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SPENT FUEL SHIPPING, REPROCESSING, AND RECYCLE FABRICATION IN THE HTGR FUEL CYCLE

by

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December 15, 1972

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ABSTRACT

The High Temperature Gas Cooled Reactor (HTGR) fuel recycle operation is described. The description includes the HTGR spent fuel shipping system and the proposed method of reprocessing the spent fuel to recover the bred U-233 and U-235. The process for refabricating the recovered fuel into recycle fuel is also discussed.

INTRODUCTION

The High Temperature Gas Cooled Reactor (HTGR), as developed at Gulf General Atomic, is a helium-cooled graphite-moderated reactor. The fuel in an HTGR consists of fissile microsphere particles containing U-235, recycle microsphere particles containing U-233, and thorium fertile particles contained in a hexagonal fuel element shown in Figure 1. The HTGR fuel recycle operation consists of shipping spent fuel to a recycle facility, reprocessing the fuel to recover the U-233 and U-235, refabricating the U-233 and U-235 into recycle fuel, shipping the refabricated fuel from the recycle facility to the reactor, and ultimately storing the radioactive fission product wastes.

In the reprocessing portion of the fuel recycle facility, the spent fuel is received and temporarily stored. The total decay time prior to reprocessing will permit essentially complete decay of the Pa-233 to U-233, about 120 days. The fuel reprocessing sequence starts with the head-end operation, where the fuel in the HTGR fuel element is separated from the graphite body by fluidized bed burning. Subsequent head-end operations separate particles containing U-235 from ash containing U-233, thorium, and fission products. The metal oxide ash is dissolved to create a solution of uranium, thorium, and fission products; the silicon carbide coated U-235 remains as a residue. The U-235 is separated mechanically and the uranium and thorium are individually recovered from the fission products. The recovered U-233 and thorium are stored for reuse as fuel. The radioactive wastes are disposed of in appropriate storage facilities.

The fuel refabrication portion of the recycle facility converts the U-233 solution received from the reprocessing plant to coated fuel particles. The fissile U-233 particles are blended with coated thorium particles, formed into fuel rods, and assembled into graphite blocks to form the recycle fuel element. Following thorough inspection and temporary storage, the fuel is shipped to the reactor.

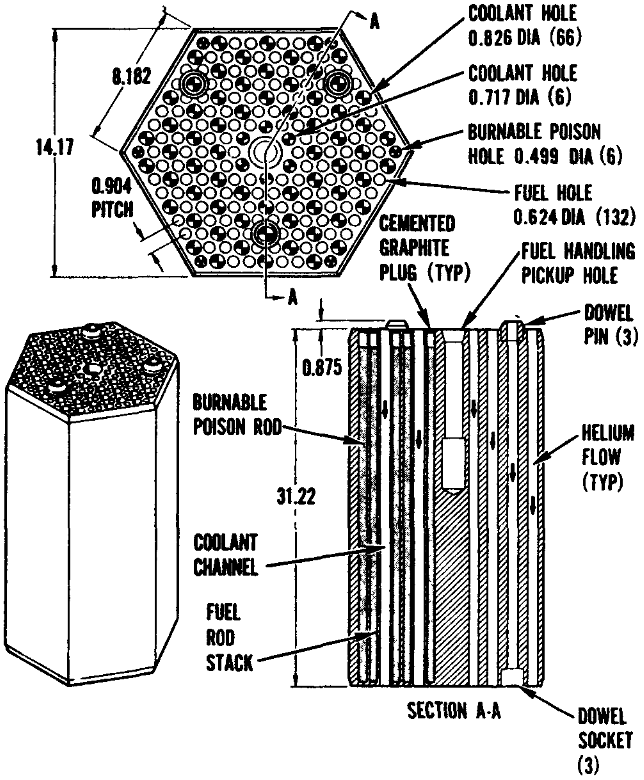

Fig. 1 HTGR hexagonal fuel element

HTGR FUEL DESIGN RELATED TO RECYCLE OPERATIONS

The HTGR fuel element has a number of design features that are of particular significance in its shipping, reprocessing, and refabrication. These are:

