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# FISSION PRODUCT GAS RETENTION PROCESS AND EQUIPMENT DESIGN STUDY

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FISSION PRODUCT GAS RETENTION  
PROCESS AND EQUIPMENT DESIGN STUDY

May 1974

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under

Subcontract 3628

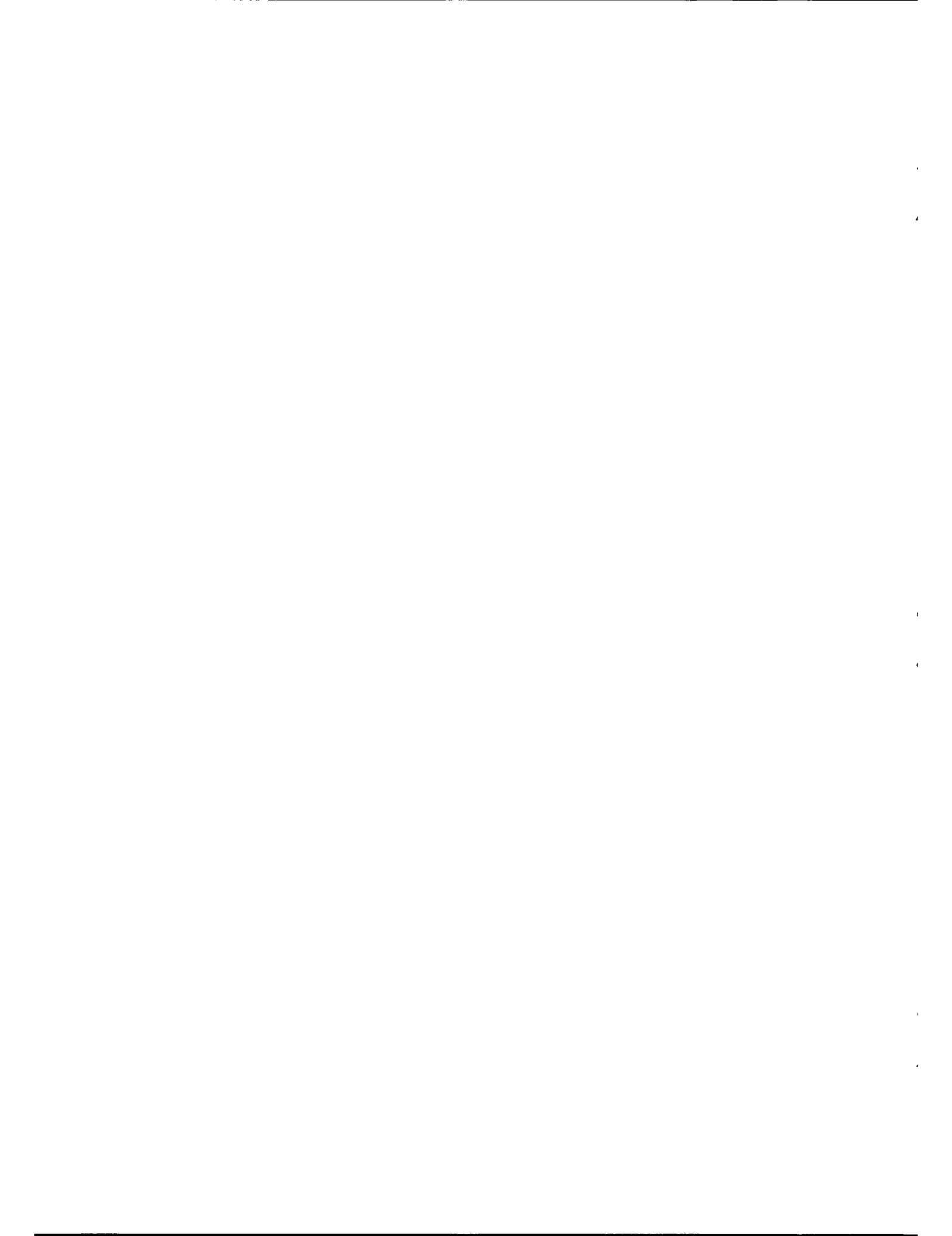
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ABSTRACT

The status of fission gas retention systems of potential application to a fuel reprocessing plant has been reviewed. Two processes currently under development at Oak Ridge National Laboratory and the Oak Ridge Gaseous Diffusion Plant were evaluated to determine their applicability to the Barnwell Nuclear Fuel Plant. These processes are fluorocarbon absorption for krypton recovery and voloxidation for tritium recovery.

Flowsheets for adaptation of these processes to the BNFP were developed and cost estimates prepared for installation of the facilities. The estimates were  $\$5 \times 10^6$  for krypton recovery and  $\$34 \times 10^6$  for tritium recovery. These costs are exclusive of storage charges.

Neither process is sufficiently developed at this time for commercial reprocessing plant installation.

## I INTRODUCTION

Allied-Gulf Nuclear Services under sub-contract to the Oak Ridge National Laboratory has carried out a study of the application of advanced fission gas retention systems to the Separations Facility at the Barnwell Nuclear Fuel Plant.

ORNL under the sponsorship of the AEC has been studying advanced off-gas systems for the LMFBR. Commercial LMFBRs will probably run at higher power levels and to higher burn-up than the current generation of LWRs, thus producing larger quantities of fission products per tonne of fuel. In addition, there probably will be economic incentive to reprocess the fuel after shorter cooling times so that the quantities of fission gases processed will be greater (assuming the fuel can be shipped to the reprocessing plant with short cooling). Also, there is new emphasis on maintaining effluent releases to as low as practicable. As a result, new systems for treatment of the off-gas from nuclear fuel reprocessing plants are being developed. This study is part of the program to evaluate the adaptation of such systems to a commercial reprocessing plant.

Fission gas retention systems have been reviewed and the ORNL processes for tritium and krypton retention were evaluated for adaptation to a commercial plant such as the BNFP.

The Separations Facility is designed to reprocess 5 MTU per day of light-water reactor fuel at an annual rate of 1500 MTU. At an assumed average burnup of 32,000 MWd/MTU and a post irradiation cooling time of 180 days, the krypton and tritium releases from the plant are estimated to be  $1.3 \times 10^7$  Ci/yr and  $5.6 \times 10^5$  Ci/yr respectively.<sup>1</sup> The radiation dose rate to the general public from these releases is very small. Further, the dose rate from releases of krypton and tritium from all nuclear installations by the year 2000 will be only a few percent of that from natural background radiation.<sup>2</sup> However, with the objective of reducing emission of radioactive gases to as low as practicable, development programs have been and are being carried out. Therefore, it is appropriate at this time to evaluate the advanced fission gas retention processes for adaptation to the Separations Facility, which is the largest commercial reprocessing plant in the United States.

Flowsheets for incorporating krypton and tritium recovery into the overall Separations Facility process were developed. The modifications to the plant to implement these flowsheets were determined and cost estimates were prepared. The information generated in this study forms the body of this, the final report on the sub-contract.

## II DESCRIPTION OF THE SEPARATIONS FACILITY

The Separations Facility at the BNFP will recover uranium and plutonium from spent light-water reactor fuels. The fuel will be processed by a modified Purex process, i.e. solvent extraction utilizing TBP in a hydrocarbon diluent as the solvent. The plant is designed to process 5 MTU daily at an annual rate of 1500 MTU. The spent fuel will be cooled a minimum of 160 days prior to processing.

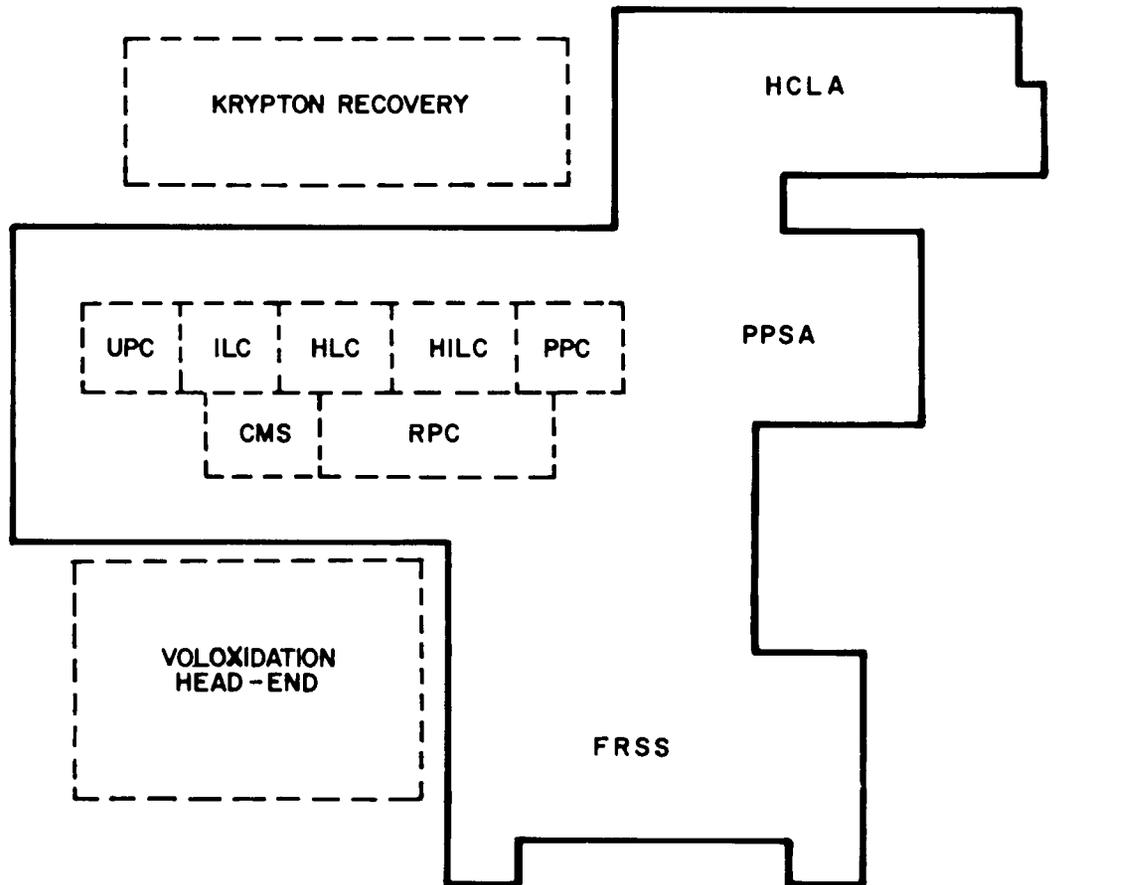
The physical layout of the portion of the plant of importance to this study is shown in Figure 1. The main process building is outlined by the solid line. The shielded process cells within the main enclosure are designated by dotted lines. The location of the krypton recovery unit and the voloxidation head end cell which would be most compatible with the process building are also shown in dotted lines.

Incoming fuel is received and stored in the fuel receiving and storage pool. When scheduled for reprocessing, the fuel is removed from the storage pool and transferred to the remote process cell for shearing and dissolution. The purification process equipment to produce uranyl nitrate and plutonium nitrate is located in the remaining shielded process cells.

The first step in the process is to chop the fuel elements into segments in order to expose the fuel to the nitric acid dissolvent. During the chopping process, any gases in the plenum of a fuel rod will be released and carried into the dissolver off-gas (DOG) system by an air sweep. During dissolution, the remaining fission product gases will be released to the gas phase. Some tritium will be released to the gas phase and some will remain in solution as tritiated water (or tritiated nitric acid).

The off-gas from the dissolver passed through the No. 1 iodine absorber, the  $\text{NO}_x$  absorber, joins the stream from the vessel off-gas system, passes through the No. 2 iodine absorber, through a silver zeolite bed and finally is discharged through the main plant stack.

All krypton is vented through the main plant stack. Tritium in the gas stream from the dissolver is also vented through the stack. Tritium which remains in the aqueous phase passes into the first cycle contactor, exits in the waste stream and is fed to the high activity waste concentrator. The split in the concentrator is approximately 10 to 1, vapor to liquid. The overheads are eventually vaporized and discharged as water vapor through the stack. The tritiated water which is stored in the high level waste will ultimately be released during



UPC - URANIUM PROCESSING CELL  
 ILC - INTERMEDIATE LEVEL CELL  
 HILC - HIGH INTERMEDIATE LEVEL CELL  
 PPC - PLUTONIUM PROCESSING CELL  
 CMS - CONTACT MAINTENANCE STATION  
 RPC - REMOTE PROCESS CELL

HCLA - HOT-COLD LABORATORY AREA  
 PPSA - PLUTONIUM PRODUCT STORAGE AREA  
 FRSS - FUEL RECEIVING AND STORAGE

Fig. 1. BNFP Separations Facility.

waste solidification so that tritium in the incoming fuel is essentially all released to the atmosphere, although a small fraction does undergo some decay prior to release. }!

The processes presently being developed which may eventually be applicable to the retention of krypton and tritium at a plant such as the BNFP are discussed in the subsequent sections.

