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# REPROCESSING DEVELOPMENT FOR HTGR FUELS

by
C. A. HEATH and M. E. SPAETH

MASTER

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## ABSTRACT

The High-Temperature Gas-Cooled Reactor (HTGR) is a thermal reactor concept now being introduced into the utility industry. It provides high efficiency, improvements in reactor safety characteristics, and the implementation of an additional energy source by the use of thorium. The HTGR uses helium as the coolant and graphite as the moderator and core structural material. The fuel cycle employs uranium highly enriched (93%) in U-235 for the initial and makeup fissile material, thorium as the fertile material, and U-233 as the bred material, which is recovered for recycle to the reactor.

The technology required for the recycle of the bred U-233 in the HTGR fuel includes reprocessing to extract the valuable fissile materials in the spent fuel from the fission product wastes and refabrication to manufacture the U-233 back into fuel elements for further use. This paper describes the development program being undertaken for reprocessing of HTGR fuels. Most of the work described is being performed at General Atomic Company as part of the Thorium Utilization Program funded by the U.S. Energy Research and Development Administration.

## INTRODUCTION

The program to develop the necessary technology for reprocessing of HTGR fuel elements and for refabrication of elements containing U-233 is known as the Thorium Utilization Program, or sometimes as the National HTGR Fuel Recycle Program. Work on the program is being performed by three contractors. General Atomic Company is primarily responsible for the development of reprocessing technology. Allied Chemical Company, operator of the Idaho Chemical Processing Plant (ICPP), has the responsibility for construction and operation of a reprocessing pilot plant to handle radioactive spent fuel elements. These fuel elements will be discharged from the Fort St. Vrain reactor. Finally, Oak Ridge National Laboratory (ORNL) is responsible for development of refabrication technology and for construction and operation of a pilot plant for refabrication of HTGR fuel. The details of this program, including a description of the reprocessing and refabrication pilot plants, which are now scheduled for operation in late 1981 or 1982, are given in Ref. 1.

## CHARACTERISTICS OF HTGR FUELS

The HTGR contains a graphite moderator, which also serves as the core structural material, and is cooled by helium gas. The basic fuel element in the HTGR is a graphite block 79.3 cm (31.4 in.) high with a hexagonal cross section 35.9 cm (14.2 in.) across the flats, as illustrated in Fig. 1. The graphite block is drilled lengthwise with two sets of holes; one allows the passage of helium coolant, the second contains the fuel rods. Fuel rods are formed by the molding of selected blends of fuel particles with a graphitic pitch; each fuel rod is 5.1 cm (2 in.) in length and has a diameter of 1.58 cm (0.625 in.). The fuel particles used in the HTGR fuel are schematically shown in Fig. 2. These fuel particles contain either uranium dicarbide (highly enriched in U-235 or recycle U-233) or $ThO_2$. The particles shown in Fig. 2 typically have diameters of 500 to 800 μm.

Two particle types are used in the HTGR fuel; they are categorized by the coatings that have been applied to them. As may be seen in Fig. 2, they are classified as BISO- or TRISO-coated particles. BISO particles are coated with a relatively porous buffer layer of carbon and then with a dense coating of pyrolytic carbon. TRISO coatings, in addition, have a silicon carbide coating placed between two layers of pyrolytic carbon. The SiC layer provides a means of separating these particles from the BISO particles in head-end reprocessing operations; it also enhances fission product retention in the fissile particles. BISO coatings are used for particles initially loaded only with thorium oxide, while TRISO coatings are used for particles loaded with uranium.

