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UC-77

CONSOLIDATED REPROCESSING PROGRAM

**QUARTERLY PROGRESS REPORT
FOR THE PERIOD ENDING FEBRUARY 28, 1979**

**Prepared under
Contract EY-76-C-03-0167
Project Agreement No. 53
for the San Francisco Operations Office
Department of Energy**

MASTER

DATE PUBLISHED: MARCH 1979

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GENERAL ATOMIC COMPANY

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GA-A13255 - September 1974 through November 1974
GA-A13366 - December 1974 through February 1975
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ABSTRACT

This publication continues the quarterly series presenting results of work performed under the Consolidated Reprocessing Program at General Atomic Company. Results of work on this program prior to June 1974 were included in a quarterly series on the HTGR Base Program.

The work reported includes the development of unit processes and equipment for reprocessing of High-Temperature Gas-Cooled Reactor (HTGR) fuel, the design and development of an integrated pilot line to demonstrate the head end of HTGR reprocessing using unirradiated fuel materials, and design work in support of Hot Engineering Tests (HET). Work is also described on trade-off studies concerning the required design of facilities and equipment for the large-scale recycle of HTGR fuels in order to guide the development activities for HTGR fuel recycle.

CONTENTS

1. SUMMARY	1-1
2. STUDIES AND ANALYSES - TASK 100	2-1
2.2. ACTIVITY	2-1
2.2.1. Evaluation of GCFR Fuel Reprocessing	2-1
2.2.2. Criticality Analysis	2-1
2.2.3. Availability Allocations	2-2
2.2.4. Spent Fuel Storage	2-2
2.2.5. Recycle Plant Data Base	2-2
2.2.6. Flowsheet and Material Balance	2-2
References.	2-3
3. HEAD-END STUDIES - TASK 310	3-1
3.1. FUEL ELEMENT SIZE REDUCTION	3-1
3.1.1. Sequential Operations	3-1
3.1.2. UNIFRAME Throughput Verification.	3-2
3.2. CRUSHED FUEL ELEMENT BURNING	3-5
3.2.1. FY-78 Sequential Operations	3-5
3.2.2. Unscreened Feed Operation - 0.40-m Primary Burner.	3-8
3.2.3. Automation Development - 0.20-m Primary Burner.	3-11
3.3. PARTICLE CLASSIFICATION, CRUSHING, AND CRUSHED PARTICLE BURNING	3-12
3.3.1. Summary	3-12
3.3.2. Air Classification	3-12
3.3.3. Particle Crushing	3-17
3.3.4. Secondary Burning	3-18
3.4. DRY SOLIDS HANDLING	3-20
3.4.1. FY-79 Sequential Operation.	3-20
3.4.2. Pneumatic Transport Component Testing System	3-23

4.	OFF-GAS TREATMENT - TASK 340	4-1
4.1.	SUMMARY	4-1
4.2.	SYSTEM DESIGN	4-2
4.3.	FABRICATION AND INSTALLATION	4-3
4.4.	PREOPERATIONAL SYSTEM CHECK	4-23
4.5.	SYSTEM SAFETY.	4-23
4.6.	SCHEDULES AND MILESTONES	4-24
	Reference.	4-24
5.	SOLVENT EXTRACTION - TASK 350	5-1
5.1.	ANNULAR PULSE COLUMN EXTRACTION TESTS.	5-1
5.2.	CYLINDRICAL PULSE COLUMN PLATE TESTS	5-2
5.2.1.	Extraction Column Tests With 12% and 24% Free Area Straight Slotted Plates	5-2
5.2.2.	Stripping Column Tests	5-3
5.3.	ROBATEL CONTACTOR TESTS	5-8
	References	5-8
6.	LABORATORY STUDIES - TASK 370.	6-1
6.1.	VERTICAL CENTRIFUGE TESTING.	6-1
6.2.	DISSOLUTION TESTS.	6-2
6.2.1.	Introduction	6-2
6.2.2.	Solid-Liquid Contact Improvement Study	6-3
6.3.	BENCH-SCALE LABORATORY STUDIES	6-3
6.3.1.	Effect of Thorium on Hydrolysis of Tributyl Phosphate	6-3
6.3.2.	Iodine and Ruthenium Volatility in Thorium Fuel Reprocessing.	6-5
6.3.3.	Three Phase Compositions in Thorium, Uranium, Nitric Acid, TBP, Diluent System	6-7
	References	6-7

FIGURES

3-1.	Size distribution of burner feed	3-3
3-2.	Size distribution comparisons for tests LF-809 and LF-810. .	3-10
3-3.	Pneumatic transport components test system	3-24

FIGURES (Continued)

4-1.	Modified CO/HT oxidizer	4-5
4-2.	Burner off-gas treatment system	4-7
4-3.	Dissolver off-gas treatment system.	4-8
4-4.	Gas mixing unit. DOG humidifier and iodine vapor generator are at left and right lower corners	4-9
4-5.	General system installation. From left to right: CO ₂ /Kr, Rn, moisture, SO ₂ , I ₂ backup, and I ₂ adsorbers	4-10
4-6.	Detailed installation of adsorption vessels with associated auxiliaries. Vessel at left is NO _x catalytic converter, vessel at right is tritium/moisture adsorber	4-11
4-7.	Typical installation of on-line process auxiliaries such as heater, flow meter, control valve, and temperature and pressure measurements	4-12
4-8.	Carbon monoxide tube pack	4-13
4-9.	General arrangement of CO/HT oxidizer system	4-15
4-10.	CO/HT oxidizer system installed	4-17
4-11.	CO/HT oxidizer, cooling air blower, and exhaust heat exchanger	4-18
4-12.	Gas-analyzer system	4-20
4-13.	Calibration gas mixing unit	4-21
4-14.	Off-gas system control room	4-22
5-1.	Onset of cyclic flooding in the stripping column	5-4
5-2.	Hold-up in the stripping column with standard nozzle plates for temperature of 54°C (130°F) and A/O ratio of 1.25	5-6
5-3.	Hold-up in the stripping column with zebra cartridge plates for temperature of 54°C (130°F) and A/O ratio of 1.25	5-7
6-1.	Pulse sparge design	6-4
6-2.	Effect of aging on TBP hydrolysis	6-6

1. SUMMARY

The evaluation of GCFR fuel and blanket reprocessing requirements was initiated in January 1979. An interim objective is to identify features of GCFR spent fuel and irradiated blanket elements that are different from LMFBR fuel systems and could therefore affect currently specified reprocessing and facility development requirements.

Two modifications were made to the UNIFRAME after the FY-77 sequential operation. A rotary valve was added to the discharge of the screener, and a prelube system was added to the primary and secondary movable jaw Pitman bearings.

Twenty crushed fuel elements, totaling 2500 kg, were burned back to yield a low carbon content fuel particle product as part of FY-78 sequential operations. All acceptance criteria were met. Peak carbon burn rates of over 900 g/min were attained during the run; the design burn rate is 800 g/min.

The air classification, particle crushing, and secondary burning processes as a part of the pilot plant sequential operations were completed. Only one operating problem was encountered in the completion of all three of these sequential operations.

The crusher product removal system was used to convey crushed fuel elements from the UNIFRAME to the crusher product bunker. A constant feed rate of 0.5 kg/s was maintained by allowing a heel to build up in the screener prior to starting the rotary valve. A rotary valve speed of 40 rpm delivered the feed at a rate so that crushing was completed before the screener was emptied.

Installation of the engineering-scale off-gas treatment system is essentially completed, with only one heat exchanger yet to be delivered and installed. The heat exchanger is a late addition to the system for the purpose of adsorption bed regeneration. Piping walkdown and system cleaning have been completed. System leak check and correction are underway and almost completed.

Electrical work was resumed in FY-79. All electrical system installation and wiring, including the Diogenes digital process control system and Hewlett-Packard data acquisition system, have been completed. Electrical system functional checkout is continuing.

The capacity of critically safe cylindrical solvent extraction columns limits plant size because of the limited cross-sectional areas of plutonium or U-233 bearing processing columns. There is a need to increase the cross-sectional area of these columns. This can be accomplished through use of a column having an annular cross section. The preliminary testing of the pilot-scale annular pulse column has been completed.

A Sharples Model P850 vertical continuous centrifuge has been installed in the General Atomic Company pilot plant. The vertical centrifuge will be tested for operability and solids-liquid separation efficiency under various operating conditions related to nuclear process applications. A vertical configuration was selected primarily because of the advantages involved with its adaptability to remote maintenance.

