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# Characteristics of Radioactive Waste Streams Generated in HTGR Fuel Reprocessing

K. H. Lin

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CHEMICAL TECHNOLOGY DIVISION

CHARACTERISTICS OF RADIOACTIVE WASTE STREAMS GENERATED IN  
HTGR FUEL REPROCESSING

K. H. Lin

JANUARY 1976

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CHARACTERISTICS OF RADIOACTIVE WASTE STREAMS GENERATED IN  
HTGR FUEL REPROCESSING

K. H. Lin

ABSTRACT

This report presents the results of a study concerned with identification and characterization of radioactive waste streams from an HTGR fuel reprocessing plant. Approximate quantities of individual waste streams as well as pertinent characteristics of selected streams have been estimated. Most of the waste streams are unique to HTGR fuel reprocessing. However, waste streams from the solvent extraction system and from the plant facilities do not differ greatly from the corresponding LWR fuel reprocessing wastes.

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INTRODUCTION

The eventual acceptance of the HTGR is inevitably dependent upon the successful closing of the thorium fuel cycle. In this fuel cycle, the major purposes of spent fuel reprocessing are (a) to recover the bred U-233 and unconsumed U-235 from the spent fuel, (b) to convert them into a form amenable to refabrication into recycle fuel elements, and (c) to convert radioactive wastes into forms suitable for safe storage and/or isolation. As a byproduct, thorium is also recovered for possible reuse in the future.

In the course of fuel reprocessing, waste streams of various forms are produced at different process steps which are grouped into three work units, namely, head-end processing, solvent extraction, and off-gas cleanup. Processing and isolation of these waste streams can be considered a separate major operation in fuel reprocessing. Most of the waste streams from the head-end and off-gas cleanup operations are characteristic of HTGR fuel reprocessing, while those from solvent extraction are similar in nature to the corresponding wastes from LWR fuel reprocessing.

The objective of this report is to present the result of a study that deals with identification and characterization of waste streams from various sources in HTGR fuel reprocessing operations. Characterization of waste streams includes their approximate quantities which serve as one of the criteria in determining which of available alternatives should be used in processing of individual waste streams. A previous study<sup>1</sup> of similar nature dealt with the waste handling requirements for a small-scale (~ 5 prismatic fuel elements/day) demonstration plant based on an earlier concept of HTGR fuel reprocessing and refabrication.

The results of this study emphasize waste streams that require further processing prior to disposal or long-term storage. These results will be used to guide subsequent studies concerned with evaluation and development of various potential methods for waste processing and isolation.

#### HTGR FUEL REPROCESSING

In order to facilitate discussion on subjects pertaining to waste streams, a brief description is presented below covering various steps involved in HTGR fuel reprocessing to recover heavy metals (U-233, U-235, and Th). Figure 1 illustrates key process steps, showing flows of fuel material as well as off-gas streams. Semi-volatile, liquid, and solid waste streams are shown in other figures presented elsewhere in this report. These process steps are based on, but not necessarily identical to, the reference flowsheet for an HTGR recycle plant.<sup>2,3</sup> Additional information on a proposed commercial-scale HTGR fuel reprocessing plant may be found in reference 3. Different types of fuel elements (i.e., 25R, 23R and 25W) are defined in Table 1 (p. 11).

##### Crushing, Primary Burning, and Classification

The first step in the fuel reprocessing is to reduce the size of fuel blocks to particles less than ~ 3/16 in. This is accomplished by crushing fuel elements by means of two jaw crushers in series followed by a double-roll crusher (step 1, Fig. 1). The crushed pieces are then transported to a primary fluidized-bed burner where the bulk of the graphite is burned to CO<sub>2</sub> in a gaseous mixture of CO<sub>2</sub> and O<sub>2</sub> (primary burning; step 2). Small

SPENT FUEL ELEMENTS  
(25 R, 23 R, 25 W)\*

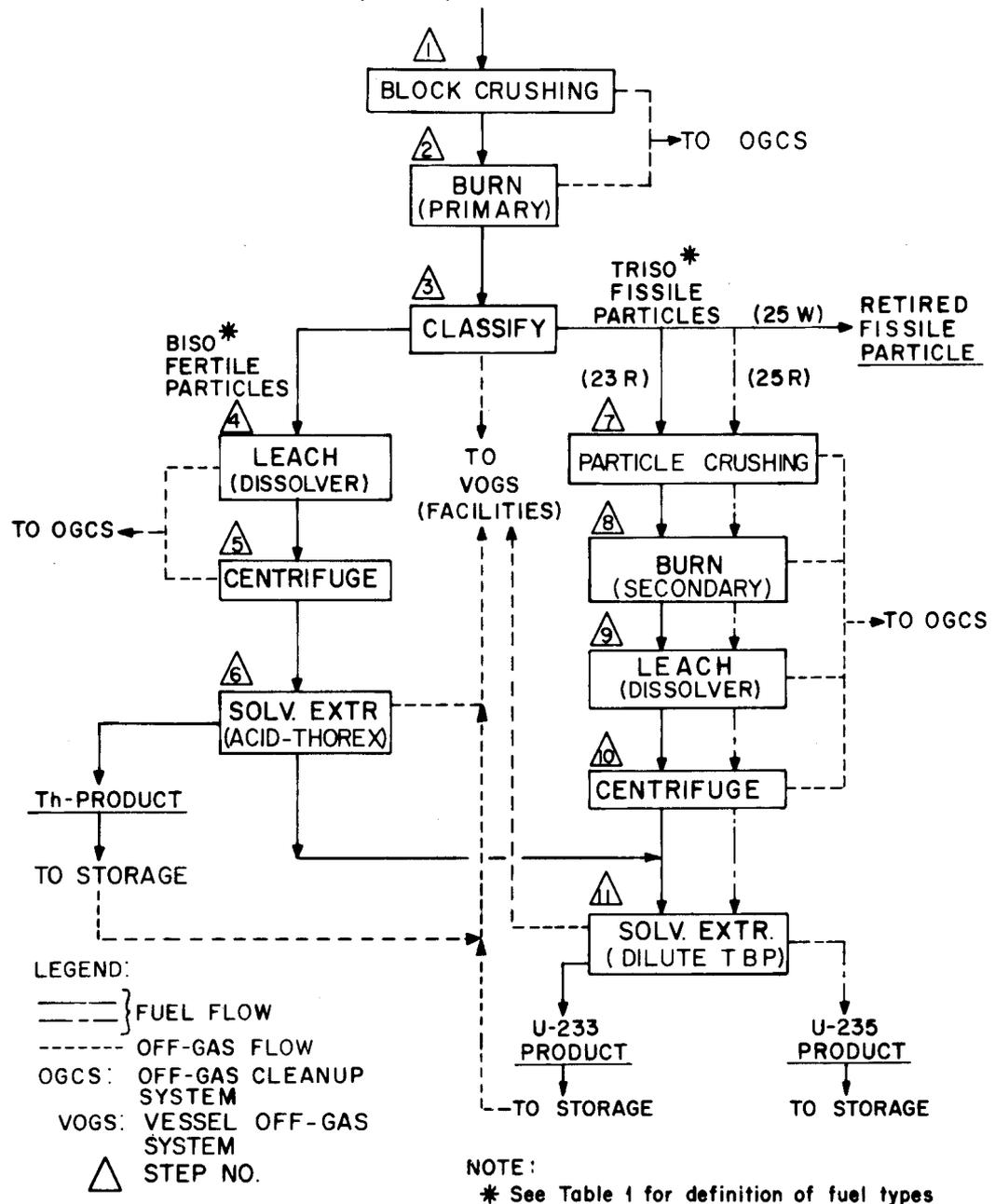


Fig. 1. Schematic Flow Diagram of HTGR Fuel Reprocessing (Head-End and Solvent Extraction Systems).

fractions of fission products, activation products, and transuranics are released during the burning step. The unburned residue, consisting mainly of fuel particles, is separated into fissile and fertile fractions by the air elutriation method (classification; step 3). After the classification, the fissile and fertile particles are processed separately according to the steps described below (steps 4 through 11).

#### Processing of Fertile Particles

Following the classification step, fertile particles (only kernels remain from these BISO particles) are leached in a dissolver by a Thorex solution (13 M  $\text{HNO}_3$ , ~ 0.05 M HF and ~ 0.1 M aluminum nitrate) to dissolve heavy metals and most fission products (step 4). After separation of insoluble residue by a centrifuge (step 5), the clarified leach solution is sent to a denitrator-evaporator for acidity adjustment prior to solvent extraction.

The solvent extraction process consisting of three extraction cycles is employed to recover the fuel values from the leach solution. They are the first Acid-Thorex cycle with partitioning, the second Acid-Thorex cycle for additional purification of thorium (step 6 represents first and second cycles), and the dilute TBP cycle for purification of uranium. The first cycle uses 30% TBP for separating the fission products, thorium and uranium by means of dilute  $\text{HNO}_3$  (0.25 M and 0.01 M acid, respectively, for transfer of Th and U to the aqueous phase). Thirty percent TBP is again the extractant in the second cycle in which thorium solution is further purified by the process steps similar to those of the first cycle. The aqueous raffinate (contains the bulk of fission products) from the first column (1AW) in the first cycle will be processed as the high-level liquid waste. The function of the third cycle is selective extraction of uranium into a dilute TBP solution (~ 5%), thus achieving separation of uranium (U-233 and other isotopes) from residual thorium and fission products (step 11). The uranium is further decontaminated through consecutive scrubbing and stripping operations. Recovery of plutonium may be made in the first cycle, if so desired; otherwise the bulk of plutonium will be in the high-level liquid waste. Thorium in the aqueous raffinate stream resulting from the initial

uranium-thorium separation step is placed in storage after concentration (no further purification for storage). Thus three major streams are produced in the solvent extraction step (steps 6 and 11) -- U-233 product, thorium product, and high-level liquid wastes.

#### Processing of Fissile Particles

Fissile particles from three different types of spent fuel elements (25R, 23R and 25W) are processed separately. Those from 25W elements are placed in the cans for subsequent storage or further processing as wastes. Fissile particles from 25R and from 23R are subject to the same sequence of processing steps (steps 7 through 11), but are processed as two independent streams to preclude crossover of  $^{235}\text{U}$  (with concomitant  $^{236}\text{U}$ ).

