliary facilities, all of which are required to support the IPD and ROMD experimental programs.

Planned experimental programs for the facility constructed by this project do not use plutonium or fission products but instead make use of natural uranium, process gases, and trace amounts of radioactivity.

This project interfaces closely with other elements of the CFRP in that process equipment being evaluated in this facility is supplied in a large part by separately funded Component Development and Systems Development efforts within the program.

The status of the Title II design and Title III construction for the three phases is as follows:

1. The IPD design effort including the Iodex system is slightly behind schedule (98% actual vs 100% scheduled). Design efforts on the Iodex and magnesium nitrate systems are being continued.
2. The design for ROMD is more than 99% complete vs 100% scheduled.
3. The GPF contractor completed the Title III contract on schedule in June.
4. The IET contract, which consists of installing major pieces of IPD and ROMD equipment, is scheduled to be awarded in September 1980.

Integrated Process Demonstration

The tanks, heat exchangers, evaporators, piping, and other IPD equipment items furnished by line-item funding are being procured by UCC-ND for installation

by the IET contractor. Title II design is 98% complete and procurement is in progress for all major pieces of equipment except the Iodex system.

Bids on the distributed data acquisition and control system (DDACS) have been reviewed, and a contract has been awarded.

The design of piping and instrumentation continues. Most of the advance procurement items have arrived. The remaining items, with the exception of DDACS and the Iodex system, will be completed by vendors during the next quarter.

Remote Operation and Maintenance Demonstration

Work on the contract with PaR Systems, Inc. for the integrated crane and manipulator system has continued. This equipment is scheduled for delivery in December 1980.

The television specification underwent revision based on input from several prospective vendors. Bids were received and a contract is expected to be issued in August 1980.

Contracts have been awarded and fabrication is proceeding on the simulated shielding walls, horizontal transfer devices, and vertical transfer devices. The System 50 master/slave manipulators, periscope, and mirrors have been received.

4.3.4 Receiving and Storage

S. A. Meacham and E. C. Bradley

Sodium removal program

Westinghouse Advanced Reactors Division has prepared a document describing test requirements for sodium wetting, flow characterizations, process development, and sodium removal testing of the liquid-metal fast breeder reactor fuel assembly test models.

The sodium wetting and removal vessels were installed in the sodium removal facility at Westinghouse. The steam turbine flowmeter and the 72-kW steam generator were also received and installed. The installation of all components of the sodium removal system is now complete, and equipment calibration has begun.

Wire-wrapping of the remaining 23 simulated fuel pins was completed. Seventeen of these pins were used to complete the assembly of the second proof-test model. The remaining six wire-wrapped pins have been designated as spares but will be used in the preparation of three-pin test trains for sodium corrosion exposure.

The first three-pin test train (dummy cladding) is now undergoing sodium exposure at 650°C.

Storage

Water exposure of the irradiated cladding specimens continued through the final ten months. (The total exposure was 22 months.) The specimens were photographed prior to shipment to Los Alamos Scientific Laboratory (LASL) for scanning electron microscopy analysis.

The draft of the final report on the corrosion III program has been prepared. The report presents results of the water exposure of irradiated cladding experiments (corrosion III) and summarizes the results of corrosion II experiments on unirradiated cladding specimens.

4.3.5 Special Remote Systems

R. Blumberg and W. F. Schaffer, Jr.

The objective of this task is to develop special remotely operable components and systems that are common to the major needs established by the requirements of the HEF conceptual design or by other tasks in Component or Systems Development. There are three systems currently being developed: remote connectors and jumpers, remote welding, and remote sampling.

Remote connectors and jumpers

Improved line connectors are being developed for use in the HEF. Since the present technology utilized Teflon seals, the primary goal of the development program is to develop a connector with a more radiation-resistant seal. Beyond this, it is desirable to improve remote handling characteristics, space requirements, overall reliability, and costs.

Remote cutting and welding

A program has been initiated to develop remote pipe cutting and welding to be used in lieu of remote line connectors for the interconnecting piping of the HEF. Ultimately, it could be used in place of remote connectors in many other situations in nuclear fuel reprocessing plants. The achievement of this goal would result in lower cost and improved reliability of piping systems.

A study has been completed by Battelle Columbus Laboratories (BCL) to investigate the concept and feasibility of developing remote pipe welding. The report concludes that remote pipe repair is feasible and with an adequate development program, there is a high probability of success. The study developed a sequence for the remote pipe repair and identified major equipment items including (1) equipment to clamp and align the piping and provide support and positioning of

functional tools, which is a straightforward design job; (2) pipe cutting equipment; (3) equipment to deburr and clean the cut ends of the piping; (4) welding equipment; and (5) inspection equipment. The welding equipment can be selected and modified from the commercially available tube welders. However, the report recommends that some development testing be made for the cutting, cleaning, and inspecting functions. Inspecting the finished weld is the most difficult development task, therefore, six nondestructive weld inspection techniques plus optical/visual examination are discussed. The problems with the remote utilization of the various inspection methods are the high radiation levels, the remote operation, and the restricted space. The report recommends the development of three methods of inspection — visual, by means of closed circuit television; radiography, using a fluorescent screen for displaying the resultant image, and ultrasonic detection.

An automatic tube welder and an automatic pipe outting device have been acquired by the CFRP. These devices will be tested and used to develop some of the information recommended in the BCL report.

HEF sampling system

Activity in the development of an HEF liquid sampling system has been concentrated on an advanced conceptual design of a prototype system to be built for testing and demonstration in ROMD. The work is being done by Los Alamos Technical Associates (LATA), and the conceptual design report draft is being reviewed.

The HEF sampling system must withdraw liquid from numerous process tanks, and then deliver that liquid to the analytical areas in identified bottles. As now conceived, the equipment to provide that function is embodied in three structural components: the sampling station, the vehicle, and the track.

The sampling station contains the piping, jet ejectors separation pot, and needle blocks that allow liquid to be withdrawn from the process tank and circulated through the bottle. There is an individual sample station (approximately 120 for the HEF) for each point to be sampled.

