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AN IMPROVED APPARATUS FOR THE PREPARATION OF PLUTONIUM HEXAFLUORIDE

by

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[redacted]

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CHEMISTRY--TRANSURANIC ELEMENTS

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CHEMISTRY - TRANSURANIC ELEMENTS

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ABSTRACT

An apparatus is described which will produce plutonium hexafluoride in greater than 85% yield by the reaction

$$PuF_4 + F_2 \longrightarrow PuF_6$$

The equipment, as described, is capable of producing plutonium hexafluoride at a rate of approximately 0.75 gm/hr, and larger production could be achieved by a simple scaling up of the apparatus.

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Introduction

The preparation of plutonium hexafluoride in fractional gram quantities has been described previously.[1,2] However, the rate of production was so low as to make it difficult to produce material in quantities adequate for experimental purposes.

An improved apparatus has been developed for the production of plutonium hexafluoride by the reaction

$$PuF_4 + F_2 \longrightarrow PuF_6$$

Apparatus

The apparatus for the preparation and purification of plutonium hexafluoride is illustrated in Fig.1. The manifold system is constructed of 1/4 in. copper tubing joined by silver solder. All of the valves in this portion of the fluorination system are of the removable bellows type with fluorothene seats and fluorothene gasketed copper bellows. The body of the valve is monel.* To enhance the utility of the system, the manifold was constructed in duplicate, thus permitting pumping through traps, and precluding the formation of vapor block by the fluorine which is continuously given off by the plutonium hexafluoride. In addition, pumping may be carried out in both directions, thus permitting samples to be transferred conveniently. Furthermore, an alternate inlet is provided in case of valve failure.

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* Crane valves, types HGP, HAP, and HTP.