The initial step in processing of 25R and 23R fissile particles involves crushing in a roll crusher or jet grinder to break the protective SiC hulls (step 7). This exposes the internal graphite coatings which are burned off by secondary burning (step 8) that follows the grinding. The exposed uranium carbide kernels are converted to the oxide form during burning. Secondary burning also releases a significant portion of gaseous and semi-volatile radioactive products. A separate step ("thermal soaking") may be required immediately following secondary burning to drive off tritium from the burner ash. Since there is no means for removal of tritium beyond the secondary burning step, this process step is highly desirable in order to drive out tritium prior to the aqueous processing steps. In the leaching step (step 9),  $\text{HNO}_3$  is used to dissolve metal oxides in the burner ash, and insolubles are separated from the mother liquor by a centrifuge (step 10).

Extraction of uranium from the clarified dissolver liquor and its decontamination from fission products are carried out based on the "Dilute TBP Process," which consists of two extraction-stripping cycles using 3-5% TBP in an organic diluent (e.g., *n*-dodecane) as the solvent (step 11). Use of the dilute TBP organic phase (i.e., 3-5% rather than 30% TBP as in Purex process) is necessitated by the low uranium concentration (in the range 0.03 to 0.1 M uranium) in the dissolver liquor.

This is because a relatively high uranium concentration in the organic phase is required in order to achieve satisfactory separation from the fission products. Thus processing of 25R and 23R fissile particles yields U-235 and U-233 products, respectively, and high-level liquid wastes.

### Alternatives to Process Steps in Fuel Reprocessing

Outlined below are possible alternatives to some of the process steps shown in Fig. 1.

1. Classification of Fertile and Fissile Particles: The gas elutriation method to separate fissile and fertile particles (step 3) might be replaced by a chemical separation method. This method involves leaching of the residue from the primary burner using a Thorex solution. The bare kernels of fertile particles will be dissolved in this step, and the undissolved SiC-coated fissile particles can then be separated. This approach essentially eliminates step 3. The clarified liquor, after acidity adjustment, is sent to the solvent extraction system. The dried fissile particles are processed following the same sequence as that described in the preceding section (steps 7 through 11).

2. Routing of Dissolver Liquor from 23R Fissile Particles: In the processing of fissile particles from 23R fuel elements, the clarified  $^{233}\text{U}$  liquor from the centrifuge (step 10) may be combined with the  $^{232}\text{Th}$ - $^{233}\text{U}$  liquor from 23R fertile particles (from step 5). The combined liquor is then sent to the Acid-Thorex solvent extraction system (step 6).

3. Plutonium Recovery: The small amount of plutonium present in the feed solution to the solvent extraction system may be recovered as a product, if economically justified or if required for other reasons. For this purpose, the feed solution may have to be acid ( $> 1 \text{ M HNO}_3$ ).  $\text{NaNO}_2$  or gaseous  $\text{NO}$  plus  $\text{NO}_2$  is added to the leacher solution to keep plutonium in the extractable Pu(IV) form. Uranium, thorium, and plutonium are co-extracted in the first column (IA; Acid-Thorex process). The organic phase from the scrubber column (IS) is fed to a partition column where plutonium is reduced to the inextractable Pu(III) by addition of a reductant (e.g., ferrous sulfamate) so as to transfer Pu to the aqueous

phase. Plutonium is thus recovered as an aqueous solution of  $\text{Pu}(\text{NO}_3)_3$ . A similar process scheme may be employed in the dilute TBP process to recover plutonium from the fissile particles.

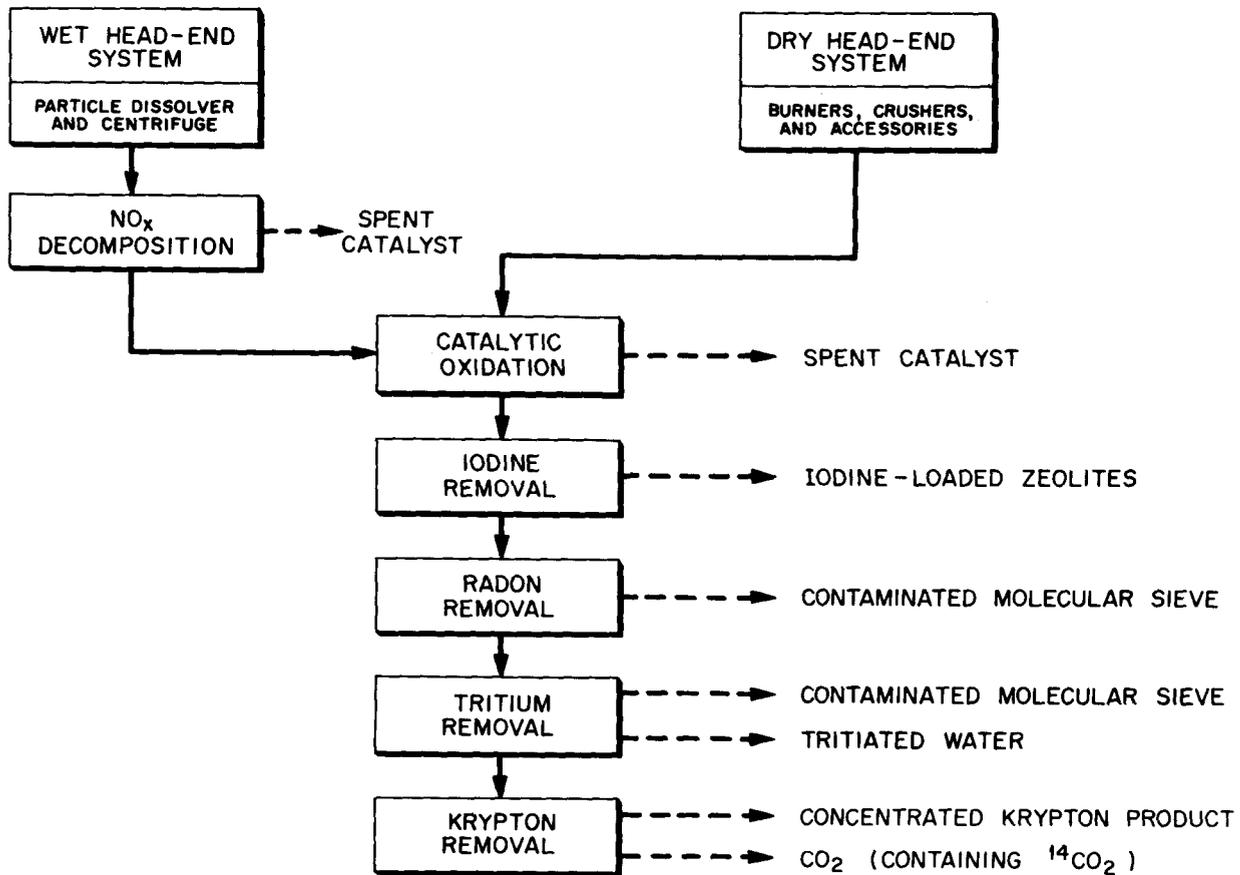
#### Decontamination of Off-Gas Streams

Off-gas streams produced during head-end operation and solvent extraction are processed through two different treatment systems before they are released to the environment. Those streams that contain radionuclides at relatively low levels are treated by the vessel off-gas system (VOGS) which consists of iodine removal and radon removal subsystems in addition to deep-bed and HEPA filters. Off-gas streams in this category are, for example, those from particle classification,\* vessel off-gas from solvent extraction, etc.

On the other hand, relatively large quantities of radionuclides are present in the off-gas streams from the fuel block and particle crushers, burners, the dissolver, and the centrifuge. These streams, therefore, are processed through the off-gas cleanup system (OGCS) primarily to remove iodine, tritium, radon, and krypton (Fig. 2). Semi-volatile radionuclides and particulates in the off-gas from the burners are removed by means of refrigerated surfaces and filters near the gas exit region. Off-gas from the wet head-end system contains  $\text{NO}_x$  which is first decomposed into  $\text{N}_2$  and  $\text{H}_2\text{O}$  in a catalyst bed. Tritium and CO in the gas from both wet and dry head-end systems are catalytically oxidized to HTO and  $\text{CO}_2$  before entering various removal subsystems (Fig. 2). Different types of waste streams resulting from off-gas cleanup operation are indicated in Fig. 2.

---

\* Provided that particle breakage is negligible.

**LEGEND:**

- OFF-GAS FLOW  
 - - - WASTE STREAM FLOW

Fig. 2. Schematic Flow Diagram of Off-Gas Cleanup System.

## GENERAL CONSIDERATIONS AND ASSUMPTIONS

### Overall Flow Scheme of Waste Streams

Shown in Fig. 3 is an overall view of the HTGR reprocessing waste streams and their sources as well as a general waste processing scheme. Only sources directly accountable for the wastes indicated are considered. These waste streams require further processing prior to disposal or long-term storage. Waste sources may be divided into four groups -- the head-end system, the solvent extraction system, the off-gas cleanup system and miscellaneous sources. The last group includes the HTGR reflector blocks and the plant facilities.

### Basic Assumptions

In the present study, assumptions were made that (a) the fuel cycle has reached steady-state condition (~ 8-10 years after the start), (b) three different types of spent commercial fuel elements (25R, 23R, and 25W; see Table 1 for definition) are reprocessed, (c) the fuel contains TRISO-coated fissile particles and BISO-coated fertile particles (Table 1), and (d) the fuel elements have been cooled for 180 days after irradiation in the reactor for 4 years. Additional assumptions, in regard to the rate of fuel reprocessing and the heavy metal (U and Th) contents of the initial and spent fuel elements, are summarized in Table 1.

Figure 4 depicts schematic flow paths of different types of nuclides, showing how they are distributed among major waste and product streams. The numbers shown on the flow paths represent estimated percents of individual nuclide types in respective paths based on the quantities originally present in the discharged fuel particles. These distribution data were used as the basis in estimating radiochemical compositions and other characteristics of selected waste streams (presented in the sections to follow).

