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Prepared for

Working Group 7

of the

International Nuclear Fuel Cycle Evaluation

September 1979

INFCE/DEP/WG.7/3
September 1979

A B S T R A C T

WASTE ARISINGS FROM A HIGH-TEMPERATURE REACTOR WITH A URANIUM-THORIUM FUEL CYCLE

This paper presents an equilibrium-recycle condition flow sheet for a high-temperature gas-cooled reactor (HTR) fuel cycle which uses thorium and high-enriched uranium (93% U-235) as makeup fuel. INFCE Working Group 7 defined percentage losses to various waste streams are used to adjust the heavy-element mass flows per gigawatt-year of electricity generated. Thorium and bred U-233 are recycled following Thorex reprocessing. Fissile U-235 is recycled one time following Purex reprocessing and then is discarded to waste. Plutonium and other transuranics are discarded to waste. Included are estimates of volume, radioactivity, and heavy-element content of wastes arising from HTR fuel element fabrication; HTR operation, maintenance, and decommissioning; and reprocessing spent fuel where the waste is unique to the HTR fuel cycle.

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WASTE ARISINGS FROM A HIGH-TEMPERATURE REACTOR WITH A URANIUM-THORIUM FUEL CYCLE

1. INTRODUCTION

The high-temperature gas-cooled reactor (HTR) described in this paper is an advanced converter system using a closed uranium-thorium fuel cycle. This cycle uses thorium and 93% enriched U-235 in the fissile makeup fuel. Thorium is recycled and mixed with makeup thorium to form fertile particles of pure thorium dioxide. The bred U-233 is incorporated into fissile particles and continuously recycled with fertile particles in U-233 recycle fuel elements. Makeup fissile uranium particles are mixed with fertile particles and fabricated into makeup fuel elements. Sufficient unburned U-235 remains in spent makeup fuel elements to warrant recovery and recycle to the reactor one additional cycle prior to retirement and disposal as waste. These recycle U-235 fissile particles are mixed with fertile particles and fabricated into recycle U-235 fuel elements. All fuel elements have essentially the same characteristics such as dimensions, reactivity, and graphite-to-heavy metal ratios.

The particle configuration of the fuel permits physical separation of the fertile particles from the fissile particles. The fissile particles from the spent makeup elements are treated in a Purex reprocessing plant to recover the partially depleted U-235 and the minor amount of plutonium (mostly Pu-238) is sent to waste with the fission products and other transuranics. The spent U-235 recycle fissile particles are not processed but are sent to waste as silicon carbide coated particles after physical separation from fertile particles. All fertile particles and the fissile particles from the U-233 recycle elements are processed in a Thorex reprocessing plant to recover a purified U-233 and thorium product. The refabrication plant processes all but the makeup fissile uranium particles and fabricates all three types of fuel elements which, in turn, require shielded carriers for shipment to the reactor.

Presented first is the equilibrium recycle condition HTR fuel cycle flow sheet which incorporates the agreed Working Group 7 assumptions regarding percentage losses to various waste streams. Following that, estimates are made of waste arisings which are unique to the HTR fuel cycle, i.e., those which were not scaled to one of the reference fuel cycles by the working group.

2. THE HTR FUEL CYCLE FLOW SHEET

A detailed study of the reactor lifetime mass flow information shows that the HTR only slowly approaches a quasi-equilibrium condition. In a high-gain converter with self-generated recycle, the amount of fissile material recycled in successive cycles over the reactor lifetime increases with time and the amount of makeup fissile material decreases accordingly. The fuel cycle which is indicated

here is an average of the latter 15 years of reactor operation out of a 26-year lifetime. Adjustments were made to incorporate the agreed Working Group 7 assumptions regarding percentage losses to various waste streams and to balance the reactor fissile uranium output to the reactor fissile uranium input plus the losses to waste streams involved in fuel reprocessing and refabrication. The waste arisings as Working Group 7 views them should very closely approximate this "average" since there will be a number of these reactors of all ages discharging fuel in a given year. The actual waste streams should, in fact, reflect this average.