2. STUDIES AND ANALYSES - TASK 100

2.1. INTRODUCTION

The objectives of this task are to (1) define unique reprocessing development and facility requirements for spent GCFR fuel compared with present LMFBR reprocessing plans and facility designs, (2) evaluate the impact of HTGR fuel design changes on reprocessing requirements and costs, and (3) confirm flowsheet requirements for HTGR reprocessing and waste streams, and prepare material balances as required.

2.2. ACTIVITY

2.2.1. Evaluation of GCFR Fuel Reprocessing (100.1)

The evaluation of GCFR fuel and blanket reprocessing requirements was initiated in January 1979. An interim objective is to identify features of GCFR spent fuel and irradiated blanket elements that are different from LMFBR fuel systems and could therefore affect currently specified reprocessing and facility development requirements.

Data are being compiled on GCFR fuel and blanket design, materials, and spent fuel composition. Information is being obtained from consolidated fuel recycle program (CFRP) elements on current reprocessing flowsheets, process and equipment development, and facilities design pertinent to breeder fuel reprocessing.

2.2.2. Criticality Analysis (100.2)

A draft of the HRRF dry head-end preliminary criticality safety evaluation has been completed. Although the evaluation consisted primarily

of isolated component reactivity parameterizations, it can be concluded that equipment comparable to that in the cold pilot plant will be adequately safe provided that suitable layout requirements are imposed. A major constraint on the dry head-end reprocessing subsystem is that flooding must be precluded downstream from the UNIFRAME size reduction discharge hoppers.

2.2.3. Availability Allocations (100.2)

A topical report on the availability of reprocessing systems in an HTGR fuel recycle facility (Ref. 2-1) was released and distributed.

2.2.4. Spent Fuel Storage (100.2)

A draft report giving the results of the HTGR spent fuel storage study (Ref. 2-2) was completed and is undergoing internal review.

2.2.5. Recycle Plant Data Base (100.2)

Update of the commercial recycle plant data base was completed for HTGR high gain fuel. A User's Manual for the computerized data base is in preparation; the draft is about 90% completed.

2.2.6. Flowsheet and Material Balance (100.3)

A topical report on the reprocessing flowsheet and material balance for MEU spent fuel (Ref. 2-3) was released and distributed.

REFERENCES

- 2.1. Holder, N. D., J. N. Sharmahd, and J. S. Rode, "Availability of Reprocessing Systems in an HTGR Fuel Recycle Facility," DOE Report GA-A15028, General Atomic Company, November 1978.
- 2-2. "HTGR Spent Fuel Storage Study," General Atomic Company, to be published.
- 2-3. Abraham, L., "Reprocessing Flowsheet and Material Balance for MEU Spent Fuel," DOE Report GA-A15145, General Atomic Company, October 1978.

3. HEAD-END STUDIES - TASK 310

3.1. FUEL ELEMENT SIZE REDUCTION (SECONDARY SUBTASK 311)

3.1.1. Sequential Operations

Two modifications were made to the UNIFRAME after the FY-77 sequential operation. A rotary valve was added to the discharge of the screener, and a prelube system was added to the primary and secondary movable jaw Pitman bearings. The rotary valve was installed to provide a constant feed rate from the crusher to the transport system so that transport velocities, and therefore particle breakage, could be minimized. The prelube system was installed to provide lubrication to the Pitman bearings during startup. Both systems operated satisfactorily.

3.1.1.1. Crushing. The first six fuel elements were individually crushed and transported to the crusher product bunker. The next four elements were crushed in a span of 24 min. During the crushing of these four fuel elements, the next fuel element was dropped into the primary crusher as soon as the previous fuel element had cleared the screener. The remaining 10 fuel elements were crushed in this semicontinuous mode as feed was required for the primary burner. During each crushing operation, the rotary valve was not started until 30 s after the block was dropped into the crusher. This allowed product to build up in the screener to provide a constant feed rate to the transport system.

3.1.1.2. Sampling. The crushed fuel was sampled at the exit of the crusher product bunker as it was transported to the primary burner feed bunker. The samples were taken using the cross-cutting sampler. A continuous sample was taken of each of the 14 batches transported to the

primary burner feed bunker. The results indicated an overall particle breakage of 8.7% for the fissile particles and 1.5% for the fertile particles. A composite size distribution for all 14 samples is shown in Fig. 3-1.

The fissile particle breakage of 8.7 wt % is not significantly different from the particle breakage experienced during the FY-77 sequential operation. No samples were taken prior to transporting the crushed fuel to the crusher product bunker so it is not known whether this breakage was caused by crushing or transport.

3.1.1.3. Material Balance. Each fuel element was weighed prior to crushing. The total weight of fuel crushed was 2469 kg. The change in the crusher product load cells was monitored each time material was added to or removed from the bunker. The weigh cells indicated a total weight gain of 2460.7 kg during transport to the bunker and a total weight loss of 2464.2 kg during transport from the bunker. Cleanout of the crusher yielded only 0.5 kg of material. The MUF for the crushing operation was -0.3% or -0.2% depending on whether the weight gain or loss in the crusher product bunker was used.

3.1.1.4. Conclusions and Recommendations. The lubrication system modification to prelubricate the Pitman bearings worked very well. This system, however, is not considered prototypical. The particle breakage was not significantly changed from the FY-77 sequential operation by the addition of the rotary valve. It is recommended that this be further investigated in the next sequential operation. The throughput rate of the UNIFRAME is well above the requirements of four blocks per hour. The material holdup of 0.5 kg and the MUF of 0.2% to 0.3% are acceptable.

3.1.2. UNIFRAME Throughput Verification

During the FY-78 sequential operations and previous crusher tests, it was noted that the residence time of a fuel block in the primary crusher was approximately 90 s, while the residence time in the secondary crusher

Fig. 3-1. Size distribution of burner feed

was approximately 270 s. It was felt that throughput could be improved by making these times closer to each other, therefore balancing the crushing load between the two crushers.

The UNIFRAME was modified by adding a shim to the primary crusher toggle assembly to change the close side setting from 51 mm (2.0 in.) to 38 mm (1.5 in.). The nip angle was readjusted to 14°, its previous setting, after the shimming was completed.

Crushing tests were made using unloaded fuel blocks. The data from these tests are being reduced and will be compared with prior data to determine the effect of changing the close side setting.

3.2. CRUSHED FUEL ELEMENT BURNING (SUBTASK 312)

3.2.1. FY-78 Sequential Operations

3.2.1.1. Summary. Twenty crushed fuel elements, totaling 2500 kg, were burned back to yield a low carbon content fuel particle product as part of FY-78 sequential operations. All acceptance criteria were met. Peak carbon burn rates of over 900 g/min were attained during the run; the design burn rate is 800 g/min. The new gravity fines recycle system performed exceptionally well with a final heel that was less than 0.02 wt % of the total throughput; the design heel allowance is 0.50 wt % of the total throughput. Product carbon content was less than 1 wt %. Average particle breakage in the burner system was 2.3 wt %.

3.2.1.2. Feed Material. Twenty crushed fuel elements comprised the burner feed. The average size distribution is shown in Fig. 3-1. Feed composition was:

Burnable carbon	83%
TRISO/BISO fuel particles	<u>17%</u>
	100%

Feed particle breakage as received by the primary burner was 8.7 wt % of the WAR TRISO particles and 1.5 wt % of the BISO particles.

3.2.1.3. Burner Operation. This burner run followed Operating Procedure 524401, Issue C, throughout. Initial burner startup proceeded by adding 200 kg of fresh feed to the burner, fluidizing it with 2300 slpm of CO₂, and heating to 700°C. The lower induction coil was used until the bed had been heated to 300°C, then both upper and lower coils were actuated. This was done because bed expansion is not sufficient to fill both coils until a 300°C bed temperature is reached. Plenum O₂ flow was then increased to ignite the bed; flow rates greater than 700 slpm O₂ were maintained thereafter to oxidize the fluid bed graphite.

Off-gas filter pressure drops were uniformly low (less than 30 in. H₂O) throughout the run.

Fines combustion was initiated after recycling fines at 6 rpm rotary valve speed to heat the fines stream to greater than 400°C. Mid-reactor O₂ flow peaked at 360 slpm during the first 18 h of the run. After this time, flows of 200 slpm were required to burn fines, indicating that the recycling fines inventory was very low and that only newly generated fines were being burned. Above-bed temperatures declined as the fines inventory decreased.

Fresh feed was continuously added to maintain bed carbon inventory. Initial bed carbon content was 83 wt % (same as crushed fuel elements); this was reduced to 0 wt % over a 26 h period so that an interim product removal could be effected. Fuel particle inventory was raised above 200 kg over this 26-h time span. This was sufficient for an induction-heated bed carbon burnout, as described in the operating procedure.

