“Analysis of helium environments in former HTR’s”

NNC Contribution - Helium purification in the Dragon Reactor

by

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Summary

This report provides a review of the purification system used for the Dragon Reactor. The report contributes to deliverable D44 of the HTR-E Project.

Information is given on the helium atmosphere and the purification process used.

1. Introduction

This document covers a review of the purification system used in the Dragon Reactor.

The purpose is to provide where possible available information on the helium atmosphere (gas content, dust particles) during operation, initial specifications and for the helium purification system.

This document contains the results of work at NNC on Deliverable D44 of the HTR-E Project.

2. The Dragon Reactor purification requirements

The Dragon Project was a 20 MW helium-cooled reactor experiment that ran from 1959 to 1976. A cross section of the Dragon reactor is shown in Figures 1 and 2. The process of helium purification comprised adsorption of both fission product and impurity gases on liquid nitrogen-cooled charcoal beds (Refs 1 & 2).

The quantities of fission products and chemical impurities within the helium coolant of a HTR must be limited to avoid neutron absorption, graphite burn-up and carbon deposition. The design specification for impurity concentration levels in Dragon is presented in Table 1. Regarding the fission product content, it was desirable from an operational standpoint to reduce the level as much as possible.

During the start-up period of the reactor, large amounts of impurities will have been liberated from the moderator and reflector graphite. And during normal operation, impurities will have been introduced continuously, either by the outgassing of exchanged fuel elements or by small water or air inleakages. Further outgassing of the whole circuit will also have continued to proceed.

Fission products escaping from the fuel boxes were continuously scavenged and transported to a trapping system by a small flow of helium, drawn from the primary cooling circuit via the interior of the graphite fuel element cans. This gas was then passed through a system of traps to remove both fission products and gaseous impurities from the helium before it was returned to the reactor. During commissioning and start-up, when the circuit would contain large quantities of impurities, it was undesirable to pass them through the fuel elements on their way to the purification system. Provision was therefore made for drawing a second stream of coolant gas directly from the primary circuit, under such circumstances, without interrupting or reducing the purge flow through the fuel elements.

3. The purification process

The purification plant was situated within the helium circuit as shown in Figure 3. A schematic diagram of the helium purification plant is presented in Figure 4. The main purge gas process consisted of the following stages:

1. **Admitting a small, controlled and approximately equal flow of scavenge gas to the top of each fuel rod.**

   The purge gas was admitted at the top of each fuel rod via a restriction designed to prevent back-diffusion of fission products and to equalise flows between the seven rods in each element.

2. **Scavenging away gaseous and volatile fission products appearing in the annular gap between the graphite fuel boxes and graphite outer fuel cans.**

3. **Trapping out fission products likely to cause corrosion or blockage of subsequent parts of the system, by means of a fission product trap situated in the bottom of each fuel rod.**

   After passing through the fuel boxes and the outer fuel cans, the scavenge gas, now bearing gaseous and volatile fission products, passed through traps in the bottom of the fuel elements. These traps, containing charcoal, were intended to capture as much as possible of the corrosive fission products such as tellurium and the halogens, together with strontium and barium which might otherwise block the purge gas piping further on in the system.

4. **Conducting the purge gas stream from each fuel element into a common purge gas manifold, via equipment for equalising and sampling streams from the individual fuel elements, and detecting and locating faulty or unseated fuel elements.**

   The purge gas left each fuel element via a mounting spike. Each spike unit contained an orifice restrictor to equalise the flows between the fuel elements and to detect changes in flow resulting from blocked, unseated or broken fuel elements. The orifices were protected from blockage by miniature cyclones incorporated in the spike units. Dust removed by the cyclones was collected in ceramic-lined pockets containing thermocouples to detect the effect of decay heat build-up. The spike units were removable, removal and replacement being carried out using a special tool on the fuel element charge/discharge machine. The purge gas left the spike unit sockets via short pipes which were connected to a purge pipe manifold suspended under the core support grid.

5. **Condensing out volatile fission products and cooling the purge stream to a temperature not exceeding 100°C.**

   Purge gas was conducted by the pipe manifold to a pre-cooler (see Figure 5) situated in the bottom of the reactor vessel. The pre-cooler cooled the purge gas from 350°C to approximately 100°C and was intended to condense out metallic fission products from the gas as solid deposits. When clean, the pre-cooler removed 10kW of heat from the purge gas but as fission products built up, an additional 60 kW of decay heat was produced. In order to maintain the correct temperature gradients for controlled solids deposition, the unit was gas-cooled. For

this purpose, clean helium was circulated around a closed circuit containing the pre-cooler and a water-cooled heat exchanger. In the pre-cooler unit the clean helium was conveyed through a finned helical pipe coil; the fission product-bearing purge stream passed over the outside of this coil in counter flow.

