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THE FACILITY 350 HELIUM-ATMOSPHERE SYSTEM

by

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ABSTRACT

The helium atmosphere system in Argonne's Facility 350 is described in detail. The system is straightforward, employing drying and carbon towers for the removal of moisture, oxygen, and other impurities. The bulk of the 15,000 ft³ of helium atmosphere is continuously recirculated at nearly atmospheric pressure. Purification is accomplished at 140 psig on a portion of the gas that is passed through the drying tower at room temperature and the carbon towers at -46°C (-50°F). The operation is continuous, requiring a minimum of maintenance and operational manpower. The helium atmosphere is supplied to the gloveboxes with impurity levels below 3,000 ppm (0.3%) nitrogen, 1,000 ppm (0.1%) oxygen, and 50 ppm moisture. Such purity levels prevent oxidation and combustion of the plutonium materials being processed. Experimental data concerning the adsorption of oxygen from helium by activated carbon over a range of temperature and pressure conditions are also reported.

I. INTRODUCTION

Facility 350 at Argonne National Laboratory, (1-7) was designed and built for the purpose of processing plutonium-bearing fuel materials. Extremely toxic and pyrophoric materials are being processed in large quantities with maximum safety to the operational personnel, the Laboratory, and the surrounding community. Much of this operational safety may be credited to the use of an inert helium atmosphere in most of the gloveboxes.

The helium atmosphere system in Facility 350 has been operating continuously for about two years. During this period of time, a complete knowledge of the system has been acquired, operational procedures have been established, and an initial report has been made.⁽⁸⁾ The system has proven its worth. It has also proven that a wide variety of production operations can be carried out with ease in an inert atmosphere of this type, and, in particular, operations that could not have been considered were not an inert atmosphere provided.

This report describes the helium atmosphere system in Facility 350 in detail. Attention is given to system design, operation, and control aspects. Selected operational data is included in the appendix, along with the results of a series of oxygen-adsorption experiments carried out over a range of temperature and pressure conditions.

II. CHOICE OF AN INERT GAS

Of the gases normally used as protective atmospheres in metallurgical operations, only argon and helium are satisfactory for all the fabrication processes performed in the Facility 350 gloveboxes. Nitrogen, although sometimes used, is not satisfactory for high-temperature operations exposed directly, or indirectly, to the atmosphere.

Comparisons of the relative merits of inert gases have been reported, including their physical properties and cost.⁽⁵⁻⁷⁾ Based on these facts, and others, helium was chosen over argon for the following reasons:

- (1) Proven regeneration cycles were available.
- (2) Helium has a high heat capacity.
- (3) Helium in a system can be useful as a built in "leak detector."
- (4) Small piping can be used in certain sections of the system with little pressure drop.
- (5) At the time the choice was made, the cost of helium was less than argon.

The main disadvantages associated with the use of helium as a protective atmosphere are the difficulties involved in pumping it, and its high rate of diffusion, making containment difficult.

III. CHOICE OF A PURIFICATION METHOD

Cryostatic purification techniques are used extensively in large helium-producing plants with final purification by adsorption on activated carbon.⁽⁹⁾ Smaller systems have been designed based on adsorption on activated carbon at temperatures near that of liquid nitrogen and at atmospheric pressure.^(7,10) Purge, or once-through inert gas systems, have been proposed. One system of this type is now in operation.⁽¹¹⁾ None of these methods were considered adaptable to the helium-purification system in Facility 350 because of its size. It was not considered large enough to justify the expense of the cryostatic purification method and yet too large to be dependent upon a liquid nitrogen or metered helium supply.

The purification method selected is based on the adsorption of oxygen on activated carbon at elevated pressures and at normal refrigeration temperatures. The work of Field and Wells,(12) and the prior work of Homfray,(13) gave support to the premise that this would be the best approach for the 15,000-ft³ system in Facility 350. Drying would be accomplished by molecular sieve material at elevated pressures and room temperature. The method chosen is not without certain disadvantages; the high-pressure system must be tight, and a certain amount of mechanical equipment is required.

IV. DESCRIPTION OF THE SYSTEM

The helium atmosphere system in Facility 350 is illustrated by the block diagram of Figure 1. For discussion purposes, it will be divided into two main sections - the recirculating system and the purification system. The bulk of the helium atmosphere is continuously recirculated through the ducts and gloveboxes by a seven-stage turboblower. About ten percent of the helium flow is through the purification system, consisting of the compressor, after cooler, drying towers, Freon heat exchanger, and carbon towers.

Fig. 1. Schematic of Helium Atmosphere System

glovebox, and some of the duct work and piping, including the discharge heat exchanger. The duct work and valves are arranged so the blowers can be operated individually, in series, or in parallel. Each blower has a capacity of 1,000 cfm at a maximum pressure differential of 17 in. of water. Two AEC high-efficiency filters (24 x 24 x 12 in.) in a bag-change filter box are provided in the helium atmosphere return (HAR) line from the gloveboxes.

The recirculating system is detailed in Figure 2. Two Hoffman seven-stage turboblowers, E-22 and E-23, are housed within a helium-ventilated glovebox (PF-27) to eliminate the need for shaft seals. Each blower is driven at 3600 rpm by a 15-hp motor. Figure 3 shows one of the blowers, a portion of the

The recirculating system is detailed in Figure 2. Two Hoffman seven-stage turboblowers, E-22 and E-23, are housed within a helium-ventilated glovebox (PF-27) to eliminate the need for shaft seals. Each blower is driven at 3600 rpm by a 15-hp motor. Figure 3 shows one of the blowers, a portion of the duct work and piping, including the discharge heat exchanger. The duct work and valves are arranged so the blowers can be operated individually, in series, or in parallel. Each blower has a capacity of 1,000 cfm at a maximum pressure differential of 17 in. of water. Two AEC high-efficiency filters (24 x 24 x 12 in.) in a bag-change filter box are provided in the helium atmosphere return (HAR) line from the

Fig. 2. Helium Atmosphere Recirculating System
(Drawing No. PF-27-104-4)