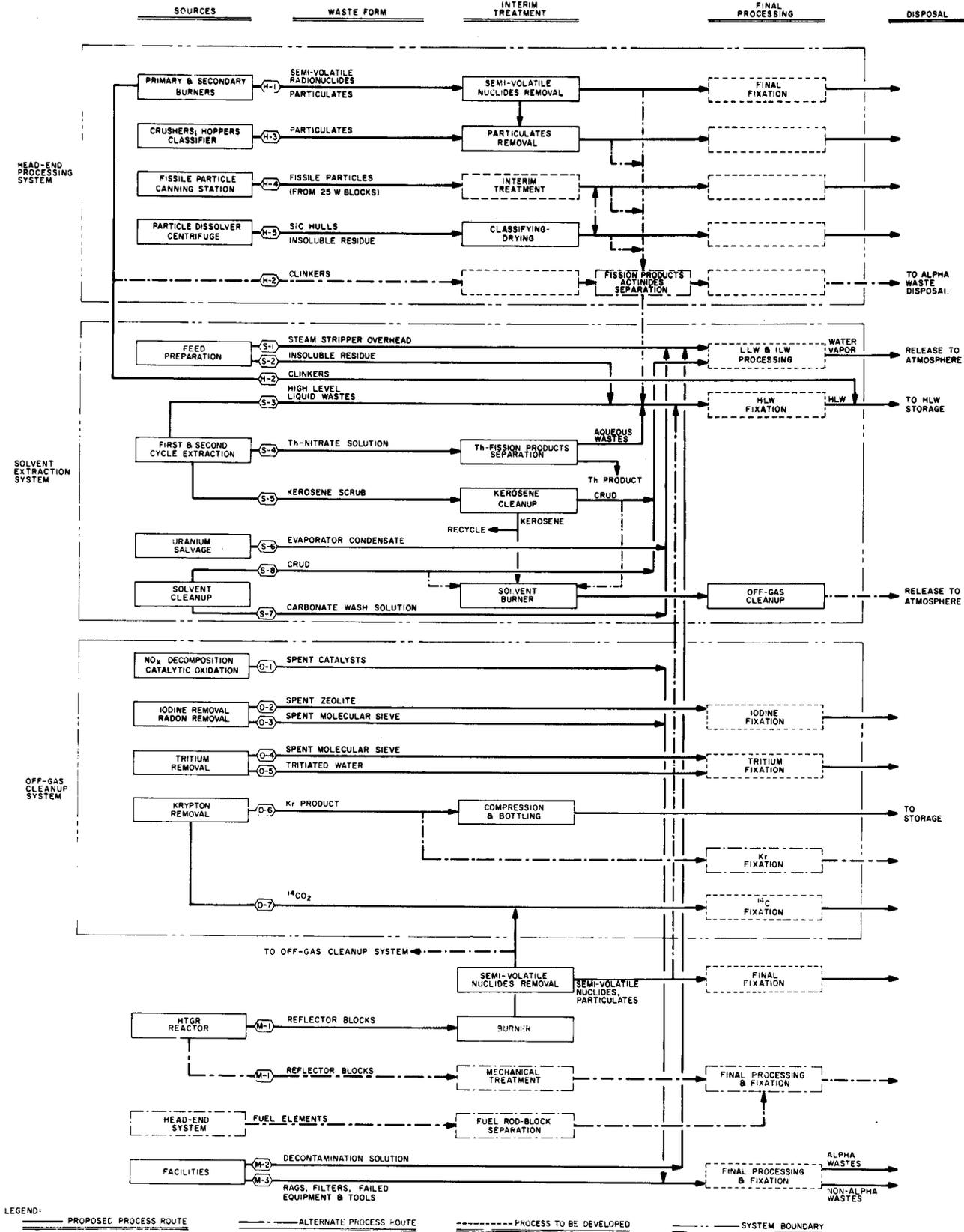


Fig. 3. Overall Schematic Flow Diagram for HTGR Fuel Reprocessing Wastes (Semi-Volatiles, Liquids and Solids).

Table 1. Estimated processing rate and characteristics of individual types of fuel elements<sup>a</sup>

Fuel Types <sup>b</sup>	Processing Rate (No. F.E./yr)	Heavy Metal <sup>c</sup>	Average Contents of Heavy Metals (kg/F.E.)				
			Charged Fuel		Spent Fuel		
			Fissile Particle	Fertile Particle	Fissile Particle	Fertile Particle	
IM-25R	11,600	U Th	0.706 0	0 8.550	0.216 0	0.222 7.926	Burned 0.592 0.0984
23R	7,800	U Th	0.742 0	0 8.830	0.226 0	0.222 7.926	1.198 0.1052
25R-25W	600	U Th	3.250 0	0 7.000	2.24 0	0.19 5.60	2.2200 H 0.216

<sup>a</sup>Based on a HTGR fuel reprocessing plant of 20,000 fuel elements per year.

<sup>b</sup>All fuel types contain BISO-coated (inner porous carbon and outer dense carbon) fertile particles and TRISO-coated (porous carbon, dense carbon, SiC, and dense carbon) fissile particles. Fertile particles contain thorium, while fissile particles contain uranium of varying isotopic compositions depending upon the fuel type as follows:

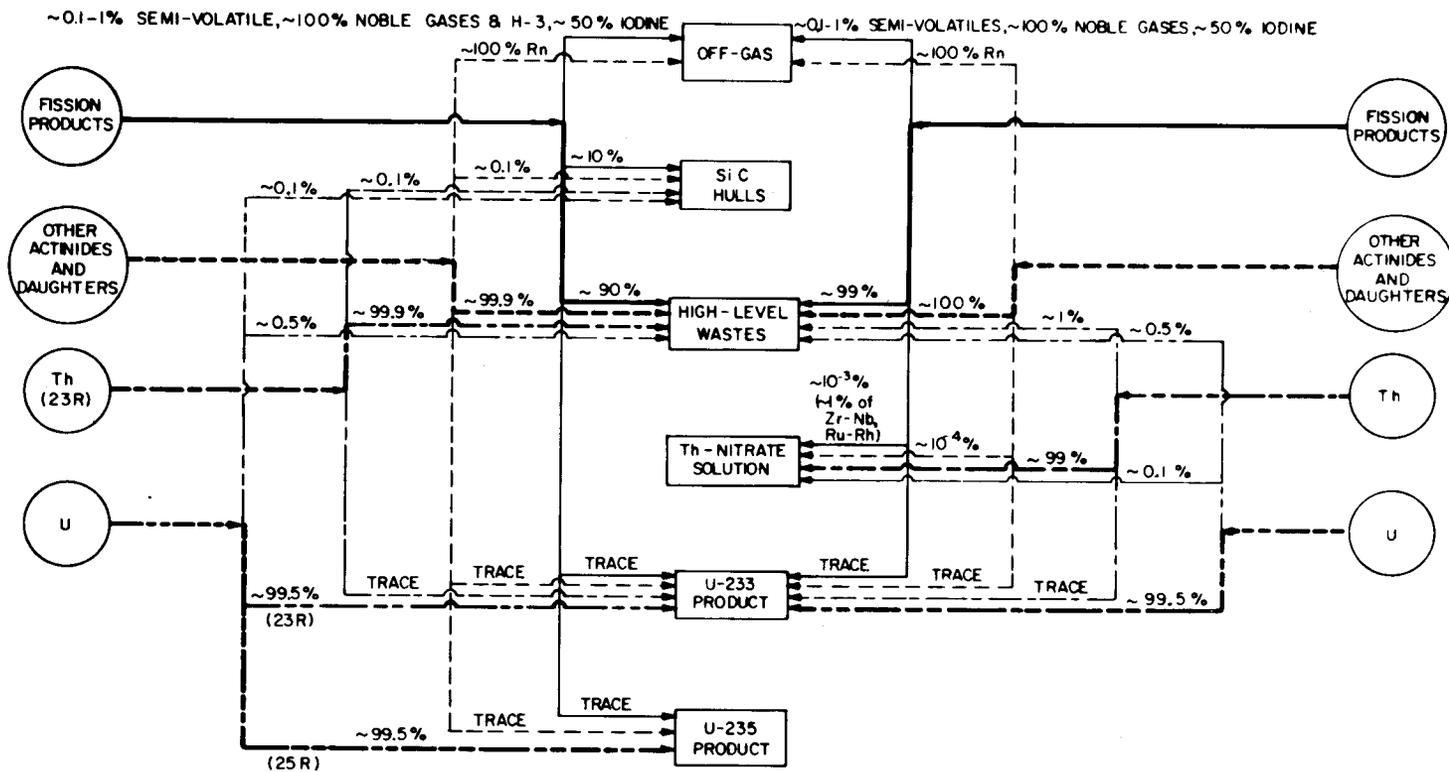
- IM: contains virgin uranium of ~ 93% U-235, initially charged to HTGR
- 25R: contains recycle uranium of ~ 30% U-235 (refabricated from burned IM)
- 23R: contains recycle U-233
- 25W: contains uranium of ~ 4-5% U-235 (burned 25R).

<sup>c</sup>Uranium is present as UC<sub>2</sub>, and thorium as ThO<sub>2</sub>.

**NUCLIDES IN  
FISSILE PARTICLES**

**WASTE OR PRODUCT STREAM**

**NUCLIDES IN  
FERTILE PARTICLES**



NOTE:  
 (1) NUMBERS INDICATED ARE APPROXIMATE PERCENT BASED ON QUANTITIES OF INDIVIDUAL NUCLIDES IN DISCHARGED FUEL PARTICLES.  
 (2) HEAVY LINES INDICATE MAJOR FLOW PATHS OF INDIVIDUAL TYPES OF NUCLIDES

LEGEND:  
 ——— FISSION PRODUCTS  
 - - - - ACTINIDES (EXCLUDE Th & U) AND DAUGHTERS  
 - - - - THORIUM  
 - - - - URANIUM

Fig. 4. Estimated Distribution of Various Types of Nuclides Among Waste and Product Streams.

## SOURCES AND CHARACTERISTICS OF WASTE STREAMS

Outlined below are brief descriptions of individual waste streams in terms of their sources and characteristics. Evaluation of methods for interim and final waste processing (shown in Fig. 3) will be presented in detail in a separate report.

Head-End Processing System

This system consists essentially of all the steps shown in Fig. 1 except for the steps associated with solvent extraction. The major units of equipment that are of concern as sources of wastes are depicted in Fig. 3 under the source column.

1. Semi-Volatile Radionuclides: The nuclides of this type are those having relatively high vapor pressures at burner temperatures (~ 900°C). Included in this type of nuclides are mainly fission products (e.g., Ru, Sb, and Cs), and much smaller amounts of others (i.e., activation products such as <sup>75</sup>Se). They are released in the burning step that takes place at ~ 900°C, and tend to plate out on exposed surfaces at lower temperature along the off-gas passage. Some condensation will occur on or in the dusts collected in cyclones or on filters.

Under normal conditions, a major portion (> 99%) of these nuclides is removed by the sintered-metal filter while a small portion (< 1%) is removed by the HEPA filter. However, some nuclides may penetrate through these filters presumably either as aerosols or as vapors. The amount of nuclides released and the removal efficiencies of filters are not the same for all the semi-volatile nuclides, but vary with individual nuclides. According to some recent results from hot-cell burning experiments,<sup>4</sup> significant amounts of Sb and Ce could penetrate through the sintered metal and HEPA filters. Table 2 shows the approximate range of radiochemical composition of semivolatile and particulate nuclides in the off-gas from the secondary burner (before sintered-metal filter). The composition has been estimated from the data in reference 4.

