

GAT-252
TECHNOLOGY –
FEED MATERIALS

OXIDE FLUORINATION TOWER

by

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SUMMARY

A 3-inch-diameter flame tower for the conversion of uranosic oxide to uranium hexafluoride with elemental fluorine was tested for possible use in the fluorination step of the present uranium recovery process. The oxide was fed from a hopper to the tower by a screw feeder. The fluorine and the oxide entered at the top and flowed concurrently down through the tower. The unreacted or partially reacted oxide was collected in an ash receiver at the bottom. Fine solid particles were removed from the gas stream by an electrostatic precipitator and a tube-type filter. The uranium hexafluoride was collected in cold traps.

Twenty-five experimental runs were conducted with average oxide feed rates from 3.73 to 19.38 pounds per hour. The average fluorine flow rates were from 7.5 percent below to 448 percent above the stoichiometric amount of fluorine required. The best operating conditions were at a feed rate of 15 pounds of oxide per hour with a minimum fluorine excess of 75 percent (10.6 pounds of fluorine per hour). The material collected in the tower ash receiver represented between 6.0 and 10.0 percent of the total amount of uranium fed during the run. The ash, combined with an equal weight of oxide, can be fed back to the tower. The electrostatic precipitator was capable of collecting up to 92 percent of the solids in the gas stream. A porous-tube filter worked best for the removal of the remaining particles in the gas stream.

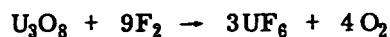
The experimental runs have shown that uranosic oxide can be satisfactorily converted to uranium hexafluoride in a 3-inch-diameter flame tower of simple design. The present conversion rate of uranosic oxide, approximately 4 pounds per manhour with the tube reactor, could be increased to 10 pounds per manhour by using the flame tower.



INTRODUCTION

In the present uranium recovery process at this plant the conversion of uranosic oxide (U_3O_8) to uranium hexafluoride (UF_6) by elemental fluorine (F_2) is carried out in small inclined-tube reactors. The original reactors each consisted of a tube about four feet long mounted in a horizontal position. The uranosic oxide was fed to the reactor from a small hopper at one end and was moved slowly down the tube by a ribbon-type agitator while the oxide reacted with the fluorine. This method was unsatisfactory because the heat of reaction deformed the agitator. Two of the three reactors were modified by removing the agitator and installing a vibrator which caused the oxide to move slowly down the length of the reactor. With this method, it takes about 12 hours to feed 11 pounds of oxide through a reactor.

In a study to improve the economics of this portion of the recovery process, an oxide fluorination system was designed and installed in a pilot plant for testing. The major unit of the system was a 3-inch-diameter tower reactor similar in construction to the towers used in the Goodyear Atomic Feed Plant for the conversion of uranium tetrafluoride to uranium hexafluoride. This oxide fluorination tower was designed for a capacity of 22 pounds of oxide per hour at a fluorine excess of 100 percent, based on the equation



EQUIPMENT

A flow diagram of the oxide fluorination system is shown in Figure 1. The uranosic oxide in the hopper is fed to the tower by a screw feeder driven by an electric motor through a variable speed transmission. The oxide is dispersed by a motor-driven stirrer as it enters the tower from the screw feeder. The fluorine is distributed evenly around the oxide inlet through a small annular opening. Coils around the tower are used either for heating or cooling. The unreacted or partially reacted oxide is collected in an ash receiver at the bottom of the tower. The fluorine and the oxide flows are concurrent downward through the column with the uranium hexafluoride, nitrogen, oxygen, and excess fluorine leaving at the bottom. Any fine solid particles formed are removed from the gas stream by an electrostatic precipitator supplemented by a porous-tube filter. The uranium hexafluoride is collected in cold traps; any residual uranium hexafluoride is removed from the gas stream by the chemical trap. The excess fluorine and other gases are exhausted from the system by an air ejector and from the building through the process vacuum header. The oxide fluorination tower and related equipment are shown in Figure 2. Details of the equipment are given in Appendix I.

