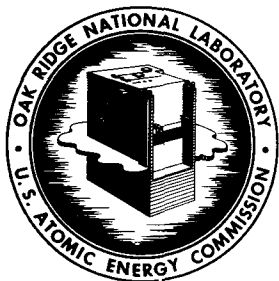


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DATE: December 7, 1960

SUBJECT: The Helium Purification System for the Proposed 800 MWT Pebble Bed Reactor

TO: A. P. Fraas

FROM: C. D. Scott and J. C. Suddath

COPY NO. 49

## ABSTRACT

A helium coolant purification system was designed for the proposed 800 MWT Pebble Bed Reactor. The purification system will operate on a coolant side stream with a flow rate 1% of the total coolant flow and there are provisions for radioactive and non-radioactive contamination removal.

Primary equipment components are dual oxidizers, an economizer heat exchanger, a gas cooler, dual adsorbers, a fission product gas delay trap, and dual filters. The fission product trap is sized to provide a hold-up of 30 minutes for Kr and 6 hours for Xe and for 99.9% retention of iodine. Non-radioactive decontamination is sufficient to maintain oxygen bearing contamination at < 30 ppm in the coolant.

Total cost of the system excluding auxiliary equipment and containment was estimated to be \$176,360.

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## 1.0 INTRODUCTION

The helium coolant of any gas cooled reactor will become contaminated with radioactive and non-radioactive gases and solids during the operation of the reactor. Some common sources of these impurities are inleakage of air and water, outgassing of both the graphite reflector and fuel, release of radioactive gases and solids from the fuel, and generation of particulate matter by attrition.

In general, the purification system for the helium coolant must include provisions for removing gaseous and particulate radioactive and non-radioactive impurities.

Radioactive contamination must be kept at a low level so that in the event of a maximum credible accident the release of activity will not exceed acceptable limits. It is also desirable to keep radioactive contamination at a low level so that direct maintenance of certain of the reactor components will be possible with minimum equipment decontamination.

The main reasons for limiting non-radioactive contamination are to prevent chemical corrosion and particulate induced erosion of the various reactor materials and to reduce the effects of poor thermal properties of the coolant.

The proposed purification system for the 800 MWT Pebble Bed Reactor which is presented in this memorandum is based on present technology; however, future developments may allow a more economical design.

## 2.0 PURIFICATION SYSTEM FLOWSHEET

The helium purification system will be composed of three major sections: (1) chemical purification consisting of dual oxidizers and adsorbers, (2) fission product delay by a charcoal delay trap, and (3) dual absolute filters for particulate removal (Fig. 2.1). An economizer heat exchanger and gas cooler are also necessary to conserve coolant heat and reduce the coolant temperature prior to its entry to the adsorbers and fission product trap.

### 2.1 Chemical Purification

The non-radioactive gaseous contaminants of interest are CO, H<sub>2</sub>, hydrocarbons, CO<sub>2</sub>, and H<sub>2</sub>O. Unit operations to be used for removal of these contaminants are (1) oxidation of all oxidizable gases to CO<sub>2</sub> and H<sub>2</sub>O and (2) sorption of the CO<sub>2</sub> and H<sub>2</sub>O from the coolant. It was decided to use a fixed bed of CuO as the oxidizer since early experimental work indicated that CuO is an excellent oxidizing agent without appreciable oxygen contamination of the purified gas.<sup>(1)</sup> The oxidizers are to operate at 750°F.

The co-sorption of CO<sub>2</sub> and H<sub>2</sub>O will be by fixed-beds of type 5-A Linde Molecular Sieves. Literature from the Linde Company indicates that H<sub>2</sub>O and CO<sub>2</sub> contamination levels can be reduced to < 1 ppm H<sub>2</sub>O and < 5 ppm CO<sub>2</sub> by use of type 5-A Sieves.<sup>(2,3)</sup> Adsorbers are to operate at ambient temperature.

