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MATERIAL CONTROL AND ACCOUNTABILITY ASPECTS OF SAFEGUARDS FOR THE USA $^{233}U/Th$ FUEL RECYCLE PLANT*

J. A. Carpenter, Jr., S. R. McNeany, and P. Angelini
Oak Ridge National Laboratory, Oak Ridge, Tennessee

N. D. Holder and L. Abraham
General Atomic Company
San Diego, California

ABSTRACT

The material control and accountability aspects of reprocessing and refabrication in a large-scale High-Temperature Gas-Cooled Reactor (HTGR) fuel recycle plant are discussed. Two fuel cycles are considered. The highly enriched uranium (HEU) cycle uses uranium enriched 93% in ^{235}U as the initial fuel. The medium enriched uranium (MEU) cycle uses uranium with a ^{235}U enrichment less than 20% as its initial fuel. In both, ^{233}U is bred from thorium. The HEU ^{235}U and the ^{233}U of both cycles are recycled. The MEU ^{235}U is retired to waste after one reactor cycle. Typical heavy metal contents of spent fuel elements from both cycles are presented.

The main functional areas of the recycle plant are Shipping, Receiving, and Storage; Reprocessing; Refabrication; and Waste Treatment. A real-time materials accountability system will manage the data provided by measurements from all four areas. Simulations of material flow used in the HTGR development program are forerunners of such a system.

The material control and accountability aspects of Reprocessing and Refabrication only are discussed. The proposed accountability areas are identified and the measurement techniques appropriate to various streams crossing the boundaries of the areas are identified. Special emphasis is placed on novel nondestructive methods developed for assaying solid materials containing ^{233}U-Th. The material form, total uranium and plutonium, and activity of selected reprocessing streams are listed. The isotopics and activity of the uranium input into Refabrication are also presented.

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INTRODUCTION

The U.S. HTGR Recycle National Program

The objective of the U.S. High-Temperature Gas-Cooled Reactor (HTGR) ^{233}U-Th fuel recycle program is the design and licensing of a large-scale demonstration recycle plant to be built and operated in the time frame of 1995–2000. Heretofore, emphasis of the program [1] was on the development of the recycle technology, much of it done in cooperation with the United Kingdom and the Federal Republic of Germany. The development effort has now progressed to the stage in which almost all the process steps of reprocessing and refabrication have been demonstrated in prototypic equipment with natural or depleted uranium. While this development work progressed, conceptual design studies of such a recycle plant were conducted and included materials control and accountability.

The fuel cycle

The general flow of materials for the HTGR ^{233}U-Th fuel cycle is indicated in Fig. 1. Enriched uranium and thorium are fabricated into elements in a fresh fuel plant and sent to the reactor. The spent fuel is sent to the recycle plant, where it is reprocessed to recover the fissile ^{233}U produced from the thorium and, in some cases, the residual ^{235}U. These fissile materials are combined with fresh thorium and refabricated into recycle elements, which are shipped back to the reactor. The unrecovered fissile material and other wastes are processed in waste treatment and sent to a repository. The spent thorium is stored for later use.

This paper examines the two nuclear fuel cycles of primary interest for implementation with the HTGR. The first is called the highly enriched uranium (HEU) cycle. It has the best economic performance and resource utilization and traditionally has been the prime candidate for HTGR use. However, in response to proliferation concerns, a number of

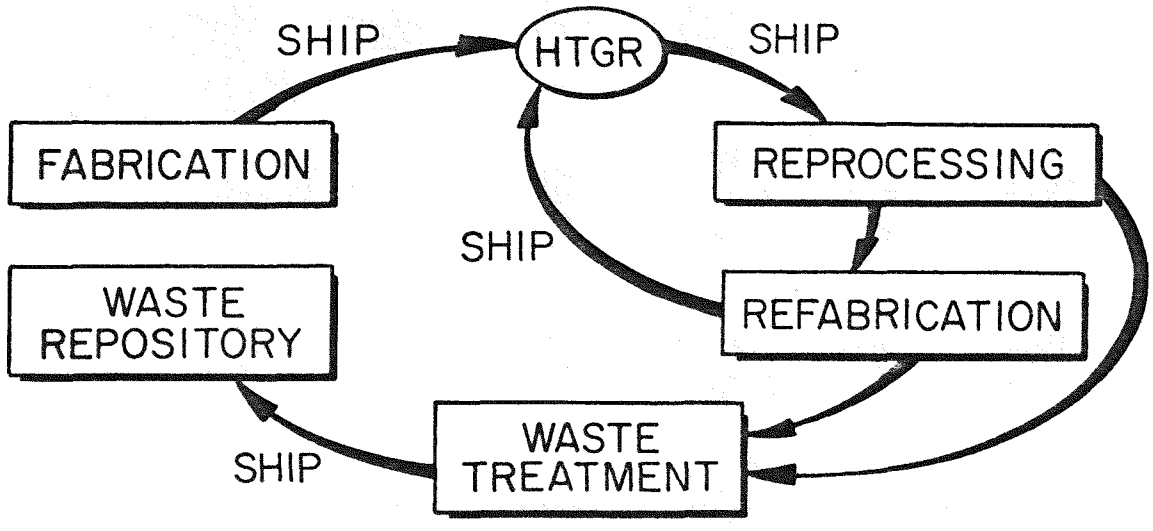


Fig. 1. Fuel Cycle.

alternate cycles have been examined. A fuel cycle that may provide more proliferation resistance is the medium enriched uranium (MEU) cycle; however, its economic performance is not as good as that of the HEU cycle. A low enriched uranium (LEU) cycle has also been considered. As there would be no recycle of the fuel at all, this cycle is not considered in this paper.

Scope of paper

Shipping, receiving, and storage; reprocessing; refabrication; and waste treatment are the main functions of the recycle plant. The majority of the materials control and accountability problems are in reprocessing and refabrication and it is there that most of our efforts have been placed; therefore, only reprocessing and refabrication are discussed. Item control will be used in shipping, receiving and storage. The materials control and accountability aspects of waste treatment are being defined. Physical security aspects are not addressed, although they have been and are being considered in our studies.

GENERAL SAFEGUARDS CONSIDERATIONS FOR THE ENTIRE FUEL RECYCLE PLANT

The fuel and the fuel cycle

The HTGRs and HTGR fuel recycle have been reviewed in detail elsewhere [2—5]. The U.S. design of the General Atomic Company uses a hexagonal graphite block 0.79 m high and 0.36 m across the flats as its fuel element. The fuel and the fuel element are depicted in Fig. 2. The fuel is contained in microspheres less than 1000 μm in diameter. The fissile particle containing the initial fuel, ^{235}U or ^{233}U, is coated with three layers of pyrolytic carbon and a single layer of silicon carbide. The fertile particle, containing thorium, from which additional ^{233}U is produced, is coated with two carbon layers only. The two types of particles are bonded by a graphitic matrix to form a fuel rod about 51 to 65 mm long and 13 to 16 mm in diameter. These fuel rods are stacked end-to-end into holes drilled longitudinally through the block parallel to the coolant holes.

Selected heavy metal compositions and characteristics of spent fuel elements are presented in Table I for both the HEU and MEU fuel cycles. The spent fuel compositions are for burnups of about 70,000 MWd per tonne heavy metal (U + Th) for the HEU fuel cycle and 85,000 MWd per tonne heavy metal for the MEU fuel cycle, both cooled 180 days from reactor discharge.