There are also indications that approximately 0.1 to 1% of fission products originally in the spent fuel may be released to the off-gas in

Table 2. Approximate radiochemical composition of semivolatile and particulate nuclides in secondary burner off-gas<sup>a,b</sup>

Nuclides <sup>c</sup>	<sup>95</sup> Zr	<sup>95</sup> Nb	<sup>106</sup> Ru	<sup>125</sup> Sb	<sup>134</sup> Cs	<sup>137</sup> Cs	<sup>144</sup> Ce
% of Total Radioactivity <sup>d</sup>	1-5	9-18	14-28	3-4	15-21	26-36	1-5

<sup>a</sup>For nuclides having significant levels of gamma radioactivity only.

<sup>b</sup>Calculated from the data in reference 4 for burning of blended fuel particles of BISO ThC<sub>2</sub>-TRISO UC<sub>2</sub>. Before sintered-metal filter.

<sup>c</sup>The behavior of Ce is not well understood.

<sup>d</sup>Based on the sum of radioactivities of nuclides listed.

the burning step. It has been observed that the frequency of replacement of the sintered-metal filter would be governed by the rate of the oxidation attack as well as by the amount of radionuclides deposited on the filter. During cold tests in a secondary burner, a 316-L stainless steel filter (~ 5- $\mu$ m pore size) lasted for about 100 hours at ~ 800°C.<sup>5</sup> In the future, Hastelloy will be used, which will have a much longer life than stainless steel. The service life of the HEPA filter is expected to be much longer than that of the sintered-metal filter. In addition to filters, refrigerated surfaces may be provided to increase the overall decontamination efficiency for the semi-volatile radionuclides.

2. Particulates: Operation of crushers and burners produces fine particulates which are carried away by off-gas streams. These particulates come from graphite fuel blocks and fuel particles as well as burner ash, and are expected to contain significant amounts of fission products, activation products, and actinides. The bulk of particulates in the off-gas is removed by cyclone and sintered-metal filters. The final removal will be carried out by HEPA filters. The amount of particulates entrained in the off-gas stream is estimated to be of the order of 0.1% of the fuel element weight when the fines recycling mechanisms are functioning properly. Otherwise, the particulate content may be higher.

3. Clinkers: This type of solid wastes refers to sintered masses ranging in size from small nodules (several millimeters) to large agglomerates (several inches). Formation of clinkers in the lower portion of the fluidized-bed burner has been observed in work conducted at GAC.<sup>6,7</sup> Similar phenomena were also observed by German researchers.<sup>8</sup>

Understanding of the causes for formation of such masses is still incomplete. Operation of the burner at excessively high temperatures is one of the causes. In addition, the presence of certain impurities (e.g., Na, K, Ca, Cr, Ni, Fe, and Al) on the outer coatings of fuel particles may contribute to lowering of melting points of the coatings

and of exposed fuel material ( $\text{UO}_2$ ,  $\text{ThO}_2$ , etc.). This situation would promote agglomeration of fuel particles under certain operating conditions of the fluidized-bed burner.

Although formation of clinkers is unpredictable, and efforts are being made to overcome the difficulties, clinkers must be considered one of the solid waste streams which require additional processing. They are expected to contain appreciable amounts of actinides as well as fission products.

4. Fissile Particles: The waste fissile particles are those discharged with the 25W fuel elements from the HTGR, and contain  $^{235}\text{U}$  that has been recycled once. The uranium in the initial (IM) fuel elements is ~ 93% enriched in U-235 which declines to ~ 30% U-235 after the initial cycle, with an appreciable buildup of U-236 (a neutron poison). After one recycle, U-235 content drops to ~ 4-5% with a further increase in U-236 content (~ 70% of uranium). Thus, reuse of uranium from the 25W fissile particles in the HTGR becomes economically unattractive.

As shown in Table 3, the 25W fissile particles contain the entire spectra of actinides and fission products, and are high-level wastes (specific radioactivity ~ 600  $\text{kCi/ft}^3$  and heat generation rate ~ 3.6  $\text{kW/ft}^3$ ). The major portion of radioactivity (> 95%) and the bulk of heat generation (> 80%) are attributable to fission products. It has been suggested that the waste fissile particles be either processed with the high-level wastes from the solvent extraction system or be stored as coated particles. These approaches may be undesirable because of the large amount (~ 64% of all nuclides by weight; ~ 750  $\text{Ci/kg}$ ) of actinides present in the fissile particles.<sup>9</sup>

5. SiC Hulls and Insoluble Residue: SiC hulls are discharged from the fissile particle dissolver after leaching of heavy metals and most fission products. Some noble metals also remain as insoluble residue. In leaching of fertile particles (no SiC coating), only the insoluble residue is left. All three types of spent fuel elements (23R and 25R; plus 25W if SiC coatings are processed separately from the fuel kernel) are the sources of SiC hulls which contain non-leachable actinides (~ 0.1% of total actinides) and fission products (~ 10% of total fission products).<sup>10</sup>

Table 3. Estimated composition of retired fissile particles  
(Basis: 600 of 25W fuel elements/yr)

Major Nuclides	Radioactivity		Weight		Rate of Heat Generation	
	Curies	%	kg	%	Watts	%
<u>Fission Products</u>						
Kr-85	$3.393 \times 10^5$	0.52	0.87	0.03	551	0.15
Sr-89	$8.090 \times 10^5$	1.24	0.03	$1 \times 10^{-3}$	2,911 <sup>a</sup>	0.78
Sr-90	$2.473 \times 10^6$	3.79	17.49	0.66	3,240	0.88
Y-90	$2.474 \times 10^6$	3.79	$4 \times 10^{-3}$	$2 \times 10^{-4}$	14,183	3.83
Y-91	$1.413 \times 10^6$	2.17	0.06	$2 \times 10^{-3}$	5,378	1.45
Zr-95	$2.332 \times 10^6$	3.58	0.11	$4 \times 10^{-3}$	12,208	3.30
Nb-95	$4.553 \times 10^6$	6.99	0.12	$4 \times 10^{-3}$	21,914	5.92
Ru-103	$5.506 \times 10^5$	0.84	0.02	$6 \times 10^{-4}$	1,815	0.49
Ru-106	$3.840 \times 10^6$	5.89	1.14	0.04	228 <sup>a</sup>	0.06
Rh-103m, -106	$4.392 \times 10^6$	6.74	5.13	0.19	40,622	10.98
Sb-125	$1.063 \times 10^5$	0.16	0.10	$4 \times 10^{-3}$	431	0.12
Te-127	$5.460 \times 10^4$	0.08	$6 \times 10^{-3}$	$2 \times 10^{-4}$	30 <sup>a</sup>	$8 \times 10^{-3}$
Te-129	$1.562 \times 10^4$	0.02	$5 \times 10^{-4}$	~ 0	31	$8 \times 10^{-3}$
Cs-134	$7.425 \times 10^6$	11.39	5.70	0.22	78,656	21.26
Cs-137	$2.784 \times 10^6$	4.27	32.00	1.21	4,554	1.23
Ba-137m, -140	$2.604 \times 10^6$	4.00	40.00	1.52	10,230	2.76
Ce-141	$3.523 \times 10^5$	0.54	0.01	$5 \times 10^{-4}$	693 <sup>a</sup>	0.19
Ce-144	$1.199 \times 10^7$	18.40	3.75	0.14	9,806	2.65
Pr-143, -144	$1.199 \times 10^7$	18.40	33.99	1.29	92,884	25.11
Pm-147	$1.179 \times 10^6$	1.81	1.27	0.05	608	0.16
Eu-154	$1.667 \times 10^5$	0.26	1.15	0.04	1,369	0.37
Eu-155	$1.420 \times 10^5$	0.22	0.11	$4 \times 10^{-3}$	120 <sup>a</sup>	0.03
Other F.P.	$2.663 \times 10^5$	0.41	820.3	31.09	1,564	0.42
<u>Actinides</u>						
U-232	3.3	~ 0	$2 \times 10^{-4}$	~ 0	0.10 <sup>a</sup>	~ 0
U-233	$3.2 \times 10^{-3}$	~ 0	$3 \times 10^{-4}$	~ 0	$1 \times 10^{-4}$	~ 0
U-234	75.3	$1 \times 10^{-4}$	12.17	0.46	2.17 <sup>a</sup>	$6 \times 10^{-4}$
U-235	0.13	~ 0	62.62	2.37	$3.7 \times 10^{-3a}$	~ 0
U-236	61.7	$1 \times 10^{-4}$	973.4	36.89	1.67	$4 \times 10^{-4}$
U-237	25.5	~ 0	$3 \times 10^{-4}$	~ 0	0.02	~ 0
U-238	0.10	~ 0	292.6	11.09	$2 \times 10^{-3a}$	~ 0
Pu-238	$1.634 \times 10^6$	2.52	96.84	3.67	54,145 <sup>a</sup>	14.64
Pu-239	1,303	$2 \times 10^{-3}$	21.25	0.80	40 <sup>a</sup>	0.01
Pu-240	2,816	$4 \times 10^{-3}$	12.78	0.48	88 <sup>a</sup>	0.02
Pu-241	$9.569 \times 10^5$	1.47	9.42	0.36	40 <sup>a</sup>	0.01
Pu-242	37.8	$6 \times 10^{-5}$	9.70	0.37	1.12 <sup>a</sup>	$3 \times 10^{-4}$
Cm-242	$1.750 \times 10^5$	0.27	0.05	$2 \times 10^{-3}$	6,450 <sup>a</sup>	1.74
Cm-244	$1.467 \times 10^5$	0.22	1.63	0.07	5,130 <sup>a</sup>	1.39
Other Actinides	4,239	$6 \times 10^{-3}$	183.2	6.94	68	0.02
Total F.P.	$6.225 \times 10^7$	95.51	963.3	36.50	$3.040 \times 10^5$	82.17
Total Actinides	$2.921 \times 10^6$	4.49	1,675.6	63.50	$6.596 \times 10^4$	17.83
Grand Total	$6.518 \times 10^7$	100.00	2,638.9	100.00	$3.6996 \times 10^5$	100.00
Per ft <sup>3</sup>	$6.328 \times 10^5$				3,591	

<sup>a</sup>Beta power only.

Practically all (> 99%) nuclides present in the spent SiC hulls are fission products (Table 4). Cesium-134,  $^{143}\text{Pr}$  and  $^{144}\text{Pr}$  are the predominant heat sources, and account for over a half (> 60%) of the decay heat generated in the SiC hulls. The characteristics of SiC hulls resulting from processing of 23R fuel elements are not very much different from those associated with 25R fuel elements. The natures and forms of these actinides and fission products have not been fully characterized. However, it is possible that SiC may have interacted with these nuclides, forming stable compounds. The intense radioactivity (total  $\sim 300 \text{ kCi/ft}^3$ ;  $\sim 0.2 \text{ Ci actinides/kg}$ ) and heat generation rate ( $\sim 1.4 \text{ kW/ft}^3$ ) of the SiC hulls imply that they should be handled as high-level wastes.