1. The moderator (graphite) is an integral part of the fuel element and represents the majority of the material shipped. A large part of the head-end reprocessing operation is devoted to disposal of the graphite.
2. The fuel material consists of small, individual spherical kernels of thorium and uranium carbide or oxide covered with multiple layers of impervious coatings and formed into fuel rods. These particle coatings, shown in Figure 2, must be removed in

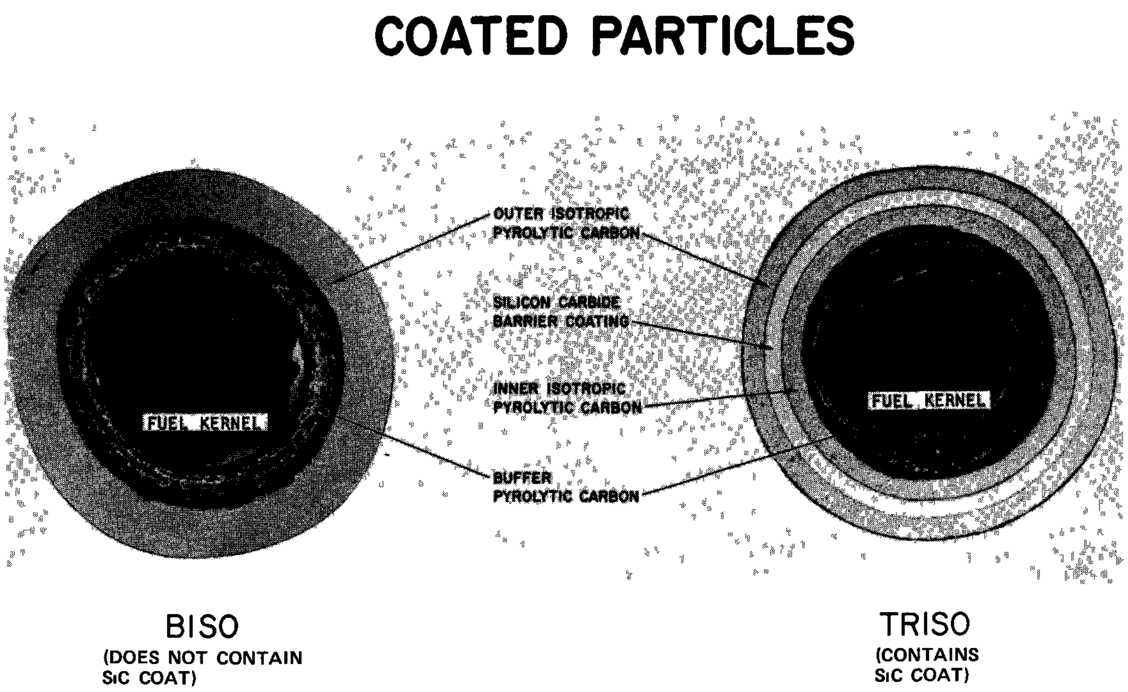

Fig. 2 HTGR fuel particles. Diameter 540 microns

the head-end operations in order to recover the uranium and thorium during subsequent processing operations.

3. In the thorium-uranium cycle, the fertile material is thorium, and the fissile material is U-235 and bred U-233. The high burn-up of the U-235 results in a high U-236 content mixed with the discharged U-235. It is desirable to recycle the uranium containing this U-235 and U-236 for only one additional cycle because of U-236 poisoning (1). Since the uranium isotopes are chemically inseparable, it is necessary that the bred U-233 and thorium be physically separated from the U-235. This is accomplished by initial appropriate sizing of the fuel particles and by inclusion of a SiC coat on all U-235 containing particles.
4. The thorium and U-233 contain the relatively short half-life isotopes Th-228 and U-232, which have daughters that decay with the release of energetic gamma radiation. The presence of the daughters of U-232 in U-233 necessitates remote shielded refabrication and shielded shipping of the refabricated fuel.