Fig. 3

PF-27 Glovebox and
Seven Stage Turboblower E-23

The purification system is illustrated in Figures 4 and 5. Helium is drawn from the recirculating system at a rate of approximately 120 cfm and compressed to 140 psig by a Gardner-Denver two-stage compressor. The heat of compression is removed by a chilled water intercooler and aftercooler, and the gas then enters one of the drying towers, which contains 91 lb of Linde Molecular Sieve (Type 4A). Figure 6 shows one of the drying towers and the chilled water condenser used during the regeneration cycle. The helium next passes through a direct-expansion, Freon 22 heat exchanger (Figure 5) which is capable of lowering the gas temperature to -46°C (-50°F). The helium is then passed through one of the purifying towers, or banks, containing National Carbon Brand "Columbia" activated carbon. Each bank is actually four towers in series with a total volume of 29 ft³. One of the carbon towers is shown in Figure 7, which was taken during installation before the enclosure chamber was packed with Styrofoam insulation. The exit gas from the carbon tower may be throttled back to the recirculating system by two routes; either direct or via the liquid nitrogen Dewar and the countercurrent heat exchangers mounted on the top of each carbon tower. This latter course has been proven ineffective as designed and it is little used.

Fig. 4. Helium Atmosphere Purification System. Part I - Drying
(Drawing No. PF-27-104-5)

- NOTES
- 1 K1 IS INTERLOCK TO PREVENT TWO STAGE COMPRESSOR E-16 FROM STARTING WITHOUT CKT-1 ENERGIZED SEE PF-27-104-4
 - 2 MOTOR CONTROL CKT. E-22 E-23 SEE PF-27-104-4
 - 3 FOR PNEUMATIC CONTROL SYSTEM SEE PF-27-104-7
 - 4 E-16 ON SVC IN CKT E-16 OFF SV-F IN CKT SEE PF-27-104-4
 - 5 FOR CKT 1 SEE PF-27-104-4
 - 6 FOR CHILLED WATER SYSTEM SEE PF-27-104-3

- ⊗ MANUAL VALVE ⊗ SOLENOID VALVE
⊗ THROTTLE VALVE ⊗ BARKSDALE PRESSURE SW.
⊗ RESISTANCE THERMOMETER BULB PF-27-103-5

FROM DRYING TOWERS
SEE PF-27-104-5

FREON 22 PRE-COOLER
NOTE 1

FROM GAS HEATER

TO RECIRCULATING SYS. SEE PF-27-104-4

TO SAMPLING SYS. (PURIFIER)
SEE PF-27-104-6

TO P.A.E.

TO P.A.E.

TANK VENT
TO H.A.R.

TANK VENT
TO H.A.R.

NOTE 1 FOR FREON LOW TEMP. SYS.
SEE PF-27-104-2

- MANUAL VALVE
- THROTTLE VALVE
- REGULATOR
- RESISTANCE THERMOMETER
PF-27-103-5

110 V CIRCUIT DIRECTORY

- CKT. 1 PURIFICATION SYSTEM SUPPLY AND RETURN VALVES (SV4) PF-27-104-4
- CKT. 2 BALLAST TANK PRESSURE CONTROL (SVF SV6) PF-27-104-5
- CKT. 3 EMERGENCY VACUUM CONTROL (SVB) PF-27-104-4
- CKT. 4 SYSTEM PRESSURE CONTROL (SVH SVI SV6) PF-27-104-5
- CKT. 5 PF-27 GLOVEBOX PRESSURE (SVC SVJ SVE) PF-27-104-4
- CKT. 6 RESISTANCE THERMOMETER SYSTEM PF-27-103-5
- CKT. 7 ANALYZER SYSTEM (CABINET) PF-27-104-3

Fig. 5. Helium Atmosphere Purification System. Part II - Oxygen Adsorption
(Drawing No. PF-27-104-6)

Fig. 6
Drying Tower E-30B, Valve Panel,
and Condenser

Fig. 7
Carbon Tower Bank E-31B and
Enclosure Chamber (Note heat
exchangers mounted above
each unit)

The Freon 22, direct-expansion heat exchanger is serviced by a low-temperature refrigeration system. A schematic piping diagram for the refrigeration system is shown in Figure 8. The complete system is detailed in Figure 9.

Fig. 8. Low-temperature Refrigeration System - Piping Diagram
(Drawing No. PF-27-104-10)

All requirements for cooling water are provided by a chilled water system consisting of a packaged chiller and a circulating system. A schematic piping diagram of the chilled water refrigeration system is shown in Figure 10. Details of the complete system are shown in Figure 11.

Conventional materials of construction were used throughout. The piping and ducts are of welded stainless steel. Standard O-ring flanged joints are common. Neoprene O-rings are used where temperature is not a problem. Teflon O-rings were originally specified in the purification (high-pressure) section. They failed under regeneration conditions and were replaced with a Silastic (silicone) material that has proven effective under all operating conditions.

Fig. 9. Low-temperature Refrigeration System
(Drawing No. PF-27-104-2)

Fig. 10. Chilled Water Refrigeration System - Piping Diagram
(Drawing No. PF-27-104-11)

Fig. 11. Chilled Water System
(Drawing No. PF-27-104-3)

On the larger pipes or ducts, Keystone rubber-lined butterfly valves are in service, operated either manually or by motor. On the high-pressure purification system, flanged Jamesbury ball valves are used. These valves are rated for vacuum service over the range of operating temperatures specified. Rockwood ball valves are used in other areas of the system.

V. OPERATION

Normal operation of the system requires that all components be in continuous service if the purity of the helium atmosphere is to be held at desired levels. The two turboblowers of the recirculating system permit uninterrupted operation. The blowers are used alternately for 7-10-day periods to provide for equal wear and maintenance. The parallel tower arrangement of the purification system permits one tower to be "on stream" while the other is being regenerated. Only one helium compressor is provided, and failure would disrupt purification procedures. No trouble has been experienced in this area, however, and planned shutdowns have been of short duration.