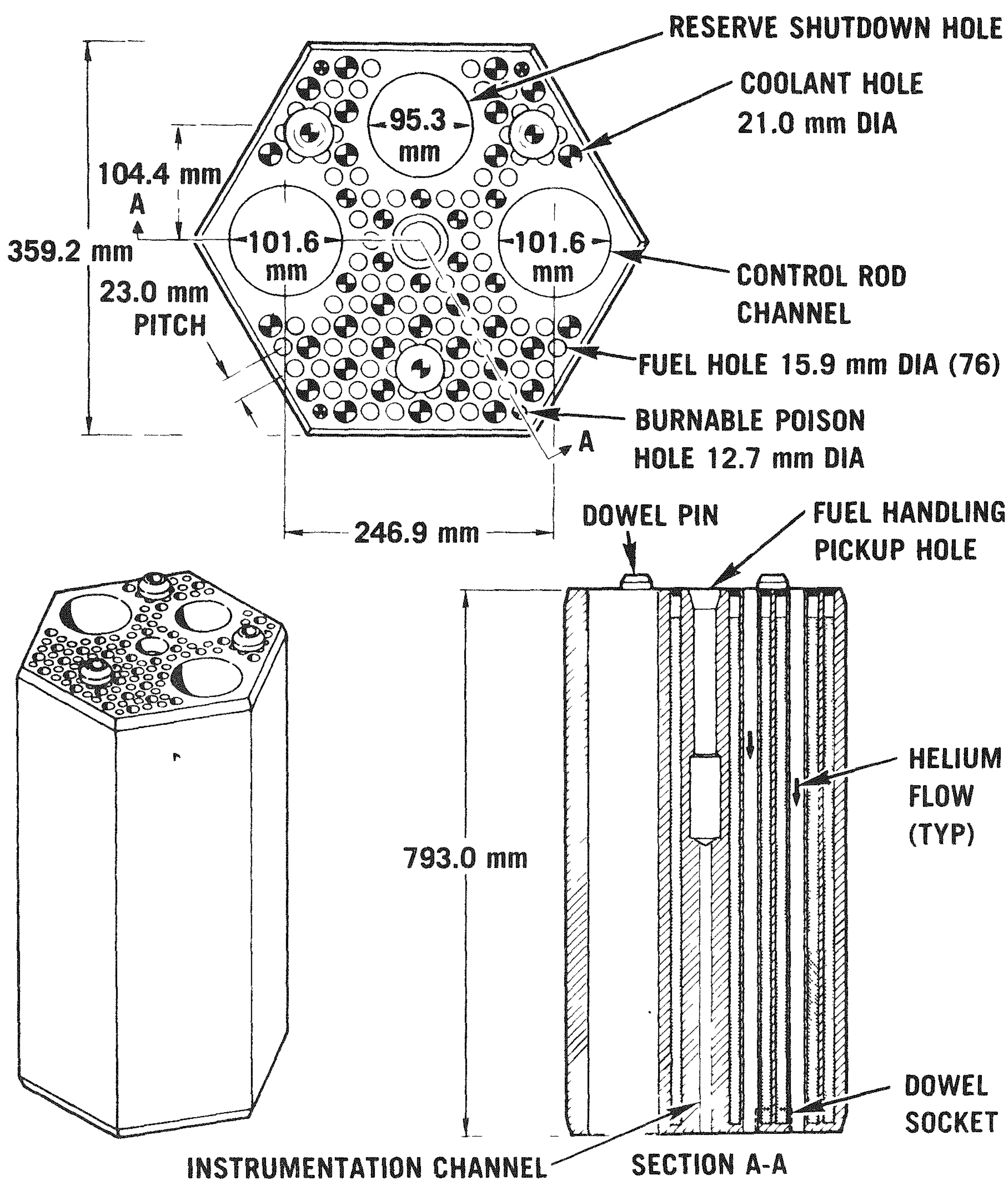


Fig. 1. HTGR fuel element

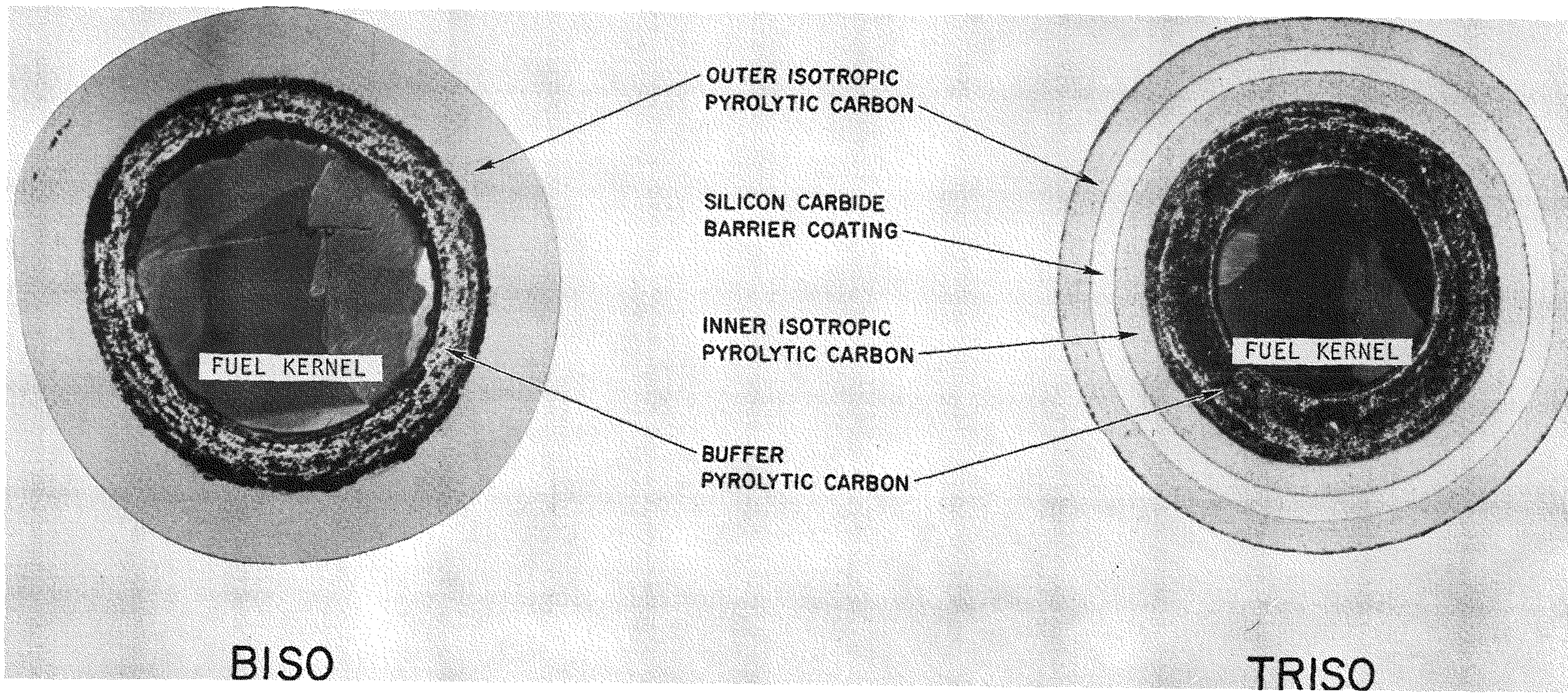


Fig. 2. HTGR coated fuel particles

During irradiation in the reactor, the fissile and fertile materials undergo one or more of the processes illustrated in Fig 3. An initial core loading of fissile and fertile particles will be irradiated to spent fuel with fissile particles containing mostly U-235, U-236, and fission products, and fertile particles containing mostly thorium, U-233, and fission products. The main objective of reprocessing the spent fuel is to recover the bred fissile U-233 in a form suitable for use in recycle fuel e.ements. Secondary objectives are to recover thorium and unused U-235 for later possible reuse in recycle fuel and to separate fission products for disposal as waste. A successful reprocessing technique must recover the bred U-233 from the fertile particle with minimal contamination by the U-236 now contained in the spent fissile particles. U-236 acts as a neutron absorber or poison in the HTGR core and significant contamination of bred fuel with U-236 reduces its value in the reactor.

The fuel design described here is different from the fuel in the Fort St. Vrain HTGR. Both fissile and fertile particles in Fort St. Vrain are TRISO coated, and the fissile particle contains a 4.25:1 ratio of Th to U in a $(U/Th)C_2$ form. Since fuel from Fort St. Vrain will be the only irradiated HTGR fuel available in large quantities during the reprocessing development program, process demonstration will be performed with TRISO-TRISO fuel. However, as will be shown, reprocessing of TRISO-TRISO fuel is considered more difficult than reprocessing TRISO-BISO fuel.

## A FLOWSHEET FOR HTGR FUEL REPROCESSING

A block flow diagram of a flowsheet under development for reprocessing of HTGR fuel as part of the Thorium Utilization Program is shown in Fig. 4. Fuel element types are classified as initial (I) or makeup (M) recycle elements containing U-235 (25R) or recycle elements containing U-233 (23R) according to the type of fissile particle contained in the fuel element. Only one type of fissile particle will be contained in a given fuel element. The fuel element types will be segregated in the receiving area of a fuel recycle facility prior to further processing.

