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: J. F. Eichelberger

: Trip Report - L. L. Bentz  
to National Lead and ORNL

: R-Building

: URANIUM HEXAFLUORIDE PRO-  
DUCTION.

URANIUM HEXAFLUORIDE PRODUCTION AT NATIONAL LEAD - FERNALD

D. L. Roesch, W. R. Amos and the writer visited National Lead at Fernald on July 2, 1958 to discuss problems concerned with the production of uranium hexafluoride.

We were met in the lobby of the Administration building by Mr. C. Pepper, who is in charge of analytical instrumentation. Mr. Pepper escorted us to the office of Mr. J. W. Robinson, head of analytical chemistry. Here we had a discussion explaining our visit to Fernald. From my telephone conversation with Mr. Robinson on July 1, I learned that they prepared uranium hexafluoride from gram quantities of uranium salt. In our discussion with Mr. Robinson and Mr. Pepper, we learned that most of their uranium hexafluoride preparations were from one-half gram quantities of  $U_3O_8$ . Occasionally they reacted as much as five (5) grams of  $U_3O_8$  to produce uranium hexafluoride. They produce this material only for mass spectrometer isotopic analysis. One-half gram of  $U_3O_8$  is a sufficient quantity for the analysis.

Production of uranium hexafluoride is on a batch scale at room temperature or colder with the aid of a catalyst composed of 75 per cent KCl and 25 per cent KI. One-half gram of  $U_3O_8$  is loaded in a one-fourth inch O.D. copper tube reactor (see Figure 1), the reactor is tapped so as to spread the sample evenly over the bottom. Catalyst, about the size of the head of a kitchen match is dumped on top of the sample. The reactor is closed by means of attaching the reactor to a Moke angle valve (C 411 A) with a flare fitting.

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The reactor is connected to a vacuum pump and evacuated while the outside of the reactor is heated to just a blue coloration with a torch to remove traces of moisture in the sample. It is pumped for five minutes and the valve is closed. The reactor is transferred to the fluorine reaction rack and attached by means of a Crawford fitting. The entire system is evacuated and the valve in the line between the reservoir and the reactor is closed. Fluorine is added to the reservoir to a pressure of four pounds. A thermocouple is attached to the bottom of the reactor and a Dewar of liquid air is placed around the reactor and cooled until frost appears on the nut of the flare fitting. The valve on the reactor is closed and the liquid air Dewar removed from the reactor. Fluorine is allowed to go into the reactor. The temperature gauge is carefully observed for sudden deflection caused by the reaction of fluorine with the  $U_3O_8$ . This reaction occurs anywhere from  $-10^{\circ}C$  up to room temperature. Then a cold trap of trichloroethylene and dry ice is placed around the reactor to solidify the uranium hexafluoride. The vent line to the reactor is opened slowly to read the pressure and the pressure of excess fluorine, HF and waste gases are released slowly to the atmosphere to a pressure of about two pounds. The remainder is pumped off through the trap system of rock salt and Drierite to remove all the waste products. The Hoke angle valve on the reactor is closed and the reactor removed and warmed under a hot water tap. Then it is connected to a vacuum rack and cooled in a trichloroethylene dry ice bath and pumped again to remove trapped gases. Then the valve on the reactor is again closed and the reactor removed for mass spectrometer analysis. (Mass Spectrometer by Consolidated Electrodynamics Corporation, Model 21-220A)

It was pointed out that it is very important that moisture, grease and organics be kept free from contacting the fluorine gas. They have had a number of violent explosions with these impurities. Shielding of approximately one-fourth inch steel

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completely enclosed the reactor chamber and Dewar flask during the fluorination. They recommend the use of Monel valves, tubing and fittings. Hoke diaphragm valves with Teflon gaskets are used. Hoke needle valves (two of them, No. M-341) are used on the high pressure fluorine lines from the fluorine cylinder which is stored outside the building in a small enclosure. The valves are outside the building with extended handles through the building wall to the inside. The needle valves have pipe threads and the threads are lubricated with a Teflon thread compound (E. Co. Engineering Company, New York 12, New York).