### III KRYPTON RETENTION

#### Review of Krypton Recovery Methods

Several processes for the removal of krypton (and xenon in some cases) have been studied at various levels of effort through the past several years. As the rare gases are essentially chemically inert, the processes investigated have been based on physical properties such as absorption, adsorption, or diffusion. The absorption processes are based on absorption in solvents such as fluorocarbons, liquid carbon dioxide and certain organic materials. Cryogenic distillation is based on initial absorption of krypton in liquid nitrogen. The adsorption processes are based on adsorption on charcoal or molecular sieves at ambient or cryogenic temperatures. Selective diffusion through membranes is based on solution of the gas under pressure in the membrane and then diffusion through the membrane and dissolution on the low pressure side. Pseudo compounds such as hydrates and clathrates which can be formed under high pressure have also been investigated.

Absorption processes based on fluorocarbons<sup>3,4,5,6</sup> and liquid carbon dioxide<sup>6</sup> have been studied at ORNL (and ORGDP). The basis of the absorption processes is the higher solubility of krypton (and xenon) in the solvent in comparison to air, or other components of the gas stream. By contacting the gas stream in a multi-stage absorber the krypton and xenon are selectively absorbed. Additional steps are then utilized to separate the absorbed gases from the solvent. The liquid carbon dioxide process is being developed primarily for the processing of HTGR fuel. The first step in processing HTGR fuel probably would be the combustion of the graphite matrix<sup>7</sup>. Carbon dioxide is condensed in the gas stream from the burner.

The heart of the fluorocarbon absorption process as developed at Oak Ridge<sup>4</sup> is a set of three columns, an absorber, a fractionator, and a stripper. The feed gas is cooled and fed to the absorber where it is contacted by a downward flow of dichlorodifluoromethane (R-12). The krypton, xenon and some air are absorbed. The stripped gas is vented. The loaded solvent is fed to the fractionator where the air is removed. Some krypton and xenon are contained in this gas stream so this stream must be recycled to recover the krypton and xenon. The bottoms from the fractionator are fed to the stripper where the krypton and xenon are removed as a purified fraction. In pilot plant runs, 99.9% of the krypton in the feed gas was removed by this process.

In the cryogenic distillation process, the gas stream containing krypton and xenon is contacted with liquid nitrogen which condenses and absorbs the higher boiling gases into the liquid phase. The lower boiling components can then be removed in a fractionating column leaving behind a product enriched in krypton and xenon which can be subjected to further purification as desired.

A cryogenic distillation unit for krypton recovery has been operated at the Idaho Chemical Processing plant on a campaign basis for several years.<sup>8</sup> Linde division of Union Carbide is developing a cryogenic distillation process for use by a fuel reprocessing plant.<sup>9</sup> Their process is also based on condensation using liquid nitrogen. Airco/BOC has developed a cryogenic distillation process for krypton and xenon retention from off-gas from a boiling water reactor.<sup>10</sup> Their process was designed for use with reactors. In order to adapt their system to a fuel reprocessing plant, extensive pretreatment of the gas stream would be required. Oxygen removal would be necessary to prevent accumulation of ozone with the attendant explosion hazards.

In the cryogenic distillation process as practiced at Idaho,<sup>8</sup> the dissolver off-gas is passed through a caustic scrubber to remove  $\text{NO}_2$ . The gas is then passed over a rhodium catalyst at  $800\text{--}1100^\circ\text{F}$  to decompose  $\text{N}_2\text{O}$ . The gas is cooled, compressed, and passed through a demister and drier. The gas is then fed to one of two alternate cold traps for pre-cooling. The cooled gas is fed to the center of the primary distillation column. Liquid nitrogen is fed into the top of the column condensing the higher boiling gases including krypton and xenon. Periodically the bottoms are transferred to a batch still where most of the nitrogen and oxygen are removed by distillation. When a sufficient quantity of product has accumulated in the batch still, the bottoms are then fractionated into krypton and xenon fractions. Demonstrated krypton recovery has been 44% for the overall process although it was reported that the cryogenic equipment achieved an efficiency of 97%.

The Linde process<sup>9</sup> consists of three main steps: oxygen removal, prepurification, and cryogenic distillation. Oxygen is removed by passing the gas stream through a catalytic recombiner where it is reacted to water with hydrogen. The hydrogen also converts the nitrogen oxides to nitrogen and water. The water and any carbon dioxide are removed from the gas in the prepurification step. The gas stream then enters the first distillation column where krypton and xenon are condensed by a stream of liquid nitrogen. The bottoms from the first column are fed to a second column for krypton and xenon separation and recovery. The system includes many heat exchangers which serve as economizers and certain streams are recycled to maintain proper concentrations.

The adsorption processes developed in the past have been primarily aimed at treating the off-gas from an operating reactor for decay holdup of krypton and xenon. The long-lived  $^{85}\text{Kr}$  would subsequently be vented to the atmosphere.

A German company (AEG) markets an ambient temperature charcoal adsorption system which reduces the radioactivity in a BWR effluent by a factor of 2000. The system for an 1100 MW(e) reactor requires five beds 6 to 9 feet in diameter by 50 feet long.<sup>11</sup> Charcoal adsorbers are a fire hazard so molecular sieves (inorganic zeolites) have been suggested as a substitute but the beds are 2 to 4 times as large as the charcoal bed.<sup>2</sup>

Ambient temperature adsorption does not provide a concentrated product,<sup>2</sup> so further process modification would be required to adapt the process to a fuel reprocessing plant.

Adsorption at liquid nitrogen temperature has the advantage of concentrating krypton and xenon in a small volume and offers the possibility of recovery of the rare gases by temperature cycling the adsorbers. Charcoal is the only adsorbing medium utilized. The process was tested at the Idaho Chemical Processing Plant and the operability was demonstrated. However, the alternate heating-cooling requirements were beyond the capacity of the liquid nitrogen facility so the process was never operated on a continuous basis.<sup>12</sup>

Because of the low temperature utilized and alternate heating-cooling cycles, refrigeration costs would undoubtedly be higher for cryogenic adsorption. Prepurification of the gas stream would be required in order to remove gases which freeze at liquid nitrogen temperatures.

Diffusion through a permselective membrane as a means of separating krypton and xenon has been evaluated at Oak Ridge.<sup>13</sup> The membrane studied was a methyl phenyl polymer containing 30% silica. Krypton and xenon diffuse through the membrane faster than oxygen or nitrogen. Therefore, this technique is a method for concentrating krypton and xenon from a gas stream.

The separation obtained is dependent on the relative solubility of gases in and diffusivity through the membrane.

According to Reference 13, development of improved membrane materials is underway at General Electric. The membranes tested in the ORNL work required operating pressures of 150 psi and many stages would be required to obtain the desired separation. The development program at ORNL has been terminated in favor of the fluorocarbon absorption process.<sup>2</sup>

The formation of solids containing the rare gases has been investigated. Hydrates and clathrates can be prepared but only under high pressures.<sup>14</sup> Certain benzene derivatives will retain krypton and xenon during condensation under pressure. The rare gases are contained interstitially in the lattice. Quinol forms clathrates with xenon and krypton of the approximate composition  $(\text{Quinol}_3) \cdot \text{RG}$  where RG is the rare gas. The clathrates decompose under radiation and elevated temperature.<sup>2</sup> The solid compounds of the rare gases appear to be little more than a laboratory curiosity so do not merit serious consideration for retention of krypton and xenon.

Based on the preceding review of krypton recovery methods, it appears that there are only two processes which are suitable for adaptation to a fuel reprocessing plant. These are fluorocarbon absorption and cryogenic distillation. The other processes either suffer from major technical problems or lack of development effort.

At the present time, there are no strong reasons for selecting fluorocarbon absorption over cryogenic distillation as the reference process for krypton recovery. It appears that when fully developed, both processes should be capable of excellent recovery of the rare gases. However, the fluorocarbon process can more readily operate with impurities in the gas stream.

#### Impact on the BNFP

The addition of a krypton recovery unit to the BNFP would require shutdown of the plant for an undetermined length of time. Cell penetrations into the top of the Intermediate Level Cell will be installed during initial construction. However, in order to make the piping connections it would be necessary to decontaminate the equipment and piping in the cell to allow access for construction.

The off-gas line from the  $\text{NO}_2$  absorber would require cutting and the addition of the piping connections to and from the krypton recovery facility. Valves to by-pass krypton recovery in case the facility was down for maintenance or repair would be installed outside the cell.

Construction of the facility should not have a major impact on the BNFP. As can be seen in Figure 1, construction would be on the opposite side of the Separations Facility from fuel receiving. Delivery of construction materials would have to be scheduled to prevent interference with regular plant shipments.

Additional security forces would probably be required both from the standpoint of safeguarding of special nuclear material and for the safety of the construction crew.

Depending on other construction activities, it might be necessary to clear additional land for construction warehousing and office space. The environmental impact of this activity would have to be assessed.

Additional operators would have to be hired, trained and licensed. Other activities in support of licensing such as preparation of the Safety Analysis Report and Environmental Impact Report would be necessary.

A storage facility for recovered krypton would be required. The area for the facility would depend on the type of storage, low pressure, high pressure, under water or in a circulating air-cooled vault. Management and surveillance of this facility would be necessary.

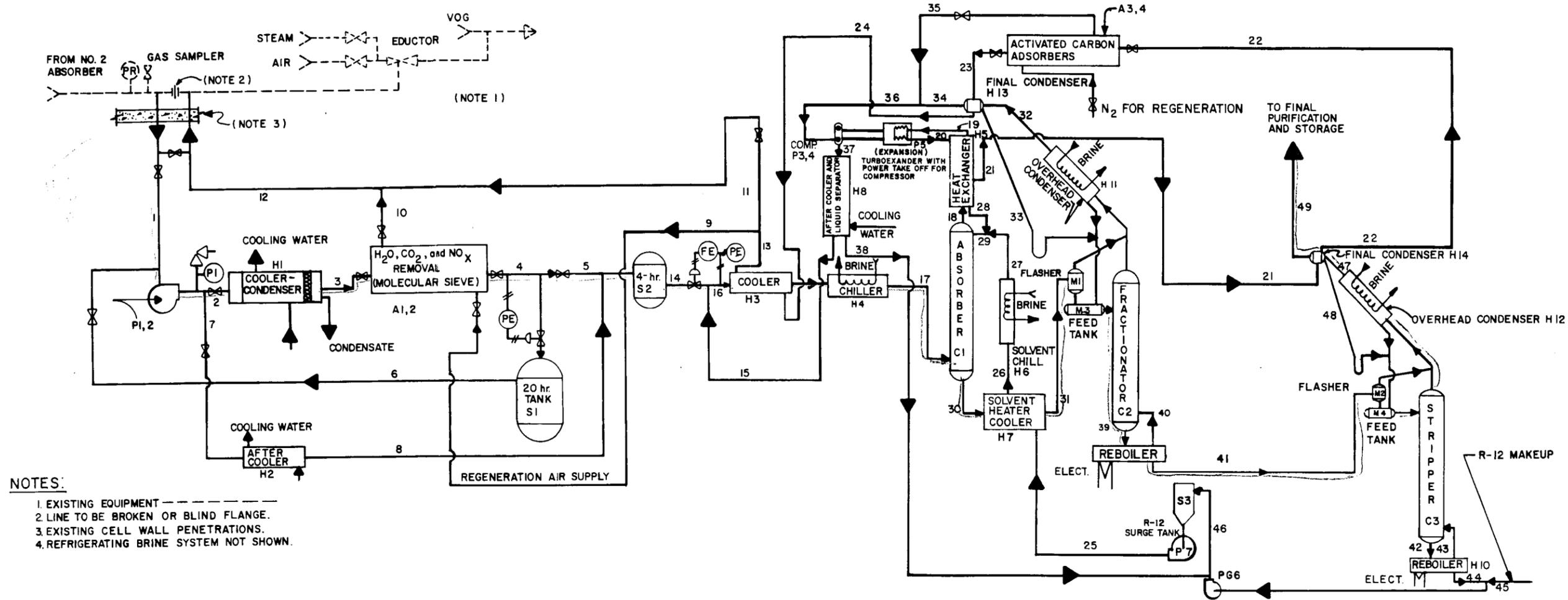
During the construction period, there would be an additional load on the supporting facilities in the area such as parking and the highways.

### Flowsheet

In order to adapt the fluorocarbon absorption process to the BNFP, some additions to the basic Oak Ridge flowsheet were required. In particular feed preparation required additional steps for moisture and  $\text{NO}_x$  removal. Gas flow rate is higher than any considered in the Oak Ridge work. Solvent recovery was also included. However, the heart of the flowsheet is the Oak Ridge process. The aim was to develop a flowsheet which could be adapted to the Separations Facility and provide krypton recovery.

The flowsheet developed is shown in Figure 2. The recovery unit would be located on a pad to the north of the existing process building as shown in Figure 1. The material balance for the flowsheet follows Figure 2 in tabular form. Component flows are shown as lb-mole/min, /mole %, /slm, i.e., as pound moles per minute or in mole %, or in standard liters per minute depending on which unit (or units) is appropriate to the stream. The type of equipment layout envisioned is shown in Figure 3.

Shearing and dissolution of the fuel are on a batch basis, however flows have been averaged to the 5 MTU per day reprocessing rate. The head end of the plant (shear - dissolver complex) is capable of operations at 6 MTU per day. In the event that the head end was operated at this rate for brief periods of time, there is sufficient



- NOTES:
- 1. EXISTING EQUIPMENT
  - 2. LINE TO BE BROKEN OR BLIND FLANGE.
  - 3. EXISTING CELL WALL PENETRATIONS.
  - 4. REFRIGERATING BRINE SYSTEM NOT SHOWN.