6. **Trapping out any remaining fission product iodine and bromine.**

7. **Delaying the fission product gases xenon and krypton in water-cooled charcoal beds for sufficient time for them to lose most of their decay heat and their most damaging activity.**

   The iodine traps and delay beds were charcoal-filled water-cooled units (see Figure 6). The process gas flowed first through the iodine traps and then through a series of interconnected charcoal-filled U-tubes, of progressively increasing diameter in the delay beds. Each delay bed contained 3 m^3 of charcoal. Cooling water was introduced at the bottom of each delay bed outer containment shell and flowed upwards at low velocity over the outside of the charcoal-filled tubes. It left at the top of each delay bed and flowed at a much higher velocity through the jackets of the iodine traps, which were mounted immediately above the delay beds. The water circuits were designed to prevent the accumulation of pockets of gas formed by radiolytic decomposition of the water. The radiolytic gases were swept out to a header from which they were circulated continuously through a catalytic recombiner by a water-driven ejector in the coolant circuit. Warm water from the header was drawn through a heat exchanger in which it was cooled by water from the cooling tower, before being pumped back to a distribution manifold feeding the delay beds. Any radioactive gas leaking from the traps was confined in the water circuit. Isolating valves on both gas and water sides permitted isolation of a faulty trap.

   The delay beds were designed to delay krypton for 15 hours and xenon for 200 hours, leaving only long-lived rare gas nuclides in the process stream.

8. **Oxidising impurity gases such as CO, H_2 and CH_4 to H_2O and CO_2, which are easier to trap.**

   The process gas leaving the delay beds could be fed to any one of three identical purification plants. Each plant consisted of a high temperature (400-600°C) section, followed by a low temperature section. In the high temperature section, the gas was heated and fed through an oxidiser containing copper oxide to oxidise hydrogen, carbon monoxide and methane.

9. **Removing water vapour and carbon dioxide in two beds of 4A molecular sieve, at ambient temperature.**

10. **Further delaying the residual xenon and krypton in a liquid nitrogen cooled charcoal bed for sufficient time to eliminate activity due to all nuclides except ^{85}Kr, which has a half-life of 10 years.**

The cold gas was then passed through a delay trap, cooled by liquid nitrogen boiling and condensing in a closed circuit. The cold delay trap contained 100 litres of charcoal and was designed to delay the rare gases sufficiently for all the radioactive gases, except ^{85}Kr, to decay.

11. Trapping the remaining stable xenon and krypton, together with ^{85}Kr, and the residual non-oxidisable impurity gases such as N_2, Ar, etc, in a regenerable charcoal trap.

The ^{85}Kr, inactive xenon and krypton, and impurity gases leaving the cold delay trap were collected in a second charcoal bed. Since little radioactive decay occurred in this bed, it was not cooled by liquid nitrogen but was kept cold by the process gas passing through it. When the trap was full, it was regenerated by recycling hot helium through a set of tubes imbedded in the charcoal.

12. Discharging the contents of the freezer unit and the regenerable charcoal trap at intervals to a system of dirty gas receivers where they will be sampled and monitored for activity. Then discharging them in a controlled fashion to the atmosphere via the stack.

During regeneration of the freezer and final charcoal trap, their contents were exhausted to previously evacuated dirty gas receivers. The contents of these receivers could be isolated from the purification plant and monitored for radioactivity before being pumped out to the stack. Water from the freezer was allowed to accumulate in a drain vessel.

13. Warming up the cleaned helium and returning it to the reactor by means of purge gas circulators.

The cleaned helium was finally chilled in a set of coils in the liquid nitrogen head tank and was then returned to the clean side of the 'freezer' heat exchanger where it was warmed to room temperature by the incoming dirty gas. Whenever ice or solid CO_2 built up, indicating that the freezer was in need of regeneration, it was warmed up by recycling hot helium through its clean gas passages.

On leaving the cold plant, the process gas was essentially radioactively and chemically pure helium. It was then routed to the compressor room to be returned to the primary circuit by the main purge gas circulators. Some or all of the flow could be routed to the dump tanks, or compressed into high pressure storage cylinders.

4. Operational helium

During the irradiation of the first charge, one purification plant was operated continuously on the primary circuit, one at 8 g s^{-1} on fuel purge and one intermittently at 4 g s^{-1} on by-pass. The purity of the primary circuit improved continuously during the run (see Table 2; Ref 3). The amount of carbon removed from the core and

reflector in the form of gaseous impurities was approximately 900 g. Since no significant carbon deposits were found on examination of the heat exchanger and circulator, this quantity of carbon was assumed to represent the total graphite corrosion.