The HEU fuel cycle uses three types of elements. One is the initial or makeup element, produced by the fresh fuel plant. This contains uranium highly enriched in ^{235}U (∿93%) as its initial fuel. The other two types of elements are products of the recycle plant. One type contains uranium highly enriched in ^{233}U (∿70%) produced from the thorium in previous reactor cycles. The third type uses uranium containing about 30% ^{235}U, which is the residual of the uranium from previous irradiation of the initial or makeup elements. These three types of elements are designated IM, 23R, or 25R elements to denote elements charged to the reactor or IMS, 23RS, or 25RS to denote spent elements, respectively. The uranium in the fissile particles of the IMS and 23RS elements and in the fertile particles from all three types of elements

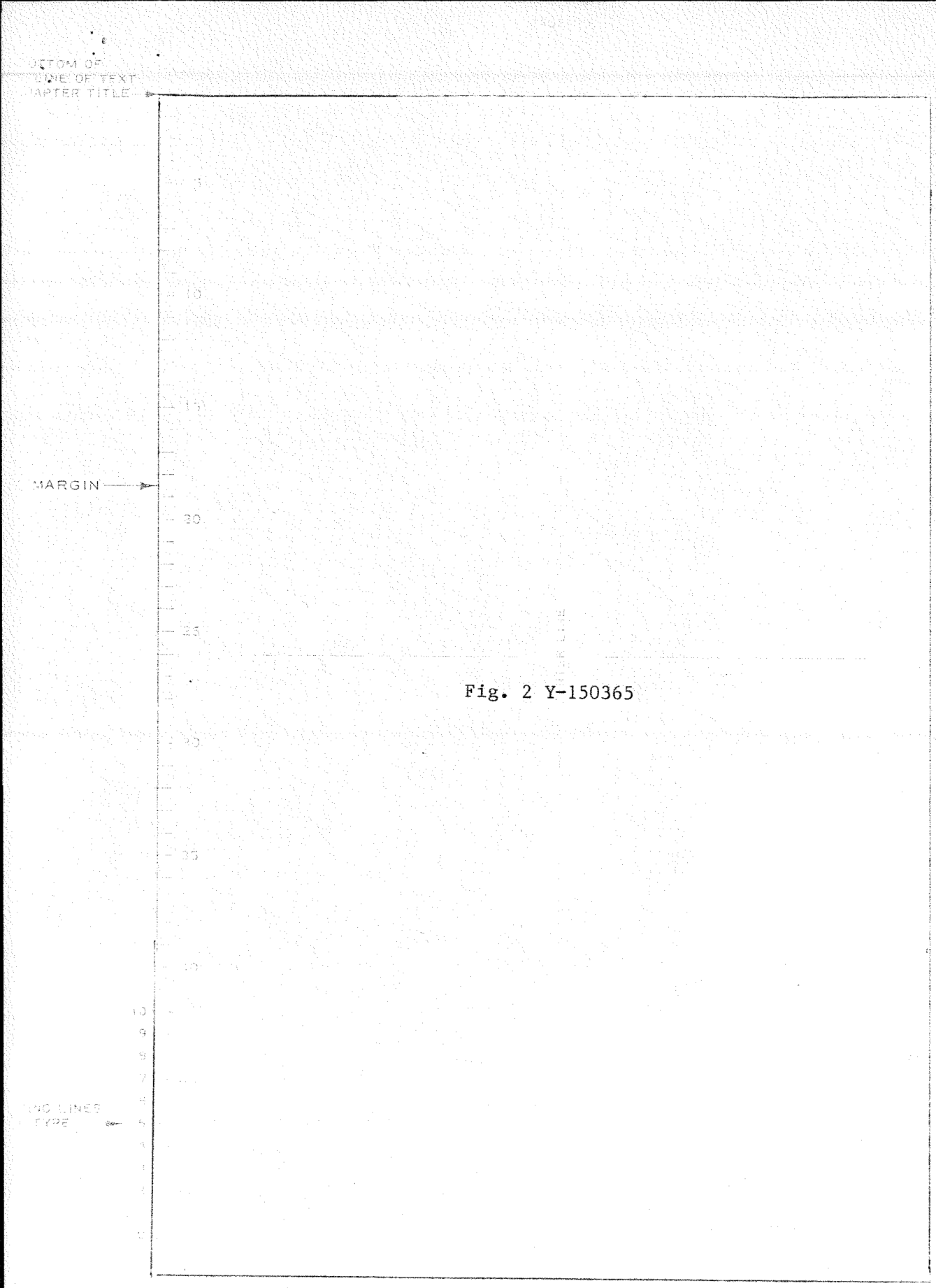
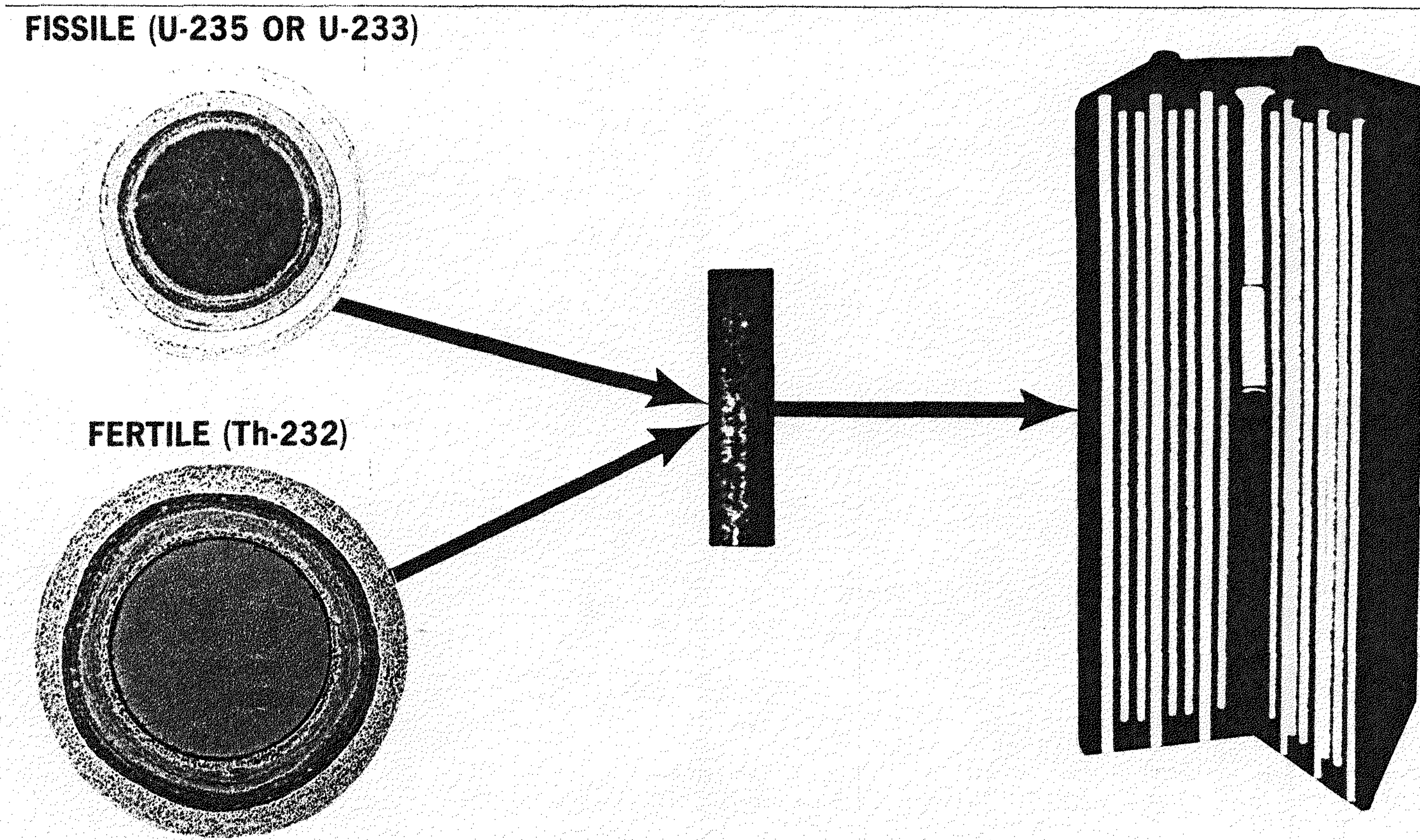


Fig. 2 Y-150365

Fig. 2. HTGR Fuel Components.



