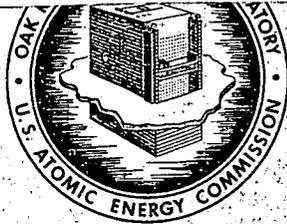




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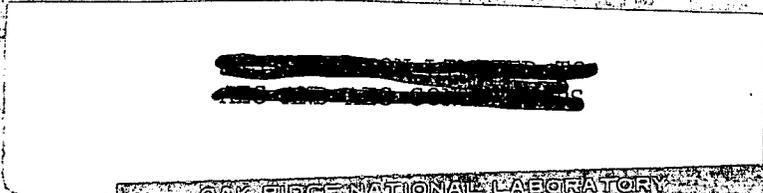


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STUDY OF THE WASTE HANDLING REQUIREMENTS FOR THE HTGR FUEL RECYCLE DEVELOPMENT PROGRAM

R. S. Lowrie



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CHEMICAL TECHNOLOGY DIVISION

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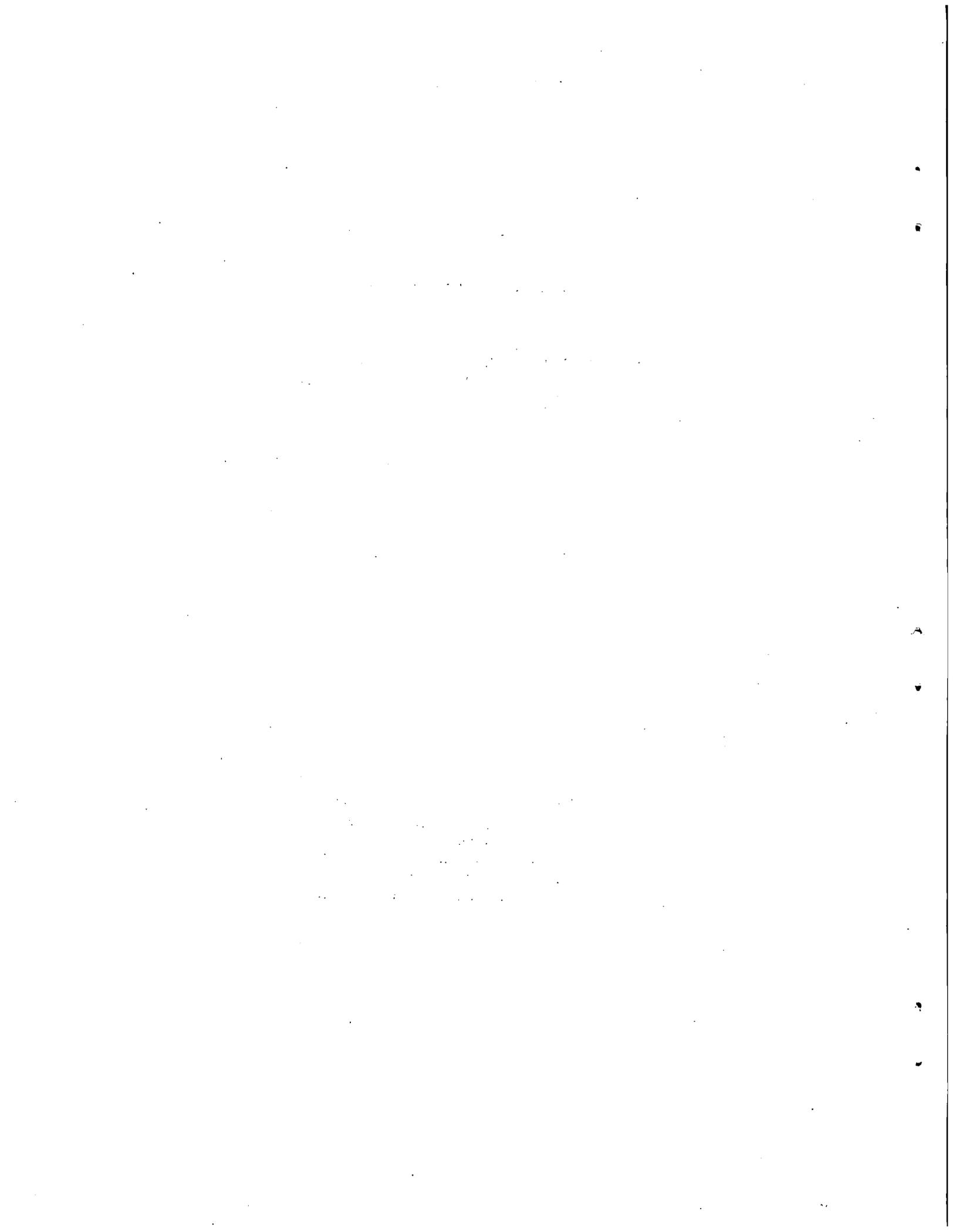
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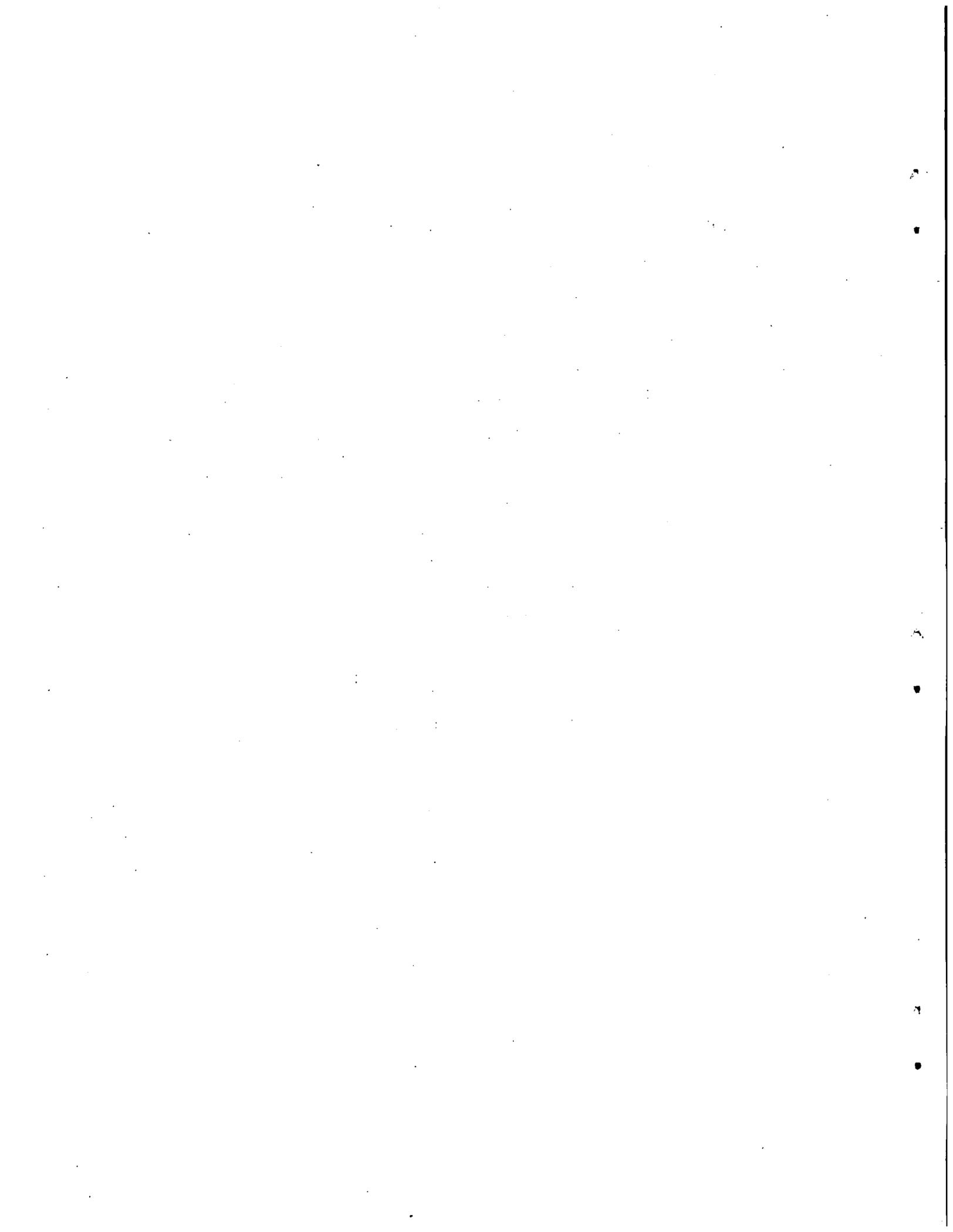


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STUDY OF THE WASTE HANDLING REQUIREMENTS FOR THE
HTGR FUEL RECYCLE DEVELOPMENT PROGRAM

R. S. Lowrie

ABSTRACT

Studies have been performed to define the problems involved in handling and disposing of the waste streams generated during the radioactive demonstration of the HTGR fuel reprocessing and refabrication facilities to be conducted at Oak Ridge National Laboratory under the National HTGR Recycle Development Program. During these studies, the waste streams generated by the Head-End, Acid-Thorex, and Refabrication Pilot Plants were identified, the processes and facilities necessary to prepare these wastes for disposal were determined, and an order-of-magnitude estimate of the cost of building these facilities was made. Where applicable, alternative processes are discussed. The estimated capital costs of the major waste disposal systems for the Head-End Pilot Plant are \$650,000 for the partial-block burning concept and \$848,750 for the whole-block burning concept. Waste facilities for the Acid-Thorex Pilot Plant would cost \$257,500; those required for the Refabrication Pilot Plant would cost \$40,500.

1. INTRODUCTION

A recycle development program for reprocessing and refabricating high-temperature gas-cooled reactor (HTGR) fuels is being conducted at the Oak Ridge National Laboratory as part of the Thorium Utilization Program. The planned program includes the design, construction, and operation of three developmental pilot plants to demonstrate the reprocessing and refabrication operations. To demonstrate the performance and determine maintenance characteristics of the process equipment, it is planned that approximately 900 Ft. St. Vrain Reactor (FSVR) fuel elements will be reprocessed during a six-month period of operation. Enough ²³³U would be recovered to meet the requirements for fabricating approximately 150 recycle elements to FSVR element specifications. The

successful implementation of this plan should provide much of the basic technology required for the design, construction, and operation of a commercial plant for recycling HTGR fuel.

