RESEARCH FACILITY FOR THE SYNTHESIS AND FABRICATION OF REFRACTORY PLUTONIUM MATERIALS

F. A. SAULINO, J. C. ANDERSEN
AND K. M. TAYLOR

The Carborundum Company
Research and Development Division
Niagara Falls, New York

This paper describes a facility for studying the synthesis and fabrication of refractory plutonium materials. The outstanding features of the facility are its compactness, reliability, low operating cost and the unusually high purity of the atmosphere in the helium glove boxes (2-3 ppm oxygen and less than 1 ppm water vapor). The high purity helium atmosphere results from the leak tightness of the system and the highly effective zirconium-titanium alloy getter system. In addition to the usual health and safety precautions, possible trouble areas are continuously monitored by an extensive alarm system.

INTRODUCTION

In 1960 The Carborundum Company completed a facility* for investigating the synthesis and fabrication of refractory materials containing plutonium in such forms as carbides, silicides, oxides and nitrides. These materials are extremely toxic, and some are quite sensitive to oxygen and moisture, especially in a finely divided state; therefore, a leak-tight glove box system having a carefully controlled inert atmosphere was required. The necessity for a highly inert atmosphere was further emphasized by the decision to fabricate mainly by sintering at atmospheric pressure rather than in vacuum. In addition, it was desirable that the facility have a low operating cost and be protected by adequate alarm and safety devices, especially since it was planned to work on a one-shift, five-day week basis with the facility in continuous operation.

GENERAL DESCRIPTION OF FACILITY

The facility is located in the Central Laboratory of The Carborundum Company at Niagara Falls, New York, and is essentially a newly constructed room in an existing reinforced concrete building. The principal unit is 15 feet wide, 48 feet long and 8-1/2 feet high, with enameled steel paneled walls and ceiling and vinyl floor covering. This area is divided into a change room, 11 by 15 feet, and a work space, 15 by 37 feet. In addition, a helium purification and recirculation system servicing the facility occupies a 12 by 24 foot space in an adjacent room. The general layout of the facility is indicated in Fig. 1, while Fig. 2 and Fig. 3 are photographs of a portion of the work area and helium purification and recirculation equipment respectively.

*The glove boxes and equipment contained therein were financed by the United States Atomic Energy Commission.