#### Solvent Extraction System

Essentially all waste streams discharged from the solvent extraction system are in liquid forms. The most important streams are the liquid high-level wastes from the first and second cycle extraction.<sup>11</sup> The remainder are mostly scrub or wash solutions and contaminated steam condensate having low to intermediate levels of radioactivity. According to reference 12, for liquid wastes, the maximum permissible concentrations (MPC) for unidentified radionuclides are:

for LLW: < 0.1  $\mu\text{Ci/ml}$   
for ILW: < 1  $\text{mCi/ml}$ .

1. Steam Stripper Overhead: The acidity adjustment of feed solution to the first extraction cycle is by steam stripping to remove  $\text{HNO}_3$  in excess of the feed specification ( $\sim 1 \text{ M HNO}_3$ ). The steam condensate leaving from the top of the stripper contains  $\text{HNO}_3$  and some fission products, such as iodine and ruthenium, and is considered as the intermediate-level waste (ILW).
2. Insoluble Residue from Feed Preparation: This is the precipitates that accumulate in the bottom of the steam stripper. The precipitates consist of insoluble compounds of such fission products as Zr, Nb, Mo and noble metals, and is removed from the adjusted feed by filtration before the first extraction cycle.

Table 4. Estimated characteristics of waste SiC hulls<sup>a,b,c</sup>

Major Nuclides	Radioactivity				Weight				Rate of Heat Generation (8 and $\gamma$ powers)			
	25R		23R		25R		23R		25R		23R	
	Curies	%	Curies	%	kg	%	kg	%	Watts	%	Watts	%
	<u>Fission Products</u>											
Sr-89	4.708(5)	1.29	7.204(5)	2.17	0.02	3(-3)	0.03	6(-3)	1,694 <sup>d</sup>	0.93	2,592 <sup>d</sup>	1.63
Sr-90	1.918(6)	5.25	1.610(6)	4.84	13.57	2.33	11.38	2.45	2,513	1.38	2,108	1.33
Y-90	1.919(6)	5.25	1.610(6)	4.84	4(-3)	7(-4)	3(-3)	6(-4)	1.130(4)	6.20	9,476	5.98
Y-91	8.052(5)	2.20	1.083(6)	3.26	0.03	5(-3)	0.04	9(-3)	3,064	1.68	4,119	2.60
Zr-95	1.103(6)	3.02	1.432(6)	4.31	0.05	8(-3)	0.07	0.02	5,773	3.16	7,492	4.72
Nb-95	2.167(6)	5.93	2.798(6)	8.43	0.06	0.01	0.07	0.02	1,043(4)	5.72	1,347(4)	8.50
Ru-103	1.847(5)	0.50	1.826(5)	0.55	6(-3)	1(-3)	6(-3)	1(-3)	609.0	0.33	601.8	0.38
Ru-106	9.516(5)	2.61	5.493(5)	1.65	0.28	0.05	0.16	0.03	56.4 <sup>d</sup>	0.03	32.6 <sup>d</sup>	0.02
Rh-103m, -106	1.136(6)	3.11	7.318(5)	2.20	2.88	0.49	1.91	0.41	1,009(4)	5.53	5,859	3.70
Sb-125	4.708(4)	0.13	1.066(5)	0.32	0.04	7(-3)	0.10	0.02	190.9	0.10	432.3	0.27
Ce-134	5.499(6)	15.06	3.935(6)	11.85	4.22	0.72	3.02	0.65	5,823(4)	31.93	4,168(4)	26.29
Ce-137	1.971(6)	5.40	1.692(6)	5.11	22.66	3.89	19.46	4.18	3,224	1.76	2,769	1.75
Ba-137m, -140	1.843(6)	5.05	1.583(6)	4.77	28.73	4.93	24.42	5.25	7,245	3.97	6,221	3.92
Ce-141	1.512(5)	0.41	2.129(5)	0.64	5(-3)	8(-4)	7(-3)	2(-3)	297.5 <sup>d</sup>	1.63	418.8 <sup>d</sup>	0.26
Ce-144	7.679(6)	21.03	6.920(6)	20.84	2.40	0.41	2.17	0.47	6,278	3.44	5,660	3.57
Pr-143, -144	7.679(6)	21.03	6.924(6)	20.84	24.60	4.22	21.72	4.67	5,949(4)	32.62	5,362(4)	33.82
Pm-147	6.604(5)	1.81	6.420(5)	1.93	0.71	0.12	0.69	0.15	340.5	0.19	331.0	0.21
Eu-154	1.143(5)	0.31	7.448(4)	0.22	0.79	0.13	0.51	0.11	938.2	0.51	611.6	0.38
Eu-155	9.487(4)	0.26	6.184(4)	0.19	0.07	0.01	0.05	0.01	79.8 <sup>d</sup>	0.04	52.0 <sup>d</sup>	0.03
Other F.P.	1.192(5)	0.33	3.465(5)	1.04	478.6	82.18	376.6	80.99	442.0	0.24	966.4	0.61
	<u>Actinides</u>											
U-232	4(-3)	~ 0	35.51	1(-4)	~ 0	~ 0	2(-3)	4(-4)	1(-4) <sup>d</sup>	~ 0	1.14 <sup>d</sup>	7(-4)
U-233	~ 0	~ 0	2.42	~ 0	~ 0	~ 0	0.26	0.06	~ 0	~ 0	0.07	~ 0
U-234	0.17	~ 0	5.37	~ 0	0.03	5(-3)	0.87	0.19	5(-3) <sup>d</sup>	~ 0	0.154 <sup>d</sup>	1(-4)
U-235	1(-3)	~ 0	1(-3)	~ 0	0.47	0.08	0.50	0.11	~ 0	~ 0	~ 0	~ 0
U-236	0.08	~ 0	0.04	~ 0	1.27	0.22	0.61	0.13	2(-3) <sup>d</sup>	~ 0	1(-3) <sup>d</sup>	~ 0
U-238	2(-4)	~ 0	~ 0	~ 0	0.58	0.10	5(-6)	~ 0	5(-6) <sup>d</sup>	~ 0	~ 0	~ 0
Pu-238	1,531	4(-3)	786.3	2(-3)	0.09	0.02	0.05	0.01	50.69 <sup>d</sup>	0.03	26.03 <sup>d</sup>	0.02
Pu-239	1.50	~ 0	0.47	~ 0	0.02	3(-3)	8(-3)	2(-3)	0.05 <sup>d</sup>	~ 0	0.01 <sup>d</sup>	~ 0
Pu-240	3.17	~ 0	0.96	~ 0	0.01	2(-3)	4(-3)	9(-4)	0.10 <sup>d</sup>	~ 0	0.03 <sup>d</sup>	~ 0
Pu-241	1,070	3(-3)	310.7	9(-4)	0.01	2(-3)	3(-3)	6(-4)	0.04 <sup>d</sup>	~ 0	0.01 <sup>d</sup>	~ 0
Pu-242	0.04	~ 0	0.01	~ 0	0.01	2(-3)	2(-3)	4(-4)	1(-3) <sup>d</sup>	~ 0	3(-4) <sup>d</sup>	~ 0
Cm-242	215.3	6(-4)	39.44	1(-4)	6(-5)	~ 0	1(-5)	~ 0	7.93 <sup>d</sup>	4(-3)	1.45 <sup>d</sup>	9(-4)
Cm-244	206.5	6(-4)	22.37	~ 0	3(-3)	5(-4)	3(-4)	~ 0	7.22 <sup>d</sup>	4(-3)	0.78 <sup>d</sup>	5(-4)
Other Actin.	4.30	~ 0	42.49	1(-4)	0.20	0.03	0.10	0.02	0.13	~ 0	1.40	9(-4)
Total F.P.	3.6512(7)	99.99	3.3225(7)	99.99	579.7	99.54	462.6	99.48	1.8228(5)	99.96	1.5851(5)	99.98
Total Actin.	3.032	8(-3)	1,246	4(-3)	2.693	0.46	2.409	0.52	66.2	0.04	31.1	0.02
Grand Total	3.6515(7)	100.00	3.3227(7)	100.00	582.4	100.00	465.0	100.00	1.8235(5)	100.00	1.5854(5)	100.00
Per ft <sup>3</sup> Mixt.	2,968(5)Ci											1,450 W

<sup>a</sup>Assume that ~ 10% of fission products and ~ 0.1% of actinides (both based on the total amounts in discharged fuels) are present in SiC hulls.

<sup>b</sup>The quantities shown are based on an assumed annual reprocessing capacity of 7,800 of 23R fuel elements, and 11,600 of 25R fuel elements.

<sup>c</sup>Numbers in parentheses represent powers of 10.

<sup>d</sup>Beta power only.

3. High-Level Liquid Wastes (HLLW): HLLW is basically the aqueous waste stream discharged from the first extraction cycle, and contains the bulk of the fission products and very small amounts of actinides ( $< \sim 0.5\%$ ) from the spent fuel in  $\text{HNO}_3$  solution. The major portion of HLLW (in terms of radioactivity) comes from this aqueous raffinate discharged from the first-cycle extraction column, and most of the remaining portion is the concentrate of the aqueous waste from the second-cycle extraction column.

The estimated characteristics of HLLW in Table 5 represents that of a mixture of HLLWs from processing of 23R and 25R fissile particles, and of fertile particles from 23R, 25R and 25W fuel elements. Fission products are, by far, the dominant nuclides, accounting for nearly 90% of the total weight of all the nuclides, and contribute over 95% to the decay heat generated. Niobium-95,  $^{134}\text{Cs}$ ,  $^{143}\text{Pr}$ , and  $^{144}\text{Pr}$  are among the major heat sources. Thorium-232 is the predominant actinide ( $\sim 9\%$  of total by weight) in the HLLW, but important heat generating actinides are  $^{238}\text{Pu}$  and  $^{233}\text{Pa}$  (included in "other actinides").

Aqueous low-level and intermediate-level wastes (LLW and ILW) from other units of equipment in the solvent extraction system (e.g., steam stripper overhead, uranium-salvage evaporator condensate, etc.) will eventually be concentrated and combined with the HLLW.

One of the features unique to the HLLW from the HTGR fuel reprocessing (as compared to the LWR fuel reprocessing) is the presence of fluoride which is required for the dissolution of thoria. This fluoride will be volatilized as HF and released in the off-gas stream during the high-temperature solidification process unless it is converted to some non-volatile compounds (e.g.,  $\text{CaF}_2$ ) prior to solidification.