Under normal operating conditions drying tower life is from 7 to 12 days. Further use results in a moisture content above 10 ppm of the tower discharge gas, which will cause frosting of the Freon heat exchanger. At least one freshly regenerated carbon tower is used daily. Two may be used on Mondays and Fridays, as well as on days when a drying tower is being regenerated.

The purification system may be operated without the low-temperature refrigeration system in service. In this case, the 140-psig helium enters the carbon tower at room temperature rather than at -46°C (-50°F), and the oxygen absorption is about $\frac{1}{3}$ the value obtained with low-temperature operation. Stated in another way, one carbon tower at low temperature is equal to 3 towers at room temperature. However, three saturated, room-temperature towers can be changed in the same time period required for one low-temperature tower to approach thermal equilibrium.

The recirculating system may be operated independently of the purification system. The helium atmosphere will be constantly recirculated but the impurity content (oxygen, moisture, etc) will gradually increase, depending upon system leakage and production operations within the gloveboxes.

Drying Tower Regeneration

A drying tower is regenerated by heating the molecular sieve bed with a stream of preheated helium flowing in a reverse direction to that employed in the normal drying operation. Moisture is removed from the

saturated gas by passing it through a chilled water condenser. When water ceases to be collected in the condenser trap, regeneration has been achieved.

The gas stream leaving the second stage of the compressor is split before it passes through the aftercooler. Between 35-40 cfm of helium are used for the regeneration operation at near atmospheric pressure, with the remaining 80-85 cfm following the normal purification cycle. The gas heater (see Figure 4), raises the temperature of the helium to 190.5°C (375°F), and the gas, after passing through the saturated bed, is cooled by the condenser to about 10°C (50°F). After leaving the condenser, the helium re-enters the intake line of the helium compressor. When the temperature of the tower discharge gas reaches approximately 66°C (150°F), water begins to condense and be collected in the trap.

After regeneration has been completed, the tower is isolated, cooled to room temperature, gradually pressurized to 140 psig, and held in readiness for operation. About 15 ft³ of gas is taken from the system when the tower is pressurized. The same amount of gas is vented to the process air exhaust at the beginning of the cycle. Drying tower regeneration is accomplished in about 7 hr with from 500 to 800 cc of water being removed from the condenser trap. It should be noted that during regeneration less helium is being dried and there is a tendency for the system moisture content to gradually increase.

A plot of water content versus dew point for helium at various pressures⁽¹⁴⁾ is shown in Figure 12. Calculation indicates that less than $\frac{1}{3}$ of the moisture is removed from the system during regeneration. The 40 scfm of helium passing the regenerating tower and chilled water coil is saturated at 12°C (53°F). Since this moist gas, carrying approximately 700 ml of H₂O per hour, is returned to the compressor suction, the moisture is readsorbed in the in-service drying tower. If the saturated helium-recirculation period is 4 $\frac{1}{2}$ hr, approximately 3 liters of water are moved from one drying tower to the second while less than 1 liter is removed from the system. A liquid nitrogen freeze-out tray has been investigated to improve the effectiveness of the regeneration.

Carbon Tower Regeneration

The carbon towers are regenerated by evacuation. The pressure in the tower to be regenerated is released to the storage tank by means of a transfer pump (see Figure 5). Any remaining helium is vented to the process air exhaust, and the tower is evacuated by two Model 148 Stokes vacuum pumps to less than 10⁻¹ torr. Overnight evacuation is the usual procedure, after which the tower is restored to operating pressure (140 psig) and held in readiness for placing "on stream."

Fig. 12. Calculated Water Content of Helium vs Dewpoint of Gas

The transfer pump is a Bell and Gossett Model SYC-17-1, canned in a helium-tight, chilled water-cooled container. Helium consumption, during regeneration, can be directly related to the performance of this pump and the time allowed, or involved, in the transfer operation. If the pump is functioning properly, with piston rings and other components recently installed, an acceptable transfer, either way, can be accomplished in less than an hour. Little helium will be vented to the exhaust system and little will be taken from the storage tank on the refill. As the rings wear and the pump efficiency decreases, longer pumping time is required, and desired pressures cannot be attained. Under the worst conditions, 90-120 ft³ of helium may be lost from the system at each end of the transfer cycle for each regenerated carbon tower.

Helium Atmosphere Purity

A satisfactory oxygen level can be maintained on a one-shift operating basis. The purity of the helium supplied to the gloveboxes is held to less than 3,000 ppm (0.3%) nitrogen, 1,000 ppm (0.1%) oxygen, and less than 50 ppm water. Normal operating conditions generally give 1200-2100 ppm nitrogen, 400-700 ppm oxygen, and 20-40 ppm water. Lower levels could be maintained (with the exception of moisture) with around-the-clock operation.

Lowering of the system oxygen content can be accelerated by pressurizing the carbon tower, after evacuation, with fresh tank helium instead of storage tank helium, which may be of higher oxygen content. This method shortens the transfer pump cycle and will permit as many as four carbon tower changes per day, but only at the expense of increased helium consumption. This procedure was used during the initial startup of the system because of the oxygen level at that time.*

The usual oxygen-leakage rate from all causes (actual leaks, diffusion, etc) averages 10 ppm/hr. If this rate is consistently exceeded to any extent, detectable leaks exist. Minimum oxygen levels can be maintained only if all leaks are promptly located and sealed.

Helium Consumption

Accurate figures on helium consumption during normal operation of the helium atmosphere system are not available. One reason is that the helium supply (tank farm) to the facility is a common system serving all

*Air was expelled from the gloveboxes by large vinyl prefabricated bags that were blown up with tank helium. After purging the remaining volume, the bags were opened to the glovebox, collapsed, and removed. Oxygen levels were reduced to approximately 2% with much less helium than would be required to purge to this level.

building requirements. Some helium is used for welding, purging, and other production operations, and no finite separation has been attempted. The usage rate is decreasing gradually, but might average about 50-60 cylinders (11,000-14,000 ft³) per month. Based on the helium system operations that have been described, one might estimate the present average helium use for the system to be about 25 cylinders (5,950 ft³) per month, or about half the overall building helium usage.