Fig. 2. Conceptual Krypton Recovery Flowsheet.

		MATERIAL BALANCE FOR KRYPTON RECOVERY FLOWSHEET				
<u>Line</u>		During Dissolution				
1	Condition for Flow					
3	Stream No.	1	2	3	4	5
5	Flow rate, std liters/min	15,600	15,600	14,600	14,400	12,000
6	lb-moles/min	1.54	1.54		1.42	1.18
8	Temp., °F	90		≤ 90		
10	Pressure, psia	11.4	485		470	
	Component flows, lb-mole/min /mole %/slm					
14	Air (O <sub>2</sub> + N <sub>2</sub> + Ar)	1.41/91.6/14,300		1.41/98.0/14,300	1.41/99.4/14,300	1.17/99.4/11,900
16	H <sub>2</sub> O	0.103/6.72/1050		/0.144/21	-	-
18	NO + NO <sub>2</sub> + HNO <sub>3</sub>	0.0174/1.12/175		/1.2/175	-	-
20	N <sub>2</sub> O	0.008/0.52/81		/0.55/81	/0.56/81	/0.56/67.5
22	CO <sub>2</sub>	0.0015/0.10/15.6		/0.11/15.6	-	-
24	Ne + He + O <sub>3</sub> + H <sub>2</sub>	/0.005/0.74		/0.005/0.74	/0.005/0.74	/0.005/0.62
26	Xe	0.0004/0.025/4.0		/0.027/4.0	/0.028/4.0	/0.028/3.33
28	Kr	0.00004/0.003/0.426		/0.003/0.426	/0.003/0.426	/0.003/0.355
30	R-12	-	-	-	-	-



## MATERIAL BALANCE FOR KRYPTON RECOVERY FLOWSHEET

Line	Normal Operation, assuming negligible R-12 loss to activated carbon, product stream or off-gas									
	13	14	15	16	17	18	19	20	21	
1										
3										
5	11,900	12,000	5,900	17,900	17,900		11,900	11,900	11,900	
6	1.17	1.18	0.58	1.76	1.76	1.21	1.17	1.17	1.17	
8					-20	-20	-120	-150	-50	
10	14	465	457	455	441	439				
14	/ /11,900	/99.4/11,900	0.58/ /5900	/ /17,800	1.75/ /17,800	1.17/ /11,900	1.17/ /	1.17/ /	1.17/ /	
16	-	-	-	-	-	-	-	-	-	
18	-	-	-	-	-	-	-	-	-	
20	-	/0.56/67.5	-	/ /67.5	/ /67.5	-	-	-	-	
22	-	-	-	-	-	-	-	-	-	
24	/ /0.62	/0.005/0.62	/ /0.31	/ /0.93	/ /0.93	/ /0.62	/ /0.62			
26	-	/0.028/3.33	/ /0.67	/ /4.0	/ /4.0	-	-	-	-	
28	-	/0.003/0.355	/ /0.075	/ /0.43	/ /0.43	-	-	-	-	
30	-	-	-	-	-	0.04/ /	-	-	-	

## MATERIAL BALANCE FOR KRYPTON RECOVERY FLOWSHEET

Line

Line	Normal Operation										
	22	23	24	25	26	27	28	29	30	31	
1											
3											
5	11,900	11,900	11,900								
6	1.17	1.17	1.17	3.48	3.48	3.48	0.04	3.52	3.87	3.87	
8	-45	-35	-30	50	-10	-20	-30	-20	-20	40	
10				500		344		439	441		
14	1.17/ /	1.17/ /	1.17/ /	-	-	-	-	-	0.58/ /5,900	0.58/ /5,900	
16	-	-	-	-	-	-	-	-	-	-	
18	-	-	-	-	-	-	-	-	-	-	
20	-	-	-	-	-	-	-	-	/ /67.5	/ /67.5	
22	-	-	-	-	-	-	-	-	-	-	
24			/ /0.62	-	-	-	-	-	/ /0.31	/ /0.31	
26	-	-	-	-	-	-	-	-	/ /4.0	/ /4.0	
28	-	-	-	-	-	-	-	-	/ /0.43	/ /0.43	
30	-	-	-	3.48/ /	3.48/ /	3.48/ /	0.04/ /	3.52/ /	3.28/ /	3.28/ /	

## MATERIAL BALANCE FOR KRYPTON RECOVERY FLOWSHEET

Line	Normal Operation									
	32	33	34	35	36	37	38	39	40	41
1										
3										
5										
6	0.88	0.10	0.78	-	0.78	0.78	0.20	3.45	0.16	3.29
8	0	0	-15					33	33	33
10	45	45	45		45	135	457	46	46	46
14	0.58/ /5,900	-	0.58/ /		0.58/ /5,900	0.58/ /5,900	-	-	-	-
16	-	-	-		-	-	-	-	-	-
18	-	-	-		-	-	-	-	-	-
20	-	-	-		-	-	-	/ /70.9	/ /3.4	/ /67.5
22	-	-	-		-	-	-	-	-	-
24	/ /0.31	-	/ /0.31		/ /0.31	/ /0.31	-	-	-	-
26	/ /0.67	-	/ /0.67		/ /0.67	/ /0.67	-	/ /3.50	/ /0.17	/ /3.33
28	/ /0.075	-	/ /0.075		/ /0.075	/ /0.075	-	/ /0.373	/ /0.018	/ /0.355
30	0.30/ /	0.10/ /	0.20/ /		0.20/ /	0.20/ /	0.20/ /	3.44/ /	0.16/ /	3.28/ /

## MATERIAL BALANCE FOR KRYPTON RECOVERY FLOWSHEET

Line

Line	Normal Operation			Make-up	Normal Operation			
	42	43	44		45	46	47	48
1								
3	42	43	44	45	46	47	48	49
5								100
6	3.83	0.55	3.28			0.01	-	0.01
8	0	0	0	75		-20	≤ -20	-35
10	24	24	24	85	100	23	23	23
14	-	-	-	-				-
16	-	-	-	-				-
18	-	-	-	-				-
20	-	-	-	-				/ /67.5
22	-	-	-	-				-
24	-	-	-	-				-
26	-	-	-	-				/ /3.33
28	-	-	-	-				/ /0.355
30	3.83/ /	0.55/ /	3.28/ /					/ /30

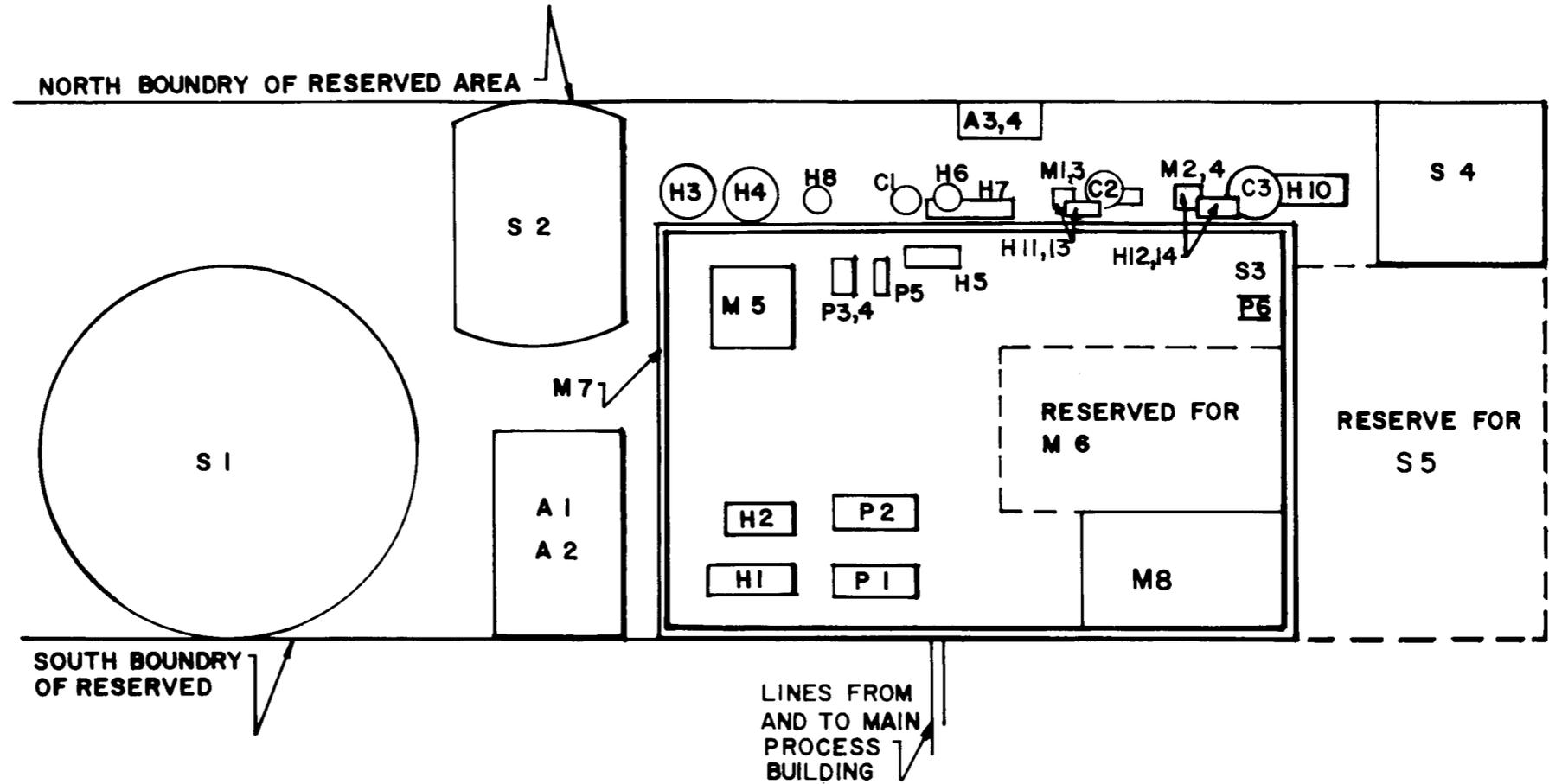


Fig. 3. Equipment Layout for Krypton Recovery.

*[Handwritten mark]*

surge capacity in the krypton recovery unit to accommodate the increased flow.

The off-gas from the fuel dissolver is passed through the #1 iodine scrubber where most of the evolved iodine is removed and then to the NO<sub>2</sub> absorber where NO<sub>2</sub> and some NO (by oxidation to NO<sub>2</sub>) are removed. The gas stream from these clean-up steps would then be piped to the krypton recovery unit.

The composition of the gas stream at this point is given in Table 1.

As can be seen, the krypton is a very minor constituent. Further, there is 8 times as much xenon as krypton.

The gas stream is compressed to 485 psi and passed through a cooler-condenser to remove the heat of compression and condense some of the water in the gas stream. It then passes to a molecular sieve unit for removal of the remaining water, NO, NO<sub>2</sub>, and CO<sub>2</sub>. N<sub>2</sub>O is not removed by the molecular sieve. NO is catalytically oxidized to NO<sub>2</sub> in order to be sorbed on the sieve. Two molecular sieve units are installed in parallel in order to permit regeneration.

①  
Emergency  
stream  
#

Table 1

Composition of Feed to Krypton Recovery

<u>Component</u>	<u>Mole %</u>
H <sub>2</sub> O (10) + Cond = 2-3	6.72
N <sub>2</sub> O 49	0.52
NO (10)-(10)	1.00
NO <sub>2</sub> (10)-(10)	0.08
CO <sub>2</sub> (1)-(10)	0.10
HNO <sub>3</sub> (10)-(12)	0.04
H <sub>2</sub> , Ne, He, O <sub>3</sub> etc, etc	0.005 (total)
Xe (49)	0.025
Kr (49)	0.003
N <sub>2</sub> , O <sub>2</sub> , Ar 22-12	balance

The gas flows from the molecular sieve to a 4-hour surge tank. The capacity of this tank is equivalent to 4 hours of DOG flow with the H<sub>2</sub>O and NO<sub>x</sub> removed. The 4-hour tank and the 20-hour tank are operated to smooth out the gas flow to the remaining process equipment. When the feed pressure to the 4-hour tank exceeds that required for downstream operation, the excess gas is stored in the 20-hour tank. Thus, 24 hours of surge capacity is available so that during the period when the dissolver is not operating, the krypton recovery equipment can be continued in operation. When the shear-leach process is not in operation the normal DOG stream will bypass the krypton recovery system. The 20-hour tank will feed the compressor to maintain the continuous operation mode.

Gas flow from the 4-hour tank is controlled by the primary flow control valve. The gas flows through a heat exchanger cooled by the "clean" air stream which is being returned to the main plant off-gas system. The gas then flows to a chiller which is cooled by mechanical refrigeration to -20°F.

The cold gas then enters the absorber where it is contacted by a counter-current flow of liquid R-12 (CCl<sub>2</sub>F<sub>2</sub>) at -20°F. The liquid absorbs greater than 99.9% of the krypton and xenon and some air (approximately 33%). The liquid stream from the bottom of the absorber passes through a heat exchanger where it is heated to about 40°F. The liquid then flows through a throttling valve to a flash pot. Part of the liquid vaporizes and joins the overheads from the fractionator. The remaining liquid is fed to the fractionator.