HTGR SPENT FUEL SHIPPING SYSTEM

The HTGR spent fuel shipping system provides the method and equipment required for removing and transporting spent fuel blocks from a nuclear electric generating plant to a storage and/or reprocessing plant. The same equipment is employed for transporting refabricated HTGR fuel blocks to the reactor plant. (Refabricated fuel requires 65% to 75% as much gamma shielding as 100-day-old spent fuel.)

The HTGR spent fuel shipping system equipment is designed to satisfy all federal regulations of the U.S. AEC and U.S. DOT. In addition, all applicable state laws and requirements of the Association of American Railroads for unrestricted interchange service are satisfied.

The shipping package is based on "double containment," and is pictured in Figure 3. Four spent fuel blocks in a spent fuel storage can are placed into a spent fuel shipping container. This container is sealed with a bolt-on lid incorporating both an elastomer and metallic O-ring seals. The shipping container provides the "primary" containment for the system. Twelve (12) spent fuel shipping containers are loaded into one spent fuel shipping cask. With the lid in place and sealed, the spent fuel shipping cask acts as the "secondary" containment vessel. The shipping cask provides the shielding, maintains the fuel elements in a subcritical condition, ensures adequate cooling of the fuel elements, and guarantees through a sound structural design that these functions can be maintained at all times.

In operation the shipping containers are loaded at the reactor, leak-rate tested, and then placed in temporary storage in an area adjacent to the fuel shipping port. When the cask and railroad car are in position, the spent fuel shipping containers are lowered through the loading port and shielding collar into the cask. The shielding collar prevents radiation streaming into the vehicle bay and isolates the exterior surfaces of the shipping cask from radioactive contamination. The cask is not removed from the railroad car.

The fuel shipping cask structure is fabricated of stainless steel. Depleted uranium is used for gamma shielding between the inner and outer walls. In three places, between the outer wall and the depleted uranium shielding, aluminum filler material

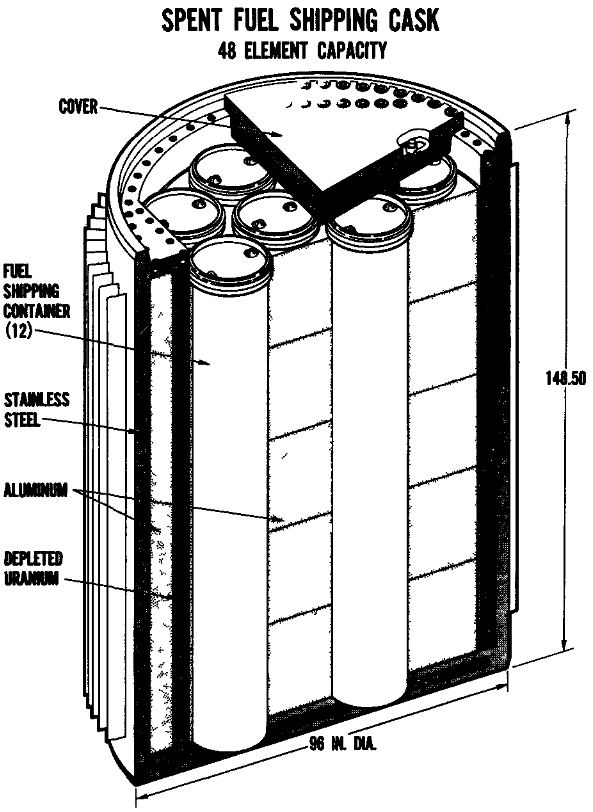

Fig. 3 HTGR spent fuel shipping cask

is used to improve heat transfer. Figure 3 illustrates the arrangement of these materials. The fuel basket, which supports the fuel shipping containers, is fabricated from an aluminum-cadmium alloy. This ensures subcriticality as well as facilitating conduction of the decay heat from the shipping containers to the cask wall.