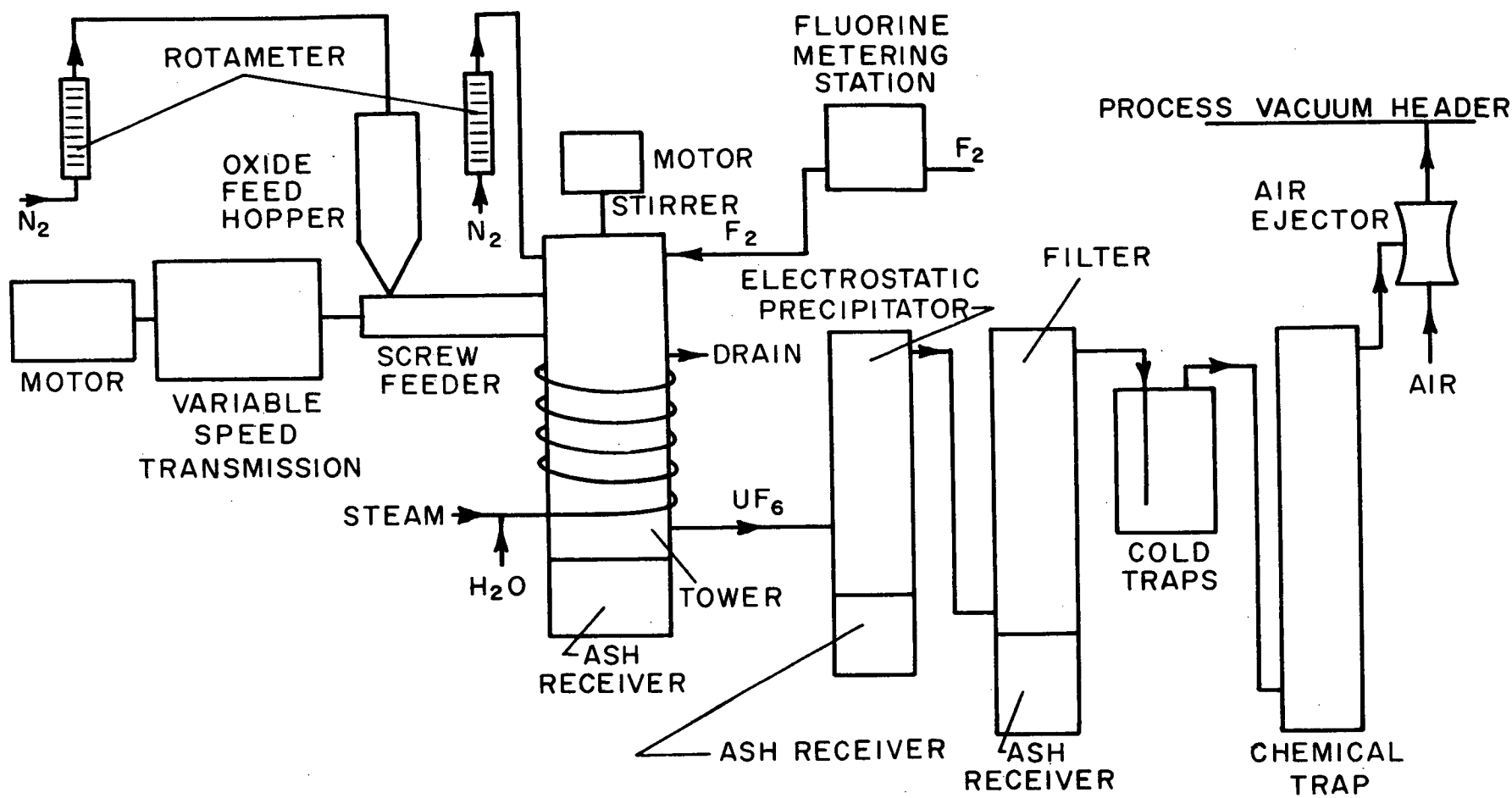


FIG. 1. FLOW DIAGRAM OF THE OXIDE FLUORINATION SYSTEM

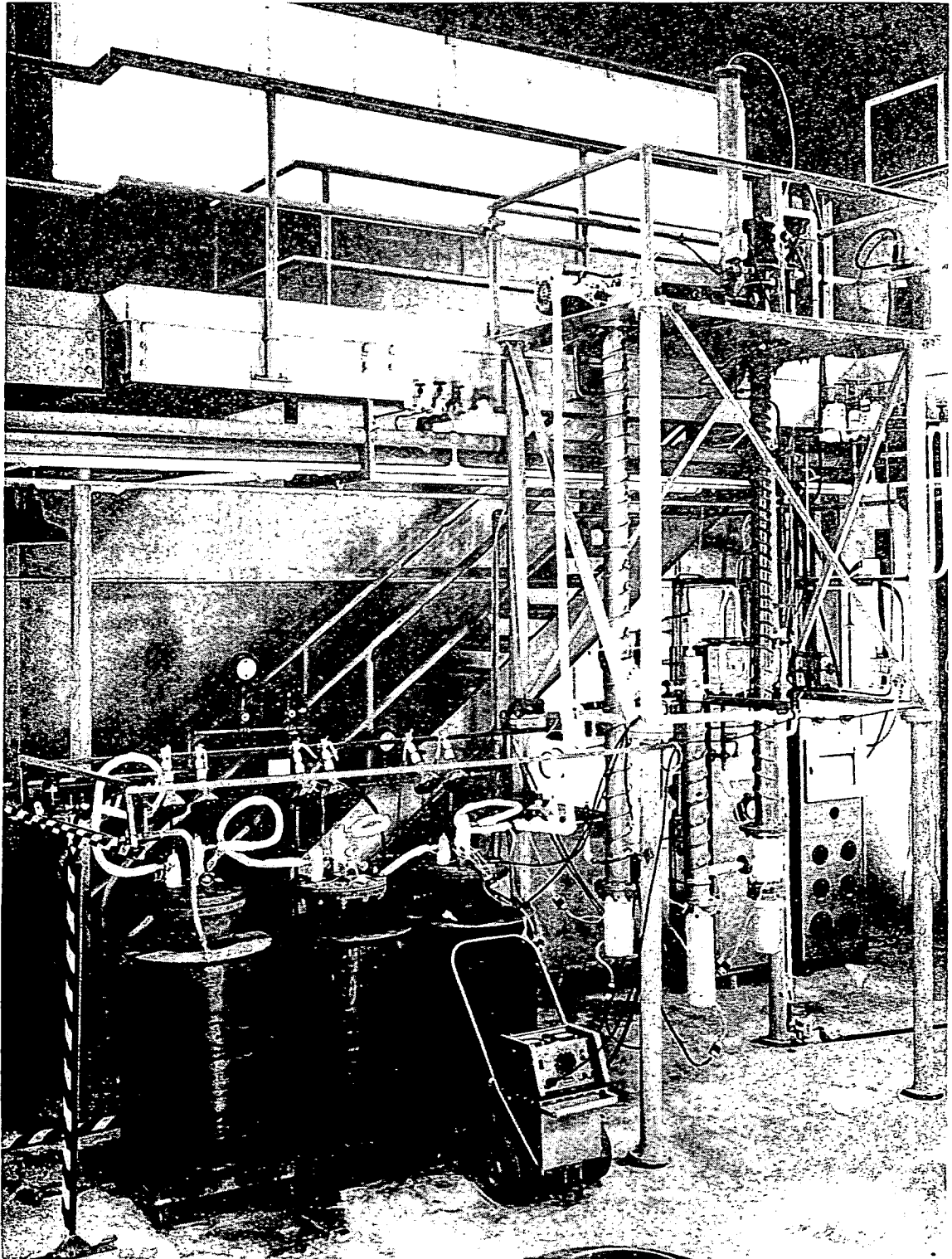


FIG. 2. OXIDE FLUORINATION TOWER AND RELATED EQUIPMENT

OPERATING PROCEDURES

EQUIPMENT PREPARATION

The system was treated with fluorine prior to the first run and after the addition of new equipment. A nitrogen flow through the system was started and fluorine was added by 20 percent increments every 20 minutes until the flow was estimated to contain about 60 percent fluorine. The nitrogen was then valved off, and the treatment was completed with a low flow rate of 100 percent fluorine for 20 minutes. The screw feeder was calibrated before the operation was started; the calibration curve is shown in Figure 3.

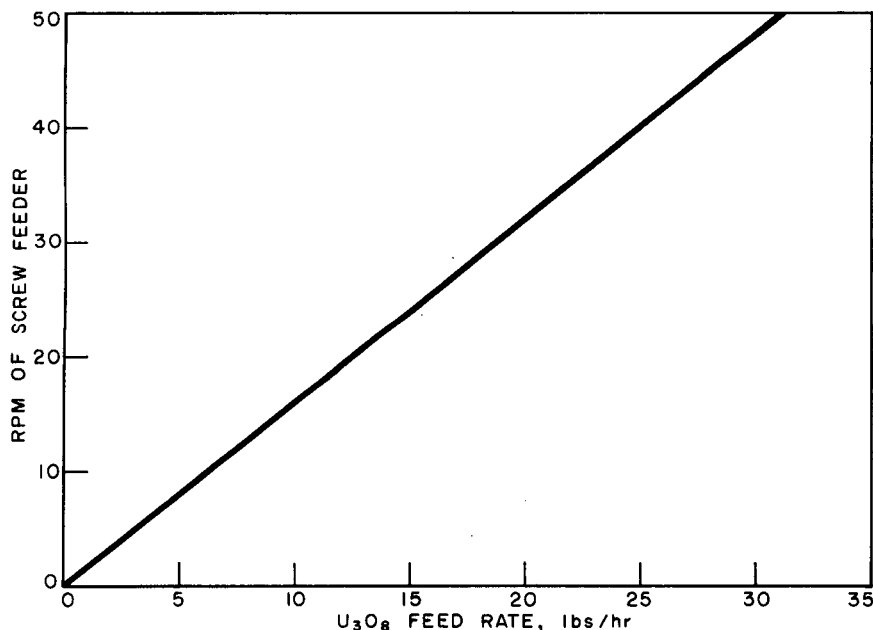


FIG. 3. SCREW FEEDER CALIBRATION GRAPH

The oxide was passed through a 12-mesh screen to remove excessively large particles before it was placed in the hopper. Before the hopper was filled with a known amount of oxide, the screw feeder was set for the desired feed rate. The tower, precipitator, filter, and lines up to the first cold traps were heated to 75°C. or higher before a run was started. In addition, the dip legs of the cold traps, the pigtails, and the valves of the cold traps were heated. The cold traps were cooled to approximately -50°C. by the addition of dry ice to a 60 percent aqueous solution of Prestone. Nitrogen was then passed through the system for approximately 15 minutes.

Fluorine was passed through the system for 15 minutes after the nitrogen purge was completed. The electrostatic precipitator, stirrer, and screw feeder were started in that order after the desired fluorine flow through the system was obtained.

EQUIPMENT OPERATION

The tower pressure was maintained near atmospheric pressure by throttling the last valve in the exhaust line before the air jet which was used to pull the gases from the system. The pressure drop through the system increased during a series of runs because

of a build-up of deposits on the filter, the filling of the cold traps, or the filling of the chemical trap. After the pressure drop through the system reached approximately 11.5 psi., the system was closed down and the necessary steps were taken to reduce the pressure drop. Usually the pressure drop across the filter was reduced by tapping the outside of the filter with a hammer. The effectiveness of the electrostatic precipitator started to decrease after approximately 15 minutes because of deposits collecting on the walls of the precipitator and the center wire. Most of these deposits were removed from the walls during the run by tapping the precipitator while the power was turned off. This operation was repeated about every 15 minutes during a run.