FUEL PARTICLES
SCALE: 100 X

FUEL ROD
1 X

FUEL ELEMENT
1/7 X

6 1/4" to 5 1/2"

10" to 8 3/4" → 88% of orig.

P. 4
fig. 2
88% of original size

Table I. Quantities of Selected Heavy Metals in Typical High-Temperature Gas-Cooled Reactor Spent Fuel Elements Cooled 180 Days from Reactor Discharge

	Highly Enriched Uranium (HEU) Fuel Cycle						Medium Enriched Uranium (MEU) Fuel Cycle			
	Initial or Makeup Fuel Element		23R Recycle Fuel Element		25R Recycle Fuel Element		Initial or Makeup Fuel Element		23R Recycle Fuel Element	
	Fissile Particle	Fertile Particle	Fissile Particle	Fertile Particle	Fissile Particle	Fertile Particle	Fissile Particle	Fertile Particle	Fissile Particle	Fertile Particle
Thorium, Total wt/Fuel Element (g)	0.01017	10,550	9.76×10^{-3}	10,550	0.01984	10,550	0.0333	3216.3	2.02×10^{-3}	1,657
-228, wt% of Total Th	0.75275	1.48×10^{-5}	1.266	1.48×10^{-5}	0.2807	1.48×10^{-5}	2.81×10^{-5}	1.36×10^{-5}	46.06	1.36×10^{-5}
-229, wt% of Total Th	0.2322	1.69×10^{-5}	20.03	1.69×10^{-5}	0.0862	1.69×10^{-5}	1.72×10^{-5}	1.72×10^{-5}	15.56	1.72×10^{-5}
-230, wt% of Total Th	91.73	7.89×10^{-5}	78.23	7.89×10^{-5}	34.717	7.89×10^{-5}	1.07×10^{-5}	1.07×10^{-5}	38.38	1.07×10^{-5}
-231, wt% of Total Th	2.04×10^{-4}		1.65×10^{-5}		2.31×10^{-4}					
-232, wt% of Total Th	7.2858	99.99989	0.4731	99.99989	64.913	99.99989	99.994	99.996		99.996
-234, wt% of Total Th	3.55×10^{-4}				2.22×10^{-3}					
Protactinium, Total wt/Fuel Element (g)	6.61×10^{-6}	0.1118	4.23×10^{-4}	0.1118	0.600	0.1118	1.3×10^{-6}	0.1104	1.67×10^{-4}	0.0569
-231, wt% of Total Pa	99.224	67.57	99.951	67.57	85.2	67.57	30.72	20.98	99.9999	20.98
-233, wt% of Total Pa	0.776	32.43	0.049	32.43	14.8	32.43	69.27	79.02		79.02
Uranium, Total wt/Fuel Element (g)	198.5	270.5	174.4	270.5	730.5	270.5	1851.9	109.45	85.135	56.384
-232, wt% of Total U	4.45×10^{-5}	0.0531	6.39×10^{-4}	0.0531	3.91×10^{-6}	0.0531	4.55×10^{-8}	0.0382	5.65×10^{-2}	3.82×10^{-2}
-233, wt% of Total U	1.31×10^{-4}	77.57	8.67	77.57	1.77×10^{-5}	77.57	5.07×10^{-5}	82.956	20.17	82.95
-234, wt% of Total U	8.40	17.34	42.64	17.34	0.7894	17.34	8.66×10^{-6}	14.171	44.44	14.17
-235, wt% of Total U	30.92	4.36	23.81	4.36	8.160	4.36	3.904	2.54	19.49	2.54
-236, wt% of Total U	45.57	0.6844	24.76	0.6844	69.025	0.6844	3.469	0.2956	15.83	0.2956
-238, wt% of Total U	15.1	9.7×10^{-4}	0.1144	9.7×10^{-4}	22.026	9.7×10^{-4}	92.624	1.08×10^{-5}	1.96×10^{-3}	1.08×10^{-5}
Neptunium, Total wt/Fuel Element (g)	13.07	0.1083	5.979	0.1083	0.1589	0.1083	8.5603	0.01836	2.114	9.46×10^{-3}
-237, wt% of Total Np	99.9999	99.9999	99.9999	99.9999	99.9999	99.9999	99.9997	99.9999	99.9999	99.9999
Plutonium, Total wt/Fuel Element (g)	8.537	0.02912	3.551	0.02912	61.21	0.02912	48.647	4.07×10^{-3}	1.2093	2.10×10^{-3}
-238, wt% of Total Pu	61.773	82.546	74.46	82.546	64.55	82.546	7.07	89.80	85.52	89.80
-239, wt% of Total Pu	16.31	10.686	12.05	10.686	15.00	10.686	37.17	6.27	7.96	6.27
-240, wt% of Total Pu	9.30	4.104	6.5	4.104	8.75	4.104	16.39	1.65	2.86	1.65
-241, wt% of Total Pu	6.48	2.000	4.25	2.000	6.17	2.000	20.20	1.03	2.57	1.03
-242, wt% of Total Pu	6.14	0.6621	2.73	0.6621	5.55	0.6621	19.16	1.25	1.09	1.25
Total Weight of Fuel Element (g)	120,600		119,900		122,700		115,180		110,050	
Total Activity of Fuel Element (Ci)	56,450		53,100		61,720		67,310		22,300	

is recovered in the recycle plant. The fissile particles from the 25RS elements, containing uranium that is about 8% ^{235}U but about 70% ^{236}U, are retired to waste.

The MEU cycle uses only two types of elements, the IM element and the 23R element containing ^{233}U (70%).* The IM element contains uranium with an enrichment less than 20% ^{235}U. This burns down to 4% ^{235}U in the spent fuel element, so the fissile particles of the IMS elements are discarded and not recycled. The uranium in the 23RS elements and that in the fertile particles of the IMS elements are recycled.

The Fuel Recycle Plant

The overall flow of material within the recycle plant is indicated in Fig. 3. The spent fuel elements enter the plant in Shipping, Receiving, and Storage and are stored before delivery to Reprocessing.

*Another MEU cycle, in which the recycle 23R uranium is denatured to less than 12% ^{233}U has also been considered but is not the current reference.