4. Thorium Nitrate Solution: An acidic solution of thorium nitrate is removed from the bottom of the partition-scrub column. The solution is the product of dilute  $\text{HNO}_3$  stripping of thorium from the solvent phase containing uranium and thorium in the partitioning column (first cycle). As shown in Table 6, residual amounts of fission products and uranium and other actinides are present in the thorium nitrate solution. Thorium-232 accounts for nearly 100% (by weight) of all the nuclides present in

Table 5. Estimated characteristics of high-level wastes<sup>a,b,c</sup>

Major Nuclides	Radioactivity		Weight		Rate of Heat Generation ( $\beta$ and $\gamma$ power)	
	Curies	%	kg	%	Watts	%
<u>Fission Products</u>						
Sr-89	4.901(7)	3.23	1.74	0.01	1.764(5) <sup>d</sup>	2.57
Sr-90	5.290(7)	3.49	374.09	2.19	6.681(4)	0.97
Y-90	5.291(7)	3.49	0.07	4(-4)	3.114(5)	4.54
Y-91	5.873(7)	3.87	2.70	0.02	2.510(5)	3.66
Zr-95	8.690(7)	5.73	4.11	0.02	4.549(5)	6.63
Nb-95	1.689(8)	11.13	4.30	0.02	8.129(5)	11.84
Ru-103	9.248(6)	0.61	0.29	2(-3)	3.048(4)	0.44
Ru-106	2.248(7)	1.48	6.70	0.04	1,332 <sup>d</sup>	0.02
Rh-103m, -106	2.151(7)	1.42	74.78	0.44	2.406(5)	3.50
Sb-125	3.552(6)	0.23	3.35	0.02	1.441(4)	0.21
Cs-134	1.224(8)	8.07	94.03	0.55	1.296(6)	18.88
Cs-137	5.525(7)	3.64	635.21	3.72	9.039(4)	1.32
Ba-137m, -140	5.134(7)	3.38	770.36	4.51	2.031(5)	2.96
Ce-141	1.434(7)	0.94	0.50	3(-3)	2.821(4) <sup>d</sup>	0.41
Ce-144	3.024(8)	19.93	94.71	0.55	2.473(5)	3.60
Pr-143, -144	3.025(8)	19.94	704.49	4.12	2.342(6)	34.12
Pm-147	3.052(7)	2.01	32.87	0.19	1.574(4)	0.23
Eu-154	2.439(6)	0.16	16.82	0.10	2.002(4)	0.29
Eu-155	2.036(6)	0.13	1.59	0.01	1,714 <sup>d</sup>	0.02
Other F.P.	1.350(7)	0.89	1.211(4)	70.87	3.260(4)	0.47
<u>Actinides</u>						
U-232	817.59	~ 0	0.04	2(-4)	26.2 <sup>d</sup>	4(-4)
U-233	206.80	~ 0	21.83	0.13	6.0	9(-5)
U-234	58.90	~ 0	9.52	0.06	1.7 <sup>d</sup>	~ 0
U-235	0.01	~ 0	6.23	0.04	4(-4)	~ 0
U-236	0.62	~ 0	9.70	0.06	0.02 <sup>d</sup>	~ 0

Table 5. (continued)

Major Nuclides	Radioactivity		Weight		Rate of Heat Generation ( $\beta$ and $\gamma$ power)	
	Curies	%	kg	%	Watts	%
<u>Actinides (continued)</u>						
Pu-238	2.331(6)	0.15	138.08	0.81	7.718(4) <sup>d</sup>	1.12
Pu-239	1,976	1(-4)	32.25	0.19	61.4 <sup>d</sup>	9(-4)
Pu-240	4,146	3(-4)	18.81	0.11	128.4 <sup>d</sup>	2(-3)
Pu-241	1.069(6)	0.07	13.61	0.08	57.4 <sup>d</sup>	8(-4)
Pu-242	54.59	~ 0	14.01	0.08	1.6 <sup>d</sup>	~ 0
Cm-242	2.546(5)	0.02	5.24	0.03	9,382 <sup>d</sup>	0.14
Cm-244	2.287(5)	0.02	2.83	0.02	8,001 <sup>d</sup>	0.12
Th-228	1,003	7(-5)	1(-3)	~ 0	32.8 <sup>d</sup>	5(-4)
Th-230	0.89	~ 0	0.05	3(-4)	4(-3) <sup>d</sup>	~ 0
Th-232	0.17	~ 0	1,565	9.16	4(-3) <sup>d</sup>	~ 0
Other Actin.	9.053(7)	5.97	317.03	1.86	1.322(5)	1.92
Total F.P.	1.423(9)	93.78	1.493(4)	87.39	6.638(6)	96.69
Total Actin.	9.442(7)	6.22	2,154	12.61	2.271(5)	3.31
Grand Total	1.517(9)	100.00	1.708(4)	100.00	6.865(6)	100.00

<sup>a</sup>Quantities shown are based on an assumed annual reprocessing capacity of 7,800 of 23R fuel elements and 11,600 of 25R fuel elements. The high-level wastes (HLW) in this table are a mixture of HLWs from reprocessing of both fissile and fertile particles.

<sup>b</sup>HLW is assumed to contain: (1) all fission products and actinides other than U (in the discharged fuel), except those retained by SiC hulls and/or lost to the off-gas, and (2) 0.5% of U not retained by SiC hulls, and (3) 1% of Th in the discharged fuel (fertile particles).

<sup>c</sup>Numbers in parentheses represent powers of 10.

<sup>d</sup>Beta power only.

Table 6. Estimated characteristics of thorium nitrate solution<sup>a,b,c</sup>

Major Nuclides	Radioactivity		Weight		Rate of Heat Generation ( $\beta$ and $\gamma$ power)	
	Curies	%	kg	%	Watts	%
	<u>Fission Products</u>					
Sr-89	382.9	0.02	1(-5)	~ 0	1.4 <sup>d</sup>	0.01
Sr-90	211.4	0.01	2(-3)	~ 0	0.2	2(-3)
Y-90	211.4	0.01	4(-7)	~ 0	1.2	0.01
Y-91	489.8	0.02	2(-5)	~ 0	1.9	0.02
Zr-95	6.408(5)	28.54	0.03	~ 0	3,355	26.97
Nb-95	1.242(6)	55.31	0.03	~ 0	5,978	48.05
Ru-103	5.942(4)	2.65	2(-3)	~ 0	195.8	1.57
Ru-106	8.971(4)	4.00	0.03	~ 0	5.3 <sup>d</sup>	0.04
Rh-103m, -106	1.492(5)	6.64	0.32	2(-4)	970.9	7.80
Sb-125	21.7	1(-3)	2(-5)	~ 0	0.1	7(-4)
Cs-134	375.0	0.02	3(-4)	~ 0	4.0	0.03
Cs-137	222.9	0.01	3(-3)	~ 0	0.4	3(-3)
Ba-137m, -140	208.6	0.01	3(-3)	~ 0	0.8	6(-3)
Ce-141	110.6	5(-3)	4(-6)	~ 0	0.2 <sup>d</sup>	2(-3)
Ce-144	1,710	0.08	5(-4)	~ 0	1.4	0.01
Pr-143, -144	1,710	0.08	3(-3)	~ 0	13.2	0.11
Pm-147	188.0	8(-3)	2(-4)	~ 0	0.1	8(-4)
Eu-154	7.4	3(-4)	5(-5)	~ 0	0.1	8(-4)
Eu-155	6.3	3(-4)	5(-6)	~ 0	5(-3) <sup>d</sup>	~ 0
Other F.P.	87.68	4(-3)	0.02	~ 0	0.2	2(-3)
	<u>Actinides</u>					
U-232	128.0	6(-3)	6(-3)	~ 0	4.1 <sup>d</sup>	0.03
U-233	38.9	2(-3)	4.1	3(-3)	1.1	0.01
U-234	6.2	3(-4)	1.0	6(-4)	0.2 <sup>d</sup>	2(-3)
U-235	6(-4)	~ 0	0.3	2(-4)	2(-5) <sup>d</sup>	~ 0
U-236	3(-3)	~ 0	0.05	~ 0	9(-5) <sup>d</sup>	~ 0

Table 6. (continued)

Major Nuclides	Radioactivity		Weight		Rate of Heat Generation ( $\beta$ and $\gamma$ power)	
	Curies	%	kg	%	Watts	%
<u>Actinides</u> (continued)						
Pu-238	0.02	~ 0	1(-6)	~ 0	5(-4) <sup>d</sup>	~ 0
Pu-239	8(-6)	~ 0	1(-7)	~ 0	2(-7) <sup>d</sup>	~ 0
Pu-240	1(-5)	~ 0	6(-8)	~ 0	4(-7) <sup>d</sup>	~ 0
Pu-241	3(-3)	~ 0	3(-8)	~ 0	1(-7) <sup>d</sup>	~ 0
Pu-242	5(-8)	~ 0	1(-8)	~ 0	1(-9) <sup>d</sup>	~ 0
Cm-242	1(-4)	~ 0	4(-11)	~ 0	5(-6) <sup>d</sup>	~ 0
Cm-244	4(-5)	~ 0	4(-10)	~ 0	1(-6) <sup>d</sup>	~ 0
Th-228	5.801(4)	2.58	0.07	~ 0	1,900 <sup>d</sup>	15.27
Th-229	12.1	5(-4)	0.06	~ 0	0.4 <sup>d</sup>	3(-3)
Th-230	88.5	4(-3)	4.6	3(-3)	2.5 <sup>d</sup>	0.02
Th-232	16.9	8(-4)	1.549(5)	99.99	0.4 <sup>d</sup>	3(-3)
Other Actin.	90.4	4(-3)	2(-5)	~ 0	0.1	8(-4)
Total F.P.	2.1871(6)	97.40	0.44	2(-4)	1.0531(4)	84.65
Total Actin.	5.8391(4)	2.60	1.5491(5)	~ 100	1,909	15.35
Grand Total	2.2455(6)	100.00	1.5491(5)	100.00	1.2440(4)	100.00

<sup>a</sup>Quantities listed are based on an annual reprocessing capacity of 20,000 fuel elements (11,600 25R, 7800 23R and 600 25W).

<sup>b</sup>The recovery of thorium nitrate solution was made by the Acid Thorex Process based on the following assumptions: (1) Th recovery efficiency is 99%, and (2) DFs are  $10^5$  for fission products (except DF = 100 for Zr-Nb and Ru-Rh),  $10^6$  for actinides excluding U, and  $10^3$  for U. DF here is defined as: DF = (Amt. of nuclide in fuel)/(Amt. of nuclide in Th-nitrate solution).