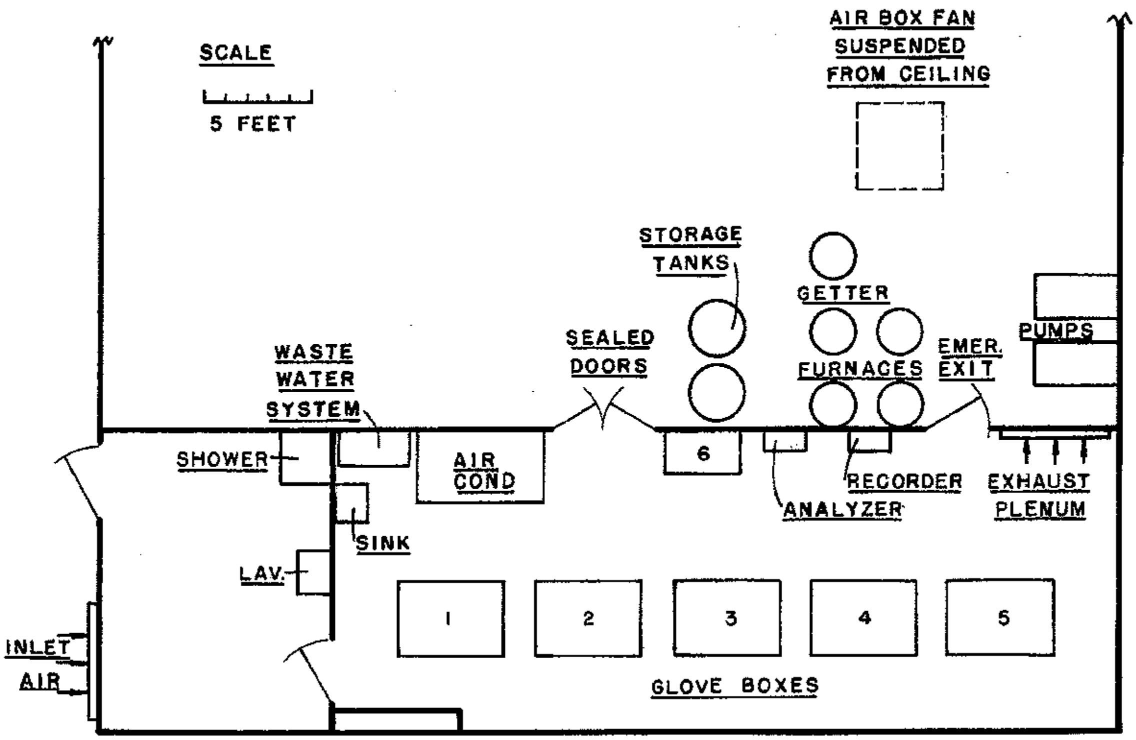

Fig. 1. Area layout

Fig. 2. Portion of work area showing glove-box arrangement

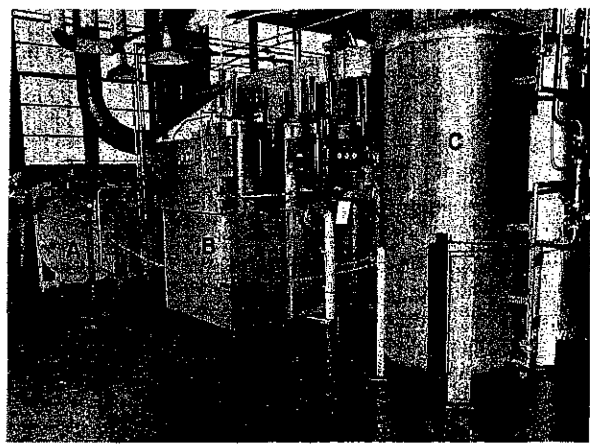

Fig. 3. Helium purification area showing helium pumps (A) getter furnaces (B) and Helium storage tanks (C)

atm
ave
alu
sys
box
Ma
or
low

per

atn
sch

par
pun
vac
pun

con
let
blo

eva
coo
aut

hig
thr
col
ten

ind
col
The
gas

thr
all
get
ten

cor

Of the glove boxes indicated in Fig. 1, Numbers 1, 2, and 6 operate with an air atmosphere and Numbers 3, 4, and 5 with high purity helium. The glove boxes have an average size of 3 feet high, 3-1/2 feet wide and 5 feet long and are constructed of welded aluminum plate with full-side safety plate glass windows. The air and helium glove box systems are connected by a vacuum lock located between boxes 2 and 3, while the remaining boxes, with the exception of No. 6, are connected by two-layered flexible plastic tubing. Materials may be moved into or out of the box system by the well-known pouching technique, or directly through the O-ring sealed sliding door of No. 1 air box which is maintained at a low level of contamination.

The change room contains a shower, lavatory, lockers, storage cabinets, personnel monitoring equipment, and an alpha counting apparatus.

DESCRIPTION OF COMPONENT SYSTEMS

Helium Glove Box System. Glove boxes 3, 4, and 5 are helium boxes and the atmosphere is supplied by a helium recirculation and purification system shown schematically in Fig. 4.

The helium from the glove boxes is circulated by two blowers operating in parallel. One blower acts as a standby to be used when servicing or repairing the other pump. These blowers, with their drive motors, are individually mounted in separate vacuum-tight tanks that have an independent helium atmosphere so that any leakage at the pump seals introduces helium rather than air.

The pump inlet suction pressure is maintained at minus 1/2 psi by a manual flow control by-pass valve which regulates the amount of helium that is by-passed from the outlet manifold, back into the inlet manifold and recirculated through the pump. Gas leaves the blower at approximately 4 psi.

Each pump tank is connected to a common system evacuation manifold for evacuation after servicing. Since the pumps are enclosed in gas-tight tanks, they are water cooled to prevent overheating. Should the operating pump fail, a change-over switch will automatically start the stand-by pump to permit continuous operation of the system.

Helium at a pressure of approximately 4 psi is purified by passage through four high temperature (1650°F) zirconium - 50 atomic % titanium chip getter columns and then through a low temperature (700°F) zirconium-titanium column. The high temperature columns remove such impurities as nitrogen, oxygen and hydrocarbons, while the low temperature column removes hydrogen.