The battery-powered vehicle travels along the perimeter of the process cells on a track supported on the walls of the cells at a level just above the service penetration plugs to which the sample stations are mounted. At the designated stations, the vehicle handling system removes a filled bottle from the needles and replaces it with an empty bottle. The vehicle then returns to the load/unload station where the filled tray

is removed. The samples are then transferred to the analytical area.

The configuration of the system allows it to be maintained without interfering with the operation of the HEF process. Almost all the moving parts and failure prone components are mounted on the vehicle and are accessible for maintenance. The vehicle itself can easily be removed from the track to allow a spare to continue the operation.

4.3.6 Remote Control Engineering

**J. Garin (Westinghouse Advanced Reactors Division),
B. J. Bolfig, H. W. Harvey, and G. E. Smith**

Servomanipulator

L. Martin and B. J. Bolfig

Servomanipulator development involves two major activities: the procurement of an industry-standard system and the development of an advanced servomanipulator system. Each system consists of a set of two manipulator arms (50-lb capacity) and an interface package that has positioning equipment for cameras and an auxiliary hoist. The industry-standard servomanipulator system is currently being procured from Central Research Laboratories. This contractual activity is 30% complete. The assembly drawings are complete, and long-lead materials are being procured.

The advanced servomanipulator system is currently starting into the conceptual design phase. The system design requirements specification was completed and incorporated into a bid package. Two independent conceptual designs are being obtained. One design contract has been awarded to the Martin-Marietta Corp.; the other is expected to be awarded in the next quarter.

Analytical efforts are also underway to support the design performance of both systems. A computer code which simulates the behavior of the industry-standard manipulator has been completed. This code is being used to verify performance of motors and other components selected for the industry-standard manipulator. This code predicts response characteristics, stability, and can be used to simulate future control strategies.

Power arm

Manipulator arms with a capacity greater than 50 lb are required to perform remote handling tasks in the HEF. The power arm will be used to perform tasks up to 400-lb capacities.

A study initiated last year with MBAssociates was completed. This study yielded a power arm concept

design of unique capabilities. The general features are as follows:

1. The method of controlling the arm or the system architecture was developed.
2. Actuator design using a pancake motor and revolute gearing system was specified.
3. A unique kinematic (linkage) arm arrangement that allows performance of the task with greater dexterity was developed.

A demonstration of control methods recommended by this study has been initiated using an existing power arm. A PaR 3000 arm (1965 vintage) was converted for control with a minimaster and is operational. This system demonstrates improvements over the switch controls previously used with this type of arm.

Actuator design is also progressing to implement the concepts developed in the above report. The present efforts are in the area of selection of candidate motors, transducers, and gear trains. These are currently being ordered for testing and evaluation. The successful candidates will then be used in the prototype arm to be procured in the future.

Viewing and hand/eye coordination

D. W. Bible, J. A. Hawk, S. Coleman, and P. E. Satterfield

The hand/eye coordination study is progressing and is 80% complete. This study is being performed by the University of Florida and is aimed at generating generic information for viewing system designs for the HEF. The literature search on the state of the art in viewing systems for remote handling systems was completed. The system design requirements for man/machine interface was completed. The testing equipment requirements for viewing system selection is also complete. The specification for a viewing system is progressing.

The installation of a closed circuit television (CCTV) viewing system at Building 7601 has been completed. This viewing system is being used with a pair of CRL model 8 mechanical master/slave manipulators to perform typical remote work tasks (Fig. 4.1). In addition, a three-dimensional viewing system (Sub-Seas, Inc.) and a large screen projection TV system (1.8-m diagonal) have been procured and are being integrated into an ongoing operator console design.

The testing program is in progress to compare and characterize the capability of the above equipment for remote handling viewing. The testing is being administered by Oak Ridge Associated Universities (ORAU). This test station is being used by 14 experienced master/slave manipulator operators to provide base-line data for viewing equipment testing. The testing station

Fig. 4.1. CRL model 8 mechanical master/slave manipulators and closed circuit television viewing system.

accommodates a variety of viewing systems, lighting, and tooling. Viewing equipment selection testing will be completed by the end of next quarter.

A colorgraphics CRT terminal has been installed in the Thorium-Uranium Recycle Facility (TURF) operator's console to assist in controlling the TURF manipulator system. A graphical cell map program has been developed which shows the location of the manipulator system in the cell in relation to cell boundaries, and allows manipulator position commands to be entered. A manipulator system user's guide has been written to instruct operators on the use of the colorgraphics terminal in both the display and command modes.

Power transmission G. E. Smith

A battery study is being conducted by the Westinghouse Research and Development Center (W-R&D).

This study is directed toward the selection of a battery system to be used as an alternate power source for remote handling equipment. The study has progressed to the selection of three electrochemical system candidates for radiation environment qualification tests.

Lead-acid, nickel-cadmium, and iron-nickel batteries will undergo radiation dosage tests at Argonne National Laboratory to determine any immediate or long-term battery performance effects and to observe any component material damage that may be sustained during anticipated battery life. The total integrated exposure on the batteries will be equivalent to that expected to occur during the normal battery life in the HEF. The batteries will be charged and discharged while undergoing the irradiation.

Measurements will be made to determine if electrochemical performance of any battery system is modified significantly and whether the battery's construction material is significantly altered. The experiment is

planned to be controlled through parallel battery tests conducted elsewhere under nonradiation conditions.

Conceptual arrangements of battery packages for demonstration in TURF have been designed; all are commercially available.

Sensor development

P. E. Satterlee

This task involves the development of in-cell equipment that will provide additional information regarding the status of the manipulator systems, or sensors that will play an active part in the control of in-cell equipment.

A study has been initiated with SRI International for a conceptual design of an obstacle avoidance system that will prevent in-cell collision. The first phase of this study, a literature survey on obstacle avoidance techniques and applicable sensor technology, has been completed.

A development is also underway to use a solid-state diode array camera as a sensor to determine a target location for manipulator positioning. This system is currently operational and will later be installed in TURF and integrated with the power manipulator for effectiveness evaluation.

