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QUARTERLY REPORT SERIES*

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GA-A13255 - September 1974 through November 1974
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ABSTRACT

This publication continues the quarterly series presenting results of work performed under the National HTGR Fuel Recycle Program (also known as the Thorium Utilization 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 and the design and development of an integrated line to demonstrate the head end of HTGR reprocessing using unirradiated fuel materials. Work is also described on trade-off studies concerning the required design of recycle facilities for the large-scale recycle of HTGR fuels in order to guide the development activities for HTGR fuel recycle.



INTRODUCTION

This report covers the work performed by General Atomic Company under U.S. Energy Research and Development Administration Contract E(04-3)-167, Project Agreement No. 53. The work done under this project agreement is part of the program for development of recycle technology for High-Temperature Gas-Cooled Reactor (HTGR) fuels described in the "National Program Plan for HTGR Fuel Recycle Development" (GCR-76/19).

The objective of the program is to provide a demonstration plant for the recycle of HTGR fuels. This plant will demonstrate facility and equipment design and operating procedures which are licensable and commercially feasible for the reprocessing and refabrication of spent fuel from HTGRs. Work at General Atomic Company is concentrating on the following National Program tasks: Program Management and Analysis (Task 100); Reprocessing Technology Development (Task 200); Refabrication Technology Development (Task 300); HTGR Recycle Demonstration Facility (HRDF) design support (Task 600).

Task 100, Program Management and Analysis, includes the functions of overall planning, scheduling, budgeting, reporting, management control of the program, and coordination of activities.

Task 200, Reprocessing Technology Development, includes the definition of flowsheets, the development of components, and the definition of operating techniques, remote maintenance and or disassembly techniques, and coordination of fuel shipping and storage activities. Operations which must be developed include crushing of the fuel elements; burning the graphite in a fluidized bed-burner; separation of the fertile and fissile

particles; crushing the SiC coating on fissile particles; burning the crushed particles; dissolution of thorium and uranium in the burned, crushed particles; separation of the undissolved solids (SiC hulls, etc.) from the leachate; separation of the thorium and uranium from the fission products by solvent extraction; separation and purification of the thorium and uranium by solvent extraction; process and facility off-gas treatments to ensure releases are environmentally acceptable and in compliance with regulations; and the primary treatment of solid, liquid, and gaseous wastes from the process.

Task 300, Refabrication Technology Development, includes the definition of flowsheets, the development of components, and the definition of operating techniques, remote maintenance and/or disassembly techniques, and coordination of fuel shipping and storage activities. The refabrication begins with aqueous uranyl nitrate solution from the reprocessing facility and ends with fuel elements prepared for shipment to the reactor. The principal operations to be developed are loading the ion-exchange resin with uranium, resin carbonization, resin conversion, coating the converted resin with pyrolytic carbon and SiC, fuel rod fabrication, fuel element assembly, fuel and fuel element inspection, scrap recovery, and waste handling.

Task 600, HTGR Recycle Demonstration Facility, includes the design, construction, proof-testing, and operation of a demonstration facility for the recycle of HTGR fuel. The plant is to include all fuel cycle operations from the receiving of spent fuel elements from the reactors to shipping the refabricated fuel elements back to the reactors. The preconceptual design studies and the early conceptual design are to be used to guide the development work for reprocessing and refabrication processes and equipment. The results of the research and development tasks will in turn be used to guide the detailed design of HRDF.

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1. SUMMARY

Program activities remained on schedule during the quarter, with the head-end reprocessing line installation and startup completed and equipment shakedown and modifications initiated. UNIFRAME design, procurement, installation, and check-out of all major equipment items and subsystems are complete.

The fuel particle crusher was successfully demonstrated by processing Fort St. Vrain (FSV) fertile fuel particles. Procurement and installation of the prototype solids handling system are complete. Check-out and shakedown are also complete except for some weigh cell electronics.

Installation and preoperational check-out of the prototype burners were completed. Experimental work has continued to concentrate on the 20-cm primary burner. Analysis of operations without fines recycle and the startup and tail-burning phases were completed. Tests on the reference fines systems have also been completed.

Bench-scale experiments were used to study the effects of graphite processed through the dissolution and feed adjustment steps on solvent extraction scrub section zirconium distribution coefficients. Only minor effects were observed when comparisons were made with control samples. Additional tests are planned with a slightly modified procedure to investigate silicon carbide and perform further studies of heavy metal carbide effects.

A preliminary study was made on formic acid denitration of uranyl nitrate solutions as an alternative to amine extraction. The results of this study show promise and additional work is recommended. Studies of solvent nitration continued during the quarter with efforts being directed toward the safety of the thorium intercycle concentrator.

Five solvent extraction runs were completed during the quarter. These runs represented the first cycle of the Acid-Thorex flowsheet.

An off-gas study was completed with the objectives of reviewing the development status of the off-gas treatment technology with regard to the needs for the HTGR Recycle Demonstration Facility (HRDF) design, reviewing the off-gas progress to date, and identifying components requiring engineering-scale demonstration. After a detailed review of the proposed process flowsheets and material flow schedules, several necessary changes in the head-end off-gas treatment scheme became apparent. Some additional laboratory and hot cell development needs were also identified.

Operational readiness efforts continued on semiremote fixtures, including testing, interface verification, and operational check-outs.

The head-end systems analysis study was completed during the quarter. The study assessed thermal limitations imposed on spent fuel particle hopper designs and operating modes by radioactive decay heat. Constraints on hopper sizing were established with respect to different modes of decay heat removal.

Work has been initiated on a Spent Fuel Element Decay Heat and Source Term analysis, which is a follow-on to the HTGR Spent Fuel Composition and Fuel Element Block Flow study completed in FY-1976. A Process Yields and Material Throughput study was also initiated wherein detailed material balances are being prepared based on recently determined spent fuel element compositions and representative production operating modes. The material balance will account for process yields, decontamination factors, process impurities, and system efficiencies which reflect the current status of the development program. Core segment history will also be considered in determining maximum heavy metal throughputs.

Support activities for HET-Reprocessing preliminary design were directed toward completion of the design criteria phase. A draft of the

HET-Reprocessing Facility Conceptual Design Criteria document was completed on July 10, 1976 and subsequently approved by ORNL for use by Ralph M. Parsons Company in preparing the final design criteria documents.

The HRDF-Reprocessing Flowsheet Review and Updating study continues as part of technology assessment to ensure that the proposed HRDF flow-sheet incorporates recent technology development improvements and new design data, and that supporting technical programs are apprised of flow-sheet design issues requiring resolution.

2. FUEL ELEMENT AND FUEL PARTICLE CRUSHING

2.1. FUEL ELEMENT CRUSHING

The major design, procurement, installation, and checkout activities are complete and minor modifications are in progress on the fuel element size reduction system (UNIFRAME) to be installed as a part of the engineering-scale reprocessing (cold) pilot plant at General Atomic.

2.1.1. UNIFRAME Prototype Fuel Element Size Reduction System

2.1.1.1. Introduction

A fuel element size reduction system is being developed to permit HTGR fuel elements to be reduced to a size suitable for removal of element and matrix graphite in a fluid-bed burner. Detailed equipment descriptions and photographs of the construction phase were presented in the previous quarterly report (Ref. 2-1). The fuel element size reduction system, designated UNIFRAME because of its single machine frame construction, consists of five major equipment items and five subsystems as follows:

Major Equipment Items

1. Primary crusher: an overhead eccentric jaw crusher for reducing the elements to <6-in. ring-sized fragments.
2. Secondary crusher: an overhead eccentric jaw crusher for further reduction of the fragments to <2-in. ring-sized fragments.
3. Tertiary crusher: a double-roll crusher for final reduction of the fragments to $\leq 3/16$ -in. ring-sized product.

4. Screener: a vibratory screener-separator for separating the acceptable product from any oversized fragments.
5. Oversize crusher: an eccentrically mounted single-roll crusher for reduction of oversize fragments to acceptable product size.

Subsystems

1. Ventilation: the enclosure that provides containment and collection of radioactive materials and dusts while minimizing the surfaces exposed.
2. Structural: the special framework replacing standard machine frames to enable an efficient array of the equipment.
3. Drive: the standard and special drive components required to make the UNIFRAME system compatible with remote operation requirements and the radioactive environment.
4. Lubrication: the standard and special lubrication and bearings for equipment that make it more reliable and compatible with the radioactive environment and the remote operation requirements.
5. Mechanical: the standard and special components required to make the major equipment items compatible with the structural, ventilation, and remote operation requirements.

A sketch of the basic system is shown in Fig. 2-1.

2.1.2. Documentation

In order to provide a record of the various aspects of the development of the fuel element size reduction system, a series of control documents

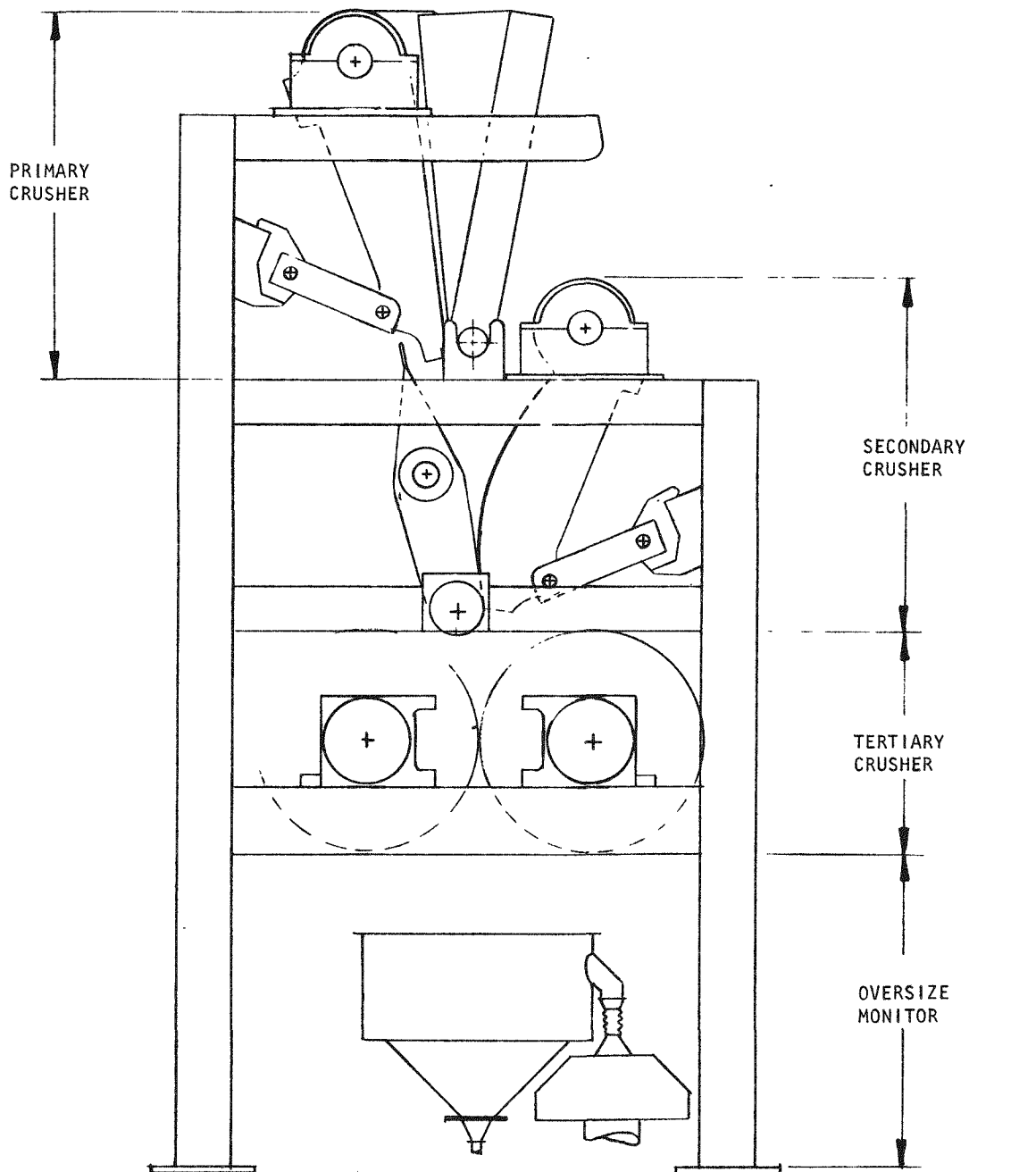


Fig. 2-1. UNIFRAME size reduction system (less drives and enclosures)

was planned. A complete list of these documents and their current status is given below:

<u>Doc. No.</u>	<u>Description</u>	<u>Status</u>
DDI-521001	Design Document Index	Complete
DC-521001	Design Criteria	Complete
SD-521001	System Description	Complete
PF-521001	Process Flow Diagram	Complete
PI-521001	Piping and Instrumentation Diagram	Complete
OP-521001	Operating Procedures	Complete
MM-521001	Maintenance Manual	Complete
Dwg No. 5210002	Equipment Assembly	Complete
Dwgs. beginning with No. 5210003	Equipment Detail Drawings	Complete
IS-521001	Implementation Specification: Primary Crusher	Complete
IS-521002	Implementation Specification: Secondary Crusher	Complete
IS-521003	Implementation Specification: Tertiary Crusher	Complete
IS-521004	Implementation Specification: Vibrating Screen	Complete
IS-521005	Implementation Specification: Oversize Crusher	Complete
AP-521001	Activity Plan	Complete - To be released
DR-521001	Design Report	Due 1/79

All of the control documents listed will be continuously upgraded to reflect changes brought about by installation and operating experiences, test results, and other programmatic results. In addition to the control documents, several supporting documents such as installation and checkout procedures, design calculation reports, quality assurance documents, and interim reports have been prepared or are in preparation.

2.1.3. Current Status

The design, procurement, installation, and checkout of all major equipment items and subsystems are complete. Upgrading to a final design which is prototypical to equipment and process requirements for a Hot Reprocessing Demonstration Facility (HRDF) is an integral part of the development program and will be carried on in parallel with the current effort.

Each of the major equipment items has been operated singly at design speeds without fuel charging. The entire system has been successfully operated by processing partial fuel elements (without fuel particles) through the entire UNIFRAME size reduction system.

Design factors and manufacturing tolerance buildups resulted in an out-of-specification close side setting (discharge gap) on the primary crusher. Newly designed toggles, which will allow the correct side setting, have been ordered, fabricated, and received. Pending quality assurance acceptance, they will be installed for continuing tests as outlined in the Activity Plan (AP-521001).

During the dismantling of the toggle assembly to ready the crusher for the new toggles, a thorough examination of the bushings (on the toggles, pitman casting, and pushing beam) and the pin joint shafts was undertaken to obtain all data available on the wear characteristics and dry film lubrication at this stage of operation. The shafts were examined both before and after the dry film lubricant had been removed. Preliminary observations indicated satisfactory lubrication and normal wear on these parts. A complete description of the condition of these components will be included in the next quarterly report.

This downtime was also utilized to replace leaking oil seals on all the outboard bearings of the primary and secondary crusher pitman assemblies. This type of seal is expected to experience some leakage even under

ideal conditions, and interim measures to avoid contamination of the product are in work. A complete redesign is planned as a part of the upgrading of the lubrication subsystem for prototypicality to the HRDF.

2.2. FUEL PARTICLE CRUSHING

2.2.1. Introduction

A crusher has been built at General Atomic for removing the SiC coatings from the Fort St. Vrain TRISO-coated fertile fuel particles to expose the inner carbon coating for fluidized bed combustion. This crusher, which is a double roll type, has consistently produced crushed product (six controlled runs, 3 kg) compatible with the fluidization requirements of the prototype secondary burner. The following discussion summarizes the crusher development activities to date.

2.2.2. Fuel Particle Double Roll Crusher

Phase I of the Activity Plan for the fuel particle crusher has been completed, successfully demonstrating the ability of the double roll crusher to process Fort St. Vrain fertile fuel particles. A series of tests were performed utilizing representative feed lots split from a single larger lot. The tests were arranged so that actual crusher performance could be compared with the performance requirements of the particle crusher design criteria (DC-520001). A comparison of the individual performance requirements of the design criteria with the actual test results is given below.

The double roll crusher is required to process fuel particles at a minimum rate of 30 kg/hr. Tests proved the throughput rate to have a linear relationship with roll speed. Throughput rates of 50, 101, and 148 kg/hr resulted from roll speeds of 25, 50, and 72 rpm. In each case, the product size distributions were virtually identical (see Fig. 2-2) even though the throughput rates varied by factors of two and three.

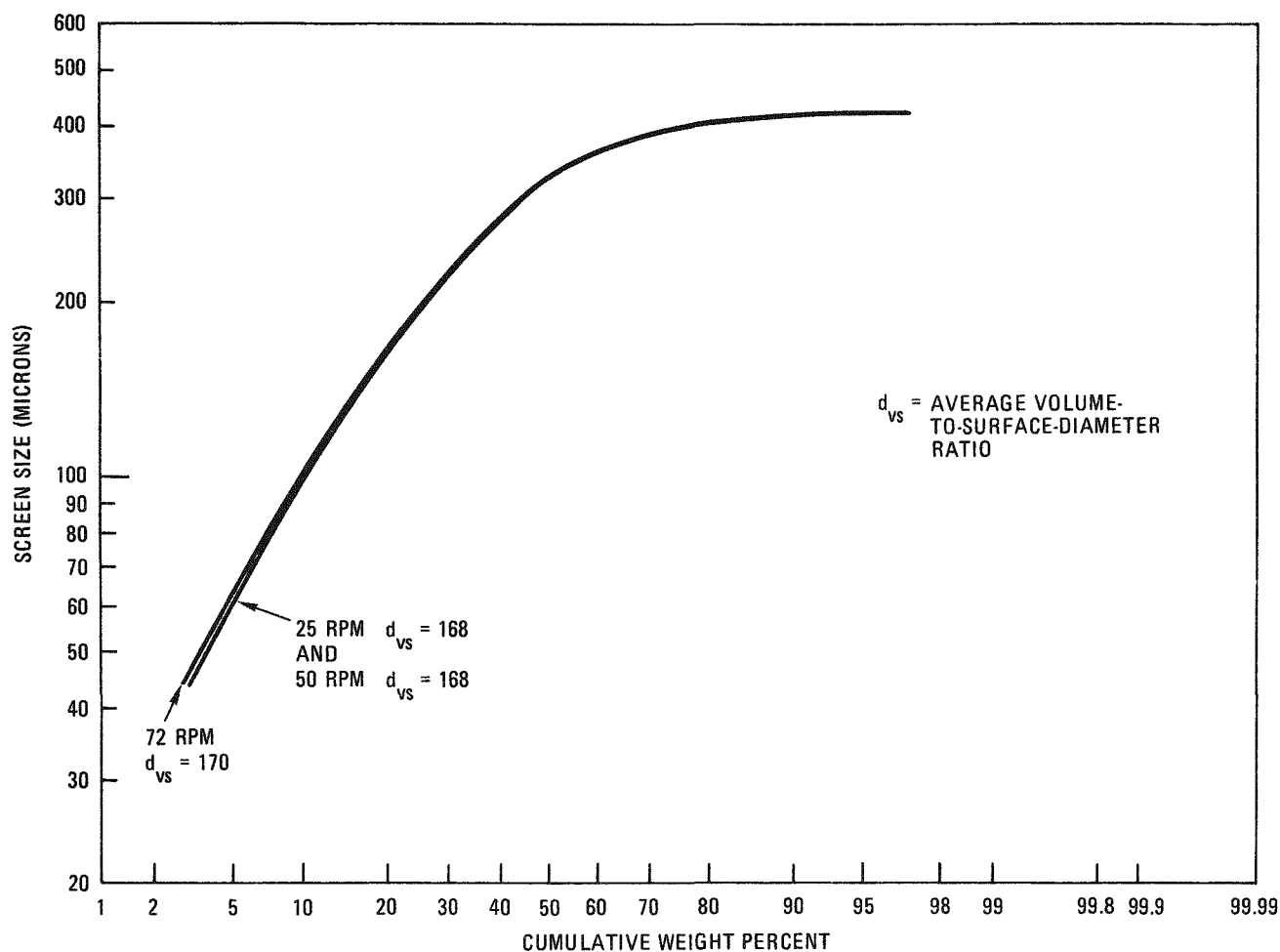


Fig. 2-2. Fuel particle crusher product size distributions for various throughput rates

In the event that the double roll crusher is shut down during operation, it must have sufficient power to be restarted without removing the feed material remaining in the crusher. Several tests were performed with feed material in the crusher prior to startup. No significant differences were observed in crusher starting characteristics or in product size distribution for the two load cases.

The crusher product must contain less than 1% unbroken particles. Throughout the test program, unbroken particles were found in small quantities in the upper two size categories ($>500 \mu\text{m}$ and $>425 \mu\text{m}$). The maximum amount of unbroken particles found in all the tests was estimated at less than 0.1%.

The size distribution requirement for the crusher product is $160 \mu\text{m}$ minimum volume-to-surface diameter. To describe usefully the size distribution of a batch of solid particles, the size distribution functions P and p were defined as follows (Ref. 2-2). Let P be the volume fraction (or weight fraction or numbers fraction) of particles smaller than size d_p and let $pd(d_p)$ be the volume fraction (or weight fraction or numbers fraction) of particles of size between d_p and $d_p + d(d_p)$. P gives the cumulative distribution of sizes and is dimensionless, whereas p gives the distribution directly and has units of reciprocal length.

The relationship between p and P is found by considering particles of any particular size d_{p_1} , for which

$$p_1 = \left(\frac{dP}{d(d_p)} \right)_1 \quad \text{or} \quad P_1 = \int_0^{d_{p_1}} pd(d_p) \quad .$$

For discrete distribution of particles with equal or unequal intervals of size (GA's sieve analysis produces a discrete distribution of unequal size intervals), the relation between p and P at any d_{p_i} is

$$P_i = \left(\frac{\Delta P}{\Delta d_p} \right)_i \quad \text{or} \quad P_i = \sum_i (p \Delta d_p)_i = \sum_i x_i ,$$

where x_i is the fraction of material in size interval i .

With the size distribution of particles defined, the specific surface and mean diameter of a mixture of different-sized particles of a given shape may be found. For pressure drop in flow-through beds (a variable in secondary burner fluidization), the surface area of the particles is the prime consideration. Thus, a mean size and shape are defined such that for a given mass of solid, the surface area remains constant. Using the size distribution, the mean specific surface is defined as

$$\bar{a}' = \sum_{\text{all } i} a'_i (p \Delta d_p)_i = \frac{6}{\phi_s} \sum_{\text{all } i} \frac{(p \Delta d_p)_i}{d_{p_i}} = \frac{6}{\phi_s} \sum_{\text{all } i} \left(\frac{x}{d_p} \right)_i$$

for a discrete distribution. Since the mean specific surface defined in terms of mean diameter is

$$\bar{a}' = \frac{6}{\phi_s \bar{d}_p} ,$$

the mean diameter becomes

$$\bar{d}_p = \frac{6}{\phi_s \bar{a}'} = \frac{1}{\sum_{\text{all } i} [(p \Delta d_p)_i / d_{p_i}]} = \frac{1}{\sum_{\text{all } i} (x/d_p)_i} .$$

For mixtures of particles of different size and shape, \bar{d}_p and d_p are replaced by $\bar{\phi_s d_p}$ and $\phi_s d_p$, respectively. Since the shape factors (ϕ_s) are unknown in our size distribution, the shape factors are assigned the value of unity for ease of calculation. The mean diameter (\bar{d}_p) is termed the volume-to-surface diameter (d_{vs}) since the surface area is the prime consideration. A description of how the equation for the volume-to-surface diameter

$$d_{vs} = \frac{1}{\sum \left(\frac{x}{d} \right)}$$

is used for a given particle batch is given in Ref. 2-3.

The main objective of the Phase I tests was to determine the roll gap at which the product size distribution meets the 160- μ m requirement. The proper roll gap was approached from the low side at 0.001-in. (25.4 μ m) intervals. The final three tests at a roll gap of 0.019 in. produced volume-surface diameters greater than 160 μ m. At successively smaller roll gaps, smaller volume-surface diameters were obtained. Figure 2-3 shows the product size distributions for various roll gaps.

The Fort St. Vrain fertile particle double roll crusher is installed and fully operational at the prototype secondary burner.

The prototype pilot plant Experimental Plan also calls for the development of a crusher to remove the SiC coatings from the Fort St. Vrain fissile fuel kernels. The crusher being developed for this purpose is the same double roll type as the fertile particle crusher. Design is complete, and bids are currently being received for the fabrication and assembly of the Fort St. Vrain fissile fuel particle double roll crusher.

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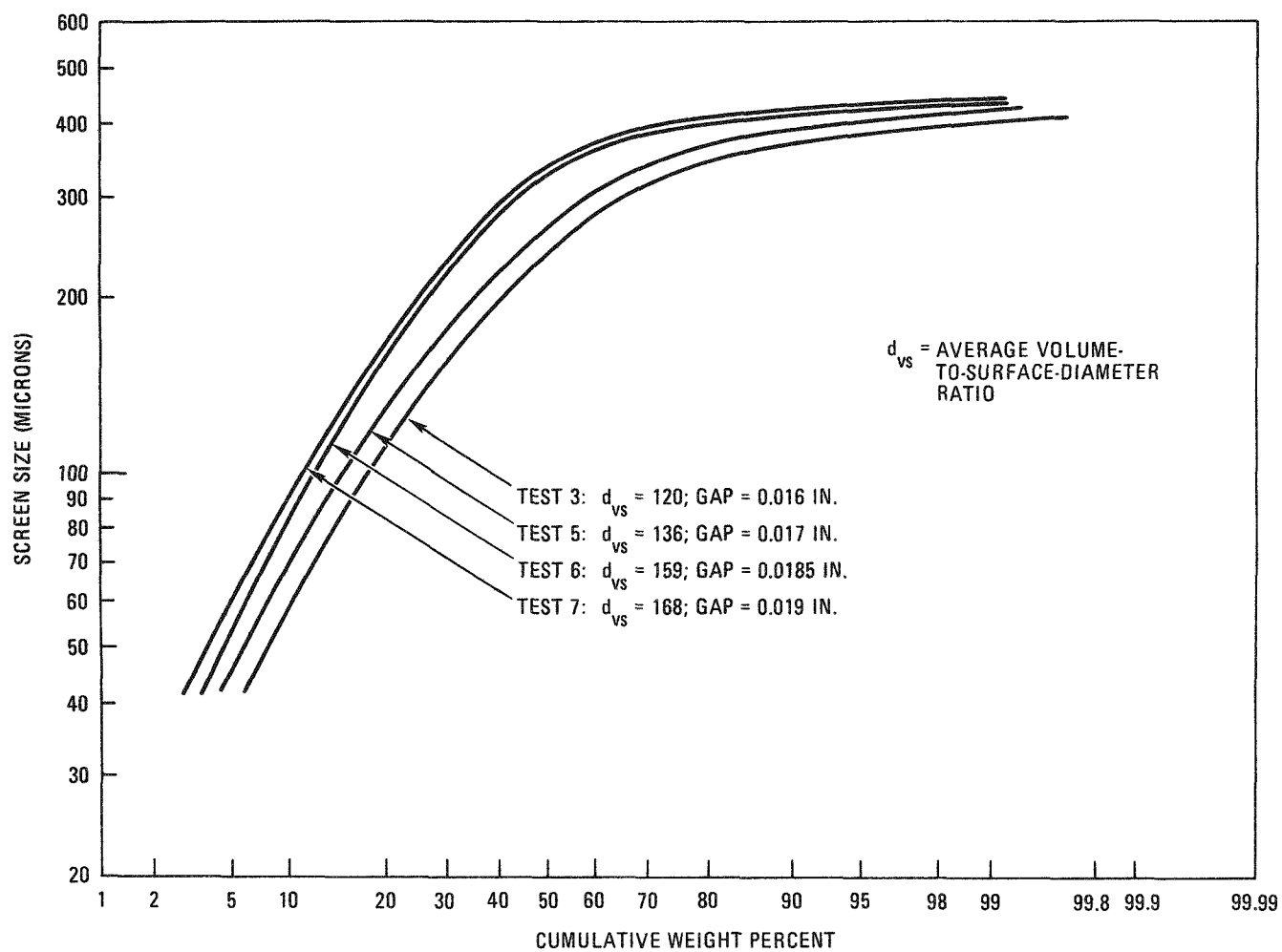


Fig. 2-3. Fuel particle crusher product size distributions for various roll gaps

3. SOLIDS HANDLING

3.1. SUMMARY

Procurement and installation of the prototype solids handling system are complete. Check-out and shakedown are also complete except for some weigh cell electronics.

In order to accommodate the non-sequential operation of the pilot plant, modifications need to be made to the solids handling system. These have been completed for both the burners.

In situ samplers will be required during future sequential operation, and a selection of different types have been ordered.

In order to prevent hazards arising from the pyrophoric nature of WAR fissile kernels, steps will be taken to install an argon dumping system connected to the bunkers.

3.2. PNEUMATIC TRANSPORT

Transfer of the particulate solids between and around the unit operations in the head-end prototype reprocessing system will be by pneumatic transport. With respect to this mode of material transfer, there is an ongoing evaluation of the criteria, equipment, and operation.

3.2.1. Pneumatic Transport Test System

Several components of the test system have been involved in the modifications to the pilot plant required for non-sequential operation (see Section 3.2.2.5).

3.2.2. Prototype Pneumatic Transport System

3.2.2.1. Description

The prototype pneumatic transport system has been designed to link the unit operations in the head-end to produce an integrated process. As a result, the transport system encompasses both the feeding and product withdrawal interfaces for each unit operation. To optimize each operation, hence the process as a whole, surge bunkers were specified where needed.

The system as designed is depicted in Fig. 3-1. However, the activities planned for the various unit operations in the coming months require non-sequential operation. This is discussed in Section 3.2.2.5.

3.2.2.2. Procurement

Procurement is complete.

3.2.2.3. Installation

Installation has been completed. With the exception of data handling electronics for the weigh cell system, everything has been checked out and shaken down. Problems with the weigh cell electronics are currently being solved.

3.2.2.4. Documentation

The status of the design documents for solids handling is given in Table 3-1.