Mr. Robinson thought that we should be able to get more information pertinent to our problem from J. C. Barton at K-25, Oak Ridge, also from Y-12 reports. He also said that Goodyear at Waverly might be able to help us.

From what we have learned of the difficulties Fernald had in getting their equipment set up for handling fluorine and reacting it with one-half to five (5) gram quantities of  $U_3O_8$ , I feel that it is quite essential that we make a thorough investigation of this problem before attempting to undertake uranium hexafluoride processing. Fernald personnel have spent much time at Goodyear gaining useful experience. Handling fluorine gas is very dangerous and it is absolutely necessary that all lines, valves and equipment be free of moisture, grease and other organic matter. I feel that it is practically impossible to set up our equipment and test it with depleted uranium in a couple of weeks. I also feel it is extremely essential to test the equipment carefully with depleted uranium, as an explosion with uranium-233 could cause a rather serious contamination problem.

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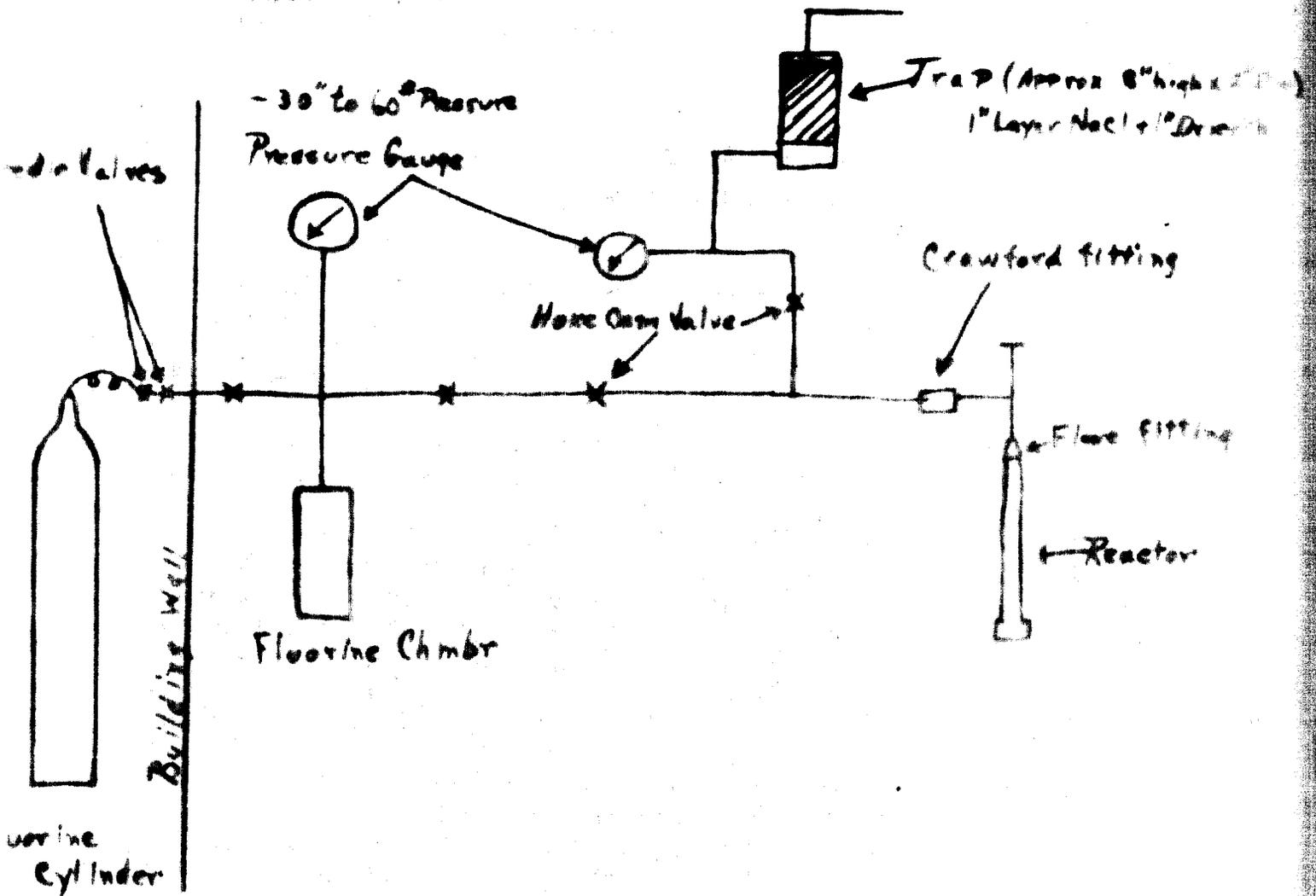