Auxiliary Components

The chilled water system (see Figures 10 and 11) operates continuously. The compressor is loaded or unloaded automatically, as the demand upon the system varies. Under normal operating conditions, the temperature control range is about 2.3°C (4°F). This differential is measured on the return water line with the maximum and minimum temperatures being 5.6°C (42°F) and 3.3°C (38°F), respectively. If the demand, or heat load, drops markedly from normal operation conditions, the unit will automatically shut down and await a rise in water temperature before restarting. The unit has a minimum refrigerating capacity of 180,000 Btu/hr. Early difficulties experienced with this unit involved control problems that were easily remedied.

The low-temperature refrigeration system (see Figures 8 and 9), is somewhat unique, not necessarily from the requirement of continuous operation, but the manner in which it operates. There is no controlled unloading feature as employed on the chilled water system. The compressor operates at full load and the system must be carefully balanced throughout at all times if the temperature at the heat exchanger is to be accurately maintained. After a period of early operation under improper control conditions, the compressor failed and a complete overhauling and rebuilding was necessary. Since that time, little difficulty has been experienced if attention is given to proper operating temperatures, pressures, and maintenance procedures.

During drying tower regeneration, the reduction in helium flow rate across the Freon heat exchanger from 120 cfm to 85 cfm causes a marked change in load on the low-temperature refrigeration compressor. The load change is beyond the normal control range, and some adjustment of the expansion valves (see Figure 8) is often necessary. Thus, it is possible to keep the low-temperature refrigeration system in operation during the regeneration operation. This was not the case initially, but better operation of the compressor under any and all conditions has been obtained by lowering the speed from 955 to 745 rpm. This change raised the product (helium) temperature from -48°C (-55°F) to -46°C (-50°F), but permitted continuous operation without undue upset and manipulation. At the present operating speed the compressor has a refrigeration capacity of approximately 18,000 Btu/hr.

Corrective Measures

Satisfactory operation of the system could not be attained without replacement of some materials of construction. Teflon O-rings and seals subjected to alternate high- and low-temperature service failed after a few months of operation. These were satisfactorily replaced by a silicone rubber compound.

Graphite piston rings and seals did not perform satisfactorily in the compressors. The first set of graphite piston rings in the Gardner-Denver compressor failed after 300 hr of operation. The second set of graphite rings was completely worn out in 600 hr. Fiberglass-filled Teflon piston and wearing rings in aluminum pistons were substituted for the graphite. The compressor was overhauled at 3800 hr. The Teflon rings showed essentially no wear. A second set of rings lasted for 5800 hr. These were replaced when blow-by became evident. A third set of rings lasted for 7800 hr.

In July 1962, the compressor was overhauled because of bearing and cross-head wear. First- and second-stage pressures were maintaining at 38 and 150 psig, respectively. The volume efficiency of the compressor had dropped about 10 percent. Radial wear on first- and second-stage Teflon rings were found to be 0.010 and 0.009 in., respectively. These values were considered extremely low. It was believed that the rings would have given some months additional service.

Other corrective operations which were done in July 1962 included replacement of valve seal, repair of frost-damaged insulation, and replacement of the molecular sieve drying beds. The latter were powdered and heavily contaminated with oily graphite. The system was returned to service in August, 1962.

Based on the success of the Teflon piston rings in the Gardner-Denver compressor, they were also substituted for the carbon piston rings in the Bell and Gossett transfer pump. This has extended the ring life by several fold, but there are other factors here, such as improper cooling, that still plague the operation. This is one of the remaining areas of the system where modification and improvement is recommended and necessary.

Original plans called for drying tower regeneration with hot compressor gas. This method was tested. It did not prove effective because the actual gas temperature was lower than originally calculated and actual heat losses exceeded original assumptions. This condition was corrected by the addition of a gas heater (see Figure 4) of ample capacity to provide for completion of the regeneration cycle during a single day shift.

Corrective measures could be applied to the design of the cooling coil in the liquid nitrogen Dewar of the purification system (see Figure 5). Improved coil design could produce slightly lower temperatures of gas entering the carbon tower. Frosting of the coil would occur at liquid nitrogen temperatures, however, which could lead to additional operational problems.

VI. INSTRUMENTATION AND CONTROL

Pressure Control

The control of pressure in the gloveboxes is dependent upon a constant pressure in the helium atmosphere supply (HAS) and return (HAR) ducts. Three pneumatically operated, static-pressure controllers are used for this purpose, as indicated in Figures 13 and 2. One unit modulates the control valve, another the bypass valve, and the third regulates the amount of helium in the system.

The control and bypass valve instruments (D1 and D2) are set and adjusted to control the static pressure in the HAS and HAR ducts at +3 and -3 in. water gauge, respectively. Both valves will modulate, depending upon the number of helium atmosphere gloveboxes in service, holding the duct pressures constant.

Helium is added to or removed from the system by the volume controller (D3) in order to maintain a preset pressure in the helium atmosphere supply (HAS) duct just upstream of the control valve. This is done by the actuation of ballast tank solenoid valves (see Figure 4). The controller floats between 7.5 and 11.5 in. water gauge. As the low limit is approached, helium is transferred from the ballast tank to the supply (HAS) duct. As the upper limit is reached, helium is removed from the purification system (at 140 psig) and transferred into the ballast tank. Transfer of gas either way is stopped after the instrument senses a change of approximately 0.5 in. water gauge from either end of the control range.

When only the recirculation system is operating, gas can be transferred from the ballast tank to the supply (HAS) duct, but when gas must be removed, it is vented to the process air exhaust (PAE) since no high-pressure gas is available for transfer to the ballast tank. The pressure control range is the same in either case.

Each of the 14 glovebox lines, ranging from 220 to 2600 ft³ in volume, is equipped with a similar pneumatic controller which will open the glovebox to the exhaust system if the glovebox pressure rises to near atmospheric pressure. When and if this occurs, the HAS and HAR lines to the glovebox are automatically closed. The glovebox ventilation and control details have been reported,⁽¹⁵⁾ and will not be dealt with here.