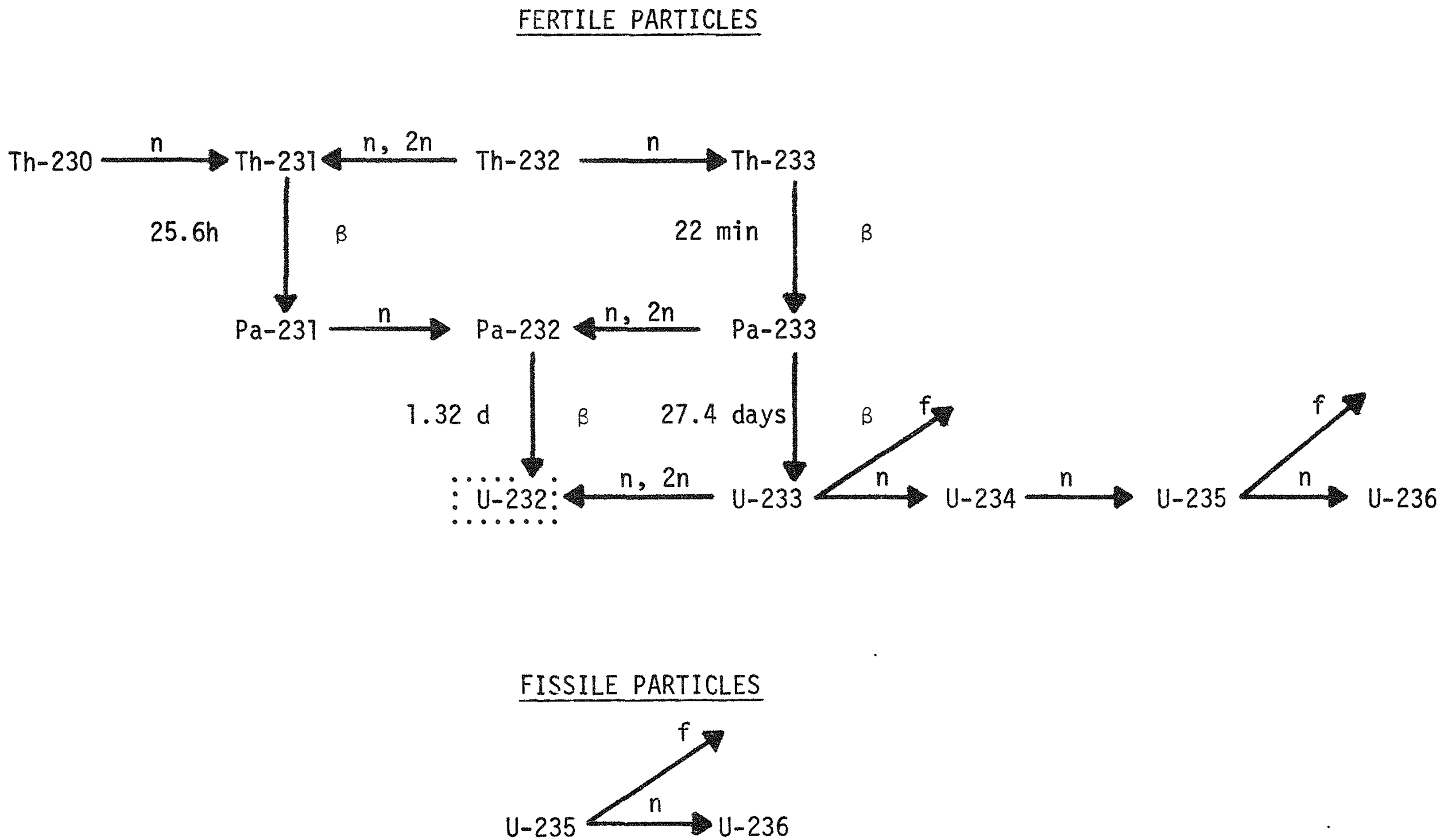


Fig. 3. Nuclear reactions in the fuel

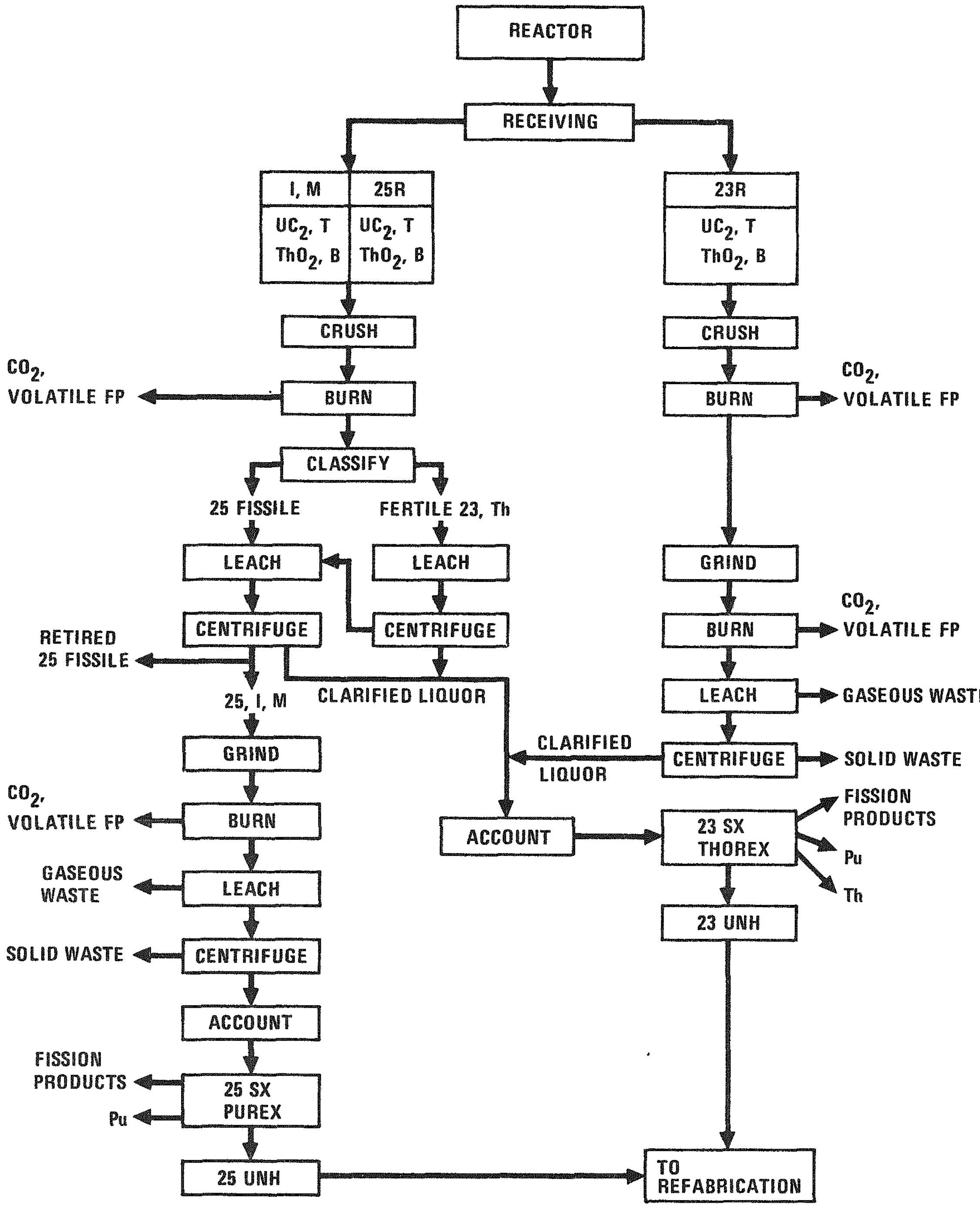


Fig. 4. Reprocessing flow diagram

Separation of fission products from fissile materials is eventually achieved by solvent extraction from a solution of these materials which is formed by dissolution in nitric acid. Effective dissolution cannot be performed if significant amounts of graphite are present. The reference method of removing this graphite is by combustion, which is most readily achieved in fluidized-bed burners. Experimental tests have shown that a feed with a maximum particle size no greater than 3/16 in. is required for successful operation in the fluidized-bed burners (Refs. 2-4). The spent HTGR fuel element received at a reprocessing facility must therefore be reduced to this size before removal of the graphite by combustion.

The initial or head-end steps of the HTGR fuel reprocessing flowsheet include size reduction, or crushing; burning to eliminate the graphite moderator; classification of particle types, crushing or grinding as required; and dissolution in nitric acid solutions. The treatment of various fuel types can be traced using Fig. 4.

In fuel elements bearing U-235 (the IM and 25R types), TRISO-coated fissile particles and BISO-coated fertile particles contain the bred U-233 to be recovered. The crushing step is designed to reduce the fuel element to fragments no larger than 3/16 in. while breaking a minimal number of SiC-coated, or TRISO, particles. During the fluidized-bed burning step, these SiC-coated particles remain intact so that they may be separated from the product ash in a classification step. The thorium and U-233 are contained in the burner ash, which is directly dissolved. Insoluble material, which will include any TRISO particles that have been mis-classified, is returned to a leaching or washing step that removes soluble materials, including U-233, which may cling to the surface of the SiC shells. The solution from the dissolving and washing steps, shown as clarified liquor in Fig. 4, is treated in an evaporator-stripper for feed adjustment prior to separation in a solvent extraction system. The acid-Thorex solvent extraction flowsheet is planned for separation and purification of the U-233 and thorium.

TRISO particles from the U-235 recycle fuel elements (25R) will contain U-235 that has been cycled through the reactor twice. After two passes through the reactor, the value of the residual U-235 will be overshadowed by the negative effect of the U-236 isotope so it is planned that these particles will be retired. TRISO particles from initial or makeup elements will contain sufficient U-235 that it may be economical to recover the material for recycle. These particles will be cracked or ground and then fed to a fluidized-bed burner for reduction to ash. A similar burn, leach, and clarify flowsheet will be followed to provide feed material for solvent extraction using a Purex-type flowsheet.

While the U-233 recycle (23R) fuel elements will also contain both TRISO- and BISO-coated fuel particles, all the uranium in these fuel elements, which is mostly U-233, will be recycled to the reactor. It will, therefore, not be necessary to classify the particle types in this stream. Because the very small particle diameters will preclude breaking of all TRISO coatings in the presence of larger fragments, a crush, burn, grind, burn sequence is also envisioned for this stream with the simplification of eliminating the classification step. It might be possible to provide only single crushing and burning steps except that experiments indicate that the burning of fine material in fluidized-bed burners is difficult (Refs. 5,6). Separation and purification of U-233 and thorium will be performed using a Thorex-type solvent extraction flowsheet.