A partial product removal of 100 kg of material was then made, followed by adding additional fresh feed to the bed to make up for the loss in bed volume. Burner restart was followed by increased fines burning due to the added carbon content of the fluid bed.

Fresh feed rate was controlled to keep a minimum of 10 wt % carbon in the bed. After all the fresh feed had been added, a final bed carbon burnout was carried out with induction heater assist to maintain temperatures.

3.2.1.4. Instrumentation. Computer programs were developed for recording and processing data during the primary burner portion of the sequential operation. Twenty-four channels of data were scanned and recorded on magnetic discs every 15 s. In addition, the burn rate and the total carbon burned were calculated and displayed every 15 s. This was the

first run during which the operator could monitor the burn rate during operation.

Selected data, including the time, gas concentration, burn rate, and total carbon burned, were printed out. The printout was used by operators to check long term trends, and to give relief operators a quick summary of previous shifts.

3.2.1.5. Product Material. Initial product removal material (99.9 kg) had the following composition:

96.2	wt % BISO kernels
2.6	wt % WAR TRISO particles
<u>1.2</u>	wt % carbon
100.0	wt %

This material was 98.5 wt % between 425 and 850 μm .

Particle breakage was 2.0 wt % of the BISO kernels and 18 wt % of the WAR TRISO particles.

Final product removal material (270.9 kg) had the following composition:

77.5	wt % BISO kernels
21.7	wt % WAR TRISO particles
<u>0.8</u>	wt % carbon
100.0	wt %

This material was 98 wt % between 425 and 850 μm .

Particle breakage was 0.3% of the BISO kernels and 13% of the WAR TRISO particles. The plenum backflow material (23.6 kg) had the following composition:

84.2 wt % BISO kernels
12.7 wt % WAR TRISO particles
<u>3.1 wt % carbon</u>
100.0 wt %

This material was 91 wt % between 425 and 850 μm . Particle breakage was 6.3% of the BISO kernels and 24% of the WAR TRISO particles.

Fines heel material, which only amounted to 520 kg, contained 94 g non-combustible material, which was 75% SiC, 7% U_3O_8 , 18% ThO_2 , and 0.1% TRISO WAR particles.

Fines heel density was 0.36 g/cm^3 . Size distribution was 0.7 wt % greater than 250 μm , 8 wt % between 63 and 250 μm , and 91.3 wt % less than 63 μm .

3.2.1.6. Conclusions and Recommendations. Gravity fines recycle has been conclusively demonstrated as a reliable and easy to control system for fines combustion in the 0.40-m primary burner. All other burner sub-systems also worked very well, as demonstrated both by their smooth operating characteristics and the product attributes of low carbon content and low breakage.

From a process viewpoint, the burner system, as now constructed, is capable of reprocessing HTGR fuel. Future work on remote maintainability and more complete automation should be pursued to bring them up to the available levels of technology.

3.2.2. Unscreened Feed Operation - 0.40-m Primary Burner

3.2.2.1. Summary. A 9-h burner run was successfully completed on the 0.40-m primary burner using feed material that was crushed through the UNIFRAME

crusher without a screener or an oversize pulverizer. No difference in operating characteristics was noted in comparing this run to those with oversize screening and crushing. The fluid bed temperature profile varied less than 10°C over the 1.7 m axial bed length. No agglomerates were noted in the product stream.

3.2.2.2. Feed Material. Initial fluid bed material consisted of a crushed FSV type fuel block (109.5 kg) loaded with TRISO fuel particles. No screener or oversize pulverizer was used during crushing of this block.

Size distribution of this material is shown in Fig. 3-2. It is of interest to note that 5 wt % is between 4.75 mm (3/16 in.) and 6.30 mm (1/4 in.). Comparative distribution of material crushed with the screener and pulverizer (Test LF-809) is also shown in the graph.

During burner operation, 220 kg of TRISO fuel particles were continuously added to build up a product bed large enough for an induction-heater-assisted final bed carbon burnout.

3.2.2.3. Burner Operation. Operating Procedure 524401, Issue E, was followed throughout the burner run.

The peak burn rate was 700 g/min. The off-gas filter pressure drop reached 23 in. H₂O. Filter temperature was controlled at 600°C while fluid bed temperature was kept at 900°C. Off-gas CO concentration averaged 12 vol % while O₂ averaged 2 vol %. Total heatup time was 1-1/2 h. Mid-reactor oxygen flow averaged 300 slpm during peak fines burning periods. The rotary fines valve was operated at 6 rpm during the burner run.

3.2.2.4. Product Material. Product carbon content was 5.3 wt %. No fuel particle agglomeration was observed. Gas distributor flow characteristics remained unchanged after the run.

Fig. 3-2. Size distribution comparisons for tests LF-809 and LF-810

The high product carbon content is due to the presence of broken fuel particles in the product sample. No free graphite was in the sample, based on microscopic examination. It is thought that a significant portion of the particle breakage may have occurred during product removal and transport, since the broken particle carbonaceous material would have oxidized in the burner during final bed burnout procedures.

It should be noted that the fuel particles used in this test were FSV types that were rejected because their coatings did not meet specifications. The high particle breakage, therefore, might not be representative of on-specification material.

3.2.2.5. Conclusions and Recommendations. The success of this burner run indicates that the 0.40-m primary burner and associated feed and product transport systems are capable of processing fuel element material that has been crushed through the UNIFRAME crusher without the screener or oversize pulverizer. It is recommended that these equipment items be removed from the UNIFRAME for all future feed preparation for the 0.40-m primary burner.

3.2.3. Automation Development - 0.20-m Primary Burner

Automation development is now proceeding on the 0.20-m primary burner. With gravity fines recycle firmly established, we are now ready to fine tune the process controller and establish a fully automatic mode of operation. New gas flow meters, with improved accuracy and immunity to induction heating interference, have been added. Accurate gas flow measurements are critical in calculating the carbon burn rate. The combined fines pipe and above-bed cooling air loops have been separated to achieve better cooling air flow distribution.

A supervisory interface link was established between the Diogenes process controller and a Hewlett Packard 9825 A minicomputer. This link allows direct access to process measurements, constants, and calculations stored in computer memory, thus further enhancing data acquisition.

Complete monitoring of a process control loop is now possible (1) remotely through software programming of the supervisory computer, or (2) locally at the Diogenes operator's console. The entire memory of Diogenes can now be read and stored on magnetic tape and then reprogrammed via the supervisory interface in 2 or 3 min.

During FY-79, we will be automating the steady-state portion of the run by July 15, the startup sequence by August 15, and be fully automated with tailburn and shutdown by September 30.

3.3. PARTICLE CLASSIFICATION, CRUSHING, AND CRUSHED PARTICLE BURNING (SECONDARY SUBTASK 313)

3.3.1. Summary

The air classification, particle crushing, and secondary burning processes as a part of the pilot plant sequential operations were completed. Only one operating problem was encountered in the completion of all three of these sequential operations.

Parametric studies on air classification have begun.

The design of an advanced fuel particle crusher was completed.

Installation of an advanced gas distributor on the 0.10-m secondary burner was completed. Checkout has begun.

3.3.2. Air Classification

3.3.2.1. Pilot Plant Sequential Operations. The classification of the approximately 384 kg of primary burner product was accomplished in three runs. The first was approximately 104 kg of product from combustion of the first 10 fuel elements. The remainder of this product was returned to the burner as a starting bed for the remaining combustion. To avoid exceeding the capacity of the product bunker's load cells, the final

burner product was removed in two parts, one of 181 kg and one of 99 kg. These were also classified separately. A gas velocity of 6.0 m/s was used in all three runs.

Feed rates were obtained from the rate of weight loss from the feed bunker measured by load cells. After the feed reached the level of the bellows on the bunker, it no longer affected the load cells; therefore, to ensure cleanout the classifier was operated for a few minutes after the readings showed no further weight loss.

At the startup of Part I, the power to the vibratory feeder was increased in steps. At 70% power, the feed reached design level, and this setting was used for the remainder of Part I and all of Part II. However, shortly after the start of Part III the feed rate dropped and the power was raised to 100% to compensate. The drop in rate at 70% power was attributable to the segregation of particles in the primary burner. This produced feed which was predominantly BISO kernels for Parts I, II, and the initial portion of Part III. As the feed became richer in the lighter TRISO particles, the rate reduced. This was substantiated by a simultaneous increase in the proportion of overheads indicated by the load cells on the product bunkers.

The total time required for classification was 7 h 26 min at an overall average feed rate of 0.81 kg/min, not including the cleanout times.