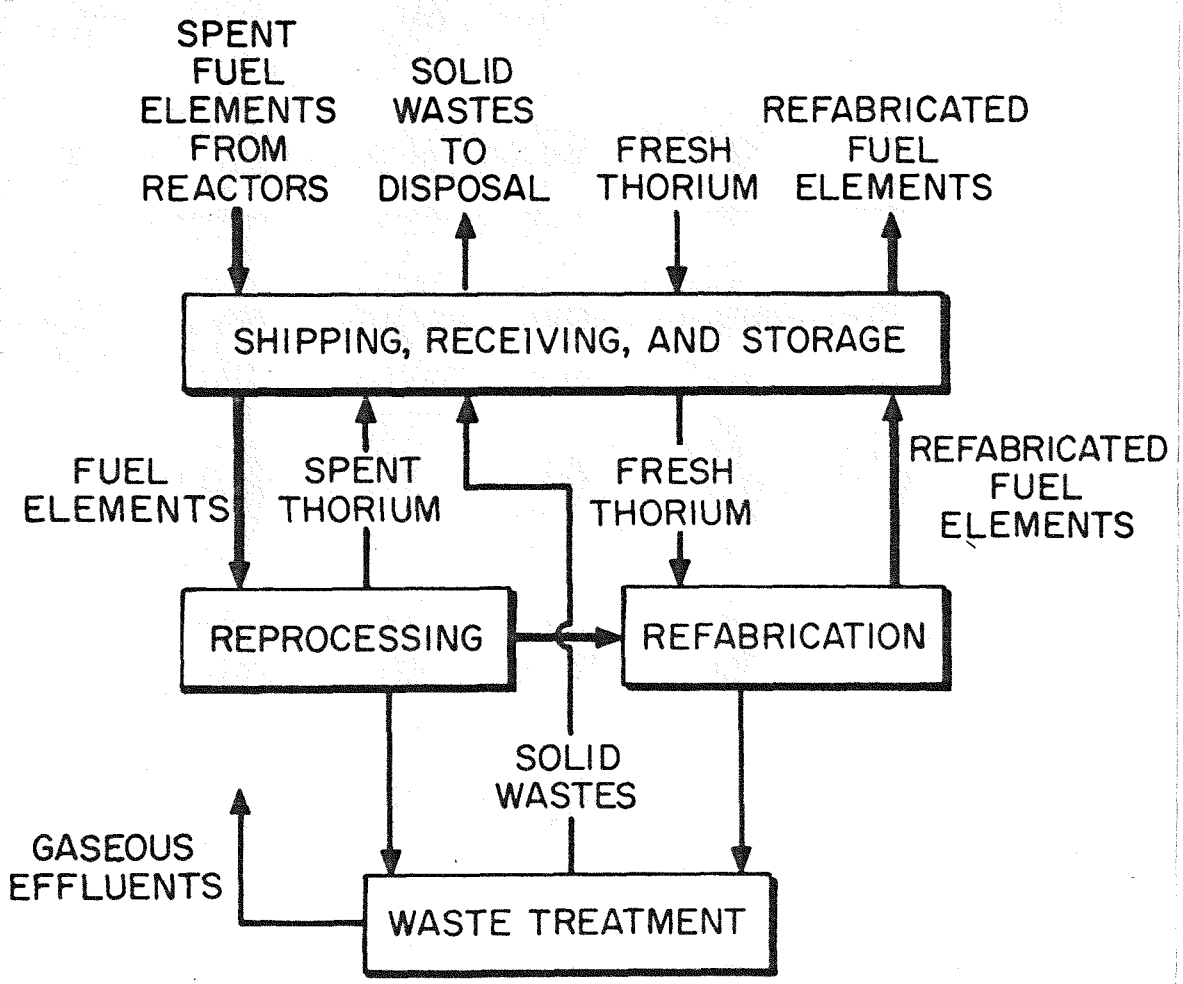


Fig. 3. HTGR Fuel Recycle Flowsheet.

From Reprocessing recovered fissile material is delivered to Refabrication. Retired fissile material and all wastes are sent to Waste Treatment. The spent thorium is solidified and placed in storage for 20 to 30 years to allow its radioactivity to decay before recycling.

In Refabrication the recovered fissile material is joined by fresh thorium in the form of coated particles, and these are fabricated into recycle elements, which are stored and eventually sent to the reactor. Refabrication wastes are also sent to Waste Treatment.

In Waste Treatment all solid and liquid and some gaseous wastes are processed to a repository-ready solid waste form. The remaining gaseous effluents are treated and vented to the atmosphere. Liquid effluents from the plant contain no nuclides from the fuel elements. The solid wastes are stored and ultimately delivered to a waste repository.

Because of the inherent radioactivity associated with most of these streams, virtually all process operations in all four main areas of the plant will be done remotely.

General Plant Material Control and Accountability

The HTGR Fuel Recycle Plant will incorporate the latest state-of-the-art techniques to implement a highly effective safeguards program. Safeguards are maintained by the inherent radioactivity of the fuel, the physical barrier of the required heavy shielding, an integrated system of measurement including destructive analysis of samples and nondestructive assay, and physical security. The data collected will be managed by a real-time accounting system similar to the DYMAC Program [6,7] developed at Los Alamos Scientific Laboratory (LASL). A schematic of the proposed system is shown in Fig. 4. A central computer collects and analyzes instrument data and operator-supplied information to continuously update the recorded status of material locations throughout the plant. Simultaneous computer simulations of plant operations continuously calculate expected amounts of material in various parts of the plant. The two values are continuously compared.

In support of the development of real-time accounting capability, several material flow models [8,9] have been developed to simulate expected mass flow patterns throughout the recycle plant. One of these, which calculates average fissile mass movements, has been used to determine accuracy requirements of measurement devices [10] to meet U.S. government material accountability standards. Other studies have yielded the time-dependent variations of these flows. Simulations of reprocessing operations are under way at General Atomic Company using the GASP IV simulation language, and similar simulations of the refabrication operations are scheduled for the near future at the Oak Ridge National Laboratory (ORNL). Total nuclide flows and the associated radioactivity through the recycle plant have been calculated at ORNL with isotope-depletion codes ORIGEN [11] and ORIGEN2 [12].

DETAILED MATERIAL CONTROL AND ACCOUNTABILITY ASPECTS OF REPROCESSING

The general flowsheet for the operations involved in reprocessing of spent HTGR fuel is shown in Fig. 5. The proposed accountability areas are indicated by the dashed lines. Material form, total uranium and plutonium, and activity for selected streams are presented in Table II. The relative attractiveness (or unattractiveness) depends more on

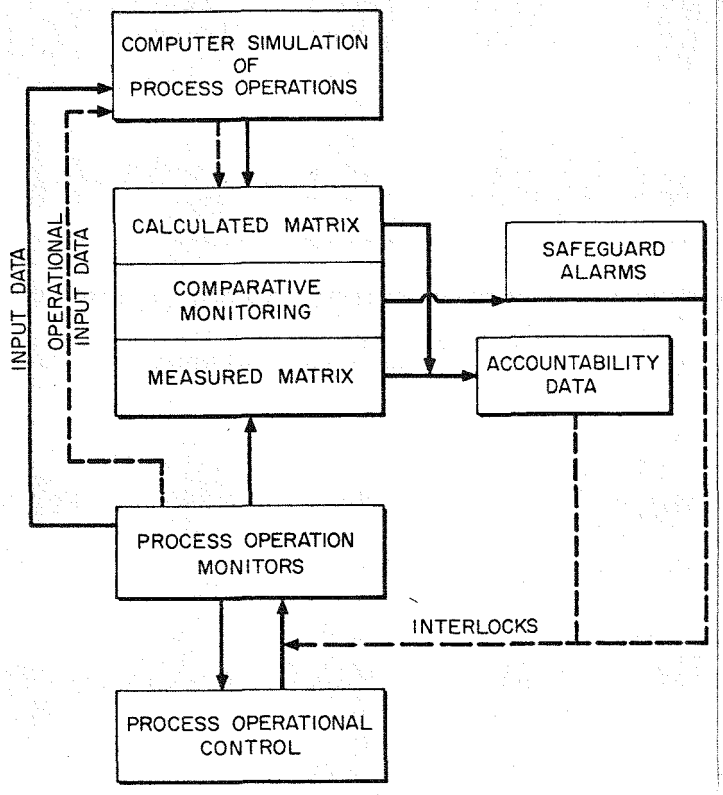


Fig. 4. Real-Time Materials Accountability Concept by Integration of Process Simulation and Monitoring.

dose-rate than simply on activity, but as the former depends upon geometry, the specific energies of the emitted radiation, and matrix, only activity is reported.