The three recycle pilot plants described in the National HTGR Fuel Recycle Development Program Plan¹ are the Head-End Pilot Plant, the Acid-Thorex Pilot Plant, and the Refabrication Pilot Plant. The Head-End and Refabrication Pilot Plants will be located in Bldg. 7930, The Thorium-Uranium Recycle Facility. The Acid-Thorex Pilot Plant will be located in Bldg. 3019, The Radiochemical Processing Pilot Plant. Each of these will generate liquid, gaseous, and solid wastes that will have to be prepared for safe discharge to the environment or will have to be delivered in a suitable form to ORNL waste disposal facilities. The studies described in this report were performed to identify the streams generated by the pilot plants during the planned demonstration program, to determine the processes and facilities necessary to prepare these wastes for disposal, and to make an order-of-magnitude estimate of the cost of building these facilities. Where applicable, alternative processes are discussed.

It is difficult to predict what the federal, state, and Laboratory waste disposal regulations will be in 1976. It is expected that allowable radioactivity release rates to the environment will be lower than those at present. Therefore, these waste studies were made using the following ground rules:

1. Gaseous wastes produced in each pilot plant will be discharged to the atmosphere after suitable treatment to remove radioactive materials (e.g. ⁸⁵Kr and ³H) to as low a level as is practical. A decontamination factor (DF) of 10^2 or greater will be required.
2. Liquid waste will be pumped to hold tanks, where it will be monitored and prepared for discharge via pipeline to the ORNL Liquid Waste System.
3. Solid wastes will be segregated according to type and activity, packaged in suitable containers, and sent to the ORNL Solid Waste Disposal System.

4. Cost estimates will include only the special equipment needed by the pilot plants to prepare the waste for discharge to the ORNL waste systems or stack.

2. IDENTIFICATION OF WASTE STREAMS

Detailed chemical process flowsheets have been prepared showing the compositions and flow rates of the process streams for the Head-End and Acid-Thorex Pilot Plants and the sol preparation and microsphere-forming steps of the Refabrication Pilot Plant.² Estimates were made of the composition and flow rates of the entering and exiting streams in the particle coating, fuel stick preparation, and fuel element assembly steps. Calculations for the flowsheets were based on the following assumptions:

1. The daily production capacity will be 12 kg of $(\text{Th}-^{233}\text{U})\text{O}_2$ sol-gel microspheres with a Th/U ratio of 4.25.
2. All of the ^{233}U needed for fabrication will be recovered from irradiated FSVR fuel elements.
3. Approximately 150 recycle fuel elements will be produced.
4. Particles containing ^{235}U and particles containing ThO_2 (or ThC_2) will be prepared and coated separately from this program and will be purchased or otherwise provided for use in recycle element fabrication.

The uranium, thorium, and fission product concentrations and isotopic compositions calculated using the ORIGEN code³ for fertile (see Tables 1 and 2) and fissile particles in the fuel discharged from the FSVR after 2 years of equivalent full-power operation and cooled for 150 days were used to represent all fuel reprocessed. These calculations indicate that reprocessing approximately ten fuel elements per day will provide sufficient ^{233}U , when combined with the internal ^{233}U recycle stream, to produce 12 kg of microspheres per day.

Table 1. Heavy-Metal Nuclide Composition of Fertile Particles in Spent FSVR Fuel

H T G R FORT ST VRAIN REACTOR -- FERTILE PARTICLE (2 YR EXPOSURE)

POWER= 9.24 MW/MT, BURNUP= 8437. MWD/MT, FLUX= 4.84E 13 N/CM**2-SEC

NUCLIDE CONCENTRATIONS, GRAMS / METRIC TON FUEL CHARGED TO REACTOR

	CHARGE	DISCHARGE	30.0 D	90.0 D	150. D	365. D	3652. D
HE 4	0.0	1.68E-03	1.80E-03	2.05E-03	2.31E-03	3.32E-03	2.35E-02
PB208	0.0	2.20E-03	2.48E-03	3.12E-03	3.85E-03	7.17E-03	1.08E-01
TH228	0.0	9.86E-03	1.08E-02	1.26E-02	1.42E-02	1.94E-02	3.79E-02
TH229	0.0	6.66E-02	7.03E-02	7.81E-02	8.59E-02	1.14E-01	5.45E-01
TH230	0.0	1.85E-01	1.85E-01	1.86E-01	1.87E-01	1.89E-01	2.19E-01
TH232	7.70E 05	7.50E 05	7.50E 05	7.50E 05	7.50E 05	7.50E 05	7.50E 05
PA231	0.0	5.42E 00	5.44E 00	5.44E 00	5.44E 00	5.44E 00	5.43E 00
PA233	0.0	9.33E 02	4.37E 02	9.58E 01	2.10E 01	1.82E-01	1.36E-08
U232	0.0	1.59E 00	1.59E 00	1.59E 00	1.59E 00	1.58E 00	1.45E 00
U233	0.0	1.05E 04	1.09E 04	1.13E 04	1.14E 04	1.14E 04	1.14E 04
U234	0.0	1.24E 03	1.24E 03	1.24E 03	1.24E 03	1.24E 03	1.24E 03
U235	0.0	1.64E 02	1.64E 02	1.64E 02	1.64E 02	1.64E 02	1.64E 02
U236	0.0	1.22E 01	1.22E 01	1.22E 01	1.22E 01	1.22E 01	1.22E 01
NP237	0.0	3.73E-01	3.94E-01	3.95E-01	3.95E-01	3.95E-01	3.95E-01
PU238	0.0	5.40E-02	5.52E-02	5.51E-02	5.50E-02	5.48E-02	5.11E-02
PU239	0.0	7.77E-03	7.77E-03	7.77E-03	7.77E-03	7.77E-03	7.76E-03
PU240	0.0	2.18E-03	2.18E-03	2.18E-03	2.18E-03	2.18E-03	2.18E-03
SUBTOT	7.70E 05	7.63E 05	7.63E 05	7.63E 05	7.63E 05	7.63E 05	7.63E 05
TOTALS	7.70E 05	7.63E 05	7.63E 05	7.63E 05	7.63E 05	7.63E 05	7.63E 05

Table 2. Fission Product Element Composition of Fertile Particles in Spent FSVK Fuel

H T G R FORT ST VRAIN REACTOR -- FERTILE PARTICLE (2 YR EXPOSURE)

POWER= 9.24 MW/MT, BURNUP= 8437. MWD/MT, FLUX= 4.84E 13 N/CM**2-SEC

ELEMENT CONCENTRATIONS, GRAMS / METRIC TON FUEL CHARGED TO REACTOR

	CHARGE	DISCHARGE	30.0 D	90.0 D	150. D	365. D	3652. D
H	0.0	1.17E-02	1.16E-02	1.15E-02	1.14E-02	1.10E-02	6.65E-03
ZN	0.0	2.88E-05	2.52E-09	4.79E-18	0.0	0.0	0.0
GA	0.0	1.88E-05	5.48E-10	1.04E-18	0.0	0.0	0.0
GE	0.0	1.93E-01	1.92E-01	1.92E-01	1.92E-01	1.92E-01	1.92E-01
AS	0.0	8.63E-02	8.33E-02	8.33E-02	8.33E-02	8.33E-02	8.33E-02
SE	0.0	3.24E 01	3.24E 01	3.24E 01	3.24E 01	3.24E 01	3.24E 01
BR	0.0	1.26E 01	1.26E 01	1.26E 01	1.26E 01	1.26E 01	1.26E 01
KR	0.0	2.23E 02	2.22E 02	2.22E 02	2.22E 02	2.21E 02	2.13E 02
RB	0.0	2.07E 02	2.07E 02	2.08E 02	2.08E 02	2.09E 02	2.17E 02
SP	0.0	3.96E 02	3.86E 02	3.75E 02	3.70E 02	3.63E 02	3.26E 02
Y	0.0	2.18E 02	2.17E 02	2.17E 02	2.16E 02	2.14E 02	2.14E 02
ZR	0.0	1.15E 03	1.15E 03	1.15E 03	1.15E 03	1.15E 03	1.19E 03
NB	0.0	2.10E 01	1.96E 01	1.34E 01	7.97E 00	1.28E 00	1.21E-03
MO	0.0	8.01E 02	8.13E 02	8.32E 02	8.45E 02	8.61E 02	8.62E 02
TC	0.0	1.71E 02	1.72E 02	1.72E 02	1.72E 02	1.72E 02	1.72E 02
RU	0.0	2.48E 02	2.45E 02	2.41E 02	2.40E 02	2.38E 02	2.35E 02
RH	0.0	6.73E-02	4.58E-03	1.60E-03	5.64E-04	1.56E-05	5.36E-09
PD	0.0	3.29E 01	3.33E 01	3.38E 01	3.43E 01	3.57E 01	3.85E 01
AG	0.0	1.38E 00	1.36E 00	1.36E 00	1.36E 00	1.36E 00	1.36E 00
CD	0.0	4.65E 00	4.67E 00	4.67E 00	4.67E 00	4.67E 00	4.67E 00
IN	0.0	4.26E-01	4.33E-01	4.36E-01	4.37E-01	4.38E-01	4.38E-01
SN	0.0	1.30E 01	1.29E 01	1.29E 01	1.28E 01	1.28E 01	1.28E 01
SB	0.0	6.69E 00	6.21E 00	6.06E 00	5.94E 00	5.54E 00	3.25E 00
TE	0.0	1.94E 02	1.91E 02	1.89E 02	1.88E 02	1.88E 02	1.90E 02
I	0.0	1.21E 02	1.19E 02	1.20E 02	1.21E 02	1.22E 02	1.22E 02
XE	0.0	1.26E 03	1.26E 03	1.26E 03	1.26E 03	1.26E 03	1.26E 03
CS	0.0	6.82E 02	6.86E 02	6.84E 02	6.81E 02	6.73E 02	5.96E 02
BA	0.0	3.54E 02	3.47E 02	3.48E 02	3.50E 02	3.59E 02	4.36E 02
LA	0.0	3.32E 02	3.31E 02	3.31E 02	3.31E 02	3.31E 02	3.31E 02
CE	0.0	8.66E 02	8.51E 02	8.24E 02	8.07E 02	7.68E 02	7.15E 02
PR	0.0	3.69E 02	3.75E 02	3.85E 02	3.89E 02	3.90E 02	3.90E 02
ND	0.0	8.79E 02	8.96E 02	9.15E 02	9.29E 02	9.66E 02	1.01E 03
PM	0.0	4.82E 01	4.89E 01	4.72E 01	4.52E 01	3.87E 01	3.58E 00
SM	0.0	1.29E 02	1.31E 02	1.33E 02	1.35E 02	1.42E 02	1.76E 02
EU	0.0	2.16E 01	2.14E 01	2.13E 01	2.12E 01	2.10E 01	1.95E 01
GD	0.0	6.73E 00	7.09E 00	7.26E 00	7.32E 00	7.53E 00	9.27E 00
TB	0.0	4.56E-02	4.50E-02	4.42E-02	4.37E-02	4.32E-02	4.31E-02
DY	0.0	1.16E-02	1.24E-02	1.33E-02	1.38E-02	1.46E-02	1.51E-02
HO	0.0	2.82E-04	2.82E-04	2.82E-04	2.82E-04	2.82E-04	2.82E-04
ER	0.0	3.78E-05	3.81E-05	3.81E-05	3.81E-05	3.81E-05	3.81E-05
TOTALS	0.0	8.81E 03	8.81E 03	8.80E 03	8.80E 03	8.81E 03	8.80E 03