Fig. 13. Pneumatic Control Systems
(Drawing No. PF-27-104-7)

Helium Flow

The flow of helium throughout the recirculating system and the gloveboxes is measured by an Alnor (Illinois Testing Laboratories) thermoanemometer system (see Fig. 14), consisting of a measuring bulb, or probe, and an indicator, the latter being either portable or panel mounted. These units are used only in the low-pressure system with the probe tip being centrally located in the pipe or duct. Probes are located in the HAR, HAS, bypass, and purifier return lines, as indicated in Figure 2. One probe is also located in the 5-in. supply line to each helium glovebox, thus permitting a measurement of flow through each box. This flow can be adjusted at the glovebox to provide between 4-15 atmospheres changes per hour, depending upon the process requirements within the line.

Chilled-water Flow

The flow of chilled water through all the various heat exchangers, cooling jackets, etc., is regulated pneumatically, as indicated in Figure 14. The system is straightforward and effective, with suitable provisions for manual regulation, if desired. Numerous directly reading thermometers are included in the system to establish and verify the setting of the controller.

Helium Temperature

A knowledge of the gas temperature throughout the purification system, as well as in some areas of the recirculation system, is an operational necessity. An Alnor resistance-thermometer system is used for this purpose. Probe-selector and -indicating instruments are panel mounted along with the three pneumatic controllers, previously described, on an instrument panel in Basement A, as shown in the photograph of Figure 15. Thirty resistance-thermometer probes located in various parts of the system are tied into this selector-indicator unit. A simplified circuit diagram of the system is shown in Figure 16. The indicator range is from 23.9°C (75°F) to 260°C (500°F) on the high scale and from 23.9°C (75°F) to -198.5°C (-325°F) on the low scale. No difficulties have been experienced with the system; only occasional maintenance is required.

Helium Atmosphere Purity

An oxygen analyzer and two hygrometers (moisture monitors) continuously analyze and record the oxygen and moisture contents of the helium atmosphere system. Two Beckman instruments, a model G-2 oxygen analyzer and a Model 179 hygrometer, are mounted in the sampling system instrument cabinet, as shown in Figure 15. A Consolidated Type 26-301 hygrometer is separately mounted. The sampling system

Fig. 14. Thermoanemometer System Wiring Diagram
(Drawing No. PF-27-104-9)

NOTE NO. 1
PROBE CONNECTIONS SAME AS FOR PORTABLE

HEATER CURRENT 266.7 MA

RESISTOR VALUES:
 $R_2 = 1.5 \Omega$
 $R_3 = 1.5 \Omega$
 $R_4 = 1.5 \Omega$
 $R_5 = 1.0 \Omega$
 $R_6 = 802.1 \Omega$
 $R_7 = 20 \Omega$ RHEO.
 $R_8 = 5.26 \Omega$
 $R_9 = 1.5 \Omega$
 $R_{10} = 3.0 \Omega$
 $R_{11} = 19.5 \Omega$
 $R_{12} = 22.5 \Omega$

Fig. 17. Analytical Sampling System
(Drawing No. PF-27-104-8)

ALL PIPING 1/4 OR 1/2 NOM.
COPPER TUBING

The oxygen analyzer measures the magnetic susceptibility of oxygen as an impurity in the helium atmosphere. The heart of the instrument is a metal-coated glass dumbbell, suspended on a thin quartz fiber in a magnetic field. A change in position of the dumbbell results from a change in the magnetic field, which is directly related to the amount of oxygen present in the sample gas passing through the test chamber. Impurities other than oxygen have little or no effect upon the operation of the instrument.

Calibration of the oxygen analyzer is carried out with tank helium and the span cell. (The span cell is an electrolytic cell in a dilute KOH solution). The unit is zeroed daily with tank helium. The span, or full-scale calibration, is carried out quarterly. A gas sample is analyzed by mass spectrometry periodically to check the accuracy of the instrument and to determine if any buildup of nonmonitored impurities is occurring within the system. Since the activated carbon will remove these impurities in varying degrees with respect to oxygen, the oxygen level can be used as a basis of comparison. Nitrogen is the largest of all impurities, and the ratio of nitrogen to oxygen is generally between 3 and $3\frac{1}{2}$. Argon is used for welding operations within certain gloveboxes, and the ratio of argon to oxygen sometimes reaches one. Hydrogen, carbon dioxide, and some hydrocarbons (alcohols and acetone) are usually found in trace quantities.

The principle of operation of the Beckman Model 179 hygrometer is based on the absorption of water from the sample gas by means of a film of phosphoric acid within the cell. Electric current produced by dissociation of this absorbed water within the cell is a measure of the water concentration in ppm (by volume). The unit was calibrated before installation.⁽¹⁶⁾ It is checked periodically with tank helium, providing an order of magnitude calibration. Actually, frequent calibration has been found unnecessary because of a built-in feature that provides a constant check; moisture content above 10 ppm will result in frosting of the Freon heat exchanger. This condition is easily detected by monitoring the exchanger discharge gas temperature - a rise in temperature indicates frosting. The Consolidated Type 26-301 hygrometer operates on the same principle as the Beckman, but it is less elaborate in structure. The calibration of this unit has been checked against the Beckman with good agreement.

Much experience in sampling and gas analysis has been gained in working with this system. The sampling systems were revised numerous times. All flowmeters had to be calibrated for helium, all lines had to be thoroughly dried, and all "in line" materials that could absorb and hold moisture had to be either removed or placed on the downstream side of the monitors. Some early pressure difficulties were experienced, and the sampling system for the Beckman instruments had to be modified to separate the two analytical system (O_2 and H_2O) as much as possible. Above all, there must be no leaks in the sampling system.

Care is required in the use and maintenance of gas-analysis instrumentation of this type if long, trouble-free service is to be obtained. The suspension in the oxygen analyzer is easily damaged and the replacement cost is high. The electrolytic cells in the hygrometers are somewhat less expensive and more rugged. However, they also can be made unservicable through improper operation. Exposure of the cell to moisture slugs or to air by leakage or otherwise should be avoided. Procedures for cell regeneration have been only partially successful. The best service from both the oxygen analyzer and the moisture monitors will be obtained from steady-state conditions. Service life will be shortened drastically by sudden or erratic changes in pressure, flow, or oxygen and moisture contents.