Reprocessing is divided into wet and dry head-end and solvent extraction. The entire head-end constitutes one large accountability area. Dry head-end consists of Primary Crushing, where the fuel elements are reduced to 5-mm-diam granules; Primary Burning, where the graphite fuel block and exposed fuel particle carbon coatings are burned away in a CO_2-O_2 atmosphere; Particle Classification, where the silicon-carbide-coated fissile particles are separated from the burned-back fertile kernels; and, for the ^{235}U particles of the HEU IMS elements and ^{233}U recycle fissile particles of the 23RS elements, Secondary Crushing and Burning to crack the silicon carbide coatings and burn away the remaining carbon. The fissile particles of the HEU 25RS and the MEU IMS elements are retired to waste after Particle Classification. A major safeguard advantage in the MEU flowsheet is that the residual ^{235}U and the plutonium bred from the ^{238}U remain in containment with fission products in intact fissile particles.

Input into the dry head-end consists of whole fuel elements, and output consists of fuel particles and CO_2-bearing off-gas from the burners. Item count identity is lost at the fuel element crushing stage. By well-planned administrative controls, material can be batched and total mass accountability maintained by weighing before and after each process step. The burning step will also require CO_2 measurements and calculations of the quantities of carbon removed in the

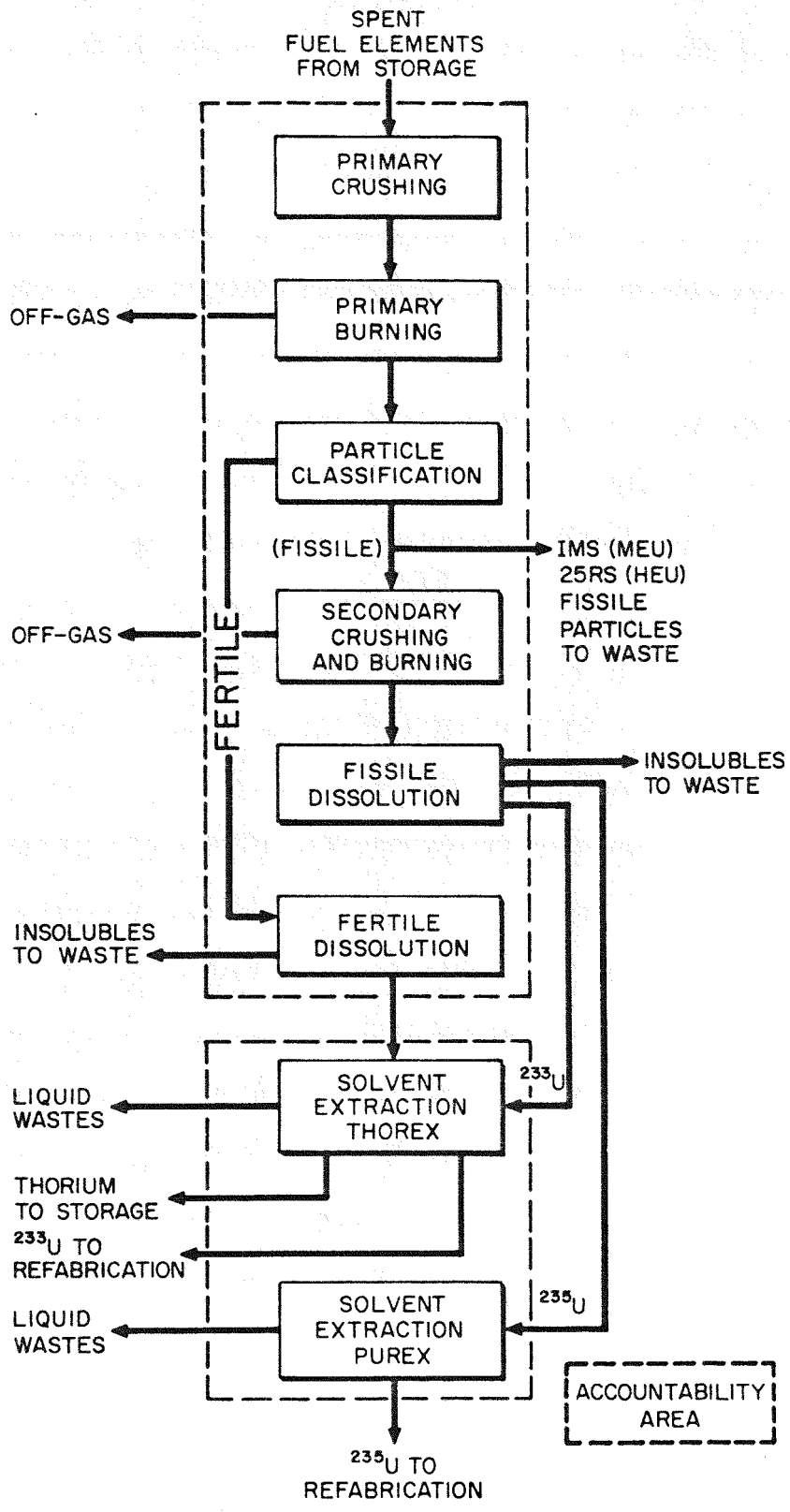


Fig. 5. HTGR Reprocessing Flowsheet.

Table II. Characteristics of Main Nuclide Streams HTGR Reprocessing HEU and MEU Fuel Cycles Fuel Cooled 180 Days from Reactor Discharge

