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WASTE MANAGEMENT CONSIDERATIONS IN HTGR RECYCLE OPERATIONS

In the recycle of ^{233}U-Th in HTGRs, some different waste management considerations are encountered compared with the LWR recycle. The purpose of this paper is to discuss the types of waste associated with HTGR recycle operations and to describe how some of the wastes will be treated. Certain assumptions have been made regarding effluent treatment requirements in the late 80's.

The types and quantities of wastes resulting from reprocessing HTGR fuel are similar in many respects to that from reprocessing LWR fuel, as one would expect since both use essentially the same solvent extraction process. The principal differences are in the wastes generated at the head-end. A simplified flow diagram for reprocessing is shown in Figure 1. During the burning operation, some of the activation and fission products become volatilized; and these products must be removed prior to release of the off gas. The products of concern are listed in Figure 2. Certain of the fission product metals and oxides are volatilized and condense as very small particles in the cooler portion of the off-gas system. These, along with minute particulates that become entrained during the burning step, may contain an appreciable amount of radioactivity. The gaseous products assumed to require containment are ^{129}I, ^{85}Kr, ^{220}Rn, ^{3}H, and possibly ^{14}C. I-131 is not included in the list because the fuel will be cooled long enough before reprocessing to allow for its decay to a negligible level. The method of collection and containment for each of these materials is different, and a possible flow scheme is shown in Figure 3.

The techniques to be used and removal requirements for semi-volatile fission products that escape the burners have not been fully defined. In addition to the sintered metal filters within the burner system, some sort of condensing device followed by HEPA filtration is a likely choice. Based on

experiments with irradiated HTGR fuel, essentially all volatile fission products are liberated from the uranium carbide fuel while most volatile fission products are retained in the thorium oxide during the burning process. Therefore, about thirty percent of the iodine, krypton and tritium may enter the dissolver. Most of the iodine and all of the krypton will be sparged from the dissolver solutions. Because the dissolver off gas will contain nitric acid and NO_x, the nitric acid will be de-entrained and the NO_x converted to nitrogen and water using ammonia over a zeolite catalyst. To protect the CO/HT oxidizer, the iodine will probably be removed first with a combination of several solid adsorbents. Cadmium- and silver-exchanged zeolites show considerable promise for this application at the present. A bed of cadmium-exchanged zeolite is used to remove the bulk of the elemental iodine and any organic iodides that may be present.

The CO/HT oxidizer will probably use a noble metal impregnated catalyst material similar to that in some of the hydrogen recombiners. Nearly all of the tritium will probably already have been converted to HTO in the burner, but an appreciable amount of CO may be present. The tritiated water will be collected on regenerable molecular sieves. Radon-220 with its 56-second half-life will be delayed until it decays. Solid adsorbents such as molecular sieves or charcoal will be used to accomplish the delay and retain the daughter products which appear to plate-out quite readily.

Krypton and xenon removal will be accomplished by adsorption in liquid CO_2 by the KALC process. Should $^{14}CO_2$ containment be necessary, the most economical approach to retaining the CO_2 from the KALC process appears to be fixation as calcium carbonate.

As indicated earlier, certain assumptions have been made regarding which nuclides will require control and the expected successful development of particular treatment processes. Any changes in these assumptions could affect the arrangement of the indicated treatment systems and the quantities of waste to be disposed.

In Figure 4 the classification of high-level solid wastes is shown. The solidified fission products are similar in form to those from reprocessing LWR fuels. The fluoride added to assist in the dissolution

of the thorium oxide follows the aqueous phase in the solvent extraction cycle to the high-level waste. If vitrification or other technique involving high-temperature operations is used to solidify the high-level waste prior to long-term storage, volatile fluoride compounds may be released resulting in corrosion of off-gas treatment system components. However, the addition of calcium compounds shows promise in stabilizing the fluoride even at temperatures as high as 1100°C.

The silicon carbide hulls are equivalent in purpose to the LWR metal hulls in that they contain the fuel and fission products until release during reprocessing. Fissile particles, in which enough of the ^{235}U has been consumed to make further recovery uneconomical, are retired with the fission products and spent fuel intact. Whether or not these materials will require fixing in an inert medium prior to storage has not yet been determined.