It was possible to determine when no oxide remained in the hopper by the sharp decline in the maximum tower temperature. After the run was completed, the system was purged with fluorine and nitrogen for 10 and 15 minutes, respectively. The system was then evacuated to 3.5 psia. The above procedure was used only when the receivers were removed for weighing and sampling purposes. It was not necessary to remove the receivers from the system after each load of oxide. Because of the size of the receivers, they had to be emptied only after every second or third run. The feed hopper was refilled with a total off-stream time of less than 30 minutes. This was accomplished by eliminating the fluorine purge at the end of the run and cutting the nitrogen purging time down to 5 minutes before the cover was removed from the top of the feed hopper. The nitrogen bleed to the top of the feed hopper was turned on approximately ten minutes before the estimated time for the feed to be gone. After the hopper was loaded, a 5-minute nitrogen purge and a 5-minute fluorine purge were given the tower before the screw feeder was started. The weights of the cold traps were determined after they were removed from service.

RESULTS AND DISCUSSION

A total of 1,529 pounds of uranosic oxide was reacted with fluorine to form uranium hexafluoride during the 25 experimental runs conducted with the oxide fluorination system. In addition, 67 pounds of tower ash from these runs were converted to uranium hexafluoride. The equipment was periodically modified during the first 19 runs, while runs 20 through 25 were made with the equipment in the final, recommended form.

OPERATING CONDITIONS

Average oxide feed rates varying from 3.73 to 19.38 pounds per hour were investigated. The feed rate for optimum operation of the system was approximately 15 pounds per hour. At oxide feed rates above 15 pounds per hour the pressure drop through the system increased very rapidly, making it difficult to maintain the desired fluorine flow rate for an extended period. Feed rates used in each run are given in Table I. Average fluorine flow rates from 7.5 percent below to 448.0 percent above the stoichiometric amount were investigated. The calculation of the excess fluorine was based on a feed containing 100 percent uranosic oxide. The tower should be operated with a minimum fluorine excess of approximately 75 percent in order to prevent the build-up of deposits inside the tower. Fluorine flow rates and the amount of excess fluorine used in each run are presented in Table I.

The reaction in the tower begins without preheating the fluorine and within 30 seconds after the screw feeder is started. It takes approximately 20 minutes for the tower to reach the normal operating temperature. The maximum temperature during normal operating conditions is usually below 600°C. The reaction area of the tower, as determined by wall temperature measurements, is about one foot long and starts just below

Table I

SUMMARY OF RUNS WITH THE OXIDE FLUORINATION TOWER

| Run | Length of Run, hr. | U ₃ O ₈ Converted, lb. | Average Feed Rate, lb. U ₃ O ₈ /hr. | Average Fluorine Flow Rate, scfh. | Excess Fluorine, percent | Quality of Feed Material, percent U ₃ O ₈ | Ash, g. | Uranium Analysis of Ash, percent U | Uranium Recovered in Ash, percent of total U in feed | Filter Deposits, g. | Uranium Analysis of Filter Deposits, percent U | Uranium Recovered in Filter Deposits, percent U in feed | Deposits in Cyclone Separator, g. |
|-----|--------------------|----------------------------------------------|-----------------------------------------------------------|-----------------------------------|--------------------------|-----------------------------------------------------------------|---------|------------------------------------|------------------------------------------------------|---------------------|------------------------------------------------|---------------------------------------------------------|-----------------------------------|
| 1 | 3.47 | 32.50 | 9.39 | 80.3 | 121.5 | 80.8 | 1,000 | 56.3 | 5.57 | 1588 | 1.74 | 0.27 | -- |
| 2 | 1.00 | 9.17 | 9.17 | 117.8 | 234.5 | 81.7 | 606 | 58.5 | 12.34 | 354 | 2.51 | 0.31 | -- |
| 3 | 2.00 | 26.38 | 13.19 | 103.7 | 126.0 | 82.8 | 868 | 54.5 | 5.64 | 653 | 1.98 | 0.16 | -- |
| 4 | 2.00 | 7.48 | 3.73 | 54.8 | 282.5 | 81.5 | 330 | 52.9 | 7.44 | 307 | 3.50 | 0.46 | -- |
| 5 | 2.18 | 20.00 | 9.17 | 53.4 | 63.0 | 79.3 | 853 | 49.6 | 6.93 | 508 | 1.58 | 0.13 | -- |
| 6 | 2.28 | 20.00 | 8.77 | 48.3 | 39.2 | 80.0 | 315 | 34.5 | 1.76 | 741 | 16.80 | 2.02 | -- |
| 7 | 0.89 | 5.17 | 5.80 | 18.4 | - 7.5 | 81.1 | 20 | 18.7 | 0.23 | 68 | 3.15 | 0.13 | -- |
| 8 | 1.00 | 8.34 | 8.34 | 66.5 | 107.8 | 81.2 | 571 | 59.9 | 13.15 | 449 | 29.1 | 5.03 | -- |
| 9 | 1.50 | 15.25 | 10.15 | 76.3 | 95.8 | 81.2 | 505 | 52.0 | 5.51 | 948 | 0.67 | 0.13 | -- |
| 10 | 1.00 | 12.02 | 12.02 | 86.8 | 92.8 | 80.2 | 375 | 53.3 | 5.39 | 588 | 6.05 | 0.96 | -- |
| 11 | 2.00 | 30.40 | 15.20 | 115.3 | 97.8 | 80.2 | 1,339 | 47.1 | 6.74 | --* | --* | --* | -- |
| 12 | 1.72 | 20.15 | 11.70 | 102.4 | 128.0 | 93.4 | 749 | 62.5 | 6.48 | 87 | 1.36 | 0.02 | 29 |
| 13 | 2.08 | 30.60 | 14.72 | 94.3 | 67.6 | 93.4 | 1,178 | 73.6 | 7.89 | 170 | 2.04 | 0.04 | 34 |
| 14 | 2.02 | 40.00 | 19.83 | 76.0 | 25.8 | 93.4 | 3,277 | 70.9 | 16.15 | 206 | 0.40 | 0.01 | 50 |
| 15 | 4.48 | 68.50 | 14.20 | 113.4 | 104.8 | 93.2 | 3,626 | 73.0 | 10.81 | 1040 | 2.42 | 0.10 | -- |
| 16 | 2.43 | 40.00 | 16.50 | 114.3 | 80.7 | 93.3 | 2,959 | 73.8 | 15.25 | 627 | 3.29 | 0.14 | -- |
| 17 | 0.92 | 17.42 | 19.00 | 115.1 | 57.6 | 92.9 | 2,010 | ---† | ---† | 341* | --* | --* | -- |
| 18‡ | 32.27 | 500.50 | 15.54 | 97.3 | 63.7 | 94.3 | 24,488 | 73.4 | 10.01 | 1441 | 0.60 | 0.01 | -- |
| 19# | 2.57 | 32.50 | 12.64 | 104.2 | 114.8 | 95.7 | 1,386 | 72.6 | 11.05 | 140 | 36.4 | 0.43 | -- |
| 20‡ | 31.87 | 480.00 | 15.05 | 106.9 | 100.1 | 95.3 | 16,283 | 73.0 | 6.73 | 1282 | 3.24 | 0.02 | -- |

*Filter burned up during the run.

†Uranium analysis was not obtained.

‡This run included 13 parts.

#This run included 4 parts.

‡This run included 12 parts. Results for the electrostatic precipitator are shown in Table V.

the level of the annular fluorine opening around the oxide inlet to the tower.