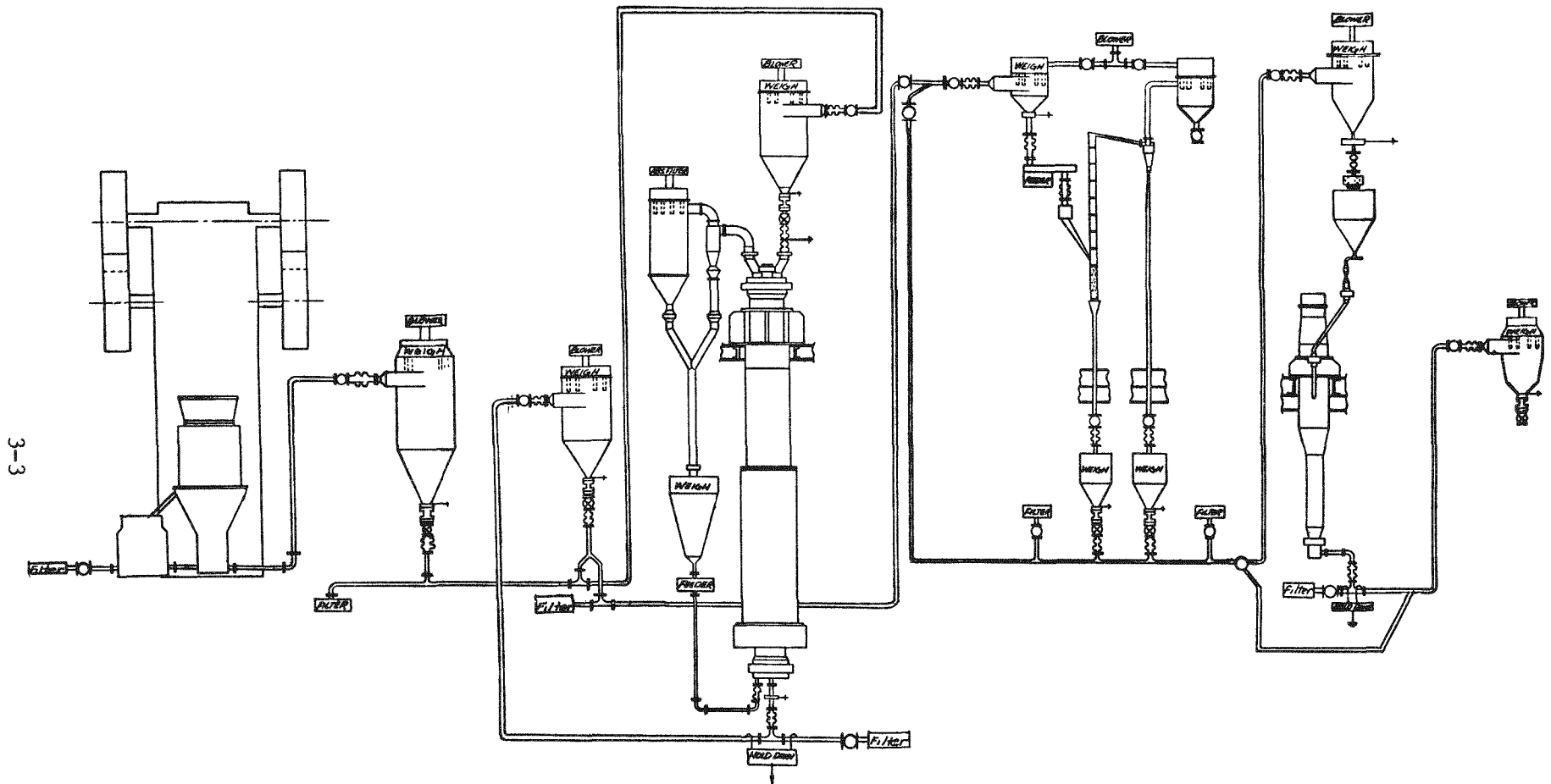


Fig. 3-1. Pneumatic transport system

TABLE 3-1
SOLIDS HANDLING SYSTEM DOCUMENT STATUS

Number	Type	Title of Document	Status
DC-520101	Doc	Design Criteria: Solids Handling	Issue A
PF-520101	Dwg	Process Flow Diagram	Issue A
PI-520101	Dwg	Piping and Instrumentation Diagram	Issue A
OP-520101	Doc	Operating Procedures: Solids Handling	Issue A
MM-520101	Doc	Maintenance Manual: Solids Handling	--
DR-520101	Doc	Design Report: Solids Handling	--
PA-500001	Dwg	Plant Arrangement	Issue B
5201002	Dwg	Equipment Assembly	Issue A
5201003	Dwg	Equipment Detail Drawings in Chronological Order	Issue A
SD-520101	Doc	System Description: Solids Handling	Issue A
IS-520101	Doc	Implementation Specification: Bunkers	Issue A
IS-520102	Doc	Implementation Specification: Blowers	Issue A
IS-520103	Doc	Implementation Specification: Filters	Issue A

Revision of the engineering detail drawings is progressing. The changes include:

1. Renumbering to bring numbers of drawings and parts into line with other pilot plant systems and to reflect the six subsystems.
2. Incorporating a relaxation in tolerancing and weld specifications in bunker and filter assemblies. This relaxation was a major cost reduction item.
3. Incorporating changes to the dimensions of bellows such that assemblies fitted together.

3.2.2.5. Non-Sequential Operation

The solids handling system was designed according to the principle that the product of one operation would be used directly as the feed for the next. In the initial phases of the operation of the pilot plant, however, this is not necessarily the case. The additional material transfer requirements are given in Fig. 3-2.

Since pneumatic product removal from the fuel element size reduction system, crushed fuel element burner, and crushed particle burner forms an integral part of the systems, it was decided that product should be removed by pneumatic transport. Each point of material addition or removal was examined in order to ascertain the most suitable technique. Another aspect of the desired operations was the need to take samples and to blend fuel particles and crushed graphite to make simulated primary burner feed.

The planned modifications to the solids handling system are shown in Fig. 3-3. Crushed fuel elements are removed pneumatically, then are separated from the conveying gas in the test rig and fall into a 55-gal drum. Fuel particles are added simultaneously to graphite to simulate

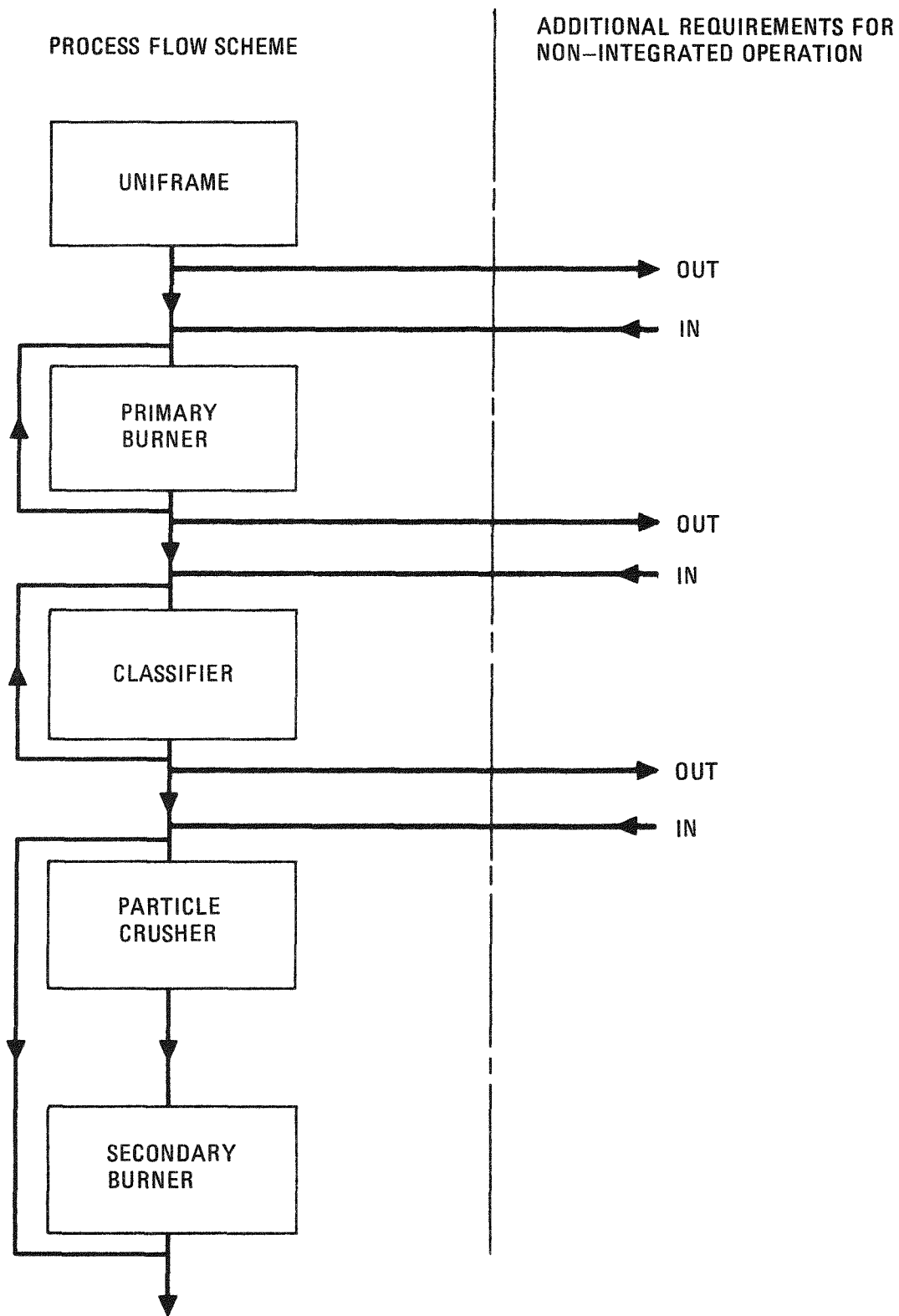


Fig. 3-2. Material transfer requirements

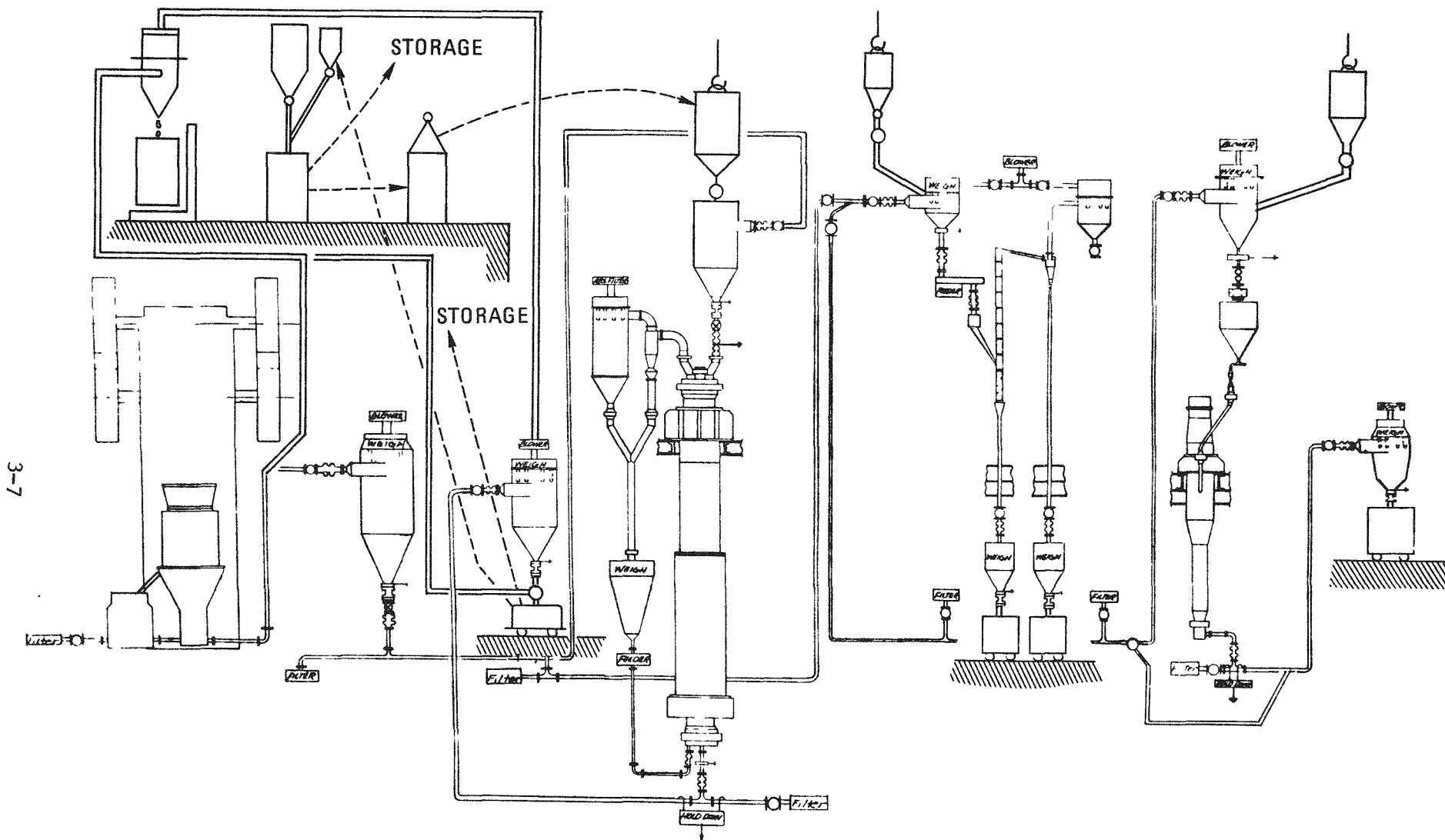


Fig. 3-3. Modified solids handling system

primary burner feed, which is fed to the primary burner feed bunker by gravity. This is made possible by removing the filters in the bunker and substituting a top with a central inlet. Primary burner product is conveyed to the product bunker, in accordance with the original design. It can be discharged by gravity into a container or conveyed pneumatically to the test rig and a 55-gal drum. The latter course is probably the most suitable for burner heating tests with graphite. Since fuel particles themselves are very free flowing, they can probably be fed into the classifier and particle crusher feed bunkers through the fittings provided for high-level detectors. Inclined feed pipes guide material to the bunkers from a fuel particle canister. Classified fuel particles can be fed by gravity into small containers. Secondary burner product is discharged by gravity from the product bunker.

The test rig variable-speed blower serves to ventilate the UNIFRAME and discharge both the primary burner and the primary burner product bunker. The variable speed is required when discharging the primary burner in order to cope with the thermal effects described in the last quarterly report (Ref. 3-1).

In order to take samples, a 16 to 1 splitter has been installed in a ventilated space such that the bulk of the sampled material falls into a drum. A 2 to 1 splitter is available for reducing the size of the sample.

To date, the modifications for feeding and removing product from the primary and secondary burners are complete. The particle addition device for making simulated primary burner feed is under construction.

3.2.2.6. Sampling

The facilities for taking samples in the pilot plant are not yet optimal. During the coming few months, when operation will be non-sequential, samples can be taken outside the line (see Section 3.2.2.5,

Non-Sequential Operation). However, studies of particle breakage and the variation in stream composition and particle size during extended sequential operation demand accurate in situ sampling. A study by the Ralph M. Parsons Company (Ref. 3-2), commissioned by the then AEC, concluded that retractable tube samplers powered by pneumatic mechanisms, and suitable for pneumatic conveying lines, were the most promising start for an equipment development program. An obvious disadvantage of such devices is that they present a collecting tube as a target to a stream of conveyed particles and may cause particle breakage in the sample. More generally, any sampler upstream of the classifier must treat the sample gently if particle breakage data are to be accurate.

Industrial samplers, which are designed to withstand prolonged usage, are quite large and fit into large ducting. The pilot plant already exists, and there are height constraints at a number of locations where sampling is desirable. Elevated temperatures will be encountered around the burners, and sampling devices suitable for such conditions will probably need to be developed.

Answering the questions relating to sampling accuracy, fuel particle breakage, suitability for elevated temperatures and hot cell use, positioning and orientation, etc., will require experimentation. Therefore, a variety of samplers, with the intent that they be interchangeable, have been ordered. The selection includes:

1. Samplers which divert a part of a stream falling under gravity.
2. Samplers which remove a part of a stream falling under gravity by using a small auger.
3. Samplers which use a retractable sampling tube.

3.2.2.7. Pyrophoricity

3.2.2.7.1. Introduction. Uranium carbide is pyrophoric. The development reference LHTGR fuel is made in a process whereby weak acid resin (WAR) beads are loaded with a uranium solution, dried, and then carbonized. The remaining skeleton ($UC_3O_{0.5}$) is highly reactive -- considerably more reactive than previous, dense kernels. This increased reactivity is attributed to the porosity and hence the high surface area of the material. Although this reactivity is expected to decrease significantly after burnup in a reactor, the Cold Head End Reprocessing Pilot Plant must be equipped to prevent any hazards arising from pyrophoricity. Although hot reprocessing facilities would normally handle less reactive material, a capability to handle hazards arising from fresh fuel may be required. On the other hand, hot facilities also have heat removal equipment in order to deal with decay heat.

The hazard arises when the SiC coating around the kernel cracks, and the kernel is exposed. At the particle crusher, this is what will occur. Upstream of the particle crusher, some breakage in the fuel element size reduction system, transport systems, crushed fuel element burner, and particle classifier is inevitable.

Uranium carbide reacts with oxygen (heat of reaction 283 kcal/g-mole UC_2), CO_2 (137 kcal/g-mole UC_2), nitrogen, and water. The temperatures attained depend on the amount of particle breakage, the supply of the oxidizing agent, and the thermal capacity of material in the neighborhood. The rates of reaction are not known with certainty. Reaction of exposed kernels can be prevented by providing a blanket of inert gas, e.g., argon.

3.2.2.7.2. Possible Precautions. The Cold Head End Pilot Plant fuel element size reduction system, transport systems, and particle classifier are run at an underpressure. This is essentially incompatible with the use of an inert gas through the process, since any leakage will be of air

inward. Conversion to pressurized transport brings with it the risk of discharging fine particles of thorium and uranium compounds.

Containing the entire process within an inert enclosure would be costly. Assuming this is done, however, one then needs to consider hands-on maintenance. The plant is not fully remote, and sooner or later equipment will have to be dismantled. If the plant had been inerted, any exposed pyrophoric material, which had been retained in nooks and crannies, would form a hazard. Assuming that particle breakage in the transport systems will be within bounds, no great hazard should result from allowing individual particles to react with the oxygen in the air conveying them. Furthermore, since reaction with oxygen will probably be almost instantaneous, and since most breakage will probably occur in the bends of the conveying systems, the heat will be liberated where the heat transfer, from particles to gas, is good. Heat transfer from the bunkers is poor, but the breakage that can be reasonably expected in a bunker is also small. Nevertheless, the chance of high temperatures in a bunker cannot be ruled out.

The particle crusher would almost certainly become very hot if it were not purged with argon. Similarly, the argon would prevent the particle crusher product from heating up. The angle of repose feeder, used to feed crushed TRISO particles to the secondary burner, should use argon to fluidize the powder. It is possible that CO_2 could diffuse up the secondary burner feed pipe and cause a temperature problem. If the feed bunker is at a sufficiently large pressure, however, this should not happen. When the batch of feed reaches the secondary burner, a substantial quantity of CO_2 , which is used as a fluidizing medium, will be available. However, heat removal facilities are present in the secondary burner. The operational procedures required to prevent excessive temperatures during the startup of the secondary burner have yet to be finalized.

3.2.2.7.3. Recommendations.

1. A continual argon purge should be provided to the particle crusher and the secondary burner feed hopper.
2. When discharging particle crusher product to the secondary burner, argon should be used in the angle of repose feeder. The argon pressure in the feed hopper must be sufficient to prevent up-flow of CO_2 from the burner. The procedure for charging the burner such that excess temperatures are not generated has yet to be finalized.
3. Thermocouples should be mounted on all bunkers and connected to a multipoint recorder in the Control Room. An alarm, set at 300°C , can then alert the operator. Apart from the secondary burner feed hopper, all bunkers should be connected to an emergency supply of argon, which can flow into the bunkers at a command from the Control Room. In the event that the plant is unmanned, provision should be made to dump argon into the bunkers, after a time delay. The secondary burner feed hopper should not be used for overnight storage.
4. As a backup to the pressure relief valves already present, bursting discs should be installed on all bunkers.

Current Activity Plans do not include WAR fissile particles until 1977.

The impact of pyrophoricity on the design of the HRDF, including the determination of rates of reaction with CO_2 , is being studied.

3.3. PNEUMATIC CLASSIFICATION

3.3.1. Prototype Installation

Installation, check-out, and shakedown are complete.

3.3.2. Documentation

The status of the documentation is given in Table 3-2.

3.4. SOLIDS PROPERTY TESTING

3.4.1. "Cold" Testing

There was no reportable activity during the current quarter.

3.4.2. "Hot" Testing

Discussions with ORNL were held in mid-August in connection with the planned shear cell tests in the hot cell. The immediate goal is procedural compatibility.

REFERENCES

- 3-1. "Thorium Utilization Program Quarterly Progress Report for the Period Ending May 31, 1976," ERDA Report GA-A13949, General Atomic Company, June 30, 1976.
- 3-2. "HTGR Fuel Reprocessing Facility Sampling and Sample Transfer Systems," Ralph M. Parsons Company, Contract AT[10-1]-1500, Job No. 5403-1, AEC Idaho Operations Office, March 1975.

TABLE 3-2
FUEL PARTICLE CLASSIFICATION DOCUMENT STATUS

Number	Type	Title of Document	Status
DC-522101	Doc	Design Criteria: Fuel Particle Classification	Issue A
PF-522101	Dwg	Process Flow Diagram	Issue A
PI-522101	Dwg	Piping and Instrumentation Diagram	Issue A
OP-522101	Doc	Operating Procedures: Fuel Particle Classification	Issue A
MM-522101	Doc	Maintenance Manual: Fuel Particle Classification	--
DR-522101	Doc	Design Report: Fuel Particle Classification	--
PA-500001	Dwg	Plant Arrangement	Issue B
5221002	Dwg	Equipment Assembly	Issue A
5221003	Dwg	Equipment Detail Drawings in Chronological Order	Issue A
SD-522101	Doc	System Description: Fuel Particle Classification	Issue A
IS-522101	Doc	Implementation Specification: Cyclone	Issue A

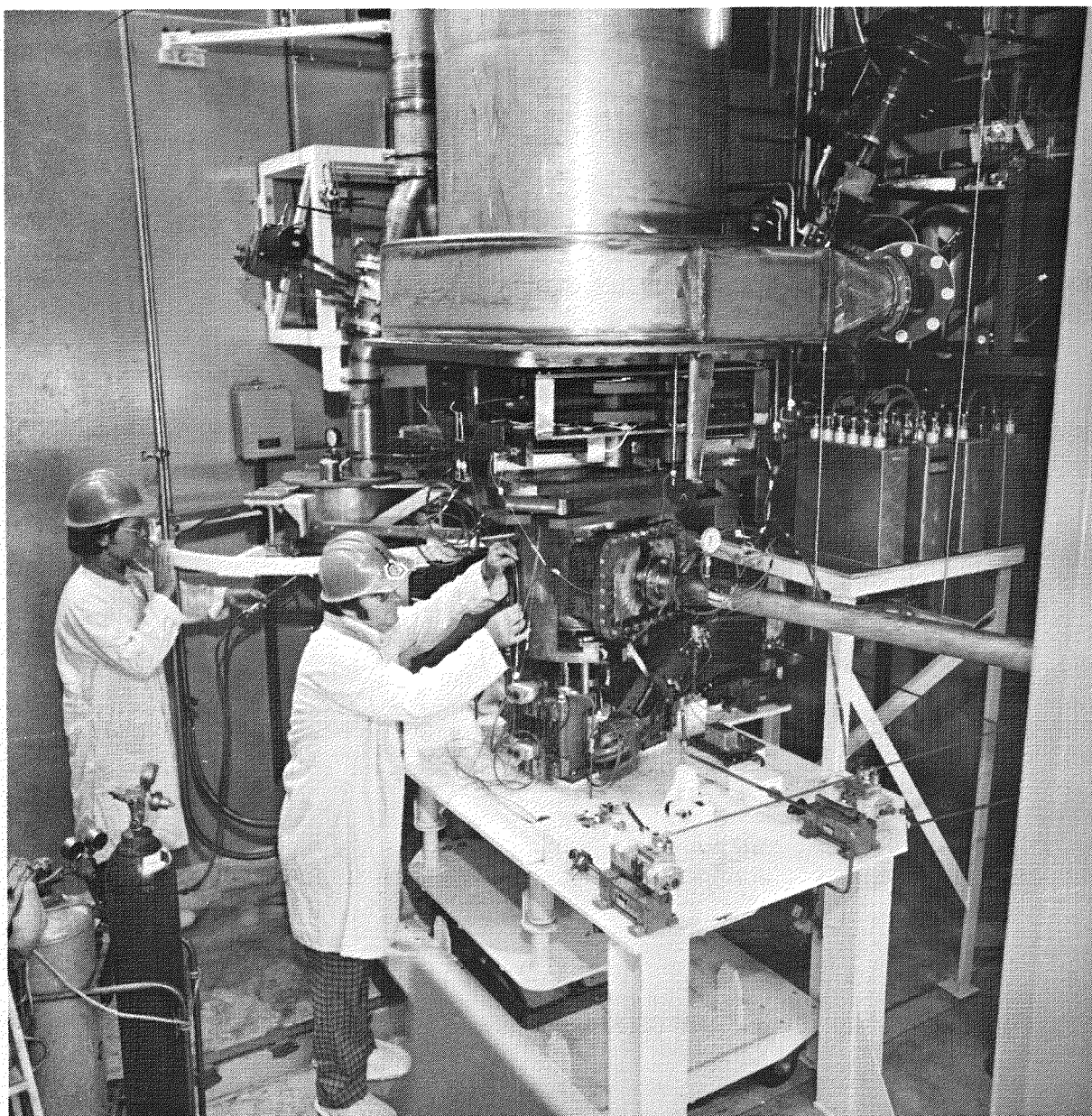
4. FLUIDIZED-BED COMBUSTION

During the past quarter the installation and preoperational check-out of the prototype burners were completed. Experimental work has continued to concentrate on the 20-cm primary burner. Analysis of operation without fines recycle, the startup and tail-burning phases, was completed. Tests on the reference fines systems have also been completed.

4.1. PROTOTYPE 40-CM PRIMARY BURNER

Installation and check-out of the prototype primary burner have been completed. Figure 4-1 shows the final installation of the primary burner plenum and remote clamp. Previously completed equipment installation was described in the previous quarterly report (Ref. 4-1). During preoperational check-out the following checks and tests were performed:

1. Leak check of the burner vessel. Pressure decay was ~ 1.8 psi at a nominal 13 psig over an 18-hr period.
2. Operation of the off-gas analyzer.
3. Setting of gas purge rates.
4. Calibration of differential pressure cells.
5. Calibration and use of control room instrumentation.
6. Check on operability of solenoid valves.
7. Operation of control valves.
8. Calibration of induction coil water flows.



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Fig. 4-1. Primary burner plenum and remote clamp

9. Correlation of gas flow versus distributor ΔP .

10. Tuning of the induction heater circuit.

Figures 4-2 and 4-3 show the instrument cabinets, which contain the control valves and differential pressure cells.

During the check-out of the induction heater, problems were encountered with excessive magnetic susception and heating of the outer metal shroud (this problem is discussed in more detail in Section 4.5). The outer induction shroud is now being removed. Inert bed heatup tests will be performed after copper shielding is applied to the inboard cooling air plenum surfaces and insulation is applied to the induction coil.

Installation of the fines recycle system was completed during the previous quarter with the installation of the fines sampling system equipment and the Orbitran weigh cell equipment. Preoperational check-out revealed no major problems in the operation of the equipment. In several cases, recalibration or adjustment of instruments was required as well as regasketing of several flanges.

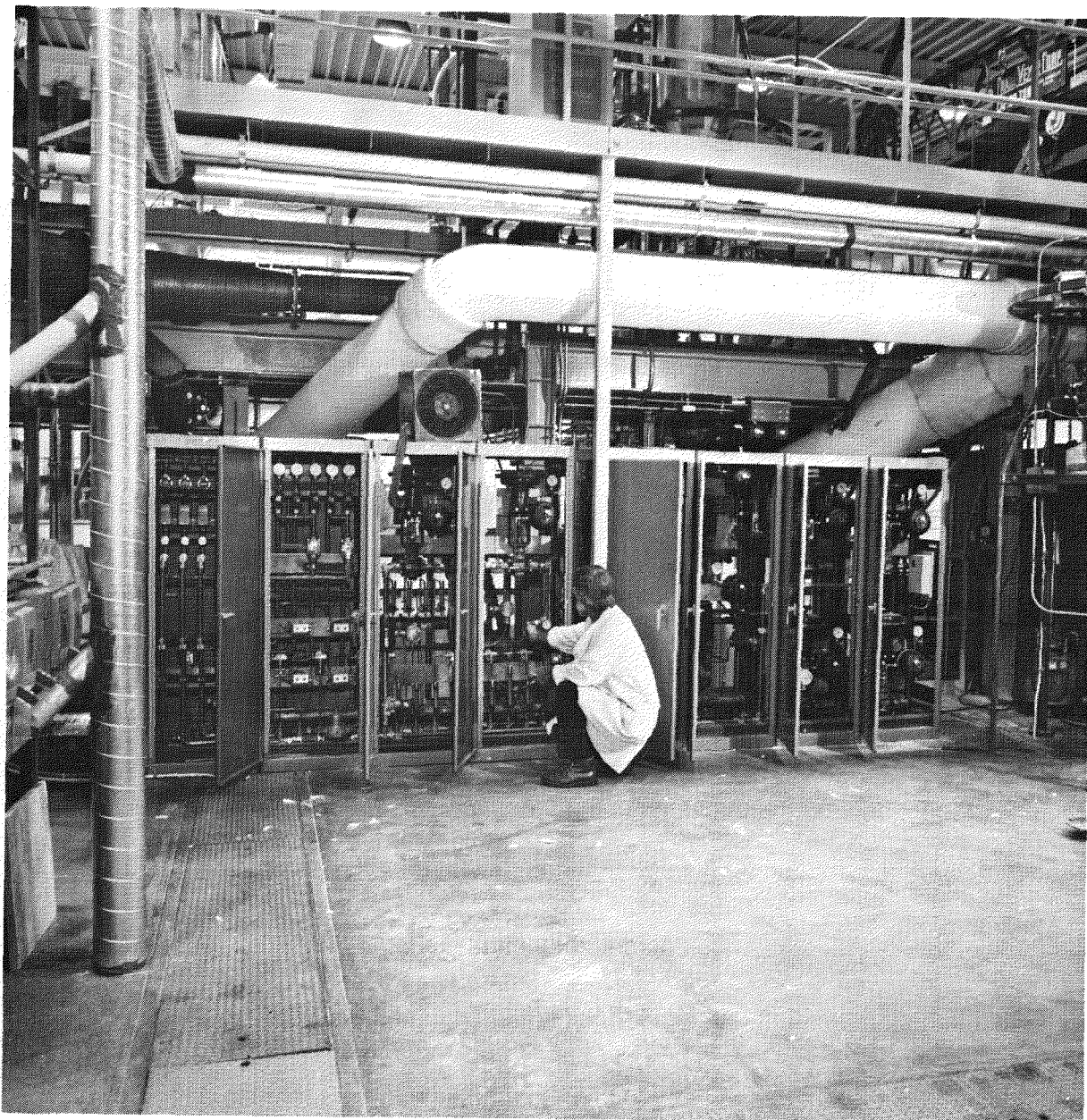
4.2. 20-CM PRIMARY BURNER

4.2.1. 20-cm Primary Burner Experimental Work

Analysis of operation without fines recycle has been completed. Nine of the 11 runs planned to test alternate systems of fines recycle have been successfully concluded.

4.2.1.1. Parametric Analysis of Operation Without Fines Recycle

The first four modules of experiments with the rebuilt 20-cm primary burner have been completed in accordance with the Activity Plan. The



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Fig. 4-2. Burner instrument cabinets



76G1523

Fig. 4-3. Burner operating console

principal objectives of these modules were (1) the determination of startup characteristics with an induction heater, (2) the determination of fines elutriation rates from a startup bed, and (3) the effect of induction heater operation during tail-burning on burnout. The results were correlated using statistical design techniques and have been used to select suitable operating conditions for the subsequent two modules of experiments (fines recycle).