The intermediate-level solid waste (Figure 5) is composed of contaminated failed equipment, highly concentrated long-lived intermediate- and low-level wastes, and some of the HEPA filters. The disposition requirements for these wastes are still being defined. Because of the unique characteristics of ^{85}Kr- and ^{129}I-bearing wastes, special treatment will likely be considered. These characteristics relate to the high heat generation rate and the relatively short half-life (10.8 years) of concentrated ^{85}Kr and the highly concentrated form expected for the ^{129}I wastes and its very long half-life of 16 million years. The storage of tritium could also be placed in a special category if it is collected in a highly concentrated form which is probable in reprocessing HTGR fuels.

The low-level solid waste categories are listed in Figure 6. The first three categories are similar to what is expected from LWR reprocessing: general trash, failed equipment, and HEPA filters. Waste from refabrication operations is included in this classification. The calcium carbonate would result should it become necessary to contain the $^{14}CO_2$ from the burner off gas.

The present HTGR recycle facility concept assumes all liquid wastes will be concentrated and solidified, and any resulting volatile radioactive species collected and contained for storage.

A look at the composition of the waste streams provides a clearer picture of the differences in the wastes from reprocessing HTGR and LWR fuels. The composition of the high-level liquid waste per GWe-year is illustrated in Figure 7. The quantity of the fission products from HTGR fuel is slightly less. Because the HTGR is more efficient than the LWR, one would expect fewer fission products in the HTGR waste. There is a large difference in the quantity of heavy metals because of the higher enrichment of the HTGR fuel. The difference would be much larger were it not for the fertile thorium present in fuel. The quantity of corrosion products is about the same, but there is an appreciable difference in the chemicals necessary to dissolve the fuel. This is because the Thorex solution contains 0.1 M aluminum nitrate and 0.05 M hydrofluoric acid in addition to the nitric acid. It will be necessary to add calcium to prevent the volatilization of fluoride, but this will only add several percent to the total. Therefore, the total quantity of high-level waste will be nearly the same for an equivalent power generation base.

The volume and composition of intermediate-level waste from the two types of fuels are about the same (Figure 8). There are fewer heavy metals in the HTGR intermediate-level liquid waste but more chemicals. The present intentions are to blend the intermediate-level liquid waste with the high-level waste just prior to solidification.

The largest difference in the high-to-intermediate-level waste will be in the quantity of hulls. As shown in Figure 9, there is a much greater quantity of metal hulls from reprocessing LWR fuels than silicon carbide hulls and retired fissile particles from HTGR fuels; however, the latter will contain more radioactivity.

The quantity of low-level waste estimated from HTGR reprocessing is not well defined at this point but is believed to be similar in quantity to that from LWR reprocessing. Should containment of ^{14}C be required, this would add appreciably to the quantity of low-level waste.

Figure 10 shows the comparative off-gas compositions from LWR and HTGR fuel reprocessing. The quantity of noble gases released during

reprocessing of LWR fuels is somewhat higher than that from HTGR fuel but the quantity of ^{85}Kr from HTGR fuel is nearly twice that from LWR fuel. The quantity of total iodine is estimated to be about the same, but the quantity of carbon dioxide is enormously different.

Although not much has been said about the wastes from refabrication operations, this should not be construed to suggest these wastes have been ignored. A simplified refabrication flow diagram is shown in Figure 11. Because of the valuable ^{233}U content in these wastes, as much recycle of the wastes will be done as is practicable. Essentially all of the waste from refabrication will be low level. A large portion of the off-gas treatment will address nonradioactive pollutants but will likely contain radioactive contaminants. These include the perchloroethylene scrubbers used to contain carbon particles from coating operations and organic compounds from kernel and fuel rod carbonization, and caustic scrubbers to contain the hydrochloric acid resulting from the decomposition of methyltrichlorosilane used to deposit the silicon carbide coating on the fuel particles. The wastes from refabrication are in the process of being more clearly defined, and waste treatment and handling procedures will be developed and modified as this work progresses.

Many details regarding how the wastes from an HTGR recycle facility will be treated and managed remain to be resolved pending a better definition of the respective waste products and waste management regulations. The objectives of the waste management portion of the development programs associated with the recycle of ^{233}U-Th include the intent to be responsive to these needs.