Telemetry system

S. Coleman, L. Martin, P.E. Satterlee, and T. Sliski

The object of this activity is to eliminate cabling between the remote manipulators, crane transporter systems, and the control stations.

In the area of remote signal transmission, the wireless transmission technique and signal multiplexing/demultiplexing are being investigated. This system will interface with a yet to be determined wireless transmission system.

4.3.7 Waste Handling

J. E. Van Cleve, Jr.

The contract with Pacific Northwest Laboratories was initiated to demonstrate continuous air melting and casting of stainless steel fuel hulls. Difficulties with the feed system, the crucible material, and billet withdrawal system have resulted in unsuccessful tests so far. The problems in each of these areas have been identified, and corrective measures have been taken. A second series of melts has been scheduled.

A contract with Rockwell International at Canoga Park, California, is being initiated. This work will determine if a metallic reaction vessel for the molten salt burner can be used to simplify maintenance problems.

4.3.8 Uranium for Tests

N. R. Grant and R. W. Knight

This task is divided into three areas: (1) the procurement of dummy assemblies as feed material for the disassembly/shear tests, (2) the procurement of depleted uranium dioxide as feed material for the voloxidizer and/or the dissolver, and (3) the disposal and/or recycle of uranyl nitrate.

The vendor fabricating dummy assemblies has resolved the problem of manufacturing type 446 stainless steel hex ducts. A problem was associated with the grain size of the raw material. The ducts are now being formed from sheet metal, and a seam along the length of the tube is being welded with an electron beam. The welded pieces will then be sized, and the two end pieces will be circumferentially electron-beam welded to the pad section. The fabrication of ten ducts to finish the base order is more than 50% complete, and delivery of the ten assemblies is expected in August. The first option for the next 27 assemblies should be negotiated during the next quarter. The delay in receiving the assemblies has not hindered the development program since the first ten assemblies delivered last year were used.

National Lead of Ohio has started to manufacture 30 000 kg of depleted U_3O_8 powder to be used in dissolver tests next fiscal year. The powder is being made by oxidizing scrap chips from machining operations performed routinely by National Lead. The order should be complete by the end of FY 1980.

4.4 HEF CONCEPTUAL DESIGN

The conceptual design of the HEF, a pilot-scale fuel reprocessing plant, is being performed by ORNL with the assistance of an architect-engineer and other subcontractors. The HEF is conceived as a versatile reprocessing test facility capable of handling the complete spectrum of breeder reactor fuels for the various fuel cycles now being considered. This will allow testing of breeder fuel cycles consistent with the overall breeder development program. The HEF will demonstrate advanced fuel reprocessing technology under plant operating conditions with irradiated fuel, and will establish a basis for the projection of reprocessing capability to the breeder industry.

The conceptual design activities are proceeding on schedule and within the budgeted cost estimate. Significant accomplishments during this report period include Department of Energy-Oak Ridge Operations (DOE-ORO) approval of the process flow diagrams, support building drawings, cost code of accounts manual, and an outline for the Conceptual Design Report (CDR).

4.4.1 Site and Support Buildings

O. A. Rogers

Drawings for the support buildings have been approved by the DOE-ORO. Site plans and site utility routing drawings are in process.

4.4.2 HEF Process Building

J. N. Herndon

The process building arrangement drawings for the CDR have been completed and submitted to the DOE for approval. These drawings depict the functional arrangement of the cell complex and out-cell process building support areas. An artist's concept of the process building can be seen in Fig. 4.2.

The process building structural drawings are being reviewed by UCC-ND. The cell liner details have been reviewed by UCC-ND and are being updated by Bechtel.

Zoning of the process building for ventilation and for radiation purposes was completed. Heating, ventilation, and air conditioning (HVAC) flow diagrams for all process building zones are being reviewed by UCC-ND.

A work task to define the instrumentation and controls concept for the general purpose cell equipment (Sect. 4.4.3) was initiated at Planning Research Corp. (PRC). This system will be required to interface and will be compatible with the balance of the plant instrumentation and controls system being designed by Bechtel.

4.4.3 General Purpose Cell System

D. Macdonald and K. L. Walker

Subcontractor engineering effort for the conceptual design of remote viewing, handling, and maintenance systems is well advanced. Planning Research Corp. has completed the design and has issued preliminary drawings of remotely operated crane and manipulator systems and the in-cell viewing and lighting equipment. The PRC task has been extended to cover design of the control system for all maintenance related equipment in the HEF.

Sundstrand Energy Systems has issued preliminary drawings of equipment concepts for decontamination systems and for scrap disposal equipment.

The General Atomic Company (GA) design group has prepared pictorial sequence diagrams of in-situ remote repair of chemical process modules and of the mechanical shear system.

Los Alamos Technical Associates (LATA) completed preliminary drawings of the HEF integrated automatic sampler system.

4.4.4 HEF Process Systems

E. D. North and F. L. Peishel

All 46 process flow diagrams have been approved by the DOE. The modifications resulting from the comprehensive review of these flow diagrams has been incorporated into the process equipment design.

ORNL DWG. 80-7109

Fig. 4.2. HEF process building.

The remainder of the conceptual design package has been issued for review and approval. This consists of the sizing of all major equipment on the process flow diagrams, the location of this equipment in logical groups within the standard HEF module frame, and the arrangement of these modules within the process cell.

4.4.5 Project Support

B. F. Bottenfield, F. C. Davis, and D. P. Reid

Final comments from UCC-ND, Bechtel National Inc., and DOE-ORO on the conceptual design criteria documents are being incorporated into these documents by UCC-ND and LATA. The approved document is

presently scheduled for publication in July 1980.

The outline for the conceptual design report has been approved by DOE-ORO and issued to the various participating organizations for use in preparing the document. Completion of the first draft of all sections is scheduled for August 1, 1980.

The cost code of accounts manual for the HEF cost estimating has been approved by DOE-ORO and issued to all participating contractors for use in preparing the cost estimate. The total project equipment list has been prepared and was reviewed in detail with the architect-engineer in mid-June.