2.1. Working Group 7 assumptions regarding losses to various waste streams

Working Group 7 adopted a number of standard assumptions regarding losses to various waste streams expressed as a percentage of the process input. Those percentage losses which were pertinent to the HTR flow sheet are listed below:

Thorium mining and milling	10% Th
Uranium mining and milling	5% U
Chemical conversions and fuel fabrication	
Ammonium diuranate (ADU) to UF_6	0.4% U
UF_6 to UO_2	0.5% U
Miscellaneous fuel fabrication	0.1% U and Th
Enrichment plant maintenance	0.02% U
Enrichment plant tails (natural feed, 93% enrichment, and 0.2% tails assay)	99.45% U
Reprocessing - total	100% Pu / 1.0% U and Th
Vitrified HLW including undissolved fuel particle coatings	95% Pu / 0.8% U and Th
Medium level waste (MLW)	5% Pu / 0.2% U and Th

2.2. The quasi-equilibrium HTR fuel cycle

Figure 1 shows the heavy element flow sheet per gigawatt-year (GWa) of electricity generated for the high-temperature reactor operating on a uranium-thorium cycle with recycle of both uranium and thorium, HEU fissile makeup (93% U-235), and plutonium discard to waste.

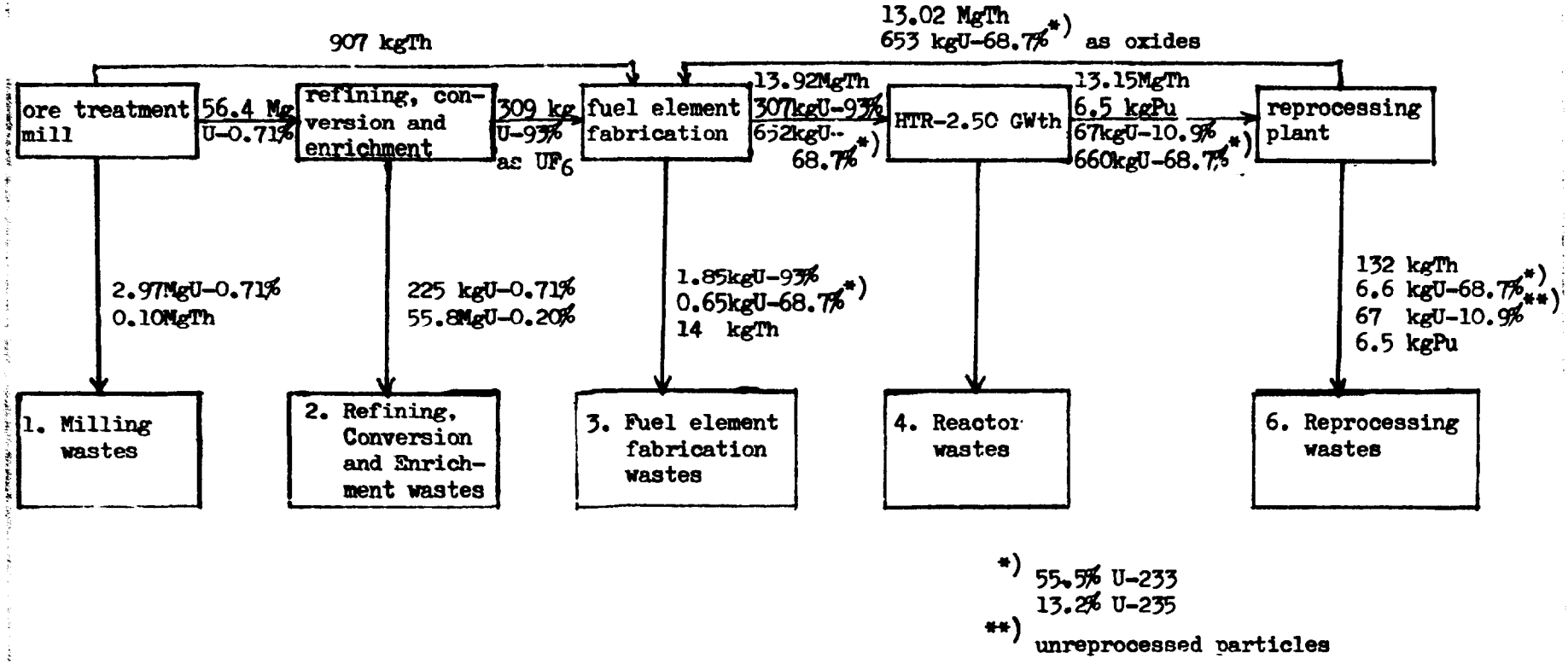
3. THE HTR FUEL CYCLE WASTE ARISINGS

As the tables of waste arisings were being developed by Working Group 7, two methods of estimating values emerged. The first involved wastes arising from fuel cycle operations which were essentially identical for all of the fuel cycles containing that operation. In this case, one fuel cycle was chosen as a reference and values for other fuel cycles were scaled to some specified fuel cycle parameter. The second involved waste arisings the values of which were not found to be dependent upon a fuel cycle parameter for all fuel cycles involved.