A reboiler at the bottom of the fractionator provides the vapor phase for fractionation. The fractionator operates at 33°F and 46 psi. Refluxing R-12 is provided by the fractionator overhead condenser. The dissolved air plus small amounts of krypton and xenon are removed in the vapor phase while the bulk of the krypton and xenon remain dissolved in the liquid. The liquid stream passes through the reboiler where about 5% is vaporized and the remaining liquid flows to the stripping column.

The gas stream which exits the fractionator is recycled to recover the small amount of krypton contained therein. The gas is compressed and cooled. Any R-12 that condenses is returned to the R-12 surge tank and the compressed gas is returned to the feed stream downstream of the flow control valve and upstream of the flow element. This will permit the flow control valve to compensate for any fluctuation in the recycle stream to maintain an even flow to the absorber.

The 95% liquid fraction from the fractionator reboiler flows to the stripping column which operates at 0°F. The liquid flows through an expansion valve into a flash tank where the pressure is dropped to 24 psi, and some of the liquid flashes to vapor and joins the overhead vapor stream from the stripper. The remaining liquid enters the top of the column and is stripped by a counter-current stream of R-12 vapor supplied by the reboiler at the bottom of the column. The overhead condenser operates at -20°F to condense part of the R-12. The final condenser cools the product stream to -35°F.

The composition of the product is given in Table 2.

Table 2

Recovered Krypton Product Composition

<u>Component</u>	<u>Mole %</u>
N <sub>2</sub> O	66.6
R-12	29.7
Xe	3.3
Kr	.35

Final purification and storage have not been specified. Removal of N<sub>2</sub>O either during pretreatment before krypton recovery or by product treatment is necessary both from the safety standpoint and to reduce storage. Removal of R-12 from the product gas is required to avoid the products of radiolytic decomposition.

The gas stream from the absorber is sufficiently purified from krypton to be returned to the DOG system. However, it contains sufficient R-12 to require treatment for recovery of the R-12. Therefore, the gas stream is passed through a heat exchanger to condense part of the R-12 which is returned to the absorber column.

The cold gas at about -120°F then passes into a turbo-expander where in expanding to near atmospheric pressure is further cooled to -150°F. The energy from the expanding gas provides part of the horsepower requirement for the recycle gas compressor.

The gas stream then serves to cool the absorber heat exchanger and the final condenser off the stripper. This stream then passes through a charcoal bed for final R-12 recovery. Two beds are operated in parallel so that one bed can be regenerated while the other is in operation.

[Note added in proof. Recent work at ORNL (W. E. Unger pvt. comm.) has shown that 13X molecular sieve is superior to charcoal as an absorber for R-12.]

The stripped off-gas then serves as the cooling medium for the final condenser on the fractionator, passes through the cooler from the 4-hour feed tank and is finally returned to the DOG system of the Separations Facility.

This treatment of the absorber off-gas recovers essentially all the R-12 plus a part of the energy required in the compression and cooling steps.

The major equipment items in the krypton recovery unit are given in Table 3. The equipment numbers are also shown in location on Figure 3.

Table 3

MAJOR EQUIPMENT LISTING

KRYPTON RECOVERY SYSTEM

Adsorbers, Molecular Sieve Units, etc.

- A1 Molecular Sieve Units (one on line, one being regenerated or on standby; piping and valves for spare unit not shown). Removes H<sub>2</sub>O, CO<sub>2</sub>, NO, and NO<sub>2</sub> from gas stream. Each sized for 14,600 std. liter./min. Shielding equivalent to 1 in. of lead required. See Note 1.
- A2
- A3 Activated carbon adsorbers (one on line, one being regenerated or on standby; piping and valves for spare unit not shown). See Note 2.

Columns

- C1 Absorber; 12-in., Sched. 40, 304 SS, ~ 28 ft high, 25 ft 1 in. packed with 316 SS Goodloe packing.
- C2 Fractionator; 16-in., Sched. 30, 304 SS, ~ 20 ft high, 18 ft packed with 316 SS Goodloe packing.
- C3 Stripper; 24 in., Sched. 20, 304 SS, ~ 16 ft high, 14 ft packed with 316 SS Goodloe packing.

Heat Exchangers, Condensers, Reboilers, etc.

- H1 Cooler-condenser; water-cooled heat exchanger with mist eliminator, Heat load: ~ 95,000 Btu/hr.

Table 3 (cont.)

H2	Aftercooler; water-cooled heat exchanger. Heat load: ~ 70,000 Btu/hr.
H3	Cooler; economizer for process gas; counter-current exchanger, ~ 30,000 Btu/hr.
H4	Process gas chiller, cooled by refrigeration, ~ 53,000 Btu/hr.
H5	Absorber effluent heat exchanger; counter-current cooler-condenser for removing bulk of R-12 from off-gas
H6	Solvent chiller; cooled by refrigeration, ~ 80,000 Btu/hr.
H7	Solvent heater-cooler; counter-current economizer ~, 310,000 Btu/hr.
H8	After cooler for compressed recycle gas and integral liquid separator for removing condensed R-12.
H9	Reboilers for supplying gas to bottom of columns. Heat requirements: H9 - 85,000 Btu/hr; H10 - 295,000 Btu/hr.
H10	
H11	Overhead condensers, cooled by refrigeration; respective capacities of about 330,000 and 480,000 Btu/hr.
H12	
H13	Final condensers; counter-current exchangers. Approximate capacities of 3,000 and 1,000 Btu/hr.
H14	

Pumps, Compressors, etc.

P1	Compressor, each capable of compressing 550 scfm of air at 11.4 psia and 90°F to 33 atmospheres. One on line, one on standby; piping and valves for spare unit not shown.
P2	
P3	Recycle gas compressor, powered by take-off from turbo-expander. See Note 3.
P4	Gas compressor for completing the compression of the recycle stream.
P5	Turboexpander, for reducing pressure and temperature of absorber effluent gas. See Note 4.
P6	Pump for transporting R-12 to pressurized storage.
P7	Pump for feeding R-12 to process.

Table 3 (cont.)

Storage

- S1 Mild steel tank, 23,500 ft<sup>3</sup>, 33 atmos. pressure. See Note 5.
- S2 Mild steel tank, 4,700 ft<sup>3</sup>, 33 atmos. pressure.
- S3 Storage tank for R-12 surge capacity; normal storage under pressure, but jacketed for intermittent cooling with refrigerant.
- S4 R-12 cylinder storage area.
- S5 Krypton cylinder storage area.

Miscellaneous

- M1 Flashers, about 4 ft<sup>3</sup>, for pressure release and adiabatic cooling of R-12.
- M2
- M3 Feed tanks, about 3 ft<sup>3</sup>, for liquid collection, liquid seal maintenance, and column feed.
- M4
- M5 Refrigeration system - cooling capacity of 1,000,000 Btu/hr with circulating refrigerant (brine) at -40°F; not shown on flowsheet.
- M6 Final Purification system (outside the scope of this report).
- M7 Cinder block building 40 x 60 ft.
- M8 Control room.

Notes

1. A1 and A2 equivalent to Union Carbide's PuraSiv N skid-mounted unit, priced at \$255,000. NO-NO<sub>2</sub> level in effluent is guaranteed  $\leq$  50 ppm, and is expected to be  $\leq$  5 ppm. Adsorbent (at a price of \$5,000, included in above) is guaranteed for two years.
2. Effectiveness of the specific type of activated carbon for R-12 retention must be verified. A molecular sieve might be preferable if effectiveness is demonstrated.
3. Using energy from turboexpander to compress recycle gas could not be economically justified from standpoint of

Table 3 (cont.)

power savings. However, energy from turboexpander (~ 50 hp) must be rejected somewhere. Compressing the recycle gas takes care of this energy rejection, and at the same time is equivalent to about 15 hp of compression.

4. A turboexpander meeting the process requirements is manufactured by the Rotoflow Corp., Los Angeles. Estimated current price is \$35,000.
5. Most economical shape probably 36-ft diam. sphere. In this case, pressure requirement can be met by 2-in. thick high-strength steel, double plated.

#### Cost Estimate and Schedule

The krypton recovery system flowsheet and layout were discussed above. The information incorporated therein was used to estimate the cost of adding krypton recovery to the BNF and to project a possible schedule for installation; this cost estimate is presented in Table 4, and the schedule is summarized in Figure 4.

The cost is estimated as \$5,000,000 in 1973 dollars; no provisions for escalation were included. This estimate does not include final krypton purification or storage. Three assumed criteria were crucial to the cost estimate: (1) only the molecular sieve units require shielding, (2) containment of process vessels and equipment is not required, and (3) Class I construction (immune to damage by design-base earthquake or tornado) is unnecessary. If any of these three criteria is not valid, the project cost would be increased significantly.

It must be realized that this cost estimate was based largely on empirical factors rather than on detailed design studies. A more detailed cost estimate would require the services of an architect-engineering firm.

The schedule is shown as a bar chart extending from an arbitrary project initiation date. We estimate that, from the time the project is approved and funds allocated, three years would be required before krypton recovery operations could begin.

Table 4

ORDER OF MAGNITUDE ESTIMATEKRYPTON RECOVERY SYSTEMMajor Equipment

Compressors & Pumps	\$ 123,000
Tanks & Drums	287,000
Special Process Equipment	298,000
Columns & Heat Exchangers	97,000
Utility & Support Systems Equipment	115,000
Subtotal Major Equipment	<u>\$ 920,000</u>

Other Directs

Equipment Foundations & Erection	98,000
Piping	490,000
Electrical & Instrumentation	353,000
Paint, Insulation & Steel	146,000
Buildings & Structures	80,000
Site Improvements	165,000
Subtotal Other	<u>\$ 1,332,000</u>

Total Direct Cost	<u>\$ 2,252,000</u>
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Indirects

Design Engineering	675,000
Construction Indirects & Fee	990,000
Home Office & Associated Capital Cost	<u>360,000</u>

Total Indirect Cost	<u>\$ 2,025,000</u>
---------------------	---------------------

Total Before Contingency	<u>\$ 4,277,000</u>
(Contingency 17%)	<u>723,000</u>

TOTAL PROJECT COST	<u>\$ 5,000,000*</u>
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- \*(1) Excludes Cost of Final Purification & Storage
- (2) Excludes Cost of Land
- (3) Estimate Based on Construction Complete in 1973
- (4) Buildings Are Not Class 1
- (5) Shielding Provided Only Around Molecular Sieve Units
- (6) Confidence Level of Estimate; -10%, +20%

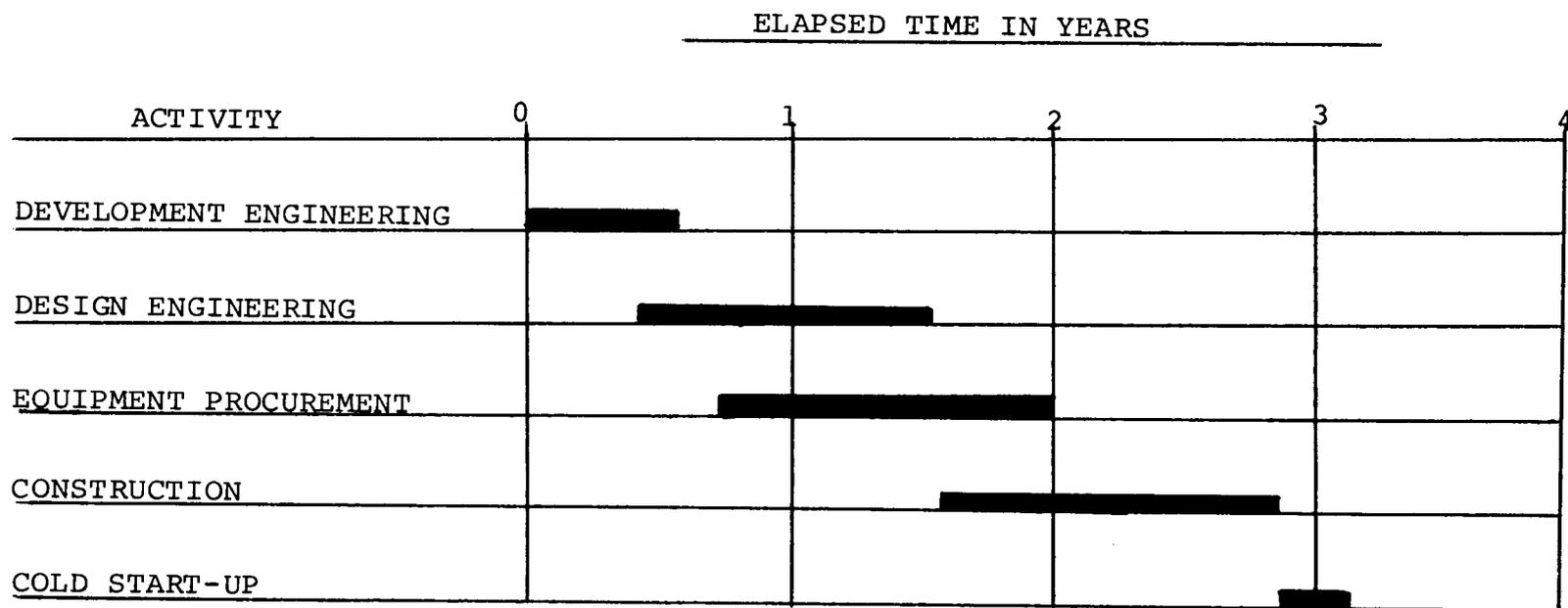


Fig. 4. Schedule for Krypton Recovery Facility.