## PROCESS REQUIREMENTS AND DEVELOPMENT RESULTS

The design requirements for the various unit operations in the reprocessing flowsheet are based upon the anticipated requirements of a commercial-scale recycle facility. Common requirements of this type will be:

1. Throughput and capacity requirements.

2. All systems should be reliable for operation in a hot cell.

3. All systems should be amenable to remote decontamination and maintenance.

4. All systems should be capable of automatic operation with control from a remote location.

5. All systems must be designed to allow cleanout for material accountability with minimum material holdup.

## SIZE REDUCTION

The purpose of the size reduction system is to reduce full-size HTGR fuel elements to a size suitable for fluidized-bed burning operations. Specific requirements of a size reduction system will be:

1. Capability to reduce full-size HTGR fuel elements to fragments less than 3/16 in. diameter.

2. The product should be near cubical in shape and contain a minimum of very fine material.

3. Containment in a shroud to minimize the spread of contamination to the environment of the operating cell and to permit operation in a $CO_2$ atmosphere.

4. Minimal breakage of TRISO-coated particles and contamination of the product U-233 with U-236.

5. A system capacity of approximately 10 fuel elements/hour.

6. A system pressure below cell ambient pressure.

Utilizing the above design requirements, a conceptual size reduction system has been formulated. This system, called the Uniframe, consists of three major crushers (primary and secondary jaw crushers and a tertiary double-roll crusher) together with an oversize reduction system consisting of a screener and an oversize pulverizer ("centrol" - single-roll eccentric) all combined in a single structure. Figure 5 is a schematic of the Uniframe. The system arrangement combines standard components from conventional crushing equipment in a compact configuration suitable for remote operation in a processing cell.

The choice of crushing mechanisms for each reduction stage was based upon experiments performed on many size reduction equipment types at several manufacturers' sites. These were scoping in nature and lead to the selection of the present system (Refs. 7-9). New and used crushers with the selected mechanisms were purchased, modified, and tested as single stages to verify the process operability of the Uniframe concept. The primary and secondary jaw crushers used in these tests are shown in Fig. 6. A continuous screener unit to process product from the tertiary stage (see Fig. 7) was also installed and tested. The experimental results to date show the process and system concepts selected are adequate to meet the design criteria.

These experiments have confirmed the following:

1. Size reduction to less than 3/16 in.

2. The presence of sufficient oversize (i.e., >3/16 in.) to warrant a screener-oversize pulverizer system.

3. Approximately cubical and predictable product size/shape.

4. Acceptably low TRISO particle breakage.

5. Acceptable quantities of fine material.

6. Capacity of at least 10 fuel elements/hour.

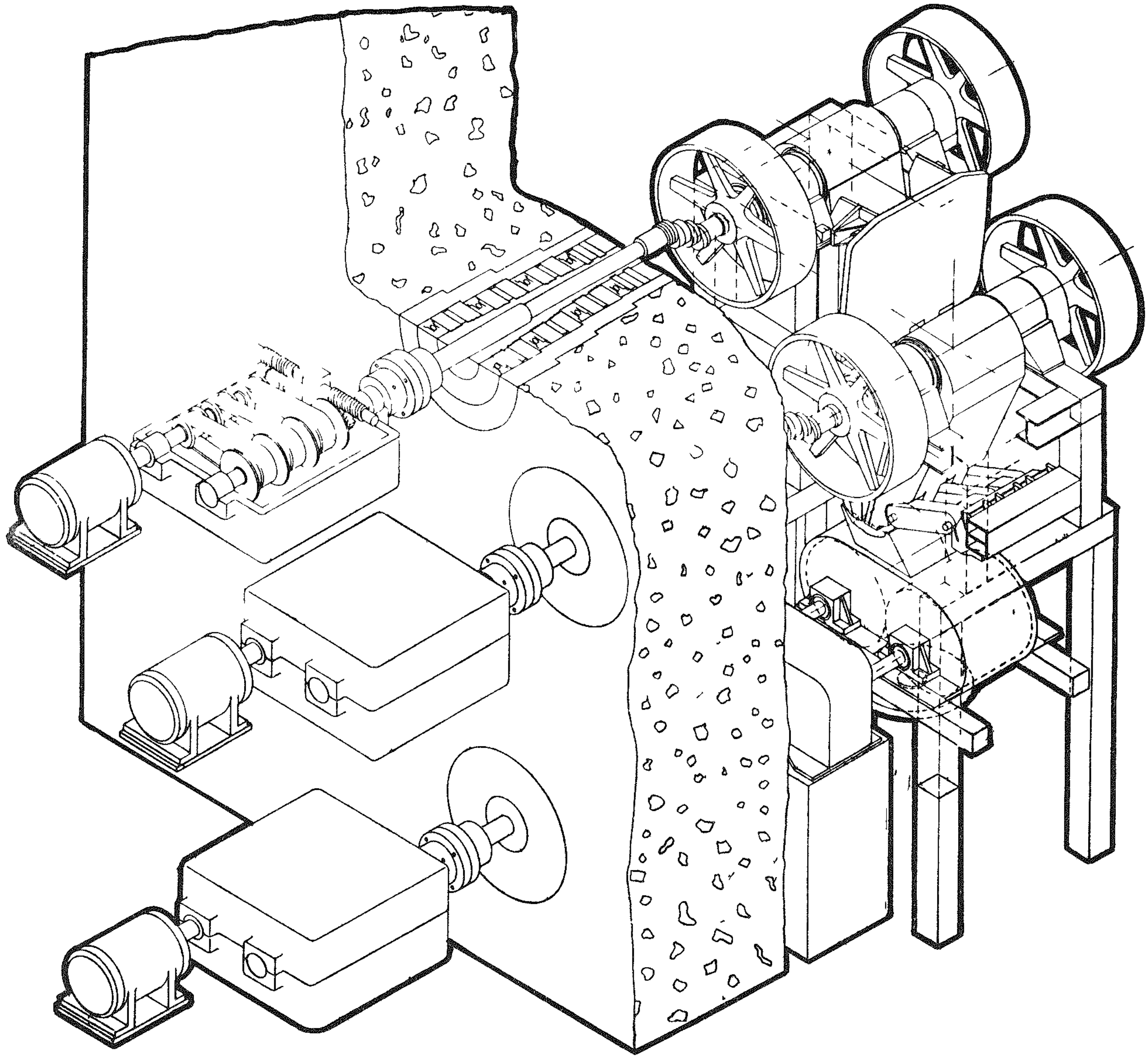

Fig. 5. Uniframe general arrangement

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Fig. 6. Primary and secondary jaw crushers