OXIDE IMPURITIES

As shown in Table I, the quality of the oxide used in the experimental work varied from 68.5 percent uranium (80.0 percent uranosic oxide) to 81.2 percent uranium (95.7 percent uranosic oxide). Typical spectrochemical analyses of the oxide feed are given in Table II. The impurities in the oxide were fluorinated along with the oxide.

Table II

TYPICAL SPECTROCHEMICAL ANALYSES OF U_3O_8 FEED*

| <u>Element</u> | <u>Batch No. 1[†]</u> | <u>Batch No. 2[‡]</u> |
|----------------|--------------------------------|--------------------------------|
| Al | 5.0 | 0.3 |
| Ca | 0.6 | 0.1 |
| Cr | 0.6 | 0.1 |
| Cu | 0.2 | 0.05 |
| Fe | 6.0 | 1.0 |
| Mg | 0.05 | --- |
| Mn | 0.06 | 0.01 |
| Mo | --- | 0.01 |
| Ni | 0.6 | 0.1 |
| Si | --- | 0.02 |
| U | >10.0 | >10.0 |

*Values are in percent and are semi-quantitative.

[†] Batch No. 1 contains 68.5 percent uranium.

[‡] Batch No. 2 contains 81.2 percent uranium.

These impurities were removed by the electrostatic precipitator and a tube-type filter. If the fluorinated impurities were not removed from the outlet gas stream, they would plug the valves in the cold traps and the bends in the piping.

The material collected on the filter was analyzed spectrochemically, and typical results for these samples are shown in Table III. The impurities that were in abundance in the oxide are the same impurities that collected on the filter. These are mainly Al, Ca, Cr, Fe, and Ni. The deposits removed from the filter and from the electrostatic precipitator usually contained between one and four percent uranium by weight. This generally represented less than one-half percent of the total uranium used in the run. The weight of the deposits, the uranium analysis of the deposits, and the percent of the total amount of uranium found in the deposits for the different runs are shown in Table I.

TOWER ASH

The ash collected in the receiver during the different runs usually contained between 50 and 75 percent uranium by weight. This normally represented between 6 and 10 percent of the total amount of uranium fed during the run. The weight of the ash, the uranium content of the ash, and the total amount of uranium recovered in the ash for the different runs are shown in Table I.

Table III

SPECTROCHEMICAL ANALYSES OF FILTER DEPOSITS*

| <u>Element</u> | <u>Filter Deposits, Run No. 1</u> | <u>Filter Deposits, Run No. 5</u> | <u>Filter Deposits, Run No. 20</u> |
|----------------|---------------------------------------|---------------------------------------|----------------------------------------|
| Ag | 0.1 | 0.03 | --- |
| Al | >10.0 | >10.0 | 4.0 |
| Ca | >10.0 | 8.0 | 1.0 |
| Cr | 0.2 | 8.0 | 6.0 |
| Cu | 0.3 | 0.5 | 1.0 |
| Fe | >10.0 | >10.0 | >10.0 |
| Mg | 0.3 | 0.2 | 0.04 |
| Mn | 0.08 | 1.0 | 0.1 |
| Na | --- | 0.3 | 4.0 |
| Ni | 8.0 | 5.0 | 2.0 |
| Pb | 0.07 | 0.05 | 0.02 |
| Sn | 0.08 | 0.2 | 0.2 |
| Ti | 0.008 | 0.03 | --- |
| Zn | 0.02 | 0.03 | --- |
| U | --- | 5.0 | >10.0 |

*Values are in percent and are semi-quantitative.

ASH CONVERSION

Runs 21 through 25 were made with feed material which contained some tower ash from previous runs. An attempt was made to start the reaction in the tower with 100 percent ash, but this was unsuccessful. However, a mixture of 50 percent ash and 50 percent oxide reacted without the need for preheating the fluorine. The mixture contained approximately 62 percent uranium by weight. The ash was ground with a rod mill and screened to finer than 12-mesh before it was used. The maximum temperature obtained during these runs was 525°C. The results are presented in Table IV.

TOWER PLUGGING

The tower plugged after 13 hours of operation. This occurred during run 7, where the average amount of fluorine fell below the stoichiometric amount needed for complete conversion of the oxide. Runs 5 and 6 were made with a calculated fluorine excess of 63 and 39 percent respectively. Small green clinkers were found in the ash in these two runs, with a greater amount in the run with the smaller quantity of excess fluorine. Before the tower plugged, it operated at an average fluorine excess over 100 percent for the first 8.5 hours. In addition, the tower operated at 63 percent excess fluorine for 2.2 hours and 39.2 percent excess fluorine for 2.3 hours before run 7. It seemed that in the runs with the lower percent of excess fluorine the plugging of the tower was accelerated. This seemed especially true in the last run during which the average fluorine flow rate was below the stoichiometric amount needed for the reaction.

Analysis of the material removed from the tower showed it to contain 3.40 percent uranium, with no tetravalent uranium present. Spectrochemical analysis showed the material

Table IV

RESULTS OF RUNS WITH ASH IN FEED MATERIAL

| Run | U ₃ O ₈ and Ash Used, lb. | Average Feed Rate, lb./hr. | Excess Fluorine, percent | Tower Ash, g. | Precipitator Deposits, g. | Filter Deposits, g. | Percent of Total Deposits Removed by Precipitation |
|-------|-------------------------------------------------------|----------------------------------|--------------------------------|------------------|------------------------------|---------------------------|-------------------------------------------------------------|
| 21 | 37.09 | 7.68 | 284 | | | 100 | |
| 22 | 21.77 | 16.11 | 69 | 8278 | 1203 | 149 | 93.7 |
| 23 | 7.48 | 5.67 | 448 | 285 | 2483 | | |
| 24 | 38.04 | 6.72 | 328 | 562 | 582 | 52 | 91.8 |
| 25 | 75.56 | 10.82 | 113 | 5030 | 1030 | 156 | 86.8 |
| Total | 179.94 | | | 14155 | 5298 | 457 | Avg. 92.1 |

to be composed mainly of Al, Ca, Fe, and Ni, the same materials that were the major constituents of the impurities of the oxide and of the deposits on the filter. The results of spectrochemical analysis are shown in Table V. The light green material from the tower could not be identified by X-ray diffraction analysis.

Table V

SPECTROCHEMICAL ANALYSIS OF PLUGGING MATERIAL
IN TOWER, RUN NUMBER 7*

| <u>Element</u> | <u>Analysis</u> | <u>Element</u> | <u>Analysis</u> |
|----------------|-----------------|----------------|-----------------|
| Ag | 0.005 | Fe | >10.0 |
| Al | >10.0 | Mg | 0.6 |
| Ca | >10.0 | Mn | 0.9 |
| Cr | 0.05 | Ni | 5.0 |
| Cu | 0.3 | U | 1.0 |

*Values are in percent and are semi-quantitative.

FILTER

A large filter that contained 37 tubes, each 88 inches long with a diameter of 0.4 inch, was used during the conversion of 1193 pounds of feed material. Of this amount, the last 713 pounds were converted after an electrostatic precipitator had been installed in the system. The filter was not plugged completely at the end of the experimental runs. It is believed that several hundred more pounds of oxide could have been handled with the filter before it would have become plugged to an extent requiring replacement of the

tubes. The pressure drop across the filter normally increased from about 0.5 psi. to about 4.0 psi. during a run. The filter was tapped with a hammer after each run to remove the deposits from the tubes and to reduce the pressure drop. Several attempts were made to blow the deposits off the filter by back-flushing with nitrogen, but this method was unsuccessful. The operation of the large tube-type filter proved to be far more satisfactory than was anticipated. It was expected that this large filter would plug after the conversion of approximately 350 pounds of oxide, based on runs made with a tube filter one-fifth the size of the large one. However, the large tube filter was still operating satisfactorily after handling more than 1,193 pounds of oxide.