<sup>c</sup>Numbers in parentheses represent powers of 10.

<sup>d</sup>Beta power only.

the solution (Table 6). The bulk of radioactivity (> 95%), however, is contributed by fission products, mostly by  $^{95}\text{Zr}$  and  $^{95}\text{Nb}$  (~ 84%). A significant portion of the radioactivity also comes from  $^{228}\text{Th}$  (~ 2.6%), and fission products Ru-Rh (~ 13%). Likewise, Zr-Nb, Ru-Rh, and  $^{228}\text{Th}$  are the major sources of decay heat. The solution will be concentrated for storage for possible future reuse, in which case further purification may be necessary.

Some of the major problems associated with storage of the thorium nitrate solution include the continuous production of  $^{220}\text{Rn}$  from  $^{228}\text{Th}$ , and the heat generation primarily from decay of residual fission products, and of  $^{228}\text{Th}$  and its daughters. Also to be considered is possible production of an explosive mixture of  $\text{H}_2$ ,  $\text{O}_2$  and other gases due to radiolysis. Therefore, the storage vessel would have to be sparged and the sparged gas cleaned through the radon removal and retention system. A cooling system may be required to dissipate the heat generated in the vessel.

5. Carbonate Wash Solution: The waste carbonate wash solution results from scrubbing of used solvent with ~ 0.5 M  $\text{Na}_2\text{CO}_3$  solution to remove the bulk of chemical and radiolytic degradation products. The solution is probably contaminated with trace quantities of fission products (e.g., Zr, Nb, Ru, Rh, I, etc.) and traces of actinides as well as degradation products.

6. Crud from Used Solvents: This type of solid waste comes from the solvent cleanup subsystem consisting of the solvent wash column and the centrifuge. The crud (a collection of chemical and radiolytic decomposition products) is separated from the solvent by centrifuging after scrubbing of the solvent with  $\text{Na}_2\text{CO}_3$  solution. Small amounts of actinides and fission products are present in the crud (estimated at ~ 0.1% of U and ~ 0.01% of fission products in the original fuel).

7. Kerosene Scrub: Waste kerosene is produced when the aqueous uranium solution from the stripping column is brought into contact with kerosene in the solvent-scrub column to remove residual TBP. The waste kerosene is subsequently transported to the kerosene cleanup subsystem,

and the clarified kerosene is either recycled to the solvent extraction system or sent to the solvent burner. It contains trace quantities of actinides and fission products as well as the TBP and solvent degradation products.

8. Evaporator Condensate: The evaporator in the uranium salvage subsystem processes aqueous waste solutions from equipment in the first and second extraction cycles, and from other units of equipment that handle appreciable amounts of uranium. The evaporator condensate which is contaminated with trace quantities of radionuclides must be transferred to the LLW and ILW processing system for decontamination.

#### Off-Gas Cleanup System

In the off-gas cleanup system, waste streams are mostly solids (i.e., spent catalysts, molecular sieves and zeolites), but liquid (tritiated water) and gaseous (Kr and  $^{14}\text{CO}_2$ ) wastes are also generated. The radionuclides associated with these waste streams are e.g.,  $^{129}\text{I}$ ,  $^{131}\text{I}$ ,  $^3\text{H}$ ,  $^{85}\text{Kr}$ ,  $^{14}\text{CO}_2$ , and  $^{220}\text{Rn}$  and its decay daughters. Some semi-volatile and particulate fission products (e.g., Sb, Ru, Cs, and Ce) from the head-end system may penetrate as far as the off-gas cleanup system.

1. Spent Catalysts: Catalysts are employed in the  $\text{NO}_x$  decomposition and in the off-gas oxidation subsystems. They are discharged as solid wastes when their performances deteriorate. The spent catalysts are expected to be slightly contaminated with iodine and some semi-volatile radionuclides. Small amounts of particulate radionuclides may also be present.

2. Spent Iodine-Bearing Zeolites: The solid wastes of this type are mostly lead- or cadmium-zeolite, which is the major adsorbent for removal of iodine from the off-gas stream. A small amount of spent silver-zeolite (a backup adsorbent following the lead- or cadmium-zeolite bed) is also discharged as the solid waste. These spent zeolites are loaded with iodine, but traces of semi-volatile nuclides

and daughters of radon may be trapped together with iodine. There is evidence to indicate that iodine isotopes are present in zeolites as relatively stable chemisorbed species. Further fixation, however, may be necessary prior to disposal of the spent zeolites.

3. Spent Molecular Sieves: These come from two different sources -- the radon loading and tritium removal subsystems. The spent molecular sieves loaded with radon and its decay daughters are not expected to present much of a problem in disposal because of the short half-lives of Rn-220 (56 sec) and its daughters ( $^{212}\text{Pb}$  has the longest half-life, 10.6 hr). However, further processing and fixation may be required if the molecular sieves should also retain iodines, tritiated water, and/or semi-volatile radionuclides.

Molecular sieves (e.g., Type 3A) in the tritium removal system, when loaded with tritiated water, are regenerated and reused until exhausted. The molecular sieves from the tritium removal system will contain residual amounts of tritiated water and traces of other fission products.

4. Tritiated Water: This is produced when the molecular sieves in the tritium removal system are regenerated. Additional processing is required to convert tritiated water into an insoluble form (e.g., fixation in concrete).

5. Krypton Product: Krypton-85 is the only significant radioactive isotope in the krypton concentrate discharged from the krypton removal system. The system uses liquid  $\text{CO}_2$  as a solvent in absorption and concentration of krypton by the KALC process (Krypton Absorption in Liquid  $\text{CO}_2$ ). The krypton product stream will probably contain appreciable amounts of  $\text{CO}_2$  and xenon, depending upon the specific mode of operation. Under certain unfavorable conditions upstream (i.e., in other subsystems in the off-gas cleanup system and in the head-end system), other fission products could also get into this system, but will be separated from the krypton product.

6. CO<sub>2</sub> Stream: The CO<sub>2</sub> in the off-gas stream entering the off-gas cleanup system from the head-end system contains ~ 0.2 ppm (by weight based on total CO<sub>2</sub>)<sup>\*</sup> of <sup>14</sup>C<sub>2</sub> which is produced in the fuel elements during their irradiation in the HTGR. The major source (> 80%) of <sup>14</sup>C is nitrogen, present in the fuel element as an impurity, and the balance is produced from <sup>13</sup>C. Because of the long half-life of <sup>14</sup>C (5730 yr), the CO<sub>2</sub> gas stream containing <sup>14</sup>C may require further processing to convert <sup>14</sup>CO<sub>2</sub> into a chemical or physical form suitable for disposal or permanent storage after krypton has been removed.

A study<sup>13</sup> has been completed on several potential methods for fixation and disposal of <sup>14</sup>C-contaminated CO<sub>2</sub> as CaCO<sub>3</sub>. The study considered two CO<sub>2</sub> fixation processes, the direct process and the double alkali process. Among the methods for disposal of CaCO<sub>3</sub> studied were: (a) shallow-land burial, (b) hydraulic fracturing, and (c) deep sea disposal. Preliminary results of evaluation indicate that the most attractive approach appears to be fixation of CO<sub>2</sub> as CaCO<sub>3</sub>, and subsequent disposal by the shallow-land burial of solid CaCO<sub>3</sub> in 55-gallon drums. Also in progress is a study to evaluate the environmental effect of <sup>14</sup>C from HTGR fuel reprocessing facilities.<sup>14</sup> The initial phase of the study is concerned with the dose rate at the site boundary, while the second phase of the study deals with the global aspect of the environmental effect.

#### Miscellaneous Waste Streams

1. Reflector Blocks: Replaceable reflector blocks are discharged from the HTGR probably on an eight-year cycle. A variety of neutron-activated radionuclides are generated in the reflector blocks as the result of impurities in the blocks. Some fission products and traces of actinides may also be transported by the helium coolant gas from failed fuel particles in the fuel elements to coolant holes in the blocks.

\*Based on ~ 30 ppm of N<sub>2</sub> present in the original fuel.<sup>15</sup>

2. Decontamination Solutions: Liquid wastes produced in decontamination of various units of equipment in the fuel reprocessing plant will have diverse chemical and radiochemical compositions. Used decontamination solutions coming from some equipment in the head-end and solvent extraction systems may contain appreciable amounts of fissile materials (i.e., uranium, thorium, etc.), for which special handling will be required. Waste solutions from other units of equipment will probably be of low- and intermediate-level types.

3. Miscellaneous Solid Wastes: Included in this category are contaminated rags, gloves, and clothes, spent filters, and failed equipment and tools. Large pieces of equipment (complete or parts, e.g., inner lining of burners) would have to be reduced in size by either mechanical or chemical means (e.g., cutting-compaction, melting-casting, chemical dissolution, etc.) prior to disposal. The extent and type of contamination on such solid wastes are variable, but those that contain high alpha radioactivities (e.g.,  $> 1 \mu\text{Ci/kg}$  solids) may have to be segregated from others and handled separately.

#### SUMMARY AND CONCLUSIONS

Table 7 summarizes information pertinent to waste streams from an HTGR fuel reprocessing plant (20,000 F.E./yr) in regard to their sources, forms, and approximate quantities. Also shown in Table 7 are radionuclides that may be present in detectable quantities in individual waste streams.

Most of the waste streams are unique to HTGR fuel reprocessing, although waste streams from the solvent extraction system and from the plant facilities are not very much different from those discharged from the LWR fuel reprocessing plant. The high-level liquid wastes from solvent extraction, however, are somewhat different from the corresponding wastes in LWR fuel reprocessing in that they contain fluorides that were introduced to facilitate dissolution of thoria. This implies that an additional processing step would be required to convert fluorides into a non-volatile form before subjecting the wastes to a high-temperature solidification process.