#### IV TRITIUM RETENTION

##### Review of Tritium Recovery Methods

There are only a few processes which have been considered for the separation and retention of tritium from the off-gas stream from a fuel reprocessing plant. Tritium is a product of ternary fission with a yield of approximately  $10^{-4}$ . Thus it will be extremely dilute in the normal in-plant gas streams so that recovery and concentration will be difficult.

In a reprocessing plant based on the Purex process, the bulk of the tritium is converted to tritiated water during dissolution of the fuel. At the BNFP, the concentration of tritium in the solution from the dissolver will be about 2 ppm.

In theory, light isotope separation processes based on isotopic exchange reactions could be applied to the separation and recovery of tritium. For example, in the electrolysis process for heavy water production tritium concentrates with the deuterium.<sup>14</sup> Thus the procedure for heavy water production could be utilized for tritium concentration.

There are certain metal hydrides in which the dissociation pressures of the hydrogen isotopes are different thus presenting a potential separation process. Metals which form a more stable tritide than hydride include vanadium, niobium and alloys of lanthanum-nickel and vanadium-niobium.<sup>15</sup> During formation of the hydride, the gas stream would be partially depleted in tritium. The gas resulting from dissociation of the hydride would be enriched in tritium. Thus by processing a gas stream containing the hydrogen isotopes through a series of hydriding-dehydriding steps, a gas stream enriched in tritium could be produced.

The light isotope separation methods although theoretically possible are not practical. For example, over 1 tonne per hour of water is vaporized in the waste concentrator. The requirements to electrolyze this quantity of water to obtain the mixture of hydrogen isotopes for separation is impressive. Further, the number of separation stages required would also be very large, in particular for the hydriding-dehydriding process.

Total retention of process water has been considered as a method for preventing the release of tritium from a reprocessing plant. An integral part of such a scheme would be extensive recycle of water and nitric acid in order to reduce volumes. Nitrogen oxides would be absorbed and converted to nitric acid for reuse. Extraneous water addition through use of steam jets would have to be reduced or eliminated. It was estimated that a potential reduction of water storage to 25 gallons per tonne was possible.<sup>16</sup>

The on-site retention of water does not appear feasible for a plant such as the BNFP. Water usage can be reduced by extensive recycle; however, some tritiated water will be lost as vapor through the vessel off-gas system from air lifts and other sources. Eventually the concentration of tritium in the recycle water would build up to some equilibrium value so that the discharged tritiated water vapor would be equal to the plant input.

A different approach to total retention has been outlined by ORNL workers.<sup>17</sup> Water input to the plant would be held to a very low quantity by extensive recycle of process water including use of recycle water for chemical make up. A new concept of sealed cells with low ventilation flow to limit water vapor introduced with the ventilation air would be required. The off-gas would be cooled to  $-140^{\circ}\text{F}$  to recover water vapor. It was estimated that total water input for a 5 MTU (U + Pu) plant could be limited to 60 Kg per day which would represent the daily storage requirement.

The technology to support the concept of low air flows, sealed cells and low water input is not yet developed. The aim of this concept is more in support of the LMFBR program. Adaptation to a plant such as the BNFP would not be practical.

One possible approach to tritium recovery is evolution of the tritium into a small stream of diluent gas followed by separation of the tritium from the diluent gas. ORNL is currently developing a process based on this principle called voloxidation.<sup>18</sup> This is a head-end process in which the fuel is sheared into short lengths in an enclosed shear in order to retain any tritium released from the gas plenum. The sheared fuel is then heated in an enclosed furnace in an oxidizing atmosphere. The  $\text{UO}_2$  in the fuel is converted to  $\text{U}_3\text{O}_8$  and the resulting phase change pulverizes the fuel so that the fission gases are released. The tritium (probably as tritiated water vapor) is contained in a sufficiently small volume that trapping from the gas phase appears to be technically feasible.

Dube and co-workers<sup>19</sup> reported on apparatus for removal of tritium from a gas stream. In their experiments, an air stream containing 0.5% tritium was passed through a catalytic oxidizer to convert tritium to water which was then adsorbed on molecular sieves. A decontamination factor of  $2.5 \times 10^5$  was obtained for tritium.

Most of the ORNL voloxidation work has been in support of LMFBR reprocessing. However, some work is applicable to LWR fuel. A Zircaloy clad  $UO_2$  fuel rod from Shippingport which had been irradiated to 32,000 MWd/tonne was sheared and then oxidized in moist oxygen for four hours at 450°C.<sup>20</sup> The oxidized fuel was then dissolved in nitric acid. Analysis of the dissolver solution showed that less than 0.1% of the initial tritium was retained in the fuel. The overall tritium balance was poor due to condensation of water in the off-gas lines. However, previous work had shown that tritium in irradiated  $UO_2$  appeared in the dissolver solution. Therefore, they feel that over 99.9% of the tritium was released by voloxidation in this experiment.

Another experiment in which stainless steel clad mixed oxide irradiated to 20,000 MWd/tonne was sheared and oxidized at 450 to 750°C showed release of essentially all the tritium.<sup>21</sup> Some other work on LMFBR fuel compositions has shown poorer release of tritium. This may be due to the higher plutonium content which inhibits phase change and crumbling resulting from the phase change.<sup>22</sup>

At the present stage of technology, voloxidation appears to be the only process which will fulfill the requirement of releasing tritium into a small gas volume so that trapping is feasible. Therefore, the requirements for adaptation of the voloxidation process to the BNFP were studied in more detail.

#### Impact on the BNFP

The equipment arrangement in the remote process cell which contains the shear and dissolver is not compatible with the addition of a voloxidation furnace. Additional process equipment is also required for the overall process. Thus an entire new head-end facility would be required which is shown in outline in Figure 1.

Major modifications to the Separations Facility would be required to retrofit the head-end facility into the plant. The system which transports the fuel to the shear would have to be extended to the new head-end facility. A shielded tunnel from the remote process cell would be required for the transfer system. A second tunnel would be required to transfer the leached hulls to the hull monitor station and to the hull disposal facilities.

Transfer piping from the dissolver system to the main reprocessing line would be added as well as connecting piping to the off-gas system.

As can be visualized, these changes would require decontamination and removal of some equipment from the remote process cell and decontamination of the remaining equipment in the cell so that construction could proceed. The cell wall would be opened for access to the transfer tunnels. Even with optimum scheduling of construction activities, an extended plant shutdown would be necessary.

A further technical problem which could have serious impact on the BNFP lies in the behavior of plutonium. In some LMFBR studies, plutonium in fuel from the voloxidation process has required fluoride for complete dissolution.<sup>22</sup> If, when processing plutonium recycle fuel, the plutonium was not immediately soluble in nitric acid a severe problem would arise in that the stainless steel equipment is not compatible with fluoride.

#### Voloxidation Flowsheet

The flowsheet for the voloxidation head-end process is shown in Figure 5. The flow rates for the various streams follow Figure 5. The equipment layout is shown in Figure 6.

When scheduled for reprocessing, the fuel is transferred from the fuel storage pool into the head-end cell. The fuel element will be allowed to dry by self-heating to minimize addition of water to the off-gas system. The element will then be moved into the shear enclosure, the enclosure sealed and the fuel chopped into short segments. During chopping, a stream of dry air will carry any fission gas released from the plenum into the tritium off-gas recovery system.

The sheared fuel drops by gravity into the voloxidation furnace. The voloxidation furnace will operate between 450 and 750°C. In this temperature range the  $UO_2$  is converted to  $U_3O_8$ . The hold-up time in the furnace is approximately three hours. The contained tritium should also be converted to tritiated water in the furnace. A sweep of dry air enters at the exit end of the furnace and joins the sweep air from the shear. This sweep prevents the loss of tritium to the dissolver system.

The oxidized fuel falls into the diverter and then to the dissolver for reprocessing through the regular process line. Any tritium remaining in the fuel at this point will eventually be discharged through the main plant stack. The combined gas stream from the oxidation furnace passes through a cyclone

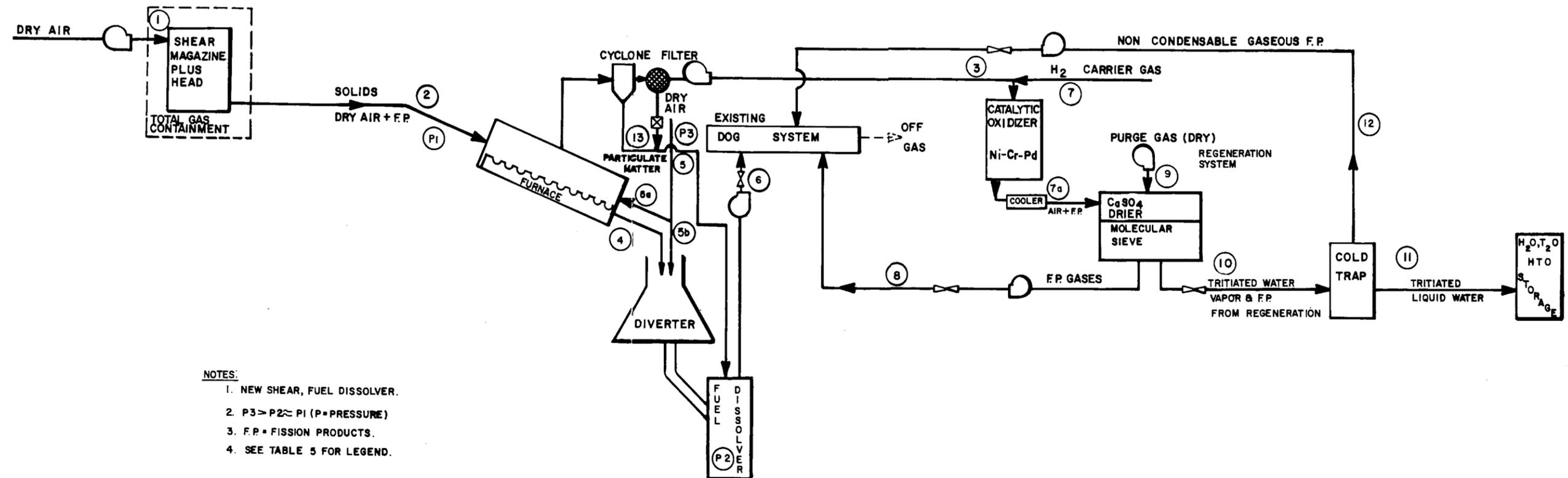


Fig. 5. Conceptual Voloxidation Head-End Process.

<u>Line</u>							
1	Condition for flow	During Chopping - Dissolution					
3	Stream No.	1	2	3	4	5	5a
5	Flow rate scfh	7500	7500	14,500	-	36,000	7,000
6	lb mole/min	0.35	0.35	0.67	-	1.67	.033
8	Temp °F	~ 150	~ 150	~ 850	-	~ 100	~ 100
10	Pressure psia	14.8	14.8	14.4	-	14.8	14.8
	component flow						
14	Air, lb-mole/min	0.35	0.35	0.67	-	1.67	0.33
16	H <sub>2</sub> O, lb/hr	-	-	P	-	-	-
18	Kr	-	P <sup>a</sup>	-	-	-	-
20	T gm/hr	-	P	0.008	-	-	-
22	Other	-	-	-	U <sub>3</sub> O <sub>8</sub> + Hulls	-	-

a) P = Present in stream, amount not known

<u>Line</u>	During Dissolution					During Regeneration				13 <sup>b</sup>
3	5b	6	7	7a	8	9	10	11	12	
5	29,000	29,0000		14,500	14,500	2,000	2,000		2,000	
6	1.34	1.34		0.67	0.67	.09	.09		.09	
8	~ 100	150		70	70	80	220	40	- 50	
10	14.8	14.4	14.7		14.4	14.8	14.6		14.4	
14	1.34	1.34		0.67	0.67					
16				.32			22	X		
18		P		P	P		P		P	
20				0.008			.53			
22				H <sub>2</sub> , 16 gm/hr						U <sub>3</sub> O <sub>8</sub> Particulate