ELECTROSTATIC PRECIPITATOR

Run 20 was made to test the effectiveness of the electrostatic precipitator which was installed in the system. This run consisted of 12 parts. A total of 480 pounds of oxide was fluorinated during the runs at an average feed rate of 15.05 pounds per hour. The average fluorine excess was 100.1 percent. A laboratory-built power supply with a maximum output of 11 kilovolts was used during parts 20A through 20E. The positive terminal of a 40-kilovolt dielectric test set was connected to the center wire of the electrostatic precipitator for parts 20F through 20I, and the negative terminal was connected to the center wire for parts 20J through 20L.

A total of 4458 grams of deposit was removed from the filter and the electrostatic precipitator during run 20. The electrostatic precipitator removed 71.2 percent of the solids in the gas stream while the filter removed 28.8 percent of the solids. The parts of the run (20F through 20L) made using the dielectric test set as the power supply showed that the electrostatic precipitator was more efficient (83.1 percent as compared to 62.5 percent of the solids in the gas stream) for removal of solids when the negative terminal was connected to the center wire of the precipitator. A summary of the amount of materials removed by the filter and the electrostatic precipitator during the run is presented in Table VI.

The current averaged 16 microamperes during part 20H when a 1/8-inch nickel rod was used as the center wire of the electrostatic precipitator. This current was much less than the current drawn by the precipitator during other parts of the run, and it was assumed that with a low current the amount of solids removed would also be low. The electrostatic precipitator collected 92.1 percent of the total amount of solids collected during runs 21 through 25 in which the feed material contained some tower ash. The negative terminal of the power supply was connected to the center wire for these runs. Samples of deposits from the electrostatic precipitator and corresponding samples of deposits from the filter showed the material from the filter to have a slightly higher uranium concentration than the material from the precipitator. The results for the samples are given in Table VII.

COLD TRAPS

The outlet gases from the cold traps in runs with two of the special 10-inch cold traps and a clean-up cold trap in series generally contained less than 0.5 mole percent uranium hexafluoride. The amount of uranium hexafluoride carry-over increased as the cold traps filled. It was possible to freeze out 180 pounds of uranium hexafluoride in one of the special 10-inch cold traps before plugging occurred.

Table VI

**MATERIAL FROM FILTER AND ELECTROSTATIC PRECIPITATOR,
RUN NUMBER 20***

| <u>Part</u> | <u>Filter Deposits, g.</u> | <u>Electrostatic Precipitator Deposits, g.</u> | <u>Percent of Total Deposits Removed by Electrostatic Precipitation</u> | <u>Average Voltage, kv.</u> | <u>Average Current, ma.</u> |
|--------------|------------------------------------|--------------------------------------------------------|-----------------------------------------------------------------------------------------|-------------------------------------|-------------------------------------|
| 20A | | | | <11 | ---- |
| 20B | 153 | 525 | 77.4 | <11 | ---- |
| 20C | | | | <11 | ---- |
| 20D | | | | <11 | ---- |
| 20E | 200 | 210 | 51.2 | <11 | ---- |
| 20F | 115 | 480 | 80.7 | 36.0 | 550 |
| 20G | | | | 36.0 | 770 |
| 20H | 233 | 416 | 64.1 | 34.6 | 16 |
| 20I | 305 | 192 | 38.6 | 36.0 | 166 |
| 20J | | | | 36.6 | 166 |
| 20K | 170 | 648 | 79.2 | 35.9 | 271 |
| 20L | 106 | 705 | 86.9 | 35.2 | 412 |
| Total | <u>1282</u> | <u>3176</u> | <u>71.2</u> Avg. | | |

*In each part 40 pounds of oxide were used.

Table VII

**COMPARISON OF DEPOSITS FROM FILTER
AND ELECTROSTATIC PRECIPITATOR**

| <u>Run No.</u> | <u>Percent Uranium in Filter Deposits</u> | <u>Percent Uranium in Precipitator Deposits</u> |
|----------------|-----------------------------------------------|-----------------------------------------------------|
| 20I | 3.24 | 2.29 |
| 24 | 9.82 | 5.60 |
| 25 | 2.51 | 0.53 |

CHEMICAL TRAP

The chemical trap became plugged several times with uranium hexafluoride and several

times with the sodium fluoride pellets which had crumbled and formed a fine powder. Usually the chemical trap was changed when the pressure differential across the trap reached approximately 9 psi.

CONCLUSIONS AND RECOMMENDATIONS

The experimental runs with the oxide fluorination tower have shown that uranosic oxide can be satisfactorily converted to uranium hexafluoride in a 3-inch-diameter flame tower of simple design. For optimum operation of the tower, the oxide feed rate should be approximately 15 pounds per hour with a minimum fluorine excess of 75 percent (10.6 pounds of fluorine per hour). The oxide must be screened to remove all lumps before it is placed in the hopper in order to insure smooth operation of the screw feeder. It is also necessary to vibrate the hopper to keep the powder from clinging to the sides. Although Teflon and nylon both have good friction characteristics, Teflon is preferable to nylon for the bearing of the screw feeder shaft since it is more inert to fluorine. A tube-type filter and an electrostatic precipitator are recommended for removal of the solid particles in the gas stream. The filter should be retubed after it is plugged; wet decontamination of the tubes is not satisfactory. A small tube filter used during several runs burned in later use after it had been cleaned by wet decontamination. It is necessary to tap the filter periodically in order to knock some of the deposits off the tubes and to reduce the pressure drop across the filter. It is best to operate the electrostatic precipitator with the center wire negative at 35 to 40 kilovolts and with a current above 150 microamperes. It will be necessary to knock some of the impurities from the walls of the precipitator approximately every 15 minutes to maintain the desired current. For future trouble-free operation of the electrostatic precipitator it will be necessary to design insulators for the top and bottom of the precipitator that will be able to withstand a fluorine atmosphere between 200° and 300°C. Until a better insulator is developed, a 1/4-inch quartz rod should be used at the bottom and a Teflon insulator at the top.

The uranium contained in the ash is not considered to be a problem since the ash can be ground up and fed back to the tower mixed with fresh oxide. The uranium in the deposits from the filter and the precipitator can be recovered by conventional wet decontamination methods.

The oxide fluorination tower and related equipment can produce a substantial savings over the present equipment in converting uranosic oxide to uranium hexafluoride. This savings in cost will result from an increase in the conversion rate from 4 pounds of uranosic oxide per manhour with the present system to 10 pounds of uranosic oxide per manhour using the tower reactor.

APPENDIX I

DESCRIPTION OF EQUIPMENT

FEED ASSEMBLY

The variable speed transmission, which had a maximum output of 90 revolutions per minute, was powered by a 1/2-hp. electric motor. A chain and sprocket drive was used between the variable speed transmission and the screw feeder shaft. A nylon bearing was used first for the bearing of the screw feeder shaft, but this bearing was replaced with a Teflon bearing after 16 runs. The diameter of the screw feeder shaft was approximately one inch. The oxide feed hopper was fabricated from a piece of 4-inch-diameter nickel pipe 40 inches long which was welded to the top of the screw feeder assembly. The hopper held approximately 40 pounds of oxide. A Teflon gasket was inserted in the cover plate of the hopper. A small nitrogen bleed was introduced at the top of the feed hopper to prevent fluorine from attacking the nylon bearing when the screw feeder shaft was emptied. A 0-2-scfh. rotameter was used to measure the nitrogen bleed. A small electric vibrator was installed near the bottom of the hopper to keep the oxide from clinging to the side of the hopper.