Electrical Controls

Numerous electrical safeguards have been incorporated within the system to handle breakdown of mechanical equipment and power failure. Most of these interlocks are shown on the figure describing the particular system, such as interlock K-1 on Figure 4.

The two-stage Gardner-Denver Compressor (E-16) cannot be started unless one of the turboblowers (E-22 or E-23) and the chilled-water pump (E-4) are in operation. Failure of either of the latter units will shut down the compressor and isolate the purification system from the recirculating system. The low-temperature refrigeration compressor (E-12) is interlocked with the two-stage compressor (E-16). Failure of the compressor (E-16) will automatically shut down the refrigeration compressor (E-12).

Only the emergency vacuum pump (E-19) (see Figure 2) and the 110-v control circuits (see Figure 5) are on emergency power. Consequently, in a complete loss of electrical power little equipment would be operable, and a loss of control of the system would no doubt be the end result. Loss of power has not been experienced since a second primary feeder was brought into the facility during the early part of 1961. This double power supply is further supplemented by a 250-KVA diesel generator which will provide minimum power during the emergency. Automatic switch gear handles the changeover from the primary to the secondary feeder, and finally to the automatic startup of the diesel generator and switchover to the building emergency circuits. Momentary power interruptions (of less than 15-sec duration) are handled, in most cases, by time-delay units in the motor starter coil control circuits.

VII. CONCLUSIONS

Operations over a period of two years with the Facility 350 helium atmosphere system has confirmed the choice of helium as the inert gas. It has also strengthened the opinion that an inert atmosphere is one of the most effective means of dealing with the problems of oxidation and pyrophoricity associated with the fabrication of plutonium-bearing fuel materials. Large-scale operations of all types have been carried out in the helium atmosphere with no problems or difficulties.

The purification system is straightforward, incorporating molecular sieve drying towers and activated carbon towers to remove moisture, oxygen, and other impurities. The system is mechanical rather than chemical in nature, with little room for error once satisfactory operational methods are established. System operation has become routine and has required limited supervision and maintenance. Oxygen, nitrogen, and other contaminant gases may be removed by adsorption on the activated carbon at room or at lower temperatures, well above that of liquid nitrogen. Tower saturation is easily detected, so that operation and regeneration procedures are simplified. Acceptable levels of oxygen (<1000 ppm) and moisture (<50 ppm) have been attained with the purification system operating on a single-shift basis. This is accomplished with only about ten percent of the total gas volume passing through the purification loop. Lower oxygen levels can be attained, if desired, with around-the-clock operation.

The design of the overall system is sound and it is versatile in operation. Recommended improvements are few. The total helium consumption could be lowered by an improvement in the gas-transfer technique (carbon tower to storage tank, and vice versa).

Some thought has been given to eliminating the low-temperature refrigeration system and cooling the gas by expansion through a special compressor. This change might have some merit but it will require considerable study and development before being applied to this or any helium atmosphere system.

It is the authors' hope that this report will contribute much to the engineering technology of helium-atmosphere systems - a technology that has been handicapped by a dearth of published information on purification methods utilizing activated carbon. Widespread useage of large-scale helium-atmosphere systems cannot be expected until this "knowledge barrier" is removed.

VIII. ACKNOWLEDGEMENTS

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The architectural engineering firm of Shaw, Metz and Dolio was responsible for the design of certain portions of the helium-atmosphere system. Installation was by the Northwestern Heating and Plumbing Company of Evanston, Illinois, The Bloomer Heating and Ventilating Company, and The Midwest Machinery Movers of Chicago, Illinois. The work was carried out under the direction of F. G. Foote, with E. S. Payne serving as Project Superintendent.

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X. APPENDICES

Appendix I

OPERATIONAL DATA

The purification system for the Facility 350 helium atmosphere operates continuously to provide an atmosphere of sufficient purity to perform the required "in hood" fabrication operations. The moisture level is monitored both in the helium-atmosphere return line (HAR) and at the drying tower exhaust. The oxygen level is monitored in the helium-atmosphere supply line (HAS) and can be monitored at the carbon tower exhaust if so desired. Each monitored point has specific significance to certain phases of the operation. The drying tower exhaust is used to determine when a tower must be regenerated. The carbon towers are changed on a routine basis, with the number of changes being governed by the HAS oxygen level. Any internal moisture leaks of water-cooled equipment within the glovebox system quickly appear as a rise in moisture level in the HAR line. External leaks into the helium-atmosphere system are first observed as a rise in oxygen level. The instrumentation is sensitive enough to detect a system leak due to a small nick in a glove or a plastic bag. Typical charts are included to show the characteristic behaviors at the various points.

Figure 18 shows two on-stream cycles for the drying tower E-30A. The most significant portion of each curve is the rapidly rising value near the end of the cycle. The limiting factor in drying tower life is the dew point value at the Freon heat exchanger. Excessive frosting, which necessitates a shutdown of the refrigeration system for defrosting purposes, will occur near 10 ppm moisture. In order to operate below this value, the drying towers are changed at the discretion of the operator rather than on a routine basis. Curve A illustrates this point. The rapid rise to

Fig. 18. Moisture in Helium Leaving
Drying Tower E-30A

5.4 ppm, which occurred on a Friday, indicated that frosting would probably occur over the weekend; therefore the tower was taken off stream. Curve B is more typical of normal operation. It also illustrates the importance of correct instrumentation when working with low moisture values. The tower was put on stream late Friday afternoon, and a value of 3.5 ppm was noted. This value was held until Monday morning, at which time a bypass around the moisture monitor was opened. The increased flow through the sampling line dries the line and gives faster reading response. The value quickly fell to 2.5 ppm and held below 3 ppm until Friday. However, over the weekend a breakthrough occurred, as shown by the rapidly rising tail of the curve. All drying tower cycles follow a similar pattern, having this characteristic rise near the end of the cycle.