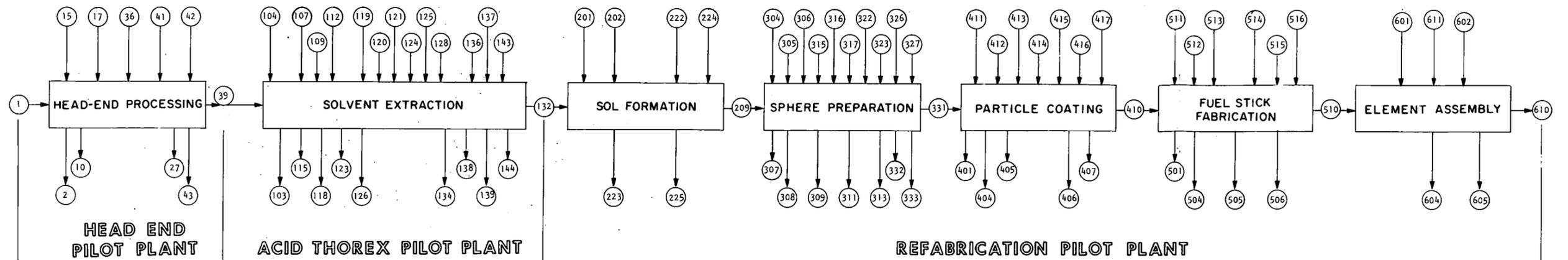
The relationship between most of the entering streams and the waste streams leaving the various processing steps is shown in Fig. 1. Streams numbered 1-99 originate in the Head-End Pilot Plant; streams numbered 100-199 originate in the Acid-Thorex Pilot Plant; and those numbered 200, 300, 400, 500, and 600 originate in one of the major processing steps in the Refabrication Pilot Plant. The waste streams are described in Tables 3 and 4.

A brief description of the process flowsheet follows. FSVR fuel consists of large hexagonal graphite blocks (14.172 in. across the flats, 31.22 in. long) containing coolant passages and fuel holes. Silicon carbide - coated fertile ThC_2 and fissile $(\text{Th-U})\text{C}_2$ particles are bonded into fuel sticks and inserted into the fuel holes. Fuel elements will be discharged from the reactor once a year and, after a suitable cooling period (which will be 150 days or longer in all cases), will be shipped to the TURF storage facility⁴ where they will be accumulated until used in the recycle demonstration.

2.1 The Head-End Pilot Plant

Fuel elements removed from the storage facility may be crushed and then screen-tumbled to remove as much "barren" graphite as possible. Such barren graphite will be sent to the ORNL waste system. The material containing the fissile and fertile particles will be burned to remove the graphite from the block, plus the outer graphite particle coating, in a fluidized-bed burner that may use Al_2O_3 as the heat transfer medium. Ash from the burner will be classified into four fractions: fissile particles, which are sent to storage; recycle Al_2O_3 ; fines; and fertile particles. The fertile-particle fraction will be crushed and then burned to remove the inner graphite coating; subsequently the ash combined with the fine fraction from the classifier will be sent to the Acid-Thorex Pilot Plant. Several alternatives to this flowsheet are being considered:

- (1) Burning the uncrushed block in a whole-block burner, thus eliminating the burner feed preparation steps and the Al_2O_3 .
- (2) Burning the crushed block in a static-bed chunk burner, thus eliminating the need for the Al_2O_3 heat transfer medium.



1 Canned Fuel Element (2 yr exposure)

39 Canned Fertile-Particle Burner Ash and Al₂O₃ Fines

132 Concentrate UO₂(NO₃)₂ Solution

209 Concentrated Sol Product

331 Specification Spheres

410 Coated Microspheres

510 Specification Fuel Sticks

610 Recycle Fuel Element to FSVR

(INPUTS)

- 15 Primary Burner Reagent Gas
- 17 Make-Up Al₂O₃
- 36 Fertile Particle Burner Reagent Gas
- 41 Cans for Fissile Particle Storage
- 42 Cans for Burner Ash and Al₂O₃ Fines Shipment

(INPUTS)

- 104 Acid to Leacher
- 107 Recycled U + Th As Oxides
- 109 Wash Water to Filter
- 112 Steam to Feed Adjustment Step
- 119 Scrub I-AS
- 120 Nitric Acid I-AX-HNO₃
- 121 Solvent I-AX 30% TBF in N-Dodecane
- 124 HNO₃ I-BX
- 125 Solvent Scrub I-B5
- 128 Uranium Strip I-CX
- 136 Na₂CO₃ Solvent Wash
- 137 HNO₃ Solvent Wash
- 143 Can for Dried Filter Cake

(INPUTS)

- 201 Th(NO₃)₂·4H₂O
- 202 Water
- 204 Water
- 222 Solvent Regeneration Solution
- 224 Solvent Wash Water

(INPUTS)

- 304 Ethyl Alcohol to Ion Exchange Column
- 305 NH₄OH Solution to Ion Exchange Column
- 306 Waste Water to Ion Exchange Column
- 315 Make-up 2-Ethyl Hexanol
- 316 Span - 80
- 317 Ethomeen S/15
- 322 Argon to Dryer
- 323 Steam to Dryer
- 326 Hydrogen to Furnace
- 327 Argon to Furnace

(INPUTS)

- 411 Argon to Purge System
- 412 Argon to Gas Distribution
- 413 Caustic
- 414 Hydrogen
- 415 Hydrocarbon Gases (C₂H₂ & C₃H₆)
- 416 Silane
- 417 Graphite Cone

(INPUTS)

- 511 Binder Materials
- 512 Fertile Particles
- 513 Argon for Carbonization Furnace
- 514 Argon for Heat Treatment Furnace
- 515 Water
- 516 Solvents

(INPUTS)

- 601 Fuel Blocks
- 602 Fuel Hole Caps
- 611 Cement for Caps

(DISCHARGES)

- 2 Empty Fuel Can
- 10 Canned Barren Graphite
- 27 Canned Fissile Particles
- 43 Combined Burner Off-Gas to Decontamination

(DISCHARGES)

- 103 Empty Cans
- 115 Condensate from Feed Adjustment
- 118 Off-Gas from Leacher
- 123 Raffinate I-AW
- 126 Thorium Product I-BT
- 134 Condensate from UO₂(NO₃)₂ Evaporator
- 138 HNO₃ to Waste
- 139 Na₂CO₃ to Waste } from Solvent Cleanup
- 144 Canned Filter Cake to Waste Storage

(DISCHARGES)

- 223 Spent Solvent Regeneration Solution
- 225 Waste Solvent Wash Water

(DISCHARGES)

- 307 Ethyl Alcohol from Ion Exchange Column
- 308 NH₄OH Waste from Ion Exchange Column
- 309 Wash Water to Waste from Ion Exchange Column
- 311 Organic Waste from Solvent Cleanup
- 313 Aqueous Waste from Solvent Cleanup
- 332 Condensate
- 333 Combined Dryer-Furnace Off-Gas

(DISCHARGES)

- 401 Particle Coater Off-Gas
- 404 Caustic Solution from Particle Coater Off-Gas Scrubber
- 405 Graphite Cones
- 406 Compressed Soot
- 407 Carbon Chips

(DISCHARGES)

- 501 Off-Gas from Carbonization Furnace
- 504 Waste Solvents (organic)
- 505 Waste Binder Materials
- 506 Waste Alumina

(DISCHARGES)

- 604 Reject Graphite Blocks (unfueled)
- 605 Carbon Scrap

Fig. 1. HTGR Fuel Recycle Summary Process Flowsheet.

Table 3. Significant Head-End and Acid-Thorex Waste Streams

Stream No.	Description	Volume	Radioactivity Level ^a	Thermal Power (kW)
27	Fissile particles	1.72 ft ³ /day	391,000 Ci of mixed FPS	1.7
43	Burner off-gas ^b	32.2 scfm	1.0 Ci of ¹³¹ I ₂ per day 16 Ci of ³ H ₂ O per day 1020 Ci of ⁸⁵ Kr per day 1 Ci of ¹⁴ C per day	
123	I-AW-raffinate	173 gal/day	211,000 Ci of mixed FPS	0.80
126	I-BT-thorium solution	455 gal/day	210 Ci of mixed FPS	
144	Filter cake	20 ft ³ /day	20,000 Ci of mixed FPS	0.08

^aBased on fuel exposed 2 years in the reactor and cooled 150 days.
FPS = fission products.

^bWill contain off-gas from primary and fertile particle burners, plus a small flow of inert blanket gas from crushing operations.

Table 4. Low-Level Alpha-Contaminated HTGR Waste Streams

Stream No.	Description	Volume Produced per Unit Time
2	Empty fuel element cans	1.6 per day
10	Canned "barren" graphite	624 kg/day (three 55-gal drums)
103	Empty fertile-particle ash shipment cans	3.7 per day
115	Condensate from feed adjustment	272 gal/day
118	Leacher off-gas	1-5 scfm
134	Condensate from $\text{UO}_2(\text{NO}_3)_2$ evaporation	282 gal/day
138	Na_2CO_3 from solvent cleanup	75 gal/day
139	HNO_3 from solvent cleanup	75 gal/day
223	Na_2CO_3 from sol formation solvent regeneration step	28 gal/day
225	Solvent wash water	8 gal/day
307	Ethyl alcohol wash from ion exchange column	11 gal/day
308	NH_4OH wash from ion exchange column	11 gal/day
309	Water wash from ion exchange column	11 gal/day
311	Organic liquid waste from solvent cleanup	19.5 gal/day
313	Aqueous waste from solvent cleanup	17.5 gal/day
332	Condensate from sphere drying and firing furnaces	7.0 gal/day
333	Combined dryer and furnace off-gas	1.5 scfm
401	Particle coater off-gas	4.0 scfm (avg.) 60.0 scfm (max.)
404	Caustic solution from particle coater off-gas scrubber	130 gal/day
405	Graphite cones	6.2 kg/day (3-1/3 cones/day)
406	Compressed soot	10 kg/day
407	Graphite chips	1 kg/day
501	Off-gas from carbonization furnace	255 scfd
504	Waste solvents (organic)	5.0 gal/day
505	Waste binder material	15 kg/day
506	Waste alumina	100 kg/day
604	Reject nonfueled graphite fuel blocks	1 per month
605	Carbon scrap	1 kg/day

- (3) Eliminating the solid barren graphite waste stream by burning it in either a secondary burner or a larger primary burner.