The stirrer, which was driven by a 1/2-hp. electric motor, was mounted on the top of the tower. This stirrer was made from a 3/8-inch rod 7-1/4 inches long with twenty-two 9/32-inch prongs welded on it to disperse the oxide. The four top prongs were slanted to give a downward motion to the oxide. A greaseless carbon bearing was used to support the stirrer. To prevent fluorine from escaping from the tower through this bearing, a small nitrogen bleed was introduced just below the bearing, and the nitrogen was measured by a second 0-2-scfh. rotameter.

REACTION TOWER

The reaction tower was made from a piece of 3-inch nickel pipe 8 feet long. Three separate copper coils were wrapped around the tower for heating and cooling purposes. The coils were spaced more closely at the top than at the bottom of the tower since the greatest amount of heat was contained in the top part of the tower. It was possible to use steam, water, or a mixture of both in the coils. Usually steam alone was used. The steam was used to heat the column before the start of the run, and after the run was started the steam acted as a coolant in the top part of the tower while it continued to heat the bottom part.

FILTER

The filter contained thirty-seven 0.4-inch-diameter porous metal tubes approximately 88 inches long inside a piece of 4-inch monel pipe 8-1/2 feet long. The filter had one tube sheet which was supported between two flanges at the top of the filter. Aluminum "O" rings were used as gaskets on each side of the tube sheet. The installation of this large filter in the system is shown in Figure 2. The bottom ends of the filter tubes were plugged. In addition, the tubes were loose at the bottom so that the deposits could be

knocked off. A receiver, made from a 12-inch section of 4-inch monel pipe, was bolted to the bottom of the filter column to collect the deposits.

ASH RECEIVER

An ash receiver for collecting the unreacted or partially reacted oxide was mounted at the bottom of the tower. It was made from a 12-inch section of 4-inch monel pipe. An aluminum "O" ring gasket with a 4-inch Pyrex glass pipe flange was used to form a seal at the top of the ash receiver.

ELECTROSTATIC PRECIPITATOR

An electrostatic precipitator was installed at the start of run 20 between the outlet of the tower and the large filter to remove some of the fine solid impurities from the gas stream before they had a chance to plug the filter, thereby increasing the operating life of the filter. The precipitator was constructed from a section of 4-inch monel pipe 4 feet long, with a receiver at the bottom fabricated from a section of 4-inch monel pipe 18 inches long. The precipitator was heated by a steam line, and the receiver was wrapped with nichrome heating wire in order to keep the uranium hexafluoride from freezing to the wall.

The center wire installed in the electrostatic precipitator was a 0.031-inch-diameter nickel wire, which was attached at the bottom with a Teflon insulator during runs 20A through 20G. The Teflon insulator is shown in Figure 4. Teflon was also used to make an insulator packing gland at the top, as shown in Figure 5. The Teflon insulator at the bottom of the precipitator was destroyed during run 20G by the hot corrosive gases in the system. This Teflon insulator was replaced with a calcium fluoride-impregnated Teflon insulator of the same design, but the impregnated Teflon insulator was destroyed before the completion of run 20G. Run 20H was made using a 1/8-inch nickel rod as the center wire with no insulator at the bottom. The remainder of the runs were made using an insulator made from a 1/4-inch quartz rod with a 0.031-inch-diameter nickel center wire.

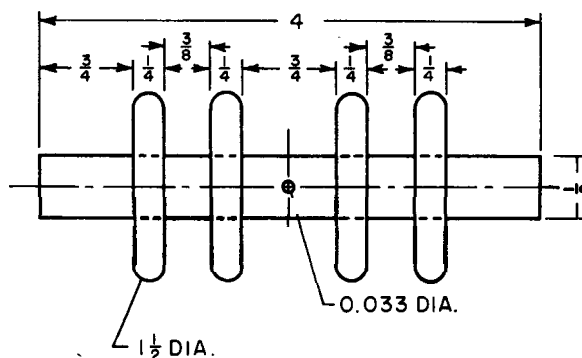


FIG. 4. TEFLON INSULATOR FOR BOTTOM OF PRECIPITATOR

The power to the electrostatic precipitator during parts 20A through 20F was supplied by a unit with a maximum output of approximately 11 kilovolts. A power supply with a maximum output of 40 kilovolts was used for the remainder of the runs with the oxide fluorination system.

COLD TRAPS

Two specially designed cold traps and one clean-up cold trap were used with the oxide fluorination system. The internal construction of the two specially designed cold traps is shown in Figure 6. The external casing was made from 10-inch steel pipe 42 inches long. Flat copper baffles were stacked inside the cold traps with 1-inch spacing between baffles and an open space of 6 inches at each end. The gas entered through a dip leg in

the center and passed up over the copper baffles with a reversal of flow between each baffle before the stream left the cold trap at the outlet valve on top.

The clean-up cold trap was added to remove uranium hexafluoride mist from the outlet gas stream. It was used as the last trap in a series of three. This trap had open spaces at the bottom and top with a series of four copper baffles next to these open spaces. Copper wire mesh was packed between the top and bottom series of baffles to prevent uranium hexafluoride from going through as mist.

CHEMICAL TRAP

A 5-inch-diameter chemical trap containing 35 pounds of sodium fluoride pellets was used to absorb the uranium hexafluoride which passed through the cold traps. Sodium fluoride pellets were used instead of activated alumina because of the large amount of fluorine that must pass through the chemical trap.

HEATERS

Asbestos-covered nichrome wire was used to heat the pigtaills, the cold trap valves, the cold trap dip legs, the tower ash receiver, the filter ash receiver, the electrostatic precipitator ash receiver, the pressure gauges, and the process piping between the reaction tower and the first cold trap. The eight variacs used to control the voltages to these nichrome wire heaters were mounted in the same relay rack as the temperature recorder; this relay rack is shown in the background in Figure 2. Six of these eight were 110-volt, 10-ampere variacs while the other two were 220-volt, 10-ampere variacs. In addition, heat lamps were used on the valves of the cold traps. For plant operations, it will be possible to use two additional 110-volt, 10-ampere variacs instead of the two 220-volt, 10-ampere variacs.