The fuel shipping cask is transported on a special-purpose railroad car. In addition to the structures required to support the cask with a loaded weight of 110-115 tons, a personnel barrier encloses the cask to prevent it from being handled by unauthorized personnel. The gross weight of the railroad shipping unit is estimated at 370,000 lb.

When refabricated fuel blocks are being shipped, the fuel storage cans are not used. Instead three (3) fuel blocks are loaded into a fuel shipping container with protective packaging material between the container and fuel block, and between each fuel block.

By providing proper standardized interface equipment at all large HTGR plants and at the fuel reprocessing and fuel refabrication plants, the fuel shipping equipment can be standardized. Lifting, handling, and decontamination of the large, heavy cask are eliminated. This significantly reduces the cask loading and unloading time.

REPROCESSING

Head-end reprocessing for HTGR fuel (Figure 4) consists of a crush-burn-leach process (2) replacing the shear, leach process for LWR fuel. Fuel element size reduction is the first step in head-end reprocessing. Two major criteria governing this step are: (1) the fuel must be crushed to a suitable size for maintaining fluidization quality in the fluidized bed burners, and (2) the crushing system should minimize the fuel particle breakage to prevent undesirable crossover of fissile and fertile product uranium.

A three-stage crushing system has been adopted for the reprocessing plant, based on the experimental testing of commercially available equipment using full-sized fuel elements. This crushing system is presently being tested.

Primary reduction is done in a large, overhead eccentric jaw crusher; secondary reduction is performed in a small, overhead eccentric jaw crusher; and tertiary crushing is done in a double-roll crusher. The tertiary crusher product, nominally minus 3/16 in., is pneumatically conveyed to the fluidized bed burners.

Crushed fuel is fed to the top or base of a continuous, exothermic* fluidized bed burner by a pneumatic feeder. The feed rate is automatically controlled by the off-gas carbon monoxide concentration, which has been shown to be proportional to the graphite surface area exposed in the bed.

Both the crushed graphite and the silicon carbide coated fissile particles serve as the fluidizing media. The heat generated by burning is removed by forced-air cooling in a clamshell jacket surrounding the burner and an off-gas heat exchanger. Fluidizing gas fed to the burner is oxygen with a small amount of inert gas (e.g., CO_2, N_2) and the flow is automatically controlled to maintain the bed temperature. The burner product removal is automatically controlled by the bed pressure drop which is proportional to the bed weight and level.

The burner off-gas with entrained fines is passed through a cyclone and a sintered metal filter for fines removal, before being cooled and proceeding to off-gas treatment. Off-gas treatment will remove noble gases by means of the KALC or another process (2A) before release to the environment. Fines are presently being recycled to the burner in our experimental program.

If the exothermic burner is operated with top feed and no fines recycle, the elutriated fines from the exothermic burner (both TRISO/TRISO and TRISO/BISO flowsheets) are also added to the feed stream for the last burning step. This mixture now constitutes the feed material for the endothermic** fluidized bed burner.

The exothermic burner product is fed to a batch operated endothermic fluidized bed burner where the remaining graphite is burned and the thorium and uranium oxide kernels are exposed. The silicon carbide coated fissile particles serve as the inert fluidizing media. The feed stream to the endothermic burner will not sustain exothermic burning to the low carbon level required in the subsequent processing steps. The burning in the endothermic burner, therefore, proceeds from exothermic conditions, with the heat removed from a clamshell surrounding the burner, to endothermic conditions with heat supplied by resistance heaters located in the clamshell. The off-gas from the burner will be treated identically to that from the exothermic burner. The product from the endothermic burner will be pneumatically conveyed to the leaching system.

The thorium and uranium oxides are dissolved in acid (13 $\underline{M}$ HNO_3 - 0.05 $\underline{M}$ HF - 0.06 $\underline{M}$ $Al(NO_3)_3$) in a steam jacketed cylindrical vessel with gas sparge

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*The term exothermic is used to describe the burner which generates sufficient heat to maintain operating temperature.