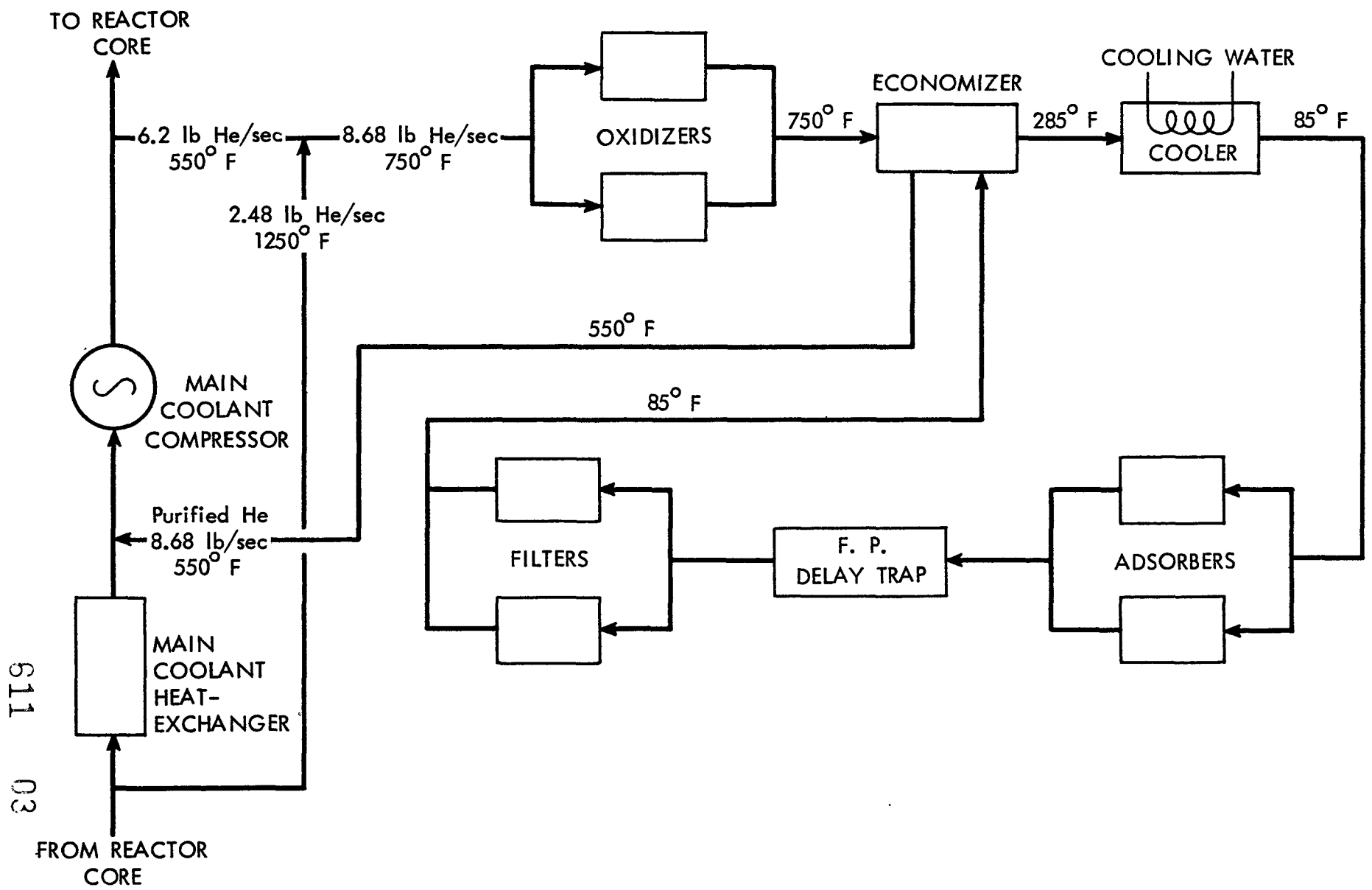


Fig. 2.1. Flowsheet for proposed Helium purification system for the 800 Mwt pebble bed reactor.

## 2.2 Fission Product Delay Trap

A charcoal trap will be used to remove or delay the fission product gases,  $I_2$ , Xe, and Kr. The  $I_2$  will be essentially irreversibly removed by the charcoal. (4) Kr and Xe will be delayed as it passes through the charcoal trap due to dynamic sorption by the charcoal. There is available information on the delay times to be expected in such a system (5) and if these are compared with the half life of each fission product, their effective removal by decay in the charcoal bed can be determined. The fission product delay trap is to operate at ambient temperatures.

## 2.3 Filters

Particulate matter removal in the coolant will be achieved by high temperature filters in the main coolant stream and by low temperature (ambient) filters in the purification side-stream. The filters in the purification system will be placed downstream from the other components to protect the main stream coolant from solids generated by attrition in the purification system.