The moisture content of the helium atmosphere is more dependent upon the operations being performed within the glovebox system than it is upon the variation of moisture content leaving the drying tower. From the beginning to the end of its 7 to 10 days on stream cycle, the variation of drying tower output is about 6 ppm, whereas the daily variation resulting from operations within the glovebox system is sometimes as much as 40 ppm. Any unexpected rise in moisture level is investigated to determine its source. An alarm system is used to detect any marked increase during the off-shift hours.

Figure 19 shows both the HAS oxygen value and that of the carbon tower E-31B discharge oxygen. This figure illustrates the two phases of the useful life of the tower. Prior to being put on stream, the tower had been regenerated by overnight evacuation, which allowed it to approach

Fig. 19. Oxygen Adsorption Curve for Carbon Tower E-31B

room temperature. Initially, oxygen is removed at a high rate until the tower approaches saturation. This phase lasts from 10 to 20 min, depending upon the regeneration previously achieved and the impurity level in the helium. The second phase is the result of refrigeration and lasts about 12 hr. Because the adsorptive capacity of carbon increases with decreasing temperature, impurities will continue to be adsorbed until thermal equilibrium has been achieved. The rate of removal during this time is low

and approximately balances the total oxygen leakage into the glovebox system. The figure shows a carbon tower discharge value higher than the HAS value for a short period of time. This could be due to an overdriving of the instrument or to other reasons which are not presently known. The dashed line indicates the probable course of the curve.

V = TOTAL SYSTEM VOLUME IN ft^3
 L = LEAK RATE OF MOISTURE IN ppm/hr .
 C = MOISTURE CONCENTRATION IN ppm .
 C_1 = MOISTURE CONCENTRATION ON EXHAUST SIDE IN ppm .
 R_1 = FLOW RATE THROUGH PURIFICATION LOOP IN ft^3/hr .
 R_2 = FLOW RATE THROUGH RECIRCULATION LOOP IN ft^3/hr .
 θ = TIME IN HOURS.
 C_0 = SYSTEM MOISTURE CONCENTRATION AT TIME ZERO.

Fig. 20. Helium System Model and Terms for Mathematical Treatment of Moisture Removal

The moisture leak rate for the whole system is LV and the removal rate is $(C - C_1) R_1$. The change in moisture level with time is the difference in these rates or

$$\frac{dC}{d\theta} V = LV - (C - C_1) R_1 \quad (1)$$

By assuming ideal mixing and that L and C_1 are constant with time, one can solve the equation to give

$$C = K e^{-R_1 \theta / V} + [C_1 + (LV/R_1)] \quad (2)$$

At time zero, $C = C_0$, and therefore the constant K can be evaluated as

$$K = C_0 [C_1 + (LV/R_1)] \quad (3)$$

or

$$C = \left(C_0 - [C_1 + (LV/R_1)] \right) e^{-R_1 \theta / V} + [C_1 + (LV/R_1)] \quad (4)$$

By defining

$$K_1 = C_1 + (LV/R_1)$$

and

$$K_2 = R_1/V \quad ,$$

we can rewrite equation (4) as

$$C = (C_0 - K_1) e^{-K_2\theta} + K_1 \quad . \quad (5)$$

Equation (5) will describe the system moisture based upon the above assumptions. The ratio of R_2 to R_1 of approximately 8 to 1 should provide ideal mixing. The leak rate L is reasonably constant during nonworking hours, but may vary slightly during the working day. Based upon actual measurements, C_1 varies but little over long periods of normal operation. From Figure 18, a rise is noted when the drying tower becomes saturated.

A system drying curve based on data taken shortly after the addition of several hood lines is shown in Figure 21. The curve has been fitted to equation (4) by least squares, and the moisture concentration is represented by

$$C = 720 e^{-1.203\theta} + 170 \quad .$$

For long periods of time, θ approaches ∞ and C approaches K_1 , the minimum moisture value attainable.

Fig. 21. System Moisture Removal under Transient Conditions

There appears to be good agreement between calculated and actual minimum values attainable under normal system operating conditions. A value of 18 ppm moisture can be calculated from the following data:

$$V = 10,730 \text{ ft}^3;$$

$$R_1 = 7,200 \text{ ft}^3/\text{hr};$$

$$C_1 = 7.3 \text{ ppm};$$

$$L \approx 7 \text{ ppm/hr}.$$

Values for V and R_1 are by measurement, and the C_1 value is obtained from the moisture analyzer. The moisture leak rate L is an average value based upon an overnight moisture rise with the purification loop shut off and all glove port covers in place. When the covers are removed and glovebox operations are in progress, the value for L will increase. Doubling L would give a value of 30 ppm for K_1 . The values of 18 ppm and 30 ppm compare very favorably with the measured values for the system (HAR) under normal operating conditions. These are the minimum moisture values attainable from the system.

An equation for system oxygen values cannot be derived until the variation of C_1 with time is known. The behavior can be divided into three periods as shown in Figure 19: the initial adsorption resulting from regeneration, the adsorption resulting from cooling, and the transition period between these two. However, the length of time which each condition exists varies from cycle to cycle so no correlation has been attempted.

Appendix II

ADSORPTION DATA

The purification system in Facility 350 was designed primarily for the removal of specific impurities from helium, namely, oxygen, nitrogen, and moisture. The design of the carbon towers was based upon the limited data of Field and Wells,⁽¹²⁾ which did not apply directly to the operating conditions of this system. Adsorption data as a function of a single pressure can be obtained from the Facility 350 system, but the temperature dependence would be difficult to determine. In order to investigate the temperature effects, an experimental adsorption apparatus was designed, constructed, and connected to the helium-atmosphere supply.