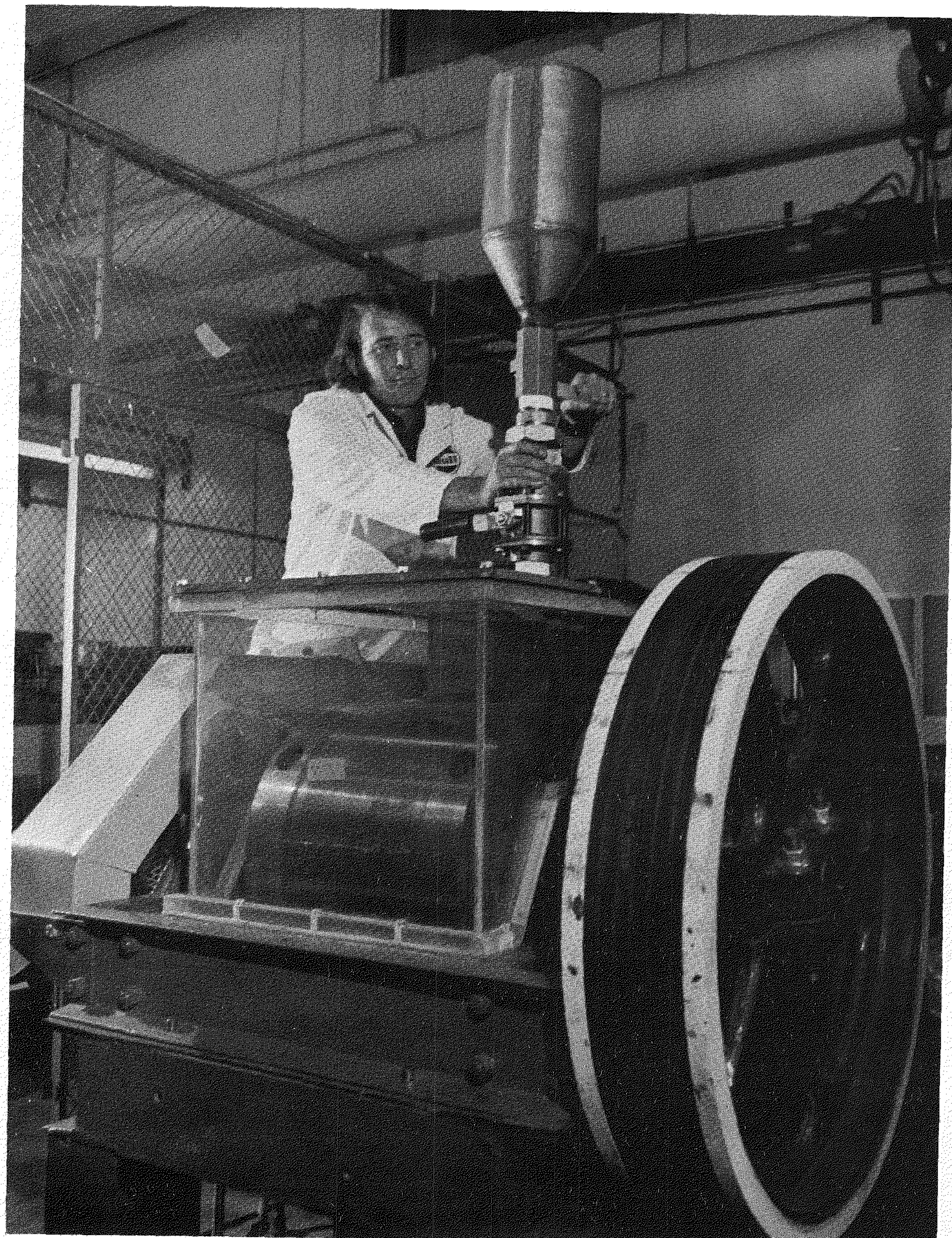

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Fig. 7. Tertiary double-roll crusher

7. An overall indication of adaptability to dust containment, reliable operation, and remote maintenance.

Final design (Ref. 10) is now underway on a full-scale, prototypical, integrated Uniframe size reduction system as shown in Fig. 5, incorporating all design requirements.

## FLUIDIZED-BED BURNING

As discussed previously, the purpose of the fluidized-bed burners is to remove the graphite moderator/fuel coatings via combustion and to oxidize carbide type fuel kernels in preparation for dissolution. A schematic diagram of a fluidized-bed burner configuration for HTGR fuel reprocessing is shown in Fig. 8. Key features of this type of system are indicated.

Two types of fluidized-bed burners are utilized in the head-end reprocessing of HTGR fuel; these are the crushed-fuel-element and crushed-particle burners. These burners differ in operating mode and configuration but have similar design requirements.

Design requirements for the fluidized-bed burners are based upon process feasibility considerations and anticipated requirements of a commercial-scale recycle facility. The following specific requirements for the burner systems have been identified:

1. The crushed-fuel-element burner must be capable of burning the equivalent of one full-size HTGR fuel element/hour to less than 5% burnable carbon in the product stream.

2. The crushed-particle burner must be capable of burning the product streams from the crushed-fuel-element burner after classification and subsequent particle crushing (where required) to less than 1% carbon and/or carbides on a capacity-matched basis.

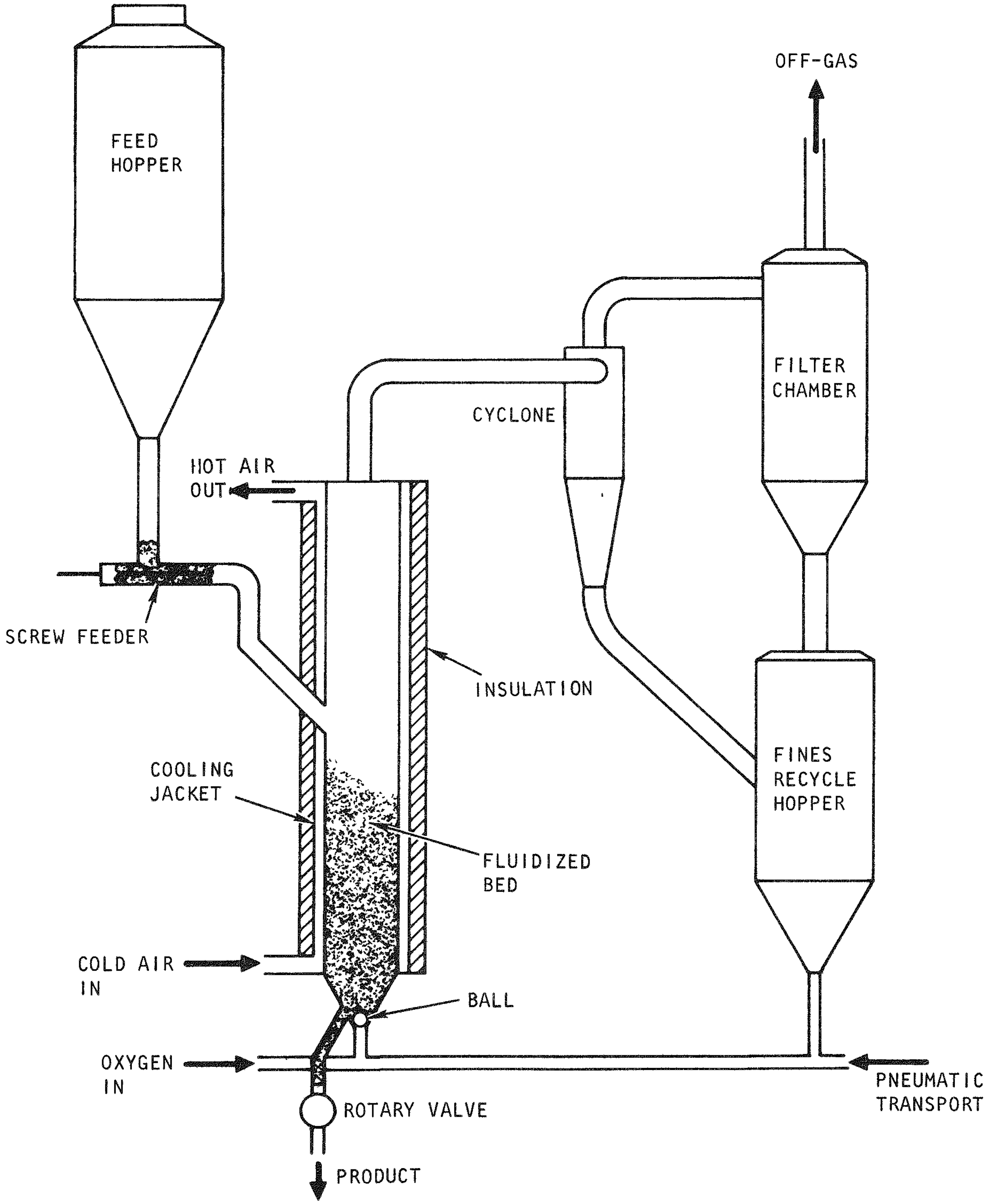


Fig. 8. Crushed-fuel-element burner configuration

3. The crushed-fuel element burner system should minimize TRISO particle breakage.

4. The operating guides for each burner type are indicated in Table 1.

5. The system configuration of the burner should include a means for recycling fines elutriated from the burner in the off-gas stream.