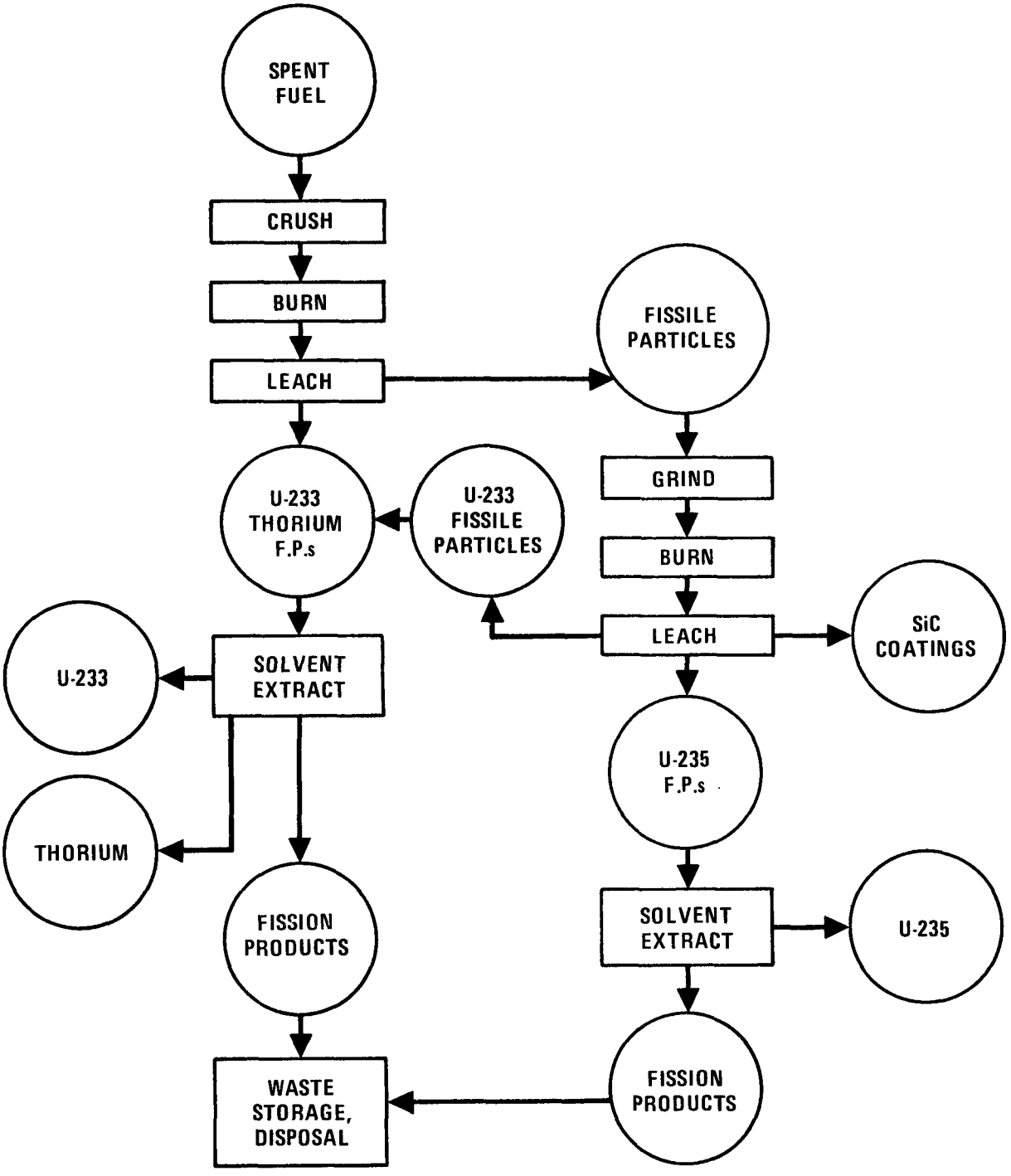

Figure 1. HTGR Fuel Processing

- **FISSION PRODUCT AEROSOLS**
- **IODINE 129**
- **KRYPTON 85**
- **RADON 220**
- **TRITIUM**
- **CARBON 14**

Figure 2. Gaseous Wastes

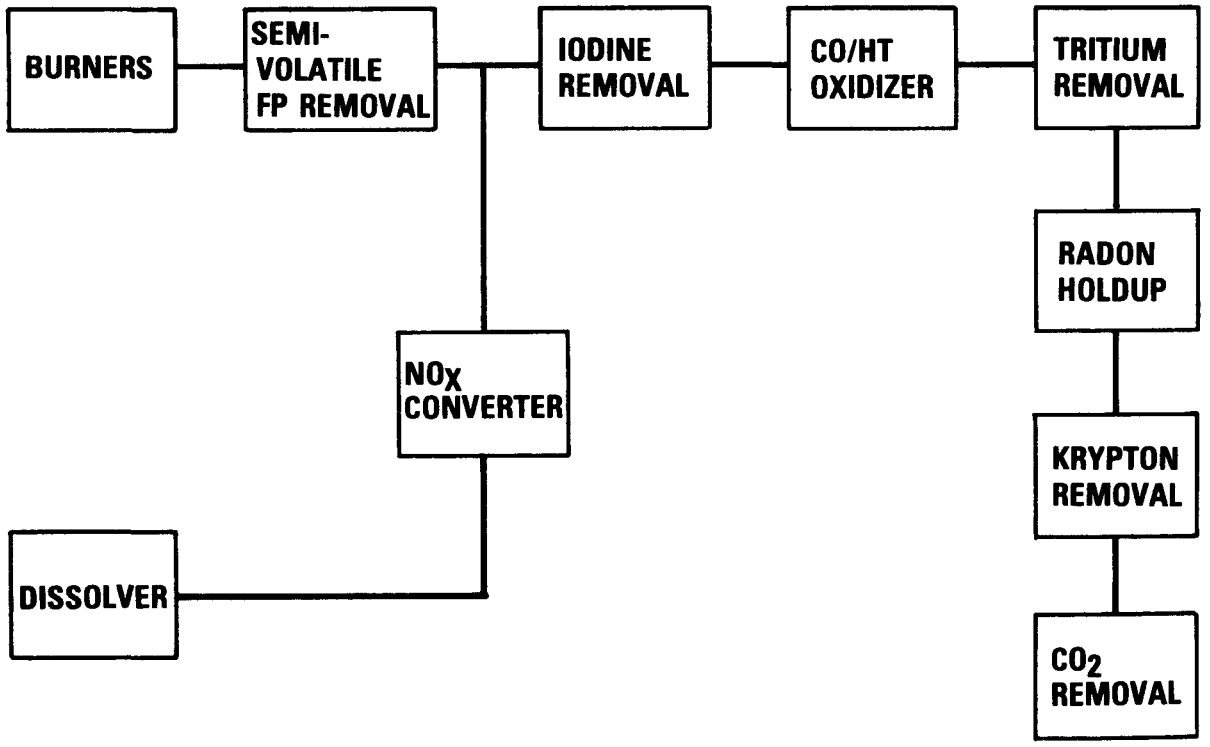

Fig. 3. Burner and Dissolver Off-Gas Treatment System

- **SOLIDIFIED FISSION PRODUCTS**
- **SILICON CARBIDE HULLS**
- **RETIRED FISSILE PARTICLES**

Figure 4. High-Level Solid Wastes

- **FAILED EQUIPMENT**
- **HIGHLY CONCENTRATED, LONG-LIVED LOW-LEVEL WASTES**
  - **IODINE 129**
  - **KRYPTON 85**
  - **TRITIUM**
- **HEPA FILTERS**

Figure 5. Intermediate-Level Solid Waste

- **GENERAL TRASH**
- **FAILED EQUIPMENT**
- **HEPA FILTERS**
- **CALCIUM CARBONATE**

Figure 6. Low-Level Solid Waste

	HTGR (KG)	LWR(KG)
FISSION PRODUCTS	950	970
HEAVY METALS	130	350
CORROSION PRODUCTS	90	80
CHEMICALS	620	370
TOTAL	1,790	1,770

IN SOLUTION WITH 20,000 ℓ (5,000 GAL) OF 1M NITRIC ACID

Figure 7. High-Level Liquid Waste Composition (per $GW_e y$)

	HTGR (KG)	LWR (KG)
HEAVY METALS	26	83
CHEMICALS	470	397
TOTAL	497	480

IN SOLUTION WITH 8,000 ℓ (2,000 GAL) OF 1 M NITRIC ACID

Figure 8. Intermediate-Level Liquid Waste Composition (Per $GW_e y$)

	HTGR (KG)	LWR (KG)
SILICON CARBIDE HULLS	990	—
RETIRED FISSILE PARTICLES	410	—
METAL HULLS	—	11,000
TOTAL	1,400	11,000

Figure 9. Quantity of High-Level Solid Waste ($GW_e y$)