Table 4-1 summarizes the results of the analysis in terms of controlled "independent" variable operating levels recommended to reduce the values of the dependent variables. The high (H, h) and low (L, l) terms indicate that the particular independent variable should be high or low within the range of values investigated. These ranges are given in Table 4-2. An upper-case letter indicates that the independent variable has more than twice as great an impact on the dependent variable as the independent variable with a lower-case letter designation.

The results given in Table 4-1 suggested that the following process control conditions be used:

Induction heated startup (Module 1). Minimum total velocity and bed weight yielded the fastest heating to ignition temperature; using minimum O_2 flow rate for ignition reduced particle breakage and agglomeration. Relatively large feed size (up to 2800- μm diameter on a Rosin-Rammler particle distribution), although not a directly controlled variable, was beneficial in reducing both heatup time and fuel particle breakage-agglomeration. Based on these initial results, it appears that the product criteria for the UNIFRAME (<4760- μm ring size) might be relaxed, although additional confirmatory work must be done first.

Operation with beds containing 83 to 30 wt % carbon and no fresh feed addition (Module 2). Immediate increase of the total velocity (CO_2

TABLE 4-1
PRIMARY BURNER OPERATING LEVELS FOR MINIMIZING DEPENDENT VARIABLES

Module and Dependent Variable	Independent Variable									Comments
	Velocity, U_T (ft/sec)	Average Feed Size, \bar{x}_F (μ m)	Bed Weight, W_B (kg)	Bed O_2 Total, V_O (SLPM)	Bed Carbon X_C (wt %)	Vertex O_2 (SLPM)	O_2 Ramp Rate, \dot{V}_O (SLPM/ MIN)	Above- Bed O_2 , (SLPM)	Feed Rate, \dot{W} (g/min)	
Module 1										Mod. 1: Induction heated startup and ignition Note inversion.
Heatup time	L	H	L							
Axial ΔT	l	L	H							
Radial ΔT	H	H	L							
Fines elutri- ation	L	H	I							
Particle breakage	H	H	H	L						
Agglomeration	H	H	H	L						Note agreement.
Module 2: High C										Mod. 2: High carbon, no fresh feed, decreasing bed carbon
CO in off-gas	L			H	L	l		l		
Axial ΔT	l			h	L	h	l			
Radial ΔT	h			L	H	h	l			
Fines elutri- ation	l			L	H	h		h		
Particle breakage	h			L	H	H	L			
Agglomeration	h			L	H	H	L			Note agreement.
Module 2: Low C										Mod. 2: Low carbon, no fresh feed, decreasing bed carbon
CO in off-gas	l			h	h	h		H		
Axial ΔT	L			h	h	L	l			
Radial ΔT	H			L	l	h	h			
Fines elutri- ation	l			l	L	h		H/L		V_S, H for $X_C > 20\%$ V_S, L for $X_C < 20\%$
Particle breakage	h			L	H	H	L			
Agglomeration	h			L	H	H	L			Note agreement.
Module 3:										Mod. 3: Fresh feed, constant bed carbon ≤ 20 wt %
CO in off-gas	l			h		h		h	L	
Axial ΔT	L			h		L	l		H	
Radial ΔT	h			l		h	h		H	
Fines elutri- ation	l			l	l	h		L	H	
Particle breakage										
Agglomeration										See Appendix, Fig. 6, Ref. 4-2.
Module 4:										Mod. 4: Fail-burning final bed See Appendix, Fig. 1, Ref. 4-2. No significant shift in temperature profile with induction heating. No hot spots.
Bed carbon			H	H						
Axial ΔT			H							
Radial ΔT										

TABLE 4-2
INDEPENDENT VARIABLE RANGES FOR 20-CM PRIMARY BURNER TESTS

Module 1: Heatup and Ignition

Independent (Controlled) Variable	Level	
	Max.	Min.
Total in-bed gas flow velocity (ft/sec)	4	3.6
Type of feed ^(a) (micron averages = d_{RR})	2800	2000
Bed size (kg)	36	32
Total in-bed O ₂ flow rate (SLPM)	150	100

Module 2: High to Low Carbon Bed Operation (No Fresh Feed)

Dependent Variables

Fines elutriation data
Radial and axial burner temperature distribution
Off-gas composition (CO, CO₂, O₂)
Particle and fines data

Independent Variable	Level			
	High Carbon (>50 wt %)		Low Carbon (<50 wt %)	
	Max.	Min.	Max.	Min.
Bed carbon (wt %)	70	50	30	15
Total in-bed gas flow velocity (ft/sec)	4.0	3.6	3.6	3.3
Total in-bed O ₂ flow rate (SLPM)	250	200	350	300
(O ₂) plenum/(O ₂) vertex	∞ (infinity)	4	∞	4
Total O ₂ ramp rate (SLPM)/min)	4	2	8	4
Secondary O ₂ addition (SLPM)	60 ^(b)	0	60 ^(b)	0

TABLE 4-2 (Continued)

Module 3: Constant Low Carbon (15-30 wt %) Bed Operation (Fresh Feed)

Independent Variable	Level	
	Max.	Min.
Fresh feed rate (g/min)	~450	~400
Total in-bed O ₂ flow rate (SLPM)	350	300
Total in-bed gas flow velocity (ft/sec)	3.6	3.3
(O ₂) plenum/(O ₂) vertex	∞	4
Total O ₂ ramp rate (SLPM/min)	4	2
Secondary O ₂ addition	50 ^(b)	0

Module 4: Tail-Burning of Final Particle Bed

Independent Variable	Level	
	Max.	Min.
Tail-burn assist	Yes	No
Percent off-gas O ₂ allowed	10	5
Bed weight (kg)	36	32

(a) The Rosin-Rammler material averages are 66.6% of the R-R plot. The feed consists of 83% graphite and 17% fertile FSV particle (TRISO).

(b) Secondary (above-bed) O₂ was added to a maximum quantity limited by off-gas O₂ increases, indicating the additional O₂ was not being consumed.

diluent) after ignition and minimal O_2 ramp rates decreased lower bed temperature changes and particle breakage-agglomeration. Use of vertex O_2 yielded decreased particle breakage-agglomeration and reduced fines elutriation; secondary O_2 reduced fines elutriation.

Operation with beds containing 15 to 30 wt % carbon and fresh feed addition (Module 3). Secondary O_2 was not beneficial in decreasing fines elutriation with bed carbon weight fractions below 20% and should not be used unless fines are being recycled; maximum rates of top-fed fresh feed were beneficial in dampening fines elutriation and temperature gradients. Other variables should be held as in the above high to low carbon phase.

Tail-burning final bed (Module 4). Induction heating and allowing maximum off-gas O_2 until off-gas CO was no longer detectable minimized final bed carbon. It also appeared that residual carbon in larger beds was more completely burned out.

The above operating conditions were selected from the results given in Table 4-1 by ranking the dependent variables in order of importance. The recommended values for the independent variables could then be chosen more easily when opposite effects on the process occurred.

Induction heated startup. For this condition, optimum operation would minimize the following dependent variables: heatup time, particle breakage-agglomeration, fines elutriation, radial ΔT , and axial ΔT . In recommending process conditions the importance of each was ranked in the order listed; that is, a reduction of heatup time was considered the most important objective, followed by particle breakage-agglomeration, etc.

Operation with beds containing 83 to 30 wt % carbon and no fresh feed addition. For this condition, optimum operation would minimize the

following dependent variables: particle breakage-agglomeration, fines elutriation, off-gas CO, radial ΔT , and axial ΔT . As before, in recommending process conditions the importance of each was ranked in the order listed. This ranking assumes no major process upset, such as explosive off-gas CO concentration, and also considers minor increases in fines elutriation to be well within the capacity of a properly functioning recycle system.

Operation with beds containing 15 to 30 wt % carbon and fresh feed addition. For this condition, optimum operation would minimize the same dependent variables as for the preceding condition. The ranking of variables was also the same.

Tail-burning final bed. For this condition, optimum operation would minimize the fraction of carbon in the final product bed, and this is the most important dependent variable.

It should be pointed out that the results were consistent in linking fuel particle breakage and particle agglomeration. This relationship has been suggested in prior studies on the 20-cm burner.

Conditions for minimizing the axial ΔT were the inverse of conditions to minimize the radial ΔT , with the exception of very high carbon (>50 wt %) operation in which both ΔT 's were minimized by the high vertex O_2 and low O_2 ramp rates.

Other significant results included the general beneficial effect that relatively large feed size and vertex O_2 had on all dependent variables except axial ΔT . Feed size larger than the range investigated (Rosin-Rammler diameter of 2000 to 2800 μm) and abrupt vertex O_2 introduction to a high carbon bed remain as parameters to be investigated further. Larger feed size will have possible implications regarding relaxation of UNIFRAME product criteria.

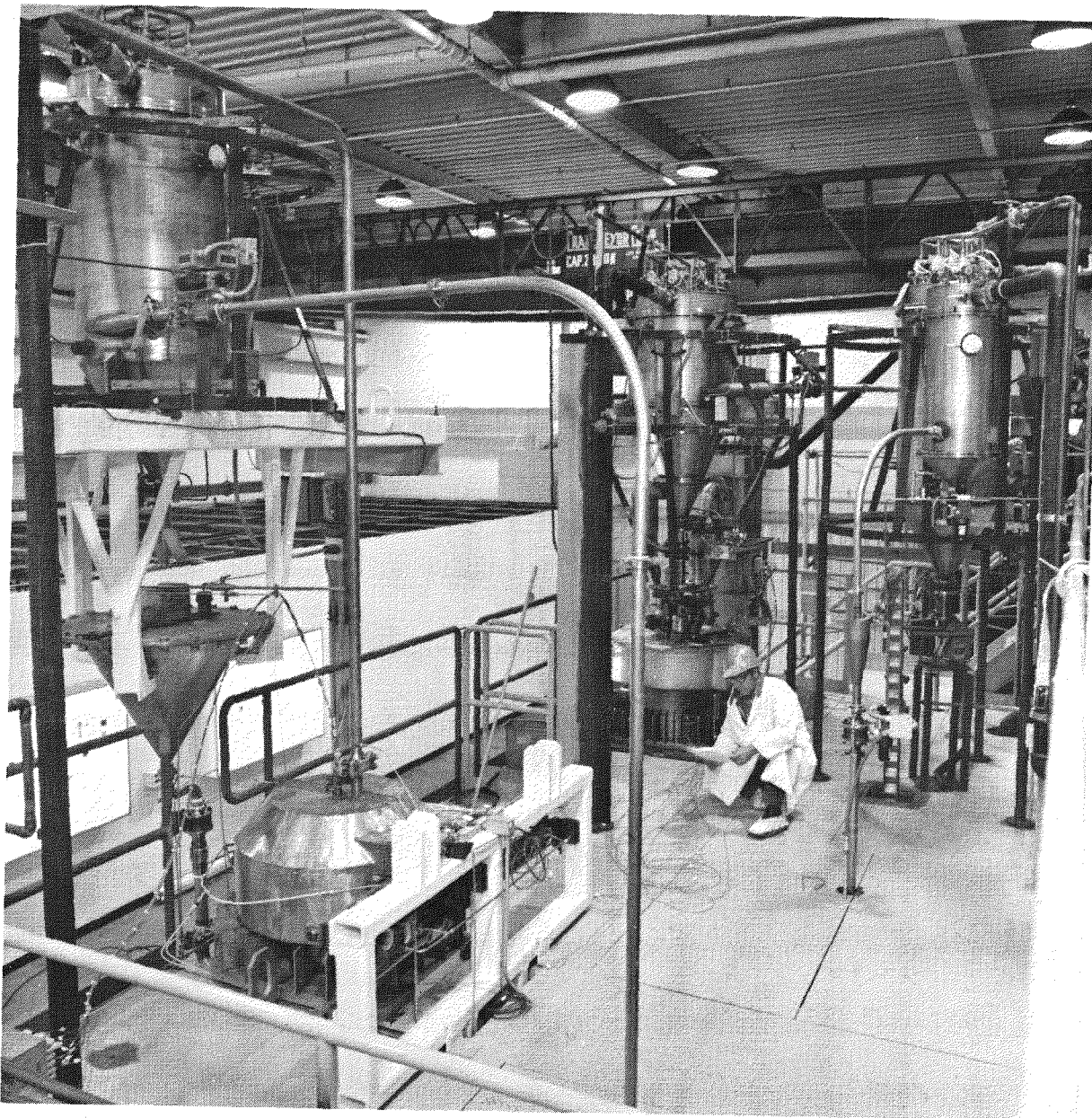
4.2.1.2. Testing of Fines Recycle Alternatives

Nine of the 11 experiments using alternate fines recycle techniques have been successfully completed (modules 5 and 6). One of the nine runs tested 40-cm primary burner type operation using vertex fines recycle with crushed graphite and TRISO/BISO LHTGR fuel particles as feed. This run successfully demonstrated the reference HTGR primary burner operating cycle, i.e., semicontinuous operation with a partial bed removal and subsequent re-ignition of the bed with the induction heater (Ref. 4-2). The other eight runs utilized crushed graphite and TRISO particles. They tested the reference parallel pressurized hopper fines recycle system, recycling fines through the bed (four vertex recycle runs) and above the bed (four above-bed recycle runs).

The remaining two fines recycle tests will be performed with two alternative fines feeding systems. The first system is a gravity system, consisting of a single aerated hopper and a rotary valve (or airlock). The second system utilizes a series of three pressurized hoppers. The gravity system is currently being installed. The parallel hopper system has been removed, and preparations are under way for installation of the series hopper system. Shakedown and hot burner operation of both of these systems should be complete by November. Preliminary results of the module 5 and 6 fines recycle alternative testing should be available for the next quarterly report. The optimum system (the simplest, most reliable fines recycle configuration which yields acceptable fines burning) may then be recommended based on the results of these tests.

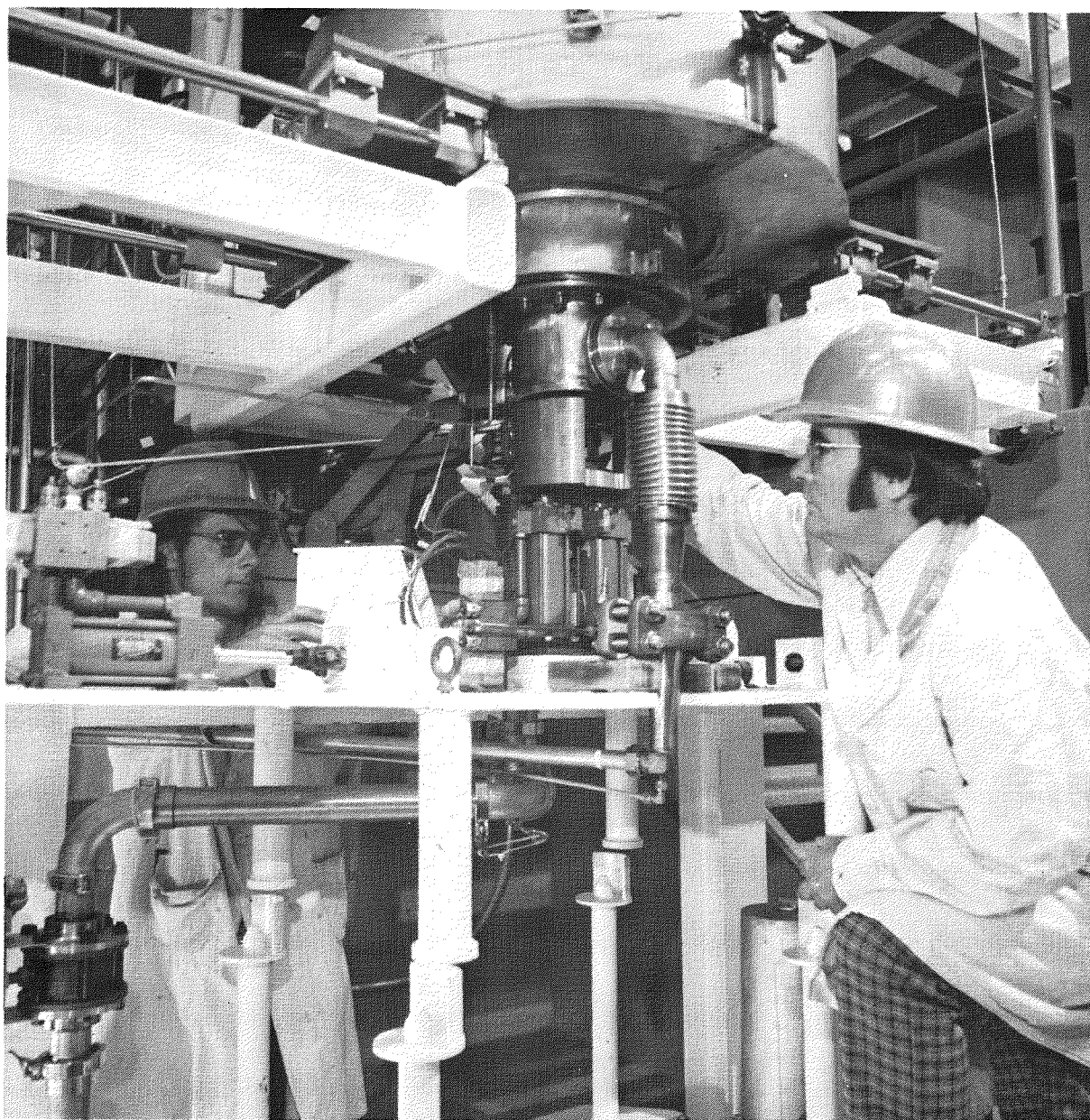
4.3. PROTOTYPE 20-CM SECONDARY BURNER

The previous quarterly report (Ref. 4-1) described the equipment installation for the 20-cm secondary burner. Photographs of the upper and lower sections of the installed 20-cm secondary burner are shown in Figs. 4-4 and 4-5, respectively. Subsequent work has included finishing the gas



76G1535

Fig. 4-4. Upper portion of 20-cm secondary burner



76G1530

Fig. 4-5. Lower portion of 20-cm secondary burner

and water line installation and proceeding through all preoperational check-out tests prior to making the initial burner run. These check-outs are as follows:

1. Leak check of burner vessel. Pressure decay was 0.2 psi at a nominal 12 psig over a 16-hr period.
2. Check on operability of thermocouples.
3. Operation of off-gas analyzer.
4. Setting of gas purge rates.
5. Calibration of differential pressure cells.
6. Calibration and use of control room instrumentation (recorders, controls, etc.).
7. Check on operability of solenoid valves.
8. Operation of control valves.
9. Calibration of induction coil water flows.
10. Tuning of the induction heater circuit.

During the check-out of the induction heater, problems were encountered with excessive magnetic susception and heating of the outer metal shroud (see Section 4.5). The outer shroud was removed to allow further experimental operation while alternate shroud configurations are investigated.

A series of heatup tests were conducted to define limits of operation on the heater system after the outer shroud had been removed. Fifty

kilograms of BISO ThO_2 fuel particles were added to the burner for the tests.

The first heatup resulted in heating the fluid bed to 700°C while fluidizing with CO_2 at a superficial velocity of 80 cm/sec. This was quite satisfactory, as was the information that the maximum support framework temperature was $<100^\circ\text{C}$ during the heatup, which showed that a stray induction field would not heat surrounding equipment to a high temperature. The noise level with the bare coil was 90 dB at 200 Hz; this was reduced in the next heatup by adding 1 in. of moldable silica insulation around the coil.

In the second heatup, all burner thermocouples were recorded to yield thermal transient data. The results indicated excessive ΔT between the lower hub and clamp. Therefore, the outer surface of the clamp was heavily insulated with 2 to 3 in. of silica insulation.

The third heatup again showed excessive, though not as great, ΔT between the lower hub and clamp. This indicated a need for lower power input rates and correspondingly longer heatup time. In addition, embedded clamp and hub thermocouples were added to more accurately reflect average temperature.

These changes were completed prior to a fourth heatup test. It operated at 50% power level (50 kW) and had the embedded thermocouples. Results indicated acceptable clamp to hub temperature gradients (maximum gradient = 130°C). However, at this low power input level, the heatup time was excessive: 2 hr was required to heat the BISO particle bed to 575°C .

Tests will next be run at higher power levels to determine the maximum level at which a clamp to hub temperature gradient does not exceed 200°C . This will be accomplished using the embedded thermocouples. Results will indicate how the actual crushed particle bed may be heated in the forthcoming burner runs.

4.4. 10-CM SECONDARY BURNER

An Activity Plan for the 10-cm secondary burner is currently being reviewed. The burner is being renovated for operation later this year. Most of the work centers on rebuilding the gas supply manifolding.

4.5. PROTOTYPE BURNER INDUCTION HEATERS

4.5.1. Introduction

Both the prototype primary and secondary burners use an induction heater around the lower burner vessel (cylindrical tube) for ignition and tail-burning of bed materials. The induction heating system consists of a solenoid induction coil wound around a metallic susceptor (RA-333 tube), with a solid thermal insulation between the two components, and a power supply system. The burner vessel is heated by the radiated heat from the susceptor (Fig. 4-6). (For details, see Ref. 4-3, Section 4.3.2.1.)

The induction heater is enclosed by a cylindrical stainless steel outer shroud and the upper and lower support plate (Fig. 4-6) and is a physically independent unit separable from the burner. The outer shroud serves the following purposes:

1. Prevents leakage of magnetic flux to the outside of the heater, which might suscept (and generate heat) on any structural material in proximity to the heater or adversely affect performance of electronic equipment.
2. Makes it easy to handle the unit by remote operation.
3. Allows a preliminary decontamination of the unit for repair or maintenance.

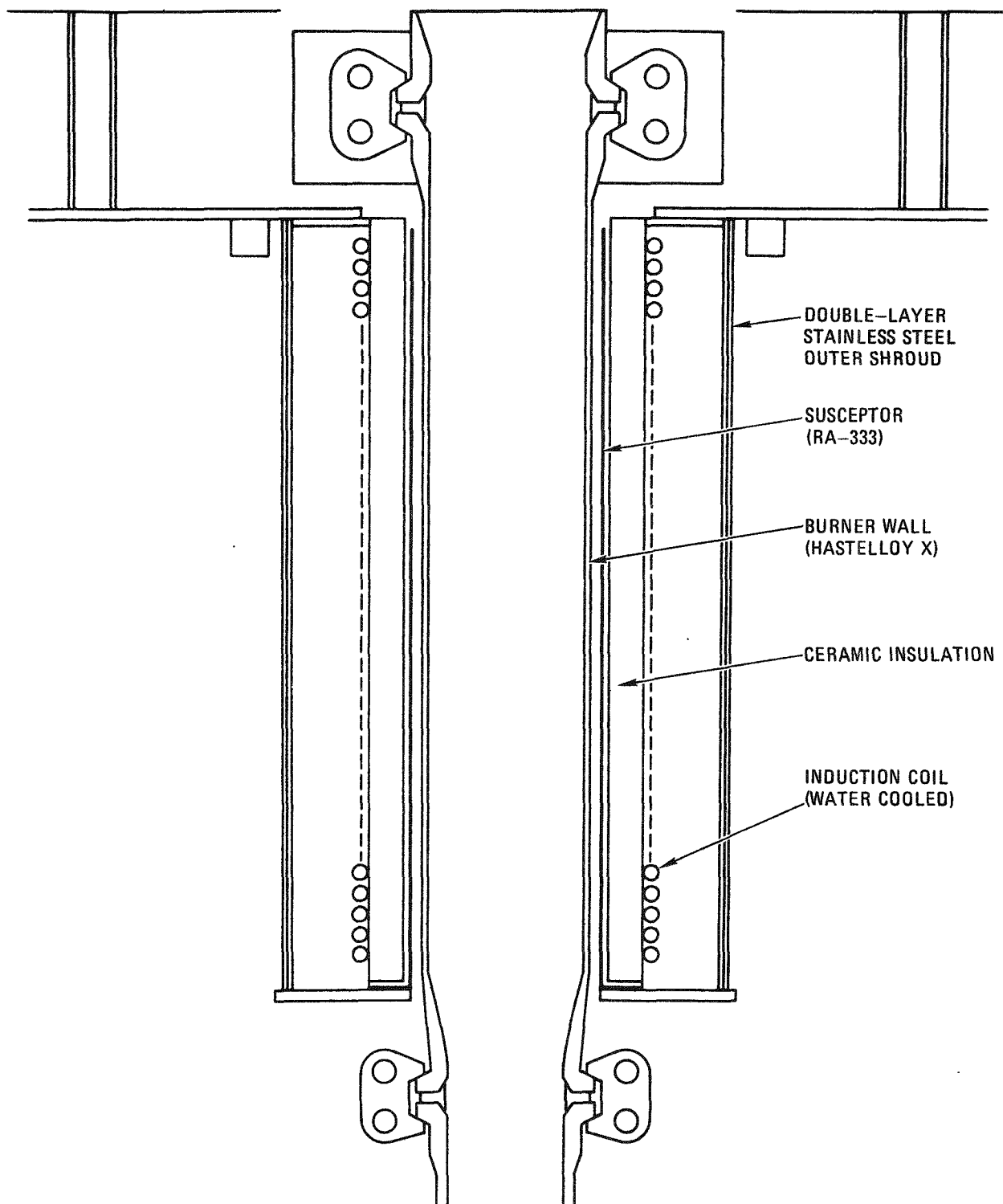


Fig. 4-6. Schematic of induction heater

During the preoperational check-out of the induction heater on both prototype burners, a significant magnetic susception was noticed on the outer shroud. The heat generation, due to magnetic susception, was judged to be unacceptably high. The following section describes the problems observed. Four alternative solutions are described and compared.

4.5.2. Problems Encountered

The heat generation on the outer shroud posed two kinds of problems:

1. Overall heating of the outer shroud.
2. A local hot spot above the slot for the exit coils.

The overall heating of the outer shroud was evidenced by its hot surface temperature. This was probably due to a high mutual induction of the coil and the outer shroud, which also necessitated an addition of large capacitance (~ 500 kVA) over the estimated amount (~ 1000 kVA) to properly balance the circuit.

The local hot spot observed at the apex of the slot was caused by a current concentration, as evidenced by subsequent tests. The direction of currents generated in the outer shroud is circumferential to the cylinder. When a cylinder is complete, the heat is evenly distributed. However, if there is any discontinuity in the cylinder, such as the slot for exit coils, currents find a path to complete the circuit, resulting in a local current concentration as shown in Fig. 4-7. This is a phenomenon similar to a stress concentration in materials.

Owing to this local hot spot, a substantial amount of heat was radiated into nearby coils. This resulted in an increase in the water temperature of the recirculating water system.

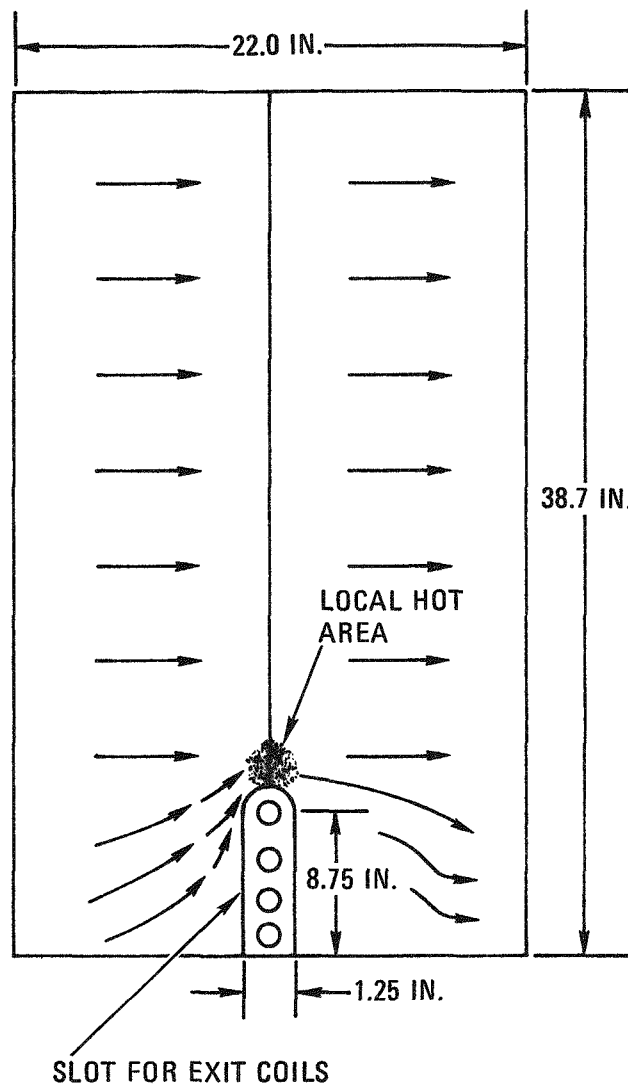


Fig. 4-7. Current concentration in secondary burner outer shroud

The amount of heat dissipated in the outer shroud could not be measured since a steady-state heat balance could not be obtained due to the local hot spot, which was glowing red hot. Additional difficulty was encountered with the coil cooling water system. Due to a flow obstruction, the recirculating water system did not have enough capacity to handle the total heat loss into the coil (this problem has since been solved).

The problems observed for both burners were almost identical. The solutions and recommendations presented below are therefore equally applicable to both burners.

4.5.3. Alternative Solutions

4.5.3.1. No Shroud

This is the simplest way to avoid the shroud heating problem. As discussed previously, the outer shroud has certain technical functions in addition to aesthetic value. A system can be designed without an outer shroud; however, this imposes certain restrictions on the design. The disadvantages should be carefully weighed against the advantages obtained by eliminating the outer shroud. Table 4-3 compares the general aspects of both cases, i.e., shroud versus no shroud.

4.5.3.2. Split Shroud

Electromagnetically, this is identical to the no shroud case. The addition of a shroud serves the purpose of decontamination and ease of handling. Because of the complexity of design required to prevent a complete circumferential circuit and the added material, the cost of fabrication is higher than for the no shroud case.

TABLE 4-3
COMPARISON OF SHROUD VERSUS NO SHROUD CASES

	Shroud	No Shroud
Decontamination	Easy (with complete or split shroud) Difficult (with slotted shroud)	Difficult
Remote handling	Easy	Difficult (exposed coils need protection for handling)
Magnetic shielding to surrounding structures	Yes (the shroud must be electrically conductive around the circumference) No (if the shroud is split with insulation)	No
Heat loss to shroud	Minimal (with low relative resistivity material such as copper or aluminum or with slotting) No loss (with split shroud)	No loss
Cost	Additional cost for shroud	No additional cost
Structural considerations	Outer shroud can add structural rigidity to the heater system	No effect

4.5.3.3. Slotting

Slotting the shroud physically increases the resistance of the circuit by letting the currents pass the detour, completing the circuit circumferentially. The currents flow a longer distance and cannot take a short-cut along the top and bottom cylinders because the electrical potential (hence the current) is developed throughout the entire segmented cylinder.

In order for the slotting method to be useful, the number of slots must be reasonably small. With a small number of slots, however, the total resistance does not increase very much and the heat generation is not reduced substantially. Unless the current is very small, local hot areas due to current concentration will develop at both ends of each slot. This has been confirmed experimentally with four slots.

A large number of slots (at least 16 to 20) may be required to bring the total heat generation to a tolerable range. As a result, the effectiveness of the outer shroud as a shield for decontamination would be lost.