b = intermittent

X = undetermined

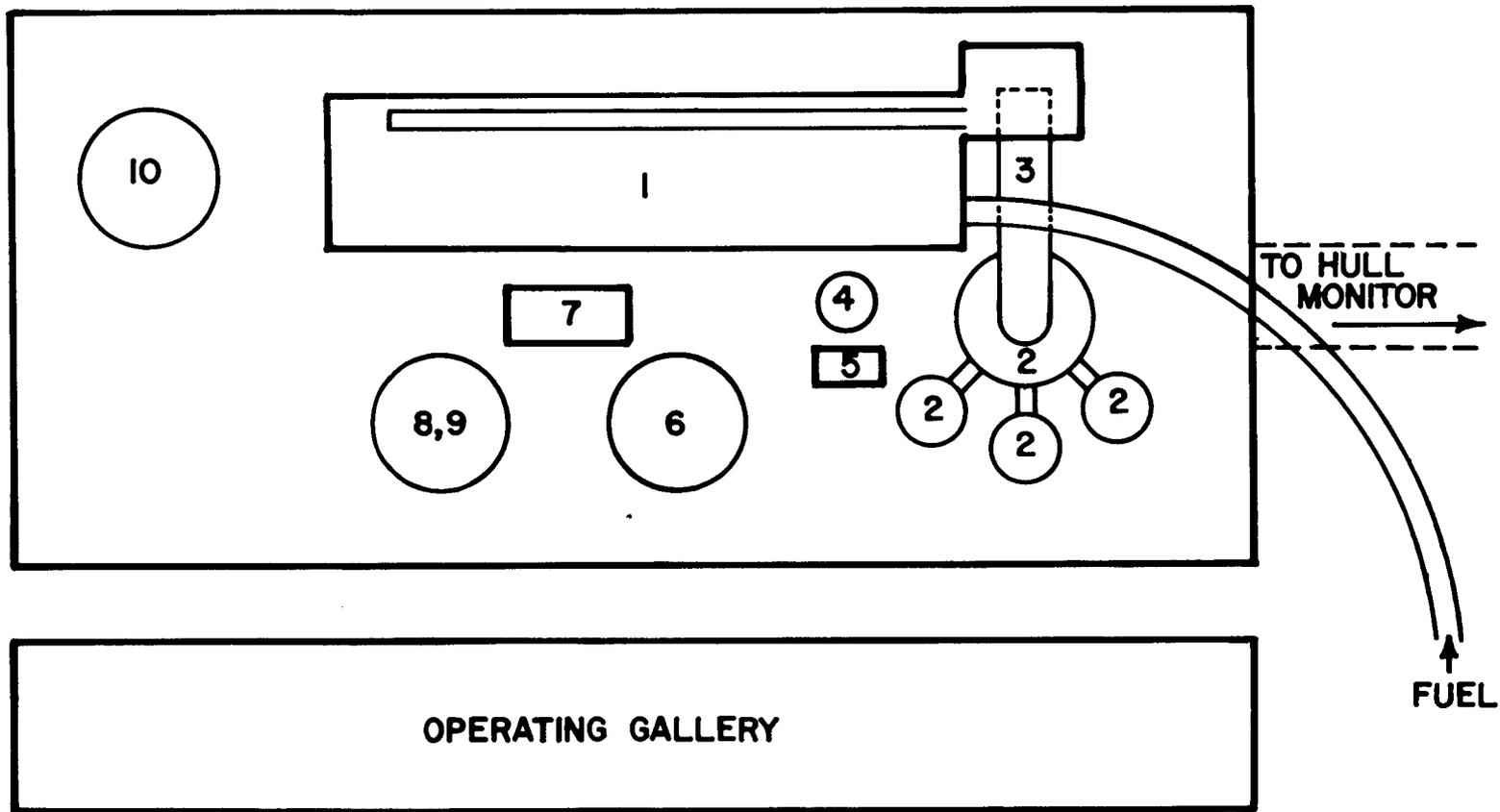


Fig. 6. Equipment Layout for Tritium Recovery.

and sintered metal filter in series to remove any particulates. The particulates could be collected in secondary containers beneath the cyclone and filter (not shown on the flowsheet). Prior to the next shearing cycle, the fines could be transferred to a dry dissolver by a rotary feeder and vibrator on the secondary container. This technique would prevent introduction of additional water vapor from the dissolver into the tritium off-gas system.

The filtered gas stream is passed into the catalytic oxidizer where any tritium gas reacts with oxygen in the presence of Ni-Cr-Pd ribbon catalyst to produce tritiated water. Hydrogen is also bled into the catalytic oxidizer to form water as a carrier for the tritiated water. The catalyst bed operates at 400 to 600°C.

Following the catalytic oxidizer, the gas stream is cooled to ambient temperature and passed through a bed of  $\text{CaSO}_4$  and then through a bed of molecular sieves to adsorb the water vapor from the gas stream. The combination of these two adsorbers in series should remove over 99.9% of the water vapor. Most of the krypton and any iodine in the gas stream should pass through the adsorber beds. The gas stream is then routed to the existing off-gas system for further processing.

The adsorbed water will be removed from the adsorbers on a bi-weekly schedule. The beds will be heated to 200 to 300°C and a stream of dry air passed through the beds to remove the water as vapor. The loaded gas stream will then be passed through a cold trap which is cooled with R-12 to approximately -20°C to freeze the water. The trap will subsequently be warmed and the tritiated water drained into a storage container. The type of storage container and the method of storage have not been specified at this time. Approximately 13 gallons of water would be generated in a two-week period, the bulk of which arises from the hydrogen bleed to the catalytic oxidizer.

The noncondensibles in the gas stream to the cold trap are vented to the Separations Facility off-gas system.

#### Cost Estimate and Schedule

The process described above, the modifications to the plant, and the equipment requirements form the basis for preparation of the cost estimate. The major equipment items are listed in Table 5. The cost estimate is given in Table 6. The cost is estimated at \$34,000,000 in 1973 dollars, again with no provision for escalation of costs. Much of the cost data are based on experience factors gained during the design and construction of the BNFP. On the other hand, the estimate of some of the equipment such as the voloxidation furnace are

much less reliable. As was the case for krypton recovery, no provisions were made for packaging, storing, and managing the tritiated water. Operational costs of the unit also are not included.

The schedule for adaptation of voloxidation head-end unit to the BNFP is shown as a bar chart in Figure 7. As can be seen, the overall schedule from project initiation through cold checkout is estimated to be 4.5 years. In comparison with the krypton recovery process, the basic flowsheet is simpler; however, the overall facility is more complex, hence the longer schedule.

Table 5

MAJOR EQUIPMENT LIST

TRITIUM RECOVERY SYSTEM

1. Shear - duplicate of existing shear at BNFP, enclosed for total gas containment.
2. Dissolver - duplicate of existing dissolver system at BNFP.
3. Voloxidation furnace - 3.5 feet diameter by 7 feet long, electrically heated to 750°C, duty ~ 150 KW, rotation speed 0-20 rpm, rotary seals for gas containment.
4. Cyclone - 1 foot diameter, 304 SS to process ~ 850 acfm air, periodic powder discharge.
5. Filter - 3 feet diameter by 4 feet long, sintered stainless steel filter elements ~ 850 acfm air, automatic blow back on filter elements, periodic powder discharge.
6. Catalytic oxidizer - 2 feet diameter by 5 feet long packed with Ni-Cr-Pd catalyst, reheat duty to 600°C if required by electrical heating jacket.
7. Cooler - fin-tube heat exchanger with brine inside tubes, capacity ~  $3 \times 10^5$  Btu/hr.
8. CaSO<sub>4</sub> Adsorber Bed - 2 feet diameter by 6 feet long, electrical heating for regeneration, duty ~ 5 KW.
9. Molecular Sieve Adsorber - 2 feet diameter by 6 feet long, electrical heating for regeneration, duty ~ 3 KW.
10. Cold Trap - 1.5 feet diameter by 3 feet long, finned tube refrigeration cooled to -20°C, heat duty ~  $4 \times 10^4$  Btu/h

Table 6

ORDER OF MAGNITUDE ESTIMATETRITIUM REMOVAL(VOLOXIDATION HEAD-END PROCESS)Major Equipment

Shear	\$1,400,000
Diverter	390,000
Dissolver System	595,000
Fuel & Hull Transfer Systems	881,000
Shielding Windows & Doors	231,000
Process Equipment	523,000
Cranes, Viewing Equip., Spec. Equip.	422,000
Hatches, Utility & Maintenance Equip.	<u>358,000</u>
Subtotal Major Equip.	\$4,800,000

Other Directs

Decon Cell, Remove & Dispose Exist. Equip.	450,000
Install New Equipment	580,000
Piping & Ventilation System	4,300,000
Electrical & Instrumentation	1,920,000
Steel, Paint & Insulation	700,000
Process Bldg. Additions & Modifications	2,150,000
Site Improvements	<u>500,000</u>
Subtotal Other Directs	<u>\$10,600,000</u>
Total Direct Cost	\$15,400,000

Indirects

Design Engineering	4,800,000
Construction Indirects & Fee	6,800,000
Home Office & Associated Capital Costs	<u>1,400,000</u>
Total Indirect Cost	\$13,000,000
Total Before Contingency	<u>\$28,400,000</u>
Contingency (20%)	<u>5,600,000</u>
TOTAL PROJECT COST	\$34,000,000*

- \*(1) Excludes Cost of Land
- (2) Estimate Based on Construction Complete in 1973
- (3) Excludes Cost of Final Waste Storage
- (4) Confidence Level of Estimate; -15%, +20%

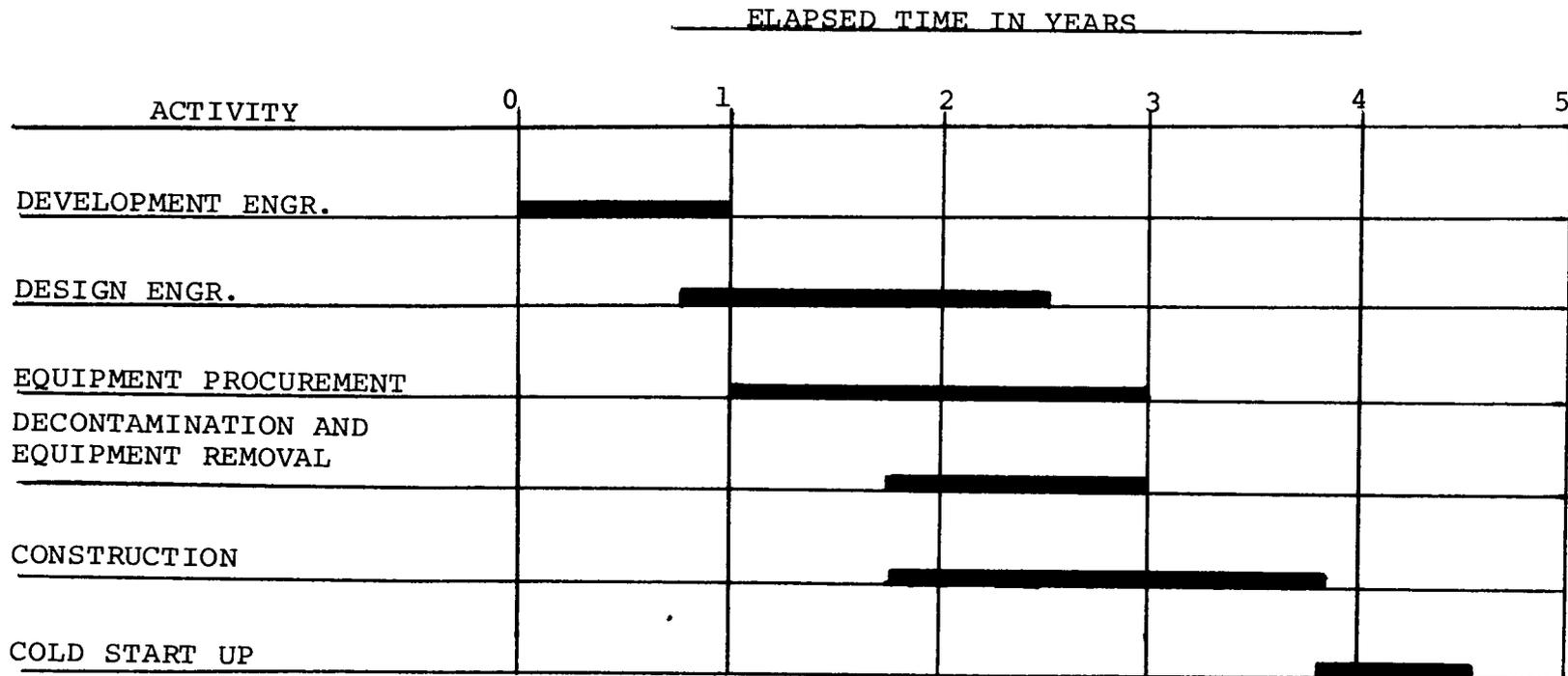


Fig. 7. Schedule for Voloxidation Head-End Facility.

## V IODINE REMOVAL

The removal of iodine from off-gas streams has been one of the important auxiliary functions at fuel reprocessing plants. In recent years with the added emphasis on environmental protection, the collection and retention of iodine has received additional attention. At the BNFP, fuel will be reprocessed with a minimum of 160 days cooling. Thus, the  $^{131}\text{I}$  will have decayed through many half lives. Even so, the fuel will contain 1.3 Ci of  $^{131}\text{I}$  per MTU as well as  $219 \times 10^{-2}$  Ci of the long-lived isotope  $^{129}\text{I}$ .<sup>1</sup> Therefore, off-gas treatment is required to reduce the release of iodine to the environs to "as low as practicable".

As a part of the work under the sub-contract, the retention of iodine was to be reviewed, however, it was agreed early in the program that adaptation of advanced iodine retention methods to the BNFP would not be included.<sup>23</sup>

Early systems for the removal of iodine from the dissolver off-gas included silver reactors, charcoal beds and caustic scrubbers.<sup>24-26</sup> The early systems were less efficient than desired so development effort has continued under the support of the AEC.