An important consideration in connection with the third alternative is the degree of contamination of the barren graphite. The off-gas from its burning may well require decontamination; in such cases, the screen-tumbling step would provide little advantage.

2.2 The Acid-Thorex Pilot Plant

Solids from the Head-End Pilot Plant will be sent to the Acid-Thorex Pilot Plant, where they will be leached with Acid-Thorex Reagent (13 M HNO_3 --0.05 M F^-). The resulting slurry will be clarified, and the composition of the aqueous solution will be adjusted to serve as feed to the solvent extraction step. The solid residue from the clarification step will be dried, canned, and sent to the ORNL Solid Waste Disposal System. The thorium and uranium will be extracted from the fission products in the first column, the thorium will be partitioned from the uranium in the second column, and the uranium will be recovered in the third column. The resulting product, $\text{UO}_2(\text{NO}_3)_2$ solution, will be concentrated and sent to the Refabrication Pilot Plant. The I-AW containing fission products and I-BT containing thorium will be discharged to the ORNL Liquid Waste Disposal System. If desired, the solvent extraction flowsheet can be modified to eliminate the thorium partitioning step, thereby producing a combined thorium-fission product aqueous waste stream.

2.3 The Refabrication Pilot Plant

The concentrated $\text{UO}_2(\text{NO}_3)_2$ solution from the Acid-Thorex Pilot Plant will be converted into sol in the sol formation step; then the sol will be formed into microspheres, which are successively dried, fired, and classified. Approximately 1 kg (as heavy metals) per day of off-specification microspheres will be returned to the leaching step. The specification-grade microspheres will be routed to the particle coater, where the inner graphite coatings, the SiC coating, and the outer graphite coating are to be applied. After inspection, the coated particles will

be formed into fuel sticks. The sticks will also be inspected and then loaded into the fuel blocks to form the loaded recycle fuel element. Reject coated particles and sticks, containing approximately 1 kg of heavy metals per day, will be recycled back to the burner.

3. HANDLING OF WASTE STREAMS

The processes and equipment necessary for preparing the gaseous, liquid, and solid waste streams generated during the radioactive demonstration for discharge to the ORNL waste systems are discussed below. No attempt has been made here to discuss either the method or the cost of handling these wastes after they leave the development demonstration facility; it is assumed that, by 1976 (the planned date of the demonstration), the ORNL waste systems will be capable of handling them.

3.1 Gaseous Waste Streams

Five gaseous waste streams are listed in Tables 3 and 4. The waste gas stream (No. 43) from the Head-End Pilot Plant consists of the off-gas streams from the primary burner and the fertile-particle burner, plus small bleed streams from the inert blanket gas in the enclosures housing the primary burner feed preparation equipment and the fertile-particle crusher. The volume of this stream depends on how much of the graphite in the fuel elements is burned. The Head-End Pilot Plant flow-sheet assumes that 75% of the graphite in each fuel block (this does not include the graphite associated with the fuel sticks or the coated particles) is physically separated and discarded as barren graphite before burning. The flow rate of the combined off-gas stream is 32.2 scfm, which would increase to about 75 scfm if all the graphite were burned.

The combined off-gas stream (No. 43) will contain both particulate and gaseous radioactive material. A metallic primary filter system, operated at a temperature of 250°C or less, will remove more than 99% of the particulate matter. Some of the fission product oxides (Ru, Cs, etc.) have significant vapor pressures at the temperatures prevalent

during the combustion steps and will be vaporized into the off-gas stream. Hot-cell studies have shown that most of the volatile fission product oxides will be removed (DF's of 10^4 to 10^5) by the primary filters.⁵ The very small amount of material passing the primary filters would be removed either by secondary fiberglass filters or, more likely, by the downstream decontamination steps for ^{85}Kr , ^3H , etc. Previous studies have shown that, when SiC-coated particles are crushed and burned, 98% of the tritium, noble gases, and iodine would be released into the composite off-gas stream.⁶ Since the reference flowsheet indicates that 75% of the graphite and all of the fissile particles will be discarded, most of the activity found in the off-gas would be contributed by the fertile particles. Thus, burning 9.67 FSVR fuel elements per day (after 2 years of exposure in the reactor and 150 days of cooling) would release 1020 Ci (0.7 Ci/min) of ^{85}Kr , 16 Ci (0.01 Ci/min) of ^3H , and 1 Ci of ^{131}I , assuming the activity of the graphite to be negligible. Table 5 shows the composition of the combined off-gas stream. The several systems proposed for decontaminating the filtered off-gas⁷ are briefly discussed below.

3.1.1 System A

In this system, the Xe, Kr, O_2 , and N_2 are separated by distilling them from liquid CO_2 . A schematic of the proposed process is shown in Fig. 2. The off-gas is first passed through a catalyst bed to oxidize any CO and $^3\text{H}_2$. Removal of the ^{131}I is accomplished in a zeolite bed containing 13X molecular sieves impregnated with silver. Next, the tritiated water is removed, probably by injecting steam and condensing it. The ^3H -containing water would be sent to the ORNL Liquid Waste System. The gas leaving the condenser is then compressed to 20 atm, cooled to -50°F , and fed to the stripping column. The decontaminated liquid CO_2 would be vaporized and discharged to the stack. The gas leaving the top of the still would be fed to cold traps (two are required) operating at 20 atm and -140°F , where most of the remaining CO_2 would freeze out. Gas leaving the cold trap would contain 1% or less CO_2 in addition to the O_2 , N_2 , Xe, and Kr. This gas can be compressed to about 2000 psi

Table 5. Composition of the Combined Off-Gas Stream

Component	Flow Rate (scfm)	Concentration (vol %)
Carbon dioxide	28.9	90.5
Oxygen	2.4	6.9
Nitrogen ^a	0.9	2.6
Total	32.2	100.0
Total Kr + Xe	23.6 ppm (1 mCi of ⁸⁵ Kr per liter)	

^a It should be noted that the nitrogen content of the off-gas can be reduced to an arbitrarily low value by eliminating its use in equipment and instrumentation, and by eliminating leaks in the burner.

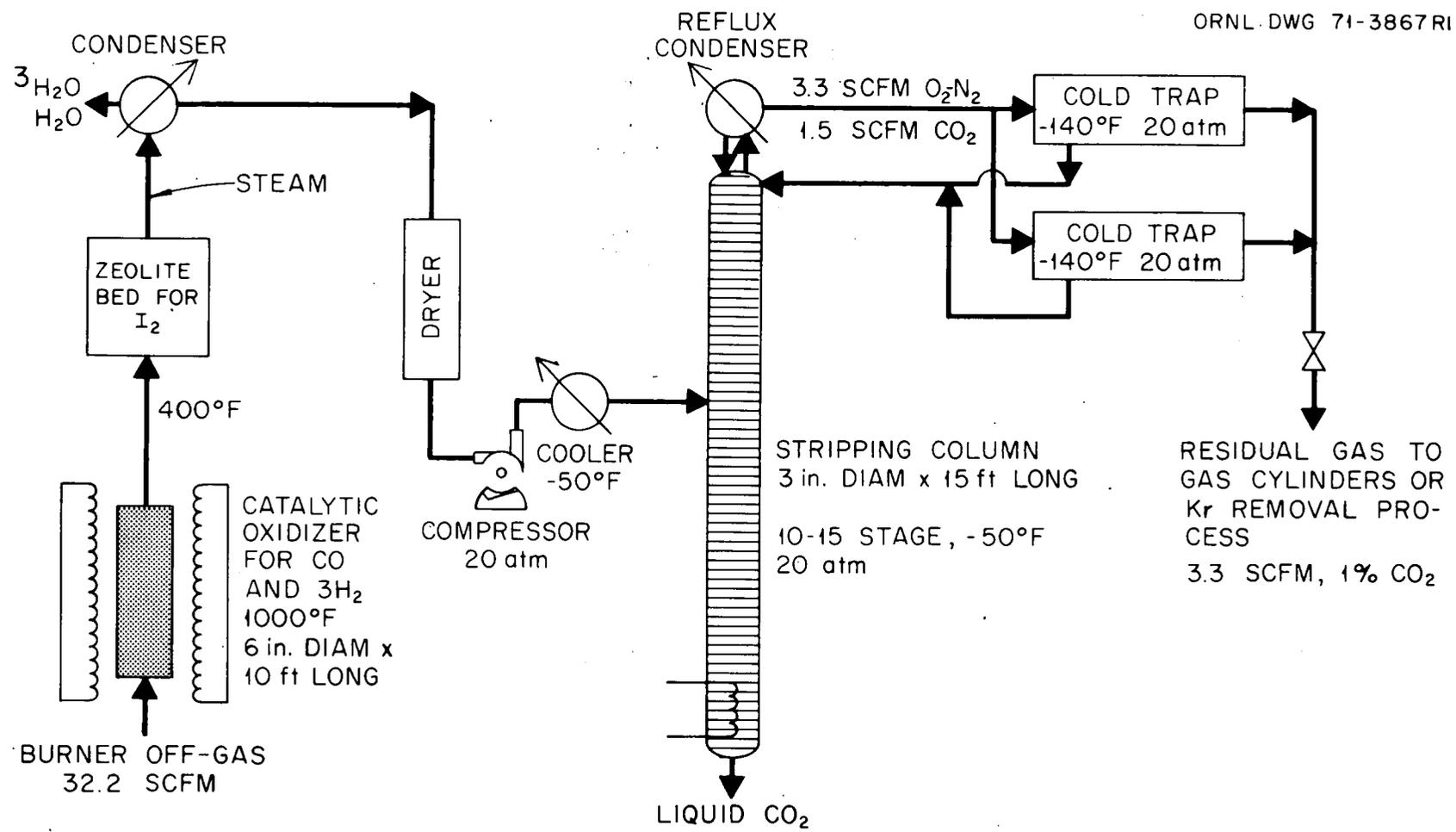


Fig. 2. System A - Distillation of Liquid CO_2 Process.

and stored in 220-scf gas cylinders (20/day) or sent to another process to further concentrate the krypton.