Fabrication of two models which depict the overall project layout and the detail of a typical process cell section has been rescheduled from FY 1981 to FY 1980.

5. Technical Support

O. O. Yarbrow

The various generic studies reported in this section are not specific to functional research and development areas but are important to the overall program. The particular areas being studied include safeguards, nuclear engineering, the development of nonspecific process instrumentation, environmental assessment relative to the HEF, and operational planning for the Integrated Equipment Test (IET) facility.

5.1 IET OPERATIONAL PLANNING

W. W. Evans

Refined and updated equipment installation schedules for the IET have been issued.

Task leaders for the shear and disassembly, voloxidizer, and the dissolver have prepared data to interface the placement of integrated head-end equipment in the Remote Operation and Maintenance Demonstration (ROMD). In addition to equipment interface requirements, a list of specific design criteria for equipment in the ROMD area will be transmitted to UCC-ND Engineering for the detailed location study.

5.2 SAFEGUARDS

H. T. Kerr

The safeguards task is responsible for ensuring the development and implementation of effective safeguards technology for reprocessing plants as may be necessary to satisfy both domestic and international safeguards objectives.

5.2.1 Material Accounting Studies

R. D. Hurt and T. L. Hebble

The safeguards staff at Los Alamos Scientific Laboratory (LASL) is continuing to evaluate the measurement capabilities required to apply dynamic accounting to a remotely operated and maintained reprocessing plant. Agreement has been reached on the most appropriate mathematical form of the measurement error models to be used in this study. Specifically, these models will include a term to represent measurement system bias that is unaffected by instrument recalibration. Random error and error caused by inexact

instrument calibration will also be included in the measurement error models. This will allow the LASL staff to calculate the performance requirements for each dynamic accounting measurement in terms of its allowable (1) imprecision, (2) calibration error, and (3) long-term bias.

5.2.2 Microscopic Process Monitoring (MPM)

R. D. Hurt, J. W. Wachter, S. J. Hurrell, T. L. Hebble, A. B. Crawford, and G. A. Cotsonis

The first MPM minirun experiment was conducted at BNFP May 14–20. The second and third plutonium cycles were operated with natural uranium feed, and the data from about 100 process instruments was accessed at 4-min intervals by the safeguards computer system. Software was written to convert the raw instrument signals to engineering units, and the prediction equations and weight matrices required for MPM were developed in detail.

This minirun was intended to provide an opportunity to identify problems in process operation and data acquisition. Minor difficulties were encountered in both areas and are currently being remedied. Three miniruns scheduled to occur during the remainder of the fiscal year will be used to test the diversion detection sensitivity of MPM under a variety of circumstances.

An article describing the basic strategy of MPM was published in the spring 1980 issue of the *Journal of the Institute for Nuclear Materials Management*.

5.2.3 Penetration Monitoring

D. W. Swindle, Jr.

An evaluation of the HEF's penetration monitoring system concept design began at Sandia Laboratories

following in-house documentation of the reference system design. Model flow diagrams to be used in the evaluation have been prepared, and computer coding of the system's instrumentation configuration has been completed.

The preliminary draft of a report devoted to a literature survey of radiation detectors for reprocessing plant safeguards applications (which includes an evaluation of their operational characteristics) has been completed by Science Applications, Inc., of Palo Alto, California.

A testing program for "liquid-in-line" devices applicable as safeguards detectors has been completed. Three sensor types, the Sensall 621S ultrasonic sensor, the LQD optical sensor, and the Honeywell R7089 conductivity sensor were chosen for extensive testing. Results of the testing program following evaluation against criteria which addressed safeguards assurance, operability, reliability, and sensitivity under varying environmental conditions indicated that the LQD optical sensor is best. However, should penetration of the line be prohibited, the ultrasonic sensor is best, although it is not reliable for monitoring small diameter (1/4-in.) stainless steel pipe.

5.2.4 Operations Monitoring (OM)

D. W. Swindle, Jr.

A scoping meeting was held with the staff of Sandia National Laboratories Advanced Facilities Protection Division to determine the cooperative research and development activities for the OM task between the CFRP and Sandia for the remainder of FY 1980 and for FY 1981. Personnel assigned to the OM task at Sandia have prepared preliminary scoping papers on the applicability of OM in reprocessing plants.

5.2.5 Safeguards Support for the HEF Conceptual Design

D. W. Swindle, Jr.

Instrumentation for the HEF's physical protection system concept design has been determined by the HEF design team in cooperation with Bechtel National, Incorporated. Architectural drawings have been prepared identifying the general location of access control devices, intrusion detection devices, and closed circuit television (CCTV).

Safeguards design criteria and guidelines for the HEF conceptual design have been finalized for submission to the DOE. These criteria and guidelines address requirements for the incorporation of an entry-control

system, an intrusion detection system, a material control and accounting system, and a safeguards response and control system in the HEF. In addition, detailed guidelines have been developed for the design of the Safeguards Control Center and the Secondary Alarm Station, which serve as the principal and backup safeguards centers of operation.

A Quality Assurance Plan has been prepared and approved which describes the responsibilities and required actions necessary to provide a high degree of confidence in the ability of the HEF's safeguards system conceptual design to meet applicable federal and international requirements.

The structure of the safeguards instrumentation and control (I&C) system for the HEF has been defined for the HEF design team and Bechtel National. Data which characterized the safeguards I&C system as part of the total HEF I&C system were determined. Drawings depicting this relationship have been initiated by Bechtel, and target completion dates for incorporation into the conceptual design report have been identified.

A meeting was held to review the conceptual design of the HEF's physical protection system. Participants included personnel from the DOE-ORO, UCC-ND, Sandia Laboratories, and the HEF design team. As part of this meeting, Sandia presented the preliminary results of effectiveness evaluations on the HEF security system design and potential sabotage targets.