One of the important developments has been the use of a silver exchanged zeolite for adsorbing iodine species from a gas stream.<sup>27-29</sup> Silver zeolite is prepared by treating a natural or synthetic zeolite with silver nitrate. Silver exchanges with sodium in the zeolite structure. The resultant material is an excellent adsorber for both organic and inorganic iodine species. A fully exchanged silver zeolite will adsorb 0.09 gm of iodine per cc of bed up to 1% breakthrough with a DF of at least 100.

Mercuric nitrate scrubbing of off-gas streams has been shown to be an effective method for retaining iodine.<sup>30</sup> In mercuric nitrate solution which is about 8 molar in nitric acid, iodine species both inorganic and organic are complexed by mercuric ion. A mercuric nitrate scrubber column is expected to have a minimum DF of 10.<sup>31,32</sup>

The advanced iodine retention method under development at ORNL is the Iodox process.<sup>33,34</sup> In this process, the off-gas stream is contacted with refluxing 16-20M nitric acid. The iodine is oxidized to iodic acid which precipitates and collects in the bottoms of the fractionating column. Experimental results have shown the process to be effective for removal of iodine species from an off-gas stream. In a six stage bubble cap column, DFs ranged from 30 in 17M acid to  $9 \times 10^5$  in 20M acid.<sup>35</sup> As mentioned earlier, LMFBR

fuels may be processed after 30 days cooling. The  $^{131}\text{I}$  activity would be orders of magnitude higher ( $\sim 12$ ) so a high decontamination process would be required.

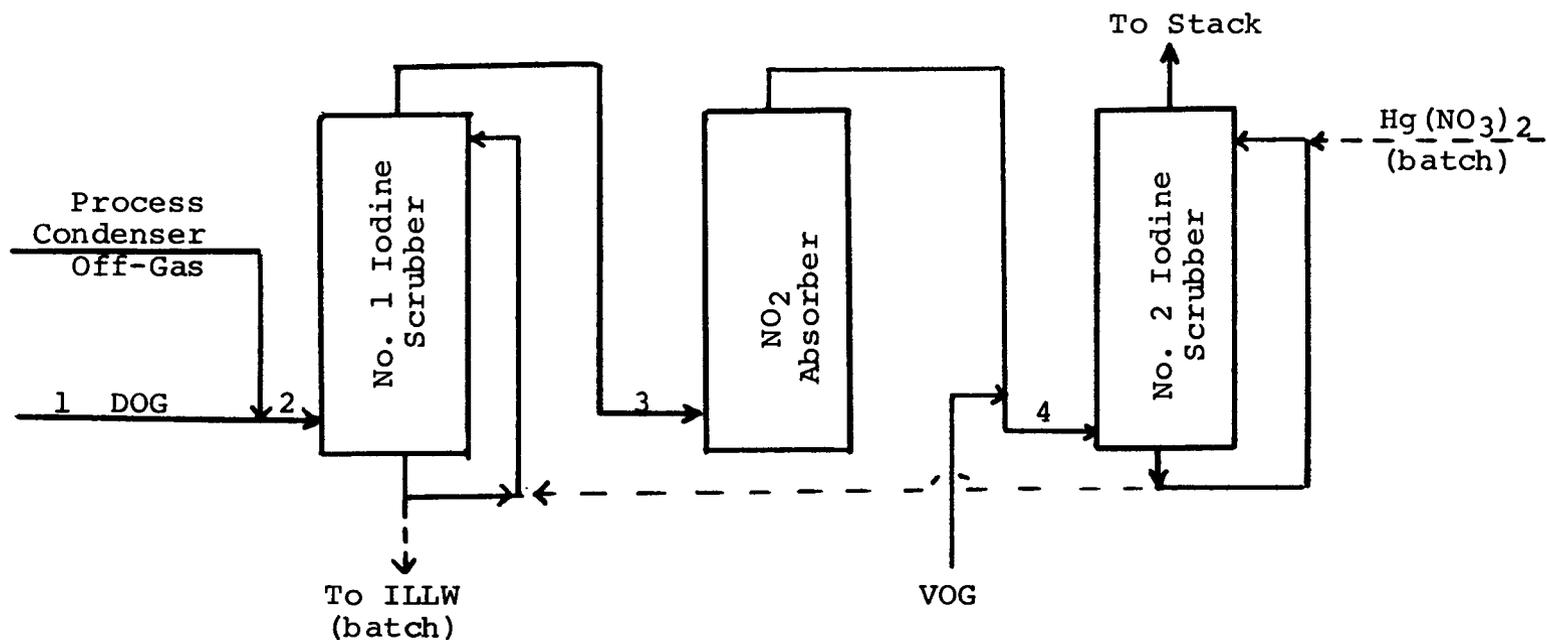
The off-gas treatment process at the BNFP includes two stages of mercuric nitrate absorption followed by a silver zeolite adsorber. The dissolver off-gas and the noncondensibles from certain evaporators are passed through the #1 iodine absorber in which the bulk of the iodine is reacted with 0.2M mercuric nitrate in 6-8M nitric acid solution. This off-gas stream is then combined with the off-gas from the vessel off-gas system and passed through the #2 iodine absorber. Additional iodine is removed by reaction with mercuric nitrate in this absorber. Each absorber is expected to have a minimum of DF of  $10^{30-32}$ .

The mercuric nitrate scrubbing flowsheet is shown in Figure 8. The initial charge of scrubbing solution is fed to the #2 scrubber. Periodically the #1 scrubber is drained to the Intermediate Level Liquid Waste (ILLW) system. Scrubbing solution is transferred from the #2 scrubber to the #1 scrubber and makeup solution is then added to the #2 scrubber.

The scrubbed off-gas is finally passed through silver zeolite adsorbers. This adsorber is expected to yield an additional DF of 100 as that the overall DF for iodine in the off-gas system is at least  $10^4$ . It has been suggested that a DF of  $10^3$  is required for 150 day cooled LWR fuel<sup>36</sup>, so  $10^4$  is more than adequate.

During this study, the path of iodine through the liquid streams was reviewed. Iodine which is not volatilized during dissolution will follow the process streams through extraction, vaporization in concentrators and evaporators and could conceivably be vaporized with the discharged water vapor. This stream enters the off-gas system beyond the silver zeolite beds so that any contained iodine will not be adsorbed. Thus, overall iodine DF could be limited by this stream.

Two process additions were identified which will alleviate this potential iodine problem.<sup>37-39</sup> These are the addition of a macroreticular resin column for treatment of the liquid stream before it reaches the acid fractionator overheads vaporizer and the addition of mercuric nitrate to the vaporizer bottoms. The resin will remove iodine from the liquid stream and the mercuric nitrate will retain any residual iodine which reaches the vaporizer. These additions are expected to improve the overall iodine retention so that the DF for the entire process will be at least  $10^4$ .



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Stream No.	lb-moles/hr	acfm	Temp. °F	Mole %					
				NO	NO <sub>2</sub>	N <sub>2</sub> O	N <sub>2</sub>	O <sub>2</sub>	H <sub>2</sub> O
1	64.4	478	106	2.72	1.52	-	69.2	18.4	8.1
2	91.22	675	102	2.17	1.11	0.52	69.8	18.5	7.7
3	95.3	713	100	2.08	1.06	0.49	70.2	18.6	7.5
4	732	5530	110	0.07	0.01	0.06	72.5	18.6	8.1

Fig. 8. Flowsheet for Iodine Removal.

The decision not to evaluate the Iodex process for adaptation to the BNFP should not be construed as an indication that the process is not workable. When fully developed, the process should be adaptable to a commercial fuel reprocessing plant. However, it was felt that the proven technology applied to the BNFP was adequate for present requirements. From the cost standpoint, the nitric acid bubble cap column should compare favorably with mercuric nitrate absorbers. Development effort on the Iodex process should be continued.

## VI CONCLUSIONS

Assuming that pilot plant development is completed satisfactorily, the fluorocarbon absorption process for krypton recovery should be adaptable to a plant such as the BNFP. The cost estimate for adaptation of the process to the BNFP is  $\$5 \times 10^6$ . However, the cost estimate does not include  $N_2O$  removal or storage of the recovered product. The cost estimate also does not include operation of the facility. As shown in Table 2,  $N_2O$  is sufficiently soluble in R-12 so that the overhead from the stripper would contain about 67%  $N_2O$ . Either the  $N_2O$  must be separated ahead of the absorption column or another process step would be required to further purify the product gas. Otherwise, storage requirements for the gas would become excessive. Also further separation of the R-12 from the product should be accomplished. Radiation decomposition of R-12 could lead to corrosion of storage cylinders. These steps should be considered in development studies.

Beyond the actual recovery of krypton lies the problem of storage. The accumulated  $^{85}Kr$  at the end of 30 years operation of the BNFP will be approximately (180) megacuries. It should be assumed that a Federal Repository will become operational during this period, but until such time, the krypton would have to be stored on site in a safe manner. The method of storage and the associated costs have not been evaluated. 1507

It does not appear that there are any other major technical problems associated with the fluorocarbon absorption process. The key steps in the process, absorption, fractionation and stripping have been demonstrated in the pilot plant program.

As pointed out in the Introduction, off-site dose rate from krypton release from the BNFP is very small. Future krypton burden to the atmosphere from reactors and reprocessing facilities may create the desirability for krypton recovery to control releases. Therefore, the development programs should be continued.

The voloxidation process does not appear to be sufficiently developed for adaptation to a plant such as the BNFP. The physical modifications to the facility would be rather extensive. Breaking into a process cell is a formidable task. Extensive decontamination procedures would be required in order to open the cell. Probably at least a year of downtime for the Separations Facility would be involved while coupling the new head end to the existing plant.

Theoretically the voloxidation process is quite simple. However, in the Atomics International study of oxidation-reduction reprocessing of  $UO_2$  fuels, during each cycle additional radioactive gas was released.<sup>40</sup> This indicates that further particle size reduction may be necessary to release the fission gas. Further development work should be completed to demonstrate the reliable release of tritium by the voloxidation process.

In spite of the potential technical problems and incomplete status of the development effort, the voloxidation process appears to be the only method applicable to a fuel reprocessing plant. Therefore, development effort on the voloxidation process should be continued.

The Iodox process could prove to be an excellent method for the removal of iodine from the dissolver off-gas stream. Coupled with iodine removal processes from the dissolver, good decontamination factors are possible. However, it is believed silver zeolite adsorber beds should also be included in any iodine treatment scheme to take advantage of the excellent retention factors demonstrated.

## VII RECOMMENDATIONS

During the course of this study, several areas were identified where further development effort is required to demonstrate the technical and engineering feasibility.

### 1) Plutonium Solubility in Nitric Acid

Dissolution studies on mixed plutonium-uranium oxide have shown that plutonium oxide will dissolve in nitric acid if the oxides are in solid solution.<sup>18</sup> However, if a second plutonium oxide rich phase is present, the dissolution rate of this phase in nitric acid is slow and fluoride ion is required for complete dissolution.

Studies on dissolution of LMFBR fuels reported by ORNL<sup>18</sup> have shown little effect of voloxidation on the dissolution rate in nitric acid when compared to dissolution of  $UO_2 - PuO_2$ . However, these tests were run with relatively high  $PuO_2$  concentration. Assuming plutonium recycle in LWRs at some future time, studies on mixed oxides containing perhaps 3-5%  $PuO_2$  should be carried out. Here a small amount of undissolved  $PuO_2$  would represent a much larger percent change in overall  $PuO_2$  solubility and could have serious effects on the reprocessing cycle.

### 2) Behavior of Zircaloy Clad Fuel

A significant fraction of LWR fuel will be clad in Zircaloy. Most of the voloxidation development studies have been carried out with stainless steel clad fuel. Therefore, similar studies should be carried out with Zircaloy clad material to determine the behavior of tritium during shearing, oxidation, and dissolution. Also, the behavior of any zirconium bearing fines generated during shearing should be determined. If some zirconium fines were to oxidize during the voloxidation step, the heat of reaction could be sufficient to cause agglomeration with some uranium oxide and to partially sinter the mixture to yield a difficultly soluble phase.

### 3) Voloxidizer

The engineering development of a reliable voloxidation furnace should be pursued. This is the key piece of equipment in the head-end process, therefore it must operate reliably for extended periods of time. One critical item on the voloxidizer is the rotary seal arrangement. A large in-leakage of air must be avoided as this would introduce additional water into the tritium retention system. Accelerated life tests of candidate seals should be carried out.

#### 4) Tritiated Water Storage

Interim on-site storage and management of tritiated water should be considered. This effort should include accident analysis and economic analysis for different schemes. Assuming that tritiated water would eventually be solidified for off-site storage, solidification methods should be developed. The obvious thought is to cast into concrete. Incorporating into a salt such as  $\text{Na}_2\text{CO}_3 \cdot 10\text{H}_2\text{O}$  and sealing in mild steel containers should also be considered.

#### 5) $\text{N}_2\text{O}$ Removal in Krypton Process

A procedure should be developed for removal of  $\text{N}_2\text{O}$  prior to the fluorocarbon absorption step. Catalytic conversion to  $\text{NO}_2$  prior to the sorption of residual  $\text{NO}_2$  on the molecular sieve is a possibility. Removal of  $\text{N}_2\text{O}$  will markedly reduce the volume of krypton storage.