The four high temperature getter columns are employed in parallel, each being individually valved and instrumented to allow independent operation or servicing. Each column is instrumented with isolation valves, a manual flow control valve and a rotameter. The high temperature furnaces are also equipped with a preheater which serves to heat the gas before entering the main furnace and permits more efficient use of the getter column.

Only a portion of the gas from the four high temperature columns is passed through the low temperature column which removes hydrogen. A manual by-pass valve allows the major portion of the gas (approximately 80%) to by-pass the low temperature getter while the remaining 20% passes through the furnace which is similar to the high temperature getters in construction and valving but has no preheater.

Each of the columns is equipped with a Sigma magnetic amplifier temperature control unit for regulating the getter column temperature. In addition, a similar separate

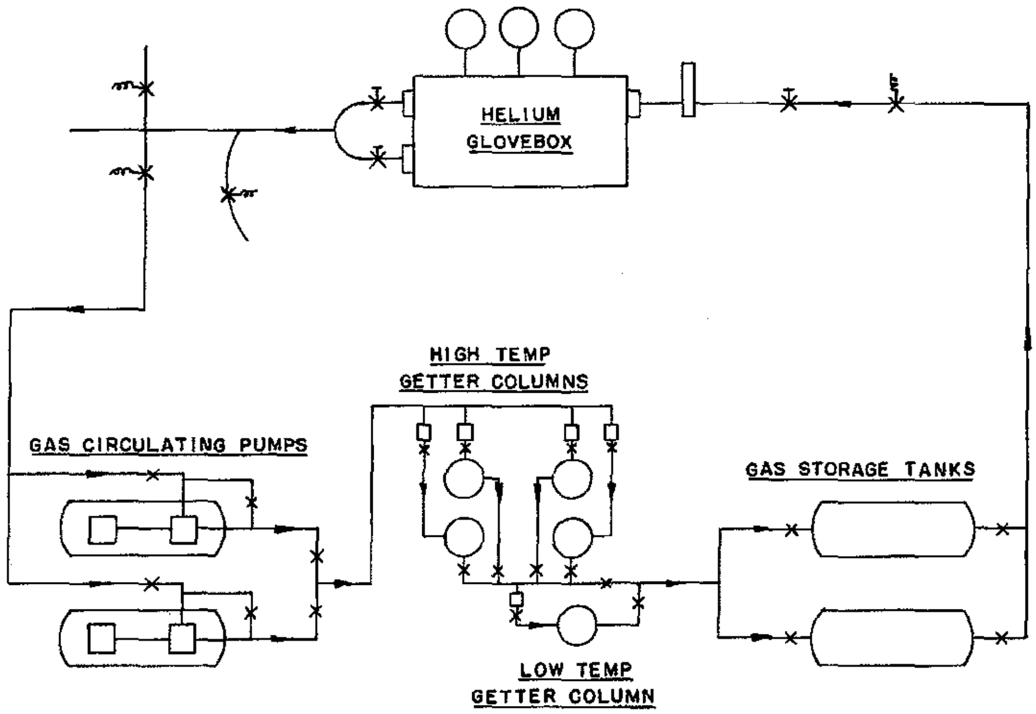

Fig. 4. Helium glove-box system

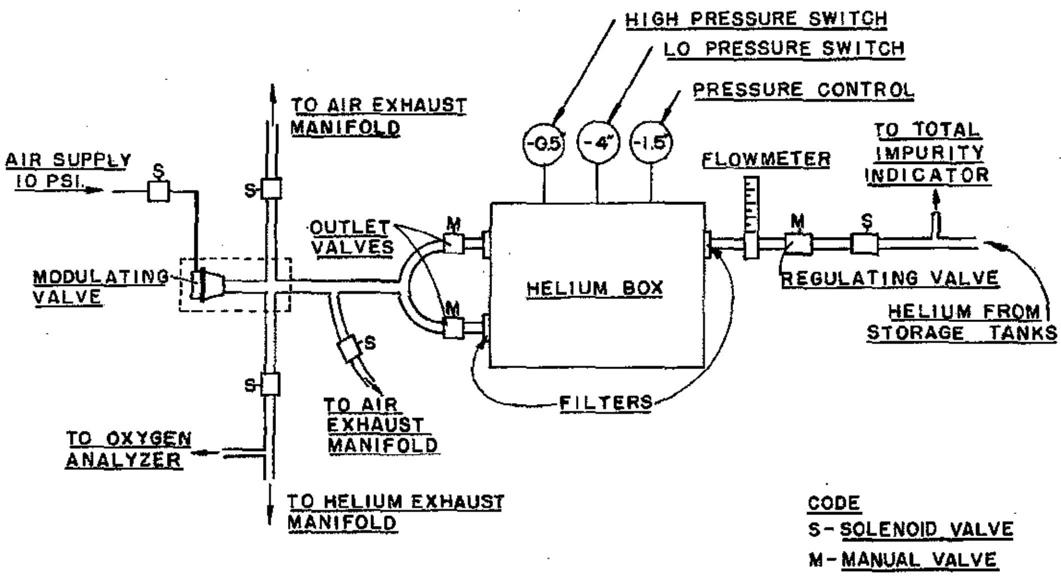

Fig. 5. Helium glove box and controls

temperature control unit, set approximately 50°F higher, provides emergency shut-off in case of column overheating. Each of the columns is also provided with a valved evacuation manifold to allow separate column evacuation.

The gas leaving the getter furnace is stored in two aluminum storage tanks, each having a volume of approximately 50 cubic feet. The pressure of the gas in the storage tanks is regulated between 14 and 15 ounces per square inch by two pressure switches mounted on the tank exit line, at which point gas is either added or bled from the system to maintain the required pressure. The tanks are provided with vacuum-tight ball valves to isolate the tanks from the system. A third ball valve leads to the evacuation manifold to allow evacuation of the tanks. A pressure gauge indicates the storage tank pressure.

The gas from the storage tanks is manifolded from the tanks to each of the three helium boxes.

The helium glove boxes are operated at a pressure of about minus 1-1/2 inches of water with respect to room pressure. Normal flow through the helium boxes is shown in Fig. 5. All components in the figure, with the exception of the oxygen analyzer and total impurity indicator, are individual for each helium box. One or more of the boxes can be isolated from the system without disturbing the operation of the remaining boxes.