6. The system configuration of the crushed-particle burner should include internal fines recycle (i.e., containment) by utilization of internal filters.

The proposed fluidized-bed burners, which will meet these design requirements, are shown in Figs. 9 and 10 for the crushed-fuel-element burner and crushed-particle burner, respectively. Further discussion of the basis for these designs may be found in Refs. 11-16.

At this time, experiments have been performed in existing smaller scale burners (10 cm and 20 cm diameter crushed-fuel-element burners and 10 cm diameter crushed-particle burner) indicating the design requirements of the burners can be met. Figure 11 shows the base of the experimental 10 cm crushed-fuel-element burners. These experiments have confirmed the following:

1. System cleanout capabilities.

2. Automatic control of burner temperature and product removal.

3. Capacities that scale-up to that required for the demonstration pilot plant.

4. Acceptably low TRISO particle breakage.

5. Acceptable product carbon content.

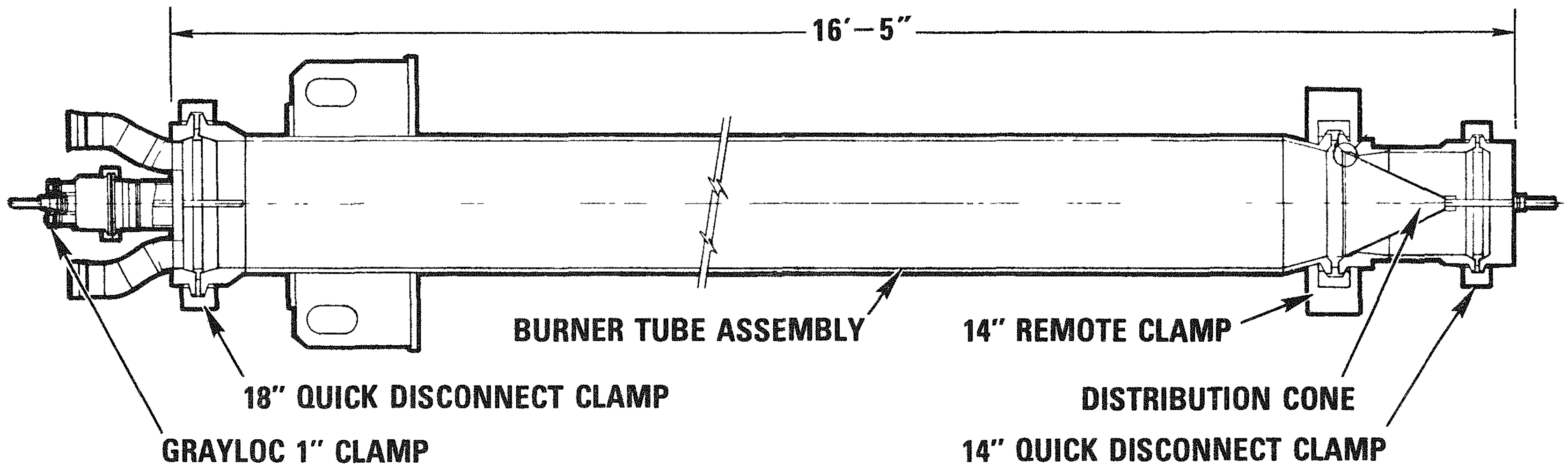


Fig. 9. Crushed-fuel-element fluidized-bed burner

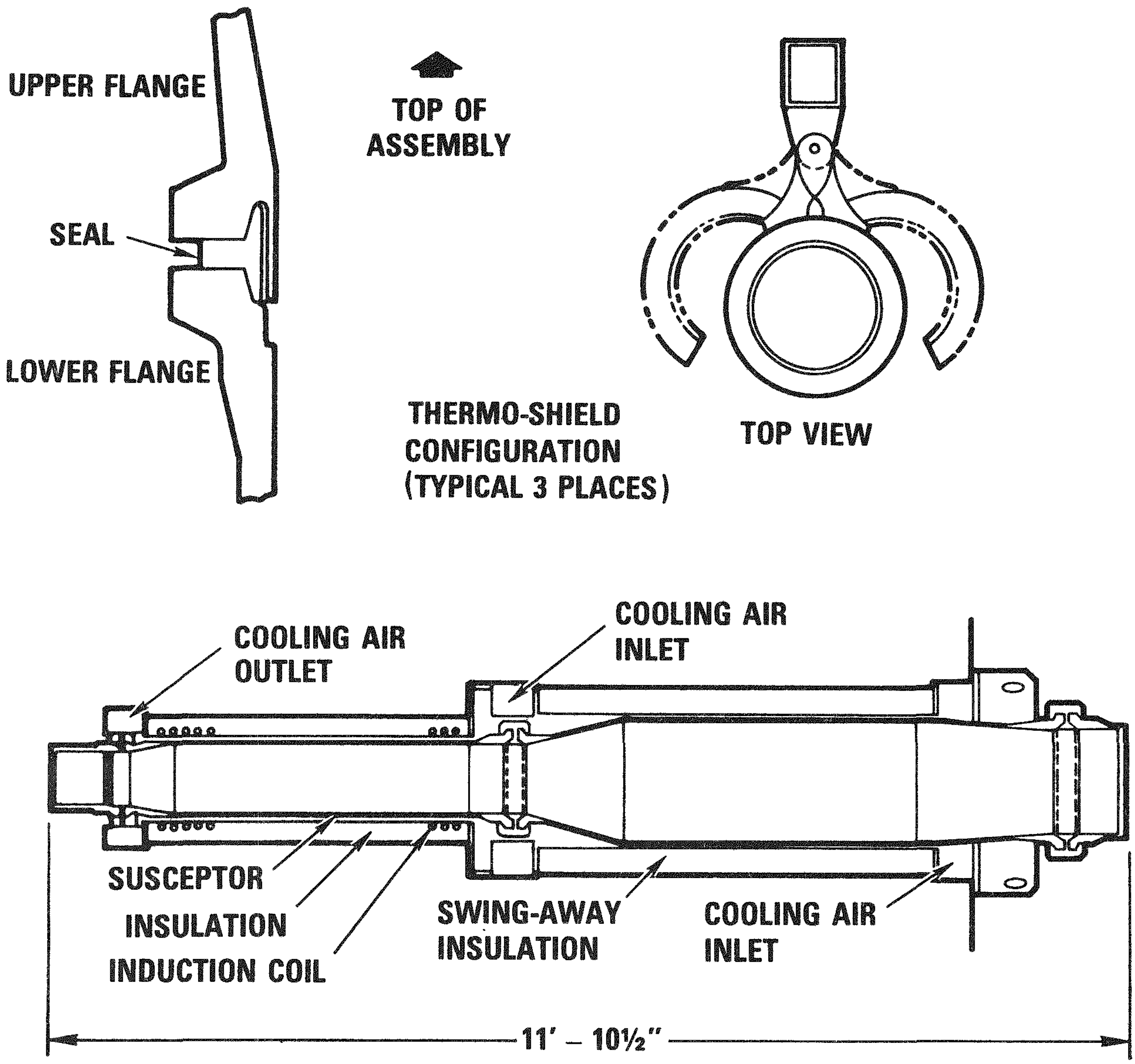


Fig. 10. Crushed-particle fluidized-bed burner

Design requirements that have not been fully verified on these smaller burners include:

1. Decontamination.

2. Remote maintenance characteristics.

3. Automatic feed control.

4. Long-term reliable operation.

5. Induction heater startup in a crushed-fuel-element burner.

6. Conical distributor plate operability on the crushed-fuel-element burner.

7. Long-term operation at 900 ± 50°C.

8. Long-term external fines recycle in the crushed-fuel-element burner.

However, the experiments to date have shown sufficient progress toward meeting the design requirements such that the final design and procurement of a large-scale, prototype burner of each variety is presently underway. These prototypical burners (40 cm diameter crushed-fuel-element burner and 20 cm diameter crushed-particle burner) will be utilized to verify the last of the design requirements; they are shown in Figs. 9 and 10, respectively.