3.1.2 System B

After the CO , $^3\text{H}_2\text{O}$, and ^{131}I have been removed by the methods described for system A, the off-gas stream would be compressed to 300 psig and the CO_2 would be absorbed in a hot (275°F) aqueous solution of potassium carbonate. The CO_2 would react to form bicarbonate, which is regenerated by heating at atmospheric pressure. The decontaminated CO_2 would be discharged to the stack. Absorption of CO_2 is accomplished in a series of scrubber columns of successively smaller diameter to compensate for the decreased flow of gas. A schematic of this process is shown in Fig. 3. The gas exiting from the absorber contains the Kr, Xe, O_2 , and N_2 , and can be stored in gas cylinders (20 per day). The concentration of CO_2 in the stored gas will be less than 1%. A second process can be used to further concentrate the krypton.

3.1.3 System C

System C uses the same methods as system A to effect removal of CO , $^3\text{H}_2$, and $^{131}\text{I}_2$. It is assumed that the amount of krypton sorbed, or otherwise held, by solid CO_2 formed in the cold trap at -140°F is very low. The off-gas is compressed to 20 atm, precooled to -50°F, and then passed into cold traps where the CO_2 is solidified at -140°F (see Fig. 4). The residual gases leaving the cold trap would contain 1% or less CO_2 . These gases would be compressed and stored in standard 220-scf gas cylinders (20 per day). Loaded cold traps would be flushed with nitrogen and warmed to -40°F, and the decontaminated liquid CO_2 would be vaporized and discharged to the stack.

The volume of residual gas is essentially the same for systems A, B, and C, and will fill 20 standard 220-scf gas cylinders per day, or about a total of 2000 cylinders, if 900 blocks are processed. This points out the need to keep the amount of nitrogen and oxygen in the off-gas to a minimum. The conservative O_2 and N_2 concentrations shown

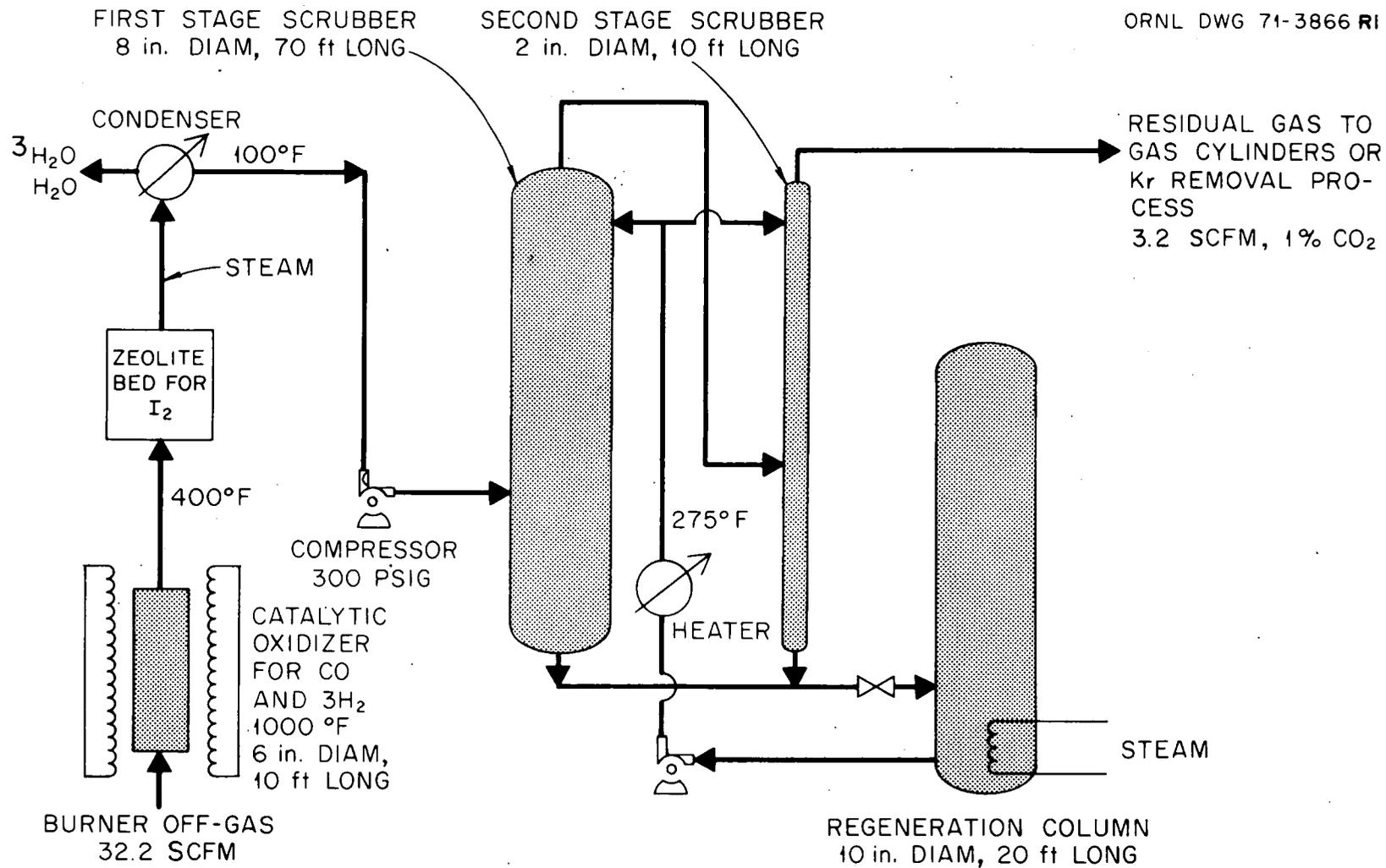
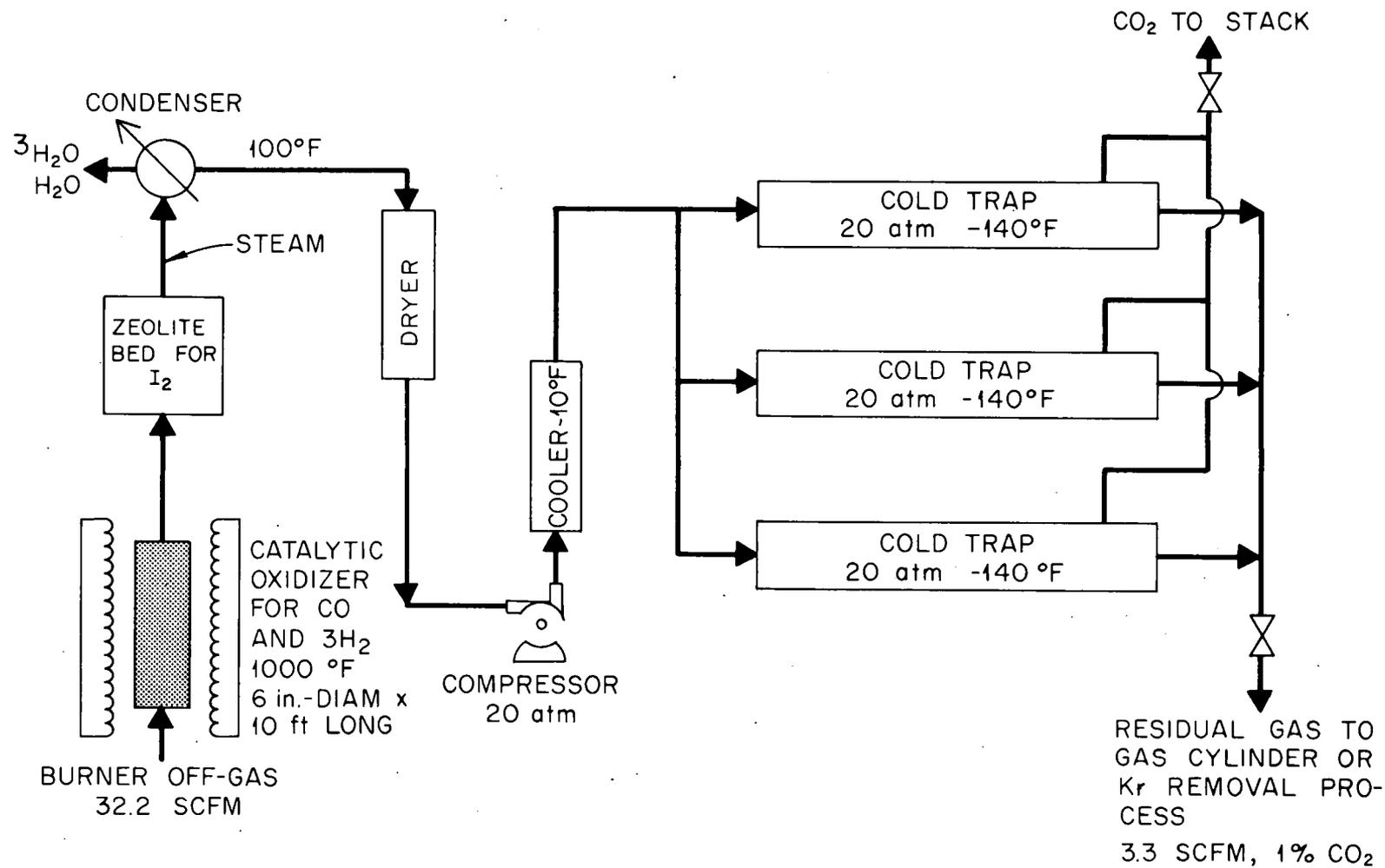


Fig. 3. System B - Aqueous Potassium Carbonate Absorption of CO₂ (KCAC).



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Fig. 4. System C - Controlled Solidification of CO_2 (CSC).

in Table 5 can possibly be reduced by a factor of 2; this would reduce the required number of cylinders per day to 10, which is still rather high. However, any further volume reduction can only be obtained by stripping the Xe-Kr fraction from the O_2-N_2 . Two systems for further concentrating the Xe-Kr are discussed below.

3.1.4 System D

Liquid nitrogen is used to strip Xe, Kr, and O_2 from the feed gas (see Fig. 5). The noble gases are then separated by distillation. Considerable work has been done on this process,⁸ and it could be adapted to the residual gas from systems A, B, and C. A maximum of two 220-scf gas cylinders should suffice to hold the product gas.