4.5.3.4. Aluminum or Copper Shroud

If copper or aluminum, which have $1/50$ and $1/25$ the relative resistivity of stainless steel, respectively, is used for the shroud material, the percent heat loss to the outer shroud can be reduced by a factor of almost 50 or 25, respectively. This is due to a phenomenon called "skin effect," which increases the actual resistance of the current path by reducing the current penetration into the material at high frequencies. This reduction virtually solves the outer shroud heating problem.

This method of lowering the shroud heat loss is compared with other methods in Table 4-2. Using an aluminum or a copper shroud provides an overall advantage over other alternatives. An outer stainless steel shroud with a copper inner liner can also be used if it is preferable to have a stainless steel outer jacket for ease of decontamination.

4.5.4. Conclusions

Based on the above discussion, the following recommendations are made:

1. For a hot cell application, an outer shroud has more advantages than no shroud.
2. Slotting the shroud can decrease the heat generation but is not a good temporary solution. This method is also not recommended as a permanent solution.
3. Splitting the shroud does not provide a magnetic shield. This method should be used as a permanent solution only when magnetic shielding is not necessary.
4. Using a low relative resistivity material for the outer shroud seems very promising. However, this must be verified using the 10-cm secondary burner induction heater before a new design for the larger burners or any hot cell application is made.
5. For pilot plant purposes, decontamination is not an important factor. For the prototype primary burner, there is no structural member in proximity to the heater. For the prototype secondary burner, the support frames are designed for a high temperature (300°F). No significant heating to the structure has been observed (for additional details, see Section 4.3). In view of these considerations, total elimination of the outer shroud for both burners may serve as both a temporary and a permanent solution.

REFERENCES

- 4-1. "Thorium Utilization Program Quarterly Progress Report for the Period Ending May 31, 1976," ERDA Report GA-A13949, General Atomic Company, May 31, 1976.
- 4-2. "Thorium Utilization Program Quarterly Progress Report for the Period Ending February 29, 1976," ERDA Report GA-A13833, General Atomic Company, March 31, 1976, pp. 4-17 through 4-23.
- 4-3. "Thorium Utilization Program Quarterly Progress Report for the Period Ending May 31, 1975," ERDA Report GA-A13510, General Atomic Company, August 15, 1975.

5. AQUEOUS SEPARATIONS

5.1. SUMMARY

Bench-scale experiments were used to study the effects of graphite processed through dissolution and feed adjustment steps on solvent extraction scrub section zirconium distribution coefficients. Only minor effects were observed when comparisons were made with control samples. Additional tests are planned with a slightly modified procedure to investigate silicon carbide and perform further studies of heavy metal carbide effects.

A preliminary study was made on formic acid denitration of uranyl nitrate solutions as an alternative to amine extraction. The results of this study show promise, and additional work is recommended.

Studies of solvent nitration continued during the quarter, with efforts being directed toward the safety of the thorium intercycle concentrator.

5.2. LEACHING

There was no reportable activity during the current quarter.

5.3. FEED ADJUSTMENT

There was no reportable activity during the current quarter.

5.4. BENCH-SCALE INVESTIGATIONS

5.4.1. Effect of Graphite Feed Content on Solvent Extraction Zr-95 Distribution

In a previous study (Ref. 5-1), reduced solvent extraction decontamination factors for ⁹⁵Zr-Nb were observed with feed solutions containing added carbide carbon following feed acidity adjustment by evaporation to 135°C and subsequent steam sparging. The current study was performed to assess the effect of the presence of graphite during solvent extraction feed preparation. The presence of graphite in dissolver solutions prepared from secondary burner ash is considered more likely than carbide carbon owing to incomplete oxidation during the burning operations.

In the current flowsheet (Ref. 5-2), a feed adjustment step is specified for removal of excess nitric acid prior to solvent extraction separation. Excess nitric acid is to be removed by evaporation to 135°C followed by steam sparging, should acid deficiency be required. Small amounts of graphite are usually present in aqueous leachates prepared for solvent extraction as a result of incomplete oxidation of fuel block fragments. Therefore, it is important to assess the effect of evaporation and steam sparging on resultant zirconium decontamination factors in the presence of potential reaction products of graphite and nitric acid.

Bench-scale experiments were performed to study the effect of (1) evaporation to 135°C and (2) evaporation to 135°C with steam sparging on zirconium decontamination values in the presence of graphite.

Solvent extraction samples were prepared for study by the addition of graphite to Zr-95 "spiked" 1M thorium-Thorex solution. Graphite was added in an amount of 0.1% by weight prior to evaporation to 135°C or evaporation to 135°C with subsequent steam sparging. This corresponds to 0.7% graphite in the secondary burner ash (SiC-free basis). For control purposes, a

sample set was prepared from feed adjustment solutions in which no graphite was added. Following adjustment of the boiler pot product to 0.4M Th^{+4} and 1M H^{+} and removal of the graphite by filtration, an initial extraction with 30% TBP/NPH was performed in a separatory funnel. A scrub solution containing 0.25M Th^{+4} and 1M H^{+} was used for all subsequent contacts. Gamma spectrometry with GeLi detection was utilized for measurement of Zr-95 and Nb-95 activity in organic and aqueous layers following phase separation by centrifugation.

The experimental design is shown in Fig. 5-1, and ^{95}Zr - ^{95}Nb gamma spectrometry data appear in Tables 5-1 and 5-2.

The net effect of the presence of 0.1% graphite feed content on calculated Zr-95 distribution coefficients is less than that observed in previous work (Ref. 5-1) for solvent extraction feed solutions containing 1000 ppm carbide carbon/gram heavy metal (equivalent to 0.008% on a weight carbide/weight feed solution basis).

Apparently, if nitric acid-graphite reaction products are formed during leaching of secondary burner ash, either (1) extractable zirconium complexes are not formed or (2) the complexes are effectively destroyed during feed adjustment.

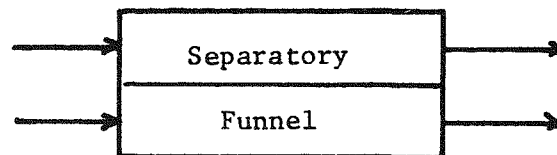
5.4.2. Denitration of Uranyl Nitrate with Formic Acid - Preliminary Study

Formic acid treatment has been shown to be effective in the denitration of thorium nitrate at 135°C (Ref. 5-2). A preliminary study was performed to assess the feasibility of formic acid denitration in the uranyl nitrate system. Production of acid deficient uranyl nitrate solutions with formic acid offers a potential alternative to the reference flowsheet amine extraction process (Ref. 5-1) currently specified for uranium loading of weak acid resins in U-233 recycle fuel flowsheets (Ref. 5-3).

Extraction

125 ml 30% TBP/NPH

125 ml Aqueous *



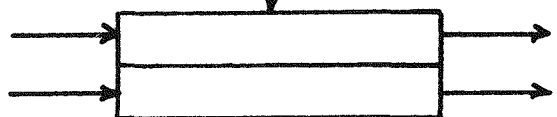
Sample 1 - 25 ml Organic Layer

Sample 2 - Entire Aqueous Layer

Scrub

100 ml 30% TBP/NPB

100 ml Scrub Solution **

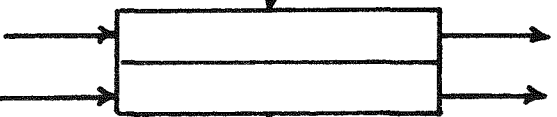


Sample 3 - 25 ml Organic Layer

Sample 4 - Entire Aqueous Layer

75 ml 30% TBP/NPH

75 ml Scrub

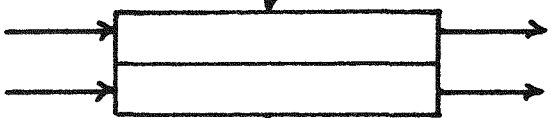


Sample 5 - 25 ml Organic Layer

Sample 6 - Entire Aqueous Layer

50 ml 30% TBP/NPH

50 ml Scrub



Sample 7 - 25 ml Organic Layer

Sample 8 - Entire Aqueous Layer

25 ml 30% TBP/NPH

25 ml Scrub



Sample 9 - 25 ml Organic Layer

Sample 10 - Entire Aqueous Layer

* 0.4 M Th⁴⁺, 1.0 M H⁺ - Feed Solution

** 0.25 M Th⁴⁺, 1.0 M H⁺ - Scrub Solution

Fig. 5-1. Experimental design for ⁹⁵Zr distribution coefficient studies

TABLE 5-1
DATA SUMMARY FOR ^{95}Zr - ^{95}Nb GRAPHITE STUDIES - EVAPORATION TO 135°C ONLY

Sample No. (a)	Dilution Factor	$\left(\frac{\text{Counts}}{\text{Unit Time}} \times \text{Dilution Factor} \right)$		Calculated K Value (b)	
		^{95}Zr	^{95}Nb	^{95}Zr	^{95}Nb
1	None	454	229	0.032	0.006
2	1/25	14,125	40,200		
3	None	12	18	0.026	0.035
4	None	464	516		
5	None	0.8	7.7	0.035	0.065
6	None	23	119		
7	None	0.5	3.6	0.062	0.044
8	None	8	81		
9	None	2.2	5	0.260	0.036
10	None	8.5	140		
1A	None	392	214	0.030	0.006
1B	None	13,225	37,075		
1C	None	11.4	17.1	0.029	0.027
1D	None	395	643		
1E	None	1.2	8.9	0.052	0.050
1F	None	23	179		
1G	None	--	--	--	--
1H	None	13	92		
1I	None	0.7	7.1	0.089	0.048
1J	None	7.9	147		

(a) Numbers designate control samples; letters designate samples containing graphite.

(b) $K = \frac{\text{Conc. Isotope Organic Layer}}{\text{Conc. Isotope Aqueous Layer}} = \text{Distribution Coefficient.}$

TABLE 5-2
DATA SUMMARY FOR ^{95}Zr - ^{95}Nb GRAPHITE STUDIES - EVAPORATION TO 135°C WITH STEAM SPARGING

Sample No. (a)	Dilution Factor	$\left(\frac{\text{Counts}}{\text{Unit Time}} \times \text{Dilution Factor} \right)$		Calculated K Value (b)	
		^{95}Zr	^{95}Nb	^{95}Zr	^{95}Nb
1	None	291	192	0.020	0.005
2	1/25	14,675	38,950		
3	None	9.6	15.7	0.032	0.032
4	None	296	498		
5	None	1.4	7.1	0.040	0.028
6	None	35	255		
7	None	1.1	4.3	0.050	0.040
8	None	22	107		
9	None	1.8	7.2	0.105	0.065
10	None	17	111		
1A	None	360	189	0.022	0.005
1B	None	15,800	38,250		
1C	None	10.7	13.6	0.030	0.023
1D	None	352	588		
1E	None	0.9	6.8	0.037	0.079
1F	None	24	86		
1G	None	0.6	6.7	0.070	0.093
1H	None	8.6	72		
1I	None	0.8	5.7	0.085	0.051
1J	None	9.6	111		

(a) Numbers designate control samples; letters designate samples containing graphite.

(b) $K = \frac{\text{Conc. Isotope Organic Layer}}{\text{Conc. Isotope Aqueous Layer}} = \text{Distribution Coefficient.}$

Denitration of uranyl nitrate was accomplished with formic acid treatment at 125° and 112°C. Precipitation occurred in some molten salt solutions at 125°C; however, dilution of the boiler pot product at ambient temperature generally resulted in precipitate dissolution and resultant nitrate to uranium molar ratios of less than 2.0. Incipient precipitation at 125°C was noted for formic acid to uranium molar ratios in excess of 0.07. Precipitation was not observed in formic acid treatments conducted at 112°C with formic acid to uranium molar ratios to 0.86, and the resultant product solutions were also found to have the required nitrate to uranium molar ratio of 1.6 without precipitation occurring.

The equipment shown in Fig. 5-2 was used for all denitration experiments described herein. A weighed quantity of reagent-grade uranyl nitrate hexahydrate (UNH) was placed in the boiler pot and heated to a specified boiling temperature.* On attainment of constant temperature, dilute formic acid solution was added to the uranyl nitrate solution with a metering pump. Following formic acid treatment, stripping water was added to the boiler pot at temperature from a second metering pump in order to remove unreacted formic acid. Finally, stripping water was allowed to accumulate in the boiler pot as a means of dilution prior to product removal at ambient temperature.

Upon completion of each experiment, the final pH of the denitrated product (diluted to a known concentration) was measured and samples were taken for uranium and nitrate analyses. The final pH value was used to estimate the nitrate to uranium molar ratio (Ref. 5-3).

Experimental results are presented in Table 5-3. Examination of the data in the table indicates a NO_3^-/U molar ratio of less than 2.0 was obtained in all cases. Precipitate formation was encountered in denitration experiments performed at 125°C. The precipitate was probably UO_3 or $\text{UO}_2(\text{NO}_3)_{2-x}(\text{OH})_x$; however, characterization of the precipitate was not attempted in this preliminary study. In most cases the precipitate formed

*In some experiments, water was added to the boiler pot to yield a UNH solution with a boiling point of 112°C.

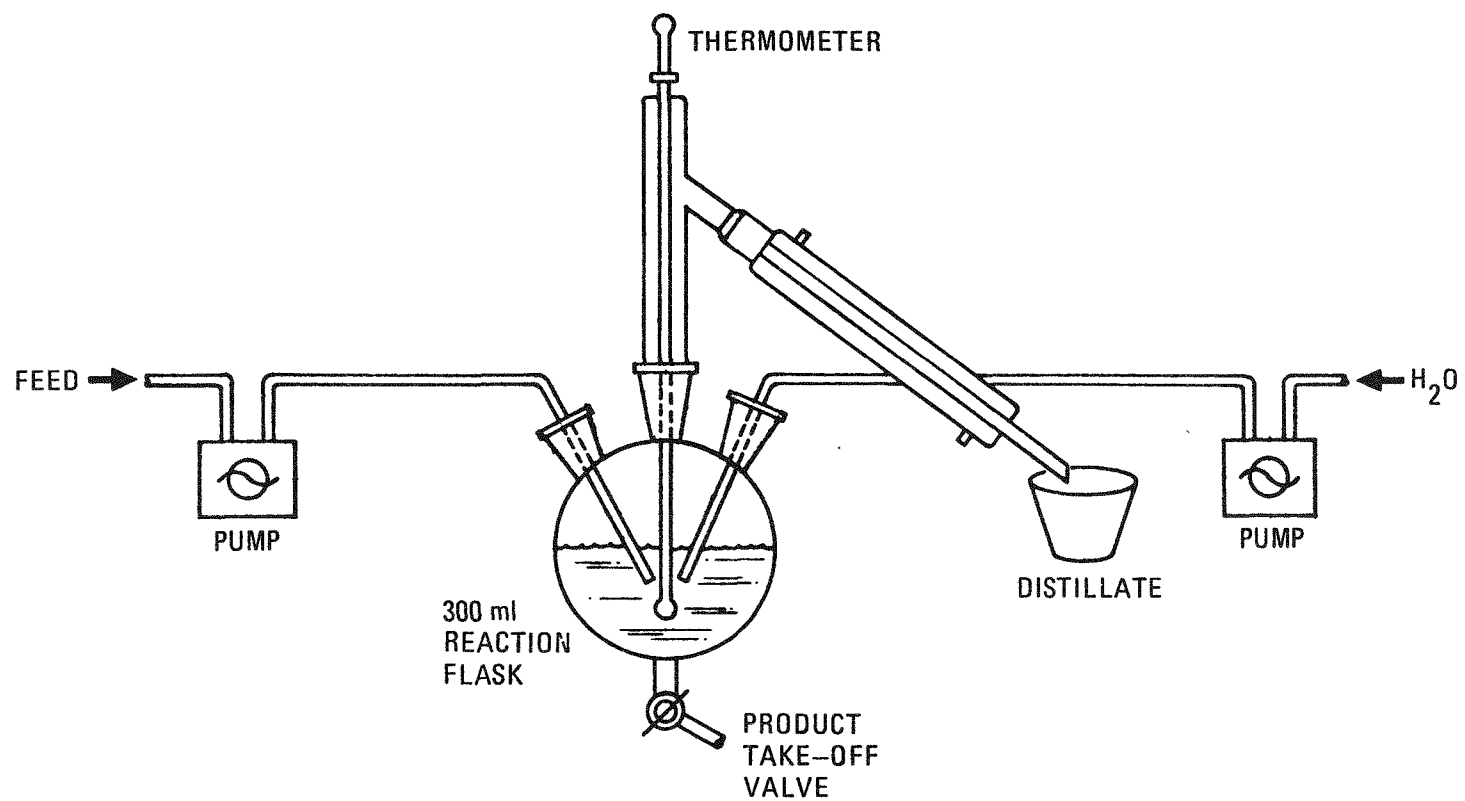


Fig. 5-2. Experimental apparatus for UNH-formic acid denitration

TABLE 5-3

DATA SUMMARY FOR URANYL NITRATE-FORMIC ACID DENITRATION EXPERIMENTS

Expt. No.	Denitration Temp(°C) (a)	UNH Weight (g)	Volume HCO ₂ H (88%) to Boiler Pot During Run (ml) (b)	Presence of Precipitate During Denitration	Total Volume Stripping Water Added to Boiler Pot (ml) (b)	Final Product Solution				
						Analytical				Predicted (c)
						pH	[U]M	[NO ₃ ⁻]M	[NO ₃ ⁻]M/[U]M	[NO ₃ ⁻]M/[U]M
1	125	150	5.0 in. 200 ml H ₂ O	Positive	50	2.70	0.929	1.377	1.48	< 1.5
2	125	100	0.61 in. 122 ml H ₂ O	Positive	10	2.05	1.050	1.940	1.85	1.8
3	112	100	5.0 in. 200 ml H ₂ O	Negative	100	2.70	0.472	0.835	1.77	1.7
6	112	100	7.5 in. 200 ml H ₂ O	Negative	57	2.83	0.463	0.781	1.69	1.6
7	112	100	7.0 in. 200 ml H ₂ O	Negative	183	2.84	0.486	0.773	1.59	1.5

(a) Temperature was held constant at the value shown during formic acid feed and stripping water addition. Temperatures indicated are the atmospheric boiling points of the respective solutions.

(b) The feed rate for formic acid and stripping water was 3.33 ml/min for all the above runs.

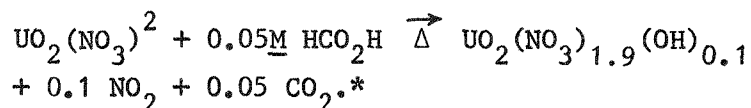
(c) Ref. 5-3.

completely dissolved on dilution at ambient temperature. Precipitation did not occur in denitration experiments conducted at 112°C, although a gradual darkening of the diluted uranium product solutions may indicate the presence of unreacted residual formic acid at this temperature.

While initial results are encouraging, additional study is required to optimize conditions for preparation of acid deficient uranyl nitrate solutions via a formic acid denitration process.

The agreement between predicted (Ref. 5-3) and analytical NO_3^-/U molar ratios is excellent. Thus, formic acid denitration of uranyl nitrate appears to be a feasible technique. Additional effort is needed to complete the UNH denitration feasibility study. Areas requiring further work include:

1. Definition of the principal denitration reaction. A general formic acid denitration reaction is as follows:



2. Characterization of denitration precipitate.
3. Definition of limiting conditions for precipitate formation during denitration.
4. Determination of conditions for removal of unreacted formic acid from denitrated product solutions. Possible approaches include:
 - a. Additional steam stripping.
 - b. Refluxing of product solutions.

*Equations can also be written with NO as a product.

5. Determination of formic acid content in distillate (overhead) and methods for its destruction.
6. Pilot plant testing of the denitration technique.
7. Preparation of uranium-loaded weak acid resins using UNH prepared by a formic acid denitration method.
8. Optimization of process parameters.

5.4.3. Solvent Nitration During Thorium Nitrate Concentration - Additional Studies

The continuing bench-scale solvent nitration study (Refs. 5-1 and 5-4) was expanded during the current reporting period to encompass safety aspects of the thorium intercycle concentrator operation.

The occurrence of vigorous reactions during concentrator operations has been reported for the uranyl nitrate/solvent (TBP in Shell Spray Base) system (Ref. 5-5). Consequently, bench-scale experiments were conducted in an attempt to find conditions where vigorous reactions between thorium nitrate and solvent (TBP in a diluent) would occur. The compositions of the aqueous solutions prepared for study approximate those of concentrator bottom solutions that would result during overconcentration due to maloperation of the thorium intercycle concentrator.

Thorium product stream (1BT) from the Acid-Thorex co-decontamination/partition cycle is concentrated prior to being fed to the extraction column of the second thorium solvent extraction cycle (Ref. 5-2). Bench-scale studies were undertaken on certain safety aspects of the thorium intercycle concentrator operation. This work was conducted to assess the potential for a vigorous reaction occurring due to the presence of TBP/diluent in the thorium intercycle concentrator and subsequent unintentional overconcentration of the thorium product. It should be noted that large amounts of TBP/

diluent would enter the thorium intercycle concentrator only during inadvertent organic flooding of the first cycle thorium partition scrub column (1BS).

Chemical analyses of the 1TC (intercycle concentrator product) solution from solvent extraction run 51 gave a composition of 1.43M Th^{+4} and 3.50M H^{+} . The boiling point of the solution was determined to be 112°C . An aqueous solution of the above composition was prepared from reagent-grade thorium nitrate tetrahydrate and nitric acid and was used for all experiments. Two organic solutions were prepared for static flooding and continuous flooding tests. These solutions were composed of 30% TBP by volume dissolved in (1) normal paraffin hydrocarbon-NPH (South Hampton Company) and (2) deodorized spray base (Chevron 450 Solvent).

5.4.3.1. Thorium Intercycle Concentrator - Organic Static Flooding Tests

A sketch of the apparatus used for static flooding tests is shown in Fig. 5-3.

In these tests an amount of TBP/diluent solution sufficient to yield a TBP/thorium weight ratio of 0.15* was added to a simulated thorium intercycle concentrator product whose boiling point had been adjusted by evaporation to a predetermined temperature prior to operation under reflux. Thorium intercycle concentrator bottom solutions with boiling points greater than 118°C would result only during maloperation of the thorium intercycle concentrator. Following addition of the organic solution, the system was allowed to reflux for several hours to monitor (1) the occurrence of a vigorous reaction and (2) the formation of "red-oil" (evidence of solvent degradation).

*This ratio was selected because work at Savannah River indicated a minimum critical ratio of 0.12 for vigorous reactions with the TBP/uranyl nitrate system (Ref. 5-6).

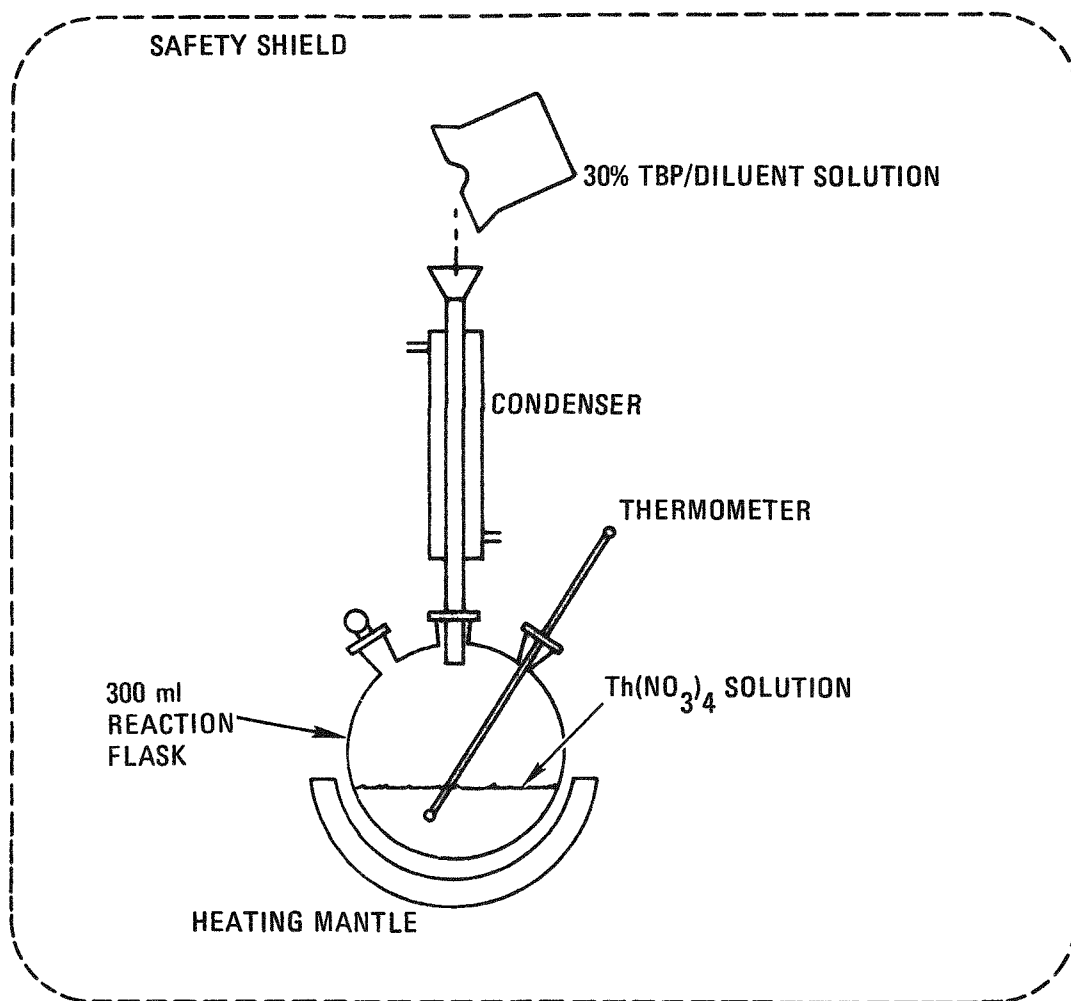


Fig. 5-3. Experimental apparatus for static organic flooding tests

5.4.3.2. Thorium Intercycle Concentrator - Continuous Organic Addition Tests

A sketch of the apparatus used for continuous organic flooding tests is shown in Fig. 5-4.

The aqueous solution described above was heated to 135°C to simulate thorium intercycle product overconcentration which could occur during maloperation of a thorium intercycle concentrator. On attainment of 135°C, 30% TBP/diluent was added to the boiler pot at a rate of 0.42 ml/min with a metering pump. The total volume of 30% TBP/diluent added was 25.2 ml, which is equivalent to a TBP/thorium weight ratio of 0.23.

5.4.3.3. Results

Experimental data obtained and observations recorded during organic static flooding tests are presented in Tables 5-4 and 5-5. No vigorous reactions were observed in these tests with either the NPH or Chevron Spray Base systems. Evolution of NO₂ was observed in all tests performed. The presence of "red-oil" was noted in all tests in which Chevron Spray Base was used as the diluent. A slight red organic coloration was observed for the NPH diluent system only after 12 hr of refluxing at 150°C.

In continuous organic addition experiments, no vigorous reaction occurred at 135°C. Examination of boiler pot contents following continuous operation revealed "red-oil" formation only in tests employing Chevron Spray Base diluent.

Bench-scale testing has given no evidence to support the existence of an undue hazard in the planned operation of the thorium intercycle concentrator. Also, no vigorous reactions were encountered in tests designed to simulate thorium intercycle maloperation (temperatures to 150°C) with either diluent. Tests described herein indicate both NPH and Chevron Spray Base are acceptable as diluents from a safety standpoint; however, solvent

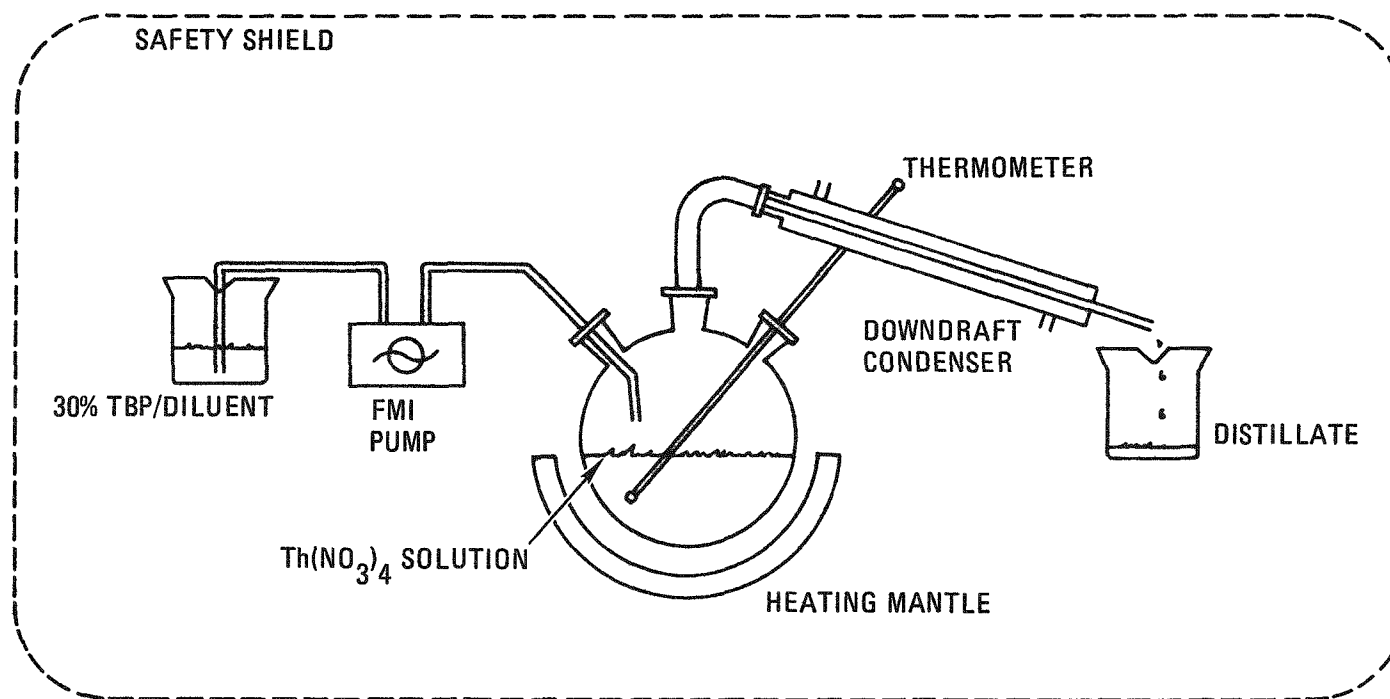


Fig. 5-4. Experimental apparatus for continuous organic addition tests

TABLE 5-4

DATA SUMMARY FOR STATIC FLOODING TESTS USING TBP/CHEVRON SPRAY BASE SYSTEM

Test No.	Boiling Point Aqueous Sol'n (°C)	Total Initial Vol. Aqueous Sol'n (ml) ^(a)	Time Period for Organic Add'n (min)	Total Volume 30% TBP/S. Base Added (ml)	Total Reflux Time (hr)	Observation ^(b,c)
1	118	67.5	10	10	12	A.
2	125	67.5	15	10	16	B.
3	135	101.3	20	15	8.5	C.
4	150	101.3	20	15	4.25	D.

(a) Initial aqueous composition = 1.38M Th^{+4} , 3.37M H^{+} .

(b) All organic layers had red colorations. The color intensity increased directly as a function of temperature. The NO_2 evolution rate appeared to decrease with increasing temperature and may be due to decreasing free acidity at higher temperatures.