## DISSOLUTION

The purpose of the dissolution system is to dissolve heavy-metal oxides and provide a clarified heavy-metal nitrate solution for subsequent solvent extraction. In addition, any insolubles must be dried and classified.

The dissolution system consists of the three major unit operations of leaching, solid-liquid separation, and insol drying and classification. The specific design requirements of these major unit operations include:

1. Minimal TRISO particle breakage.

2. Operating criteria as presented in Table 2.

The proposed unit operations that will meet these design criteria are gas-sparged tank-type leachers, a continuous solid-bowl centrifuge, and a fluidized-bed dryer-classifier.

At this time, experiments have been performed in two small leachers (13.5 cm and 20 cm in diameter) and a small batch centrifuge. Figure 12 is a photo of this experimental leaching system. Additional testing in glassware leachers and a vendor's test on a continuous centrifuge were also performed. Early experimental testing on continuous dissolvers and alternate solid-liquid separation methods aided in the early selection of the unit processes. These experiments have confirmed and verified the design requirements given previously with the exception of the demonstration of:

1. The insol dryer-classifier.

2. The continuous solid-bowl centrifuge using representative leacher product for long periods; a small batch centrifuge has, however, been demonstrated.

3. Adequate decontamination.

4. Remote maintenance.

5. Long-term reliability.

However, the experiments and methods of testing have shown sufficient promise that design of equipment is proceeding based upon these unit operations. A

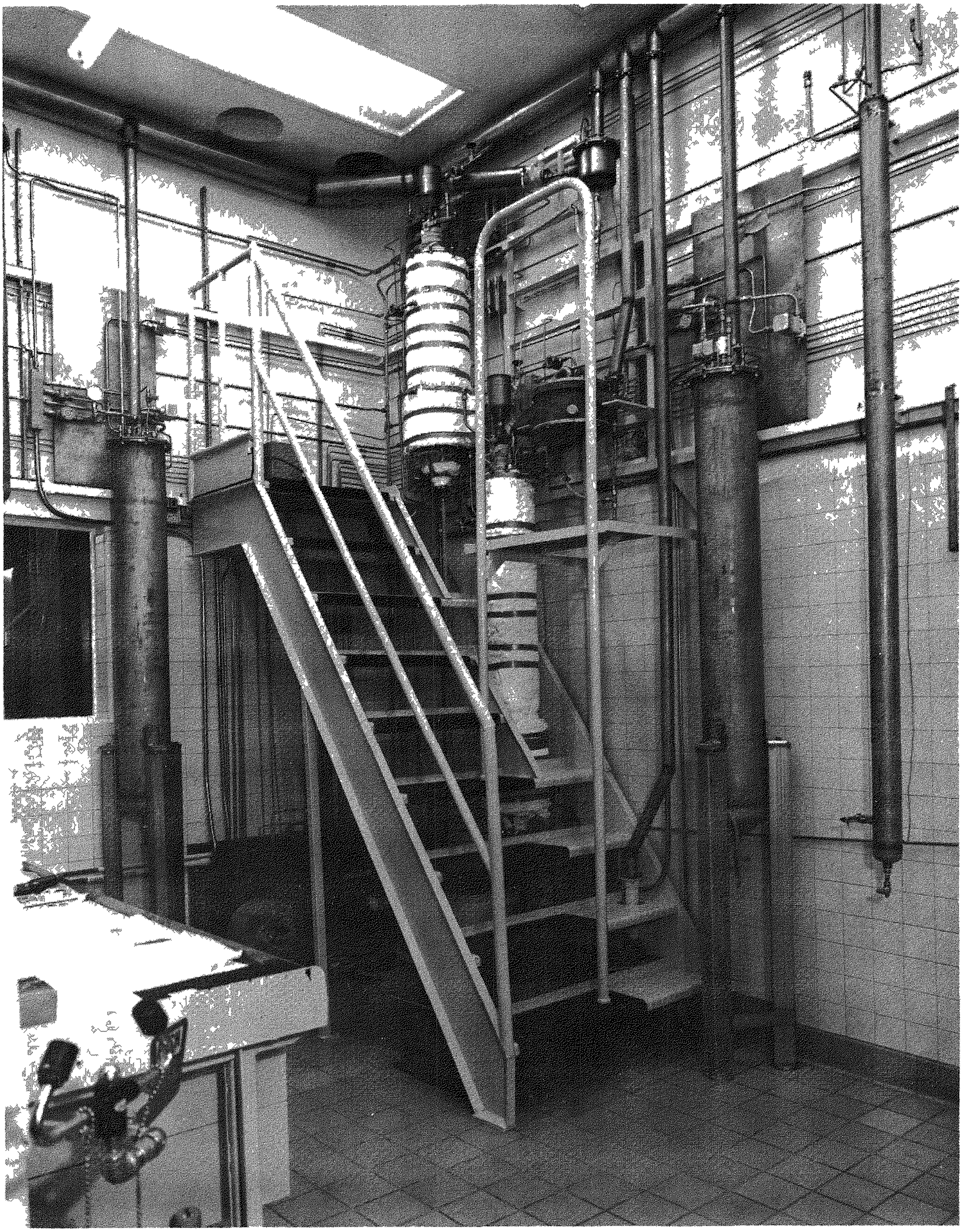

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Fig. 12. Experimental leaching system

TABLE 2
OPERATING CRITERIA - LEACHING SYSTEM

| | Leachers | Solid-Liquid Separation | Insol/Dryer/ Classifier |
|---|---|---|---|
| a. Operating mode | Batch | Batch | Batch |
| b. Startup | Electric heaters | -- | Fluidizing gas preheater |
| c. Feed system | Gravity/pneumatic | Jet eductor | To be defined |
| d. Product removal | Jet eductor | Gravity | Gravity/pneumatic |
| e. Feed/product control | Batching system | Batching system | Batching system |
| f. Temperature | 115 ± 10°C | 35 ± 10°C | 200 ± 20°C |
| g. Pressure | Less than 15 psig with all vessel locations | Negative relative to cell ambient | Less than 15 psig with all vessel locations |
| h. Materials | 304 L SST | 304 L SST all wetted parts | 304 L SST |
| i. Fluid composition | Thorex<br>14 M $HNO_3$<br>0.05 $\underline{M}$ HF<br>0.10 $\underline{M}$ $Al(NO_3)_3$ | -- | Air |

large-scale, prototypical leacher-centrifuge system is being considered to demonstrate that all design requirements can be met.

## SOLVENT EXTRACTION

The purpose of the solvent extraction system is to accept heavy-metal nitrate solutions and separate them into pure nitrate streams of waste fission products, uranium and, in the case of the fertile particle, thorium. Figure 13 depicts a simplified solvent extraction flowsheet. The solvent extraction system described in this section is the acid-Thorex system, which is used to recover and purify U-233 from the thorium-rich fertile fuel stream. The Purex system has similar requirements but is used to recover and purify U-235 from a uranium-rich fissile fuel stream in the absence of thorium. The state of the art of the Purex system is considered to be sufficient so that no cold pilot plant experimental work is needed.

The solvent extraction system consists of a feed adjustment step, pulsed-column solvent extraction contactors, and a product concentrator. Design requirements for the solvent extraction system are those identified for all systems in the reprocessing plant and a requirement that the above liquid system pressure be below cell ambient pressure.

An experimental solvent extraction system has been designed and fabricated. This system was designed to be capable of demonstrating several alternative flowsheets for HTGR fuels. These include acid-Thorex, modified-acid-Thorex, and modified Interim-23. The experimental system consists of an evaporator stripper, five 2-in. i.d. pulse columns (capable of extraction, partition, stripping, and solvent recovery), and a product concentrator.