5.2.6 CFRP Participation in the IAEA's International Working Group for Reprocessing Plant Safeguards (IWG-RPS)

H. T. Kerr, R. D. Hurl, and D. W. Swindle, Jr.

Meetings of the IWG-RPS were held in Dunblane, Scotland, and Chester, England, during March and April. The CFRP provided a conceptual description of a reprocessing facility that will be used as a common basis for comparing various safeguards methodologies. The safeguards team also submitted papers describing the basic strategy of the microscopic process monitoring concept and the concept of an integrated system approach to effective safeguards.

5.3 NUCLEAR ENGINEERING

J. F. Mincey

The nuclear engineering task function is to provide guidance and computational support in the areas of criticality, safety, radiation-level estimation, and shielding design.

5.3.1 Interim Validation Report Using Existing Breeder-Typical U+Pu Critical Benchmark Data

R. T. Primm III, E. T. Tomlinson, and J. F. Mincey

Any major design effort involving criticality evaluations relies on the accuracy of an assortment of neutronics codes. These codes are used to predict the critical condition (k_{eff} values) for systems of interest as well as for processing neutron cross-section data used in the k_{eff} calculations. If these codes and libraries are to be used for setting subcritical limits, the American National Standards Institute (ANSI 16.1 and 16.9, 1975) requires that such codes and cross-section data be frozen or archived and then gauged or validated against well-defined critical experiments.

The validation of existing benchmark criticality data against frozen codes and cross-sections continued; three classes of existing experimental data have been reported. The validation effort is about 90% complete.

5.3.2 Interim HEF Criticality Philosophy Report

J. F. Mincey

A preliminary draft has been prepared for review and comment.

5.3.3 Interim HEF In-Cell Radiation-Level Report

E. J. Allen, S. N. Cramer, and J. V. Pace

In order to provide timely radiation-damage estimates for the design of in-cell remote handling equipment, an engineering-design computer code, GHF, is being developed. The GHF will be tested against sample problem results obtained with more elaborate (and less design-suited) calculations.

One limitation of the GHF is that it does not directly account for wall scattering when calculating in-cell radiation levels. This effect, when estimated with more rigorous (and expensive) methods, was found to be fairly predictable at various locations in the cell with a rather simple methodology. By incorporating this result into GHF, a rather significant improvement in accuracy is possible.

A frozen version of the ORIGEN point-depletion code was obtained, along with supporting cross-section libraries. A procedure is being developed to facilitate the performance of these calculations with properly weighted cross-section data.

5.3.4 U+Pu Solution Experiments

R. T. Primm III

Two changes have been made to the general description of these experiments. Several steel-reflected experiments will be added to the list for the unpoisoned

300- to 500-g/l U+Pu solution experiments. Also, calculational studies are in progress to design a vessel suitable for conducting some of the needed FTR fuel pin/fissile solution experiments in order that the poisoned U+Pu solution can be used to maximum advantage.

5.3.5 U+Pu Oxide Experiment Design

J. F. Mincey and E. T. Tomlinson

Fabrication has begun on prototype equipment for these experiments. The equipment will be used in tests to ensure that the U+Pu oxide will be sufficiently uniform in density and moderation. The equipment will also be used to confirm proposed solutions to the decay-heat removal problem caused by the presence of plutonium.

5.4 INSTRUMENTATION AND CONTROL SYSTEMS

S. M. Babcock

This task is responsible for the development of process instruments and controls and data management systems for fuel reprocessing applications in the areas of process control, accountability, and safeguards.

5.4.1 Systems Development

J. A. McEvers, R. M. Tate, and R. W. Tucker

A first version of a graphics package which allows the plotting of data transmitted to the DEC PDP-11/40 computer system in either real time or from archive magnetic tapes has been completed. The utility of this package has been delayed because of problems created during conversion to a new operating system.

Intercomputer communications studies have been delayed because of delays in the delivery of purchased DECNET software.

5.4.2 Sensor Development

H. H. Ross, J. E. Strain, D. T. Bostick, M. L. Bauer, R. Harper, D. D. McCue, R. L. Anderson, G. N. Miller, W. W. Johnston, and A. W. Renshaw

Analytical sensor development

The effects of gamma radiation on quartz optical cables needed for the prototype in-line photometer are being investigated. No measurable interference due to fluorescence of quartz was seen in the 270- to 700-nm spectral region of interest when 50 cm of 3-mm-diam quartz cable was exposed to a gamma-ray flux of 1.3×10^6 R/h.

During bench tests of the prototype in-line photometer, magnetic interference with the behavior of the

photomultiplier tube occurred. Magnetic shielding is being investigated.

The spectral determination of uranium in organic streams is essentially complete. The effect of acid concentration, TBP concentration, and temperature have been evaluated and combined into a simple algorithm. The phase separation observed at elevated temperatures can be compensated for by use of a blank wavelength. The nitration of TBP in strong nitric acid to produce a yellow color may require the determination of the "uranium equivalent" of the organic phase prior to its introduction into the partitioning column. The TBP-nitration compound is now under study.

Low-level uranium standards have been prepared for use in the laser-excited fluorescence determination of uranium in aqueous waste streams. The objective will be to measure uranium at the 10-to-100-ppm level independent of foreign ions.

Electrochemical detection of hydrazine and hydroxylamine was investigated in flowing streams by cyclic voltammetry and platinum electrodes. The detection of hydrazine and hydroxylamine appears practical at flow rates up to 5 l/min.

Laboratory tests of a mock-up of the prototype free-acid monitor have demonstrated satisfactory operation. Components of a prototype unit are being fabricated.

Physical sensor development

A new ultrasonic torsion wave level and density sensor that can be operated in both torsional and extensional modes, thereby allowing temperature compensation, has been received. Tests performed on an air-water mixture with up to a 15% void fraction indicated that the change in density in a two-phase mixture can be correctly measured.

Testing has begun on a capacitance level detector and an air-launched ultrasonic level detector. Initial tests of a commercial thermal flowmeter (designed for radiation resistance up to 10^8 rads) have uncovered a large error transient upon initiation of low flows following a period of zero flow resulting from a constant heater power.

A nuclear magnetic resonance flowmeter sensor has been fabricated and is ready for interfacing to associated electronics.