Stream	Description[a]	Type of Element	Uranium Content of Stream, wt. %, Based on		Plutonium Content of Stream, wt. %, Based on		Specific Activity of Nuclides in Stream (Ci/g)	Specific Neutron Emission Rate (n/s·g)
			Fuel-Element-Derived Material	Uranium Input to Plant	Fuel-Element-Derived Material	Plutonium Input to Plant		
Spent fuel elements	Whole graphite blocks	HEU-IMS	0.39	100	0.007	100	4.7 E-1	1.69 E+1
		-25RS	0.82	100	0.050	100	5.0 E-1	9.73 E+1
		-23RS	0.37	100	0.003	100	4.4 E-1	2.5
		MEU-IMS	1.70	100	0.042	100	7.5 E-1	2.05 E+2
		-23RS	0.13	100	0.001	100	2.7 E-1	5.80 E+1
Retired fissile particles	Burned-back fissile particles	HEU-25RS	9.51	72.4	0.79	98.1	3.68	1.53 E+3
		MEU-IMS	20.8	92.9	0.55	98.2	6.33	2.58 E+3
Fissile dissolution Product solution	Liquid, 24 kg/m^3	HEU-IMS	15.63	43.1	0.647	97.67	1.51 E+1	1.52 E+3
		-23RS	14.66	40.1	0.286	97.2	1.39 E+1	2.15 E+2
		MEU-23RS	14.39	60.1	0.201	97.8	2.02 E+1	1.00 E+4
Insolubles	SiC hulls	HEU-IMS	0.0082	0.045	0.00339	0.102	5.4 E-1	8.0 E-1
		-23RS	0.0095	0.042	0.00018	0.101	5.4 E-1	1.4 E-1
		MEU-23RS	0.0045	0.063	0.00006	0.102	3.3 E-1	3.13
Fertile dissolution Product solution	Liquid, 240 kg/m^3	HEU-IMS	2.13	55.5	0.00024	0.38	2.59	2.28
		-25RS	2.13	26.3	0.00048	0.10	2.59	2.70
		-23RS	2.13	59.1	0.00024	0.84	2.59	2.28
		MEU-IMS	2.66	5.47	0.00072	0.059	7.36	1.64 E+2
		-23RS	2.27	38.8	0.00110	2.19	6.32	1.40 E+2
Insolubles	Noble metals, carbon, few fissile particles	HEU-IMS	5.60	0.76	0.114	1.79	6.08	8.56 E+1
		-25RS	10.70	1.31	0.896	1.79	4.04	1.74 E+3
		-23RS	5.60	0.75	0.114	1.78	6.08	8.56 E+1
		MEU-IMS	22.25	1.70	0.585	1.80	6.73	2.76 E+3
		-23RS	3.24	1.08	0.046	1.79	5.03	2.30 E+3
Solvent extraction-Thorex 1A column aqueous wastes	Liquid, 4.3 kg/m^3	HEU-IMS	0.014	0.056	0.00176	0.378	1.73 E+1	1.01 E+1
		-25RS	0.014	0.026	0.00322	0.097	1.73 E+1	1.28 E+1
		-23RS	0.014	0.099	0.135	98.0	1.89 E+1	1.07 E+2
		MEU-IMS	0.013	0.0055	0.00362	0.059	3.72 E+1	8.25 E+2
		-23RS	0.012	0.099	0.0980	98.0	2.24 E+1	5.17 E+3
^{233}U Product	Liquid, 233 kg/m^3	HEU-IMS	100	54.7	[b]	[b]	4.6 E-3	3.05 E+1
		-25RS	100	25.6	[b]	[b]	4.6 E-3	3.05 E+1
		-23RS	100	97.4	[b]	[b]	4.6 E-3	3.05 E+1
		MEU-IMS	100	5.42	[b]	[b]	1.7 E-2	2.51 E+1
		-23RS	100	97.8	[b]	[b]	1.7 E-2	2.63 E+1
Thorium product	Liquid, 464 kg/m^3	HEU-IMS	0.00694	0.15	[b]	[b]	1.3 E-4	2.3 E-1
		-25RS	0.00694	0.068	[b]	[b]	1.3 E-4	2.3 E-1
		-23RS	0.0113	0.267	[b]	[b]	1.3 E-4	2.3 E-1
		MEU-IMS	0.0093	0.147	[b]	[b]	8.2 E-4	2.1 E-1
		-23RS	0.023	0.265	[b]	[b]	6.2 E-4	1.07
Solvent extraction-Purex 5A column aqueous wastes	Liquid, 12.5 kg/m^3	HEU-IMS	0.019	0.043	0.00078	0.098	1.80 E+1	1.70 E+3
5D column aqueous wastes	Liquid, 0.57 kg/m^3	HEU-IMS	6.28	0.214	48.96	91.5	1.13	8.44 E+3
5F column aqueous wastes	Liquid, 0.007 kg/m^3	HEU-IMS	7.57	0.013	16.3	1.51	3.61	2.93 E+3
^{235}U Product	Liquid, 470 kg/m^3	HEU-IMS	100	42.24	1.00	0.149	2.3 E-3	2.42

[a] Liquids are nitrate solutions. Concentrations are of material originating from fuel element.

[b] Less than 1×10^{-6} wt. %.

burning process to combine with weight data for total mass accountability. Gross gamma activity measurements may be used to monitor various areas; however, background activity in the process cells may make such measurements of little value. From time to time, material will be removed from the process in failed equipment. There is also a steady flow of process control samples to sample analysis areas. For proper administrative control, both the decontamination and maintenance areas and hot laboratories must be included in the head-end material balance area. Differing requirements for reprocessing and refabrication imply separate maintenance and laboratory facilities, so that separate material balance areas should pose no real problem.

The wet head-end consists of fertile and fissile dissolution, where the burned-back fuel kernels are dissolved and insoluble materials such as the SiC hulls are separated from the product nitrate solutions. At this point, the material is in a form where uranium isotopic content can

be measured. Several chemical methods exist; however, most are not well suited for a high throughout operation. In particular, plutonium and other fission products may interfere with uranium isotopic resolution, necessitating chemical separation before the uranium can be measured. Development work on ^{233}U measurements without chemical separation is necessary for high throughput.

The uranium content measured at the dissolving stage for an entire customer lot must be balanced against the fresh fuel uranium and thorium loadings and reactor burnup calculations for a special nuclear material balance. This balance must include the uranium content of any fissile particles and insoluble materials removed for waste disposal. Again, since the purpose of particle disposal is containment of special nuclear material and fission products in the repository, the development of non-destructive assay methods on irradiated particles is preferable to chemical dissolution and separation for assay.

The solutions from the dissolution of the fertile particles are combined with the solutions from the dissolution of the 23RS fissile particles and sent through the Thorex line, where the uranium and thorium are separated from the fission products and each other. The products of the dissolution of the fissile particles from the HEU IMS elements are sent through a standard Purex line.

When the measurement of ^{233}U in the presence of plutonium and fission products is solved, the material balance initial inventory for the solvent extraction area will be straightforward. A good isotopic inventory can be provided at solvent extraction product storage. This inventory then becomes the balance transfer to refabrication. Some minimal amount of nuclear material will be lost to solvent extraction waste streams, and the development of assay techniques will include uranium measurements at very low concentrations. Assay of plutonium in the waste streams should be possible with techniques being developed for LWR reprocessing.

DETAILED MATERIAL CONTROL AND ACCOUNTABILITY ASPECTS OF REFABRICATION

The general flowsheet for the operations involved in HTGR refabrication is shown in Fig. 6. The isotopic content and the activity associated with the uranium for each of the input streams in the two fuel cycles are presented in Table III. The two types of streams (^{235}U and ^{233}U) are never mixed. The products of refabrication are separate fuel elements containing either the ^{235}U stream or the ^{233}U stream. The high activities in the ^{233}U stream due to the buildup of decay products of the inherent ^{232}U content requires that all the steps in the refabrication of the ^{233}U stream must be done remotely. Because of crossover of some ^{233}U in reprocessing, the refabrication of the ^{235}U stream of the HEU cycle must also be done remotely. Refabrication has many more measurements and accountability areas than has Reprocessing. Though dictated mainly by process and product quality control concerns, the great number of measurements serves accountability purposes as well. The sampling techniques and philosophy have been discussed in detail elsewhere [13].