During normal operation, the gas leaves the storage tanks at approximately 1 psi and flows through an inlet solenoid valve, regulating valve, flowmeter, inlet filter, glove box, outlet filters, manual outlet valves, modulating valve, solenoid valve and then to the helium exhaust manifold which returns the gas to the pump. Pump suction pressure is maintained at minus 1/2 psi.

During normal operation, the inlet solenoid and outlet solenoid valves are open. The inlet flow is constant at 4 cfm while the exit flow is throttled by the modulating valve which continuously opens or closes to increase or decrease the flow from the box.

The modulating valve consists of a 2-way valve, the stem of which is connected to a spring loaded actuator. The spring tends to pick up the valve stem and consequently opens the valve; however, injecting air at 10 psi into the actuator overcomes the spring force and closes the valve.

The modulating valve is operated and controlled by a 3-way solenoid valve. The control signal for this solenoid valve comes from the pressure control switch, one of three pressure switches mounted on the box. The switches compare box pressure with room pressure.

Operation of the control system is as follows: As the box pressure decreases below the control switch set point (minus 1-1/2 inches of water), the switch contacts close, energizing the air supply solenoid valve, which opens and allows 10 psi control air to enter the modulating valve actuator, closing the modulating valve. With the exit side of the box closed and the inlet flow of gas constant, the box pressure will increase. As the box pressure rises above the switch set point, the switch contacts are opened and the air supply solenoid valve is de-energized. This shuts off the control air and simultaneously bleeds the air from the actuator to the atmosphere and the actuator spring forces the modulating valve stem up, opening the valve and permitting the gas to flow out of the box. The exit flow continues until the box pressure falls and the above steps are repeated.

The high and low pressure switches serve as protection from excessive pressure or excessive suction should the control system fail.

Should the pressure in the box rise to minus 1/2 inch of water which is the setting of the high pressure switch, the following events occur: the inlet solenoid valve closes shutting off the inlet flow of helium; the exit solenoid valve also closes, isolating the box from the rest of the helium system; a large solenoid valve in a line leading to the air exhaust manifold opens which will evacuate the box until the high pressure switch opens and the box attempts to operate normally. The box is exhausted to the air exhaust manifold because of the possibility of the increase in box pressure being caused by a leak in the box, in which case the purification system would be contaminated with air.