## 2.4 Heat Exchangers

Since the gas must be cooled after it leaves the oxidizer at 750°F prior to its entry to the adsorbers at 85°F, a heat economizer will be used to transfer some of the heat from the impure helium to the purified helium. It will also be necessary to include a gas cooler (cooled by water) to cool the helium down to ambient temperature.

## 3.0 DESIGN OF PURIFICATION SYSTEM COMPONENTS

The design of each of the major purification system components has been based on the various reactor requirements, properties and criteria.

The following list shows the specification of each component:

<u>Components</u>	<u>Type</u>	<u>Size</u>
Oxidizer (2 each)	Vessel with external heating and insulation containing 1/8-in.-dia CuO pellets	Each 12-in.-I.D. with 22-in. O.D. x 56-in.
Adsorber (2 each)	Vessel with external heating and insulation containing 1/8-in.-dia type 5A Molecular Sieves	Each 24-in. I.D. 34-in.-O.D. x 75-in.
Fission product delay trap	Pipe containing 6-16 mesh charcoal	10 sections of pipe each 24-in.-O.D. x 35-ft
Filter (2 each)	Absolute type with fiber glass	30-in.-dia pipe x 12-ft

<u>Components</u>	<u>Type</u>	<u>Size</u>
Economizer	Shell and tube heat exchanger with insulation	28-in.-O.D. by 17-ft long
Gas cooler	Shell and tube heat exchanger	24-in.-O.D. by 18 ft long

### 3.1 Reactor Requirements, Properties and Criteria Pertinent to Design of Purification System

Following is a list of the reactor requirements, properties and criteria which were used in designing the purification system:<sup>(6)</sup>

1. Coolant will be helium.
2. Coolant working pressure is 700 psia.
3. Coolant temperature to generator will be 1250°F.
4. Coolant temperature to core will be 550°F.
5. Coolant volume will be 30,000 ft<sup>3</sup>.
6. Circuit pressure drop will be 12 psi.
7. Purification system flow rate will be 8.68 lb/sec.
8. Maximum operable H<sub>2</sub>O inleakage will be 30 lb/day.
9. Maximum out gassing from graphite will be 20 lb/day of the following composition:
  - CO<sub>2</sub> - 50% (vol)
  - CO - 10% (vol)
  - H<sub>2</sub>O - 15% (vol)
  - Hydrocarbons - 25% (vol)
10. Maximum oxygen bearing gas contamination in the coolant will be 30 ppm. The composition of this contaminant will be assumed to be 20 ppm CO<sub>2</sub> and 10 ppm H<sub>2</sub>O.
11. The dual oxidizers will operate on a 24 hour cycle at maximum impurity rates.
12. The dual adsorbers will operate on an 8 hour cycle at maximum impurity rates.

### 3.2 Design of Oxidizers

The oxidizers must have capacity for oxidizing  $H_2$  to  $H_2O$  ( $H_2 + CuO = H_2O + Cu$ ),  $CO$  to  $CO_2$  ( $CO + CuO = CO_2 + Cu$ ), and hydrocarbons (represented by  $CH_4$ ) to  $CO_2$  and  $H_2O$  ( $CH_4 + 4CuO = CO_2 + 2H_2O$ ). Following is a list of the  $H_2$ ,  $CO$  (assuming all C-O<sub>2</sub> compounds may be present as  $CO$ ) and  $CH_4$  inputs to the coolant for a period of 24 hours:

$H_2$  - 1.763 lb moles/day

$CO$  - 0.380 lb moles/day

$CH_4$  - 0.160 lb moles/day

This would require 2.78 lb-mols of  $CuO$  for the oxidation and assuming 50% effectiveness of the  $CuO$ , a total of 5.56 lb-mols or 444 lb of  $CuO$  would be needed for each oxidizer. Harshaw Chemical Company type Cu-0307  $CuO$  pellets which may be used, have a pour density of 156 lb/ft<sup>3</sup>; therefore, 2.85 ft<sup>3</sup> of oxidizer capacity is needed. This volume would be available in 12-in. I.D. x 44-in. vessel. Including 8-in. free space in the oxidizer and external electrical heating with 2-in. of insulation, the over-all dimensions would be 22-in.-O.D. x 56-in.

### 3.3 Design of Adsorbers

The dual Molecular Sieve adsorbers were designed for 8-hour continuous operation to adsorb 12.5 lbs of  $H_2O$  and 7.0 lbs of  $CO_2$  each.