- (c)
- A. Considerable NO_2 evolution. No foaming.
 - B. Considerable NO_2 evolution. No foaming.
 - C. NO_2 evolution. Some evidence of foaming.
 - D. NO_2 evolution. Some evidence of foaming. Boiling point (constant) of aqueous layer dropped to 135°C by end of reflux period (due possibly to lower boiling point of the denitrated salt).

TABLE 5-5

DATA SUMMARY FOR STATIC FLOODING TESTS USING TBP/NPH SYSTEM

Test No.	Boiling Point Aqueous Soln (°C)	Total Initial Vol. Aqueous Soln (ml) ^(a)	Time Period for Organic Addn (min)	Total Volume 30% TBP/NPH Added (ml)	Total Reflux Time (hr)	Observation ^(b,c)
1	112	67.5	5	10	19.5	A
2	117	67.5	5	10	16.0	B
3	124	67.5	5	10	18.75	C
4	130	101.2	10	15	14.75	D
5	135	101.2	15	15	13.5	E
6	150	101.2	5	15	12.0	F

^(a)Initial aqueous composition = $1.38M$ Th^{+4} , $3.37M$ H^{+} .

^(b)Only the organic layer from test 6 had a slight red coloration. The NO_2 evolution rate appeared to decrease with increasing temperature and may be due to decreasing free acidity at higher temperatures.

- ^(c)
- A. Slow NO_2 evolution throughout reflux period. No foaming.
 - B. NO_2 evolution more rapid than test 1. Gelatinous ppt observed on return to ambient temperature. It is speculated that ppt is a reaction product of thorium and TBP hydrolysis products. No foaming.
 - C. NO_2 evolution. No foaming.
 - D. NO_2 evolution. No foaming.
 - E. NO_2 evolution. Boiling point (constant) of aqueous layer dropped to $122^{\circ}C$ by end of reflux period.
 - F. Slow NO_2 evolution. Boiling point (constant) of aqueous layer dropped to $135^{\circ}C$ by end of reflux period.

degradation effects (as evidenced by the presence of "red-oil") were judged to be more pronounced in the Chevron Spray Base system.

The current bench-scale effort was undertaken to serve as a guide for pilot- and plant-scale thorium intercycle concentrator operations.

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6. SOLVENT EXTRACTION

6.1. SUMMARY

Five solvent extraction runs were completed during the quarter. These runs represented the first cycle of the Acid-Thorex flowsheet (Fig. 6-1). Runs 51, 52, and 53 were made using the facilities in the pilot plant five-pulse-column system. These runs included the addition of nonradioactive and radioactive zirconium. Dibutyl phosphate (DBP) was added to the 1A column to simulate solvent degradation.

Runs 54 and 55 utilized the five pulse columns plus the Robatel centrifugal contactor. The centrifugal contactor was used as the 1A extraction section. The five pulse columns were used for the other first-cycle operations.

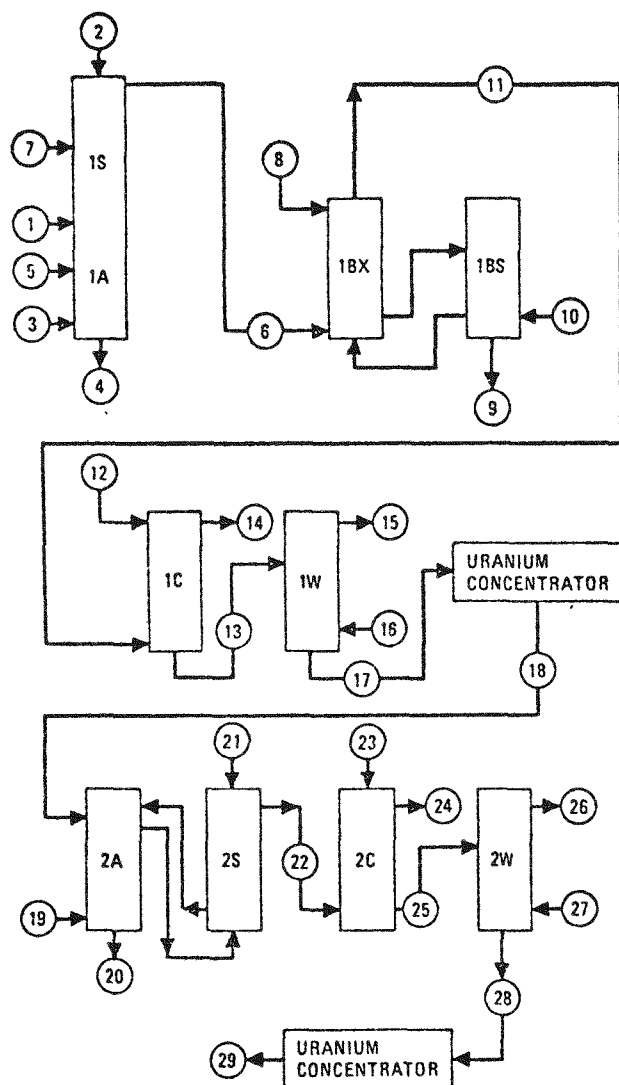
6.2. PROCESS MODIFICATIONS

For runs 52, 53, 54, and 55, the length of the cartridge section of the 1BX column was extended from 4.57 to 5.79 meters.

For runs 54 and 55, the Robatel centrifugal contactor was connected to the pulse column system. An airlift system was used for the 1AP stream from the contactor to the 1S column. This system is shown in Fig. 6-2.

6.3. RESULTS AND DISCUSSION - RUNS 51, 52, 53

Table 6-1 gives the operating conditions for runs 51, 52, and 53. Table 6-2 contains column and cartridge descriptions.



STREAM	STREAM NO.	RELATIVE FLOW	COMPOSITION		
			U (G/L)	Th (G/L)	HNO ₃ (M)
1AF	1	100	35	348	1.0
1AS	2	104 *			0.01
1AX	3	1000		(30% TBP)	
1AW	4	262		(FISSION PRODUCTS)	
1AA	5	32			13.0
1SP	6	1000	3.5	35	
1AIS	7	26			5.0
1BX	8	600			0.2
1BT	9	600	Trace	102	
1BS	10	179		(30% TBP)	
1BU	11	1180	2.98		
1CX	12	593			0.01
1CU	13	593	5.93		
1CW	14	1180		(30% TBP)	
1WW	15	59		(NPH)	
1WS	16	59		(NPH)	
1WU	17	593	5.93		
2AF	18	6	233		
2AX	19	118		(5% TBP)	
2AW	20	41		(WASTE)	
2AS	21	34			2.0
2AP	22	118	11.8		
2CX	23	59		(ORGANIC)	0.01
2CW	24	118			
2CU	25	59	23.6		
2WW	26	6		(NPH)	
2WS	27	6		(NPH)	
2WU	28	59	27.5		
U PRODUCT	29	6	233		

*VARIED

Fig. 6-1. Partition flowsheet

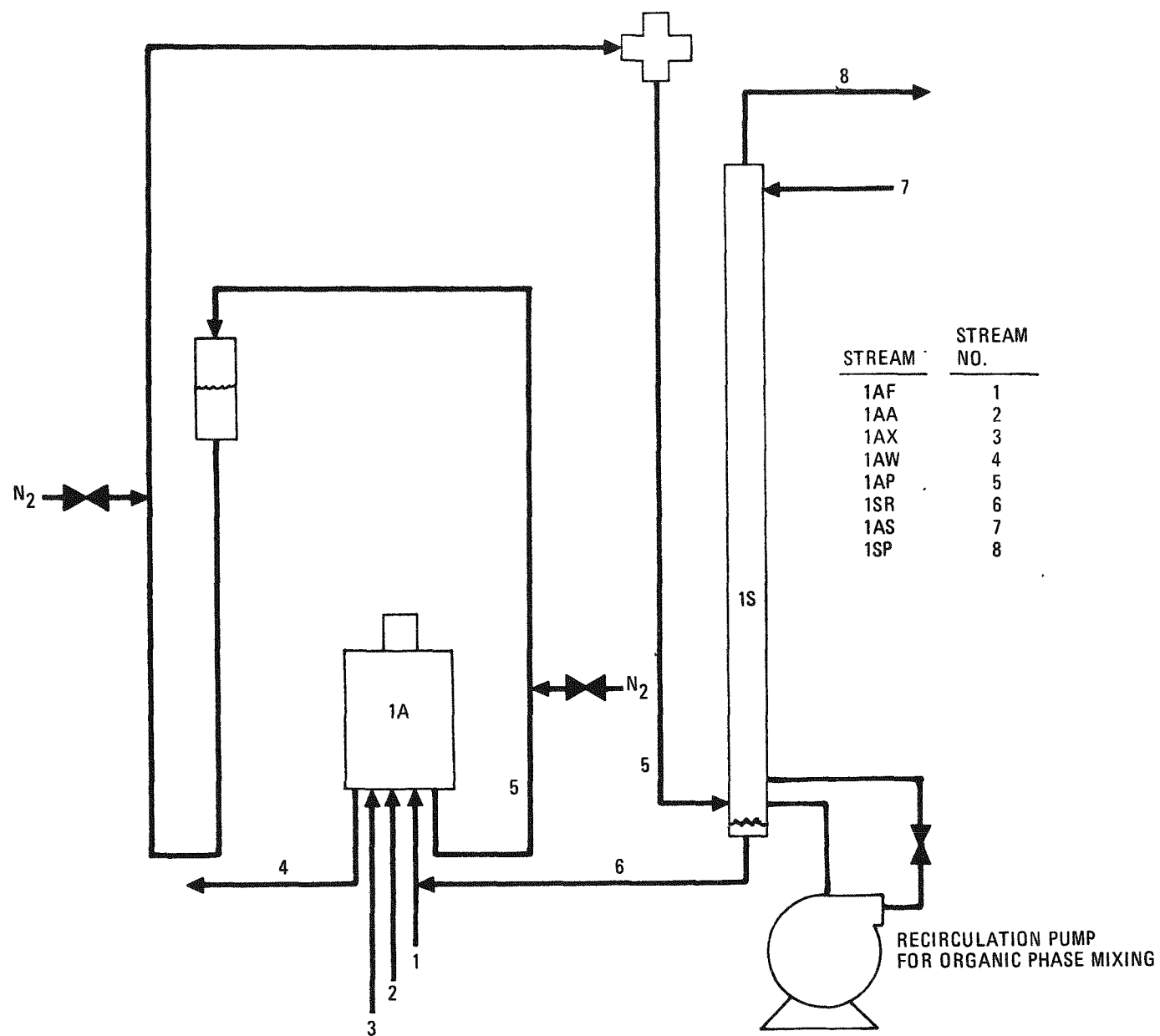


Fig. 6-2. 1A centrifugal contactor and 1S column with air lift transfer

TABLE 6-1

ZIRCONIUM DECONTAMINATION FACTOR AND FLOODING DATA FOR SOLVENT EXTRACTION RUNS 51, 52, AND 53^(a)

Run 51						
Column	Purpose	Vol Velocity (gal/hr/ft ²)	\bar{V}_a (b) (cm/sec)	\bar{V}_o (b) (cm/sec)	Flooding Freq (cycles/min)	Continuous Phase
1A	Extraction	871 (991) (917)	0.24 (0.23) (0.24)	0.74 (0.89) (0.80)	97 (87) (93)	Organic
1S	Scrub	781 (894) (829)	0.14 (0.12) (0.14)	0.74 (0.89) (0.80)	105 (95) (101)	Organic
1BX	Partition	585 (641) (595)	0.25 (0.25) (0.25)	0.41 (0.47) (0.42)	82 (81) (81)	Aqueous
1BS	Partition- Scrub	659 (659) (631)	0.57 (0.57) (0.56)	0.18 (0.18) (0.15)	82 (82) (85)	Aqueous
1C	U-Strip	553 (601) (570)	0.22 (0.21) (0.22)	0.41 (0.47) (0.42)	97 (94) (96)	Aqueous
Column	Aqueous to Organic Ratio	Zr-Nb DF		% Flooding Frequency	Temp.	
		U Basis	Th Basis			
1A	0.330 (0.264) (0.295)	13.2 (7.2) (13.4)	20.5 (24.7) (13.5)	82 (92) (86)	Ambient	
1S	0.191 (0.140) (0.170)	7.8 (3.5) (1.9)	4.2 (1.2) (1.6)	77 (85) (80)	Ambient	
1BX	0.617 (0.534) (0.582)	2.2 (3.8) (4.6)	- - -	60 (60) (60)	Ambient	
1BS	3.18 (3.21) (3.57)	- - -	0.72 (0.94) (1.15)	66 (66) (64)	Ambient	
1C	0.530 (0.437) (0.515)	1.37 (3.9) (2.6)	- - -	77 (80) (78)	52°C	

(a) The data in parentheses correspond to a second and third set of operating conditions with 0.25 g/liter DBP added to solvent at feed point.

(b) Where $\bar{V}_t = \bar{V}_a + \bar{V}_o$, volume velocity in gal/hr/ft² $\approx 885 \times \bar{V}_t$; \bar{V}_a is the superficial velocity of the aqueous phase; \bar{V}_o is the superficial velocity of the organic phase.

TABLE 6-1 (Continued)

Run 52

Column	Purpose	Vol Velocity (gal/hr/ft ²)	\bar{V}_a (b) (cm/sec)	\bar{V}_o (b) (cm/sec)	Flooding Freq (cycles/min)	Continuous Phase
1A	Extraction	936 (904) (996)	0.26 (0.26) (0.24)	0.80 (0.76) (0.89)	91 (94) (86)	Organic
1S	Scrub	833 (800) (881)	0.14 (0.14) (0.11)	0.80 (0.76) (0.89)	100 (103) (95)	Organic
1BX	Partition	618 (597) (663)	0.25 (0.25) (0.26)	0.45 (0.43) (0.49)	81 (81) (80)	Aqueous
1BS	Partition- Scrub	682 (674) (714)	0.56 (0.57) (0.60)	0.21 (0.19) (0.21)	72 (72) (71)	Aqueous
1C	U-Strip	591 (565) (635)	0.22 (0.21) (0.23)	0.47 (0.43) (0.49)	94 (96) (92)	Aqueous

Column	Aqueous to Organic Ratio	Zr-Nb DF		% Flooding Frequency	Temp.
		U Basis	Th Basis		
1A	0.322 (0.339) (0.271)	18.1 (8.2) (3.9)	23.7 (11.8) (5.0)	90 (87) (95)	Ambient
1S	0.177 (0.185) (0.123)	3.2 (2.3) (3.0)	2.1 (1.5) (2.7)	84 (82) (88)	Ambient
1BX	0.560 (0.599) (0.543)	6.4 (8.6) (19.4)	- - -	73 (73) (74)	Ambient
1BS	2.72 (3.00) (2.82)	- - -	0.83 (0.99) (1.06)	76 (76) (77)	Ambient
1C	0.493 (0.508) (0.473)	1.7 (2.3) (1.7)	- - -	73 (72) (75)	51°C

(a) The data in parentheses correspond to a second and third set of operating conditions with 0.25 g/liter DBP added to solvent at feed point.

(b) Where $\bar{V}_t = \bar{V}_a + \bar{V}_o$, volume velocity in gal/hr/ft² $\equiv 885 \times \bar{V}_t$; \bar{V}_a is the superficial velocity of the aqueous phase; \bar{V}_o is the superficial velocity of the organic phase.

TABLE 6-1 (Continued)

Run 53

Column	Purpose	Vol Velocity (gal/hr/ft ²)	\bar{V}_a (b) (cm/sec)	\bar{V}_o (b) (cm/sec)	Flooding Freq (cycles/min)	Continuous Phase
1A	Extraction	925 (861) (881)	0.243 (0.191) (0.177)	0.802 (0.782) (0.818)	92 (97) (96)	Organic
1S	Scrub	838 (769) (786)	0.144 (0.086) (0.087)	0.803 (0.783) (0.801)	100 (107) (105)	Organic
1BX	Partition	617 (607) (608)	0.258 (0.262) (0.273)	0.439 (0.424) (0.414)	82 (81) (81)	Aqueous
1BS	Partition- Scrub	680 (674) (658)	0.581 (0.591) (0.615)	0.187 (0.171) (0.129)	72 (72) (73)	Aqueous
1C	U-Strip	576 (564) (545)	0.212 (0.214) (0.203)	0.439 (0.423) (0.413)	95 (97) (97)	Aqueous

Column	Aqueous to Organic Ratio	Zr-Nb DF		% Flooding Frequency	Temp.
		U Basis	Th Basis		
1A	0.303 (0.244) (0.216)	11.6 (7.7) (3.0)	16.7 (13.1) (4.6)	86 (81) (82)	Ambient
1S	0.180 (0.110) (0.109)	10.0 (3.3) (4.4)	6.2 (1.8) (2.8)	83 (78) (79)	Ambient
1BX	0.587 (0.619) (0.661)	4.6 (12.3) (7.6)	- - -	71 (72) (72)	Ambient
1BS	3.10 (3.46) (4.76)	- - -	0.75 (0.82) (1.2)	74 (74) (73)	Ambient
1C	0.483 (0.505) (0.490)	1.1 (1.8) (3.0)	- - -	75 (73) (73)	47°C

(a) The data in parentheses correspond to a second and third set of operating conditions with 0.25 g/liter DBP added to solvent at feed point.

(b) Where $\bar{V}_t = \bar{V}_a + \bar{V}_o$, volume velocity in gal/hr/ft² $\equiv 885 \times \bar{V}_t$; \bar{V}_a is the superficial velocity of the aqueous phase; \bar{V}_o is the superficial velocity of the organic phase.

TABLE 6-2

CARTRIDGE DESCRIPTIONS FOR RUNS 51, 52, AND 53

Column	Column Diameter [in. (cm)]	Cartridge Height [ft (m)]	Plates			Plate Spacing [in. (cm)]
			Nozzle Direction	Hole Size [in. (mm)]	% Free Area	
1A Extraction	2 (5)	13 (3.9)	Down	1/8 (3)	23	2 (5)
1S Scrub	2 (5)	27 (8.2)	Down	1/8 (3)	23	2 (5)
1BX Partition	3 (7.6)	15 ^(a) (4.6)	Up	3/16 (5)	23	Graded ^(b)
1BS Partition- Scrub	2 (5)	17 (5.2)	Up	3/16 (5)	23	2 (5)
1C U-Strip	3 (7.6)	15 (4.6)	Up	3/16 (5)	23	Graded ^(b)

(a) 1BX column cartridge was 19 ft for runs 52 and 53.

(b) Graded cartridge is, from the bottom, 8-1/2 ft with 4-in. spacing, 1-1/2 ft with 3-in. spacing, and remainder with 2-in. spacing.

The objective of runs 51, 52, and 53 was to provide additional data on the effects of pilot plant feed, scrub section flow ratio, and DBP addition on zirconium-niobium decontamination in the first cycle of the Acid-Thorex solvent extraction process. The 1AS flow rate was varied to determine its effects. The feed for runs 51 and 52 was prepared from purchased thorium nitrate. About half of the thorium in the feed in run 53 was the product of pilot plant leaching and feed adjustment operations.

For each of the runs, two columns were used for the extraction-scrub operation (1A-1S) and one column was used for partition (1BX), partition-scrub (1BS), and uranium strip (1C) operations.

In these runs an exaggerated amount of DBP was added over that expected to occur in processing 180-day-cooled reference fertile particles through solvent extraction. From five to 10 times the normal concentration was used to accelerate observed effects and to change the decontamination factor significantly so as to better determine what the decontamination loss at expected HRDF conditions will be. The decontamination loss in runs 49 and 50 (Ref. 6-1) was small enough when adding the HRDF projected amount of DBP that the change could not be discerned from the normal experimental variations without DBP being added. The decontamination factors measured in all Zr-95 tracer runs through run 51 are plotted in Fig. 6-3. From this plot it can be seen that the scrub section flow ratio is a more important variable in Zr-95 separation from thorium than the DBP content of the scrub section organic phase. Also, from these data it was observed that the pilot plant feed does not appreciably lower Zr-95 decontamination through multi-run periods where the reproducibility is in the same range with either pilot plant feed or feed prepared from purchased thorium nitrate. To verify this observation, two additional runs are needed with pilot plant feed to obtain more data at the higher scrub section aqueous flow with DBP addition.

The radiation effects observed by G. L. Richardson (Ref. 6-2) on the Purex system show a greater loss in zirconium decontamination per unit of

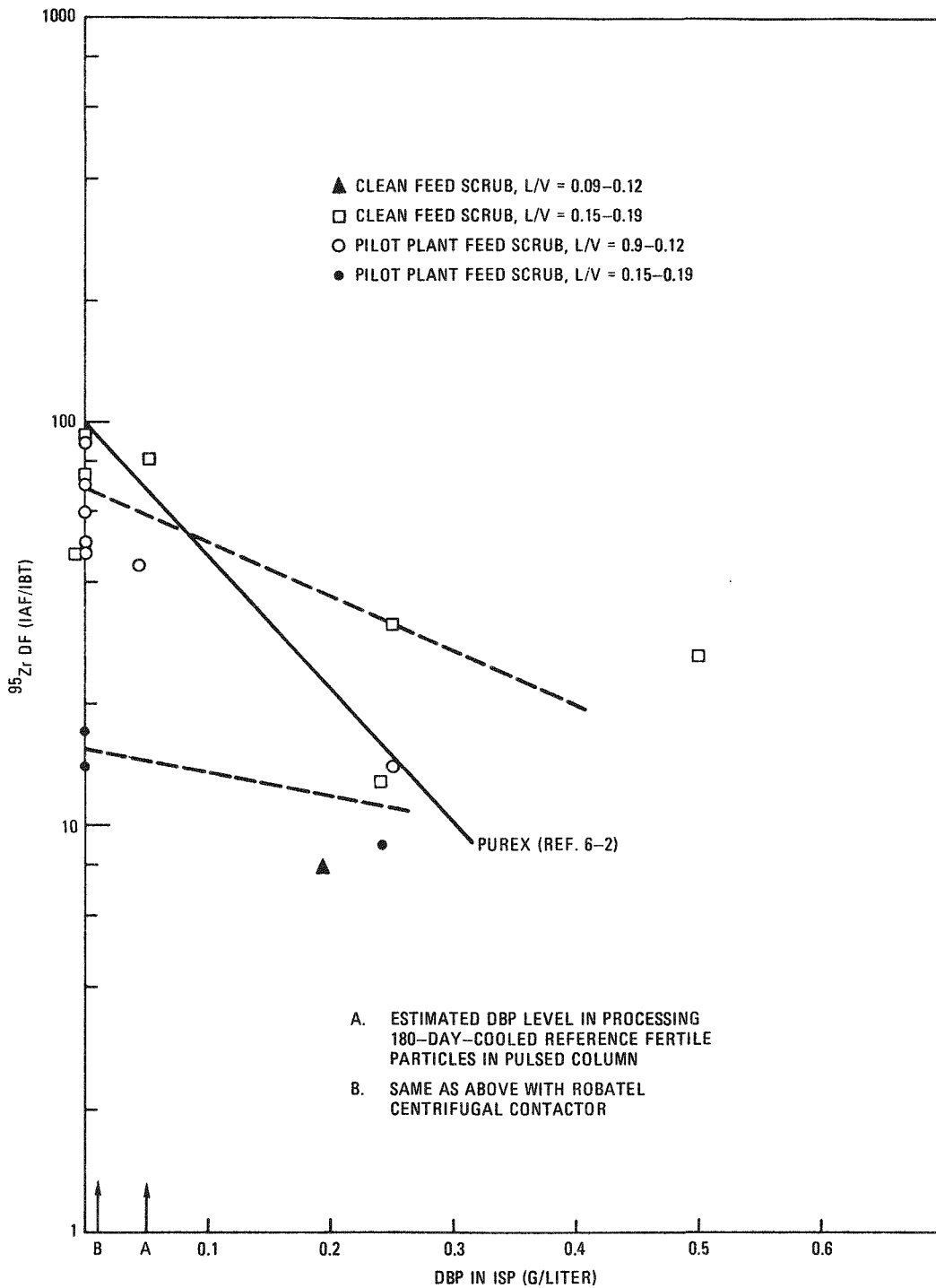


Fig. 6-3., Measured Zr-95 decontamination factors

radiation damage (watt-hr/liter or g/liter DBP) than does the Thorex system (Fig. 6-3, solid line). The reason for this difference is that thorium (IV) is a better reagent for DBP complex or compound formation than uranyl ion; thus, less DBP is available for raising the equilibrium distribution coefficient of zirconium in the Thorex system.

6.4. RESULTS AND DISCUSSION - RUNS 54 AND 55

The objective of runs 54 and 55 was to demonstrate coupling of the centrifugal contactor with the pulse column system and to study the feasibility of its use as the first-cycle extraction section in the Acid-Thorex flowsheet.

Runs 54 and 55 consisted of extraction, scrub, partition, partition-scrub, uranium strip, and solvent wash operations using the Robatel centrifugal contactor and the five-column solvent extraction system in the pilot plant. The flowsheet tested was the first cycle of the Thorex co-extraction - partition flowsheet. These were the first runs which utilized both the centrifugal contactor and the pulse columns. The centrifugal contactor was used as the 1A extraction section. No Zr-95 or DBP was added to the feed streams during either run.

Table 6-3 contains the stream analyses and flow rates for each run, and Table 6-4 contains loss data and operating conditions for each run. Table 6-5 contains descriptions of the contactor, columns, and column cartridges.

Control problems developed in both runs. At relatively high thorium loadings, the organic used splits into a dense, thorium-rich phase and a lighter, thorium-poor phase. No operational difficulties are encountered when the formation of the two organic phases occurs only in the pulse column plate section. In run 54 the control of the 1S column interface was difficult owing to the presence of the dense second organic phase. Some of this dense organic phase was returned to the 1A contactor with the 1SR aqueous stream.

TABLE 6-3A

SOLVENT EXTRACTION STREAM ANALYSES AND FLOW RATES FOR RUN 54

Stream	U (g/l)	Th (g/l)	HNO ₃ (M)	Flow (ml/min)	Relative Flow
1AF	30.78	333.0	0.257	98	100
1AS			1.01	177	181
1BX			0.177	683	697
1CX			0.011	573	585
1BS			[30%TBP]	210	214
1AW	4×10^{-3}	1.16×10^{-2}	1.89		
1AP	5.21	40.2	0.157		
1SR	0.62	63.5	1.49		
1SP	3.95	14.1	0.111 ^(a)		
1BX	0.65	37.7	0.435		
1BU	2.53	6.5×10^{-3}	0.052		
1BT	2.8×10^{-3}	26.9	0.355		
1BSU	4.42	11.7	0.094		
1CU	6.29	6.0×10^{-3}	0.086		
1CW	0.9×10^{-3}	1.8×10^{-3}	0.007		
1AA			~13.0	~25	25.5
1OS				116	118
1OW	3.0×10^{-3}	0.9×10^{-3}	-		
100	-	-	-	-	-
1AX			[30%TBP]	878	896

(a) Poor material balance caused by thorium losses to drain of centrifugal contactor due to airlift problem. Drain stream was not analyzed.

TABLE 6-3B

SOLVENT EXTRACTION STREAM ANALYSES AND FLOW RATES FOR RUN 55^(a)

Stream	U (g/l)	Th (g/l)	HNO ₃ (M)	Flow (ml/min)	Relative Flow
1AF	30.98	359.0	0.558	87	100
1AS			1.12	174.5	200.6
1AA			~13.0	29	33.3
1BX			0.210	710	816
1CX			0.041	597	686
1BS			[30%TBP]	174	200
1OS				122	140
1AW	1.25×10^{-2}	4.9×10^{-2}	1.73		
1AP	2.58	46.3	0.179		
1SR	0.49	91.6	1.08		
1SP	3.78	15.3	0.105 ^(b)		
1BXT	1.93	18.5	0.343		
1BU	0.56 (3.13)	1.6×10^{-3} (2.3×10^{-3})	0.035 (0.041)		
1BT	1.53×10^{-2}	15.0	0.327		
1BSU	5.56	2.3	0.058		
1CU	0.47 (2.08)	1.4×10^{-3} (4.5×10^{-3})	0.043 (0.038)		
1CW	0.8×10^{-3}	1.9×10^{-3}	0.014		
1OW	1.8×10^{-3}	1.6×10^{-3}	-		
1OO	1.0×10^{-3}	2.1×10^{-3}			
1AX			[30%TBP]	940	1080

(a) The data in parentheses correspond to a second set of operating conditions.

(b) Poor material balance caused by thorium losses to drain of centrifugal contactor due to airlift problem. Drain stream was not analyzed.

TABLE 6-4A

LOSS DATA AND OPERATING CONDITIONS FOR RUN 54

Contactor	Purpose	Vol Velocity (gal/hr/ft ²)	\bar{V}_a (cm/sec)	\bar{V}_o (cm/sec)	Flooding Freq (cpm)	Continuous Phase	Aqueous to Organic Ratio	Percent Loss		% Flooding Frequency	Temp.
								U	Th		
1A Centri- fugal	Extrac- tion				(<500 RPM)		0.342	0.04	0.01	(1850 RPM)	
1S Pulse Column	Scrub	767	0.146	0.721	105	Organic	0.202	-	-	76	Ambient
1BX Pulse Column	Parti- tion	572	0.249	0.397	84	Aqueous	0.628	-	0.02	67	Ambient
1BS Pulse Column	Parti- tion- Scrub	649	0.561	0.173	88	Aqueous	3.25	0.06	-	57	Ambient
1C Pulse Column	U-Strip	537	0.209	0.397	98	Aqueous	0.527	0.03	-	67	
1O Pulse Column	Solvent Wash	875	0.097	0.892	121	Organic	0.107	-	-	63	

TABLE 6-4B
LOSS DATA AND OPERATING CONDITIONS FOR RUN 55

Contactor	Purpose	Vol Velocity (gal/hr/ft ²)	\bar{V}_a (cm/sec)	\bar{V}_o (cm/sec)	Flooding Freq (cpm)	Continuous Phase	Aqueous to Organic Ratio	Percent Loss		% Flooding Frequency	Temp.
								U	Th		
1A Centri- fugal	Extrac- tion				(< 500 RPM)	0.309		0.13	0.05	(2070 RPM)	Ambient
1S Pulse Column	Scrub	810	0.144	0.772	101	Organic	0.186	-	-	80	Ambient
1BX Pulse Column	Parti- tion	589	0.259	0.407	83	Aqueous	0.637	-	0.006 (0.009)	80	Ambient
1BS Pulse Column	Parti- tion- Scrub	643	0.584	0.143	74	Aqueous	4.08	0.40	-	62	Ambient
1C Pulse Column	U-Strip	553	0.218	0.407	97	Aqueous	0.536	0.03	-	71	46°C
1O Pulse Column	Solvent Wash	899	0.101	0.915	120	Organic	0.110	-	-	65	~45°C

TABLE 6-5

CONTACTOR, COLUMN, AND CARTRIDGE DESCRIPTIONS FOR RUNS 54 AND 55

1A Contactor Extraction	180 mm Diameter	320 mm Height	8 Stages With 0.4-Liter Holdup Per Stage			
Column	Column Diameter (mm)	Cartridge Height (m)	Plates			Plate Spacing (mm)
			Nozzle Direction	Hole Size (mm)	% Free Area	
1S Scrub	51	6.71	Down	3.2	23	51
1BX	76	5.79	Up	3.2	23	Graded ^(a)
1BS	51	5.18	Up	4.8	23	51
1C	76	4.57	Up	4.8	23	Graded ^(a)
10 Solvent Wash	51	5.49	Down	3.2	23	51

(a) Graded cartridge is, from the bottom, 2.6 m with 100-mm spacing, 0.46 m with 76-mm spacing, and remainder with 51-mm spacing.