Levels of impurities in the primary circuit during the periods when continuous operation was possible with Charge II are shown in Table 3 (Ref 4). Impurities were controlled well within specified limits except during a short period following start-up and during injections. Even after loading a new core an equilibrium level of impurities was rapidly attained in the primary circuit. The most significant change was in the hydrogen level which fell from 2 vpm to 0.7 vpm following the repair of corroded heat exchanger tubes. The total amount of carbon transferred from the system to the purification plant by gaseous impurities was less than 0.75 kg, excluding injection periods.

When a heat exchanger was replaced, the primary circuit helium was replaced by nitrogen. Peaks of up to 5% O_2 occurred in the primary circuit and an average level of 2% O_2 was present over a period of approximately 20 days. The inleaking air carried with it about 0.5 kg of water vapour. At the end of the shutdown, the nitrogen was replaced by helium which was purified at ambient temperature before start-up. As the power was raised, oxidising impurities were rapidly removed, as shown in Table 4 (Ref 4), with the loss of only 100 g of graphite. During the approach to power, the fuel element purge was switched off to avoid drawing impurities directly over the fuel.

Typical impurity levels in the primary circuit helium during 1969-1970, excluding start-up transients and impurity injections, are presented in Table 5 (Ref 5). The hydrogen level shows the effect of adding lithium hydroxide to the secondary coolant. Before this treatment in 1968 the primary circuit hydrogen level did not fall below 2 vpm.

5. References

1. OEEC Dragon High Temperature Reactor Project, Second Annual Report, European Nuclear Energy Agency, 1960-1961.

2. OECD Dragon High Temperature Reactor Project, Fourth Annual Report, Nuclear Energy Agency, 1962-1963.

3. OECD Dragon High Temperature Reactor Project, Eighth Annual Report, Nuclear Energy Agency, 1966-1967.

4. OECD Dragon High Temperature Reactor Project, Ninth Annual Report, Nuclear Energy Agency, 1967-1968.

5. OECD Dragon High Temperature Reactor Project, Eleventh Annual Report, Nuclear Energy Agency, 1969-1970.

Table 1 Tolerated impurity levels (vpm) in the Dragon design specification

O_2	CO_2	H_2O	CO	CH_4	H_2
0.001 - 0.01	5 - 10	5 - 10	0 - 100	0 - 100	0 - 100

Table 2 Impurity levels (vpm) during the irradiation of the first charge

Month of operation, 1966	H_2	CH_4	H_2O	CO	CO_2	N_2
April	4.1	0.8	0.03	6.4	0.60	7.3
May	2.2	1.2	0.01	3.9	1.2	0.59
June	2.4	0.62	0.01	2.1	0.22	0.43
July	3.0	0.39	0.06	0.83	0.10	0.17
August	2.1	0.32	0.17	0.79	0.04	0.18

Table 3 Impurity levels (vpm) in Dragon primary circuit, 1967-68

Month of operation	H_2	CH_4	H_2O	CO	CO_2	Days at power
Jan 1967*	2.2	0.16	0.05	1.8	0.02	23
May 1967	0.87	0.08	0.04	0.7	0.02	57
Jun 1967	0.75	0.10	0.06	0.85	0.02	82
Oct 1967*	1.4	0.15	0.4	1.5	0.47	129
Nov 1967	0.58	0.12	0.07	0.65	0.03	142
Dec 1967	0.62	0.09	0.07	0.56	0.02	172
Jan 1968*	1.0	0.07	0.07	0.4	0.02	197
Feb 1968	0.75	0.12	0.09	0.55	0.02	226

* Start-up months

Table 4 Oxidising impurity removal on start-up after heat exchanger maintenance, October 1967

Power (MW)	CO_2 (vpm)	H_2O (vpm)	Date
2	-	40	17 Oct
7	19.7	8.7	17/18 Oct
8	12.6	6.1	17/18 Oct
9	11.3	6.2	17/18 Oct
10	12.1	6.0	17/18 Oct
16.5	2.5	1.17	19 Oct
16.5	1.3	0.68	20 Oct
16.5	0.66	0.39	21 Oct
17.9	0.12	0.09	25 Oct