Fig. 1. Sketch of  $UF_6$  Production Equipment at Fernald.

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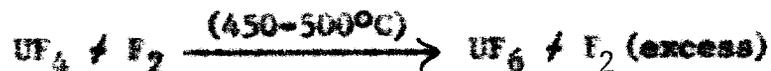
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LAB SCALE PRODUCTION OF URANIUM HEXAFLUORIDE  
(Oak Ridge National Laboratory)

On July 14, 1958, I visited the Laboratories of the K-25 Gaseous Diffusion Plant at Oak Ridge, Tennessee for the purpose of obtaining information on how to produce uranium hexafluoride on a lab scale.

I was escorted to the office of Dr. Harvey Bernhardt by a plant guard. I told Dr. Bernhardt our problem and the purpose of my visit to K-25. He first started by drawing a rough sketch on the blackboard of the "set-up" he felt would be satisfactory for our  $UF_6$  production (see Figure 1a). He suggested the use of a piece of Monel pipe with threaded ends for a reactor and pipe caps with tubing for the ends. The cap could be taken off and a nickel boat containing the  $UF_4$  put in the Monel pipe to be reacted with fluorine gas. Teflon or aluminum gaskets could be used as seals, depending upon the amount of heat present. The reactor could be put in a tube furnace and reacted with fluorine at a temperature of 450 to 500°C. The tube furnace connected to a cylinder of fluorine gas with a valve and joint (flare fitting) between the cylinder and reactor. Following the reactor with a joint and then a trap made of Monel to freeze out the  $UF_6$ . The  $UF_6$  is solidified by surrounding the trap with a dry ice trichloroethylene bath. Following the  $UF_6$  trap is a chamber containing  $Al_2O_3$  or soda lime to neutralize the excess fluorine gas before going to the atmosphere or vacuum pump. The chemical reaction of fluorine with  $UF_4$  is as follows:



K-25 obtains fluorine in 35-pound containers filled on the site. Dr. Bernhardt thought that we would probably be using General Chemical fluorine in cylinders of 700 to 750 pounds

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pressure. As for materials of construction in contact with fluorine or uranium hexafluoride, Inconel is good, Monel is better and nickel is best. For gasket materials, Teflon, 2S and 3S aluminum are very good. Teflon will stand temperatures up to 150°C. Fluoroethane is supposed to be all right but will eventually burn up and carbon-up the system. Monel containers are much more preferred because of its ease of machining. Closed containers are heliarc welded. Plain copper is fine, they have some in use for 13 years. Brass is OK, but not as good as copper. Stainless steel is not too good. Syphon was suggested for bellows for temperature expansion (Fulton Syphon, Knoxville, Tennessee). Copper bellows would probably be OK. Silver solder is used for joining materials. (No. 1800 Eutectic flux and EZE-FLO ASTM Grade 6, Eutectic Welding Alloy Corporation, 172nd St. and Northern Blvd, Flushing, New York.)