A schematic diagram of the adsorption apparatus is shown in Figure 22. A copper "U" tube containing 147 gm of National Carbon brand "Columbia" activated charcoal was used for the experiments near room temperature, and a stainless steel "U" tube containing 42 gm was used for the low-temperature experiments. The carbon is the same material used in the carbon towers of the purification system in Facility 350. The bulk density of the carbon was measured as 0.460 gm/cc. Both "U" tubes were

Fig. 22. Schematic Diagram of Adsorption Test Apparatus

constructed from 1-in. tubing, 12 in. in height and $5\frac{1}{2}$ -in. in width. They were sized to fit into a 4300-cc Dewar, which was used as a constant-temperature bath. Various mixtures of methanol, water, and dry ice were used to maintain the desired temperature. Regeneration of the beds was accomplished by evacuation. The flow rate for an experimental run was measured with two rotameters operating in parallel; both had been calibrated previously with helium. One was used as a bypass and the other fed directly into the Beckman Model G-2 oxygen analyzer which was used to determine the oxygen level in the effluent gas. The experimental system was leak checked with a mass spectrometer leak detector before any data were taken.

Each experimental run was started by first evacuating the "U" tube at room temperature to a pressure $< 10^{-2}$ torr. The charcoal was then precooled for a minimum of 2 hr. The bypass was opened, and a portion of the piping was purged until the purge gas gave the same oxygen value as the HAS gas. During this time, the gas pressure and flow rates were regulated to give the desired values. The "U" tube was pressurized, and an experimental run was then started by simultaneously closing the bypass valve and opening the exhaust valve on the "U" tube.

An experimental run consisted of allowing the carbon bed to adsorb until it became saturated. This was indicated by a return of the oxygen level to the original HAS value. The results of a typical run are shown in Figure 23. The oxygen level initially made a rapid drop to a

Fig. 23. Typical Experimental Run with Adsorption Test Apparatus

point near zero; then, as the carbon became saturated, the level returned to the original value. The area above the curve and below the HAS value is directly related to the amount of oxygen adsorbed. For example, in Figure 23 the area was graphically integrated to give a value of 6350 ppm-min, and the total flow rate was 965 cc/min. Therefore,

$$\text{Volume adsorbed} = \frac{(965)(6350)}{10^6} = 6.13 \text{ cc O}_2$$

Since this "U" tube contained 319 cc of carbon, the adsorption ratio was 0.0192 for the conditions of -20°C and an oxygen partial pressure of 0.02 psia.

The maximum pressure available was 150 psig, and the oxygen content of the helium ranged between 800 ppm and 1500 ppm. Consequently, the area of investigation ranged between 0.02 psia and 0.25 psia for the partial pressure of oxygen. The results of experiments at +23°C, -20°C, -50°C and -72°C are presented in Figure 24. The data were fitted to an equation

Fig. 24
Adsorption of Oxygen
upon Activated Carbon

of the type $x/m = kp^{1/n}$, as suggested in McBain,⁽¹⁷⁾ where p is the pressure of the gas being adsorbed, x is the weight of gas adsorbed upon m grams of solid, and k and n are constants. In this case x and m were replaced by the ratios of the volumes of the respective materials. The following values for k and n were found at the temperatures investigated:

Temperature, °C	n	k
+23	1.01	0.309
-20	1.03	0.771
-50	1.02	1.869
-72	1.06	5.58

The value of n is essentially constant over the range of temperatures investigated, and the variation is not larger than the experimental errors. Since the values for n are essentially unity, the oxygen adsorbed was proportional to its partial pressure. The values for k are the extrapolated values at the various temperatures for a partial pressure of one psia.

Figure 25 shows the adsorption as a function of temperature for four different partial pressures. The most significant observation which

Fig. 25
Oxygen Adsorption
as a Function of
Temperature

can be made from the data is the marked increase in adsorption at the lower temperatures. Although adsorption of a gas will increase with decreasing temperature, the rate at which the oxygen adsorption increased became much larger at temperatures below -40°C .

The data should prove useful in the design of helium-purification systems in which air is the main contaminant. However, the data have some restrictions which must be recognized before applying it to another system. The results are dependent upon the type of carbon used and its previous history. The values represent the amount of oxygen adsorbed by "Columbia" activated carbon after regeneration by vacuum (10^{-2} torr) at room temperature when the impurities are nitrogen and oxygen. The residual adsorbed gas can vary appreciably with different conditions of regeneration.

The supply helium contained oxygen, moisture, and nitrogen (and in some cases argon) as contaminants. All components are adsorbed to some extent depending upon the partial pressure of each component and the selectivity of the charcoal at a given set of conditions. Both the individual and total amounts of gas which carbon can adsorb from a gas mixture will be different than for the same conditions with a pure gas. The data are intended to be of use in the removal of air from helium, and the small amount of helium adsorbed is neglected. The supply gas always contained less than 10 ppm moisture, and its effect is not significant. Both nitrogen and oxygen were adsorbed, but no attempt was made to measure the nitrogen value. The measured oxygen values would be somewhat different if no nitrogen were present. However, most practical applications will involve both nitrogen and oxygen as contaminants, and the data can be applied directly.

No kinetic factors were investigated and neither was the surface effect of the containing vessel. These factors should be considered in the design of a small system requiring a helium product of high purity.

The more important design considerations which must be evaluated are the desired purity level, the identity of the contaminants, the operating temperature and pressure, and the regeneration conditions. The data presented will aid in selecting the optimum operating temperature and pressure once an acceptable impurity level has been determined. Since the oxygen was adsorbed proportionally to its partial pressure, increasing the total pressure increases the amount of oxygen removed. The limiting factor therefore becomes the maximum pressure which can be conveniently attained.

The marked increase in adsorption at low temperatures suggests operating at as low a temperature as possible. However, several factors

cannot be overlooked in low-temperature operation. Extremely dry gases are necessary or frosting will become a serious problem, especially when temperatures near that of liquid nitrogen are considered. Low-temperature refrigeration systems require a large amount of equipment which is usually specially designed for the purpose. Low-temperature operation is usually accompanied with elevated-temperature regeneration, since much of the gain would otherwise be nullified. Therefore, the heat transfer aspects must be critically examined. Over a long period of time the removal by low-temperature operation may be equalled by increasing the size of the carbon bed and operating at near room temperature, thus increasing the number of cycles possible during this time.