Table 5 Steady-state impurity levels (vpm) in Dragon, 1969-1970

H_2O	CO_2	H_2	CO	CH_4	N_2	O_2
<0.05	<0.02	0.5	0.03	<0.04	0.05	<0.02

Figure 1 Section through Dragon Reactor

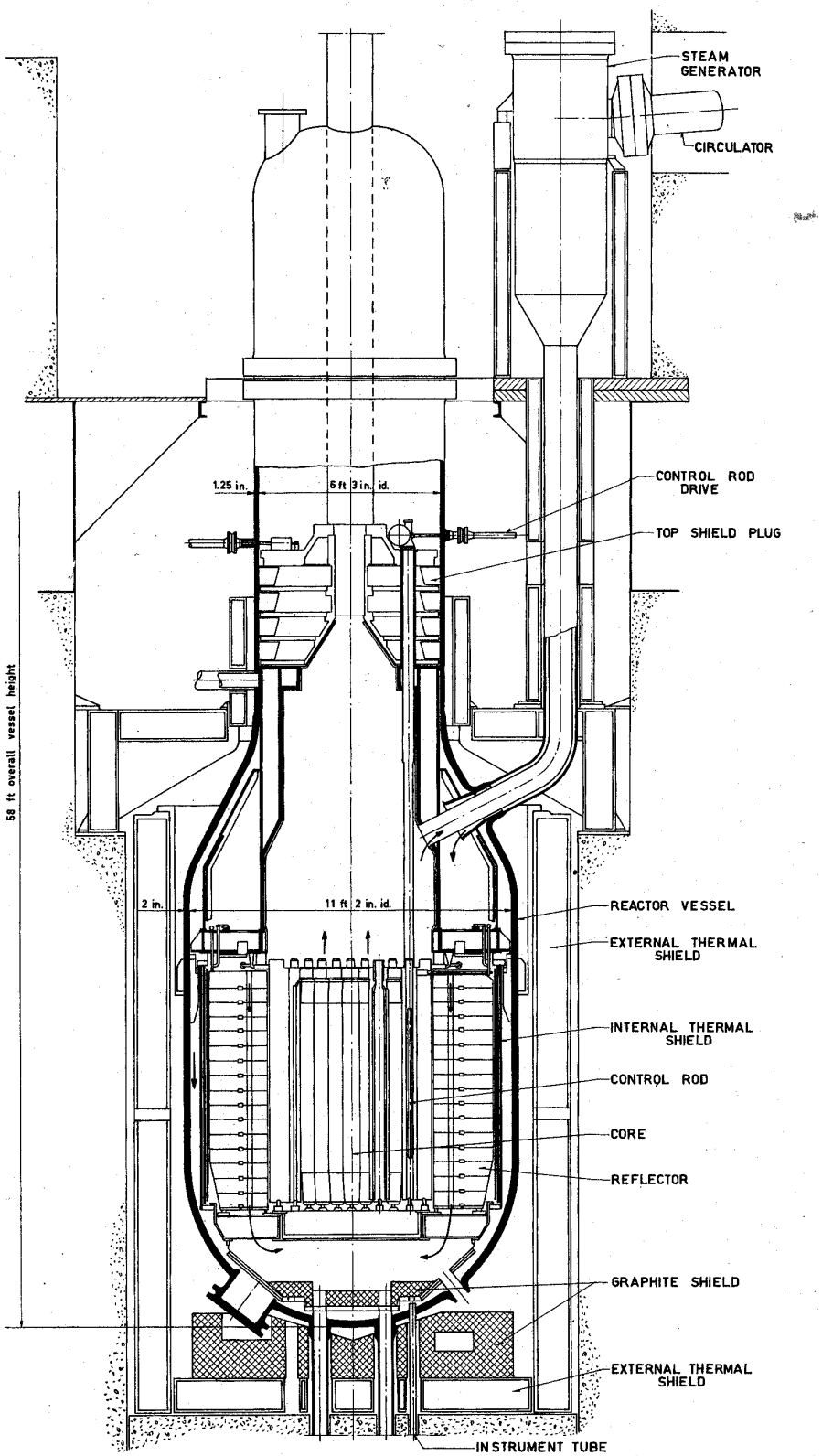

Figure 2 Layout of Dragon Reactor