Valves are cleaned after soldering with dilute nitric acid. Uranium hexafluoride is moved by B-4 pumps operating at 850 to 900 rpm with a capacity of 40 to 80 liters per minute. These pumps can be completely throttled down. They are a bellows (Syphon) type pump. Phosphor bronze, nickel plate, manufactured by Stewart Metal Fabricators, 710 West Walnut Street, Johnson City, Tennessee (price \$1500). For moving small quantities, approximately 1-1/2 liters per minute, magnetic pumps can be used. For information see report K-833, "Magnetic Pump for Corrosive Gases and Liquids", Hoke 413 diaphragm needle valves are used and Crane HGP (bellows type, body-Monel, bellows-phosphor bronze) valve are used quite widely. This Crane valve corresponds very closely with the Hoke bellows type valve we used on the ANP salt purification work. Kerotest valves are also used. They are diaphragm valves with brass body (\$15.00 to \$16.00). Pressures are measured by means of an Ashcroft Bourdon gauge. These gauges must be silver soldered and equipped with bronze gears (Lonergan Company, Philadelphia, Pennsylvania; 30 lb to 30" vacuum, \$14.30). Vacuum pumps that might be

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contaminated with fluorine use MFL oil, a fluorinated hydrocarbon, this oil will not burn up. Pumps using MFL oil must either be kept operating or the pump heated before starting. Veeco valves may be used in temperatures up to 160°C (Vacuum Electronic Corporation; one-half inch - \$18.00). Needle valves are used for throttling purposes.

In preparing to start up the equipment for the production of UF<sub>6</sub> the pieces should be carefully degreased. The system can be degreased with Freon 113 or trichloroethylene. The system can be leak tested with helium and a leak detector. It is imperative that all grease or oxidizable materials be removed, as fluorine in contact with grease or oxidizable material will explode. Finger prints on a Teflon gasket in a bellows valve is sufficient to cause an explosion when contacted with fluorine. After having degreased the equipment, it should be pumped down and flamed with a torch to help drive off the moisture. The equipment should be conditioned or pretreated with fluorine (five-inch vacuum to allow for expansion) and heating at 150°C overnight. Fluorine and UF<sub>6</sub> react with water as follows:



UO<sub>2</sub>F<sub>2</sub> is a solid and can be removed by passing chlorine trifluoride (ClF<sub>3</sub>) over it, however, it is very important to keep the system dry and thus avoid its formation. Pretreatment of the metal system with fluorine forms the fluoride of the metal.



Failure to pretreat the system with fluorine will cause the formation of UF<sub>5</sub>, a solid residue that is insoluble.

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NiF<sub>2</sub> once formed on the surface of a nickel vessel will protect it indefinitely as it has a similar coefficient of expansion as nickel. Dr. Bernhardt will be giving a paper at the Fall ACS meeting in Chicago on uranium fluorides. For chemical properties of UF<sub>6</sub>, he suggested the Journal of Chemical Society (London); "Some Physical Properties of UF<sub>6</sub>", January 1953, D. R. Llewellyn. Dr. Bernhardt also gave some formulas for calculating vapor pressures of liquid UF<sub>6</sub> at various temperatures.

Between 64°C and 92°C

$$\log P_{mm} = 6.99464 - \frac{1126.288}{t^{\circ}C + 221.963}$$

Between 90°C and 230°C

$$\log(\text{psi}) = 5.77865 - \frac{1494}{t^{\circ}C + 274.145}$$

Dissociation temperature (UF <sub>6</sub> )	- over 1200°C
Critical temperature	- 230.2 ± 0.2°C
Critical pressure	- 45.5 ± 0.5 atm.
Vapor pressure UF <sub>6</sub> (25°C)	- 120 mm

Following are the solid fluoride compounds of uranium; UF<sub>3</sub>, UF<sub>4</sub>, UF<sub>5</sub>, U<sub>2</sub>F<sub>9</sub>, U<sub>4</sub>F<sub>17</sub>, UF<sub>6</sub>(gas). UF<sub>5</sub> has the lowest melting point, 353°C. It is necessary to keep UF<sub>6</sub> below 120 mm to keep it as a gas. At 64.05°C, 1133 mm of UF<sub>6</sub> is a solid, liquid and vapor in equilibrium. The apparent boiling point is 56.5°C.