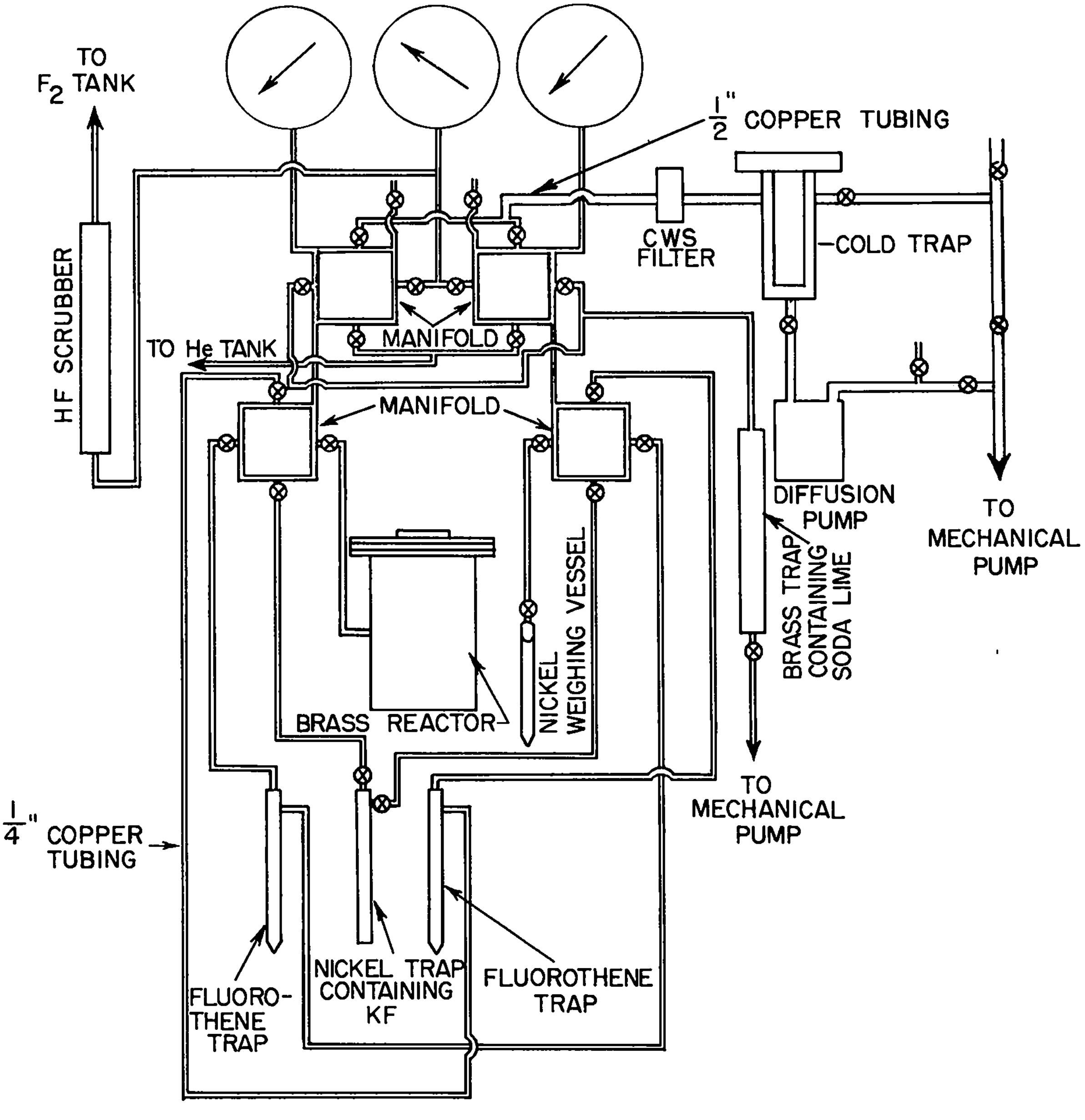

Fig. 1. Fluorination system.

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The upper of the two sets of manifolds provides the inlets for the various services required for the preparation of plutonium hexafluoride. These manifolds bring fluorine, helium, high vacuum, and a rough vacuum to the lower manifolds. The rough-vacuum line is used to remove the major part of any gases that are left in the system after reaction. To remove the fluorine, the gases are taken through a soda-lime trap before going to the pump. All pump exhausts are hooded.

The lower manifolds are used to distribute the services brought in through the upper manifolds. Connected to the lower manifolds are a brass reactor, a nickel weighing vessel, and several fluorothene and nickel traps. The choice of these materials for the construction of the fluorination system was dictated because of their relative inertness to corrosion by fluorine. In the case of fluorothene, its transparency was also a dictating factor.

The reactor, Fig. 2, is a brass can 4 in. I.D. x 6 in. high, exclusive of the 3 in. extension at the top. The top to the reactor is teflon gasketed and is secured by six machine bolts. At the top of the extension is a fluorothene window through which the reaction can be observed. The reactor originally had no extension for the fluorothene window; however, it was discovered that the window became very much overheated when located close to the hot region. The window mounted in the extended top, as shown in the figure, remained cool when the reactor was running at high temperatures.