**The term endothermic is used to describe the burner which requires heat input from a furnace to maintain operating temperature.

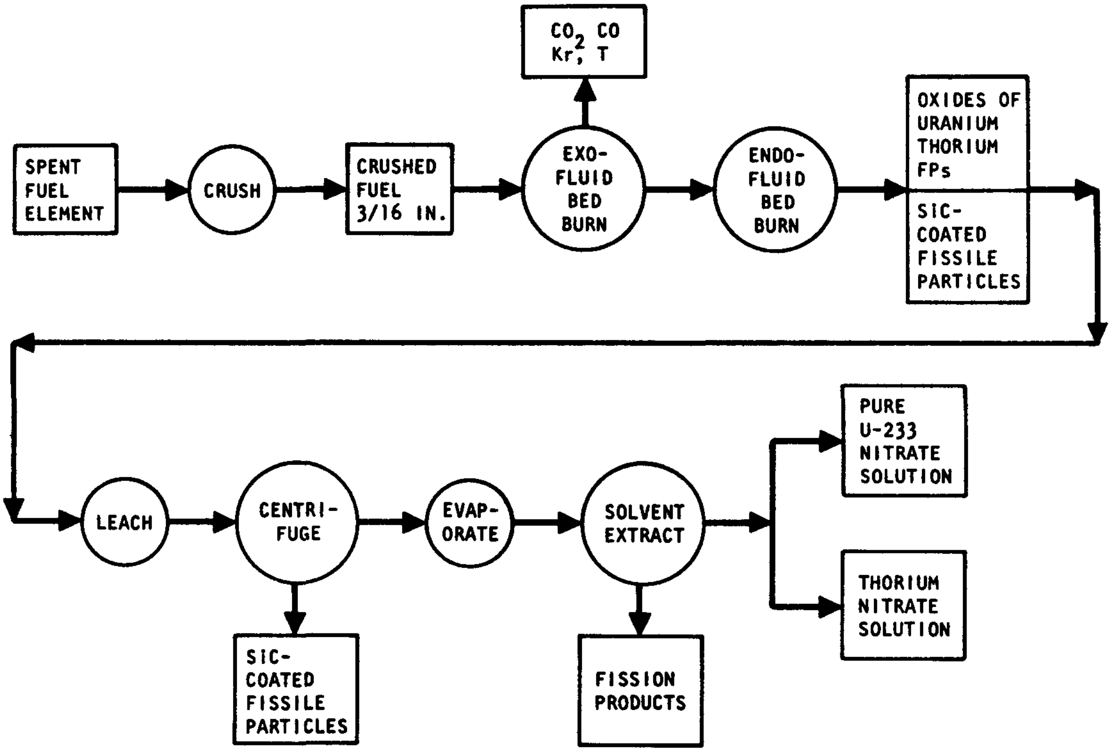

Fig. 4 Simplified reprocessing flow diagram for TRISO/BISO feed

mixing. This leaching vessel will be run as a refluxing, batch leacher.

The insoluble silicon coated fissile particles, and unburned carbon, must be separated from the mother liquor before the solution can be fed to the solvent extraction system for uranium purification and thorium recovery. A centrifugal separator will receive the entire slurry from the leacher. Solids retained on the centrifuge screen will be washed with fresh leach solution which would become the leach solution for the next batch of solids from the endothermic burner. The washed solids from the leacher are then air-dried and transferred to a screen classifier where the fissile particles are separated from the silicon carbide hulls. The waste solids are processed as wastes and the fissile particles are stored for later processing similar to the fertile particles to recover and purify the uranium.

The clarified leacher solutionn will be evaporated and steam stripped to an acid deficient condition for feed to the extraction process (3,4) During the evaporation and stripping phase, zirconium oxide hydrate will precipitate (5,6). The zirconium oxide hydrate will be removed by centrifuging and washed. The wash solution will be added to the feed to the extraction process and the solution will be adjusted to a thorium concentration of about 1.5 molar.