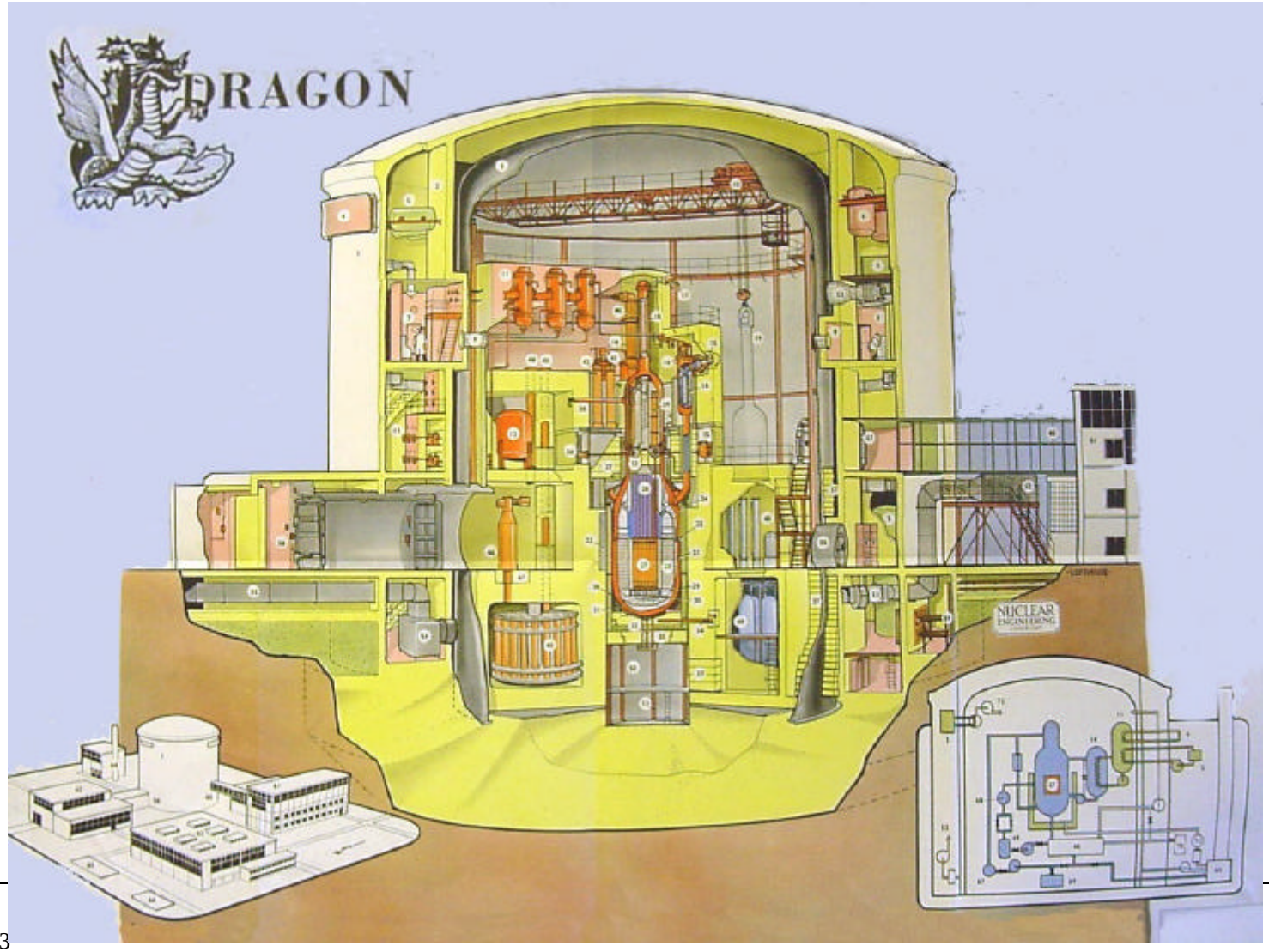

Figure 3 Simplified Helium Flow

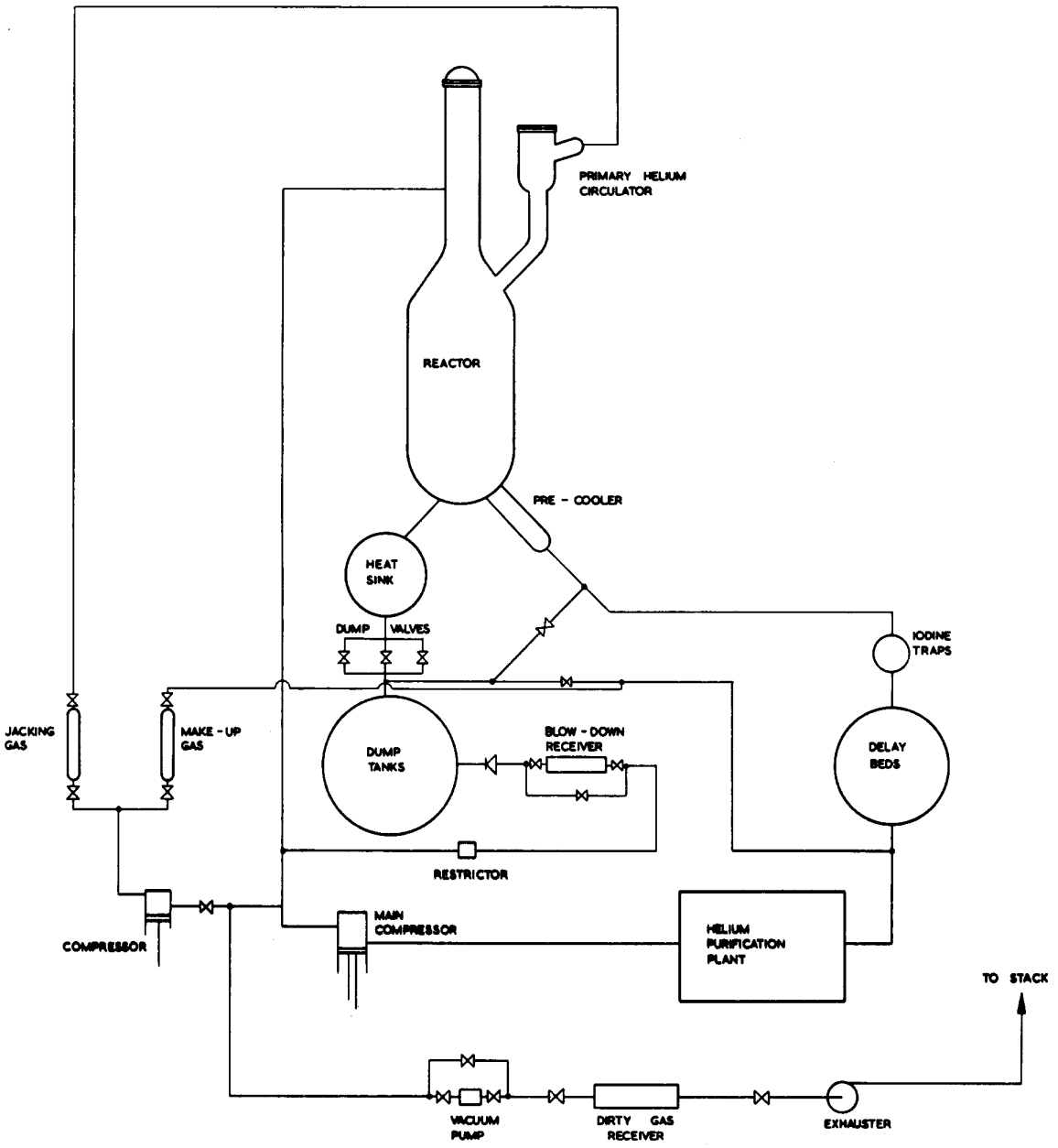

Fig. 9. SIMPLIFIED HELIUM FLOW SHEET.