According to the Linde Company, dynamic loading of the sieves should be 1/2 of the equilibrium loading.<sup>(2,3)</sup> (Loading is defined as lb of  $H_2O$  or  $CO_2$  adsorbed per 100 lb of Molecular Sieves).

If it is assumed that the adsorber off-gas may contain 1.0 ppm  $H_2O$  and 15 ppm  $CO_2$ , the loading of  $H_2O$  and  $CO_2$  on Type 5A Molecular Sieves assuming stripping down to 0.5 lb/100 lb will be 9.5 lb  $H_2O$ /100 lb sieves and 3.1 lb  $CO_2$ /100 lb sieves.<sup>(2,3)</sup> Effective dynamic loading will then be  $9.5/2 = 4.75$  lb  $H_2O$ /100 lbs of sieves and  $3.1/2 = 1.55$  lbs  $CO_2$ /100 lbs of sieves. The necessary amount of Type 5A Molecular Sieves needed per adsorber will then be,  $(12.5/4.75 + 7.0/1.55) \times 100 = 716$  lb.

A vessel 24-in.-I.D. x 63-in. will contain this amount of Molecular Sieves<sup>(2)</sup> and adding 8-in. free space, and external heating with 2-in. insulation will result in over-all dimensions of 34-in. O.D. x 75-in.

### 3.4 Fission Product Traps

In order to provide a delay of 30 minutes for krypton and 6 hours for xenon, 32,000 lbs of charcoal will be needed in the fission product delay trap. The following relationships for dynamic adsorption were used for determining the necessary amount of charcoal:

$$\tau = km/Q$$



$\tau$  = mean delay time in bed for Xe or Kr, min.

k = dynamic adsorption coefficient for Xe or Kr, cc-atm/g-atm.

m = mass of charcoal, g.

Q = flow rate of gas through bed, cc/min.

Therefore,  $m = \tau Q/k$  where,

$\tau = 30$  min for Kr or 6 hr for Xe.

Q =  $2.84 \times 10^7$  cc/min.

k = 59 cc/g for Kr or 700 cc/g for Xe.

or

$$m_{(\text{Kr})} = \frac{(30)(2.84 \times 10^7)}{(59)} = 1.44 \times 10^7 \text{ g and}$$

$$m_{(\text{Xe})} = \frac{(360)(2.84 \times 10^7)}{700} = 1.45 \times 10^7 \text{ g or}$$

$$m_{(\text{maximum})} = 1.45 \times 10^7 \text{ g} = 32,000 \text{ lbs.}$$

Since the bed density is 32 lb/ft<sup>3</sup>, 1000 ft<sup>3</sup> of bed is needed. This material can be divided into ten - 100 ft<sup>3</sup> beds in parallel. Each bed should be 35 ft of 24 inch O.D. pipe filled with charcoal to 32 lb/ft<sup>3</sup>.

### 3.5 Design of Filters

The filter must have a capacity of 8.68 lb He/sec at all times. Therefore, two full size units are provided so that one may be replaced while the other is on stream. After determining the volumetric flow rate of 1000 cfm, the filter was sized (by Flanders rated capacities for 12 inch deep units)<sup>(7)</sup> to require 3.1 ft<sup>2</sup> face area. A circular unit 2-ft-dia enclosed in a 30 inch diameter pipe will suffice.

### 3.6 Design of Heat Exchangers

It was decided to base the design of the necessary heat exchangers on 24-in.-O.D. shell and tube heat exchangers with 296 3/4-in.-O.D. tubes on 1-in. square pitch with 8 passes and 25% cut baffles.

Economizer. Gas is taken from the oxidizer at 750°F and is to be cooled to 85°F for the adsorber and delay trap, after which it will be heated back up to 550°F. These temperatures fixed the economizer exit hot-gas temperature at 285°F by a heat balance. This leads to the following design:

heat transfer area needed,  $A = \frac{Q}{(\Delta T)(U)}$

where,

$A$  = heat transfer area in heat exchanger,  $\text{ft}^2$

$Q$  = heat transferred,  $\text{Btu/hr}$

$\Delta T$  = log mean temperature difference,  $^{\circ}\text{F}$

$U$  = over all heat transfer coefficient,  $\text{Btu/hr-ft}^2\text{-}^{\circ}\text{F}$