The solvent extraction process is used for the decontamination and purification of the U-233 and thorium and for the separation of the U-233 and thorium from each other. Three solvent extraction cycles are used. In the first cycle, the uranium and thorium are co-decontaminated, using a 30% TBP solution. The addition of a reductant to the extraction contactor scrub stream removes the small amount of plutonium present. In the second cycle, the uranium and thorium are separated, by utilizing the inherently greater solubility of the uranium in the solvent relative to that of thorium. The solvent has a low (3% to 5%) TBP content. By properly controlling the extraction contactor acidity and the relative flow rates of the solvent, feed, and scrub streams, only the uranium is extracted. The thorium remains in the aqueous raffinate stream. The U-233 is processed through an additional solvent extraction cycle for final purification. Following concentration and assay, the U-233 is sent to the HTGR fuel refabrication plant. The partially decontaminated thorium is concentrated and placed in storage.

Various waste streams arise from the head-end and solvent extraction operations. All these streams contain radioactive fission products in widely differing concentrations. Most of the fission products are in the aqueous raffinates from the first cycle solvent extraction contactor. These high-level radioactive waste streams are concentrated by evaporation and then converted to stable solids by calcination. The calcined waste is sealed into high-integrity containers and placed in a water basin for decay. This waste after appropriate decay will be shipped to an approved repository.

REFABRICATION

The refabrication process (Figure 5) in brief consists of taking the 2 molar U-233 uranyl nitrate solution from the reprocessing plant, blending it with thorium nitrate and converting this solution into gelled microspheres by the sol-gel process. These microspheres are then dried, calcined, and given a pyrocarbon coating. These coated particles are blended with coated fertile particles and formed into fuel rods which are assembled into the graphite fuel blocks to form the finished fuel element.