Figure 4 Schematic of Dragon Reactor Helium Purification Plant

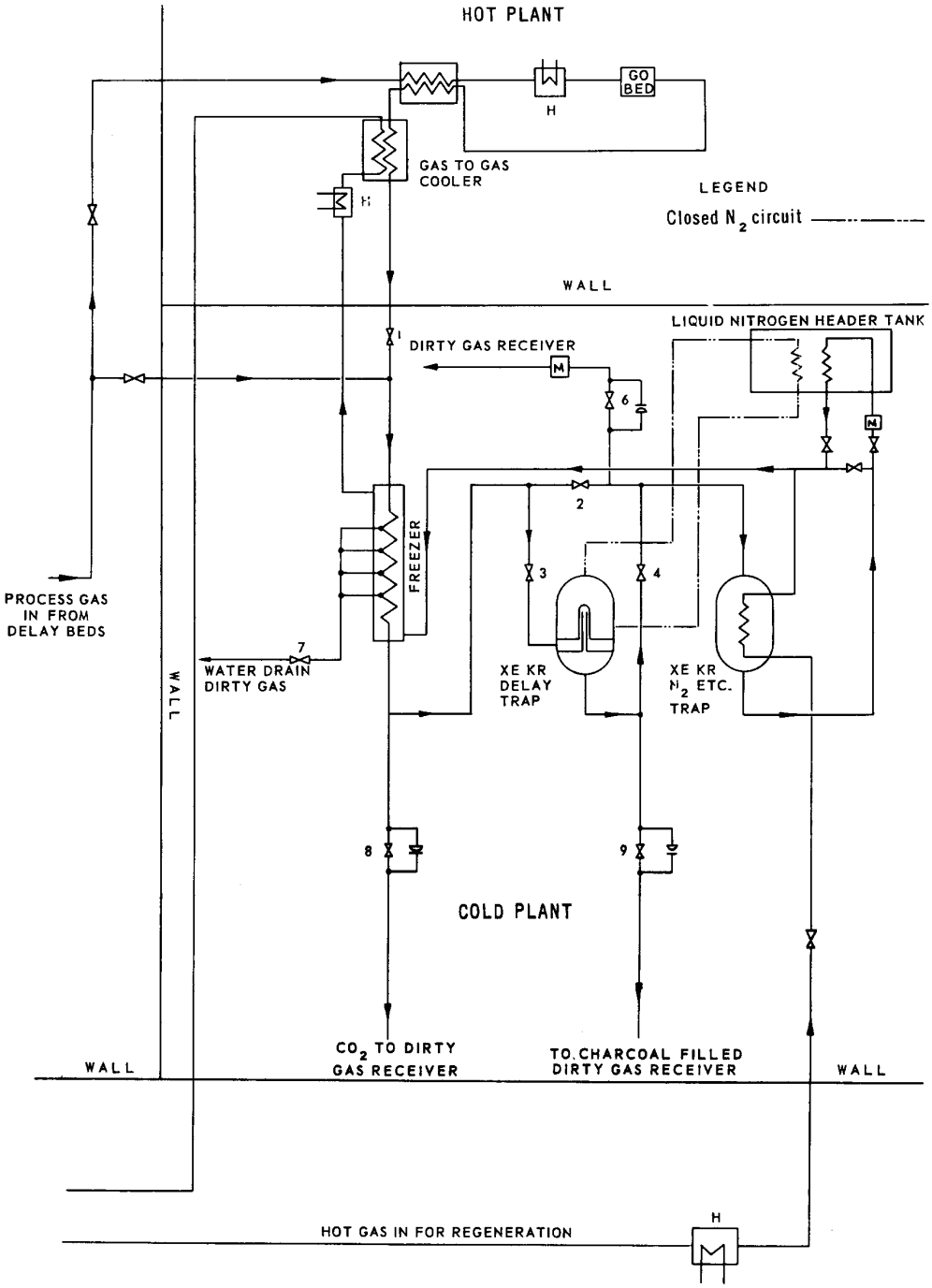

Fig. 5. HELIUM PURIFICATION PLANT

Figure 5 Assembly of Purge Gas cooler