If the pressure in the box becomes abnormally low (below minus 4 inches of water), the low pressure switch closes; this de-energizes and closes the helium outlet solenoid valve. With this valve closed and a constant inlet flow of helium, the box pressure rises until the low pressure switch opens, whereupon the normal box control system takes over.

Each helium box also has an oil-filled manometer which is connected to the same line as the pressure switches. The manometer serves a dual function. First, it provides a visual indication of the pressure in the box; and, secondly, it provides for emergency relief in case the box is over or under pressured and there is a failure in the ordinary protection system. The manometers will release at approximately $\pm$ 6 inches of water.

The purity of the helium in the system is continuously monitored by two devices. The first is a Beckman model 80 oxygen trace analyzer. The analyzer is capable of measuring oxygen content from 0-1000 ppm. A sample of the helium from the system is taken at a point between the pump tanks and getter furnaces at approximately 80 ml per minute and is vented into box No. 6 and thence to the air exhaust line. A second sample line is available at the inlet to the storage tanks which makes it possible to analyze the gas before and after the getter furnaces. Oxygen contamination within the system is normally 2-3 ppm. The analyzer is electrically connected to the helium glove box circuit. Should the oxygen impurities in the helium reach 100 ppm, the helium boxes are isolated from the helium purification system by the solenoid valves and are simultaneously vented to the air exhaust manifold. This is to protect the purification system against gross oxygen and nitrogen impurities due to such incidents as leaks in the boxes (ruptured gloves, broken glass, etc.).

A total impurity indicator also monitors the gas for purity. Briefly it works as follows: A high DC voltage is placed across a gap through which a system gas sample flows. Any impurities in the gas will result in a change in current flow across the gap. Consequently, the current flow for a particular gap potential indicates the relative purity of the helium in the system. A visual indication of the gas purity is also available by the color of the gap arc. This analyzer has been quite useful in that the removal of oxygen by the getter furnaces takes place quite readily while the ability of the furnaces to remove nitrogen decreases relatively fast with use of the getter. The Beckman analyzer may show low oxygen level in the gas while the total impurity indicator shows high gas impurity level. This indicates an apparent high nitrogen content and a need for a getter change.

<u>Air Glove Box System</u>. The air boxes in the system are Nos. 1, 2, and 6 (<u>Figs. 1</u> and <u>6</u>). Air is aspirated through each box at the rate of 20 cfm by a 100 cfm fan mounted from the ceiling in the adjoining room. Air enters the box through a nylon filter backed up by a Cambridge Absolute filter. The pressure drop of the inlet filter maintains a negative pressure in the boxes. At 20 cfm and with clean filters, the box pressure is approximately minus 1/2 inch of water. As the filters get dirty, the pressure decreases until at minus 4 inches of water, the filters are changed. The air leaving the boxes passes through two Cambridge Absolute filters and two ball-type isolation valves, in parallel sets. The air then flows through a 2 inch solenoid valve and into the air exhaust manifold where it

passes through another Cambridge Absolute filter to the fan, and thence to the exhaust stack on the roof of the building. A separate line from each box is connected to a pressure switch and an oil-filled manometer. The pressure switch is set for minus 4 inches of water and will de-energize the solenoid valve, closing it when the pressure falls to minus 4 inches of water in the box. Each box has a manual toggle switch which will de-energize and close the solenoid valve.

Waste Water Disposal System. All facility waste water is stored in two large tanks and checked before being released to the drain (Fig. 7). Water from the shower, change room lavatory, work room sink, and condensation from the air conditioner, enters the sump and is pumped through two filters into the upper tank. The second filter is much finer than the first. Pressure gauges across the two filter banks indicate when the filters need changing. When the top tank becomes approximately 3/4 full, a 300 cm^3 sample is checked for alpha contamination. If this value falls below the maximum allowable limit, the tank is drained to the sewer. If the count is above the maximum allowable limit, the water is drained into the bottom tank and diluted before draining to the sewer.