3.1.5 System E

This method uses a fluorocarbon solvent (e.g., Freon 12) to scrub the Kr and Xe from the entering gas stream (see Fig. 6). Since CO_2 is also absorbed, reporting with the Xe and Kr in the product gas stream, its concentration in the entering gas stream should be as low as possible, typically 1% or less. Considerable development work has been done on the process,⁹ and results indicate that it could be adapted to handle the residual gas streams (i.e., after CO_2 removal) typical of those of systems A, B, and C.¹⁰ The product gas from burning 900 blocks could probably be stored in three or four 220-scf gas cylinders.

A variation of System E has been proposed in which liquid CO_2 is used to scrub the Kr and Xe from the entering gases while rejecting 99.9% of the O_2 and N_2 . The Kr-rich liquid CO_2 leaving the scrubber would be distilled, with the Kr and Xe reporting to the product gas leaving the top of the still. The decontaminated liquid CO_2 leaving the bottom of the still would provide the scrub stream. Excess CO_2 would be vented to the stack. Since this process appears to be capable of concentrating the Kr in one step to the same level achieved by the two-step process described previously, an engineering evaluation will be made as soon as sufficient information becomes available.

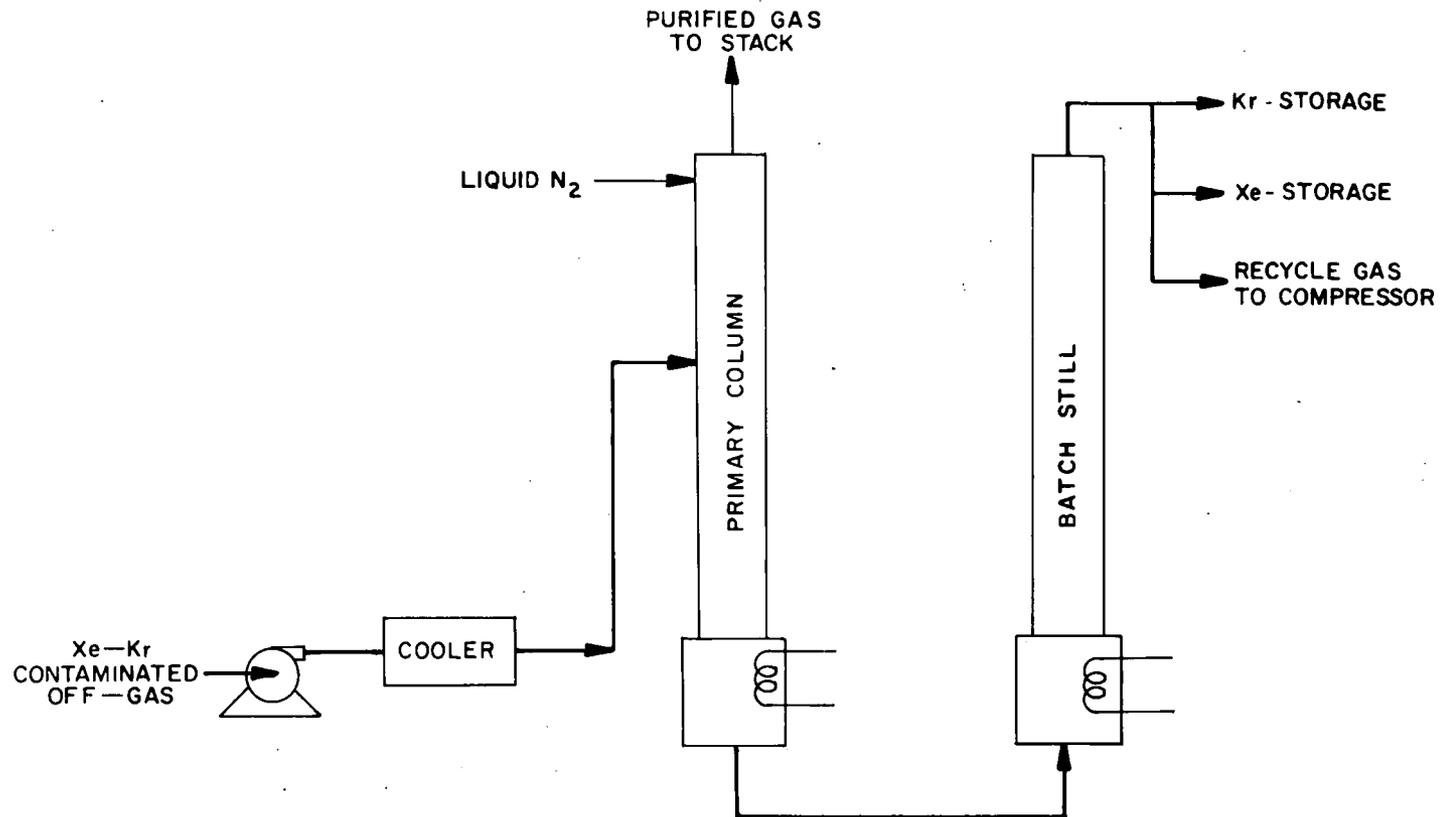


Fig. 5. Scheme D - Removal of Xenon and Krypton by Cryogenic Distillation.

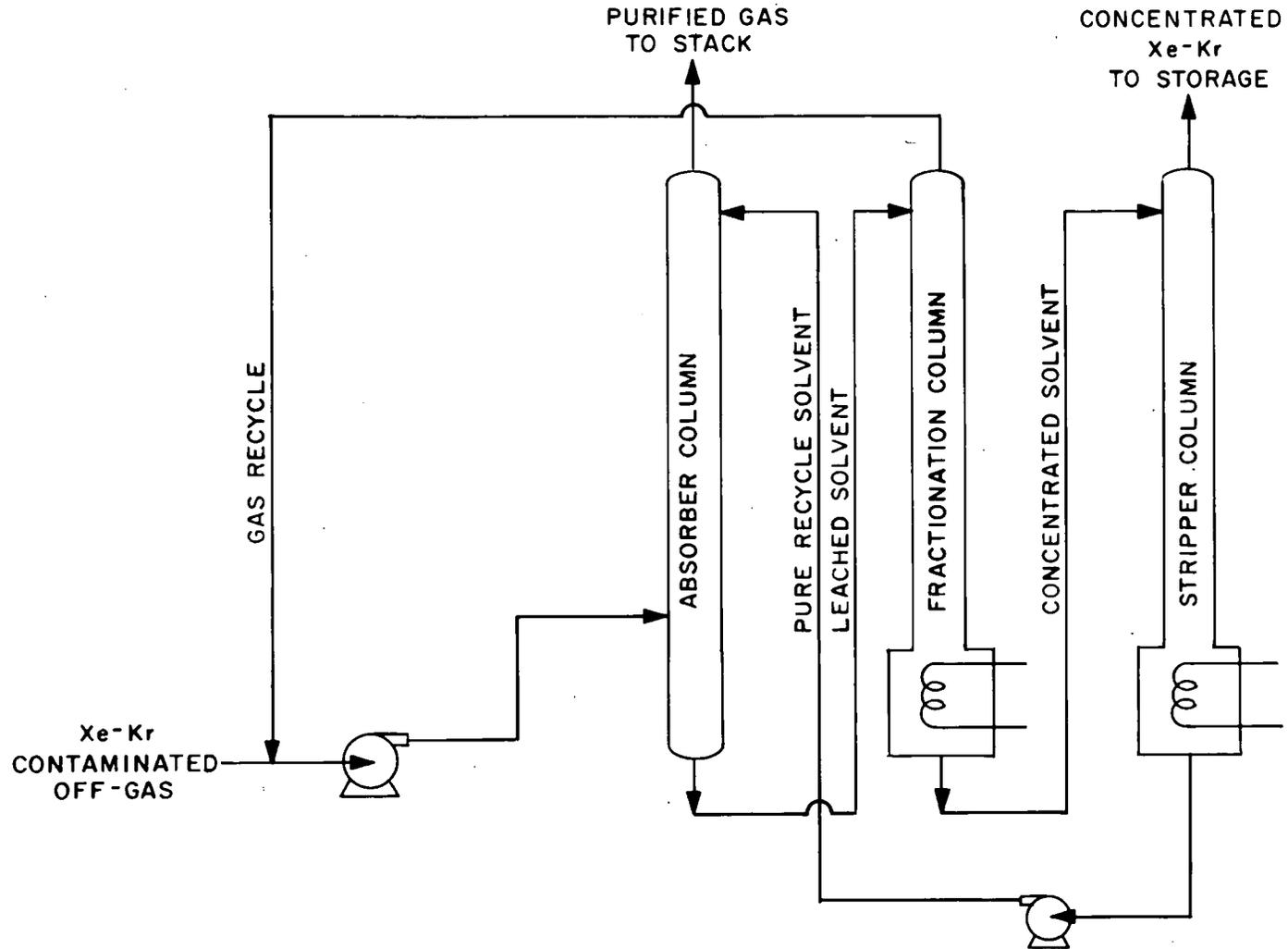


Fig. 6. Scheme E - Removal of Kr-Xe with Fluorocarbon Solvent.

The off-gas from the leacher in the Acid-Thorex pilot plant may contain significant amounts of ^{131}I , ^3H , and ^{85}Kr if these are not completely removed during the burning step. If hot-cell tests indicate that incomplete removal will cause problems, either system D or system E could be adapted to remove ^{85}Kr from this off-gas (which contains no CO_2). In the absence of these radioactive contaminants, passing the off-gas through a caustic scrubber to remove acid vapors, followed by venting to the stack, should be sufficient.

Streams 333, 401, and 501 comprise the gaseous waste streams from the Refabrication Pilot Plant. Stream 333 is the combined off-gas from the gel sphere drying and sphere-firing steps and consists primarily of Ar containing less than 5% H_2 . Stream 401 is the gas from the caustic scrubber that removes the HCl from the particle coater off-gas. Its flow rate varies from 1-2 scfm to a maximum of 60 scfm. It contains Ar, H_2 , and hydrocarbon gases. Stream 501 is the off-gas from the fuel stick carbonization furnace and contains Ar, hydrocarbons, H_2 , CO, CO_2 , and N_2 . All three streams will be combined, filtered, mixed with air or CO_2 from the off-gas decontamination step, and discharged to the stack. An alternative method would be to burn the H_2 , etc., before discharge.

4. LIQUID WASTE STREAMS

Both aqueous and organic liquid wastes are generated in the fuel recycle operations (see Tables 3 and 4). All of the aqueous wastes will be collected in the hold tanks where their volumes, compositions, and radioactivity levels will be measured. After their concentrations have been adjusted to meet the activity level specified, these liquids will be pumped to the ORNL liquid waste disposal system. The waste disposal system will also provide storage if it is decided to retain the thorium product solution. Stream 404, the aqueous waste from the particle coater off-gas scrubber, contains chloride ions; however, it can be pumped through the existing system provided its pH is greater than 11.0.