Table 7. Sources and estimated flow rates of waste streams from HTGR reprocessing plant

[Basis: 20,000 Fuel Elements/yr; 58% 25R, 39% 23R, 3% 25W]

Stream No.	Source (Subsystem)	Waste Form	Approx. Quantities per yr (Est.)	Probable Key Radionuclides Present (Est. Amt/yr)
<u>Head-End Processing System</u>				
H-1	Primary and secondary burners	Semi-volatile nuclides, particulates	110 kg, 10 MCi; incl. in H-3	Fission products (e.g., Zr, Nb, Ru, Sb, Cs, Ce, etc.), actinides
H-2	Primary and secondary burners	Clinkers	0-6 MT <sup>a</sup>	Fission products, actinides
H-3	Crushers, hoppers, classifier	Particulates	12 MT	Fission products, actinides
H-4	Fissile particle canning station	Fissile particles (from 25 W blocks)	4 MT	Actinides (U = 1.3 MT; 170 Ci); fission products (1 MT; 62 MCi)
H-5	Particle dissolver, centrifuge	SiC hulls, insol. residues (incl. noble metals)	20 MT	Actinides (5 kg, 4300 Ci); fission products (~ 1 MT; 70 MCi) (esp. noble metals)
<u>Solvent Extraction System</u>				
S-1	Feed preparation	Steam stripper overhead	1,100,000 gal <sup>b</sup> (solid content ≈ 0.5-1% wt)	I, Ru
S-6	Uranium salvage	Evaporator condensate		I, Ru
S-7	First and second cycle extraction	Carbonate wash solution		Zr, Nb, Ru, Rh, I
S-2	Feed preparation	Insol. residue	1-3 MT	Zr, Nb
S-3	First and second cycle extraction	High-level liquid waste	200,000 gal <sup>b</sup>	Fission products (15 MT, 1400 MCi) actinides (2 MT, 95 MCi)

Table 7. (continued)

Stream No.	Source (Subsystem)	Waste Form	Approx. Quantities per yr (Est.)	Probable Key Radionuclides Present (Est. Amt/yr)
<u>Solvent Extraction System (Cont'd)</u>				
S-4	First and second cycle extraction	Thorium nitrate solution	50,000 gal <sup>b</sup>	Th (155 MT, 60 kCi), actinides; Zr, Nb, Ru, Rh (total F.P. ~ 2 MCi)
S-5	First and second cycle extraction	Kerosene scrub	5,500 gal	Fission products, actinides
S-8	Solvent cleanup	Crud	~ 1 MT	Fission products (~ 2 kg, ~ 150 kCi), actinides (~ 170 kg, ~ 95 kCi)
<u>Off-Gas Cleanup System</u>				
0-1	NO <sub>x</sub> decomposition, catalytic oxidation	Spent catalysts	140 ft <sup>3</sup>	Fission products (esp., semi-volatile)
0-2	Iodine removal	Spent zeolite	Pb zeolite = 1200 ft <sup>3</sup> Ag zeolite = 150 ft <sup>3</sup>	Iodine isotopes (60 Ci, 35 kg)
0-3	Radon removal	Spent molecular sieve	440 ft <sup>3</sup>	Rn daughters, traces of I and HTO
0-4	Tritium removal	Spent molecular sieve	150 ft <sup>3</sup>	HTO
0-5	Tritium removal	Tritiated water	3,700 gal	HTO ( <sup>3</sup> H = 277,000 Ci)
0-6	Krypton removal	Kr-product stream	Kr: 460 kg	<sup>85</sup> Kr (11 MCi)
0-7	Krypton removal	CO <sub>2</sub> gas	6,680 MT	<sup>14</sup> C (~ 1 kg <sup>14</sup> CO <sub>2</sub> ), traces of <sup>85</sup> Kr

Table 7. (continued)

Stream No.	Source (Subsystem)	Waste Form	Approx. Quantities per yr (Est.)	Probable Key Radionuclides Present (Est. Amt/yr)
<u>Miscellaneous</u>				
M-1	HTGR	Reflector blocks	5600 ft <sup>3</sup> (350 MT)	<sup>14</sup> C, neutron-activation products
M-2	Facilities	Decontamination solution	1-2 x 10 <sup>6</sup> gal	Variable
M-3 <sup>c</sup>	Facilities	Rags, spent filters, failed equipment and tools, etc.	74,000 ft <sup>3</sup>	Variable

<sup>a</sup> Assume ~5% of the fuel material formed clinkers.

<sup>b</sup> Unconcentrated solutions.

<sup>c</sup> Calculated based on the data from reference 16.

Solid waste streams from the head-end system are unique to HTGR fuel reprocessing. This is especially true of waste fissile particles (from 25W fuel elements), SiC hulls, and clinkers. Selection of specific processing methods (e.g., whether to separate actinides from fission products or not) for these wastes will be governed largely by future Federal regulations and other non-economic factors. Also unique is the  $^{14}\text{C}$ -containing waste stream which may have to be converted into a form acceptable for disposal.

Additional characteristics for major waste streams are presented in Table 8. Fission products account for most (> 80%) of the radioactivity and the decay heat in all four streams. Both the 25W fissile particles and the SiC hulls must also be handled as high-level wastes because of their intense radioactivities and heat generation rates.

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Table 8. Profile of selected waste streams<sup>a</sup>

Waste Stream	Nuclide Types	Estimated Characteristics <sup>c</sup>							
		Radioactivity			Rate of heat generation			Weight	
		CI	% of Total	CI/ Unit Quantity	Watts	% of Total	W/ Unit Quantity	kg	%
Retired Fissile Particles (25W)	Fission Products	6.225(7)	95.51	6.044(5)/ft <sup>3</sup>	3.040(5)	82.17	2,951/ft <sup>3</sup>	963.3	36.50
	Actinides	2.921(6)	4.49	2.836(4)/ft <sup>3</sup>	6.596(4)	17.83	640/ft <sup>3</sup>	1,675.6	63.50
	Total	6.518(7)	100.00	6.328(5)/ft <sup>3</sup>	3.700(5)	100.00	3,591/ft <sup>3</sup>	2,638.9	100.00
SiC Hulls	Fission Products	6.974(7)	99.99	2.968(5)/ft <sup>3</sup>	3.408(5)	99.97	1,450/ft <sup>3</sup>	1,042.3	99.51
	Actinides	4.278	0.01	18.2/ft <sup>3</sup>	97.3	0.03	~ 0	5.1	0.49
	Total	6.974(7)	100.00	2.968(5)/ft <sup>3</sup>	3.409(5)	100.00	1,450/ft <sup>3</sup>	1,047.4	100.00
High-Level Liquid Wastes <sup>b</sup>	Fission Products	1.423(9)	93.78	7,115/gal	6.638(6)	96.69	33/gal	1.493(4)	87.39
	Actinides	9.442(7)	6.22	472/gal	2.271(5)	3.31	1/gal	2,154	12.61
	Total	1.517(9)	100.00	7,587/gal	6.865(6)	100.00	34/gal	1,708(4)	100.00
Thorium Nitrate Solution <sup>b</sup>	Fission Products	2.187(6)	97.40	43.7/gal	1.053(4)	84.65	0.21/gal	0.44	2(-4)
	Actinides	5.839(4)	2.60	1.2/gal	1,909	15.35	0.04/gal	1.549(5)	~ 100
	Total	2.246(6)	100.00	44.9/gal	1.244(4)	100.00	0.25/gal	1.549(5)	100.00

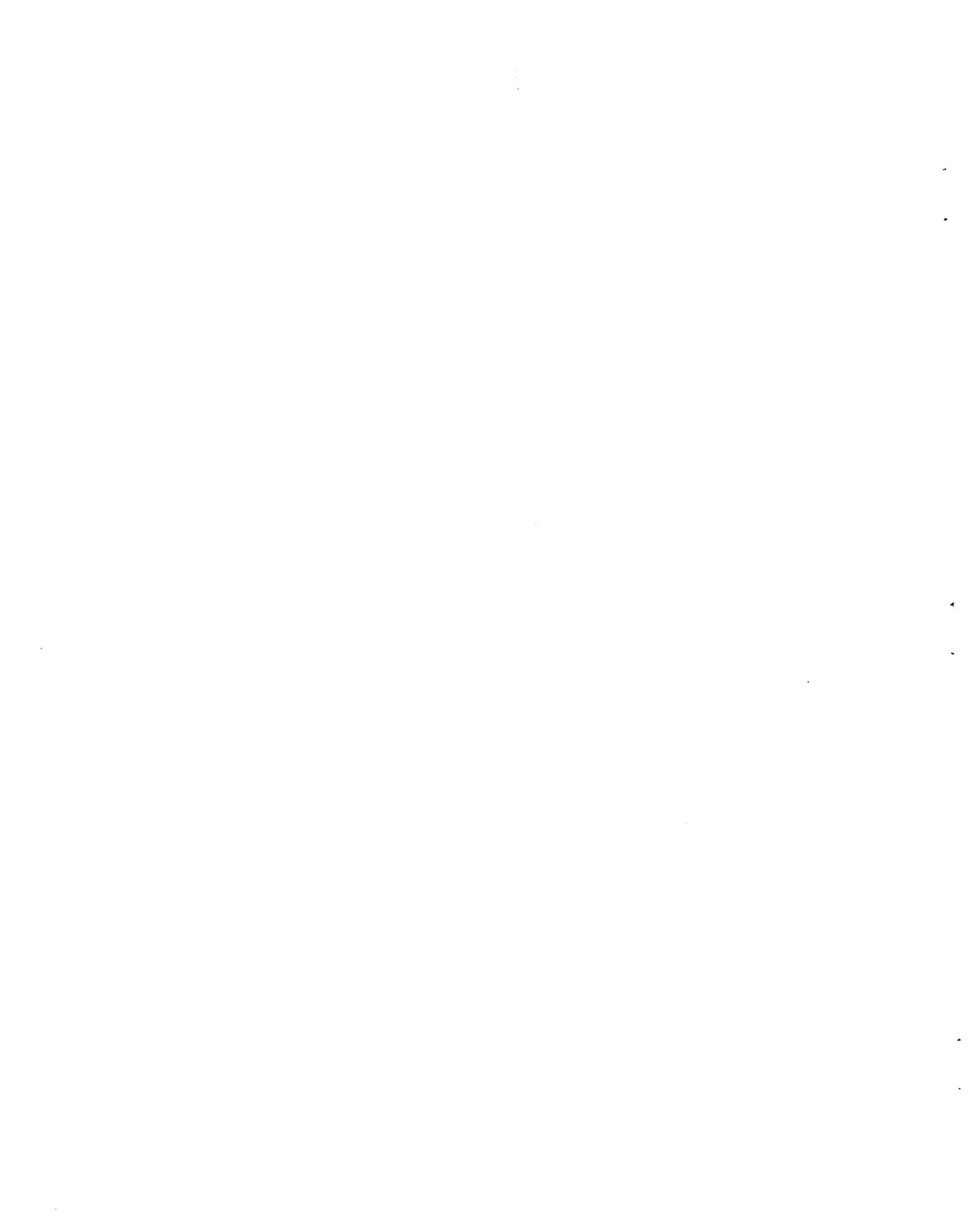
<sup>a</sup>Assumed reprocessing capacity: 20,000 F.E./yr (11,600 of 25R, 7,800 of 23R, and 600 of 25W).

<sup>b</sup>The specific radioactivity (CI/gal) and specific heat generation rate (watts/gal) are based on unconcentrated aqueous solutions.

<sup>c</sup>Numbers in parentheses represent powers of 10.

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