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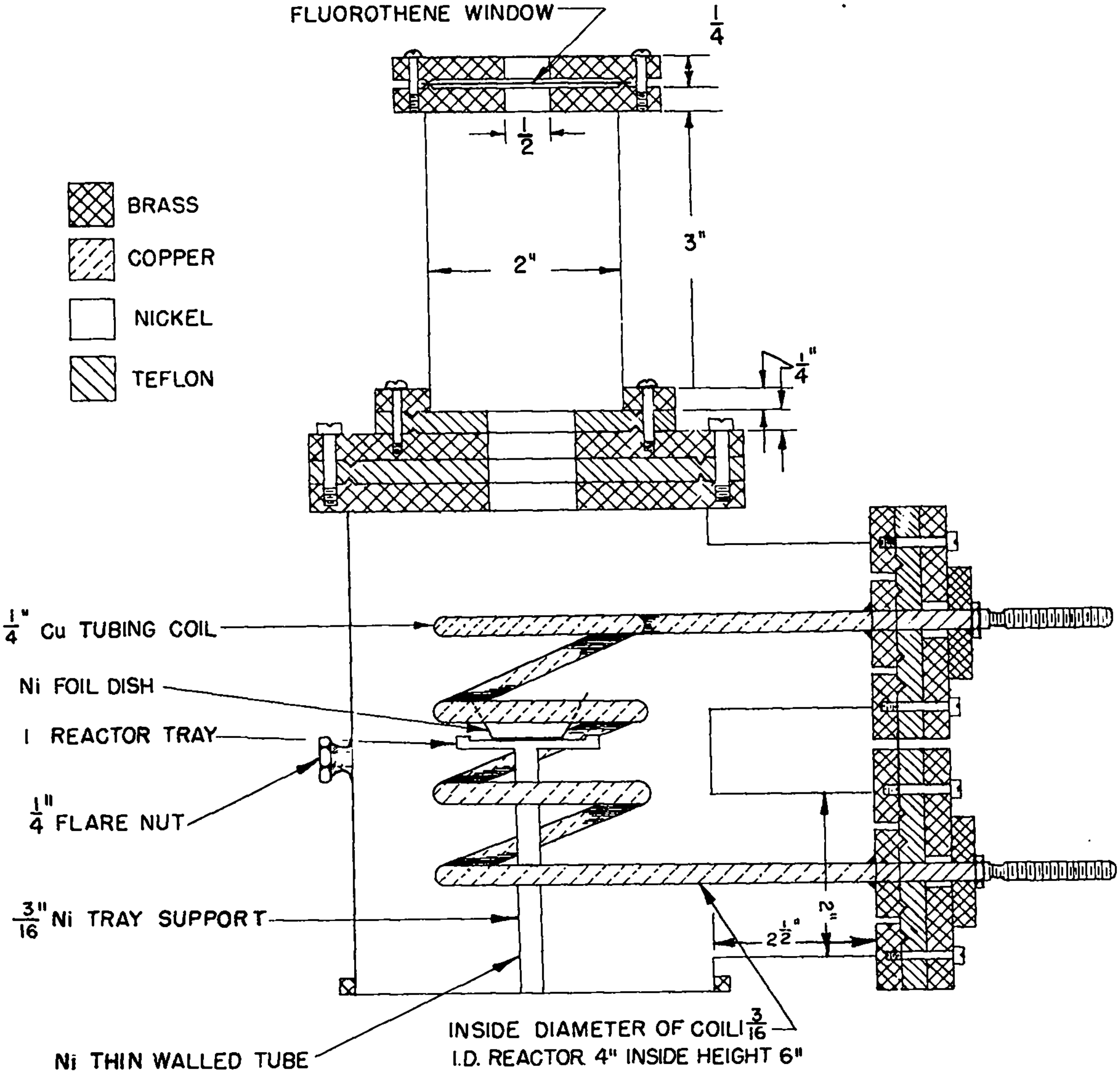

Fig. 2. Fluorination reactor.

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In the center of the reactor is an inductively heated nickel dish on which the plutonium tetrafluoride is placed. The heating coil is a 1/4 in. copper tube wound in a coil with an inside diameter of 1-3/16 in., leaving approximately 1/32 in. clearance between it and the reactor dish. The heating coil serves also as a condenser for the plutonium hexafluoride, since liquid nitrogen is circulated through it while the reaction takes place. The plutonium hexafluoride is frozen out almost immediately after being formed, thus reducing any tendency toward subsequent decomposition of the compound. The high temperature gradient and the large difference in the temperatures between the reactor dish and the heating-cooling coil set up strong convection currents which promote the rapid transfer of plutonium hexafluoride from the dish to the collection coils. For this reason, it is possible to get an efficient reaction by charging the reactor with a single load of plutonium tetrafluoride and fluorine; hence, the problems of accumulation of impurities from the fluorine and the disposal of large quantities of fluorine in a simple flow system are obviated.

Experimental Data

Several preparations of plutonium hexafluoride have been carried out using this apparatus. All of these runs have been done at an absolute fluorine pressure of 33 cm. This is the vapor pressure of fluorine at the temperature of the liquid nitrogen which is forced through the heating-cooling coil. The temperature of the plutonium tetrafluoride at the time of reaction is approximately 700°C as determined by an optical pyrometer.