The total weights of the feed, underflow, and overheads were obtained from load cell readouts taken before and after completion of each classification run with all equipment turned off to reduce vibration. After each run, the samples were removed and materials in the filter bunker were discharged after blowing back the filters. The samples and filter materials were weighed on a balance.

The segregation of BISO and TRISO fuel particles was evident from the material balance results. In Parts I and II, the overheads content was only 4% of the product, while in Part III it was 78%.

A consistently positive MUF of over 3% was obtained in all three parts. This was due to errors introduced by the feed bunker's suspension system.

Two types of continuous in-line automatic samplers were used to obtain samples concurrently with classification. The overheads (fissile streams) were sampled with a gravity feed sampler designed at GA. The underflows (fertile streams) were sampled with a Vezin laboratory sampler.

The materials discharged from the filter bunker were split-sampled in a 12-to-1 riffle splitter. The larger underflow samples were also split to obtain a more reasonable size sample for analysis.

The Vezin sampler took a reasonably sized sample of about 1% of the total underflow. To reduce the quantity of samples taken by the gravity sampler, one of the sample tubes was sealed off prior to Part III. Upon removal to accomplish the operation, it was discovered that a fragment of graphite was blocking one of the tubes. This apparently occurred during Part II, and resulted in a smaller sample from that of Part I. The deliberate blocking of the tube reduced the sample size from 6.1% in Part I to 2.4% in Part III.

Each of the overhead, underflow, and filter material samples was analyzed for size distribution, carbon content, and broken fuel particles. The size distributions were determined by screen analysis. After screening, the mesh fractions were recombined and ignited in a muffle oven overnight at 900°C to obtain the weight loss from carbon combustion. The residues were screened into plus and minus 425 μm fractions, and each fraction was leached for uranium and thorium content. The insoluble materials were crushed, reburned, and leached again for uranium and thorium content. Uranium present from broken TRISO fuel particles was detected in the initial leaching. Comparison of this result with the total uranium from both leachings provided an estimate of the percentage of broken particles.

To obtain an estimate of broken BISO fuel kernels, it was assumed that the thorium in the +425 μm fractions was from whole kernels and the thorium in the -425 μm fractions was from broken kernels. Since whole BISO kernels generally are larger than 425 μm , and crossover is expected to be almost entirely from fragments less than 425 μm , the assumption is considered reasonable.

As a result of normal segregation, the quantity of fines was expected to be greater in the last material removed from the burner. This was substantiated by the size distribution results. Parts I and II each contained less than 3% product smaller than 425 μm , while Part III contained over 6%. Also, as expected, the overheads contained more fines than the underflows, and the largest percentage of fines was in the filter materials. However, a relative inefficiency of the cyclone was indicated by the quantity of product larger than 425 μm in the filter materials.

A low overall percentage of product over 850 μm indicated few agglomerated particles or large fragments of graphite were present in the burner product. A small overall percentage of product less than 425 μm indicated that particle breakage may not be excessive.

The total burnable carbon in all of the classifier product was calculated to be 1.6%. Since the carbon content is unchanged by classification, it is also representative of the carbon in the burner product, plus any burnable carbon coatings subsequently exposed by TRISO particle breakage during pneumatic transport and classification.

After primary burning, the residual carbon was expected to be present mainly as fines and, as a result of segregation, present mainly in the last material removed from the burner. This was substantiated by the carbon analyses. The overheads and filter materials contained a total of 5.8 kg of carbon and the underflows contained only 0.5 kg. The percent carbon was almost 35 times greater in the overheads than the underflows, and 74 times greater in the filter materials than the underflows. The products from Part III contained 5.6 kg of carbon while the products from Parts I and II combined contained only 0.7 kg.

The overall particle breakage was calculated to be 3.2% of the fertile and 22.8% of the fissile particles. Of the fertile particles found in the fissile streams (overheads and filter products) 68.22% and 40.26%, respectively, were broken while only 1.98% of the fertile particles were broken in the fertile streams. A higher percentage of broken particles was also shown for fissile particles in the fertile stream than for fissile particles in the fissile stream. These results clearly show the tendency for broken particles to result in crossover.

The uranium and thorium results from the analyses for broken fuel particles were also used to calculate the heavy metal content and distribution in the classifier products.

Overall thorium recovery was 98.85% in the fissile streams (underflows) and 1.15% in the fertile streams (overflows and filter materials). In both Parts I and II, over 99.5% of the thorium was recovered in the fissile streams. This reduced to 89% in Part III.

Overall uranium recovery was 93.26% in the fertile streams and 6.74% in the fissile streams. In Part III, over 99.4% of the uranium was recovered in the fissile streams. This reduced to 71% and 57% in Parts I and II, respectively.

The increase in the crossover of thorium in Part III and uranium in Parts I and II is attributed to the tendency of entrainment of the minor component in the major component stream and to broken particles.

3.3.2.2. Classification Studies. The design of advanced fuel types will affect the efficiency of classification proportionally to the differences in the densities and diameters of the fissile and fertile fuel particles. These effects are being studied to establish parameters for the separability of advanced fuel types, such that fuel particle designs will be able to take separability into account.

Velocities at which various mesh fractions of WAR TRISO and PSC type TRISO fissile and fertile fuel particles become overflow or underflow in the engineering scale classifier have been determined. Study of the data generated and comparison with calculated terminal velocities is underway.

3.3.3. Particle Crushing

3.3.3.1. Pilot Plant Sequential Operations. A 45.6-kg portion of the classifier overhead fissile stream from Part III of the classification was used as feed to the fuel particle crusher as a continuing part of the pilot plant sequential operations.

The roll speed throughout the crushing run was 40 rpm.

The feed weight was obtained from load cells on the feed bunker. These readouts were taken before and after crushing with all equipment turned off to reduce vibration. Crushing rates were not obtainable from the rate of weight loss from the feed bunker because normal vibrations from the crushing caused large variations in the load cell readouts. Instead, the average crushing rate was obtained from the total weight crushed and the total run time. Forty-four minutes after startup, the crusher stopped due to a tripped circuit breaker. The apparent average crushing rate to that time was 1.0 kg/min.

It was assumed that crushing had been completed because the load cell readouts were constant and tapping on the feed lines indicated they were empty. Therefore, a restart was not attempted. However, during dismantling to investigate the cause of the stoppage, 0.3 kg of uncrushed feed was found.

The crusher's gap was 0.055 cm (0.0215 in.) prior to the startup and 0.06 cm (0.0235 in.) after the run. This sample was obtained by crushing 0.5 kg of the Part III overheads material after an initial inspection of the crusher and at the conclusion of the crushing and checking the final gap. Since all of the material was crushed at the

final 0.06 cm gap, it may be somewhat coarser and contain more unbroken fuel particles than the average material crushed for secondary burner feed.

3.3.3.2. Advanced Particle Crusher Design. The present design of the fuel particle crusher requires very close manufacturing tolerances, a high quality assurance follow up, and precision assembly. A new design has been completed which avoids these problems, provides increased bearing protection, and reduces side plate wear. In addition, an in-line sampler has been designed to facilitate determination of crusher product characteristics during testing. A complete design report is in progress.

3.3.4. Secondary Burning

3.3.4.1. Pilot Plant Sequential Operations. The feed material for the secondary burning portion of the sequential operations was 45.6 kg of the product from crushing a portion of the overheads (fissile stream) from Part III of the classification. A gravity pneumatic feeder was used to add the material to the burner and this was accomplished in about four minutes.

No operating problems were encountered throughout the burning. Heat-up to ignition temperature took about 45 min. Oxygen was introduced at that time and ramped up to 70% of the inlet gas concentration in about 10 min. Total oxygen utilization, the main burn period, lasted 1 h 35 min at 280 lpm oxygen flow. The fluidizing gas velocity during this period was 0.8 m/s. After oxygen breakthrough in the off-gas, the oxygen was gradually reduced to 20% of the feed concentration to keep the oxygen concentration below 20% in the off-gas. After 50 min, the fluidizing gas velocity was reduced over a period of 10 min to 0.45 m/s for tail burning. The run was terminated after 20 min of tail burning. The total run time after ignition was 3 h 5 min.

Total product recovered was 31.30 kg. It contained only 0.30% burnable carbon. However, due to the widening of the crusher gap 8.72% of the fissile fuel particles remained unbroken through secondary burning.

3.3.4.2. Remote Gas Distributor. In FY-78, a remotely replaceable gas distributor was designed and constructed. This unit, in addition to its remote features, locates the potentially troublesome flange connection outside of the high-temperature zone. The distributor has been installed on the 0.10-m secondary burner. Checkout of the burner for testing of the remote distributor has begun.