At this time the experiments have been performed in a two-column system. However, a five-column system with a concentrator and evaporator stripper will be tested in the very near future. The experimental tests to date have been performed to provide pulse column efficiencies and capacities for the previously described flowsheet varieties, and, in addition, flooding data,

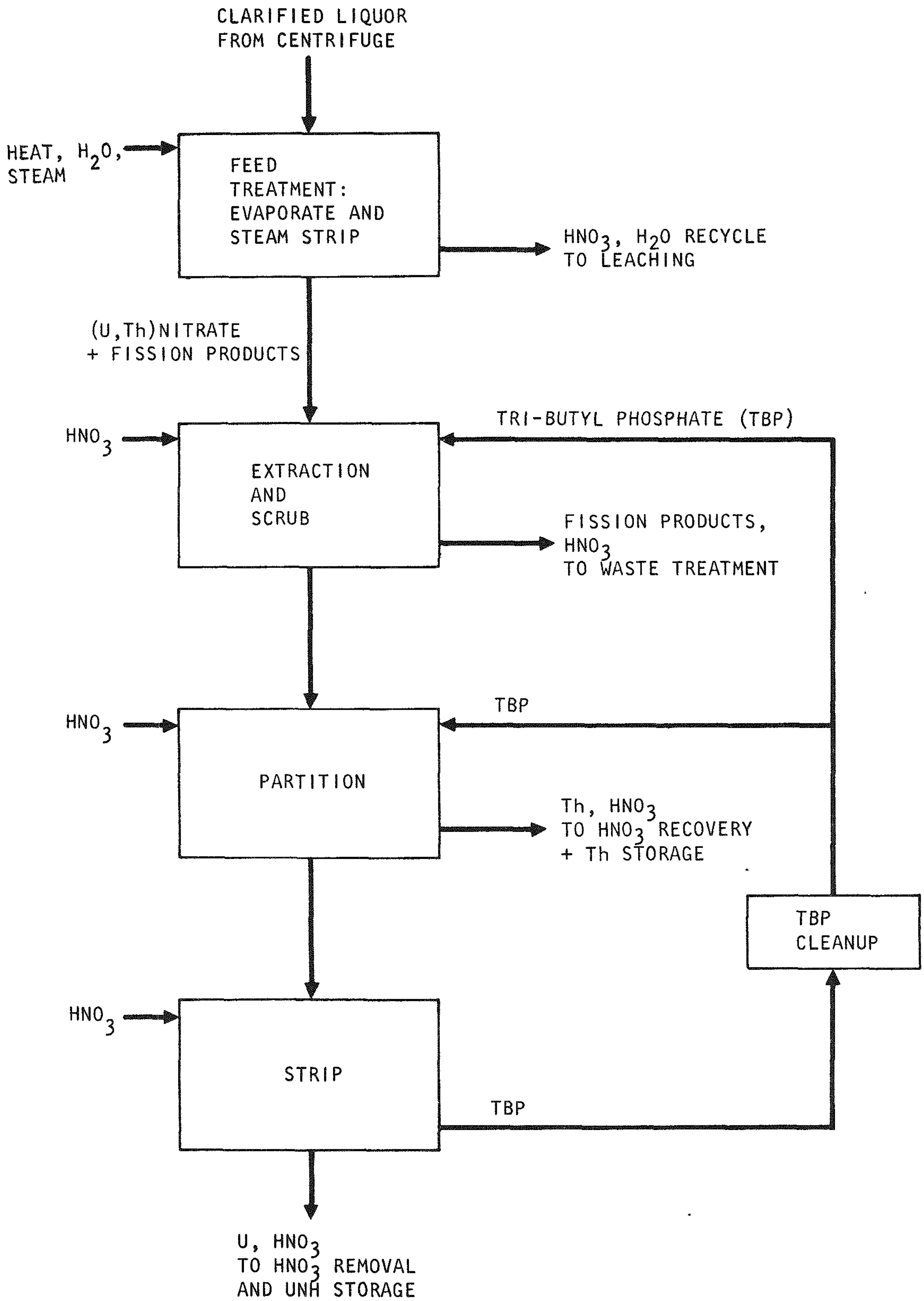


Fig. 13. Solvent extraction flowsheet

height equivalent to a theoretical stage, frequency—amplitude effects on pulse column operation, aqueous—continuous versus organic—continuous operation, nozzle plate free area, and materials effects and overall system performance as related to the ability to scale-up the data to commercially sized equipment.

Experiments to date have verified that the planned solvent extraction installation will provide adequate data to meet the design requirements for a commercial Thorex solvent extraction system.

The experimental crush-burn-leach-extract equipment described in the previous sections is being demonstrated as automatically controlled unit operations from a remote location by utilizing the control room concept shown in Fig. 14.

## WASTE DISPOSAL REQUIREMENTS ASSOCIATED WITH HTGR FUEL REPROCESSING

Several types of radioactive wastes that will result from the reprocessing of irradiated HTGR fuel are:

1. High-level liquid waste resulting from the concentration of the aqueous waste streams from the primary solvent extraction columns.

2. Intermediate-level liquid waste from concentration of solvent wash and equipment decontamination wastes.

3. Silicon carbide hulls from fuel particles.

4. Retired U-235 particles coated with silicon carbide.

5. Zeolites from off-gas cleanup bearing iodine and tritium.

6. Semi-volatile fission products released in the burning operations.

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Fig. 14. Control room concept

7. Krypton released during burning and leaching.

8. Failed contaminated equipment.

With the exception of 4 and 6, all categories have counterparts from light-water reactor (LWR) fuel reprocessing. Disposal methods established for these wastes may be applicable to HTGR reprocessing facilities, although this must be demonstrated. It should be noted that many radioactive wastes from the HTGR flowsheet will be released in gaseous rather than liquid form which, it is hoped, will aid in their separation and recovery. The Thorium Utilization Program includes work on a system known as KALC (Krypton Absorption in Liquid Carbon Dioxide) which is being developed to remove krypton from the $CO_2$ off-gas from the fluidized-bed burners. Work on this process is being pursued even though existing AEC regulations permit the release of krypton to the atmosphere.

The high-level liquid waste from HTGR fuel reprocessing contains fluorides which are used in the Thorex solution required to dissolve thorium oxide. The present AEC program for the development of glasses for ultimate disposal of solid waste does not presently include fluoride-bearing wastes, and there is some question as to whether these glasses will contain fluorides. Preliminary conversations on this and other HTGR waste problems have been held with the AEC Division of Waste Management and Transportation and additional work on HTGR wastes may be started soon.

The silicon carbide hulls in category 3 are analogous to fuel element hulls from LWR fuel. These hulls will also include that fraction of fission products from high burnup fuel that does not dissolve and that will be removed by the centrifuge after the leaching step. The retired U-235 particles removed from the U-235 stream will contain a large fraction of U-236 and some amounts of Np and Pu-238. The silicon carbide hulls are very effective barriers to dissolution; they could serve the purpose of an impermeable coating against release of fission products and actinides. The fact that these coatings have resisted combustion at 900°C and leaching with 14 $\underline{M}$ $HNO_3$ is an indication of their protective capability. Silicon carbide coatings are being investigated for disposal of all radioactive solid wastes. It appears reasonable that a conglomeration of these particles can be put into an acceptable form for ultimate disposal.

As the process technology for the reprocessing of HTGR fuels is becoming better defined, the need for better definition of waste forms and disposal requirements is being recognized. An aggressive effort to investigate these problems well in advance of actual construction of large-scale HTGR fuel reprocessing facilities is clearly needed.

ACKNOWLEDGMENT

The work described herein is being performed at General Atomic Company by a number of lead engineers and individual contributors; the authors of this paper are the Program Manager and Task Leader for the Thorium Utilization Program at General Atomic Company. The work of these individuals is gratefully acknowledged and we regret that space does not permit listing all of them except by reports used as references.

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