Fig. 1

FUEL CYCLE 7: HIGH-TEMPERATURE REACTOR WITH U-Th-CYCLE

HEAVY ELEMENT FLOW SHEET PER GIGAWATT-YEAR ELECTRICITY



*) 55.5% U-233
13.2% U-235

**) unreprocessed particles

3.1. Data scaled to a fuel cycle parameter for all fuel cycles

For those waste streams from the HTR fuel cycle which were scaled by some fuel cycle parameter to a reference value, the Working Group 7 report identifies the method used to determine these waste arisings. The wastes in the HTR fuel cycle which fall into this category are simply listed here for identification. They are: ore processing wastes (both uranium and thorium mill tailings); refining, conversion, and enrichment wastes; and, for reprocessing wastes, the volume of vitrified high-level waste (HLW) and both the volumes and radioactivities of medium-level (MLW) and plant maintenance waste, low-level waste (LLW), and plant decommissioning waste.

3.2. *HTR waste arisings not scaled to a fuel cycle parameter*

The following sections identify the waste arisings for the HTR fuel cycle which were not scaled to a fuel cycle parameter. Values for the volumes, heavy element contents, and radioactivities of these wastes are given.

3.2.1. Fuel element fabrication wastes

The volume is estimated to be 35 m^3/GWa when concreted in steel drums. This includes process wastes from both the UF_6 to UO_2 chemical conversion and the fabrication of fertile and fissile fuel particles. The heavy element content is as follows:

0.1% of the total thorium input from both the reprocessing plant and the thorium mill (14 kg Th/GWa),

0.1% of the recycle uranium from the reprocessing plant (0.65 kg U/GWa containing 55.5% U-233 and 13.2% U-235), and

0.6% of the fissile makeup HEU from the enrichment plant (1.85 kg U/GWa containing 93% U-235).

3.2.2. Reactor wastes

Working Group 7 identifies four classes of waste arising from the operation, maintenance, and decommissioning of reactors.

3.2.2.1. Waste from operation. During the operation of the HTR, most of the fission products generated remain within the coated fuel particles. However, small quantities may escape through the pyrolytic graphite coatings into the graphite structure of the fuel elements and then diffuse into the primary coolant helium. A helium purification system is provided to remove these fission products as well as chemical impurities from the primary coolant system. The coolant purification process produces gaseous wastes which are in turn handled by a radioactive gas waste system.

Some radioactive liquid waste results from diffusion of tritium into the secondary coolant system. Other liquid wastes include condensate from the helium-purification cooler, decontamination fluid, laundry and contaminated-shower liquid waste, and floor drains. The liquid wastes are processed through ion-exchange units until the activity level is low enough to permit release from the plant. Radioactive filter elements, filter sludges, reverse osmosis concentrates, and spent ion-exchange resins are incorporated into cement for disposal.

Dry material that becomes contaminated from use during plant operation is compressed into 200-liter drums by means of a hydraulically driven compactor. Spent titanium sponge is vacuumed from hydrogen-getter units, incorporated in cement, and deposited into shielded drums. Spent demineralizer resins, diatomite filter cake, and soda-lime absorbent from the carbon dioxide absorber in the radioactive gas recovery system is incorporated in cement and put into unshielded drums. Noncompactible items such as failed equipment are cut into pieces small enough to fit into drums.

The quantities of reactor operating waste shipped for each 1 GWa of electricity generated are as follows:

	m^3/GWa	drums/GWa
Cemented liquids and solids	325	1625
Compactible/combustible trash	321	1605
Noncompactible/noncombustible trash	34	170
Total HTR operating waste	680	3400

The total radioactivity is estimated to be 3 kCi/GWa at one year. The division of the drums between shielded and unshielded was taken to be the same as for LWR operation (10% shielded and 90% unshielded). This results in 340 shielded drums/GWa and 3060 unshielded drums/GWa.