3.4. DRY SOLIDS HANDLING (SECONDARY SUBTASK 314)

3.4.1. FY-79 Sequential Operation

3.4.1.1. Crusher Product Removal System. The crusher product removal system was used to convey crushed fuel elements from the UNIFRAME to the crusher product bunker. A constant feed rate of 0.5 kg/s was maintained by allowing a heel to build up in the screener prior to starting the rotary valve. A rotary valve speed of 40 rpm delivered the feed at a rate so that crushing was completed before the screener was emptied. The oversize crusher valve was opened after the screener was emptied to empty the oversize crusher.

The blower was sheaved to provide the 18 m/s transport velocity selected as optimum during the FY-77 sequential operation. No samples were taken during this operation. The weight of the fuel elements crushed was 2469 kg. The weighing system on the crusher product indicated a weight gain of 2460.7 kg when the material was transported to the bunker, and a weight loss of 2464.2 kg when the material was removed.

3.4.1.2. Primary Burner Feed System. Crushed fuel was transported from the crusher product bunker to the primary burner feed bunker using the primary burner feed system. The blower on this system had been sheaved to provide the 18 m/s transport velocity selected as optimum during the FY-77 sequential operation. A feed rate of 0.25 kg/s was used. The primary burner feed system was operated by the burner operators from the control room during the entire run.

A sample taken during each transport operation was analyzed for size distribution, burnable carbon, thorium, uranium, and broken particles. The results indicated an overall particle breakage of 8.7% of the fissile particles and 1.5% of the fertile particles. The particle breakage is a combination of particles broken by crushing, transport, and the rotary valves below the UNIFRAME and crusher product bunker.

3.4.1.3. Primary Burner Product Removal System. The primary burner product removal system worked very well during the sequential operation. The system was operated from the control room by the burner operator.

The primary burner product was removed from the burner in three separate operations. The product from the first 10 fuel elements burned was removed; 103.1 kg were transported to the classifier feed bunker and the remaining 74.2 kg were transported back to the burner to form a bed for Part II of the sequential operation. A sample taken during this operation indicated a particle breakage of 10.6% of the fissile particles and 0.6% of the fertile particles. The product from Part II was removed in two batches of 179.7 and 98.4 kg due to a 200-kg limit on the bunker weigh cells.

The material was fed into the transport system at a feed rate of 0.5 kg/s using the variable position knifegate valve. The variable speed blower was adjusted to the minimum speed that would permit reliable transport. No samples were taken during the transport from the burner to the product bunker.

3.4.1.4. Classifier Feed System. The classifier feed system functioned normally during the sequential operation. The variable speed blower was adjusted to provide the 15 m/s transport velocity determined in the FY-77 sequential operation. The material was fed to the transport system at a rate of 0.25 kg/s.

The material from the primary burner product bunker was transported to the classifier feed bunker in three separate operations. The loss indicated by the primary burner product bunker was 382.8 kg, and the weight gain indicated by the classifier feed bunker was 373.0 kg.

Samples were taken during the last two transport operations. A sampler malfunction caused a loss of the sample during the first transport. The first sample indicated 47.1% fissile particle breakage and 1.75% fertile

particle breakage. The samples of the second transport indicated 15.1% fissile particle breakage and 4.13% fertile particle breakage. The average breakage was 18% of the fissile particles and 2% of the fertile particles. The large difference in breakage between the samples can be attributed to particle segregation in the primary burner. The primary burner tends to discharge the fertile particles, because of their higher density, before the fissile particles are discharged. The first transport operation contained mostly fertile particles and a few broken fissile particles. The second transport contained mostly fissile particles and a higher-than-average percentage of broken fertile particles. Another point of interest is to compare the fissile particle breakage in the last two batches transported (18.0%) with the fissile particle breakage in the batch recycled to the burner (10.6%). The particles recycled to the burner contained 8.3 kg of uranium, and the final two batches transported to the classifier contained 14.9 kg of uranium. This means that the majority of the fissile particles in the last two batches transported had recycled to the burner from Part I of the sequential operation, and a definite increase in particle breakage, 10.6% to 18.0%, occurred because of the longer residence time in the burner.

3.4.1.5. Particle Crusher Feed System. Classified fissile particles were conveyed from the fissile particle hopper to the particle crusher feed hopper. The weight loss in the fissile particle hopper was 45.6 kg. The weight gain in the particle crusher feed hopper was 45.2 kg. The weight loss in the particle crusher feed hopper when the particles were crushed was 46.4 kg. These readings were all within 3%. The blower had been sheared to provide a conveying velocity of 15 m/s, and the feed rate was 0.1 kg/s.

3.4.1.6. Secondary Burner Removal System. The improved secondary burner system installed after the FY-77 sequential operation was used to transport the secondary burner product to the secondary burner product bunker. The system operated satisfactorily with the total operation conducted in the automatic mode. The weight gain in the secondary burner product was

31.2 kg. When the product was removed, the weight loss was 31.0 kg and the material weight was 31.3 kg. These readings agreed within 1%.

3.4.1.7. BISO Product Removal System. The newly installed BISO product system was used to transfer particles from both the fissile and fertile particle hoppers to the BISO product hopper for removal from the system. Four separate transfers were made, two fissile and two fertile. The weights of fissile and fertile particles removed were 279.9 kg and 27.2 kg, respectively.

3.4.1.8. Conclusions and Recommendations. The solids handling systems performed satisfactorily during the sequential operation. The automatic control systems all functioned correctly and permitted all transport operations to be made from the control room.

A sampler should be installed on the crusher so that particle breakage in the crusher can be separated from that in the transport system. The weighing systems demonstrated repeatability within 1% to 3%; this must be improved if nuclear material accountability is to be made using these samples.

3.4.2. Pneumatic Transport Component Testing System

The pneumatic transport component testing system is a recirculating loop designed to test system components in long-term and/or cyclic tests. The system is computer-controlled, and operating modes can be selected by software. The system configuration can be changed to test a variety of pneumatic transport components.

3.4.2.1. System Description. The test system is shown in Fig. 3-3. Two 0.2 m³ hoppers are vertically stacked with a knifegate valve separating them. Gas enters the transport loop through the inlet filters. Material to be transported is metered from the lower hopper into the loop by the rotary valve. The material is transported through the 0.05-m piping to

Fig. 3-3. Pneumatic transport components test system

the upper hopper where the solid material is separated from the motive gas by sintered metal filters. The gas outlet line is connected to a variable speed blower, which provides the suction for the transport loop. The transported material can then be drained to the lower hopper through the knifegate valve and retransported around the loop ad infinitum.

The knifegate valve, filter blowback, rotary valve, and blower are actuated by the control program in the computer. Presently, the top level sensor is the only parameter monitored by the computer. A lower level sensor and some loop differential pressure monitors will be added to the control system. The computer records operating times and counts the cycles of operation for the equipment tested.

3.4.2.2. Present Operation. The present configuration of the test loop and software controls is aimed at establishing the lifetimes of the rotary valve, knifegate valve, and level sensor. Crushed graphite with a size distribution ranging from 9 mm to less than 10 μ m is being circulated through the loop at a rate of 2000 kg/h. Wear in the transport piping is also being monitored.

The system has presently completed 3200 cycles in 200 h of operation. The only problem encountered so far was graphite fines compacting in the vanes of the rotary valve and stopping flow. This was attributed to the roughness of the cast surfaces of the vanes. The rotor was removed and every other pocket was polished and the valve returned to service. Future inspection will determine if the problem has been solved.

It is intended to run the system until a major component failure occurs, or the expected service life of the components in an HRRF is exceeded.

4. OFF-GAS TREATMENT - TASK 340

4.1. SUMMARY

Installation of the engineering-scale off-gas treatment system is essentially completed, with only one heat exchanger yet to be delivered and installed. The heat exchanger is a late addition to the system for the purpose of adsorption bed regeneration. Piping walkdown and system cleaning have been completed. System leak check and correction are underway and almost completed.

Electrical work was resumed in FY-79. All electrical system installation and wiring, including the Diogenes digital process control system and Hewlett-Packard data acquisition system, have been completed. Electrical system functional checkout is continuing.

Most of the design activities were completed in FY-78. The CO/HT oxidizer design has been modified. The length of the catalyst bed has been increased to 178 mm and 356 mm (7 in. and 14 in.) from 76 mm and 152 mm (3 in. and 6 in.) for the first five sections and the last section, respectively. The change was made to compensate for the loss of residence time that resulted from the reduction of bed diameter from 229 mm (9 in.) to 152 mm (6 in.). Recent test results from the Allied Chemical Corp. (ACC-INEL) confirm the GA design at the same operating conditions. Plenums and diffusers are also added to the CO/HT oxidizer heat exchanger sections in order to reduce the excessive entrance and exit losses observed during a preliminary system functional check. Test operation after the modification indicates that the pressure losses are drastically reduced and that the original design flow through the heat exchanger is fully restored.