Uranium, as liquid nitrate, enters the refabrication line from Reprocessing and from Scrap Recovery. The first system is Uranium Feed, where the liquid is stored, isotopically blended, and chemically adjusted. The liquid goes to Resin Kernel Preparation, where the uranium is loaded onto resin microspheres. These then go to Resin

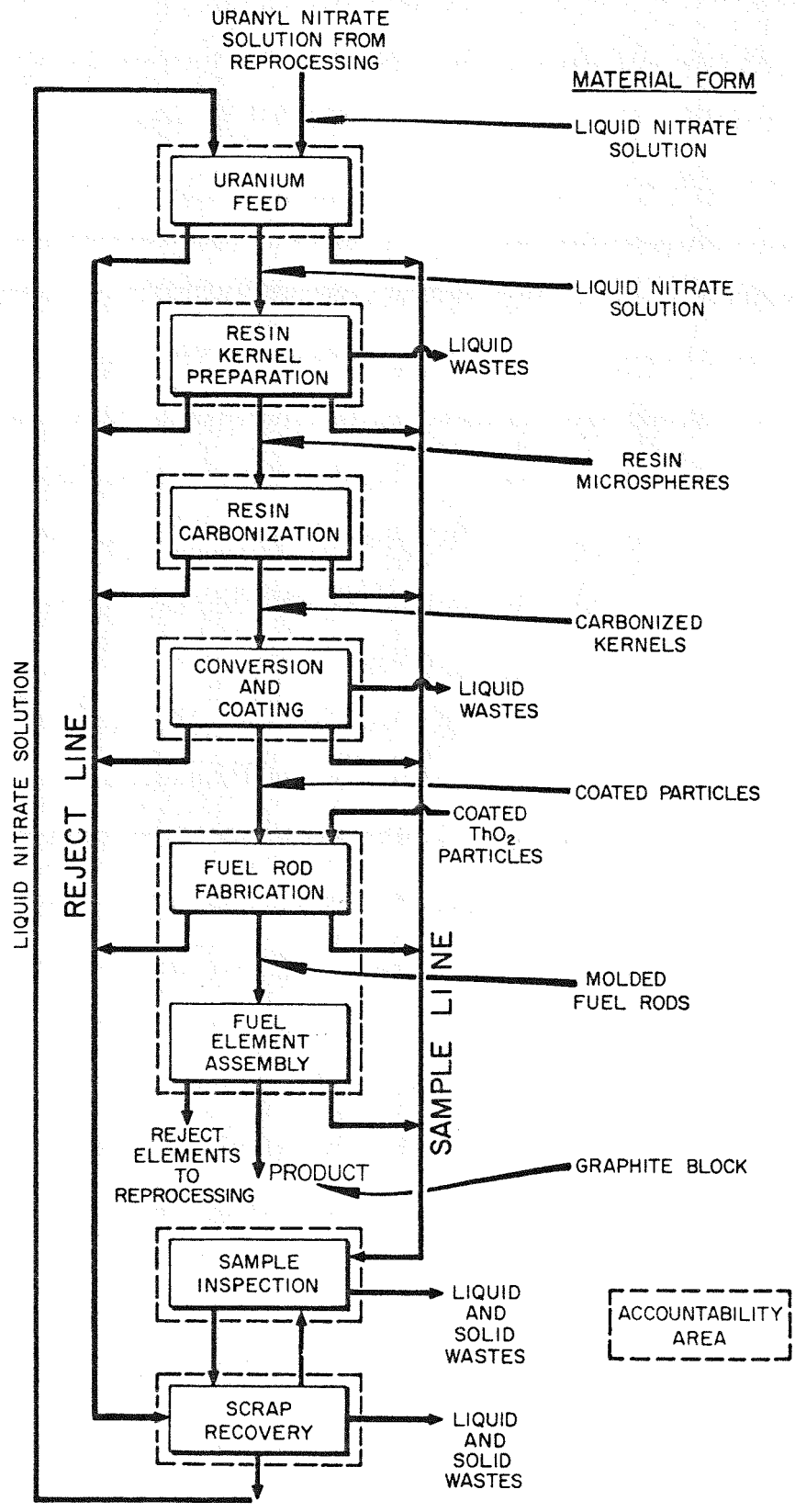


Fig. 6. HTGR Refabrication Flowsheet.

Table III. Characteristics of Input Streams
HTGR Refabrication

Fuel Element Type	Uranium Isotope Content, %						Specific Activity (mCi/g)	Specific Neutron Emission Rate (n/s·g)
	232	233	234	235	236	238		
HEU-235	0.002	2.93	1.62	29.96	48.78	16.72	2.3	2.621
HEU-233	0.045	66.73	21.30	7.42	4.48	0.025	19.2[a]	30.97[a]
MEU-233	0.043	67.55	21.33	6.58	4.01	0.48	18.5	29.81

[a]Assumes 30 d since solvent extraction cleanup.

Carbonization, where the resin is decomposed (carbonized) to produce a kernel consisting of uranium dioxide in a carbon matrix. The kernel goes to Conversion and Coating, where, in conversion, the UO_2 is caused to react with the carbon matrix to produce a mixture of the UO_2 and UC_2 and where, in coating, three layers of pyrocarbons and one SiC coating are applied to produce the coated fissile particle. These coated fissile particles then go to Fuel Rod Fabrication, where they are blended to homogenize any slight differences in coating batches and mixed with carbon-coated ThO_2 particles from a fresh-fuel plant. The mixture is molded to form the "green" fuel rod held together by a pitch-base binder. The green fuel rods go to Fuel Element Fabrication, where they are inserted into the graphite fuel blocks. The assembled fuel elements are heated to carbonize the pitch binder of the rods. Finally, the fuel elements are cleaned, inspected, and sent to Shipping, Receiving, and Storage. Scrap fuel elements are temporarily stored, then campaigned to Reprocessing for recovery.

Sample Inspection and Scrap Recovery are major systems in Refabrication. They receive streams from all the systems mentioned previously and each other. The material exiting Sample Inspection is routed to Scrap Recovery or to Waste Treatment. The material entering Scrap Recovery exits principally as recovered uranyl nitrate solution returned to Uranium Feed or as various forms sent to Waste Treatment.

Uranium Feed is the first accountability area in Refabrication. Uranyl nitrate solutions are received from Reprocessing or Scrap Recovery. Once in Refabrication there is no transfer of material back to Reprocessing with the exception of the whole reject blocks. Liquid samples are transferred to Sample Inspection to assess the impurity levels of the feed and to verify the results of the isotopic blending and chemical adjustments. Unacceptable liquid feed is transferred to Scrap Recovery and the product is delivered to Resin Kernel Preparation. Accountability for these input and output streams is by volume measurement and uranium assays of the samples.

Resin Kernel Preparation, Resin Carbonization, and Conversion and Coating are also separate accountability areas. Accountability for the liquid stream received by the Resin Kernel Preparation is by volume measurement and uranium determination of a liquid sample. Accountability for the product of Resin Kernel Preparation and the inputs and outputs of Resin Carbonization and Conversion and Coating are by means

of automatic remote weighing devices and destructive and nondestructive analyses of samples. Special passive samplers suitable for remote handling have been developed [14]. Solid particles are conveyed pneumatically between and within systems. Besides the main product streams, solids or liquids are transferred to Scrap Recovery or Waste Treatment.

Fuel Rod Fabrication and Fuel Element Assembly together constitute another accountability area. The basic accountability approach is to nondestructively assay 100% of the acceptable fuel rods produced in Fuel Rod Fabrication. Accountability in Fuel Element Assembly depends on knowledge of the location and weight of assayed fuel rods and upon the weight of a fuel block before and after loading.