The airlift system on the 1AP stream between the 1A contactor and 1S column was operated at near its capacity limit. Significant losses occurred via the contactor overflow drain owing to the inherent fluctuations of a solvent extraction system. Under normal operating conditions no liquid leaves the contactor via the drain.

The liquid loss from the contactor was much more severe in run 55 than in run 54. This was due to the inability of the 1AP airlift to handle the flow. The 1S column interface was difficult to control. In run 55 the 1S column flooded. The flooding occurred owing to the instability caused by the second organic phase and poor interface control.

The interface and flow control difficulties were resolved in later runs and will be discussed in future reports.

Except for the problems associated with the second organic phase in the 1S column and the drainage losses from the 1A contactor, the operation of the solvent extraction system was satisfactory.

In run 55 the 1BXT control valve failed. However, it was replaced without affecting the operation.

In run 55 the contactor was run at several selected rpm's in an attempt to alleviate losses via the drain. These changes in rotating speed caused no noticeable change in the operation.

In run 55 a mixer in the disengaging section above the 1S column interface was used to reduce the third phase effects (Fig. 6-3). Very little benefit was derived from its operation. The mixer may have contributed to the flooding which occurred in the 1S column.

The flooding which occurred in the lower half of the 1S column may have been caused not only by the erratic behavior of the interface control,

but also by the presence of two organic phases and some gas bubbles. The gas (N_2) from the gas-liquid lifters was entrained in the 1AP stream added to the bottom of the column.

The feed (1AF) and 1SR combined stream was filtered prior to the addition to the centrifugal contactor. Filtration was used to remove particles from the stream since the centrifugal contactor is susceptible to plugging in the channels between stages. In a full-sized plant, a centrifuge would be used for feed clarification.

In run 55 the low thorium concentration in the 1BT stream and the low uranium concentration in the 1CU stream were a result of the losses from the centrifugal contactor via the overflow drain. These losses occurred owing to insufficient capacity in the 1AP airlift system, which caused backup of the 1AP stream into the contactor and overflow of the stream to the drain. Only small losses occurred via the 1AW stream.

No HETS (height equivalent to a theoretical stage) calculations are included for these runs because they never reached steady state owing to the operating difficulties cited above.

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7. OFF-GAS STUDIES

7.1. INTRODUCTION

A study was completed with the objectives of reviewing the development status of the off-gas treatment technology with regard to the needs for the HTGR Recycle Demonstration Facility (HRDF) design, reviewing the off-gas progress to date, and identifying components requiring engineering-scale demonstration. After a detailed review of the proposed process flowsheets and material flow schedules, several necessary changes in the head-end off-gas treatment scheme became apparent. Some additional laboratory and hot cell development needs were also identified.

As a design basis for the study, the preconceptual design (Ref. 7-1) for the HRDF prepared by General Atomic was used to estimate throughput requirement. The fuel composition, fuel particle and coating contamination levels, and quantities and distribution of activation and fission products are based on GARGOYLE code calculations (Ref. 7-2). The fuel particle basis is BISO-coated ThO_2 fertile particles and TRISO-coated UC_{40} fissile particles.

7.2. ACTIVATION AND FISSION PRODUCT DISTRIBUTION

The off-gas flow from each of two 60-cm (24 in.) diameter HRDF primary burners will be about $4.53 \text{ m}^3/\text{min}$ for a total of about $9.06 \text{ m}^3/\text{min}$ ($320 \text{ ft}^3/\text{min}$), and the burners will operate essentially continuously at a combined rate of 43 FE/day (FE = fuel elements). The single secondary burner will be a 20-cm (8 in.) diameter burner and will perform two 6-hr burns per day to process the fissile particles. The secondary burner will have an off-gas flow of about $0.42 \text{ m}^3/\text{min}$ ($15 \text{ ft}^3/\text{min}$) for about 12 hr per operating day. The fertile dissolver will receive a charge of about 420 kg

of fertile particles per day and will be operated on a 24-hr batch basis with an average purge and sweep gas flow rate on the order of $0.28 \text{ m}^3/\text{min}$ ($10 \text{ ft}^3/\text{min}$). The fissile dissolver is designed for 50 kg of burned fissile particles and will be operated on two 6-hr cycles each day with an average purge and sparge off-gas flow of $0.084 \text{ m}^3/\text{min}$ ($3 \text{ ft}^3/\text{min}$) during normal operation. A detailed description of the process steps is given in Ref. 7-1.

The estimated fractional release of activation and fission products to the burner and dissolver off-gases during reprocessing operations is based on reported observations by German researchers (Ref. 7-3) and discussions with ORNL personnel (Refs. 7-4, 7-5). Fractional release values are given in Table 7-1. For other fuel, such as that from the Fort St. Vrain HTGR, the fractional releases would differ.

The distribution of H-3, I_2 , and krypton in the various off-gas streams was estimated from the quantities of activation and fission products in the fertile and fissile particles calculated by the GARGOYLE code (Ref. 7-2) and the fractional releases given in Table 7-1. It was also assumed that 4% of the fissile particles would be broken either during irradiation or during the crushing and burning steps and that the activation and fission products of interest were all released from these broken particles to the primary burner off-gas. A summary of the estimated off-gas distribution of the various species in the off-gas streams is shown in Table 7-2, and the approximate concentrations are given in Table 7-3.

From the results of Tables 7-2 and 7-3, several important observations can be made: (1) the primary burner off-gas represents nearly 95% of the total off-gas flow, but less than 10% of the total indicated activation and fission products are released to this stream; and (2) the concentrations of the indicated activation and fission products are two to three orders of magnitude greater in the secondary burner and fertile particle dissolver off-gases than in the primary burner off-gas. The values given are daily averages.

TABLE 7-1
FRACTIONAL RELEASE OF CERTAIN ACTIVATION AND FISSION
PRODUCTS DURING REPROCESSING OPERATIONS

Operation	Quantities Released
Primary burning	20% H-3; 2% I ₂ ; and 10% Kr
Secondary burning	100% H-3, I ₂ , and Kr
Fertile particle dissolution	100% I ₂ and Kr; H-3 stays in solution
Fissile particle dissolution	Nil (only trace quantities of H-3, I ₂ , and Kr present)

TABLE 7-2
DISTRIBUTION OF CERTAIN ACTIVATION AND FISSION
PRODUCTS IN OFF-GAS STREAMS

Off-Gas	Percent of Total Off-Gas Flow	Percent		
		H-3	I ₂	Kr
Primary burner	94.4	13	3	7
Secondary burner	2.2	44	44	53
Fertile dissolver	3.0	-	53	40
Fissile dissolver	0.4	-	-	-

TABLE 7-3
CONCENTRATIONS OF CERTAIN ACTIVATION AND FISSION
PRODUCTS IN OFF-GAS STREAMS

	ppm		
	H-3	I ₂	Kr
Primary burner	0.005	0.07	1.1
Secondary burner	0.73	42	384
Fertile dissolver	--	38	220

The secondary burner and fertile dissolver are batch operated, and their off-gas compositions will vary within a batch cycle. There will be portions of their operating cycles when the concentrations of the species indicated in Table 7-3 will be considerably higher than the averages. Because the primary burner will be operated semicontinuously, the H-3, I₂, and krypton concentrations in its off-gas will be relatively uniform.

7.3. OPERATIONAL CONSIDERATIONS

In view of the estimated distributions and concentrations of H-3, I₂, and krypton shown in Tables 7-2 and 7-3, there is clearly some merit in providing separate off-gas treatment trains for each of the burner types and one train for the dissolver off-gas. The concentration of krypton in the primary burner off-gas is nearly that expected in the combined burner off-gas after treatment for krypton removal. On this basis, combining the secondary burner off-gas with the primary burner off-gas before the primary burner off-gas krypton removal step should be reconsidered.

Most of the components in the off-gas treatment train involve the use of solid adsorbent packed beds. For these components to perform properly, the superficial face velocity must be in the turbulent flow region. The minimum superficial face velocity for the particle size range normally used in such packed beds is about 30 ft/min. Because the secondary burner must be operated after the primary burner is shut down, for example during cleanout, over 8.5 m³/min (300 ft³/min) of CO₂ would have to be provided for recycle if a common off-gas train were used. The period of recycle would be about 6 hr on a 20- to 25-day cycle. The situation is similar for the dissolvers. These components will be operated up to 24 hr after the primary burner is shut down for cleanout or maintenance. Therefore, a similar quantity of CO₂ would have to be recycled to provide the minimum packed bed flow for this entire time.

7.4. PROPOSED OFF-GAS CONFIGURATION

The sources of the burner and dissolver off-gases are shown in Fig. 7-1, and the proposed off-gas treatment configuration for these sources is shown in Fig. 7-2. A discussion of each off-gas train, including the rationale for component selection and order, is presented below.

7.4.1. Primary Burner

7.4.1.1. Semivolatiles Removal

In addition to the radioactive contaminant gases H-3, Kr-85, and I-129 shown in Table 7-2, the primary burner off-gas will contain semivolatile activation and fission products, CO, O₂, SO₂, Rn-220, and C-14 contaminated CO₂. All of these species can represent an adverse situation through either direct emissions or interference in one or more of the treatment processes.

The rate of emission of the semivolatiles into the primary burner off-gas will depend on the fluidizing velocity, bed temperature, method of handling fines recycle, and quantity of broken fissile particles. The lack of clear understanding of the effect of these variables makes accurate estimates of how much of the semivolatiles will be liberated and in what forms impossible at this time. The location where the semivolatiles plate out is controlled primarily by temperature and available surface area. The results of studies by Allied Chemical at INEL (Ref. 7-6) and by ORNL (Ref. 7-7) show that most of the semivolatiles penetrating the sintered metal filters plate out immediately downstream of the filters in the off-gas lines. Therefore, a removable large-surface-area heat exchanger close-coupled to the sintered metal filters that will cool the off-gas down to around 100°C would appear to be a desirable design. Such a device should remove essentially all the semivolatiles. However, the need for additional particulate removal to protect and/or replace HEPA filters at this point in the off-gas train remains to be determined in hot cell tests.

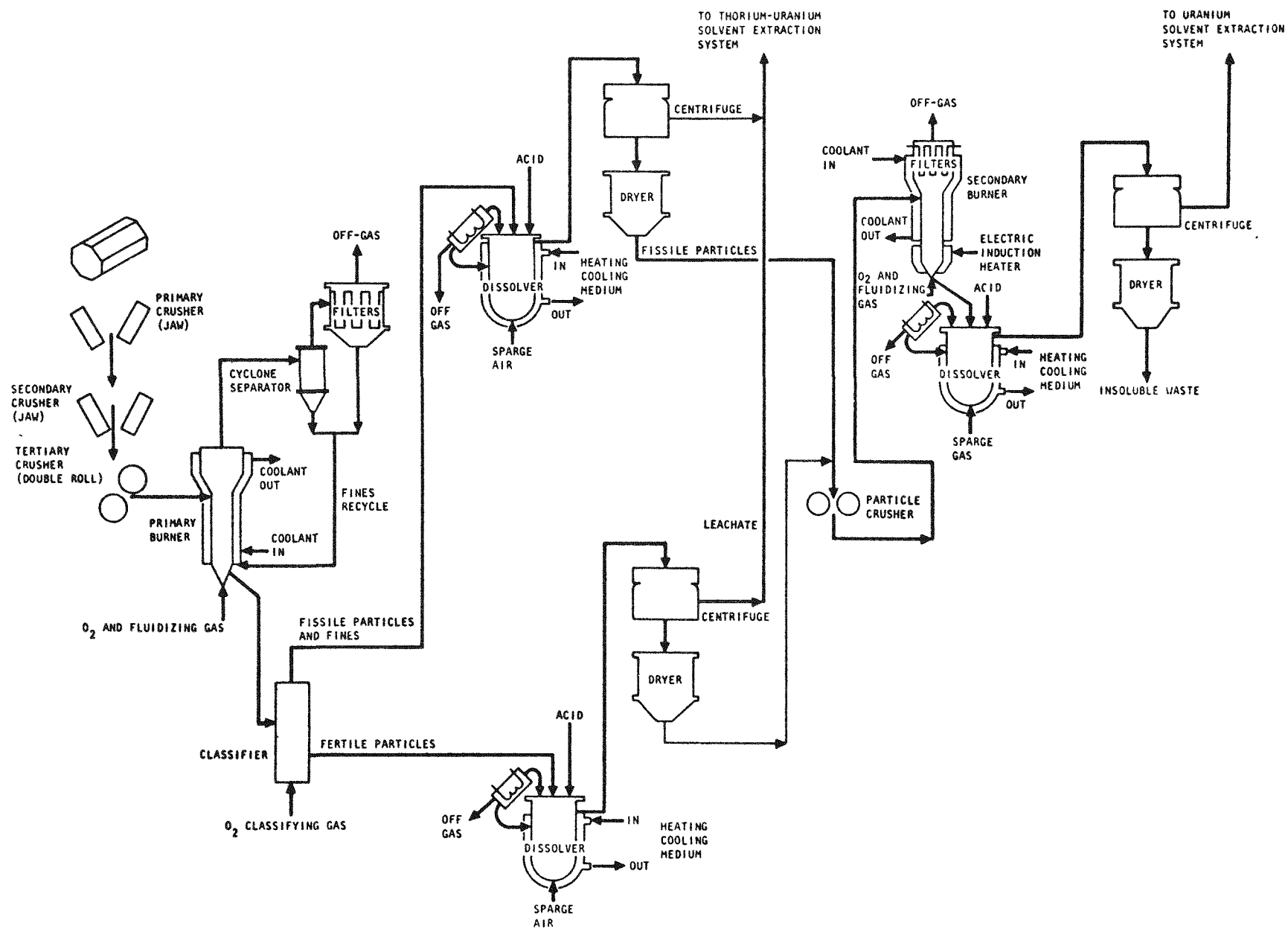


Fig. 7-1. Simplified TRISO-BISO reprocessing flow diagram

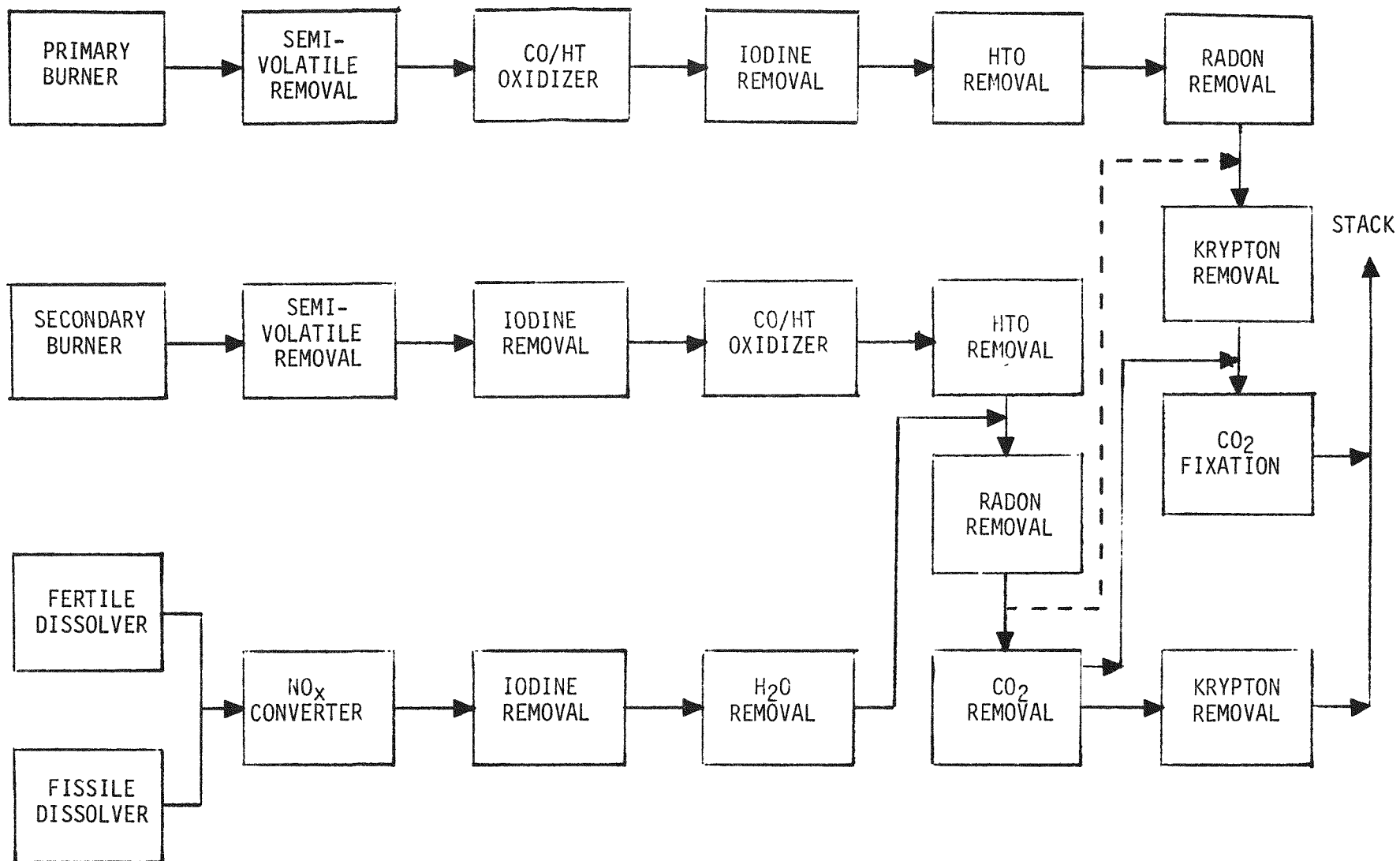


Fig. 7-2. Proposed off-gas treatment scheme

7.4.1.2. CO Oxidizer

The concentration of CO in the primary burner off-gas varies somewhat and is estimated to reach concentrations as high as 30% (Ref. 7-8). This quantity of CO could represent an explosion hazard, interfere with the proper performance of other off-gas treatment components (I_2 adsorption, radon holdup, and krypton recovery), and result in an appreciable loss in C-14 recovery if it is not treated. Several techniques have been considered for the conversion of CO to CO_2 : (1) the use of an afterburner, (2) a fluidized bed converter, and (3) several types of catalytic converters.

Catalytic converters using noble metal catalysts perform efficiently at about 175°C, but temperatures above about 400°C tend to destroy the catalyst. The CO oxidation reaction is quite exothermic, and the temperature in the catalytic oxidizer increases about 85° to 90°C for every 1% increase in CO content (Ref. 7-9). Therefore, internal recycle of the oxidizer exhaust is necessary to limit the incoming gas stream CO concentration to about 1% CO so that the catalyst bed does not reach damaging temperatures.

Because a dilution of 20- to 30-fold would be necessary for a noble metal catalyst for a complete, one-step conversion of the CO in the HRDF design, such a design does not appear to be the best option. A design incorporating several stages using either internal recycle or interstage heat exchangers is the most appealing from a control point of view. The stage efficiency, and hence the heat removal requirements, can be controlled by the amount of O_2 injected. Also, ceramic monolith catalyst support devices appear to be more suitable than the packed bed type. They offer less pressure drop but are not as efficient in the very low (less than a few hundred ppm) concentration range, which is fortunately not a requirement.

The precise level of CO removal required is not well defined. The limiting requirements for CO conversion will probably be dictated by the recovery requirements for C-14, since the proposed C-14 control techniques are based on CO₂ fixation and are ineffective for CO. Tolerance of the krypton removal system (KALC) to CO is not clear, but it will probably be higher than that required for C-14 recovery. The development and demonstration of appropriate process control loops to maintain the CO and O₂ concentrations within concentration ranges acceptable to the KALC is needed.

Several of the chemical contaminants in the primary burner off-gas stream could interfere with the proper performance of the CO oxidizer, namely I₂ and SO₂. The concentration of iodine in the primary burner stream will probably be sufficiently low, about 0.1 ppm, that its effect will be negligible. However, SO₂ represents a potential problem. Assuming a 2% sulfur content in the fuel rod pitch, the concentration in the primary burner is estimated to be on the order of 10 to 20 ppm. SO₂ concentrations this high are expected to noticeably reduce the efficiency of the CO oxidizing catalyst. Tests need to be performed to evaluate the tolerance of monolithic noble metal catalyst materials and to evaluate the feasibility of SO₂ removal upstream of the CO oxidizer.

At least preliminary testing of a CO oxidizer needs to be completed and the results analyzed before a choice of converter type can be made.

7.4.1.3. Iodine Removal

The original concept for iodine removal was to use a lead zeolite adsorber bed to remove the bulk of the airborne iodine followed by the more expensive and reactive silver zeolite bed to ensure collection of all the iodine. However, the performance of lead zeolite has proved disappointing in subsequent tests, indicating that silver zeolite or another type of silver-impregnated material may be more suitable for both beds.

On the other hand, the concentration of iodine in the primary burner off-gas is expected to be sufficiently low that only about 10 g of I_2 is expected for each day of HRDF operation. The size of adsorbent bed necessary to give adequate residence time would have enough iodine adsorbent capacity to last through several years of operation even if lead zeolite were used. Because the cost of the lead zeolite is estimated to be about eight times less than the silver zeolite cost for large quantities, the use of lead zeolite may be justified even though the static loading capacity of lead zeolite appears to be about 15 times less than that of silver zeolite (Ref. 7-10).

Because of the high cost of the iodine adsorbents, considerable thought should be given to the design of the iodine adsorbent beds. The best design for the iodine adsorbent beds is not completely resolved. Ideally, the best bed depth and configuration are those that will provide the most efficient utilization of the adsorbent (greatest loading capacity) while providing an adequate DF. The efficiency of silver zeolite beds tends to decrease during long exposures to atmospheric and industrial contaminants (Refs. 7-11, 7-12). It has also been shown that, when the loading capacities of 5, 10, and 15 cm (2, 4, and 6 in.) silver zeolite adsorption beds were measured at varying face velocities of test gas, the loading capacities increased per gram of adsorbent when the bed depth was increased from 5 to 10 cm but remained about the same when it was increased from 10 to 15 cm (Ref. 7-13). The concentration of iodine in the test gas was quite high in these tests, about 500 mg/m^3 ; and although the overall loading capacities would probably be slightly greater at lower concentration (Ref. 7-14), the trend would probably be about the same.

Some loss of iodine removal efficiency with increasing bed depth has been noted under certain conditions (Ref. 7-14), and was presumably caused by channeling. For obtaining the best utilization of the iodine adsorbent, the best bed depth appears to be 10 to 15 cm using 12- to 16-mesh adsorbent material. The diameter of the bed is dictated by the face velocity. The

superficial velocity must be in the turbulent flow regions, or premature breakthrough will occur; but if the flow is too high, appreciable loss of DF and adsorbent utilization will occur. A superficial velocity in the range of 12 to 18 m/min (40 to 60 ft/min) appears to yield the best DFs and adsorbent utilization.

As with most packed beds, the best operation of the bed to minimize the possibility of voids, bypass, and fluidization is a down-flow position. However, this arrangement does not lend itself to the best position for remote bed changeout. A horizontal bed would appear to be the best choice for bed changeout, but care must be taken in the design to ensure that adsorbent bypass cannot occur.

7.4.1.4. Tritium Removal

Considerable technology exists in the design of molecular sieve moisture removal beds. Between the primary burner and CO/HT oxidizer, essentially all the tritium should be in the form of HTO. The concentration of tritium is estimated to be less than 0.01 ppm in the primary burner off-gas. Because the water adsorbents lose efficiency rapidly below about 1 ppm, steam will have to be added to bring the moisture content to about 1000 ppm before moisture removal. This should provide a tritium DF of about 1000. Two tritium beds will be operated in parallel so that one can be regenerated while the other is in service. These beds are totally regenerable and should not require replacement for at least several years of operation.

There does not appear to be any possible contaminants that could interfere with the tritium removal adsorbents. Because the pore diameters of the Type-3A molecular sieve adsorbent are only large enough to allow adsorption of H_2O and too small for CO_2 and SO_2 , these contaminants will not interfere.

7.4.1.5. Radon Holdup

The use of synthetic mordenite molecular sieves to hold up radon long enough for the radon to decay to nongaseous products has been demonstrated at INEL (ACC). Apparently, the holdup of radon is somewhat adversely affected by organic impurities and moisture but not by CO_2 , SO_2 , or noble gases at the concentrations expected in the primary burner off-gas. Also, the radon daughter products evidently remain on the mordenite bed.

A report describing the radon holdup studies is being prepared for publication (Ref. 7-14) and will include the information necessary to design a larger system.

The fact that molecular sieve adsorption beds tend to adsorb many airborne contaminants that may be present in the off-gas makes the eventual poisoning of the bed for radon holdup probable. Therefore, two parallel beds should be included in the design such that one can be regenerated while the other is in service.

Because of the adverse effect of water on radon holdup, the radon bed should clearly follow the HTO removal bed.

7.4.1.6. Krypton Removal

The description and development progress of the krypton removal system (KALC process) are well documented (Refs. 7-15, 7-16). A clearer definition of the tolerance of the KALC process to CO , I_2 , water vapor, and SO_2 is needed before the final design of the primary burner off-gas treatment can be completed. Because of the apparent sensitivity of the radon holdup bed to contaminants, it is unlikely that the KALC process would be exposed to very high levels of contaminants. Nevertheless, the demonstrated high tolerance of the KALC process to these contaminants would offer more options in the design off-gas treatment train.

7.4.2. Secondary Burner

The off-gas flow from the secondary burner will be considerably less than that from the primary burner. However, as shown in Table 7-3, it will contain much higher concentrations of radioactive contaminants.

7.4.2.1. Semivolatiles Removal

Because the oxycarbide fuel is reduced to a powder as it burns, a much greater surface area of fissile fuel than ThO_2 fertile fuel will become exposed during burner operation, and a larger percentage of semivolatiles will become airborne. Most of the general remarks made earlier concerning the semivolatiles in the primary burner apply to the secondary burner, but containment of the semivolatiles will be much more severe in the secondary burner. In tests conducted at ORNL (Ref. 7-5), 26% of the ruthenium and 6% of the cesium were collected in the off-gas when carbide fuel was burned under quiescent conditions. This high concentration of radioactivity will impose significant operating constraints in the HRDF. Better definition of the expected semivolatiles behavior is clearly needed. Alternate secondary burner operating modes may be required to reduce the quantity of semivolatiles generated.

The development of techniques to control, collect, and treat semivolatiles for transport and storage is a major element in the HTGR reprocessing development program. Because of the high cost of performing large-scale hot cell tests and the lack of representative high-burnup fuel available at the scheduled time of operation, most of the semivolatiles control development will have to be done with unirradiated fuel and simulated fission products. Close coordination with hot cell tests using irradiated fuel in which the actual burner conditions are simulated as closely as possible will be necessary to ensure a reliable design.

7.4.2.2. Iodine Removal

The iodine concentration in the secondary burner off-gas is expected to be several hundred times greater than in the primary burner. The CO/HT oxidation catalyst is adversely affected by iodine concentrations greater than about 1 ppm, so the iodine removal system must precede the CO/HT oxidizer. The disadvantage of this arrangement is that the off-gas from the semivolatiles removal system will need to be cooled to about 150°C for the CO/HT oxidizer. Because the quantity of secondary burner off-gas is relatively small, its cooling and reheating are easily accomplished.

At the expected airborne iodine concentration, the estimated life of a 20-cm-diameter by 15-cm-deep (8 in. by 6 in.) silver zeolite adsorption bed will be only about 4 days of HRDF operations. The required frequent changeout of the silver zeolite adsorption bed suggests that careful design of this system is important.

7.4.2.3. CO/HT Oxidizer

As with the primary burner, essentially all the tritium is expected to be converted to HTO before it leaves the secondary burner. The maximum CO concentration in the secondary burner is expected to vary considerably, peaking at about 25% (Ref. 7-17). The use of a multistage CO oxidizer as described earlier will probably be desirable in the secondary off-gas train also, even though the total flow is much less. The highly variable CO concentration requires the development of adequate control systems to maintain the appropriate CO content in the effluent stream.

7.4.2.4. Tritium and Radon Removal

The tritium removal and radon holdup systems for the secondary burner off-gas train will be virtually the same as described for the primary burner, except they will be of smaller dimensions.

7.4.2.5. Krypton Removal

The estimated krypton concentration in the secondary burner off-gas is several hundred times greater than that in the primary burner off-gas. It would not be prudent to dilute the secondary burner off-gas with the primary burner off-gas prior to krypton removal.

The krypton could be removed from the secondary burner off-gas using developed cryogenic technology, but this would require prior removal of CO_2 . However, the removal of CO_2 is not a major endeavor. The CO_2 from a 6-hr run in the secondary burner could be contained on a 1-m-diameter by 2-m-long (3 ft by 6 ft) zeolite adsorption bed. Air could be used as the carrier gas to sweep the noble gases through the CO_2 adsorber bed. The noble gases could be separated cryogenically from the small flow of gas, several tenths of $1 \text{ m}^3/\text{min}$ (several ft^3/min). The CO_2 adsorbent bed would be regenerated and the desorbed CO_2 sent to the CO_2 fixation unit.

If the small flow from the secondary burner off-gas treatment system could be efficiently processed through the KALC process without being diluted with the primary burner off-gas, the arrangement shown in Fig. 7-2 would be the most desirable. The practicality of this approach should be pursued. The costs of operating a cryogenic separation unit and associated components for krypton removal from the secondary burner off-gas should be compared with corresponding costs for the KALC processing equipment for an equivalent separation.

7.4.3. Dissolvers

The only unique feature of the dissolver off-gas treatment system is the NO_x converter. Although not shown in Fig. 7-2, it is assumed that some sort of combined condenser-adsorption tower is used to remove a large fraction of NO_2 from the off-gas prior to processing through the NO_x converter. The NO_x converter reduces the NO_x to nitrogen and water by the zeolite-catalyzed reaction of NO_x with ammonia. The catalytic bed must be maintained at a temperature of about 400°C to ensure complete reaction. The

system must incorporate the necessary monitors and control loops to maintain low levels of residual NO_x or NH_3 .

Very little, if any, of the iodine from the dissolvers is expected to be retained in the zeolite NO_x catalyst. The iodine will be collected on a silver zeolite adsorbent maintained at a temperature in the range of 100° to 150°C . The adsorbent bed design and change-out features will be similar to those in the burner off-gas streams.

The moisture present in the dissolver off-gas must be removed to protect the radon removal beds. Zeolite adsorption beds will be used for this purpose.

7.5. CARBON DIOXIDE FIXATION

Carbon dioxide fixation techniques have been reviewed and reported elsewhere (Ref. 7-18). Because CO_2 fixation schemes are being studied by ORNL for engineering-scale demonstration and because this unit can operate independently of the rest of the off-gas treatment system, no consideration was given to the CO_2 fixation portion of the system in this study.

7.6. CONCLUSIONS

Based on the expected distribution of iodine and krypton during the primary burning operation and the assumed fraction of broken fissile particles (4%), several changes should be made in the proposed (Ref. 7-1) reprocessing plant off-gas treatment design. These changes should include the following:

1. Individual off-gas treatment systems should be provided, as shown in Fig. 7-2, for the primary and secondary burners, and a combined system should be provided for the dissolvers.