Recommendations from the system safety review meetings have been implemented in the system installation. Four CO monitors have been relocated, hazardous gas piping has been color coded, toxic gas bottles have been relocated outside the plant, and the control room has been slightly pressurized.

During the next reporting period, all system functional checkout and instrument calibration will be completed. After a system shakedown, the burner off-gas (BOG) system component testing will be initiated.

4.2. SYSTEM DESIGN

All scheduled design activities for FY-78 have been completed. A compressor, which is required for an integrated operation of the dissolver off-gas (DOG) system with the engineering-scale dissolver, could not be purchased due to high cost and a material problem. Most of the commercially available compressors are designed for a high-pressure delivery, are large in size, and are expensive. A low-pressure (205 kPa) and low-volume (20 Nm³/h) unit suitable to our application has been designed using a Teflon piston and Teflon bellows. Detailed design drawings for construction were originally scheduled for FY-79. Due to a lack of funds, however, this activity was postponed to FY-80.

The CO/HT oxidizer design has been revised. The bed diameter has been reduced from 229 mm (9 in.) to 152 mm (6 in.) in order to operate the system at a higher face velocity (0.7 m/s $\equiv$ 200 ft/min) and a higher Reynolds number (about 7000), which will enhance the mass transfer in the catalyst bed. In order to compensate for the loss in residence time, the bed lengths have been increased to 178 mm (7 in.) and 356 mm (14 in.) from 76 mm (3 in.) and 152 mm (6 in.) for the first five sections and the last section, respectively. ACC-INEL recently ran a test using their laboratory unit at 673 K and 0.7 m/s (Ref. 4-1). The test results

confirm the reaction kinetics model and the reaction rate used in our design calculations to determine the size of catalyst bed needed. Furthermore, our design is considered to be very conservative even in the case of minor poisoning of the catalyst since the system will actually operate at a much higher temperature, i.e., 973 K instead of 673 K, under the maximum load condition they simulated, and the reaction rate will be at least an order of magnitude higher. The CO/HT oxidizer consists of a series of catalyst beds with compact heat exchangers between them. Hot gases coming out of the catalyst bed are aircooled in the heat exchanger. Preliminary system checkout operation on the heat exchanger section showed a larger-than-expected pressure drop, resulting in a lower cooling-air flow than the original design calculation. Further tests revealed that the pressure loss was due to the entrance and exit losses. Entrance and exit plenums and plenum diffusers were subsequently added to heat exchanger sections. Test results after the modification showed a drastic reduction in pressure drop, and the design flow rate was fully restored. The design modification also corrected a flow maldistribution across the heat exchanger cross section which existed with the original design. The modified CO/HT oxidizer is shown in Fig. 4-1.

System descriptions for the burner off-gas (SD 565101) and for the dissolver off-gas (SD 565201) systems have been revised and issued. Changes in the process flow and the system design have been incorporated in order to reflect the as-built condition.

4.3. FABRICATION AND INSTALLATION

Fabrication of all system components and their installation have been completed except for one heat exchanger yet to be delivered and installed. The heat exchanger is a late addition to the system for the purpose of adsorption bed regeneration; however, regeneration is planned for FY-80. A material shortage caused a slippage of the production schedule of the heat exchanger supplier. Delivery and installation are expected in late February 1979.

System piping and tubing for gas supply, sampling, and monitoring have been completed. Utility lines are all connected to the off-gas system. Electrical work resumed in FY-79. All electrical system installation and wiring have been completed.

Figures 4-2 and 4-3 show the schematics of the process flow for the BOG and the DOG systems. Figures 4-4 through 4-14 show the system as installed.

Figure 4-4 shows the gas mixing unit. The vessel at the lower left corner is the DOG humidifier, and the one at the lower right corner is a constant temperature water bath that houses an iodine vapor generator. The system consists of rotameters, control valves, and digital mass-flow controllers. Eight different gases can be mixed in varying proportions, and moisture and iodine vapor can be added separately. The unit is designed to supply simulated burner and dissolver off-gases to the system until the system is integrated with the burner and the dissolver.

Figure 4-5 shows the general system installation. A typical adsorption vessel installation with associated auxiliaries; i.e., heaters, heat exchangers, and control valves; sampling and measurement devices and lines are shown in detail in Figs. 4-6 and 4-7.

Carbon monoxide for the CO/HT oxidizer is supplied from a tube pack mounted on a skid outside the pilot plant. The tube pack, which consists of 35 tubes with relief valves, has a total capacity of 991 m^3 [$35,000 \text{ ft}^3$ (STP)] as shown in Fig. 4-8. The tube pack has been leak checked with N_2 , evacuated, and charged with CO.

Figure 4-9 shows the general arrangement of the CO/HT oxidizer system, and Figs. 4-10 and 4-11 show the CO/HT oxidizer as installed. The first section of the oxidizer was taken down for modification at the time the pictures were taken and is missing in the figures. The pictures show the heat exchanger cooling air blower, inlet and exit headers, control valves,

Fig. 4-1. Modified CO/HT oxidizer

Fig. 4-2. Burner off-gas treatment system

Fig. 4-3. Dissolver off-gas treatment system

Fig. 4-4. Gas mixing unit. DOG humidifier and iodine vapor generator are at left and right lower corners

Fig. 4-5. General system installation. From left to right: CO_2/Kr , Rn, moisture, SO_2 , I_2 backup, and I_2 adsorbers

Fig. 4-6. Detailed installation of adsorption vessels with associated auxiliaries. Vessel at left is NO_x catalytic converter, vessel at right is tritium/moisture adsorber

Fig. 4-7. Typical installation of on-line process auxiliaries such as heater, flow meter, control valve, and temperature and pressure measurements

Fig. 4-8. Carbon monoxide tube pack

Fig. 4-10. CO/HT oxidizer system installed

Fig. 4-11. CO/HT oxidizer, cooling air blower, and exhaust heat exchanger

mass flow control valves for controlled oxygen injection to each stage, exhaust heat exchanger, sample lines, and a sample line cooler.

The reliability of the off-gas treatment system depends on the sensitivity and accuracy of the gas-analysis system. A total of 57 sample streams are analyzed by 17 gas analyzers mounted in a console and 3 field-mounted analyzers. Figure 4-12 shows the gas-analyzer console designed jointly by GA and Beckman. Due to a limited number of gas analyzers used in the system, sample-line switching is necessary to analyze all 57 sample points. The sample-line switch panel is seen at the upper left corner of the console. An additional switch panel is installed in the control room for a remote operation. The console includes a necessary sample conditioning system; namely, filters, pressure and flow regulators, and a phosphoric acid scrubber to remove NH_3 .

The gas analyzers cover a wide range of concentration of the gases used in the system. Approximately 85 cylinders of calibration gases at different concentration levels are needed to calibrate the gas analyzers. In order to alleviate this problem, a calibration-gas mixing device was designed jointly by GA and Beckman using digital mass flow controllers. With this device and a supply of 11 calibration gas cylinders, all of the 85 required calibration gas compositions can be mixed and fed to the gas analyzers. Figure 4-13 shows the calibration-gas mixing unit.

The entire process is monitored, controlled, and recorded in the control room using the Diogenes digital process controller. The data gathered can be directly fed to a Hewlett-Packard micro-computer system for data storage, reduction, and analysis. Figure 4-14 shows the completed control room.

Fig. 4-12. Gas-analyzer system

Fig. 4-13. Calibration gas mixing unit

Fig. 4-14. Off-gas system control room

4.4. PREOPERATIONAL SYSTEM CHECK

After completion of the vessel and piping installation, the entire system pipe walkdown was performed against the P&I diagrams in order to correct any possible errors. All detectable deficiencies were corrected.

Procedures have been developed and released for system cleaning (IP 565201) and leak checking (IP 565202). The system cleaning, excluding the vessels which were solvent cleaned, was done using a mild detergent to remove any organic contaminant that might poison the adsorbents, followed by a water rinsing and air blowing to dryness. No significant difficulties have been encountered during the cleaning process.

The system is now undergoing the system leak checking process. A significant amount of total leakage was found in threaded connections, valve stems, and faulty valves. Corrective measures are being employed. At present, the leakage has been reduced to a tolerable level, i.e., approximately 0.15% of the normal flow rate of 34 Nm³/h at 205 kPa. The actual leakage will be even smaller considering the fact that all the vessels are expected to operate between 101 kPa (0 psig) and 170 kPa (10 psig). Additional efforts are being made to further reduce the leaks.