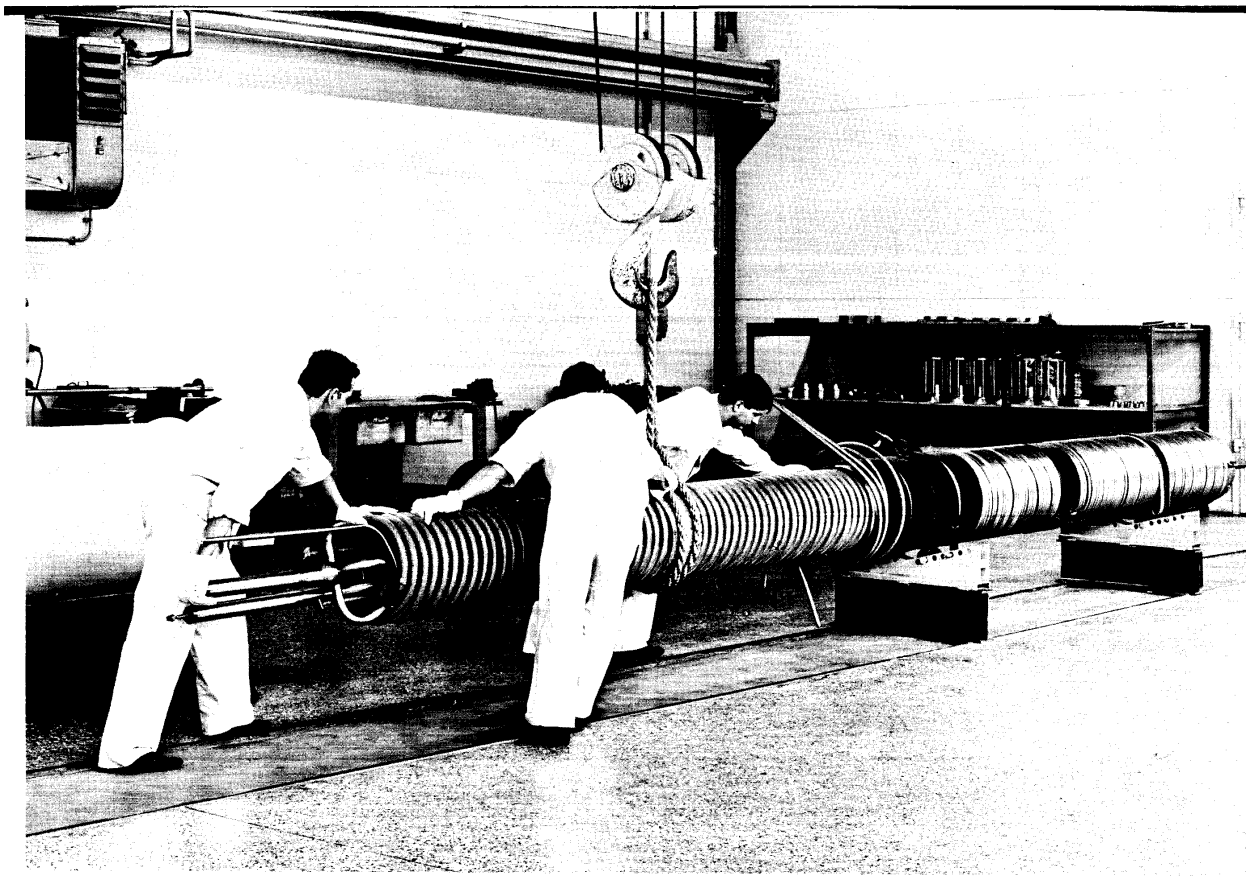

Fig. 11 THE PURGE GAS PRECOOLER

Figure 6 Filling of delay beds with charcoal

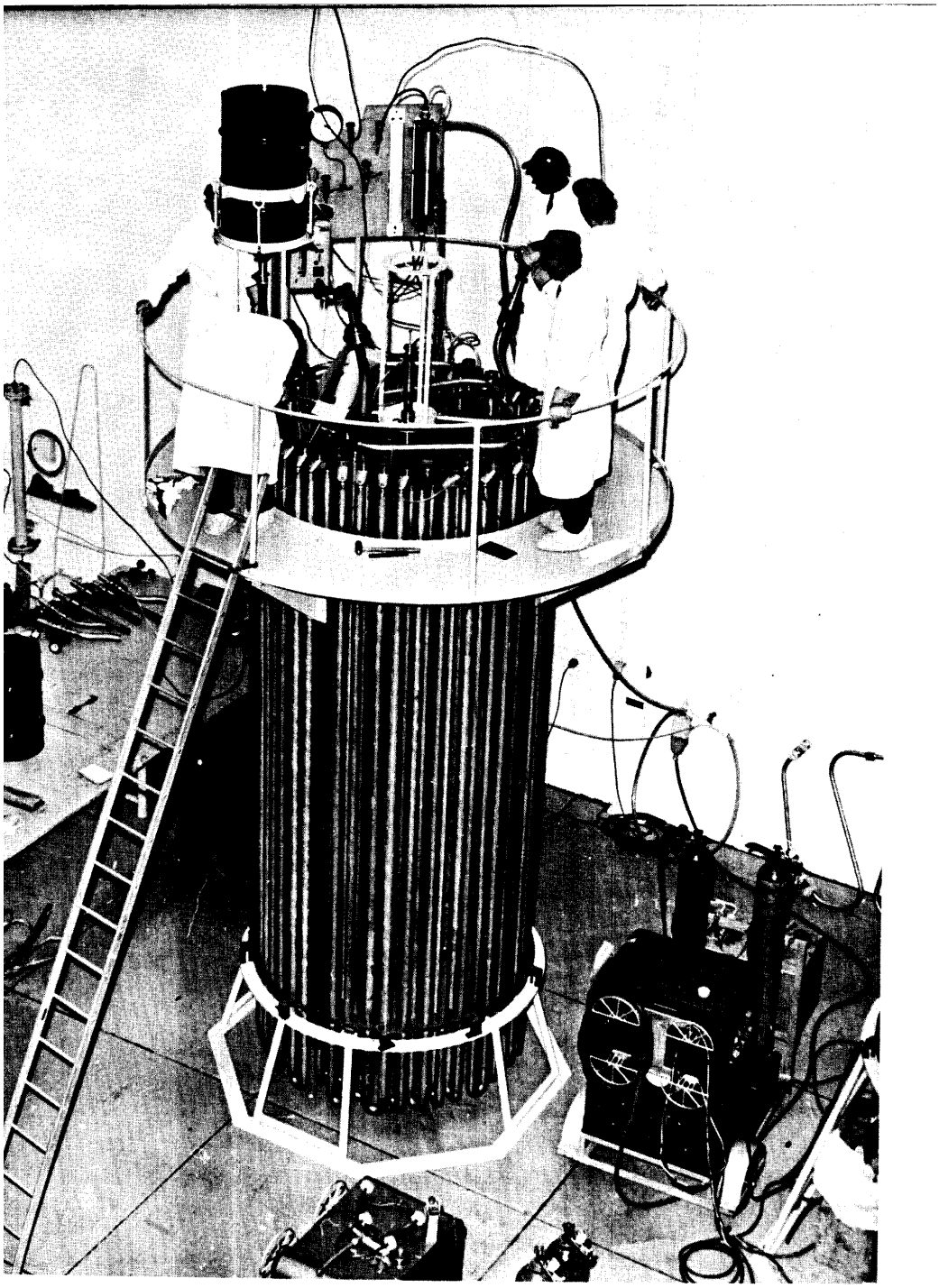

Fig. 18 DELAY BEDS - FILLING WITH CHARCOAL OF TUBE BUNDLE