Small volumes of alpha-contaminated liquid organic waste are produced in the sphere-forming process (i.e., stream 311) and in the stick

making process (i.e., stream 504). The present practice is to combine organic wastes with larger volumes of aqueous waste and send them to the liquid waste disposal system. Unfortunately, most of stream 311 consists of surfactants, which would cause problems during evaporation. This particular waste can be disposed of by burning in a fluidized-bed type of burner, or it can be absorbed on Microcel-E and sent to the waste disposal system in the form of a solid packed in drums.

5. DISPOSAL OF SOLID WASTES

The streams of solid wastes that are generated are listed in Tables 3 and 4. Criticality and fission product decay heat considerations dictate that the cans holding the fissile particles (stream 27) be 6 in. in diameter or less. The concept of a canning station in which aluminum cans would be filled and the end closure welded into place is shown in Fig. 7. The full cans of fissile particles would be stored either in cell F, Bldg. 7930, or in the fuel storage basin during the demonstration run. They would then either be returned to the Head-End Pilot Plant for reprocessing to recover the uranium or transferred to the ORNL solid waste disposal system using the existing Pu-Al carrier (ORNL carrier No. 10570-153).¹¹ The fertile-particle burner ash, while not a waste stream, would be packaged and shipped to Bldg. 3019 using a similar can and the same canning station and carrier. A similar canning station would be required in Bldg. 3019 to package the dried filter cake [stream 144 (see Table 3)]. The empty cans for shipping fertile-particle burner ash would be utilized to hold the filter cake. Any cans not needed, plus the ends that were removed when the cans were opened, could be dissolved to provide aluminum nitrate for salting purposes in the solvent extraction feed; this would also serve to eliminate a solid waste stream.

The barren graphite [stream 10 (see Table 4)] from the screen-tumbling step constitutes the largest volume of graphitic solid wastes, approximately 18 ft³/day. This stream would be combined with the other graphitic wastes (streams 405, 406, 407, 505, 506, 604, and 605) for disposal. Only two disposal methods appear feasible for these wastes:

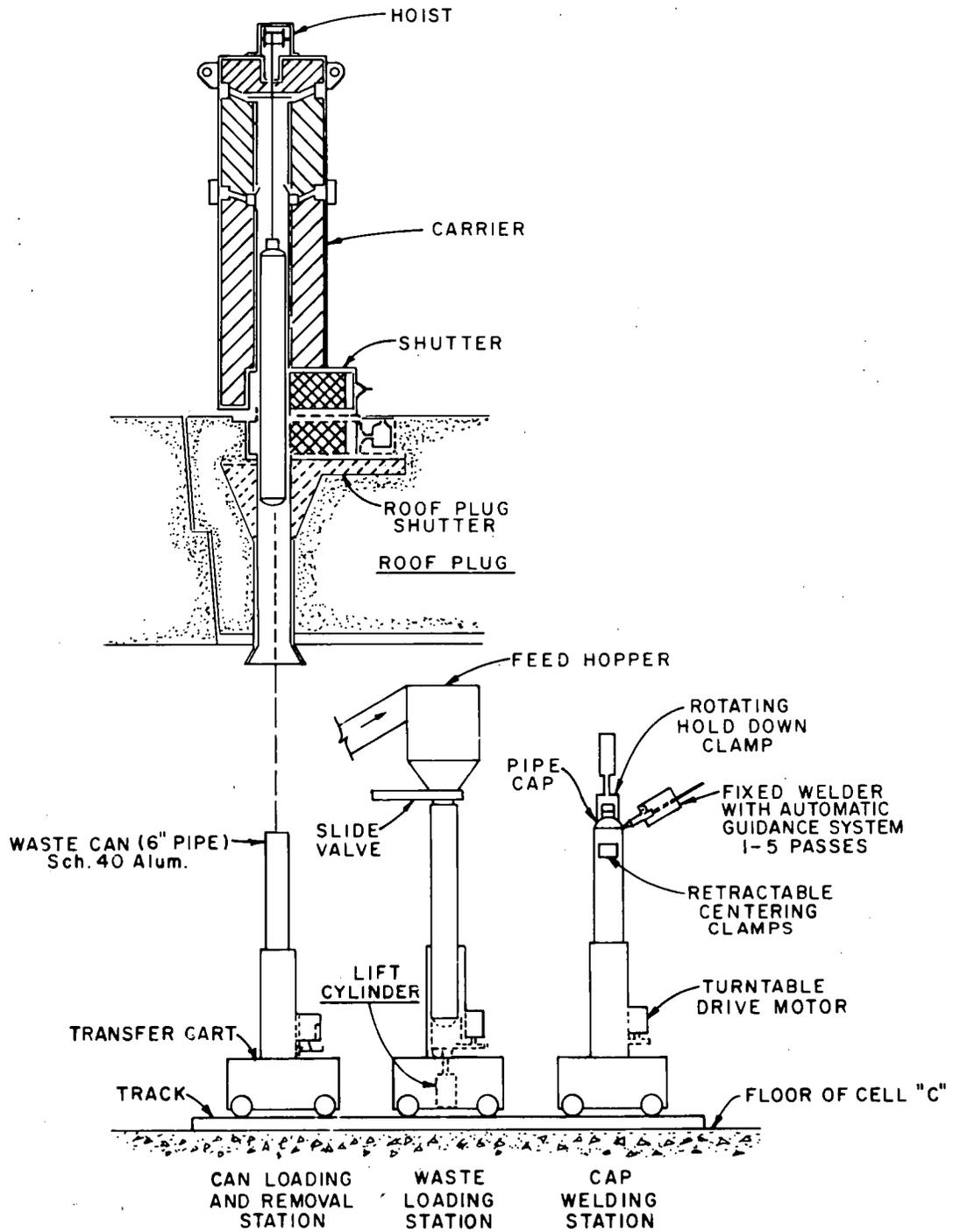


Fig. 7. Fissile Particle Removal Concept.

burning in an auxiliary burner, or removal from the cell and subsequent discard to the ORNL solid waste disposal system. The amount of the combined graphite waste streams is about twice that handled by the primary burner, and space limitations preclude the use of an auxiliary burner and off-gas decontamination system in Bldg. 7930. The effect of burning all the graphite in the primary burner was considered in estimating the cost of the off-gas decontamination equipment.

A concept for removing all solid waste except the fissile and fertile particles is shown in Fig. 8. The waste material would be placed in a can, which would be removed from the cell using a "bag-out" technique. The bagged can would then be sealed in a 55-gal drum, which would be sent to the ORNL solid waste disposal system. The radioactivity level of this waste material would depend largely on the quantities of fertile and fissile particles associated with the barren graphite leaving the screen-tumbler. Thus, a loss of 0.1% of the uranium and thorium in a fuel block would result in a barren graphite containing approximately 50 Ci of mixed fission products per cubic foot and necessitating several inches of lead shielding around the can. Miscellaneous solid wastes (gloves, contaminated equipment parts, etc.) can also be removed using this system.

6. COST ESTIMATES FOR THE WASTE HANDLING SYSTEMS

Order-of-magnitude capital cost estimates have been prepared for the major waste handling concepts using the following ground rules:

1. Unless otherwise noted, the material of construction is high-quality stainless steel.
2. The equipment is assumed to be fabricated and purchased from commercial vendor(s).
3. The equipment is assumed to be installed by the CPFF contractor.
4. Costs are given in 1971 dollars.
5. Facility space is assumed to be available, without modification, except in the case of solid waste.
6. All remote viewing and handling equipment (windows or TV cameras, cranes, manipulators, etc.) are already installed and operable.

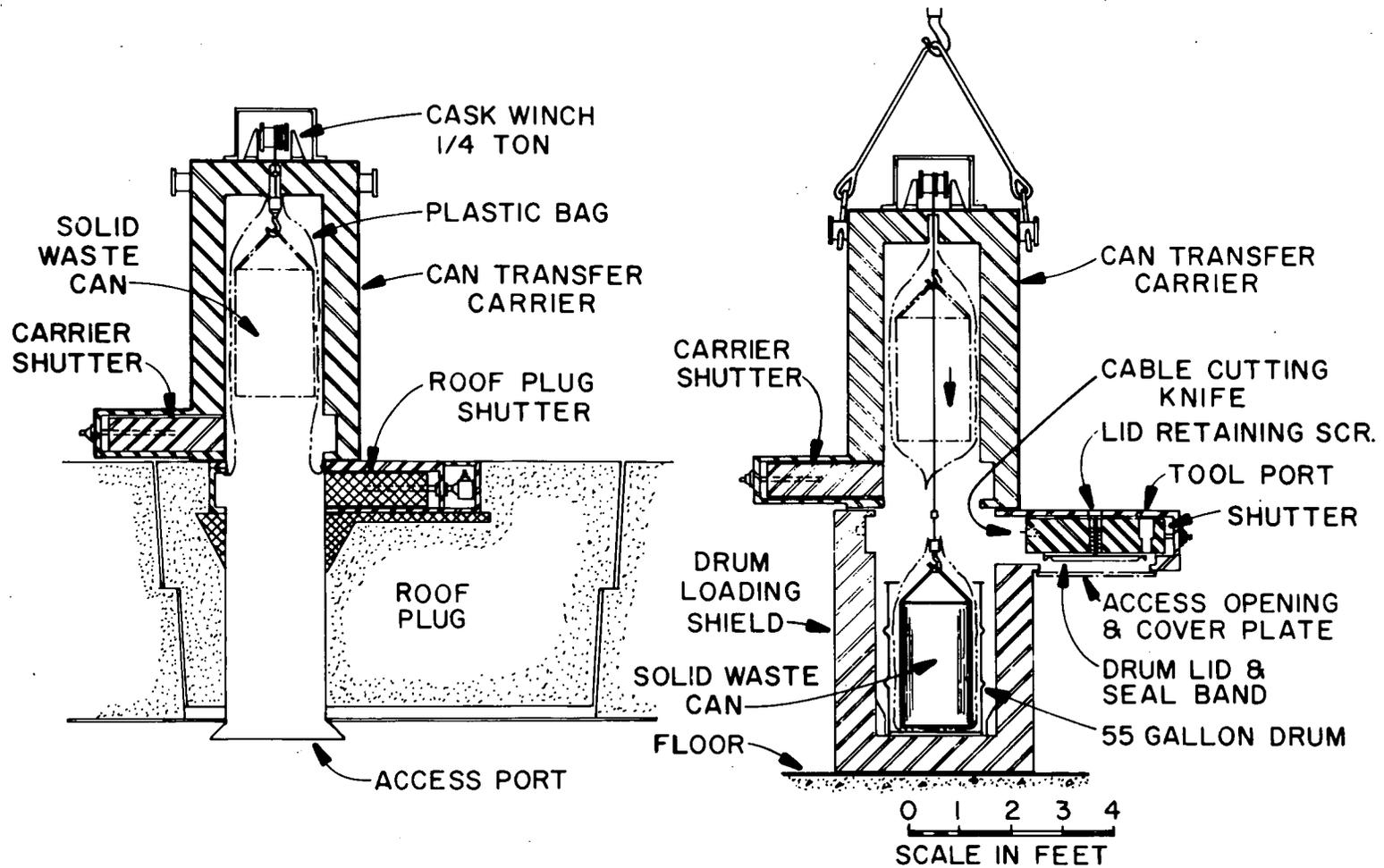


Fig. 8. Concept for Removal of Solid Wastes.