	HTGR (KG)	LWR (KG)
NOBLE GAS	153	194
IODINE	10.5	9.1
CARBON DIOXIDE	400,000	—

Figure 10. Off-Gas Composition (Per $GW_e y$)

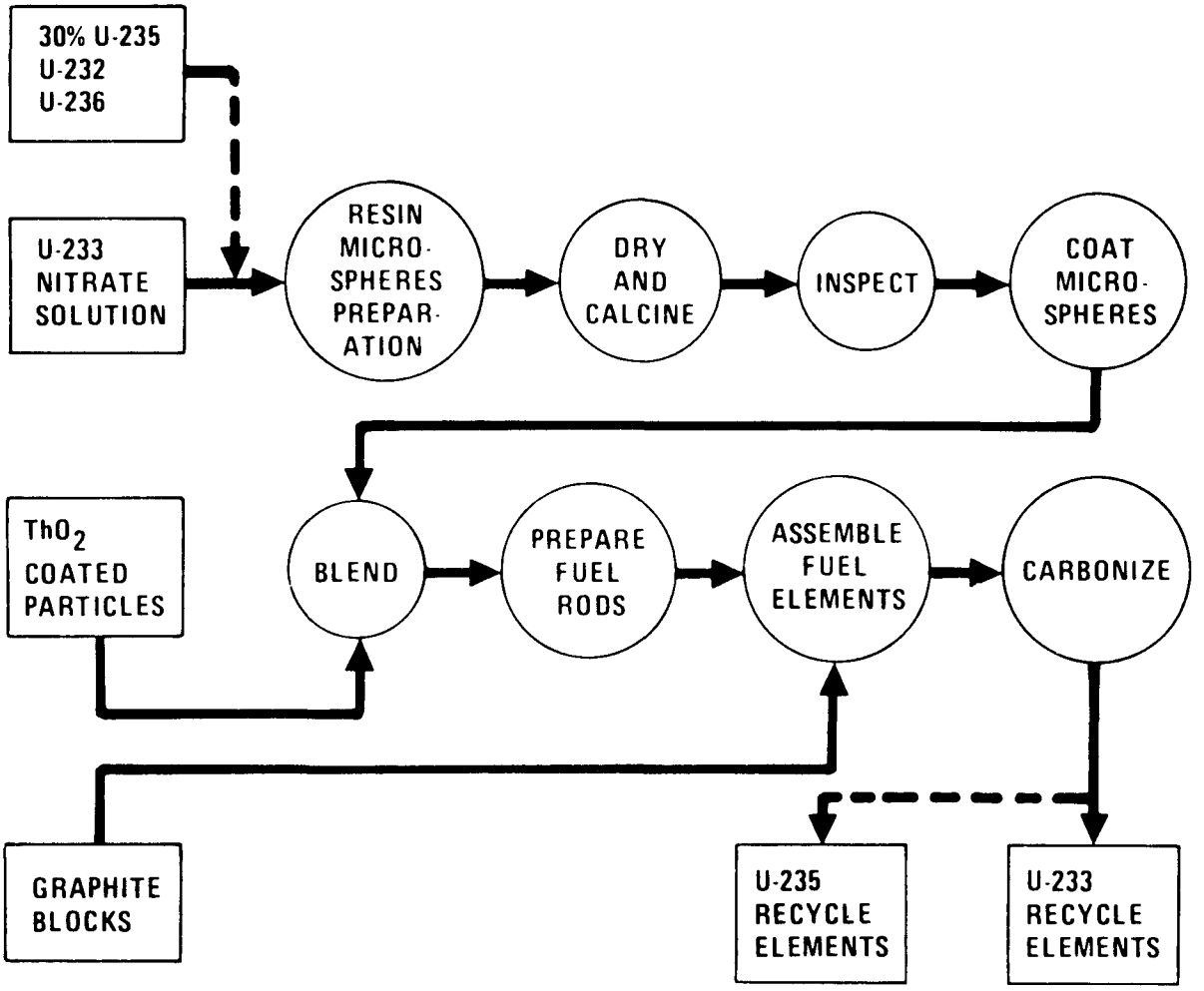

Figure 11. Simplified Refabrication Flow Diagram