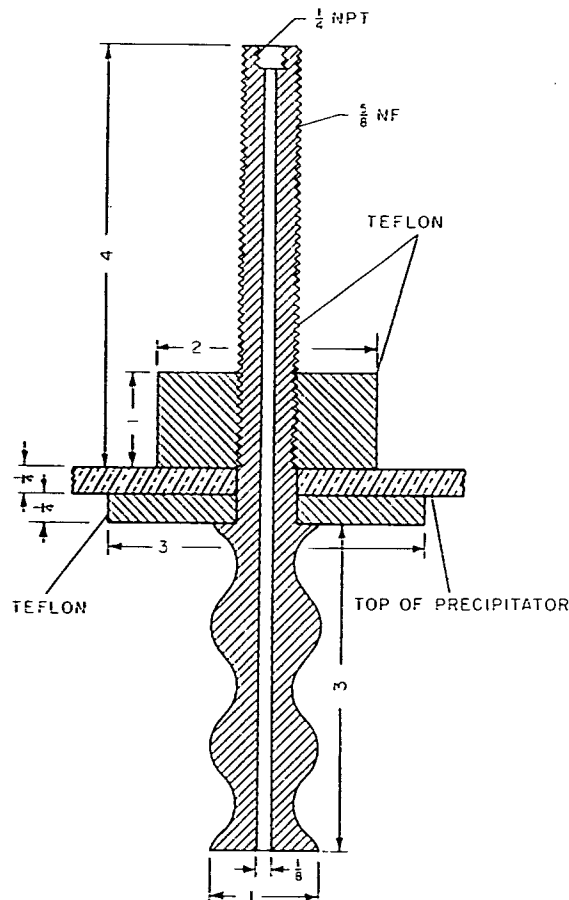


FIG. 5. TEFLON INSULATOR PACKING GLAND FOR TOP OF PRECIPITATOR

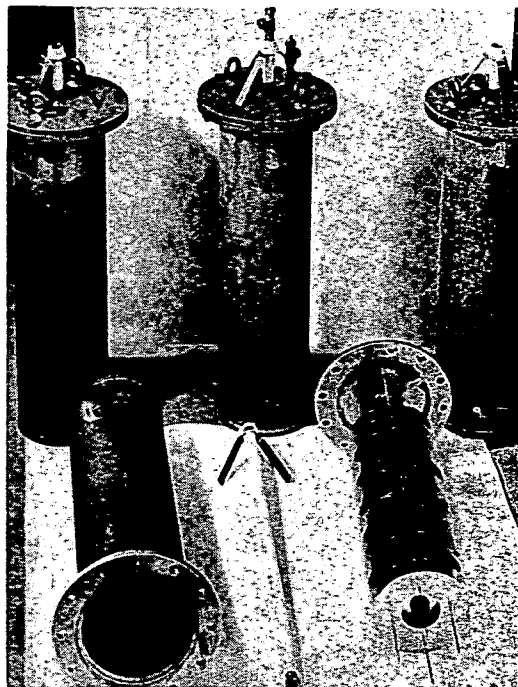


FIG. 6. SPECIALLY DESIGNED 10-INCH COLD TRAPS

TEMPERATURE

Twenty chromel-alumel thermocouples were used to obtain temperature readings in different parts of the system. Ten of these thermocouples were placed on the reaction tower. Three of these ten were in a small area near the top of the tower and the other seven were about one foot apart on the tower. Temperatures were obtained on a single-point recorder by the use of a twenty-point switch.

PRESSURE

The pressure in the oxide fluorination system was controlled by an air ejector. Bourdon-tube pressure gauges were installed at the nitrogen bleed entry to the tower, between the electrostatic precipitator and filter, between the filter and the cold traps, between the cold traps and the chemical traps, and between the chemical traps and the air ejector. Conditions of plugging at any point in the system were determined by the differences in gauge readings.

FLUORINE METERING STATION

The fluorine flow rate was measured by the pressure drop across an orifice. A PBM was used to control the fore pressure to the orifice between 0 and 20 psia., and a DBM was used to control the pressure drop across the orifice between 0 and 1.0 psia. The control valves, the PBM, and the DBM were enclosed in the relay rack shown in the background in Figure 2.

The oxide feed rate was limited at 22 pounds per hour by the capacity of the fluorine metering station when 100 percent excess fluorine was used. The tower could operate below 5 pounds per hour, but with the present instruments in the metering station the fluorine flow rate readings were very inaccurate below the 50 standard cubic feet per hour which were needed for oxide feed rates under 5 pounds per hour.

APPENDIX II

EQUIPMENT DEVELOPMENT

FEED ASSEMBLY

The original feed and stirrer mechanisms installed on the oxide fluorination tower are shown in Figure 7. The Graham Variable Speed Transmission was powered by a 1/4-hp. electric motor, and the screw feeder was connected directly to the transmission. The transmission had a maximum output of 18 rpm., which limited the feed rate to 11 pounds per hour. The screw feeder shaft was supported by a ball bearing contained in the housing at the end of the shaft. This ball bearing plugged up with uranosic oxide after 3.5 hours of operation and the resultant torque stripped a small gear in the variable speed transmission. The feed drive mechanism described in Appendix I was then installed.

The original hopper, shown in Figure 7, was approximately one foot high and held about 12 pounds of oxide. This hopper was filled several times during a run by the use of the two slide valves. These slide valves plugged with the oxide during the initial run and it was necessary to hammer the valves open and shut. During a run it was necessary to remove the hopper container and install a new container before the oxide was gone from the stationary part of the hopper, since there was a possibility of fluorine leakage around the bottom slide valve. The hopper was held fast by ring clamps which proved to require too much time to install because the bolt holes had to be aligned properly and the copper gasket had to be installed between the flanges.

FILTER

The original installation contained a filter section at the bottom of the tower fabricated from a piece of 4-inch nickel pipe 12 inches long. A cylindrical filter, 12 inches long and 3-1/2 inches in diameter, made from a porous nickel sheet, was hung from a lip around the inside of the filter section. After the first run was made, monel rings were fitted at the top and bottom of the filter section to prevent the gases from passing around the edges of the filter.

Deposits caked on the side of the cylindrical filter in the bottom of the column to a thickness of 1/2 inch or greater during a normal run. After ten runs the cylindrical filter was removed and cleaned with a sodium carbonate solution to remove some of the fine particles that seemed to be imbedded in the pores of the filter. The filter burned up at the start of the next run. The system was modified by placing a 2-inch-diameter cyclone separator at the outlet of the reaction tower, followed by a rectangular filter. This rectangular filter had 72 square inches of sintered nickel filtering area compared to the

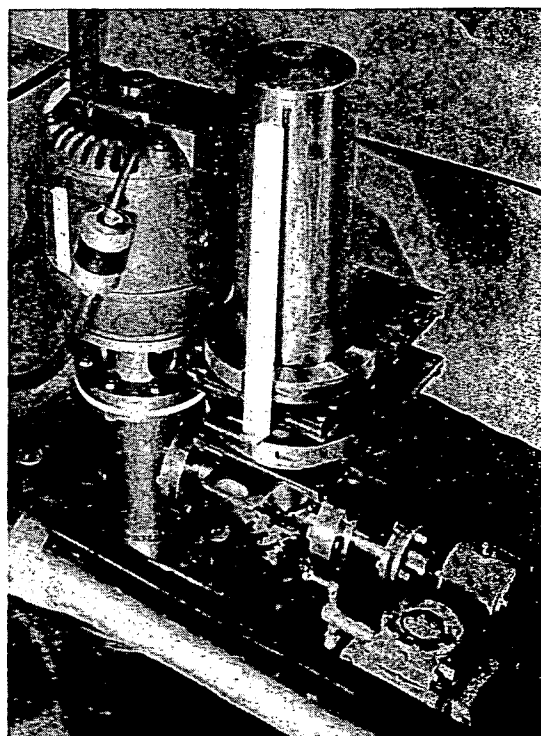


FIG. 7. ORIGINAL FEED ASSEMBLY FOR THE TOWER

cylindrical filter with 132 square inches. This rectangular filter was made from an 18-inch section of 4-inch monel pipe split lengthwise, with flanges welded around the edges. The rectangular filter and cyclone separator arrangement was used for runs 12 through 14. It was possible to complete the runs by tapping the filter section periodically to remove some of the deposits, thereby reducing the pressure drop. A photograph of the rectangular filter with the deposits is shown in Figure 8.