The as-coated fissile particles are pneumatically transferred from Conversion and Coating to a precision weigher and then to a batch blender, which blends up to 24 coating batches, each containing about 3 kg U. The blended particles are passed through a sampler, and the sample is nondestructively and destructively chemically analyzed in Sample Inspection to determine the uranium assay, isotopic contents, and the mass of the particles. The fertile particles are blended, sampled, and analyzed similarly to the fissile particles before being transferred into the hot cells. The mass of the incoming fertile material is determined. The particles are then molded into fuel rods. At this point in the process, the scrap is in the form of particles, fuel rods, and pieces of rods. The scrap is assayed by nondestructive methods or by total mass measurements in those cases where accurate uranium weight factors are known, before being transferred to Scrap Recovery.

The accepted rods then undergo fuel homogeneity inspections. All the rods are analyzed in a gamma scan system. The overall mass distribution is determined and the fissile and fertile content verified semiquantitatively. A second system samples approximately 10% of the fuel rods and determines the total heavy metal, thorium, and uranium distributions in the rods. Such a system, which operates on the principle of multi-energy radiation attenuation with selective K-edge absorption, has been developed at ORNL in the HTGR program [15]. It uses the radioisotopes ^{169}Yb and ^{177m}Lu, which emit gamma rays of energies between the respective thorium and uranium K-absorption edges. Such gamma rays permit separation of the thorium and uranium contribution because the attenuation coefficients of the thorium and uranium are very different in that energy range.

The next inspection is fuel rod assay. Two nondestructive assay devices are used for this purpose. One is an on-line device capable of assaying 100% of the fuel rods produced from two machines. The identity, location, and disposition of rods are monitored by the computer in subsequent storage and fuel element loading. A second device accepts a sample from the main product line and nondestructively assays the fuel rod in a laboratory. In addition, a limited number of rods is also chemically assayed. The two nondestructive assay devices are used for product verification and for determining the ^{233}U, ^{235}U fissile contents of the fuel rods. Both devices use active neutron interrogation with a ^{252}Cf neutron source in each irradiator assembly. The on-line assay device detects the prompt fission neutrons from the irradiated sample. A device of this type has also been developed at ORNL in the HTGR program [16].

Sample Inspection is a separate accountability area. This system comprises the analytical laboratories and equipment necessary to perform analyses on samples transferred from the other systems of Refabrication.

Samples are analyzed to characterize the main batches of material present in the other systems. The mass of each sample entering and leaving the system is determined. Both chemical and nondestructive methods are used for the analyses. Uranium, thorium, and the isotopic contents of samples are determined by potentiometric, volumetric, and other techniques. The nondestructive analyses are performed by gamma- and alpha-ray counting and by neutron interrogation. A nondestructive device that assays particles and fuel rod samples has been developed and is being tested at ORNL [17]. The device uses a ^{252}Cf neutron source in the irradiator assembly and detects the delayed fission neutrons emitted from the irradiated sample. The device yields accurate assay information and complements other assay devices and methods.

Scrap Recovery receives material in a variety of forms from all the other systems. This system is in effect a mini-reprocessing operation largely provided to avoid the inherent accountability problems associated with transfers of material back to Reprocessing. Incoming accountability is via techniques appropriate to the material form. The major exiting stream is the recovered uranium nitrate product, which is directed back to the front end of the refabrication line. Accountability for this stream is by volume measurement and sample analyses including isotopic analyses. The other exiting streams are various aqueous wastes for which the accountability is by volume measurement and sample analyses. A final stream consists of insolubles, mainly coated particles. Accountability for this stream is by means of weighing and sample analyses.

SUMMARY

The materials control and accountability aspects of the Reprocessing and Refabrication of a conceptual large-scale HTGR fuel recycle plant have been discussed. Two fuel cycles were considered. The traditional highly enriched uranium cycle uses an initial or makeup fuel element with a fissile enrichment of 93% ^{235}U. The more recent medium enriched uranium cycle uses initial or makeup fuel elements with a fissile enrichment less than 20% ^{235}U. In both cases, ^{233}U bred from the fertile thorium is recycled.

Materials control and accountability in the plant will be by means of a real-time accountability method. Accountability data will be derived from monitoring of total material mass through the processes and a system of numerous assays, both destructive and nondestructive.

REFERENCES

[1] LOTTS, A. L., et al., "HTGR Fuel and Fuel Cycle Technology," Nuclear Power and its Fuel Cycle (Proc. Int. Conf. Salzburg, 1977) 3, IAEA, Vienna (1977) 433–52.

[2] Project Staff, *Conceptual Design Summary and Design Qualifications for HTGR Target Recycle Plant,* General Atomic Company Rep. GA-A13365 (April 1975).

[3] Project Staff, *Preconceptual Design and Estimate Summary for HTGR Recycle Demonstration Facility (HRDF),* General Atomic Company Rep. GA-A13502 (July 1975).

[4] LOTTS, A. L., COOBS, J. H., *HTGR Fuel and Fuel Cycle Technology,* ORNL/TM-5501 (August 1976).

[5] NOTZ, K. J., *An Overview of HTGR Fuel Recycle*, Oak Ridge National Laboratory Rep. ORNL/TM-4747 (January 1976).
[6] AUGUSTSON, R. H., "Development of In-Plant Real-Time Materials Control: The DYMAC Program," *Nucl. Mater. Manage.* 5 3 (1976) 302–16.
[7] KEEPIN, G.R., MARAMAN, W. J., "Nondestructive Assay Technology and In-Plant Dynamic Materials Control — DYMAC," Symposium on Safeguarding Nuclear Materials (Proc. Int. Symp. Vienna, 1975 IAEA, Vienna (1976).
[8] McNEANY, S. R., Oak Ridge National Laboratory, unpublished computer program, September 1974.
[9] McNEANY, S. R., Oak Ridge National Laboratory, unpublished computer program, September 1976.
[10] JENKINS, J. D., McNEANY, S. R., RUSHTON, J. E., *Conceptual Design of the Special Nuclear Material Nondestructive Assay and Accountability System for the HTGR Fuel Refabrication Pilot Plant*, Oak Ridge National Laboratory Rep. ORNL/TM-4917 (July 1975).
[11] BELL, M. J., *ORIGEN — The ORNL Isotope Generation and Depletion Code*, Oak Ridge National Laboratory Rep. ORNL-4628 (May 1973).
[12] CROFF, A. G., Oak Ridge National Laboratory, personal communication, October 1977.
[13] PECHIN, W. H., et al., *Inspection of High-Temperature Gas-Cooled Reactor Recycle Fuel*, Oak Ridge National Laboratory Rep. ORNL-5165 (June 1977); also "Quality Control Tests for High-Temperature Gas-Cooled Reactor Recycle Fuel," *Nuclear Fuel Quality Assurance*, (Proc. Sym. Vienna), IAEA, Vienna (1976), 425–27.
[14] SUCHOMEL, R. R., LACKEY, W. J., *Device for Sampling HTGR Recycle Fuel Particles*, Oak Ridge National Laboratory Rep. ORNL/TM-5739 (March 1977).
[15] ANGELINI, P., et al., Oak Ridge National Laboratory Rep. ORNL-5266 (1977) 181–84.
[16] RUSHTON, J. E., Oak Ridge National Laboratory Rep. ORNL-5266 (1977) 186–89.
[17] KNOLL, R. W., Oak Ridge National Laboratory Rep. ORNL-5266 (1977) 189–90.