7. All equipment is installed in clean areas; that is, no radioactive working conditions are assumed for the construction.
8. All services are assumed to be available at the outside cell wall or immediately adjacent to it.
9. No changes in the cell ventilation systems are assumed.

The volume of off-gas handled by systems A, B, and C depends on how much graphite is actually burned; accordingly, two cost estimates were made for these systems. One estimate assumed that 75% of the block graphite is removed via the screen-tumbler prior to burning (i.e., partial-block burning), resulting in an off-gas flow rate of 32.2 scfm. The other estimate assumed that the entire fuel element (plus the graphite waste from the Refabrication Pilot Plant) was burned (i.e., whole-block burning), giving an off-gas flow rate of approximately 75 scfm. The CO₂-free Kr-O₂-N₂ stream from these three systems varies from 3 to 6 scfm, but was assumed to be 5 scfm for estimating the cost of systems D and E. Sufficient information was not available to estimate the cost of a system using liquid CO₂ to scrub out the Kr and Xe from the off-gas. A caustic scrubber will also be required to remove HCl from the particle coater off-gas. The estimated cost of these concepts are shown in Table 6.

The estimated costs of the solid waste removal systems are shown in Table 7. Since the radioactivity level of the solid wastes may make shielding necessary, the cost of the solid waste removal station was estimated assuming the transfer carrier to be fabricated either from 1-in. steel or from lighter steel covering 4 in. of lead shielding. Also shown in this table is the cost of a fluidized-bed burner for disposing of liquid organic wastes. Although it will be necessary to modify the cell roof plugs to provide the penetrations required for the solid waste removal systems as well as those for the fuel element loading station, the cost of this modification was not estimated.

The costs associated with the major waste handling systems needed for each pilot plant are shown in Table 8. The Head-End Pilot Plant requires a fissile-particle removal station, which also handles the fertile-particle burner ash. The solid waste removal system will be needed to

Table 6. Capital Cost Estimates for the Off-Gas Decontamination Concepts

System	Type	Cost (thousands of dollars)	
		Partial-Block Burning	Whole-Block Burning
A ^a	Equipment	230	368
	Engineering	35	56
B ^b	Equipment	230	368
	Engineering	35	56
C ^c	Equipment	210	336
	Engineering	25	40
D ^d	Equipment	125	125
	Engineering	25	25
E ^e	Equipment	125	125
	Engineering	25	25

Caustic Scrubber	Equipment	12	
	Engineering	2	

^a Removal of CO₂ by liquefaction-distillation.

^b Removal of CO₂ by hot potassium carbonate solution.

^c Removal of CO₂ by freezing.

^d Concentration of krypton by Freon absorption process.

^e Concentration of krypton by cryogenic distillation.

Table 7. Capital Cost Estimates for the Solid
and Liquid Waste Removal Concepts

System	Type of Cost	Cost (thousands of dollars)
Fissile-particle removal station	Equipment	38
	Engineering	18
Solid waste removal station (4 in. lead shielding)	Equipment	93
	Engineering	10
Solid waste removal station (1 in. steel)	Equipment	39
	Engineering	10
Fluidized-bed burner for liquid organic waste disposal	Equipment	15
	Engineering	3

Table 8. Capital Cost for the Waste Disposal Systems Required
for the HTGR Fuel Recycle Demonstration Pilot Plants

System	Cost	
	Whole-Block Burning	Partial-Block Burning
Head-End Pilot Plant		
Solids handling		
Fissile-particle removal	\$ 56,000	\$ 56,000
Solid waste removal	49,000	49,000
Liquid Waste	0	0
Off-gas decontamination		
System A	424,000	265,000
System D	<u>150,000</u>	<u>150,000</u>
Subtotal	<u>679,000</u>	<u>520,000</u>
25% for contingencies	<u>169,750</u>	<u>130,000</u>
Total	\$848,750	\$650,000
Acid-Thorex Pilot Plant		
Solid waste removal	\$ 56,000	
Liquid waste	0	
Off-gas decontamination		
System D	<u>150,000</u>	
Subtotal	206,000	
25% for contingencies	<u>51,500</u>	
Total	\$257,500	
Refabrication Pilot Plant		
Solid waste removal		Cost shown in Head-End Pilot Plant
Liquid waste system		
Organic waste furnace		\$ 18,000
Off-gas decontamination		
Caustic scrubber		<u>14,400</u>
Subtotal		32,400
25% for contingencies		<u>8,100</u>
Total		\$ 40,500

handle the other solid wastes, even if the graphitic wastes are burned. There are no liquid wastes. The off-gas decontamination system was chosen to consist of systems A and D. The estimated cost of the major waste handling system for the Head-End Pilot Plant would be \$650,000 for the partial-block burning concept and \$848,750 for the whole-block burning concept. A further charge of \$54,000 would be incurred if lead shielding is required for the solid waste removal system.

The Acid-Thorex Pilot Plant needs a solids removal system similar to the fissile-particle removal system required for the Head-End Pilot Plant, and possibly a system (system D) for removing ^{85}Kr from the dissolver off-gas. The solids removal system would share space with the material handling cubicle, but the cost of this shared space was not estimated. The estimated cost for the Acid-Thorex Pilot Plant waste handling system is \$257,500.

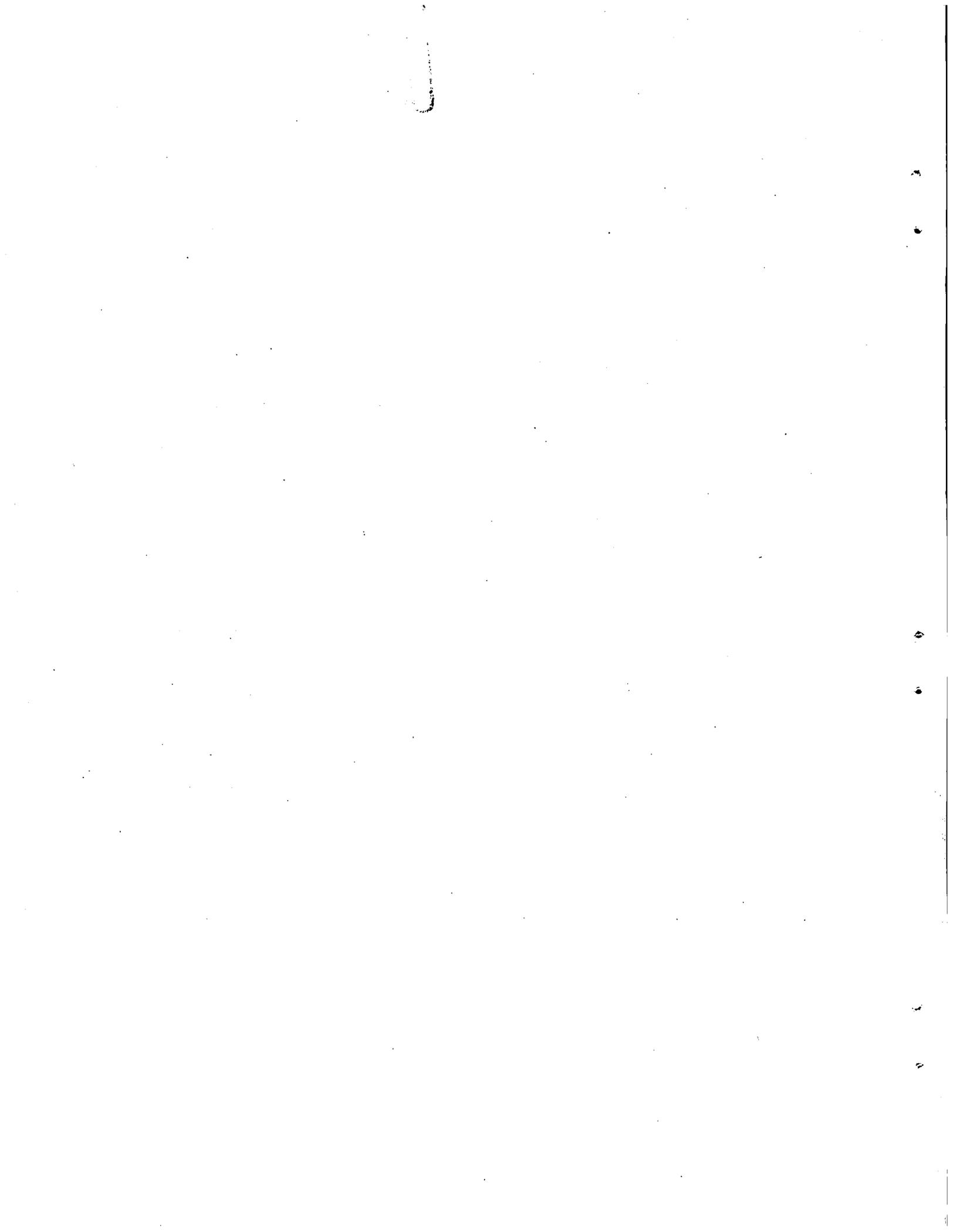
Two major waste disposal systems are required by the Refabrication Pilot Plant: a burner to handle the liquid organic waste, and a caustic scrubber for the particle coater off-gas. The estimated cost for these systems is \$40,500.

7. ACKNOWLEDGEMENTS

Fission product calculations were made, using the ORIGEN code, by M. J. Bell. W. G. Stockdale made the order-of-magnitude cost estimates of the off-gas decontamination systems. C. H. Odom, of the General Engineering Division, contributed to the design of the solid waste handling facilities, and the cost of these facilities was estimated by General Engineering Division personnel. J. M. Holmes and H. W. Godbee contributed valuable advice and information throughout the preparation of the report.

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