The over-all heat transfer coefficient,  $U$ , can be determined from,

$$\frac{1}{U} = \frac{1}{h_i} + \frac{1}{h_o} + R_d$$

where,

$h_i$  = tube side heat transfer coefficient,  $\text{Btu/hr-ft}^2\text{-}^{\circ}\text{F}$

$h_o$  = shell side heat transfer coefficient,  $\text{Btu/hr-ft}^2\text{-}^{\circ}\text{F}$

$R_d$  = fouling factor,  $\text{hr-ft}^2\text{-}^{\circ}\text{F/Btu}$

With the following values of those functions,

$$Q = 1.8 \times 10^7 \text{ Btu/hr}$$

$$\Delta T = 200^{\circ}\text{F}$$

$$h_i = 710 \text{ Btu/hr-ft}^2\text{-}^{\circ}\text{F}$$

$$h_o = 257 \text{ Btu/hr-ft}^2\text{-}^{\circ}\text{F}$$

$$R_d = 0.0035 \text{ hr-ft}^2\text{-}^{\circ}\text{F/Btu}$$

$$U = 114 \text{ Btu/hr-ft}^2\text{-}^{\circ}\text{F}$$

the heat transfer area needed is,

$$A = \frac{1.8 \times 10^7}{(200)(114)} = 790 \text{ ft}^2$$

This amount of heat transfer area is contained in approximately 15 ft of the 24-in.-O.D. heat exchanger. Addition of 2 ft of length for head covers results in an over-all length of 17 ft and addition of 2-in. insulation results in a 28-in.-O.D.

Cooler. In the cooler the gas temperature must be reduced from  $285^{\circ}\text{F}$  to  $85^{\circ}\text{F}$  by cold water at  $75^{\circ}\text{F}$ , with an arbitrary  $50^{\circ}\text{F}$  temperature rise. This leads to the following design:

$$Q = 7.75 \times 10^6 \text{ Btu/hr}$$

$$\Delta T = 54^\circ\text{F}$$

$$h_i = 710 \text{ Btu/hr-ft}^2\text{-}^\circ\text{F}$$

$$h_o = 1000 \text{ Btu/hr-ft}^2\text{-}^\circ\text{F}$$

$$R_d = 0.0035 \text{ hr-ft}^2\text{-}^\circ\text{F/Btu}$$

$$U = 169 \text{ Btu/hr-ft}^2\text{-}^\circ\text{F}$$

and the amount of heat transfer area needed is,

$$A = \frac{7.75 \times 10^6}{(54)(169)} = 850 \text{ ft}^2$$

This amount of heat transfer area can be contained in approximately 16 ft of the 24-in.-O.D. heat exchanger. Addition of 2 ft for head covers results in an over all length of 18 ft.

#### 4.0 OPERATION AND MAINTENANCE

The proposed helium purification system will operate continuously with cyclic operation of the dual oxidizers and adsorbers. It is hoped that the purification system will utilize the main coolant loop pressure drop for fluid flow. Thus, there will be no moving parts in the major equipment components.

Since there are no major moving parts, it is expected that the system will be essentially maintenance free. However, the CuO oxidizer will have to be regenerated every 24 hours and the Molecular Sieve Adsorbers will have to be regenerated every 8 hours. Regeneration will be accomplished by flowing dry air through the vessels at 600°F for adsorbers and 750°F for oxidizers. The life of the CuO pellets, Molecular Sieves, and charcoal will probably exceed 2 years.

#### 5.0 COST ESTIMATE OF PURIFICATION SYSTEM

The cost estimate for the coolant purification system was prepared for the installed equipment and initial chemical reagents only and does not include the enclosure or auxiliary services. The cost estimating procedure used is a modification of the one presented by Aries and Newton<sup>(8)</sup> in which the purchase cost of major process equipment is used to determine the cost of other components of the facility by use of factors which have been empirically determined. The total cost of the purification system was determined to be \$176,360.

##### 5.1 Purchase Cost of Major Process Equipment

Major process equipment was decided to be any piece of process equipment whose total installed cost was greater than \$1000. There are a total of 14 such

pieces of equipment (Table 5.1 and Fig. 5.1) which have a total purchased cost of \$47,170.