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To prepare uranium hexafluoride at atmospheric pressure it is necessary to keep the lines carrying the gas around 70°C. This can be done by means of Nichrome tape (0.9 ohm/ft) insulated by placing in glass tape (Raybestos, Manhattan Inc.)

Compression fittings are used with brass ferrules or Teflon ferrules. With Teflon ferrules a new ferrule must be used each time a connection is broken.

The UF<sub>6</sub> flow can be measured by means of a pressure transmitter made by Taylor Instrument Company (see Figure 2). Two pieces of equipment are used:

1. Absolute pressure (PBM-Pressure Blind Multiplier).
2. Differential pressure (DBM-Differential Blind Multiplier - \$250 each).

An orifice made of Monel is used with such an instrument. Formulas for calculating size of orifices:

$$F(\text{cc/min}) = A \sqrt{B P_f \Delta P}$$

F = Flow  
 A = Constant  
 B = Constant  
 P<sub>f</sub> = Fore Pressure  
 ΔP = Pressure drop across plate

Formula from Perry - Chemical Engineering Handbook, section on orifice starting (page 403).

$$F = A \sqrt{(B P_f^2)^* \sqrt{C P_f \Delta P}} \sqrt{D \frac{\Delta P}{P_f} \sqrt{P_f \Delta P}}$$

\* Correction term added.

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Vena Contracta Pressure taps, upstream tap at least two (2) pipe diameters away from the orifice, downstream tap one (1) pipe diameter from the orifice. A 0.1" hole is equal to 8 to 20 liters per minute.

### Isotopic Analysis

Isotopic analysis is performed by 6" radius Nier type mass spectrometers built by General Electric. They have just received two 5" radius, 180° Model 21-220, Mass Spectrometers (\$50,000). They are having a special instrument built by Nuclide Analysis Associates, State College, Pennsylvania; L. F. Herzog, President. This instrument will handle solids and it is a large surface ionization spectrometer. It will handle small samples at most milligram or microgram quantities. Uranium-234 is determined by alpha counting. Uranium-235 can be determined by fission counting, however with less precision. Mass ratio of alpha energy is listed in the budget report for work to be done. Optical spectrometry is being used at K-25 for "Gunk" solutions of 5 to 10 per cent, however this method is a long way off. They can resolve uranium-233 from uranium-235, but uranium-234 may give them a little trouble. For a separation factor of 1.005, the limit of error for a 95 per cent confidence for a single measurement is plus or minus 0.0005 to 0.0008. It would be very difficult to obtain plus or minus one per cent. Twenty to thirty minutes are required for one reading.

$$R = \frac{\left(\frac{U^{233}}{U^{238}}\right)_T}{\left(\frac{U^{233}}{U^{238}}\right)_B} = 1.005$$

R = Separation factor

For R = 1.05, 9 measurements = ± 0.00017 or 0.3 per cent.

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A two-gram sample container is equal to 10 determinations. As much as 100 ppm of uranium-235 would have no effect on a uranium-233 determination. An analysis performed on a sample with a separation factor of 1.05 is not difficult but for an analysis on a sample with a separation factor of 1.005 it is quite difficult.

K-25 will be glad to run our preliminary samples. Mound should check Oak Ridge Operations Office requesting sample containers and analysis. Prints of their sample containers can be furnished. They were wondering if our uranium-233 contained any fission products, also the question of alpha activity was raised. Hoke 411 valves with Hoke 1/8" cone connectors are used on the 1/4" and 1/8" Monel sample bulbs. The 3/8" bulb will accommodate approximately 20 grams of liquid UF<sub>6</sub>.

Air tolerance on uranium bearing materials they normally work with is 3.2 disintegrations per minute per cubic foot.

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Research Division

LLB:la

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