5.4.3 Special Measurements

C. W. Ricker, G. L. Ragan, G. G. Slaughter,
E. D. Blakeman, M. M. Chiles, C. M. Smith,
F. S. Sweeney, and D. N. Fry

Nondestructive assay

An experiment has recently been completed in which spent nuclear fuel was nondestructively assayed

by a proof-of-principle active neutron interrogation device. The fuel assayed was 4.5-year-cooled BWR fuel from the Dresden I reactor. The original enrichment of the fuel was 2.337% ^{235}U . After 27 000-MWd/MT burnup, the fuel contains about 1.1% fissile isotopes. For this experiment, the fuel rods were chopped into 5-cm segments and loaded into each of three 1-l cans (3 kg of heavy metal equally distributed). The cans, designated A, B, and C were assayed in the following seven configurations: ABC, AB, BC, CA, A, B, and C. Similarly, three standard cans were prepared by chopping and loading rods of available 1.5% fresh fuel. These cans were assayed in the same configurations as the spent fuel cans.

The fissile contents of each of the spent fuel cans is being determined by comparing spent fuel and fresh fuel assays and then checking against chemical analysis following dissolution in the Solvent Extraction Test Facility. To date, can C has been chemically analyzed, and agreement with preliminary assay determinations has been within 2%. A correction for the presence of fission products in the spent fuel that are not in the fresh fuel standard is being investigated using neutron transport calculations.

Efforts to develop a method for determining the isotopic content of spent fuel subassemblies continued with calculations for the absorber method. A new library of cross-sections was prepared, including the fission product isotopes and other materials of interest. Calculations indicate that gadolinium and europium will enhance the proportion of signature neutrons in ^{241}Pu relative to ^{239}Pu .

Another possible method of measuring isotopic ratios is called the "resonance self-indication method," which makes use of resonant detectors in a neutron-transmission-type measurement. The presence of a particular isotope in a subassembly will result in maximum attenuation of the neutron flux at the energies of maximum response of the detector coated with that particular isotope. A comparison between this measurement and a similar measurement made with a nonresonant detector provides a means for determining the relative isotopic composition of the fuel in a subassembly.

Measurements that were differential in energy were performed on thick samples of ^{235}U and ^{238}U using a ^{235}U -coated fission chamber, a boron-coated chamber, and the Oak Ridge Electron Linear Accelerator as neutron sources. These measurements indicate that ^{235}U and ^{238}U can be unequivocally distinguished and measured.

Work on the development of a leached hulls monitor continued with the investigation of data regarding self-shielding effects. Comparative counts were made of four pellets varying in fissile content from 500 mg

^{235}U (1.3 cm in diameter by 5 cm long) to 2.7 g ^{235}U (1.3 cm in diameter by 1.3 cm long). As expected, self-shielding effects are severe for the 2.7-g pellet (corresponding to an enrichment of 19.8% ^{235}U). However, self-shielding effects were moderate for a 1.5% enriched pellet, for which the count rate was suppressed less than 30% below the expected value from a linear extrapolation.

By placing a cadmium sleeve around the sample cavity, it was observed that the self-shielding effect was virtually eliminated, although the signal/source background ratio was severely reduced. Nevertheless, this approach shows promise for the quantitative assay of containers with large lumps of enriched material.

Mechanical surveillance

Vibration data analysis from a 32-h dissolver run indicates that accelerometers mounted on the outside of the shroud are capable of monitoring the mixing and transfer of material within the dissolver. The signatures from the sensors have been characterized with respect to the level of angular segmentation required and the frequency bands, which are dominated by mixing phenomena, feed material, the exit of material, and material transfer from stage to stage. Typical frequency spectra expected from the motor and bearings have also been defined.

A plan has been written for tests to resolve remaining questions regarding the required angular sensitivity, cross-talk between accelerometers, vibration paths between the drum and the shroud accelerometers, and the dependence of results on the position of the out-of-shroud accelerometers. The fabrication of electronics to allow the simultaneous signal conditioning of all 14 channels of data is underway.

5.5 ENVIRONMENT AND SAFETY

M. B. Sears

This task is responsible for providing guidance and assessments to ensure that public health, safety, and environmental concerns are addressed in the HEF design.

The results of an HEF siting study within the Oak Ridge Reservation were presented to the DOE-ORO Land Use Committee. The DOE has informally indicated that the Land Use Committee has ruled that any of the five primary sites identified in the study would be compatible with the Land Use Plan.

Savannah River Laboratory continued its safety assessment of the HEF. Significant accomplishments to date include (1) the completion and review of a draft report on the off-gas treatment and aqueous waste processing systems, and (2) the completion of a final draft report on the atmosphere control and electrical distribution systems.

6. HTGR Fuel Reprocessing

B. L. Vondra

Process research and development activities on high-temperature gas-cooled reactor (HTGR) fuel reprocessing technology are being conducted at General Atomic Company (GA) with supporting hot-cell studies at ORNL. These programs are devoted to studies and analyses of reprocessing requirements for HTGR and gas-cooled breeder reactor (GCBR) fuels and HTGR-specific head-end studies.

6.1 STUDIES AND ANALYSES

H. C. Carney and L. Abraham, General Atomic Company

The FY-1980 task objectives are to (1) complete an interim assessment of Hot Experimental Facility (HEF) processes and systems for GCFR fuel reprocessing, (2) assess the applicability of HEF process systems to demonstrate HTGR spent fuel processes, and (3) complete an interim assessment of potential methods to reduce the volume of HTGR reprocessing head-end solid waste.

6.1.1 Evaluation of GCFR Fuel Reprocessing

V. H. Pierce, J. S. Rode, and L. Abraham

The task group completed and documented the interim assessment of HEF processes and systems for GCFR fuel reprocessing.

6.1.2 HTGR Fuel Reprocessing Development Requirements

L. Abraham, V. H. Pierce, H. C. Carney, and B. B. Haldy

As part of an HEF suitability assessment to demonstrate HTGR fuel reprocessing, studies were begun to define spent fuel storage and process system and equipment requirements. The proposed HTGR spent-fuel reprocessing rate is consistent with the 200-kg/d HEF thorium-uranium fuel separation processing capability.