### 5.2 Initial Cost of Chemical Reagents

The chemical reagents which are necessary for initial charging of the equipment in the purification system are activated charcoal, CuO, and Molecular Sieves. The total cost of this initial reagent charge is \$22,220 (Table 5.2). It is estimated that this material will be effective for a minimum of 2 years.

### 5.3 Total Cost of the Facility

Total facility cost was determined by use of the factors given by Aries and Newton in which the cost of equipment installation is 43% of the purchased equipment cost, piping is 36%, instrumentation is 50%, insulation is 8%, and electrical is 15% (Table 5.3). Addition of initial reagents cost results in a physical plant cost of \$141,090 and addition of 25% for contingency results in a total facility cost of \$176,360.

## 6.0 RESEARCH AND DEVELOPMENT REQUIREMENTS

It was necessary in the preliminary design of the helium coolant purification systems for the proposed pebble bed reactor and for the pebble bed reactor experiment<sup>(12)</sup> to make a number of assumptions because of lack of complete design data. Following is a brief list of the areas in which more complete data is required so as to facilitate a more rigorous design of such a purification system:

1. Dynamic adsorption of Kr, Xe, and I on charcoal at relatively high pressures (1000 psi) including the effect of iodine poisoning.
2. Kinetics of oxidation by CuO.
3. Kinetics of co-sorption of H<sub>2</sub>O and CO<sub>2</sub> by Molecular Sieves at high pressures and low H<sub>2</sub>O and CO<sub>2</sub> levels.
4. Kinetics and reaction mechanism of the graphite-gas reactions at pressure, temperature, and flow rates of interest.
5. Kinetics of the graphite mass transfer reaction,  $2\text{CO} \rightarrow \text{C} + \text{CO}_2$ , at conditions of interest.
6. High temperature filter efficiency.
7. Amount of particulate activity to be expected in coolant circulating stream.
8. Effect of iodine sorption on Molecular Sieves.

Table 5.1. Specifications and Purchase Cost of Major Process Equipment for the Purification System

Equipment Number (Fig. 5.1)	Quantity	Description	Size	Material of Construction	Maximum Temperature, °F	Maximum Pressure psi	Material Flow Rate, lb/sec	Total Purchase Cost for 1960	Cost Reference
1	2	Oxidizer vessel	12-in.-I.D. x 44-in. extra heavy pipe w/4 fittings	mild steel	750	700	8.68	1,730	8, p. 49
2	2	Oxidizer heater	5 KW of electrical of heat	--	1500	--	--	1,100	10
3	1	Economizer shell and tube heat exchanger	24-in.-O.D. x 17-ft long with 790 ft <sup>2</sup> of heat transfer area	mild steel	750	700	8.68	6,880	8, p. 50
4	1	Gas cooler, shell and tube heat exchanger	24-in.-O.D. x 18 ft long with 850 ft <sup>2</sup> of heat transfer area	mild steel	285	700	8.68	7,160	8, p. 50
5	2	Adsorber vessel	24-in.-O.D. x 71-in. extra heavy pipe w/4 fittings	mild steel	600	700	8.68	4,580	8, p. 49
6	2	Adsorber heaters	10 KW of electrical heat	--	1500	--	--	1,760	10
7	1	Fission product delay trap	350-ft of 24-in.-O.D. extra heavy pipe w/20 fittings	mild steel	100	700	8.68	19,400	8, p. 49
8	2	Absolute filter, Rlanders type - CC21-C	30-in.-O.D. x 12-in.	fiber glass in mild steel	100	700	8.68	2,000	9
9	1	Chromatograph, gas adsorption	---	--	100	30	--	2,500	11
Total Purchased Equipment Cost								<u>647,170</u>	