2. The order of treatment components should also be as indicated in Fig. 7-2 for reasons discussed previously.
3. A cryogenic distillation separation for krypton removal from the dissolver off-gas should probably be incorporated.

Considerable laboratory and hot cell development work is clearly needed; however, these programs are still in progress. Specific areas in which more development data are required and the types of information needed are as follows:

1. CO oxidation. Determine contaminant level tolerance on catalyst (I_2 and SO_2), heat removal requirements, and tentative design (type and configuration of support).
2. SO_2 removal. Establish the need for or provide justification for the omission of an SO_2 removal system. If removal is needed, as is probable, identify techniques and demonstrate applicability.
3. I_2 loading capacities. Determine dynamic loading capacities for iodine adsorbent selected at predicted operating conditions. This must be done to ensure that unanticipated conditions will not cause unsatisfactory performance.
4. Semivolatile fission product behavior. To assist in the design of a semivolatile fission product removal device, obtain more data on semivolatile behavior at anticipated actual burner conditions.
5. Tritium behavior. Obtain further data on the liberation of tritium during a possible heat treatment step before the fertile particles are dissolved. Near-complete removal of tritium during such a step would be very important should tritium control be required at reprocessing facilities.

Finally, the need for engineering-scale demonstration for as much of the proposed off-gas treatment as is practicable is clear. Because an appreciable portion of the process design uncertainties relate to chemical and physical rather than radiochemical phenomena, these uncertainties can be resolved with cold engineering-scale development tests. Some of the design issues that can be resolved with integrated cold engineering-scale tests include:

1. Verification of the mechanical and operational design of various components. Examples of this would be the demonstration of the gas-tight switching mechanisms for the iodine, radon, and tritium mechanisms; efficiencies of scale-up adsorbent bed designs; and the remote change-out mechanisms for the adsorbent-replaceable components.
2. Coupling and decoupling designs for certain units; for example, verification of heat removal and moisture removal designs for the entire range of anticipated process operating conditions.
3. Verification of on-line control instrumentation. These tests are necessary to prove the sensitivity and response of the analytical instrumentation as well as the flow, pressure, and temperature measuring devices used for process control.
4. Integrated operation of those portions of the off-gas treatment system requiring integrated control. This involves the burner, semivolatiles removal device, CO/HT oxidizer, and iodine and moisture removal beds. Because the off-gases vary in flow rate, temperature, and composition, integration of the off-gas system with operating equipment is an essential element in process and equipment demonstration.

For chemical process operations equivalent to the level of sophistication involved in this off-gas treatment system, engineering-scale demonstration with scale-up factors as small as practicable is prudent. In the

case of the primary burner off-gas treatment train, the availability of a 20-cm burner makes tests with this equipment the most practicable. The off-gas flow rate from this primary burner simulating anticipated operating conditions will be about $0.5 \text{ m}^3/\text{min}$ compared with about $9 \text{ m}^3/\text{min}$ for the combined primary burner operation for the HRDF.

For the reasons mentioned above, it is recommended that engineering-scale demonstration tests be performed for the primary and secondary burner and dissolver off-gases using the treatment schemes given in Fig. 7-2. Because of the complexity and uniqueness of the system design, these tests should be performed on the largest scale practicable, which will probably be dictated by the availability of supporting process equipment and suitable feed material. Integrated operation should be demonstrated for as many of the components as is practicable. This would include all the components shown in Fig. 7-2 up to the radon removal beds. The effectiveness of the radon removal beds must be demonstrated in hot cell operations, and fortunately these beds do not require integrated control.

The importance of hot engineering-scale demonstrations should not be underestimated. On the other hand, the cost is much greater for hot operation than for cold engineering tests. If the needed design information can be obtained with cold development tests, this option should be judiciously pursued.

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- 7-17. Rickman, W. S., "Interim Development Report for Secondary Burning," ERDA Report GA-A13540, General Atomic Company, December 25, 1975.
- 7-18. Croft, A. G., "An Evaluation of Options Relative to the Fixation and Disposal of ¹⁴C-Contaminated CO₂ as CaCO₃," ERDA Report ORNL/TM-5171, Oak Ridge National Laboratory, April 1976.

8. SEMIREMOTE HANDLING SYSTEMS

Handling fixtures designed to demonstrate capabilities for remote maintenance of selected prototype equipment have been designed and fabricated as part of the overall head-end development program. During the current quarter, major efforts have been directed toward establishing operational readiness for all the semiremote fixtures. This has involved the necessary testing, interface verification, and operational check-outs. Specific fixture status and work descriptions are given below.

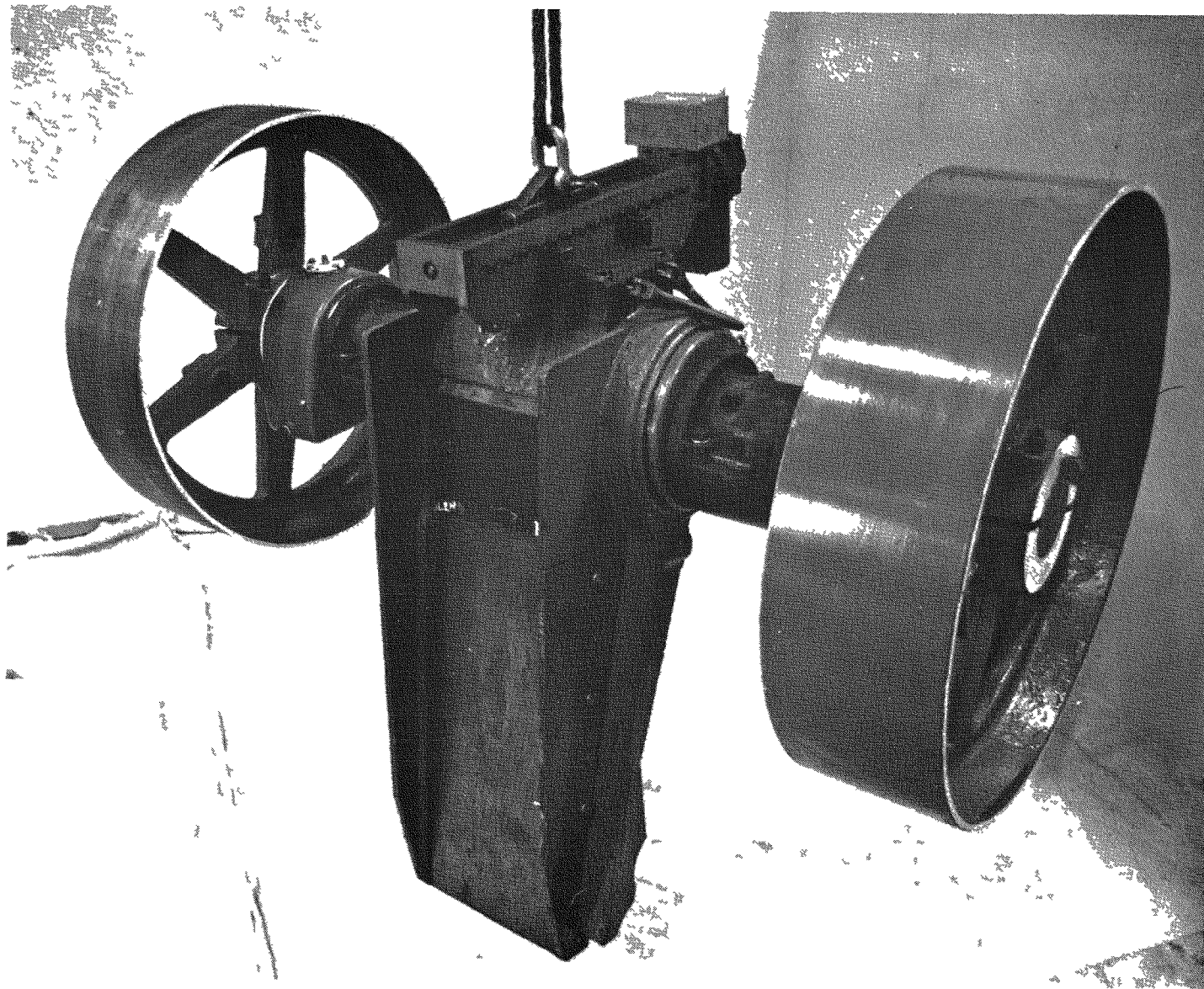
8.1. PROTOTYPE SIZE REDUCTION SYSTEM

Interface check-out and functional verification of the UNIFRAME jaw semiremote lift fixtures were started approximately one month ahead of schedule.

8.1.1. Lift Fixture - Primary Pitman Assembly

The lift fixture was interfaced without problems (see Fig. 8-1); however, the Hydraset could only be used for initial lifting to assess hang-up and to gauge the total weight lifted. No hang-ups occurred during initial lifting, and the total lifted weight of 4.9 tons confirmed the calculated weight.

On assembly of the UNIFRAME, it was found that the minimum gap between the primary pitman and stationary jaws was less than the designed value. This was due to an accumulation of tolerances and resulted in an interference between the jaws during the removal of the pitman assembly. In order to proceed with preliminary tests while a fix was being implemented, the toggle assembly was manually lifted and secured against the body of the



35-135-200-2A

Fig. 8-1. Lift fixture and primary pitman assembly

pitman. This prevented a full test from being carried out. However, the jaw was successfully lifted from the frame to the floor of the experimental area by the fixture.

During this operation, both the manual and automatic modes of attitude correction were successfully used. The only adjustment required was a tightening of the torque-limiting clutch on the lead screw drive.

8.1.2. Lift Fixture - Primary Fixed Jaw

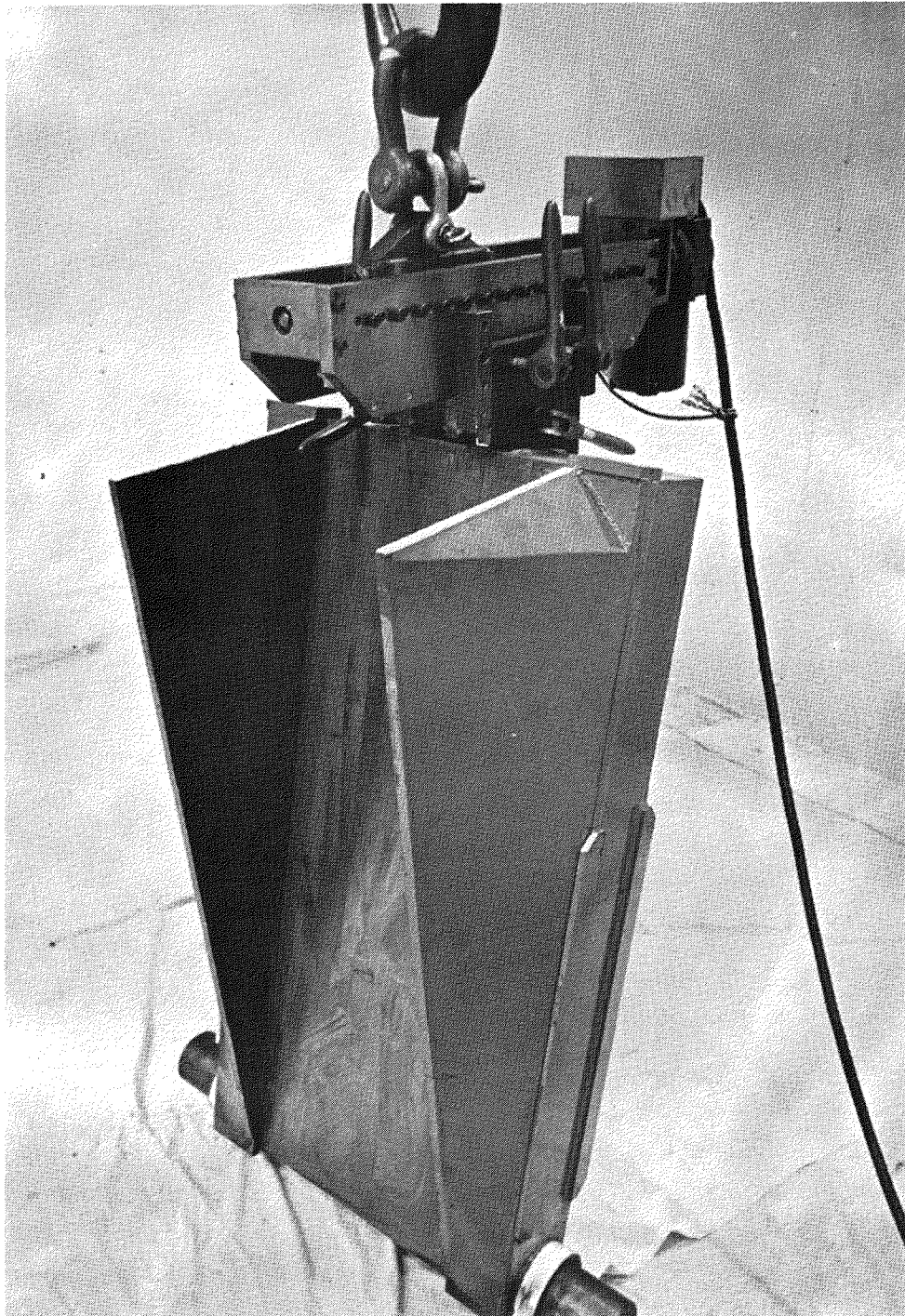
An adapter is attached to the primary pitman assembly lift fixture for lifting the primary stationary jaw. This assembly is shown in operation in Fig. 8-2. The jaw removal operation went smoothly as designed, and the jaw was removed from the frame. Attitude maneuvers were performed manually and automatically without problems.

8.1.3. Lift Fixture - Secondary Pitman Assembly

One of the reasons for removing the secondary pitman assembly from the UNIFRAME was to weld onto the toggle assembly a pin which interfaces with the lift fixture. This pin is grappled by the vertical claw on the fixture to raise the toggle before removal of the jaw. Without this feature in place, the full procedure could not be performed. However, a temporary strap was attached and the jaw was lifted out by the fixture. The weight, as read on the Hydraset, was 5.5 tons, as calculated (see Fig. 8-3).

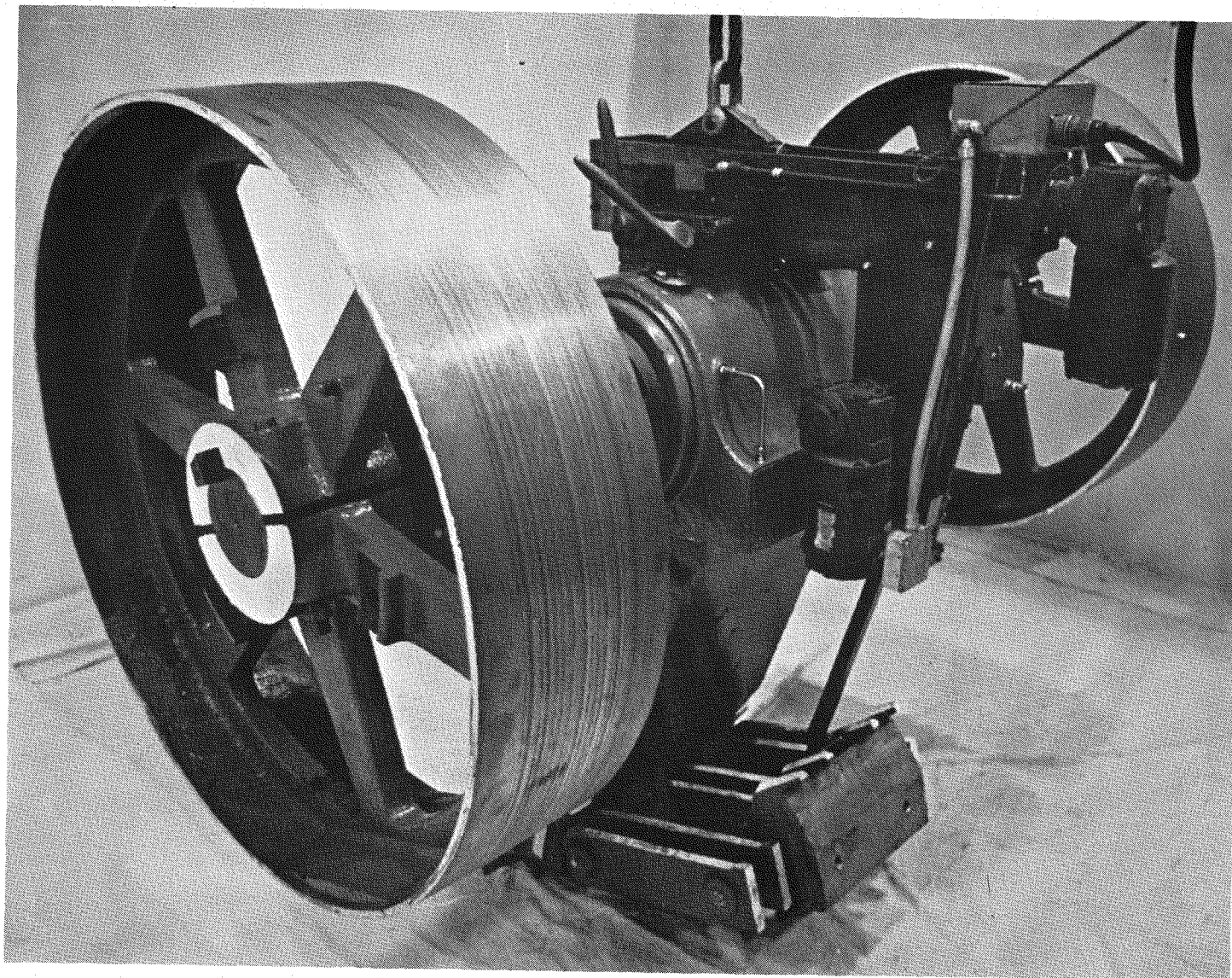
Although a slight interference with the UNIFRAME shroud side plates was experienced, the removal and replacement of the jaw were considered successful as far as the procedure could be followed.

During reassembly, several lines of approach were tried to evaluate the advantages of each. The UNIFRAME shroud has been modified to provide the correct designed clearance, and the interface pin has been welded to the toggle. Full trials will be performed shortly.



35-135-200-11A

Fig. 8-2. Lift fixture and primary fixed jaw



35-135-200-6A

Fig. 8-3. Lift fixture and secondary pitman assembly

8.1.4. Horizontal Removal Fixture - Secondary Fixed Jaw

The design package for this fixture was completed, and the fixture is ready for fabrication. Operation and test procedures have been prepared and are in review. Since this fixture is not required for operation of the prototype line, fabrication has been deferred. This will permit additional time to be spent on the evaluation of the overall remote maintenance requirements of the size reduction system. Fabrication of additional remote maintenance equipment will be considered after the maintenance requirements for the system have been reevaluated as part of an overall reexamination of the remote maintenance philosophy for the head-end reprocessing flowsheet.

8.1.5. Crusher Shroud Shutoff Valve

This valve is presently being fabricated. The valve body had previously been partly machined and temporarily installed in the crusher to expedite check-out of the size reduction system. It is now being fully machined. The gate valve materials are now available, and fabrication is proceeding on schedule.

8.2. HANDLING EQUIPMENT - PRIMARY AND SECONDARY BURNERS

All scheduled design and development work on the handling equipment was completed during the previous quarter. The development activity for the quick disconnect on the burner induction coils was deferred. Current activities have been directed toward preparation of design and operating reports (see Section 8.3).

8.3. SEMIREMOTE HANDLING SYSTEMS - DESIGN AND OPERATING REPORTS

Design and operating reports for the semiremote handling systems are in progress and are scheduled for completion by September 30, 1976, in accordance with milestone requirements. Separate remote handling design

and operating reports are being prepared covering the size reduction and burner systems. Each report will discuss the specific fixture design chosen and will document the design basis, analysis, testing, and operational check-out. These reports will include an assessment of fixture reliability, availability, and safety and an evaluation of overall equipment performance. Recommendations for future remote equipment development will be provided.

9. ALTERNATIVE HEAD-END REPROCESSING

No work was done under this subtask during the current quarter.

10. FUEL RECYCLE DESIGN

10.1. PROGRAM ANALYSIS - SPENT FUEL ELEMENT DECAY HEAT AND SOURCE TERM ANALYSIS

Work has been initiated on this study, which is a follow-on to the HTGR Spent Fuel Composition and Fuel Element Block Flow study completed during FY-1976 (Ref. 10-1). Radioactive decay heat and source term data will be calculated based on the detailed nuclide activities determined in the earlier study. Decay heat data will be calculated for the fissile and fertile fractions of each fuel element type based on the beta/gamma/alpha contribution from each nuclide. Radioactive source terms will be calculated based on the gamma energy spectrum, neutron source, and beta and alpha decay for each nuclide. Data tabulations will be prepared for five HTGR reload cases and for several decay periods after discharge from the reactor.

10.2. PLANT MANAGEMENT

10.2.1. Analysis of Decay Heat Problems in Reprocessing Spent Fuel Particle Hoppers

This head-end system analysis study was completed during the current reporting period, and the draft report on the study (Ref. 10-2) is presently in management review. The purpose of the study was to assess thermal limitations imposed on spent fuel particle hopper designs and operating modes by radioactive decay heat. Constraints on hopper sizing were established with respect to different modes of decay heat removal.

The work reported in the previous quarterly report (Ref. 10-3) established maximum particle temperatures as a function of hopper diameter,

with free convection cooling of the hopper by air. Decay heat removal problems were considered for two types of particles:

1. Crushed fuel element particles, i.e., the feed to the primary burner.
2. Mixed fuel particles, i.e., the primary burner product, which consists of burned-back ThO_2 fertile particles and SiC-coated uranium oxycarbide fissile particles.

10.2.1.1. Free Convection Cooling of Hoppers by Air

During the current reporting period, this study was extended to the consideration of two additional types of particle beds, namely hopper beds containing only fissile particles and beds containing only burned-back fertile particles. Maximum allowable hopper diameters as a function of volumetric heat generation rate for the particles were calculated on the assumption of a maximum allowable temperature of 600°C. Carbon dioxide, air, and helium were considered as the particle bed interstitial gas. Table 10-1 summarizes the maximum allowable hopper sizes for all particle types. These data show that the maximum allowable hopper diameters could be greatly extended by using helium as the interstitial gas.

It is of interest to examine the rates at which the particle hoppers approach thermal equilibrium. Using time-averaged effective thermal conductivities and heat transfer coefficients, temperature transients starting from ambient temperatures have been computed. Figure 10-1 shows the transient wall and centerline temperatures of a 914-mm (3 ft) diameter hopper containing crushed fuel elements. As indicated by the figure, because of the large mass of graphite, several days will pass before the hopper finally reaches thermal equilibrium. On the other hand, the transient time lapse is short, only a few hours, for hoppers containing mixed fuel particles, as shown in Fig. 10-2.

TABLE 10-1
MAXIMUM ALLOWABLE DIAMETERS UNDER NATURAL CONVECTION COOLING

Interstitial Gas	Max. Allowable Temp. (°C)	Maximum Hopper Diameter (mm)			
		Crushed Elements	Mixed Fuel Particles	Fissile	Fertile
CO ₂	600	700	90	105	80
Air	600	750	100	113	87
He	600	1200	140	165	125

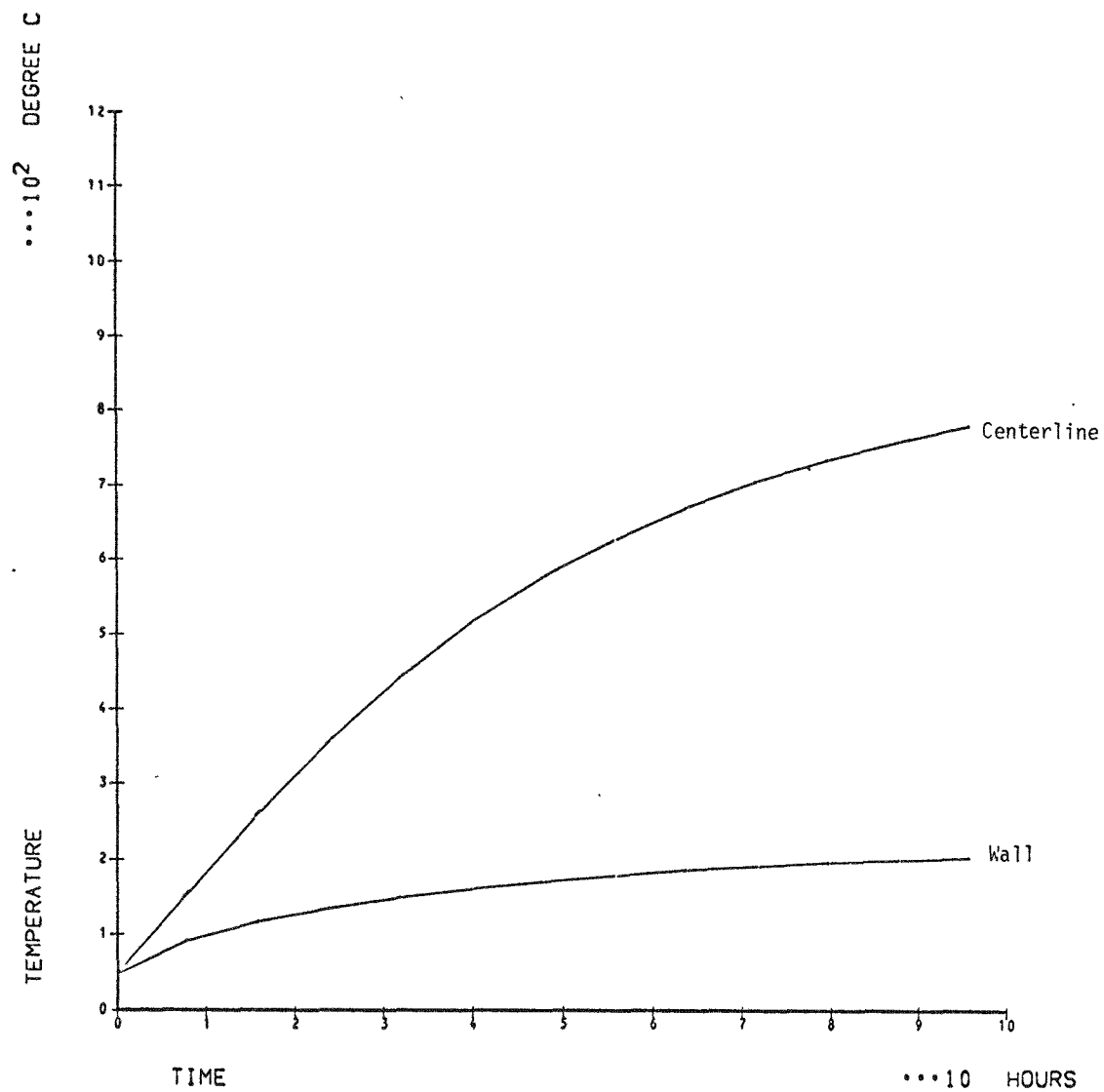


Fig. 10.1. Temperature transient for crushed fuel element bed

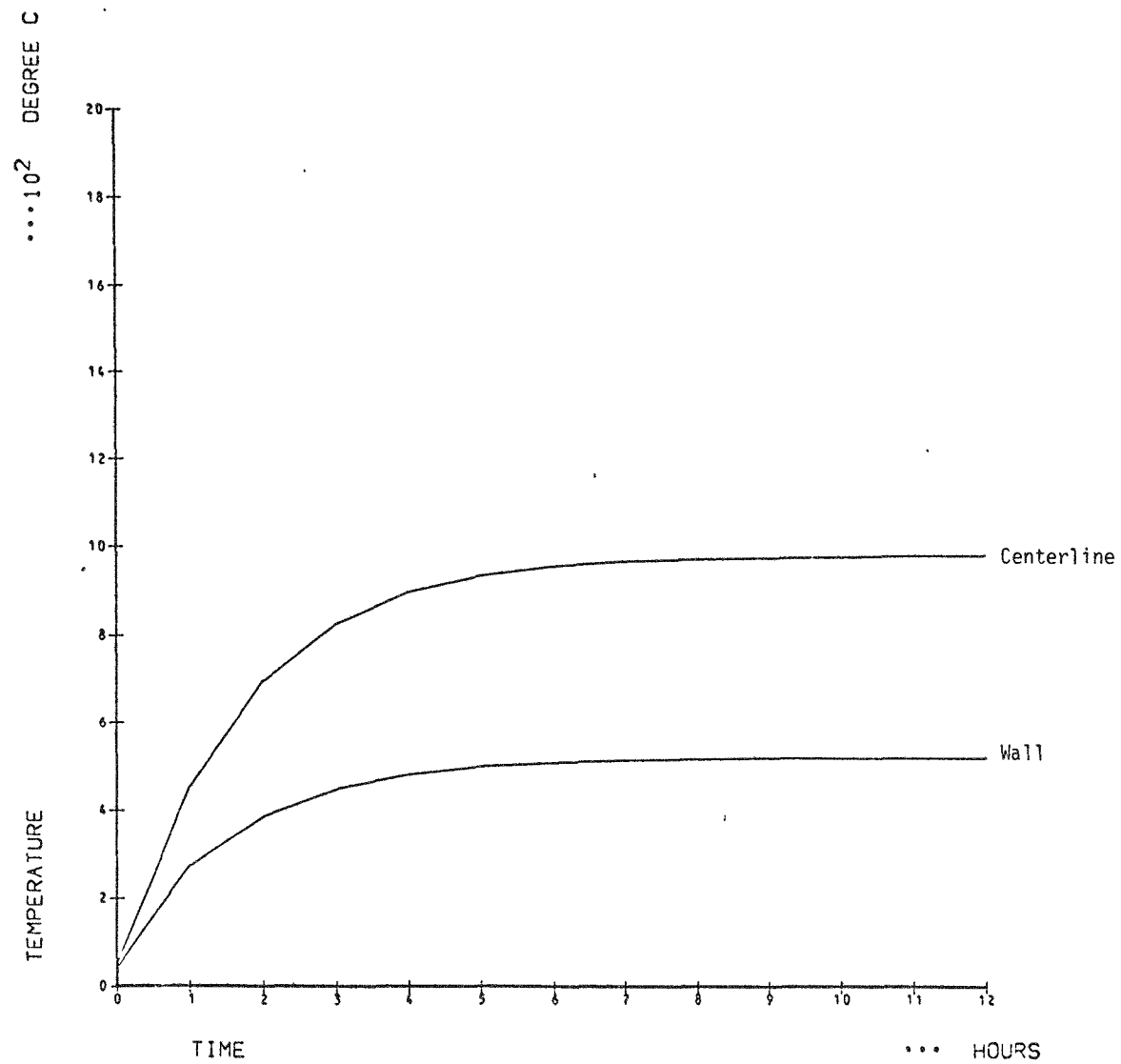


Fig. 10.2. Temperature transient for mixed fuel particle bed

10.2.1.2. Forced Air Cooling in a Jacketed Vessel

The effect of forced air cooling of hoppers was analyzed. It was concluded that, although capable of lowering the average bed temperature, forced convection cooling in jackets does not greatly lower the maximum centerline temperature of a long hopper (~ 3000 mm). Thus, this mode of cooling allows only moderate increase of maximum allowable hopper diameters. The maximum allowable hopper diameters for the mixed fuel particle beds under natural and forced convection cooling are compared in Table 10-2.