6.1.3 HTGR Fuel Reprocessing Head-End Alternatives

J. S. Rode and L. Abraham

The task group completed and documented the interim assessment of potential methods to reduce the volume of HTGR reprocessing head-end solid waste.

6.2 HEAD-END STUDIES

W. S. Rickman, General Atomic Company

6.2.1 United States-Federal Republic of Germany Cooperation on Gas-Cooled Reactor Spent Fuel Treatment

W. S. Rickman, J. B. Strand, and N. D. Holder

The United States and the Federal Republic of Germany (FRG) are cooperatively developing spent fuel treatment technology as part of the February 1977 Umbrella Agreement to develop gas-cooled reactor technology. As a part of this agreement, a Joint Prototype Head-End Facility (JPHF) is in conceptual design. It will be a prototypical-scale, cold facility to demonstrate remote operation and maintenance, and will be cooperatively designed, built, and operated using both U.S. and FRG reference fuel types.

Approximately 2000 FRG spherical fuel elements were successfully processed in the GA cold pilot plant. Three German engineers observed and participated in the tests. A number of minor differences in processing characteristics between U.S. and FRG fuel were identified, and equipment will be modified for the next demonstration test. The preliminary test results are summarized under unit operation sections.

6.2.2 Fuel Element Crushing

J. B. Strand and N. D. Holder

Minor modifications to the UNIFRAME crusher allowed FRG fuel spheres and graphite spheres to be successfully crushed at a throughput rate of 100 spheres/min. Oversize crushing was required.

6.2.3 Crushed Fuel Element Burning

W. S. Lessig and D. E. Fields

In two preliminary burner campaigns, the 0.20-m (8-in.) primary burner verified the feasibility of burning crushed FRG material in a fluidized-bed combustor. Although both runs were terminated early due to a net buildup of fines, the final bed product contained only half the maximum carbon content permissible and satisfied the acceptance criteria for that burner system.

Based on the results of the preliminary burning tests using crushed FRG fuel spheres, modifications for the 0.40-m (16-in.) primary burner system were designed to prepare it for the U.S.-FRG Joint Fuel Processing Demonstration (JFPD) this summer. The modifications will enhance fine carbon combustion, improving the overall combustion efficiency. Equipment modifications in progress include a longer burner vessel, additional above-bed gas entry ports, and water cooling between the cyclone and the off-gas filters.

6.2.4 Particle Classification, Crushing, and Crushed Particle Burning

J. B. Strand and N. D. Holder

Crushed FRG fuel particles were processed in the zigzag classifier to determine the extent of kernel-hull separation. Microscopic examination of the product indicated excellent separation. Burn tests indicated that the kernels had less than 1% burnable carbon.

Silicon carbide hulls were also separated from burned-back crushed particles to reduce the quantity of hulls for a fuel dissolving experiment.

About 17 kg (38 lb) of primary burner product from FRG fuel-sphere burning was processed in the new Mark 2 crusher. Microscopic examination detected no unbroken particles. Part of this crushed product was burned in the 0.1-m (4-in.) secondary burner to furnish material for two dissolver experiments.

6.2.5 Dry Solids Handling

N. D. Holder, H. H. Yip, and K. Higuchi

Component testing

The test loop which cyclically transports 115 kg (254 lb) of FRG crushed graphite spheres has successfully completed more than 200 cycles.

Shear cell tests on FRG crushed fuel, primary burner product, and secondary burner product were completed.

HEF voloxidizer isolation valves

Testing continued in the knifegate-Gemco-valve test rig for light water reactor (LWR) shear product. The

shear product jammed the Gemco valve. A 152-mm (6 in.) cast-iron Gemco valve was ordered so that extensive modifications could be made to alleviate the jamming.

A vibratory feeder was tested and works well for smaller pieces of shear product. However, large pieces hang up on the throttle mechanism. Wear tests will be conducted using only the smaller material.

The shear product hung up on the knifegate valve seat. To eliminate this problem, the edge of the knifegate valve seat was slanted.

6.3 LABORATORY DEVELOPMENT

R. D. Zimmerman and G. E. Benedict,
General Atomic Company

6.3.1 Inertial Filter Tests

G. J. Hostbjoer

Inertial filtration is achieved by pumping the solution stream to be clarified through the interior of a porous metal tube at a linear velocity of ~ 6 m/s (20 ft/s). A pressure gradient across the tube transfers clear effluent into the annular section; particulate matter smaller than the nominal 0.5μ pore size is trapped in the interior. A subsurface membrane develops when very fine particles penetrate a short distance into the porous tube wall. This membrane filters the particles even finer. During blowback, the annular space is pressurized to 138 kPa (20 psi) higher than the system inlet pressure, forcing particles from the pores back into the main flow stream. The interval and duration of the blowback can be set as required by the solution being processed.

Shakedown tests were initiated using $\text{Zr}(\text{OH})_4$ as the particulate (50% smaller than 0.1μ). During an 8-h period, about 100 l of a 0.1 wt % $\text{Zr}(\text{OH})_4$ suspension was run through the filter. Approximately 80 l of clear filtrate was recovered. Further work will try to shorten filtration time without appreciably decreasing filtrate quality.

6.3.2 Dissolution Tests

Two dissolution runs with FRG fuel were conducted in the 0.13- and 0.20-m (5- and 8-in.) pilot-plant-scale dissolvers. For each test, Thorex dissolvent was added in stoichiometric proportions to yield a 1 M heavy metal nitrate product. In one run, the SiC portion of the secondary burner ash was included in the feed charge, and in the second run, the SiC hulls were removed by air classification prior to dissolution. Approximately 56 h of no-heel mode operation were required to dissolve 3.17 kg (7 lb) of JFPD fuel kernels. For the 0.2-m (8-in.) pilot-plant-scale dissolver, this represents a minimum heavy metal throughput rate of 56.6 g/h (0.125 lb/h).