3.2.2.2. Maintenance waste. The maintenance wastes are associated primarily with the decontamination of equipment (fuel-handling machinery and control rod drives following plant refueling, associated systems, circulators, etc.). The volume when packaged in shielded drums is estimated to be 9 m^3/GWa with a total radioactivity of 0.2 kCi/GWa at one year.

3.2.2.3. Control rods, etc. Included in this category is the average volume of reactor items which need replacement along with the associated radioactivity after decontamination. The volume is 5 m^3/GWa with a total radioactivity of 1 kCi/GWa at one year. When packaged in HWR canisters that contain 0.60 m^3 of waste each, 8 canisters/GWa are required.

3.2.2.4. Decommissioning waste. The decommissioning wastes are based on removal of the reactor core reflector, reactor auxiliary equipment such as helium-purification systems, core auxiliary cooling systems, steam generators, circulators, and other reactor system components, along with disposal of the above components and decontamination of remaining plant components. The volume is estimated to be 30 m^3/GWa with a total radioactivity of 5 kCi/GWa at 25 years. Shielded drums are used for all of this waste.

3.2.3. Reprocessing wastes

Many of the fuel cycles considered by Working Group 7 involve fuel rods or pins in which a metallic cladding of considerable volume is present and remains in reasonably large, undissolved pieces after the dissolution of the fuel. In the HTR cycle, however, the fuel elements contain a large amount of carbon (graphite) which is burned off as carbon dioxide. This is a C-14-contaminated waste stream that does not appear in any other reference fuel cycle. Burning exposes the fissile and fertile particles (microspheres) which are then separated in a particle classifier by elutriation using an inert gas stream. The larger and denser fertile particles consisting of carbon-coated thorium dioxide (and the bred U-233) exit the bottom of the classifier and are sent to a dissolution step for preparation of Thorex solvent extraction feed. The fissile particles are carried overhead in the gas stream.

Of the three different types of fissile particles present in the HTR fuel cycle, only one type of fissile particle is present in a given fuel element. Spent fissile particles from recycle U-235 fuel elements are retired after physical separation from fertile particles. These unreprocessed, twice-through U-235 particles constitute a waste which is assumed to be added to the vitrified HLW.

Prior to dissolution, the other two types of fissile particles which are coated with silicon carbide must be crushed to expose the inner fuel kernel and burned to convert uranium oxycarbide (UCO) fuel to U_3O_8. Following dissolution, the undissolved fissile kernel coatings of silicon carbide and any undissolved fission product or actinide residues are removed from the dissolver solution by filtration and are assumed to be added to the vitrified HLW.

Working Group 7 has assumed a 1.0% loss of heavy elements being recycled in the reprocessing plant wastes. Furthermore, this loss is apportioned between the various reprocessing wastes for most of the fuel cycles as 0.3% to hulls, spacers, insolubles; 0.5% to vitrified HLW; and 0.2% to MLW. For the HTR cycle, however, a separate waste stream identified as hulls, spacers, insolubles has not been used. Rather, the normal 0.3% loss to this waste is combined with the normal 0.5% loss to vitrified HLW, the result being a loss of uranium (either once-through U-235 or recycle U-233) and thorium to the vitrified HLW amounting to 0.8%. The usual 0.2% of uranium and thorium being recycled is lost to the MLW.

All of the plutonium is discarded as waste. The initial Thorex separation results in less than complete separation of thorium and uranium from plutonium, other transuranics, and fission products. Up to 5% of the plutonium may be carried over with the uranium and thorium and subsequently removed in the second extraction step where the waste is discarded as MLW. The split of plutonium between vitrified HLW and MLW has been taken to be 95% and 5%, respectively. This choice is not particularly sensitive from a safeguards analysis standpoint because the quantities of plutonium involved are so small (only 6.5 kg Pu/GWa) and the volumes of waste in which it is being placed are so large.

3.2.3.1. Vitrified HLW. The volume of vitrified HLW for all fuel cycles was scaled to that of the LWR by assuming an equal mass concentration of fission products in the glass. The 10-year radioactivity of the HLW is calculated to be 10.5 MCi/GWa. The heavy element content of the vitrified HLW is as follows:

95% of the plutonium (6.2 kg Pu/GWa),

0.8% of the fissile uranium and thorium being recycled (106 kg Th/GWa and 5.3 kg U/GWa containing 55.5% U-233 and 13.2% U-235), and

all of the unreprocessed, twice-through U-235 fissile particles (67 kg U/GWa containing 10.9% U-235).