A functional check of the CO/HT oxidizer heat exchanger was performed. A larger-than-expected pressure drop was observed. The problem has been solved, and the design flow has been fully restored as discussed in Section 4.2.

Electrical system functional checking is continuing on schedule.

4.5. SYSTEM SAFETY

Preliminary hazard analysis (PHA) and fault tree analysis (FTA) of the engineering-scale off-gas treatment system were done previously. Recommendations on physical installation of safety devices, such as alarms, interlocks, and safety valves, have been implemented.

Safety review meetings were held with the GA Safety Department, Industrial Hygiene Department, and the Traveler's Insurance Company. As a result, additional safety measures have been taken to ensure the safety of working personnel in the pilot plant. Four carbon monoxide monitors have been relocated, toxic gas cylinders have been moved outside, gas lines have been color coded, and the central control room has been pressurized slightly positive.

Further safety measures will be employed in the system operating procedures yet to be developed. The inherent system leakage under normal and abnormal operating conditions is judged not to pose any significant health hazard to plant personnel.

4.6. SCHEDULES AND MILESTONES

Installation of the entire system is essentially complete except for one heat exchanger not yet delivered. The system is undergoing functional checkout and calibration; this work is expected to be completed by June 30, 1979. After a total system shakedown, the BOG system component testing will be started during the next reporting period.

REFERENCE

- 4-1. "Monthly Technical Progress Report," Allied Chemical Corporation Report ACC-INEL-ROEH-58-78, November 7, 1978.

5. SOLVENT EXTRACTION - TASK 350

5.1. ANNULAR PULSE COLUMN EXTRACTION TESTS

The limited size of critically safe plutonium or U-233 bearing cylindrical solvent extraction columns limits plant capacity. There is a need to increase the capacity of these processes. This can be accomplished through use of a column having an annular cross section. The preliminary testing of the pilot-scale annular pulse column has been completed.

The column is fabricated from 15 cm (6 in.) i.d. glass pipe, and has an operating length of 2.7 m (9 ft). The center post is 8.9 cm (3.5 in.) o.d. stainless steel tubing, giving an annular width of 3.2 cm (1.25 in.). For column internals, stainless steel nozzle plate material was cut to a doughnut shape, and flat sheet stock in matching dimensions was fabricated with 10 equally spaced louvers bent up from the plane by 30° resulting in a free area of 23%. One louver plate is used for about every 0.46 m (1.5 ft) of column length.

The use of 23% free area louver plates instead of the 14% free area louver plates reported from earlier work in Ref. 5-1 raised the flooding frequency of the column using a 5.08 cm (2 in.) amplitude by 18% but did not significantly change the flooding frequency with a 2.54 cm (1 in.) amplitude. The average column height equivalent of a theoretical stage (HETS) for uranium was 0.55 m (1.8 ft) at 900 gallons/hr/ft² sum of flows and 0.61 m (2.0 ft) at 1250 gallons/hr/ft² at 80% of flooding.

The 23% free area louver plates did a more than adequate job of swirling the dispersed phase completely around the annulus from a single feed addition point.

No evidence has been seen of any nonuniform pulsing action from the one side of the column to the other.

The top three plates in the annular column scrub section should be nozzle plates rather than louver plates to minimize entrainment in the solvent overflow stream (1AP).

5.2. CYLINDRICAL PULSE COLUMN PLATE TESTS

5.2.1. Extraction Column Tests With 12% and 24% Free Area Straight Slotted Plates

Flat straight slotted plates were tested in a cylindrical pulse column in the organic continuous extraction mode in order to compare their capacity and efficiency with the flat rosette pattern plates tested earlier (Ref. 5-2).

The flat slotted plates are fabricated with 1.3 cm (1/2 in.) by 0.08 cm (1/32 in.) staggered straight parallel slots versus the crossed slots in the case of the rosette pattern plates tested earlier. The flooding frequencies when using the 24% free area straight slotted plates average about 10% higher than when using the 27% free area rosette plates. However, the efficiency of the straight slotted plates is lower and deteriorates with increasing amplitudes. Standard 0.48 cm (3/16 in.) hole 23% free area nozzle plates do not show changes in efficiency with changing amplitudes of between 2.54 and 7.62 cm (1 and 3 in.) (Ref. 5-1). The 27% free area rosette pattern plates show only marginal changes in efficiency with changing amplitude (Ref. 5-2). Additional studies will be needed to determine if amplitudes lower than 2.54 cm (1 in.) would be beneficial with the straight slotted plates.

In a 1.8 m (6 ft) extraction column, the 24% free area straight slotted plates flood at 47 cycles per min with a 7.6 cm (3 in.) amplitude, and 5.08 cm (2 in.) plate spacing, and at 80% of flooding, the average

uranium HETS was 0.76 m (2.5 ft). The straight slotted plates therefore may have application in high capacity pulse columns and some additional experimentation with them is warranted.

5.2.2. Stripping Column Tests

5.2.2.1. Stripping Column Capacity. The onset of cyclic flooding in the stripping column is a function of the throughput and pulse energy input (pulse amplitude x pulse frequency). Figure 5-1 describes this relationship, also known as the flooding curve, for three types of plates.

One hundred and thirty measurements were taken using flat shielded slot plates, angle shielded slot plates, or combinations of the two types. Eight different configurations were investigated at eight different plate spacings - 5.08 to 10.16 cm (2 to 4 in.) - and with both orientations (slots up or slots down).

Neither spacing nor orientation had significant effect on capacity.

Forty-five measurements were taken using standard nozzle plates, with two different spacings. Both spacings gave comparable results.

The zebra cartridge was adapted from the cartridge presently installed in the Hanford Purex Plant uranium stripping columns. By interleaving flourethene plates among the standard nozzle plates, coalescence of the dispersed phase is promoted. This enhances the migration of the organic phase up the column. This cartridge exhibits the highest capacity of any plate tested at a given pulse amplitude and pulse frequency.

5.2.2.2. Stripping Column Efficiency. The SEPHIS computer code (Ref. 5-3) was used to predict the theoretical steady-state uranium concentration profile within the stripping column at the recorded experimental conditions: flows, temperature, and concentrations. Analysis of aliquots

Fig. 5-1. Onset of cyclic flooding in the stripping column

from samplers at heights of 1.5 m (5 ft) and 3.05 m (10 ft) provided measured uranium concentrations. By comparison of the analyzed concentrations with the predicted concentrations, the number of theoretical stages at each sample point was determined. Fractions of a stage were calculated by interpolation between integer numbers of stages.

The average height equivalent to a theoretical stage (average HETS) is easily determined by dividing the distance from the bottom of the column to the sample point by the number of stages. The calculated HETS for two of the three types of plates tested is presented below. The differences are not significant, since there is considerable variability in the individual HETS calculations.

The average height equivalent to a theoretical stage was:

<u>Type of Plate</u>	To 3.05 m (10 ft) Sample Point <u>m (ft)</u>	To 1.5 m (5 ft) Sample Point <u>m (ft)</u>
Shielded slot	0.58 (1.9)	0.39 (1.3)
Standard nozzle	0.52 (1.7)	0.37 (1.2)
Zebra cartridge	Data reduction not complete	

5.2.2.3. Dispersed Phase Holdup Measurements. With LASL funding, some additional measurements and samples were taken to determine the vol % of dispersed phase within the column as a function of operating parameters (throughput, A/O ratio, solvent uranium loading, pulse amplitude, temperature).

Figures 5-2 and 5-3 present a summary of dispersed phase holdup measurement for standard nozzle plates and for zebra cartridge plates, respectively. The data are partially reduced with continued work suspended pending receipt of additional funding.

Fig. 5-2. Hold-up in the stripping column with standard nozzle plates for temperature of 54°C (130°F) and A/O ratio of 1.25

Fig. 5-3. Hold-up in the stripping column with zebra cartridge plates for temperature of 54°C (130°F) and A/O ratio of 1.25

5.3. ROBATEL CONTACTOR TESTS

The Robatel contactor is being reconnected into the solvent extraction pilot plant in preparation for testing in the extraction mode.

REFERENCES

- 5-1. Benedict, G. E., et al., "Alternate Fuel Cycle Technologies, Technical Progress Report for the Period July 1, 1978 - September 30, 1978," DOE Report GA-A15155, General Atomic Company, to be published.
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- 5-3. Watson, S. B., and G. H. Rainey, "Modifications of the SEPHIS Computer Code for Calculating the Purex Solvent Extraction System," ERDA Report ORNL/TM-5123, Oak Ridge National Laboratory, December 1975.