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Run 1. A charge of 0.5915 gm of plutonium tetrafluoride was permitted to react with fluorine for a period of 15 min. As soon as the dish showed a red color, a white condensate was noticed on the cooling coil. The amount of condensate appeared to increase with time. After the 15-min reaction period, the bulk of the fluorine was removed by pumping while liquid nitrogen flow was maintained through the heating coil. To minimize the loss of plutonium hexafluoride, the fluorine was taken through a liquid nitrogen trap. The coil was then allowed to warm to room temperature, and the white product was distilled into the trap. The outer tube of this trap was constructed of transparent fluorothene to facilitate observation. The product deposited as a brownish powder, characteristic of plutonium hexafluoride. This powder was purified by distillation into a liquid nitrogen trap while the system was being evacuated. The purified product was then distilled into a nickel weighing vessel. It was found that 0.358 gm of plutonium hexafluoride was made during this run. The yield is 54% of theoretical.

Upon inspection of the reactor dish, it was seen that some tetrafluoride was unreacted. This was treated with fluorine for 1 additional hour, at the end of which time all the tetrafluoride had been used. There was then a total of 0.561 gm of hexafluoride and a yield of 85%.

Run 2. A charge of 0.8176 gm of tetrafluoride was fluorinated for 45 min. At the end of this time 0.1240 gm was unreacted. The hexafluoride was purified by pumping away non-condensible gases while the product was distilled into a liquid nitrogen trap. It was further puri-

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fied by distillation into a trichloroethylene-dry ice trap. The yield of plutonium hexafluoride was 0.660 gm, which is 72% of theoretical. The yield was 85% on the basis of the plutonium actually used in the reaction.

Run 3. Fresh tetrafluoride (1.0238 gm) was then added to the reactor dish (which still contained the somewhat caked tetrafluoride left unreacted from Run 2). There was present at the start of the reaction 1.1478 gm of tetrafluoride. A run of 1 hr duration produced 1.0758 gm of hexafluoride. On the basis of all the tetrafluoride present, this is a yield of 84%. Based on the fresh plutonium tetrafluoride added just before the run, the yield is 94%. Considering only the material which disappeared from the reactor dish, the yield is 96%.

Run 4. This run was designed as an exploratory experiment on the rate of formation of plutonium hexafluoride. The reaction took place in two parts: an initial run of 15 min duration, followed by a more extended reaction period of 1 hr. At the beginning of the reaction period, the reactor dish contained 1.1319 gm of plutonium tetrafluoride, of which 0.9880 gm was added after the completion of the last reaction. The 15-min reaction period produced 0.460 gm of plutonium hexafluoride, which represents a yield of 36% on the basis of all tetrafluoride present and a yield of 42% on the basis of the fresh tetrafluoride. The apparatus was then run for 1 hr more without refilling, and 0.638 gm of plutonium hexafluoride was produced. In this portion of the run, 50% of the total tetrafluoride was converted to hexafluoride, and 58% of the fresh tetrafluoride was converted.

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The entire run of 1 hr and 15 min duration produced 1.098 gm of hexafluoride. This is 87% of all the plutonium and 99% of the fresh material added. Of the plutonium tetrafluoride used in this reaction, 87% was recovered as plutonium hexafluoride.

No conclusions on the rate of formation of plutonium hexafluoride have been drawn from these results because of the preliminary nature of the experiment.

Data of the foregoing experiments are tabulated in Table 1.

References

1. A. E. Florin, Los Alamos Scientific Laboratory Report LAMS-1118 (1950).
2. A. E. Florin, Los Alamos Scientific Laboratory Report LA-1168 (1950).

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TABLE 1

SUMMARY OF DATA ON PREPARATION OF PLUTONIUM HEXAFLUORIDE

Run	Duration, min	PuF_4 Present, gm		PuF_6 Produced, gm	Percentage Converted		Yield, %*
		Total	Fresh		Total	Fresh	
1a	15	0.5915	0.5915	0.358	54	54	-
1b	60	left from 1a		0.203	31	31	-
1a + b	75	0.5915	0.5915	0.561	85	85	85
2	45	0.8176	0.8176	0.660	72	72	85
3	60	1.1478	1.0238	1.0758	84	94	96
4a	15	1.1319	0.9880	0.460	36	42	-
4b	60	left from 4a		0.638	50	58	-
4a + b	75	1.1319	0.9880	1.098	87	99	87

* This yield is calculated on the basis of the plutonium tetrafluoride actually used in the reaction.

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