10.2.1.3. Internal Direct Cooling of Particle Beds in Hoppers

The effect of hopper internal contact cooling was analyzed for two operating modes: (1) the mass velocity of the coolant gas was held at a value approximately corresponding to the incipient fluidization velocity of the fissile particles in a mixed particle bed; and (2) the velocity of the coolant gas was increased beyond that required for incipient fluidization of the particles in the hopper.

For a 914-mm (3 ft) diameter hopper containing crushed fuel elements, with the coolant gas mass velocity held at approximately the incipient fluidization velocity of fissile particles, the bed and hopper temperatures were found to be well within tolerable limits.

Under fluidized bed conditions for either mixed fuel particle beds, fissile particle beds, or fertile particle beds, the temperature of the beds was found to be approximately uniform. Figure 10-3 shows the transient fluidized bed temperatures for the three types of particle beds. In all cases it is seen that the bed temperatures reach equilibrium in about 2 hr. The equilibrium temperatures are established by the volumetric decay heat generation rates.

TABLE 10-2
COMPARISON OF MAXIMUM ALLOWABLE HOPPER DIAMETERS FOR MIXED
FUEL PARTICLES

Interstitial Gas	Maximum Allowable Temp. (°C)	Maximum Allowable Hopper Diameter (mm)	
		Forced Convection	Natural Convection
CO ₂	600	110	90
Air	600	120	100
He	600	170	140

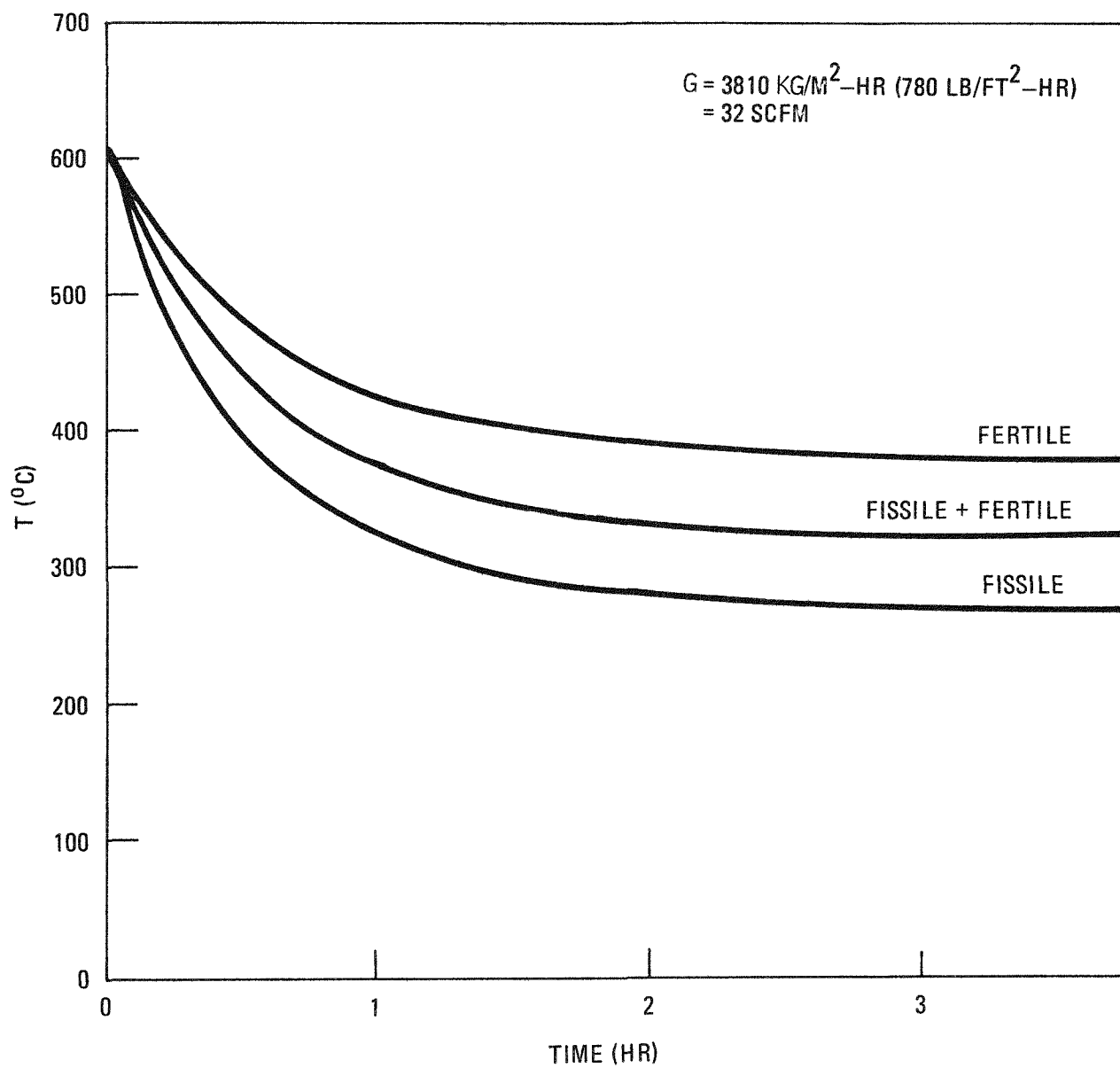


Fig. 10-3. Transient fluidized bed temperatures of 152-mm-diameter fuel particle hoppers

At a mass velocity of $3810 \text{ kg/m}^2\text{-hr}$, for mixed fuel particles of 300-cm bed height, the equilibrium fluidized bed temperatures are shown in Fig. 10-4 as a function of hopper diameter. It can be shown that in this case, the fluidized bed temperature approaches $\sim 510^\circ\text{C}$ as the hopper diameter approaches infinity.

Direct contact cooling by passing a gas such as CO_2 through the particle bed eliminates the constraint on hopper diameters. Instead, for each given maximum allowable temperature, the maximum bed height is a function of the mass velocity of the cooling gas. It was found that for a large-diameter mixed fuel hopper, the maximum allowable bed height increases linearly with the mass velocity of the cooling gas. The results will not significantly be changed by whether CO_2 or air is used as the cooling gas because both gases have similar specific heats.

Using 600°C as the upper temperature limit, the maximum allowable bed heights for infinitely large-diameter hoppers with internal cooling were determined and are presented in Table 10-3.

10.2.1.4. Summary and Conclusions

Temperature distributions in particle hoppers required in an HTGR head-end reprocessing facility have been computed. Constraints on hopper diameters and bed heights have been established, based on a proposed upper temperature limit of 600°C .

The effective thermal conductivities of the packed beds of particles are estimated using the Kunii-Smith correlation, which is found to be the best among existing correlations (see Ref. 10-3, Section 10.1.1.3).

Hopper sizing constraints for the mixed fuel particles, i.e., the primary burner product consisting of a mixture of SiC-PyC coated fissile kernels and bare fertile kernels, are presented in Table 10-4. Similarly, for the crushed fuel element particles, i.e., the primary burner feed, the sizing constraints shown in Table 10-5 apply.

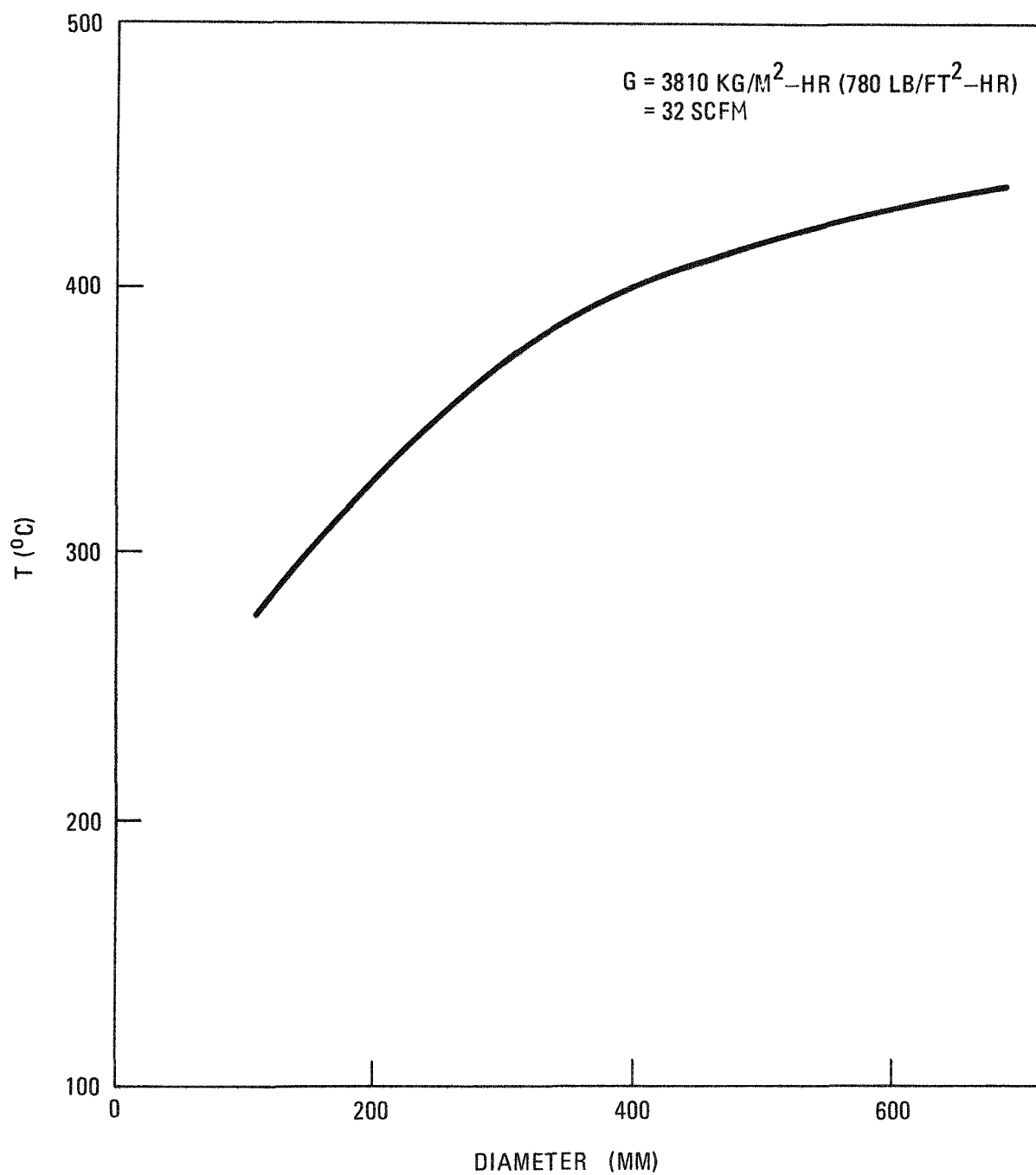


Fig. 10-4. Equilibrium fluidized bed temperatures of mixed fuel particle beds

TABLE 10-3
MAXIMUM ALLOWABLE BED HEIGHTS WITH INTERNAL COOLING

Mass Velocity, G (kg/m ² -hr)	Maximum Allowable Temp. (°C)	Maximum Allowable Bed Height (mm)	
		Crushed Fuel Elements	Mixed Fuel Particles
635	600	16,500	600
3810	600	99,000	3500

TABLE 10-4
MAXIMUM HOPPER SIZES FOR MIXED FUEL PARTICLES

Cooling Mode	Cooling Gas Mass Velocity (kg/m ² -hr)	Interstitial Gas	Maximum Diameter (mm)	Maximum Bed Height (mm)
Natural convection	0	CO ₂	90	∞
Natural convection	0	Air	100	∞
Natural convection	0	He	140	∞
Forced convection	33,200	CO ₂	110	3,000
Forced convection	33,200	Air	120	3,000
Forced convection	33,200	He	170	3,000
Direct contact internal cooling	635	CO ₂	∞	600
Direct contact internal cooling	3,810 ^(a)	CO ₂	∞	3,500

(a) Particle bed vigorously fluidized

TABLE 10-5
MAXIMUM HOPPER SIZES FOR CRUSHED FUEL ELEMENT PARTICLES

Cooling Mode	Cooling Gas Mass Velocity (kg/m ² -hr)	Interstitial Gas	Maximum Diameter (mm)	Maximum Bed Height (mm)
Natural convection	0	CO ₂	700	∞
Natural convection	0	Air	750	∞
Natural convection	0	He	1,200	∞
Direct contact internal cooling	635	CO ₂	∞	16,500

The analysis of decay heat problems in reprocessing spent fuel particle hoppers also included consideration of hopper beds containing only fissile particles and beds containing only burned-back fertile particles. The ashes from the secondary burner, consisting of burned-back fissile particles, were not included owing to the large uncertainties of the decay heat generation rate and the thermophysical properties of the ashes.

It can be concluded that for all particle types considered in this study, constraints imposed on hopper sizing from thermal considerations are not very restrictive. The decay heat in particle hoppers can be adequately removed by any one of three cooling modes, provided that the sizing constraints on hopper diameters or bed heights as determined in the study are observed. While the thermal constraint for the case of direct internal cooling is not restrictive, the constraints imposed on hopper sizing under natural or forced external convection cooling conditions can be largely removed by using helium as the interstitial gas. It is anticipated that criticality constraints (Ref. 10-4) in many cases may be more severe than thermal constraints. However, a more detailed criticality analysis will be required to adequately define the criticality constraints for spent fuel particle hoppers.

10.2.2. Process Yields and Material Throughput

Work on this study was initiated during the current reporting period, and detailed material balances are being prepared based on recently determined spent fuel element compositions and representative production operating modes. The material balance will account for process yields, decontamination factors, process impurities, and system efficiencies which reflect the current status of the development program.

10.2.3. Hot Engineering Test (HET) Reprocessing Preliminary Design

Support activities for HET-Reprocessing preliminary design continued during this reporting period and were directed toward completion of the

design criteria phase. A draft of the HET-Reprocessing Facility Conceptual Design Criteria document was completed July 10, 1976 and was subsequently approved by ORNL for use by Ralph M. Parsons Company in preparing the final design criteria documents. General Atomic Company HET-Reprocessing design criteria responsibilities included the following systems:

1. Fuel Element Size Reduction (System 1100).
2. Primary Burning (System 1200).
3. Particle Classification and Material Handling (System 1300).
4. Particle Crushing and Secondary Burning (System 1400).
5. Dissolution and Feed Adjustment (System 1500).
6. Solvent Extraction (System 1600).
7. Product Handling (System 1800).
8. Process Support (System 1900).

Responsibility for Systems 1100, 1800, and 1900 will be reassigned to Ralph M. Parsons Company during the conceptual design phase.

10.2.3.1. HET-Reprocessing Design Bases

The design bases for the HET-Reprocessing conceptual design criteria are summarized below.

10.2.3.1.1. Reprocessing Systems.

System 1100 - Fuel Element Size Reduction

1. This system crushes segmented Fort St. Vrain (FSV) spent fuel (7-in. o.d. by 31 in.) to $\leq 3/16$ -in. ring size with an average particle size of $\leq 2800 \mu\text{m}$. The FSV elements are segmented elsewhere (TBD) by another system prior to input into System 1100.
2. The jaw crusher first reduces fuel segments to $\leq 3/4$ -in. ring size.

3. The scalper then separates $>3/4$ -in. material and recycles it to the jaw crusher.
4. The double roll crusher reduces material to final size.

System 1200 - Primary Burning

1. The fluidized bed reactor (20-cm i.d.) oxidizes graphite and outer fuel particle coatings to less than 2 wt % residual carbon in product.
2. The entrained carbonaceous fines ($>1\text{ }\mu\text{m}$) are separated from the off-gas and recycled to the burner.
3. The design burn rate is 200 g C/min (nominal three FSV elements per 24 hr).

System 1300 - Particle Classification and Material Handling

1. This system separates fertile and fissile particle fractions with $<10\%$ cross-over to the fissile stream and $<15\%$ to the fertile stream.
2. It transports solid particulates between head-end process equipment.
3. It provides surge for independent head-end operations and back-cycle capability.
4. It removes characteristic samples for process control and accountability.
5. It provides weighing stations for process control.

6. The classifier is a zig-zag chamber (1.5 by 1.5 by 40 in.).
7. The system uses the recirculation pneumatic transport system to reduce the load on the off-gas system.

System 1400 - Particle Crushing and Secondary Burning

1. The double roll particle crusher breaks the outer SiC coating of fuel particles to expose inner carbon for burning.
2. The fluidized bed reactor (10-cm i.d.) oxidizes residual graphite and the inner carbon coating of fuel particles, and converts fuel kernels to oxides.
3. The burner processes a 12-kg batch of crushed feed (nominal fissile or fertile fraction of one FSV element in a 5-hr cycle).

System 1500 - Dissolution and Feed Adjustment

1. This system dissolves oxidized fuel kernels in nitric acid and separates insoluble materials from solution.
2. It denitrates high-acid feed solution.
3. It prepares 1.5M heavy metal feed (1M HNO₃) for Thorex solvent extraction and 0.043M uranium feed (2M HNO₃) for Purex.
4. One-half batch per day of Thorex SX operations and 1.5 batches per day of Purex SX operations are required.

System 1600 - Solvent Extraction

1. A five-column Thorex solvent extraction train (1.0- to 1.5-in. diameter) separates fission products, partitions uranium and thorium into separate products, and continuously regenerates and recycles solvent.
2. The same solvent extraction train is used for Purex (with IBX and IBS columns bypassed).
3. The fertile fraction from 1 FE/day is required for Thorex SX operations and the fissile fraction from 4 FE/day for Purex.
4. This system provides interim storage for HLW from three 7-day Thorex campaigns or one 7-day Purex campaign.

System 1800 - Product Handling

1. This system receives U/Th product solutions from Thorex operations and concentrates them to 1.3M heavy metal.
2. It receives uranium product from Purex operations and concentrates it to 1.3M uranium.
3. It provides interim product storage for three Thorex and three Purex 7-day campaigns.
4. It provides interim storage for ILW from one Thorex 7-day campaign (Purex ILW volume is about 20% of Thorex waste).

10.2.3.1.2 Process Support (System 1900).

Subsystem 1901 - Unirradiated Feed Material Handling (GA). This subsystem includes preparation of unirradiated fuel feed material for

acquisition of cold test data for comparison with hot test data.

Activities included are:

1. Loading scrap FSV core fuel rods into excess H-327 machined graphite blocks (GA-SVA Facility).
2. Segmenting loaded fuel elements to System 1100 size requirements.
3. Loading segmented fuel into FSV-3 licensed shipping containers.
4. Transporting fuel elements to ORNL via common carrier.

Subsystem 1906 - Irradiated Feed Material Handling (ICPP). This subsystem includes preparation of irradiated fuel feed material for acquisition of hot test data. Activities included are:

1. Retrieving spent FSV fuel elements from IFSF storage.
2. Transporting FSV spent fuel elements to the fuel segmenting facility at ICPP.
3. Segmenting FSV spent fuel to System 1100 size requirements.
4. Loading segmented fuel into one- or two-element shipping canisters.
5. Transporting fuel canisters to IFSF for interim storage and/or loading into FSV-1 or PB-2 shipping casks.
6. Transporting fuel canisters to ORNL via FSV-1 or PB-2 shipping casks and transporters.

Subsystem 1911 - Feed Material Handling (ORNL). This subsystem includes receiving, unloading, storage, and transfer to process cells of irradiated and unirradiated fuel feed material at TURF. Activities include:

1. Receiving, cleaning, and unloading a FSV-1 or PB-2 fuel cask outside the TURF facility.
2. Transferring the cask into the TURF receiving area.
3. Raising the cask to the TURF third floor level.
4. Lowering the cask into the Fuel Storage Basin.
5. Unloading and temporarily storing fuel canisters in the Fuel Storage Basin.
6. Preparing the cask and loading it for the return trip to ICPP.
7. Transferring spent fuel canisters from the Fuel Storage Basin to Cell F via Cell B.
8. Receiving unirradiated fuel drums and temporarily storing them in the TURF loft area.
9. Unpacking unirradiated fuel canisters from drums and transferring the canisters to Cell F via Cell A airlock and Cell B.
10. Transferring unirradiated or irradiated fuel from Cell F to Cell D via Cell B (airlock under door).
11. Transferring unirradiated or irradiated fuel into the crusher feed tray.

Subsystem 1921 - Burner Off-Gas. This subsystem includes collection of gaseous effluents from the primary and secondary burners, fuel element and particle crushers, and transport and classification bleed streams for pretreatment prior to discharge to the ORNL hot off-gas system. Activities include:

1. Removing semivolatiles and particulates.
2. Removing iodine.
3. Oxidizing CO/HT.
4. Removing tritium.
5. Performing radon hold-up for decay of Rn-220.
6. Performing gas sampling and analysis.

Subsystem 1931 - Dissolver Off-Gas. This subsystem collects gaseous effluents from heavy metal dissolution and aqueous processing vessel vents for pretreatment prior to discharge to the ORNL hot off-gas system. Activities include:

1. Converting NO_x.
2. Removing iodine.
3. Removing water vapor.
4. Performing radon hold-up for decay of Rn-220.
5. Performing gas sampling and analysis.

Subsystem 1956 - Solid Waste Handling. This subsystem includes handling of solid waste and contaminated equipment associated with reprocessing processes located in Cells D, E, and G. Activities included are:

1. Transferring insols from Cell G to Cell D for canning.
2. Transferring waste drums through the Cell D hatch into the transfer cask.
3. Decontaminating and assaying waste drums in the TURF loft station.
4. Transporting casks to a storage or disposal area.
5. Transferring failed equipment from Cell D to the disposal station in Cell B.
6. Decontaminating, cutting, and packaging failed equipment in Cells B, E, and G.
7. Transferring failed equipment drums through the roof in Cells E and G and through the Cell A airlock from Cell B.
8. Transporting failed equipment drums to the burial ground.

Subsystem 1961 - Solid Product Handling. This subsystem includes transfer and interim storage of burner product material derived from the primary and secondary burning of both irradiated and unirradiated fuel. Activities included are:

1. Transferring burner product from Cell D to Cell F via Cell B (airlock under door).
2. Storing burner product in fuel shipping canisters in concrete sleeves in Cell F.

3. Transferring product canisters from Cell F to Cell B for preparation for transport to final processing or storage.

4. Transferring secondary burner product from Cell D to Cell G via transfer cask.

Subsystem 1971 - Sample Handling. This subsystem includes transfer of samples from Cell D and E to analytical laboratories or sample inspection stations. Activities included are:

1. Preparing samples received from process for transport.
2. Transferring a loaded sample capsule into the TURF sample handling system.
3. Transferring the capsule to a shielded carrier.
4. Transporting the carrier to a laboratory or sample inspection station.

10.2.3.2. HET-Reprocessing Test Program

A summary HET-Reprocessing hot operations flow diagram and material balance was prepared which describes the hot operations test program (campaign basis) and product storage and waste disposal requirements (Fig. 10-5). The test program is scheduled for 2 yr of hot operations and is based on processing a total of 96 Fort St. Vrain spent fuel elements through the primary head-end systems in eight separate 4-day burner campaigns. The fissile fraction from 35 fuel elements and the fertile fraction from 36 fuel elements are processed through the secondary head-end systems to obtain feed for the aqueous processing systems. A total of eight Thorex solvent extraction campaigns (7-day continuous operation) are planned. Six campaigns will be performed using the fertile particle fraction and two using the fissile particle fraction. The HET-Reprocessing material balance is based on the fuel element composition shown in Table 10-6.

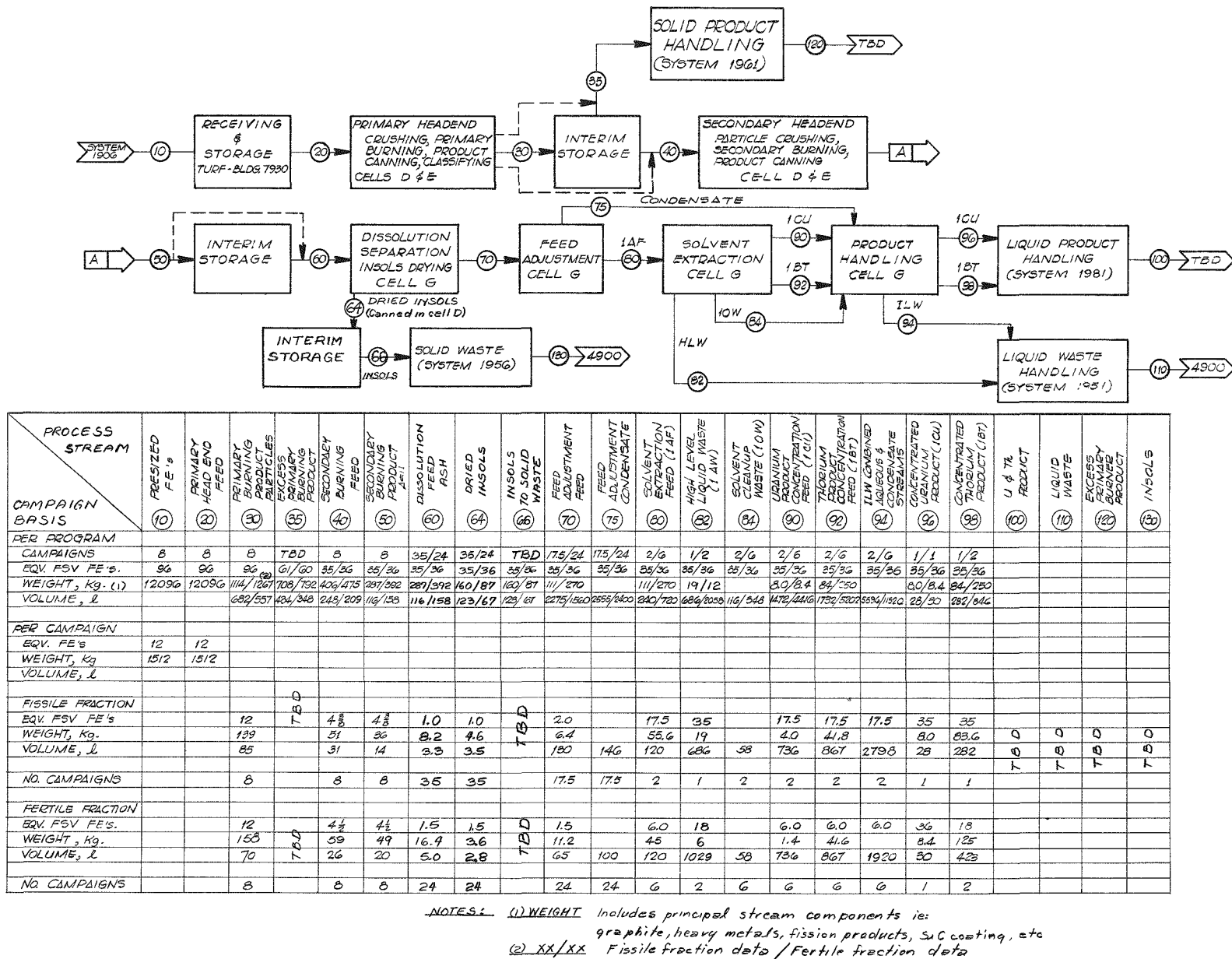


Fig. 10-5. Summary HET-Reprocessing hot operations flow diagram and material balance

TABLE 10-6
SPENT FUEL ELEMENT DEFINITION

Average Standard FSV Element, Segment 4 As Built

TRISO-TRISO

VSM Th-UC₂ Fissile

VSM ThC₂ Fertile

FUEL ELEMENT COMPOSITION

<u>Component</u>	<u>Weight (kg/FE)</u>	
Graphite block including fuel plugs and dowels		88.4
Fuel rod matrix		4.21
Fissile particles (avg $\rho = 2.35 \text{ g/cm}^3$)		17.1
Outer PyC coating	5.54	
SiC coating	4.58	
Inner PyC coating	1.95	
Buffer coating	1.55	
Kernel (avg $\rho = 9.01 \text{ g/cm}^3$)	3.50	
	<u>Fresh</u>	<u>Spent</u>
Uranium	0.62	0.23
Thorium	2.56	2.39
Fission products		0.54
Other heavy metals		0.02
Carbide	0.32	0.32
Fertile particles (avg $\rho = 3.16 \text{ g/cm}^3$)		16.2
Outer PyC coating	3.03	
SiC coating	2.42	
Inner PyC coating	1.43	
Buffer coating	1.14	
Kernel (avg $\rho = 8.81 \text{ g/cm}^3$)	8.22	
	<u>Fresh</u>	<u>Spent</u>
Uranium		0.23
Thorium	7.49	6.93
Fission products		0.33
Other heavy metals		0.00
Carbide	0.73	0.73
Burnable poison rod (avg $\rho = 1.6 \text{ g/cm}^3$)		0.10
Boron		0.004
Carbide		0.001
Matrix		0.095
TOTAL WEIGHT		<u>126.0</u>

TABLE 10-6 (Continued)

RADIOACTIVITY^(a)

	Ci/FE	
	<u>180 Days Decay</u>	<u>1-1/2 Years Decay</u>
Fissile fraction	39,400	17,800
Fertile fraction	36,300	16,400

DECAY HEAT^(a)

	Btu/hr•FE	
	<u>180 Days Decay</u>	<u>1-1/2 Years Decay</u>
Fissile fraction	810	360
Fertile fraction	740	340

(a) Radioactivity and decay heat calculated for full burnup, six equivalent full power years.

10.3. HTGR RECYCLE DEMONSTRATION FACILITY (HRDF)

10.3.1. Reprocessing Flowsheet Review and Updating

This study is part of the continuing technology assessment to ensure that (1) the proposed HRDF flowsheet incorporates recent technology development improvements and new design data, and (2) supporting technical programs are apprised of flowsheet design issues requiring resolution. The updated reprocessing flowsheet is intended to become an approved baseline document for HRDF design definition and to provide guidance for technical development activities.

Pertinent topical and progress reports issued by ORNL, GA, and ACC in 1976 have been reviewed for input to this study. Joint GA recycle engineering/technology development group review of the reprocessing flowsheet is being initiated.

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- 10-3. "Thorium Utilization Program Quarterly Progress Report for the Period Ending May 31, 1976," ERDA Report GA-A13949, General Atomic Company, June 30, 1976.
- 10-4. Yuill, W. A., "Criticality Evaluations for the HRDF," ICPP Report, December 1975.

APPENDIX A
PROJECT REPORTS PUBLISHED DURING THE QUARTER

Wong, H. W., and L. Abraham, "HTGR Spent Reflector Block Disposal Study," ERDA Report GA-A13860, May 14, 1976.

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Pierce, V. H., and M. P. Rothstein, "An Economic Analysis of U-235 Recycle in the HTGR," ERDA Report GA-A13836, July 15, 1976.

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Commissariat a l'Energie Atomique
Centre d'Etudes Nucleaires de Saclay
BP No. 2
91190 Gif-sur-Yvette
France</p> | <p>1 J. L. McElroy
Battelle Northwest Laboratories
P.O. Box 999
Richmond, Washington 99352</p> <p>1 Dr. K. Hackstein*
HOBEK
6450 Hanau/Main
Postfach 787
Germany</p> <p>1 Dr. D. Stoelzl*
Hochtemperatur Reaktorbau GmbH
Gottlieb-Daimler-Strasse 8
D-68 Mannheim - 1
Postfach 5360
Germany</p> <p>1 K. Notz
Oak Ridge National Laboratory
Oak Ridge, Tennessee 37830</p> <p>1 A. L. Lotts, Program Manager
Thorium Utilization Program
Oak Ridge National Laboratory
P.O. Box X
Oak Ridge, Tennessee 37830</p> |
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