6. LABORATORY STUDIES - TASK 370

6.1. VERTICAL CENTRIFUGE TESTING

A Sharples Model P850 vertical continuous centrifuge has been installed in the General Atomic Company pilot plant. The vertical centrifuge will be tested for operability and solids-liquid separation efficiency under various operating conditions related to nuclear process applications. A vertical configuration was selected primarily because of the advantages involved with its adaptability to remote maintenance.

A series of initial calibration tests were performed for several submerged dissolver steam jets. These tests determined which jets were capable of transferring simulated dissolver slurries, in the range of 3.8 lpm (1 gpm) to 15.1 lpm (4 gpm), without causing excessive liquid carry-under with the solids during centrifugation. The steam jet test results determined that the 3.8 lpm (1 gpm) steam jet had excessive liquid carry-under with both 0.41 MPa (60 psig) and 0.55 MPa (80 psig) steam supply. The 2-gpm jet also had excessive liquid carry-under with 60 psig steam supply but with 80 psig steam supply the 2 gpm jet had only 1% carry-under of the total feed volume. The 3 gpm and 4 gpm steam jets provided satisfactory results (<1% carry-under of the total feed volume) with both 60 psig and 80 psig steam supply. The maximum liquid holdup in the centrifuge during these tests was measured as 1.75 liters.

The solids-liquid separation efficiency will be evaluated in a series of statistically designed tests. A 2^{6-3} saturated fractional factorial design, with resolution IV, was developed to minimize the number of vertical centrifuge tests to be performed. By incorporating the foldover principle into the statistical design, a total of only 16 screening tests

need to be performed to isolate the main effects of variables under investigation. The influence of the main effects on the separation efficiency of the centrifuge can then be evaluated independently of each other. There will be six variables under investigation: slurry feed rate, centrifuge g-force, solids concentration, particle density, liquid density, and particle size. A full factorial design would require 64 tests to provide a similar analysis to the statistical design described above.

6.2. DISSOLUTION TESTS

6.2.1. Introduction

A goal of the dissolution activities is to improve the ThO_2 dissolution rate by developing a suitable method to increase contact between the ThO_2 kernels and the thorex acid solution. Various devices for enhancing the solids-liquid contact will be examined. These methods will first be evaluated in a laboratory 105-mm glass dissolver. Attempts will be made to simulate the pilot-plant-scale dissolver operating conditions in the glass dissolver. These bench-scale tests also provide visual checks on the solid-liquid mixing pattern in the various dissolver configurations. Based on the results from the laboratory tests, the best contacting device will be further evaluated in the pilot-plant-scale stainless steel dissolver.

Scale-up effects and dissolution characteristics of various thorium fuel particles will also be investigated. One thoria kernel dissolution run will be made in the 0.5-m dissolver to evaluate scale-up effects. Experiments will be performed in the bench-scale and pilot-plant-scale dissolvers to determine the scale-up effects and dissolution characteristics of various fuel types, such as ThO_2/UO_2 fuel pellets with a weight ratio of approximately 3, as well as silicon-doped ThO_2 particles.

The dissolver operating cycle in the 0.5-m dissolver system for SiC-coated driver fuel particles will be demonstrated. Particular attention will be placed on reliable solids feeding and product removal.

Scale-up effects will also be determined. The separation of SiC hulls from the dissolver product will be examined with the Sharples vertical centrifuge.

6.2.2. Solid-Liquid Contact Improvement Study

A 105-mm glass dissolver was fabricated and installed in the laboratory for the solid-liquid contact study. The glass dissolver allows visual observation of the liquid and gas circulating pattern.

The first contacting device under study will be the pulse sparge design as shown in Fig. 6-1. A timer controls the frequency and duration of the pulse. By pulsing the sparge gas, increased liquid agitation at the solids-liquid boundary was observed. A reference 40% heel dissolution of ThO_2 kernels without pulse sparge will be made first to provide baseline data, followed by the improved dissolution method.

6.3. BENCH-SCALE LABORATORY STUDIES

6.3.1. Effect of Thorium on Hydrolysis of Tributyl Phosphate

Experiments are in progress to study the effect of thorium on the hydrolysis of tributyl phosphate (TBP) to dibutyl phosphate (DBP). The presence of dibutyl phosphate in solvent extraction operations results in a lowering of fission product-heavy metal decontamination factors.

Earlier work at ORNL (Ref. 6-1) has demonstrated the feasibility of measuring DBP in TBP/dodecane solvent. The method is based on gas chromatographic measurement of a DBP-methyl ester derivative. Experiments performed at GA indicate the gas chromatographic method is not applicable to the TBP/normal paraffin hydrocarbon (NPH) solvent system. NPH contains a significant $\text{C}_{14}\text{H}_{30}$ (tetradecane) component (about 28 vol %) which was found to have a retention time similar to the DBP derivative on the recommended

Fig. 6-1. Pulse sparge design

Dexsil 300 column. In addition, the presence of nitric acid in the solvent was found to retard the formation of the required DBP-methyl ester.

An alternative procedure was developed to allow study of the hydrolysis rate of TBP in thorium-nitric acid - TBP/NPH systems. The method is based on: (1) selective extraction of DBP into sodium carbonate solution, (2) oxidation of organo-phosphorus to phosphate ion (PO_4^{-3}) with nitric and perchloric acids, and (3) colorimetric determination of PO_4^{-3} following reduction of a phospho-molybdate complex.

Samples were prepared for study by adding thorium nitrate tetrahydrate and concentrated nitric acid directly to 30% TBP/NPH solvent. These components were added to yield final thorium and nitric acid concentrations of 0 to 35 g/l and 0 to 0.63 M, respectively. The samples were thoroughly mixed and stored at ambient temperature in the dark to eliminate solvent degradation due to exposure to ultraviolet light. Following elapse of the aging period, samples were again thoroughly mixed and representative aliquots were removed for DBP analysis.

Results of experiments performed to date show a marked increase in DBP formation as a function of time. Typical data for 2-day, 6-day and 13-day aging periods are presented in Fig. 6-2.

Additional experimentation is in process to establish a DBP formation rate for longer time intervals and at elevated temperatures. It is possible that the apparent reduction in DBP in samples containing thorium and nitric acid is related to the formation of a second organic layer (third phase). Tests are being conducted to determine whether third phase formation is a factor in TBP hydrolysis.

6.3.2. Iodine and Ruthenium Volatility in Thorium Fuel Reprocessing

Reference 6-1 contains the experimental results from pilot plant and bench-scale studies on the behavior of iodine and ruthenium during dissolution and feed adjustment operations. Solutions used in the work were

Fig. 6-2. Effect of aging on TBP hydrolysis

prepared from simulated HTGR and LWR-Th fertile fuels. The major findings of the study are summarized below:

1. Iodine volatility is $\leq 10\%$ during simulated batch dissolution of full burnup, 1 year cooled, four-segment HTGR fertile fuel.
2. Iodine volatility is $\sim 70\%$ during simulated batch dissolution of $(5\% \text{ U-Th})\text{O}_2$ and $(50\% \text{ U-Th})\text{O}_2$ light water reactor-thorium (LWR-Th) fuel pellets.
3. Greater than 96% iodine volatility may be achieved through the introduction of a reducing agent, e.g., nitric oxide (NO), during dissolution of HTGR fertile and LWR-Th fuels.
4. Low ruthenium volatility is encountered in the preparation of 1M thorium-Thorex solution from HTGR fertile kernels through dissolution and feed adjustment.

6.3.3. Three Phase Compositions in Thorium, Uranium, Nitric Acid, TBP, Diluent System

Solutions with known nitric acid, uranium, and thorium concentrations were contacted with 30% TBP/NPH forming three phases: one aqueous and two organic layers. The phases are separated and the volumes recorded. The organic phases are analyzed for uranium and thorium concentration, TBP concentration, and acidity. The aqueous phases are analyzed for uranium and thorium concentration and acidity.

Data are being obtained and analyzed to characterize the system and study the effect of uranium on third-phase thorium-TBP formation.

REFERENCES

- 6-1. Wilbourne, R. G., "Iodine and Ruthenium Volatility in Thorium Fuel Reprocessing, Interim Development Report," General Atomic Company, to be published.

- 6-2. Manning, D. L., "Gas Chromatographic Study of Organophosphorous Compounds Using Dexil-300 as a Stationary Phase and a Hydrogen Flame Ionization Detector," Oak Ridge National Laboratory Report ORNL/TM-6423, September 1978.