The sump pump does not operate continuously, but is turned on and off by a water level switch. The switch actuates the pump when the water reaches a certain level, and the pump will operate until the water level has dropped to another pre-set level. The difference in this level is essentially a dead band, in that the pump will not turn on until the level has reached the high point setting. The sump tank is provided with a safety device which gives an alarm when the level of water in the sump is above normal operating height. This circumstance could arise from a faulty pump, dirty filters, full storage tank, etc.

Room Ventilation. The room ventilation is supplied by a 7-1/2 hp fan mounted on the roof of the building. Air is drawn in from the outside area through a cooling coil and filters housed directly above the main entrance to the facility. (See Fig. 1.) Air flow is approximately 6000 cfm. The air enters the change room and is filtered a second time before entering the work room. These two banks of filters are composed of Fiberglas®. The air is then swept across the length of the room, passes through a bank of Fiberglas® filters and Cambridge Absolute filters in series and exits through an exhaust duct to the roof fan and stack. The air box fan exhaust is also tied into this room exhaust duct.

An auxiliary air conditioning unit inside the work area recirculates and cools the room air when necessary.

Emergency Power. A 15 kw generator is situated on the roof of the building to provide 440 volt-3 phase standby power in the event of a power failure. The generator will automatically start up and carry the load should a power failure occur, and will automatically shut off once power has been restored.

The emergency generator operates the following pieces of equipment: room ventilation fan, air box exhaust fan, helium pumps, and a 440-110 step-down transformer which reduces the voltage to 110 volts to operate the glove box solenoid valves, the oxygen analyzer and recorder, and the continuous air sampler in the work area.

The emergency generator motor operates on propane with a total of 12 tanks in 2 banks of 6 each mounted on the roof. When one bank of tanks is empty, an automatic switchover device will open the manifold to the other bank and also show a red disc to indicate a change-over has taken place.

The motor can be started manually from the roof under no load or the generator can be tested under full load by means of a test switch. Full load tests of the generator are made twice a week.