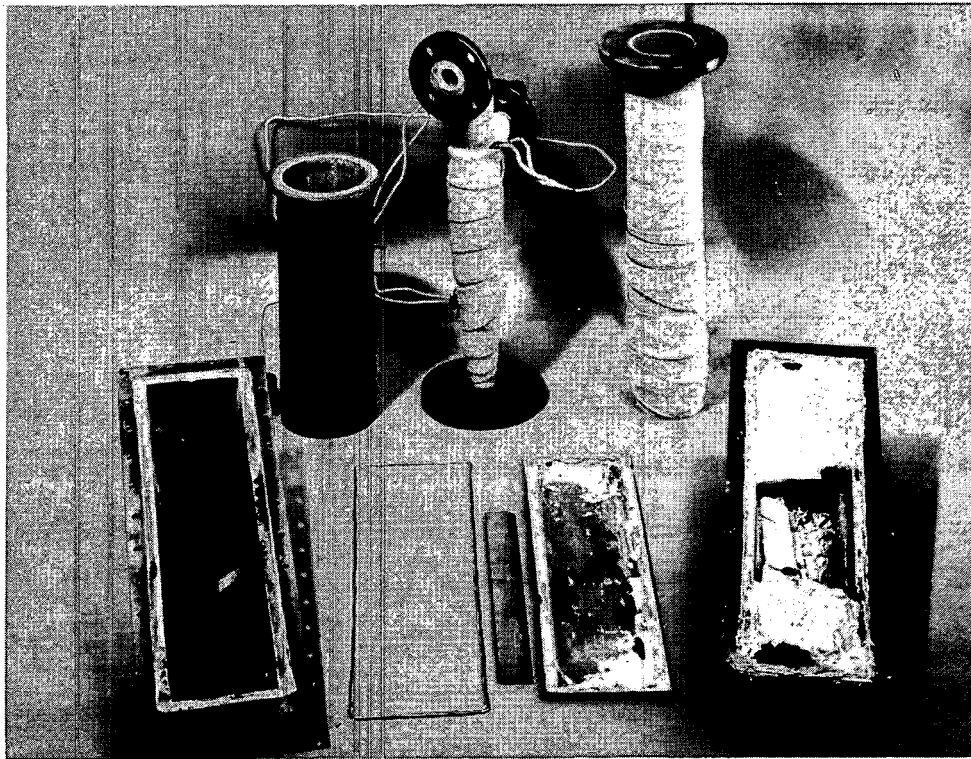


FIG. 8. CYCLONE SEPARATOR AND RECTANGULAR FILTER

The deposit on the filter was approximately 1/4-inch thick. The cyclone separator and rectangular filter were removed from the system and were replaced with a spare tube filter which was used with the reactors in the uranium recovery process. Three runs, 15 through 17, were made using this filter which contained 37 tubes 18 inches long and 0.4 inch in diameter. It had a tube sheet only at one end, the tubes being loose at the other end and plugged. This tube-type filter proved to be very successful in removing the solid impurities from the gas stream, but the filtering area was too small for plant-scale operation of the tower. Two runs were attempted after the tube filter was decontaminated by wet procedures. One run was successfully completed with this filter after wet decontamination, but the filter burned up during the second run.

CYCLONE SEPARATOR

A 2-inch-diameter cyclone separator was installed between the tower and the rectangular filter during runs 12 through 14 to remove the solid impurities from the gas stream before they plugged the filter. The cyclone separator proved to be unsuccessful in removing the impurities from the outlet gas stream of the tower, and less than 20 percent of the deposits were collected in the cyclone ash receiver during the three runs in which it was used. The weights of the material collected in the ash receiver for the cyclone separator were listed in Table I. The cyclone separator is shown in Figure 8 with its

receiver. The separator was made of copper and was wrapped with nichrome heating wire.

COUNTERCURRENT FLOW

A conversion spool piece was installed at the top of the tower during run 19 in order to modify the system for countercurrent flow between the fluorine and the oxide. This conversion spool piece is shown in Figure 9. The fluorine inlet was at the bottom of the tower and the oxide inlet was at the top, while the uranium hexafluoride was withdrawn near the top. It was thought that this arrangement might lengthen the flame area and give a lower ash-to-oxide ratio. The countercurrent experimental run consisted of four parts which were made at an average feed rate of 12.64 pounds per hour with a total operating time of 2.4 hours. The average fluorine excess during the run was 104.2 percent. Three parts of the run were terminated when plugging occurred at the bottom of the oxide feed pipe extension. A large nitrogen bleed was used in the third part to produce a downward nitrogen velocity in the 1-inch oxide feed pipe greater than the upward fluorine velocity in the column, but this proved to be unsuccessful in preventing plugging in the oxide feed pipe. Shortly after the start of the fourth part, plugging occurred around the uranium hexafluoride exit from the column.

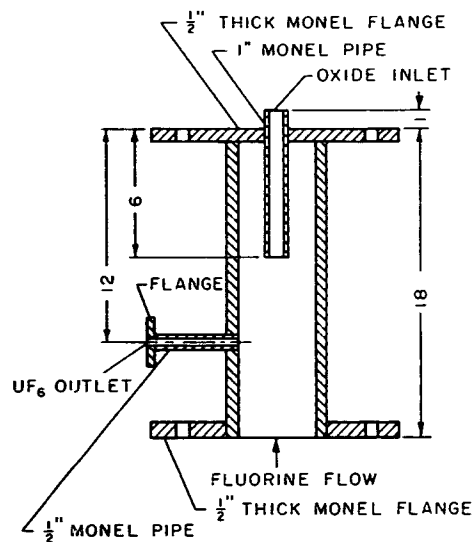


FIG. 9. CONVERSION SPOOL PIECE FOR COUNTERCURRENT FLOW

The results from the countercurrent flow experiment showed that 11.05 percent of the total uranium feed to the tower was found in the ash. This was slightly higher than the majority of the runs with concurrent flow between the oxide and fluorine. The filter deposits contained approximately 0.4 percent of the uranium used in the run. This was much higher than the normal amount of uranium found in the filter deposits. Analyses showed that the filter deposits contained 36.4 percent uranium, which was the highest uranium concentration reported in any sample of filter deposits. Samples of the plugging material from the conversion spool piece were taken for uranium analysis, spectrochemical analysis, and X-ray analysis. The uranium analysis showed it to contain 41.8 percent uranium, including 11 percent tetravalent uranium. The results of the spectrochemical analyses are shown in Table VIII.

The X-ray analysis showed the major crystalline components present to be UO_2F_2 and UF_6 . Further experimental work with countercurrent flow was not undertaken because of the plugging difficulties encountered.

COLD TRAPS

Four cold traps were placed in the original installation in series-parallel flow. However, only one pair in series was used during a run. These original cold traps were made by installing an extra valve and a dip leg in a 10-inch uranium hexafluoride cylinder. These traps were unsuccessful in trapping the uranium hexafluoride, and they were replaced with the specially designed cold traps described in Appendix I. Flexible monel

Table VIII

SPECTROCHEMICAL ANALYSES OF SAMPLES FROM THE RUN
WITH COUNTERCURRENT FLOW, RUN NUMBER 19*

| <u>Element</u> | <u>Filter Deposits</u> | <u>Spool Deposits</u> |
|----------------|------------------------|-----------------------|
| Al | 4.0 | 1.0 |
| Ca | 1.0 | 0.5 |
| Cr | 6.0 | 1.0 |
| Cu | 1.0 | 0.5 |
| Fe | >10.0 | 10.0 |
| Mo | 0.4 | 0.1 |
| Mn | 0.1 | 0.01 |
| Na | 4.0 | --- |
| Ni | 2.0 | 0.3 |
| Pb | 0.02 | --- |
| Si | ND [†] | --- |
| Sn | 0.2 | --- |
| U | >10.0 | >10.0 |

*The values are in percent and are semi-quantitative.

[†]Not detected.

hose was used for the pigtails, but was replaced with copper tubing after a hole developed in one of the pigtails.

ASH RECEIVER

The ash receiver was originally fastened to the filter section with slip-on clamps such as were used with the original feed hopper. The original feed hopper container and ash receiver were interchangeable. The slip-on clamps did not provide enough pressure to form a gas-tight seal with a copper gasket between the flange; therefore, an aluminum "O" ring gasket was used along with a 4-inch Pyrex glass pipe flange clamp.

CHEMICAL TRAPS

The original chemical traps in the system were two nickel traps 2-1/2 inches in diameter and 3 feet long installed for parallel flow. These traps held a total of 14 pounds of sodium fluoride pellets. After the first run these traps were replaced with one 5-inch chemical trap to increase the capacity for removal of uranium hexafluoride.