Based on the results of these tests, air classification of SiC hulls from the JFPD secondary burner product has several advantages: (1) the operational upsets caused by SiC hulls could be eliminated; (2) separating the insoluble hulls from the mother liquor would be unnecessary; and (3) since the insoluble content of the remaining heel would be considerably reduced, heel mode operation could be used. Heel mode operation can significantly increase the dissolver throughput.

6.3.3 Bench-Scale Laboratory Studies

R. G. Wilbourn

Zirconium scrubbing kinetics study

G. J. Hostbjoer and R. G. Wilbourn

A study on the effect of uranium saturation and acid concentration in a 30% TBP-normal paraffin hydrocarbon (NPH) system on zirconium removal from solvent extraction scrub sections was completed. Two opposing factors which influence the zirconium transfer rate were apparent. The aqueous-to-organic (A:O) equilibrium distribution ratio favored the low acid 1 M HNO_3 system, and the calculated system kinetic constants favored the higher acid 3 M HNO_3 system. The system kinetics seem to be the dominating influence on zirconium removal. The data further indicated that more than one species of zirconium was involved in the system studied.

Dissolution rate of FRG fuel kernels

R. G. Wilbourn and S. A. Dunlap

The JFPD secondary burner ash used in this study was ~75 wt % (10Th:1U) O_2 kernels (500- μm diam) and ~25 wt % inert SiC hulls. The JFPD fuel dissolution rate in Thorex was considerably slower in preliminary tests than the sol-gel ThO_2 kernels previously studied at GA. Microscopic examination of partially dissolved JFPD kernels revealed relatively large crystals in the (10Th:1U) O_2 fuel spheres. X-ray diffraction analysis confirmed these findings. The sol-gel ThO_2 kernels were subjected to similar treatment with Thorex, but no large crystals were observed. The slower dissolution rate of the JFPD kernels may reflect the difference in kernel fabrication, particularly the sintering temperature. The large crystals may be responsible for the slower dissolution rate of JFPD fuel spheres. An increase of ~200 in the JFPD (10Th:1U) O_2 surface area (produced by crushing the spheres) yielded a dissolver feed solid with a Thorex dissolution rate approximately equal to whole sol-gel ThO_2 kernels.

Alternate solvent evaluation

R. G. Wilbourn

The phase separation times of 1.09 M (51 vol %) TEHP:NPH organic from aqueous phases were measured for application in solvent extraction pulsed column operations. Extraction and stripping simulation tests found organic:aqueous separation time to be essentially the same for 1.09 M TBP or TEHP organic layers. The phase separation times for 1.09 M TEHP in contact with aqueous layers of 0.2 M sodium carbonate or hydrazine oxalate (to simulate solvent washing) were consistently longer than the 1.09 M TBP solvent. Prior mixing of 1.09 M TEHP with 3 M nitric acid and 0.2 M sodium carbonate did not eliminate the emulsion problems of simulated solvent washing tests.

Solvent cleanup studies

The feasibility of regenerating process solvent by passing it through pretreated silica gel has been studied. In one test a solvent (30% TBP-NPH) feed solution containing 150 ppm uranium as the DBP complex was prepared and fed to the sorbent bed at a volume velocity of 0.64 $\text{ml min}^{-1} \text{cm}^{-2}$; uranium breakthrough occurred. Apparently, the kinetics of uranium-DBP sorption on pretreated silica gel are slow and will require low solvent flow rates for efficient loading of the silica gel bed.

The ability of silica gel to remove uranium-DBP from an aqueous hydrazine oxalate solution was studied in a separate set of experiments. In these tests, the solvent, which contained 150 ppm uranium as the DBP complex was contacted with 0.2 M hydrazine oxalate solution to transfer the uranium-DBP to the aqueous phase. These studies showed that silica gel sorption of uranium-DBP from the hydrazine oxalate solution is more efficient than removing the complex from the solvent phase. Silica gel treatment of the hydrazine oxalate wash solution may allow wash solution reuse, reducing waste stream volume and radioactive element content. Similar tests are planned to determine the path of the zirconium-DBP complex.

$\text{ThC}_2 - \text{HNO}_3$ reaction

S. A. Dunlap

Carbide- HNO_3 reactions may form organo-metal complexes with heavy metals and fission products which could affect the solvent extraction cycle. Two fuel kernel types are used to study these reactions: (1) Fort St. Vrain (FSV) fissile with a 4:1 ratio of thorium-to-uranium carbide, and (2) weak acid resin (WAR) fissile with uranium oxycarbide only. Both types of kernels

are crushed in a simulated 10:1 ratio thorium-to-uranium feed solution, which is spiked with ^{95}Zr tracer. The aqueous phase is refluxed and/or mixed before being batch-contacted with 30% TBP-NPH solvent. Then, the TBP organic phase is stripped with three equal volumes of 0.01 *M* nitric acid contacts. The Davies-Gray titrimetric method analyzes the strip solution samples for uranium. Uranium and ^{95}Zr distribution are measured for heavy metal and fission products retained in the solvent phase. Initial results show that carbide reaction products have little effect on solvent retention of uranium and zirconium.

6.4 HOT LABORATORY STUDIES

V. C. A. Vaughn, Oak Ridge National Laboratory

6.4.1 Fuel Rod Crushing and Primary Burning

C. L. Fitzgerald and O. L. Kirkland

Six experiments on the behavior of a BISO-BISO carbide fuel combination (FBTE-2) during reprocessing

were completed. The fuel (50 fuel rods per experiment) were crushed and burned at 850°C in 5% O_2 –95% N_2 at 0.8 m/s fluidizing velocity. The fluidized bed burner was difficult to control. Temperature excursions of 1050°C, followed by “flame-outs”, occurred in half of the experiments. The burner products were free-flowing with no evidence of sintering. The weight of primary burner products (burner + cyclone + filter fines) averaged about 50% of that of the crushed fuel feed.

The burner products were dissolved using a procedure designed to test its selective dissolution in 8 *M* HNO_3 . Reflux times in HNO_3 were varied from 1 to 24 h in the six experiments. The residues from the dissolutions of the fertile fractions were refluxed in Thorex reagent for 12 h.

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