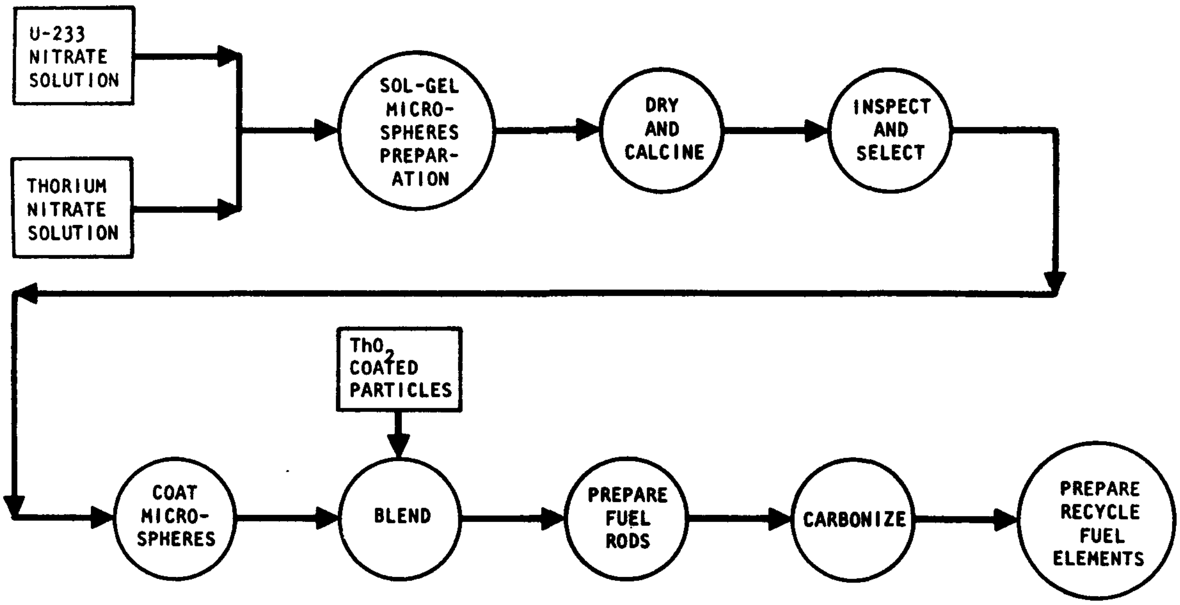

Fig. 5 Simplified refabricating diagram

The sol-gel process for forming microspheres has been developed at Oak Ridge (7). The feed to the sol-gel process in the refabrication plant consists of a blend of the recovered U-233 uranyl nitrate solution and thorium nitrate. This nitrate solution is converted to a hydrosol by extraction of the nitrate ion into a long-chain secondary amine diluted with a normal paraffin hydrocarbon. The extraction is carried out in two stages with a digestion at elevated temperature between stages. About half the nitrate is extracted in the first stage; the digestion step releases additional nitrate, which is removed in the second extraction stage.

The sol produced in the amine extraction process is too dilute, about 0.2 M uranium plus thorium, for use in the following gel formation step. Therefore, the sol is concentrated by evaporation to about 1.5 M uranium. To avoid overheating the sol, which results in thickening or flocculation, the sol is evaporated at reduced pressure.

The concentrated oxide hydrosol is introduced into the sphere-forming system. Conversion of the sol into fuel spheres is accomplished by extracting water from hydrosol drops with a countercurrent flow of an alcohol [2-ethyl-1-hexanol (2EH)]. Surfactants are added to enhance operational control and product yield. The gelled spheres are then dried with superheated steam at about 130°C to remove residual 2EH. During the drying process, shrinkage of about 8% to 10% occurs. The dried particles are then calcined by a carefully controlled heating cycle. The calcined particles are screened and inspected for size, shape, uranium and thorium content, density, and impurities.

The 2EH medium used in dehydrating the sols must be regenerated by removing the water absorbed in the gelling process by single-stage distillation. The distillate condenses into a water phase and an alcohol phase. The water is discharged to waste, and the alcohol is recycled to the gel column.

The inspected uranium thorium oxide microspheres are conveyed to fluidized bed coaters. In these coaters a low-density carbon coating, buffer coating, is deposited on the particles by a high-temperature vapor phase deposition. By changing the coating conditions, the low-density coating is followed by a high-density isotropic carbon coating. This type coating is called a BISO coating. This coating step is followed by a thorough inspection of coating thickness and coating density.

The fissile particles produced in the refabrication plant are blended with coated fertile fuel particles mixed with pitch and graphite flour and formed into fuel rods by pressing techniques. The resulting green rod is carbonized and then heat-treated to produce a strong rod with a high heat conductivity. The rods are inspected for dimensional accuracy and uniformity of fuel loading and then sent to the assembly step.

During assembly the rods are loaded into the fuel holes in the graphite fuel blocks. The fuel holes then have graphite plugs glued in place and inspected and then sent to storage. Following storage the refabricated fuel elements are packed into the shipping containers described before and returned to the reactors in the fuel shipping cask.

Aqueous wastes from the various refabrication process steps are collected in hold tanks, monitored, concentrated by evaporation, packaged in drums, and shipped to an off-site disposal area for burial. Organic wastes, primarily n-dodecane and 2-ethyl-1-hexanol from the sol-gel process, are also produced. The n-dodecane is contacted with an aqueous waste stream to extract uranium and then is combined with the hexanol stream, pumped into absorbent material in drums, and shipped off-site for burial. Uranium waste solutions are returned to the reprocessing plant.

Solid wastes consist primarily of the graphite sleeves used as liners in the carbon coating furnaces and off-specification coated particles. The sleeves contain small amounts of U-233. The graphite liners from the coaters are moved in a transfer container to a crushing machine, crushed, sampled, counted, and then burned. The uranium-containing ash is leached with nitric acid and shipped back to the reprocessing plant for purification.

ACKNOWLEDGMENT

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