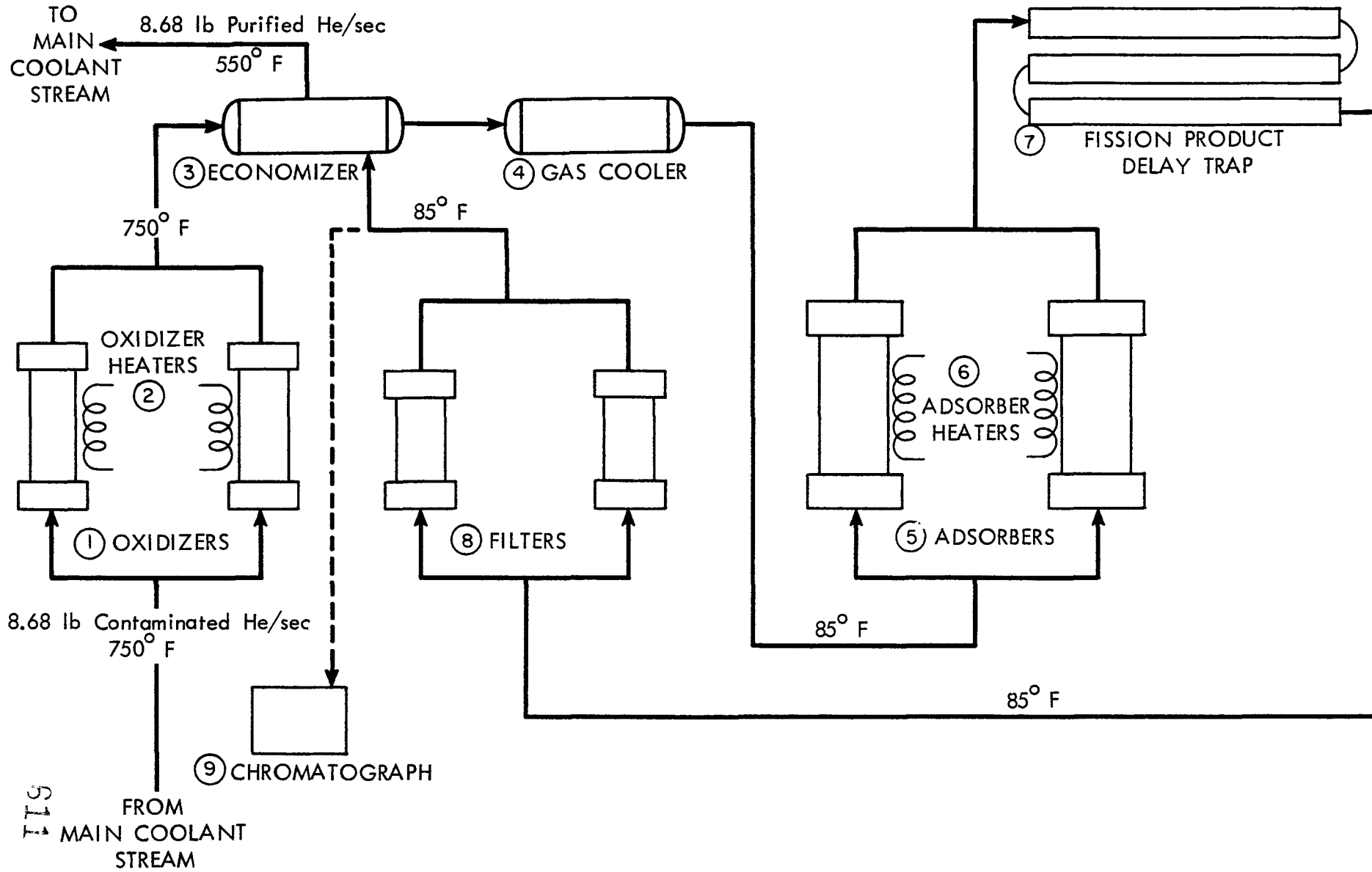


Fig. 5.1. Pebble bed reactor Helium coolant purification system flowsheet showing major process equipment.

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Table 5.2. Cost of Initial Chemical Reagent Charge

Reagent Description	Amount, lbs	Unit Cost \$/lb	Total Cost	Cost Reference
Type PCP activated charcoal 6-16 mesh	32,000	0.51	\$16,320	13
CuO pellets, 1/8-in.-dia	888	3.50	3,110	14
Type 5A Molecular Sieves, 1/8-in.-dia	1,432	1.95	2,790	15
	Total reagent cost		<u>\$22,220</u>	

Table 5.3. Total Cost of Purification Facility\*

Purchased equipment cost	\$ 47,170
Installation (43% of purchased equipment cost)	20,280
Piping (36% of purchased equipment cost)	16,980
Instrumentation (50% of purchased equipment cost)	23,590
Insulation (8% of purchased equipment cost)	3,770
Electrical (15% of purchased equipment cost)	7,080
Initial chemical reagent charge	<u>22,220</u>
Physical plant cost	\$141,090
Contingency (25% of physical plant cost)	<u>35,270</u>
Total cost of facility	<u>\$176,360</u>

\* Does not include cost of building, enclosures, utilities, etc.

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