3.2.3.2. Medium-level and plant maintenance waste. The numbers of drums and contained radioactivity are calculated in proportion to the heavy metal throughput of the reprocessing plant with the plant serving the HWR on plutonium recycle taken as the reference waste producing facility. The heavy element content of this waste is as follows:

5% of the plutonium (0.3 kg Pu/GWa) and

0.2% of the fissile uranium and thorium being recycled (26 kg Th/GWa and 1.3 kg U/GWa containing 55.5% U-233 and 13.2% U-235).

The carbon-14-contaminated waste described in the next paragraph must be included with this waste stream for the HTR fuel cycle only.

3.2.3.3. Carbon-14-contaminated waste. Two processes for recovering carbon dioxide (CO_2) from reprocessing plant off-gas streams and fixing it as a stable solid compound have been identified and studied. The first process involves contacting the off-gas containing CO_2 with an aqueous slurry of calcium hydroxide (slaked lime). The reaction product is solid calcium carbonate ($CaCO_3$) which is removed from the slurry by filtration, dried to the point that there is no free water, and immobilized and/or packaged for disposal. The second process involves passing the humidified off-gas through a packed or fluidized bed of barium hydroxide. The reaction product is solid barium carbonate ($BaCO_3$) which is physically or pneumatically removed from the process vessel and immobilized and/or packaged for disposal. Both of

these processes have been investigated on a cold laboratory scale to some extent, and both appear to work well. The disadvantage of the calcium process is that the handling of slurries can be messy and filtration is required. The barium process eliminates these concerns, but the barium hydroxide reagent is substantially more expensive than the lime used in the calcium process.

Immobilization processes have not been investigated for either of these processes. Concrete or bitumen (asphalt) appear to be the most likely candidates—concrete is assumed here because of its widespread use with other wastes. Although the concrete will always result in a waste product weight increase, it may result in a volume reduction if the unconcreted product has a very low density. Such is the case with the $BaCO_3$ product. It should be noted that most of the densities used in characterizing the volumes of the final products, both concreted and unconcreted, are estimates based on little or no experimental data and are subject to change. These estimates are as follows:

C-14-contaminated product	Density, Mg/m^3	Volume, m^3/GWa
$CaCO_3$ - as produced	2.0	670
$CaCO_3$ - concreted	3.0	670
$BaCO_3$ - as produced	0.67	2300
$BaCO_3$ - concreted	3.0	1020

The C-14 radioactivity is 0.15 kCi/GWa. Working Group 7 selected $CaCO_3$ concreted in unshielded steel drums for the disposal of the C-14-contaminated waste. An additional 3350 unshielded drums/GWa are reflected in the MLW and plant maintenance waste stream for the HTR fuel cycle.

3.2.3.4. Radioactivity in reprocessing wastes. The reprocessing wastes contain the following isotopes and activities ten years after discharge from the reactor. Included are 1% of the thorium and uranium being recycled, all of the fission products and transuranics, and all of the unreprocessed, twice-through U-235 fissile particles.

Kr-85	2.7 E 5	Th-228	4.0 E 1	Pu-238	6.8 E 4
Sr-90	2.1 E 6	Th-229	3.6 E-2	Pu-239	5.6 E 1
Tc-99	4.9 E 1	Th-230	1.0 E-1	Pu-240	9.5 E 1
I-129	7.0 E-1	Th-232	1.5 E-2	Pu-241	2.6 E 4
Cs-134	1.6 E 5	U-232	4.3 E 1	Pu-242	1.6 E 0
Cs-135	4.7 E 0	U-233	3.6 E 1	Am-241	5.6 E 2
Cs-137	2.0 E 6	U-234	9.4 E 0	Am-242m	2.2 E 0
Pm-147	1.5 E 5	U-235	1.8 E-2	Am-243	2.4 E 1
Eu-154	3.7 E 4	U-236	3.0 E 0	Cm-242	1.8 E 0
		U-238	4.3 E-3	Cm-243	3.8 E 0
		Np-237	8.5 E 0	Cm-244	3.1 E 3