#### 6) Krypton Storage

Evaluation of krypton storage methods and determination of interim storage requirements should be completed. This study should include low pressure vs. high pressure storage and the effect of xenon removal on storage requirements. Consideration should also be given to ultimate disposal requirements of retained krypton.

#### 7) Iodine Retention

Development effort on the Iodex process should be continued. The removal of the precipitated iodine compound from nitric acid solution and the storage of the iodine compound require study.

The treatment of mercuric nitrate scrubber solutions for either mercury recovery or iodine removal could simplify subsequent solidification of stored waste. Therefore, some development work is needed in this area.

Additional work on removal of iodine from aqueous streams by macroreticular resins is needed as this treatment method will increase the overall plant DF for iodine.

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APPENDIXSafety Analysis Outline

The Final Safety Analysis Report (FSAR), Docket No. 50332, for the Separations Facility has been submitted to the Atomic Energy Commission. Should additional facilities such as krypton recovery or tritium recovery be required, an appropriate Safety Analysis report will be submitted.

An outline of a projected Safety Analysis Report is presented below. A more detailed discussion of potential accidents and abnormal events which might arise during operation of these facilities is included following the outline.

CHAPTER 1.0 INTRODUCTION AND GENERAL DESCRIPTION OF PLANT

The first chapter of the Safety Analysis Report should present an introduction and general plant description. This chapter should enable the reader to obtain an overall understanding of the facility without having to delve into the subsequent chapters. Review of the detailed chapters which follow can then be accomplished with better perspective and with recognition to the relative safety importance of each individual item to the overall facility design.

CHAPTER 2.0 SITE CHARACTERISTICS

This chapter should provide information on the geological, seismological, hydrological, and meteorological characteristics of the site and vicinity, in conjunction with population distribution, land use, and site activities and controls. The purpose is to indicate how these site characteristics have influenced plant design and operating criteria and to show the adequacy of the site characteristics from a safety viewpoint.

CHAPTER 3.0 DESIGN CRITERIA - STRUCTURES, COMPONENTS, EQUIPMENT AND SYSTEMS

This chapter of the Safety Analysis Report should identify, describe, and discuss the principal architectural and engineering design criteria that represent the broad frame of reference within which the more detailed design effort of those structures, components, equipment, and systems important to safety is to proceed and against which attainment of the design objective will be judged.

Where the need arises in other chapters of the SAR to refer to design criteria included in this section, only cross reference is necessary.

#### CHAPTER 4.0 PROCESS SYSTEMS

This chapter should include a description of the process systems including mechanical processes, chemical processes, and the process support systems. Emphasis should be on safety related aspects of the systems.

#### CHAPTER 5.0 PLANT FACILITIES

This chapter should include a functional description of the facility and a discussion of the facility components and operation. Safety criteria and assurance should be outlined for the facility.

#### CHAPTER 6.0 ENGINEERED SAFETY FEATURES

Engineered safety features are provided to mitigate the consequences of postulated serious accidents, in spite of the fact that these accidents are very unlikely. This chapter of the SAR should present information on the engineered safety features provided in the proposed plant. The information provided should be directed primarily toward showing that:

- (1) the concept upon which the operation of the system is predicated has been, or will be, proven sufficiently by experience, tests under simulated accident conditions, or conservative extrapolations from present knowledge;
- (2) the system will function during the period required and will actually accomplish its intended purpose.

#### CHAPTER 7.0 INSTRUMENTATION AND CONTROLS

The information provided in this chapter should emphasize those instruments and associated equipment which constitute the protection system (as defined in IEEE Std 279-1971 "IEEE Standard: Criteria for Protection Systems for Nuclear Power Generating Stations"). The discussion of regulating systems and instrumentation should be limited to considerations of regulating system-induced transients which, if not terminated in a timely manner, would result in radiation release or other public hazard. Details of seismic design and testing should be provided in Section 3.9.

#### CHAPTER 8.0 WASTE MANAGEMENT

The purpose of the information to be provided in this chapter is to provide assurance that the nuclear plant has sufficient

installed capacity and treatment equipment in the radioactive waste (radwaste) systems to reduce the radioactivity to levels which will not be in excess of the appropriate limits for the general public or plant personnel and are as low as practicable. Wherever appropriate, summary tables should be provided.

#### CHAPTER 9.0 WASTE COOLING SYSTEM

Requirements for cooling the stored waste products should be determined. It is expected that these requirements will not be elaborate, hence a system to insure an adequate and uninterrupted supply of coolant should suffice.

#### CHAPTER 10.0 ELECTRIC POWER

The electric power system is the source of power for the process and auxiliaries during normal operation, and for the protection system and engineered safety features during abnormal and accident conditions. The information in this chapter should be directed toward establishing the functional adequacy of the emergency power sources, and assuring that these sources are redundant, independent, testable and otherwise in conformity with current criteria. Details of seismic design and testing should be provided in Section 3.9.

#### CHAPTER 11.0 AUXILIARY UTILITY SYSTEMS

This chapter of the Safety Analysis Report should provide information concerning the auxiliary systems included in the facility, some of which are required to perform certain functions during accident or emergency conditions.

#### CHAPTER 12.0 RADIATION PROTECTION

The purpose of the information to be provided in this chapter is to permit a determination that direct radiation exposures to persons at the site boundary from sources contained within the plant and on the site, and external and internal exposures to plant personnel will be kept as low as practicable and within applicable limits.

#### CHAPTER 13.0 CONDUCT OF OPERATIONS

This chapter of the Safety Analysis Report should provide information relating to the framework within which operation of the facility will be conducted.

The operation of the facility entails a myriad of instructions and procedures of varying detail for the operating staff. The details of such procedures should not be included in the Safety Analysis Report, but information should

be provided to indicate generally how the applicant intends to conduct operations, and to assure that the licensee will maintain a technically competent and safety-oriented staff.

#### CHAPTER 14.0 INITIAL TESTS AND OPERATION

This chapter should provide the information relating to the period of initial operation, with particular emphasis on tests planned to demonstrate the degree to which the facility does, in fact, meet the design criteria. Explanations for any special limits, conditions, surveillance requirements, and procedures to be in force during the initial period of operation and until such time as acceptable design performance is demonstrated should be included.

#### CHAPTER 15.0 ACCIDENT ANALYSES

The evaluation of the safety of a reprocessing plant is accomplished, in part, by studies made of the response of the plant to disturbances in process variables and to postulated malfunctions or failures of equipment. Such analyses provide a significant contribution in the selection of the design specifications for components and systems and subsequently serve importantly in showing that a design consistent with public safety has been achieved. These analyses are a focal point of the Commission's construction permit and operating license reviews of reactor facilities.

#### CHAPTER 16.0 TECHNICAL SPECIFICATIONS

In accordance with the Atomic Energy Act and Section 50.36 of 10 CFR Part 50, each operating license issued by the Atomic Energy Commission must contain Technical Specifications that include those technical operating limits, conditions, and requirements imposed upon facility operation in the interest of the health and safety of the public. The applicant for an operating license proposes Technical Specifications and bases for his facility which are reviewed by the AEC regulatory staff and modified as necessary before becoming a part of the operating license.

#### CHAPTER 17.0 QUALITY ASSURANCE

In order to provide assurance that the design, construction, and operation of the proposed facility are in conformance with applicable regulatory requirements and with the design bases specified in the license application, it is necessary that a Quality Assurance Program (QAP) be established by the applicant. In this chapter, the applicant should provide a description of the QAP to be established and executed during the design and construction of the facility.

## SAFETY AND ACCIDENT ANALYSIS

Krypton RecoveryRupture of Full 20-Hour Storage Tank

The upper limit accident considered in this study is the rupture of the 20-hour storage tank in the krypton recovery facility. At capacity, the tank could contain  $4.5 \times 10^4$  Ci of krypton. The dose rate at the site boundary was calculated for the worst meteorological conditions. The assumption and results are as follows:

Assumptions

Total activity	$4.5 \times 10^4$ Ci Kr-85	
Tank volume	23,000 ft <sup>3</sup>	
Tank pressure	33 atm.	
Distance to site Boundary	2,000 M	
Night Inversion Conditions	Point source X/Q $\sim 2.6 \times 10^{-4}$ sec/m <sup>3</sup> Volume source X/Q $\sim 1.8 \times 10^{-4}$ sec/m <sup>3</sup>	
<u>Potential Dose at Site Boundary</u>	<u>Point Source (1)</u>	<u>Volume Source (2)</u>
Whole Body Gamma	8.3 mrem	5.7 mrem
Skin Beta	340 mrem	230 mrem

(1) Contents leak through a small hole in 30 minutes.

(2) Large rupture.

Failure of Molecular Sieve Unit

In this context, failure of the molecular sieve unit refers to cessation of proper functioning (as opposed to a mechanical break in the system, which is discussed below). In the event of such failure, H<sub>2</sub>O, CO<sub>2</sub>, and NO<sub>x</sub> would no longer be removed. The results of this are not completely understood, but the following comments are applicable.

Even under normal conditions, NO<sub>x</sub> removal is incomplete, in that N<sub>2</sub>O is unaffected. NO is oxidized to NO<sub>2</sub>, and it is the NO<sub>2</sub> that is absorbed. It is believed that even under conditions resulting in failure of the unit to adsorb gas, the NO would still be oxidized to NO<sub>2</sub>. Thus, the failure discussed would result in H<sub>2</sub>O, CO<sub>2</sub>, and NO<sub>2</sub> flow downstream of the unit.

The presence of the 13 standard liters per minute of CO<sub>2</sub> in the stream would reduce the efficiency of operations, but would have no other safety-related effect. On the other hand, the relatively large quantities of H<sub>2</sub>O and NO<sub>2</sub> would freeze and plug the system; the minimum result of this is system shutdown. A possible (although improbable) situation would be that freezing and deposition would be at a location and in a manner such that a component or pipe ruptures. At the level of "extremely unlikely but requiring further study" is the possibility of an explosion similar to the occasional unexplained explosion in cryogenic plants.

### Line Rupture

Depending on its location, a line rupture could release radioactive gas, refrigerant (R-12), or both. R-12 is nontoxic, but at high concentrations can irritate the lungs; it is very dense, so that in confined spaces it could exclude oxygen. The radioactive gas is only the off-gas that would be released through the stack in the absence of krypton recovery; the only additional hazard is the exposure to operators resulting from release inside the krypton recovery building. In case of rupture, valves would be reset to by-pass the recovery system (as is done when the dissolver is not operating).

### Loss of Temperature Control

Loss of Heating - Loss of heating ability (such as at a re-boiler) would necessitate shutdown, but there would be no hazard involved. The only effect would be the loss of decontamination of the off-gas stream.

Loss of Cooling - Loss of refrigeration would have the same results as the loss of heating, plus an increase in system pressure (to the vapor pressure of R-12) and the possible loss of R-12 to the off-gas stream. Some typical vapor pressures of R-12 are:

<u>Temperature, °F</u>	<u>Gage Pressure, psi</u>
- 20	0.6
0	9.2
32	30
50	47
75	77
90	100
100	117

### Tritium Recovery

#### Loss of Gas Containment Around Shear

The consequences of loss of gas containment around the shear

would be relatively minor. Only the fission gases in the plenum would be vented to the cell atmosphere. The ventilation air is discharged through the main plant stack. The amount of tritium and krypton released would be a small fraction of that released in the current design of the BNFP.

The worse condition would be if loss of containment occurred at the start of shearing an element. It would be necessary to complete the shearing operation simply to dispose of the element in order to carry out the required maintenance. The largest fuel element envisioned is about 0.8 MTU and if 10% of the fission gases were released to the cell atmosphere and subsequently discharged through the stack, the total release would be  $\sim 7 \times 10^2$  Ci of  $^{85}\text{Kr}$  and 6 Ci of  $^3\text{H}$ .

#### Loss of Containment Around Voloxidizer

Loss of containment in the voloxidation furnace would result in release of all the contained tritium and krypton to the cell atmosphere and subsequent discharge through the main stack. Assuming one MTU in the voloxidizer and loss of all fission gas, the release would be approximately  $9 \times 10^3$  Ci of  $^{85}\text{Kr}$  and 38 Ci of  $^3\text{H}$ . These are, of course, the design release rates for the BNFP with no fission gas retention.

#### Poisoned Catalyst

If the catalyst in the catalytic oxidizer became poisoned, the unreacted tritium would pass into the DOG system of the main plant and be discharged through the main stack. It is expected that the bulk of the tritium will be converted to tritiated water in the voloxidizer so that only traces of tritium would be lost to the DOG system.

The hydrogen carrier gas would not be oxidized; however, the hydrogen in the gas stream is only 0.4% which is well below the lower explosive limit for hydrogen-air mixtures. Therefore, no hazard would result from the unreacted hydrogen.

#### Loss of Cooling to Cold Trap

If cooling to the cold trap was lost at the start of the molecular sieve regeneration cycle, one week's accumulation of tritium could be vented through the main stack. The release would be approximately  $2.6 \times 10^4$  Ci. The off site dose from this release would be as follows:

##### Assumptions

Total Activity	$2.6 \times 10^4$ Ci H-3
Distance to Site Boundary	2000 meters



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