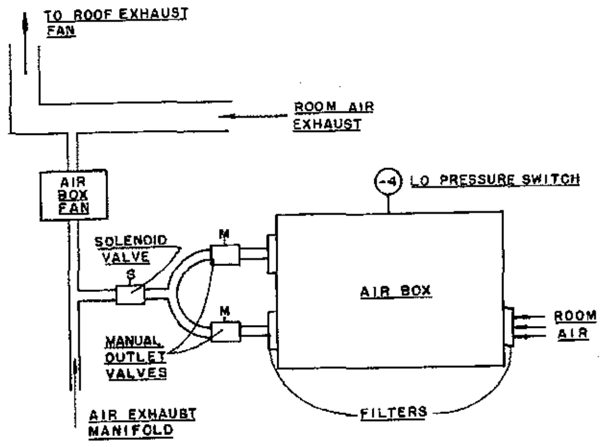

Fig. 6. Air box system

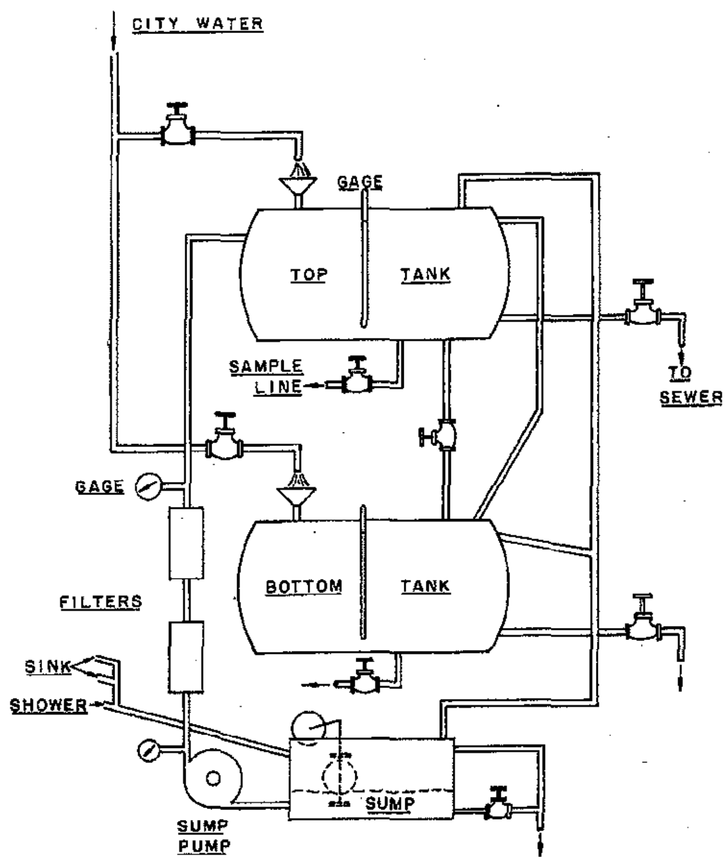

Fig. 7. Waste water disposal system

Safety Devices. Due to the toxic and pyrophoric nature of the plutonium materials being handled and possibility of equipment failures and personnel errors, great care has been taken to make the system as safe as possible. Some of the safety devices have already been mentioned in previous sections of this text; others will now be described.

An alarm system has been installed in the facility by an organization supplying electric protection services. A signaling system in the plutonium area is connected by wire to the organization's control station which maintains a continuous 24 hour electrical supervision of the system and will transmit any alarms to personnel on a call list. A total of 10 separate circuits are supervised by the organization. The following is a list of these circuits and the abnormal conditions which produce an alarm:

1. Failure in the 440 volt-3 phase and 220 single phase circuits.
2. Air pressure in the work area greater than minus 1/4 inch of water with respect to surrounding areas.
3. Gas pressure in the helium boxes greater than minus 1/2 inch of water and gas pressure in the air boxes lower than minus 4 inches of water.
4. Temperature in any of the glove boxes greater than 100°F.
5. Oxygen impurity in the helium boxes greater than 100 ppm as indicated by the gas analyzer. Under these conditions the purification system is automatically isolated and a negative gas pressure is maintained in the helium boxes by the air glove box exhaust system.
6. A temperature greater than 1700°F in the getter furnaces.
7. Gas pressure greater than 3/8 psi in the helium manifold line entering the pumps, indicating pump failure.
8. Unscheduled entry into the facility.
9. Rapid temperature rise in the work area.
10. Abnormally high water level in the sump of the waste water disposal system.

OPERATING EXPERIENCE AND DISCUSSION

The facility has been in continuous operation with plutonium materials since March, 1961. No major incidents have occurred. Studies have been made of the synthesis of plutonium carbide and of solid solutions of uranium carbide - plutonium carbide, the fabrication of the products into fuel pellets, surface grinding the pellets to dimensional tolerances, the determination of carbon content and x-ray diffraction patterns of the various products*. In addition to these operations, glove box space is available for installation of equipment for metallographic studies and for a limited amount of wet chemical analysis.

Plutonium materials have been transferred into or out of the glove box system more than 200 times with no detectable alpha contamination outside the glove boxes. To date,

* Work for the United States Atomic Energy Commission done under subcontract to United Nuclear Corporation.

there has been no alpha contamination in any part of the helium purification system since helium from the glove boxes passes through two sets of Cambridge Absolute filters before entering the purification system. Absence of contamination greatly facilitates servicing the helium pumps and the getter furnaces which are located outside the work area.

Oxygen in the helium leaving the helium glove box system is normally 2-3 ppm with water vapor less than 1 ppm. Nitrogen is not measured directly, but based upon oxygen analysis, is estimated to be less than 20 ppm. Under normal operating conditions, helium make-up in the system is about 100 ft^3 per week.

A preventive maintenance program has been set up for periodic inspection or servicing of the various components which are likely to need attention. The five getter furnaces are recharged at two month intervals with about 6 pounds each of zirconium-50 atomic percent titanium alloy in the form of thin metal chips or turnings. The bearings in the helium pumps are replaced after 1500 hours of continuous use. The three solenoids controlling the modulating valves on the helium boxes are replaced at about three month intervals. The electric alarm and monitoring system is routinely inspected and maintained by the organization providing this service. The above items represent the chief maintenance costs in operating the facility.

The emergency power functions as intended, but was not designed to heat the getter furnaces. Although oxygen in the system rises only about 15 ppm during an eight hour power failure, sufficient emergency power to operate all parts of the system is considered desirable.

The alarm system has been quite valuable in promptly alerting personnel to malfunction of equipment, particularly in the initial operation of the facility. Although difficulties now rarely occur, the alarm system is considered an essential safety feature, especially since the facility is used on a one-shift, five-day week basis, and operates continuously.