90
7/25/67

AEC RESEARCH AND DEVELOPMENT REPORT

K-1713

MASTER

REMOVAL OF IMPURITIES FROM URANIUM HEXAFLUORIDE
BY SELECTIVE SORPTION TECHNIQUES:
PROGRESS REPORT FOR 1966

AUTHORS:

M. J. Stephenson
J. R. Merriman
H. L. Kaufman

**UNION
CARBIDE**

UNION CARBIDE CORPORATION
NUCLEAR DIVISION

Operating the

OAK RIDGE GASEOUS DIFFUSION PLANT	• OAK RIDGE NATIONAL LABORATORY
OAK RIDGE Y-12 PLANT	• PADUCAH GASEOUS DIFFUSION PLANT

*For the Atomic Energy Commission
Under U.S. Government Contract W7405 eng 26*

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

Printed in the United States of America. Available from Clearinghouse for Federal
Scientific and Technical Information, National Bureau of Standards,
U.S. Department of Commerce, Springfield, Virginia 22151
Price: Printed Copy \$3.00; Microfiche \$0.65

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

- A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or
- B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

H.C. \$ 3.00; MN 65

Date of Issue: July 17, 1967

Report Number: K-1713

Subject Category: Chemical Separations
Processes for
Plutonium and Uranium

REMOVAL OF IMPURITIES FROM URANIUM HEXAFLUORIDE
BY SELECTIVE SORPTION TECHNIQUES:
PROGRESS REPORT FOR 1966

M. J. Stephenson
J. R. Merriman
H. L. Kaufman

Engineering Development Department
Technical Division

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assume any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

UNION CARBIDE CORPORATION
NUCLEAR DIVISION
Oak Ridge Gaseous Diffusion Plant
Oak Ridge, Tennessee

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

A B S T R A C T

For several years, the U. S. Atomic Energy Commission has placed limits on the types and amounts of various impurities permitted in uranium hexafluoride returned to its gaseous diffusion complex. To aid in providing the technology required to meet these specifications, an experimental program is being conducted at the Oak Ridge Gaseous Diffusion Plant to test various solid sorbent materials to determine their effectiveness for selectively removing volatile impurities from uranium hexafluoride. The testing procedure currently used in this work involves passing a uranium hexafluoride vapor stream, spiked with one or more impurity fluorides, through a 3-inch-diameter trap containing a 36-inch-deep sorbent bed. Sample pairs are periodically withdrawn at the trap inlet and outlet and are subsequently analyzed for impurity concentrations to establish sorbent performance. The beds are also removed and analyzed following the tests to provide saturation values and data for material balance checks. In the recent test series, most of the runs were conducted using various combinations of the impurities, niobium pentafluoride, titanium tetrafluoride, ruthenium pentafluoride, tantalum pentafluoride, and antimony pentafluoride, and the sorbents, sodium fluoride at 650°F and magnesium fluoride at 250°F. The results of these simulated process sorption tests have been very encouraging. It was found that magnesium fluoride could selectively remove significant quantities of niobium pentafluoride, tantalum pentafluoride, titanium tetrafluoride, and antimony pentafluoride but was less effective than sodium fluoride for removing ruthenium pentafluoride. Scoping studies designed to evaluate new sorbents were also begun, and results obtained to date indicate that lithium fluoride and aluminum fluoride are particularly promising.

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

TABLE OF CONTENTS

	<u>Page</u>
INTRODUCTION	13
SUMMARY	14
EXPERIMENTAL DETAILS	16
EQUIPMENT	17
SORBENT PREPARATION	20
Sodium Fluoride	20
Magnesium Fluoride	22
Other Sorbents	24
IMPURITY PREPARATION	24
TESTING PROCEDURE	25
TEST RESULTS	26
Sorption of Niobium Pentafluoride	26
Sorption of Titanium Tetrafluoride	32
Sorption of Ruthenium Pentafluoride	36
Sorption of Tantalum Pentafluoride	39
Sorption of Antimony Pentafluoride	42
Performance of Other Sorbents	48
CONCLUSIONS AND RECOMMENDATIONS	50
ACKNOWLEDGEMENT	52
APPENDIX - Present Specifications for Uranium Hexafluoride Delivered to the U. S. Atomic Energy Commission	53

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

LIST OF TABLES

<u>Table</u>		<u>Page</u>
I	Effect of Preparation Temperature on the Physical Structure of Sodium Fluoride Pellets	22
II	Selected Physical Properties of Test Impurities	25
III	Selective Sorption of Niobium Pentafluoride from Uranium Hexafluoride	27
IV	Simultaneous Sorption of Niobium Pentafluoride and Tantalum Pentafluoride by 250°F Magnesium Fluoride Pellets	30
V	Sorption of Niobium Pentafluoride by 250°F Magnesium Fluoride Pellets	31
VI	Selective Sorption of Titanium Tetrafluoride from Uranium Hexafluoride	34
VII	Sorption of Titanium Tetrafluoride by 250°F Magnesium Fluoride Pellets	36
VIII	Selective Sorption of Ruthenium Pentafluoride from Uranium Hexafluoride	37
IX	Sorption of Ruthenium Pentafluoride by 250°F Magnesium Fluoride Pellets	39
X	Selective Sorption of Tantalum Pentafluoride from Uranium Hexafluoride	40
XI	Sorption of Tantalum Pentafluoride by 650°F Sodium Fluoride Pellets	41
XII	Sorption of Tantalum Pentafluoride by 250°F Magnesium Fluoride Pellets	43
XIII	Selective Sorption of Antimony Pentafluoride from Uranium Hexafluoride	44
XIV	Sorption of Antimony Pentafluoride by 250°F Magnesium Fluoride Pellets	45
XV	Sorption of Antimony Pentafluoride by 650°F Sodium Fluoride Pellets	46
XVI	Sorption of Antimony Pentafluoride by 250°F Magnesium Fluoride Pellets	47

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

LIST OF TABLES (Continued)

<u>Table</u>		<u>Page</u>
XVII	Analyses of Sorbent Samples Used in Scoping Tests	49
XVIII	Specifications for Uranium Hexafluoride Delivered to the AEC	54

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

LIST OF FIGURES

<u>Figure</u>		<u>Page</u>
1	Uranium Hexafluoride Impurity Trapping System	18
2	Uranium Hexafluoride Sample Cylinder	19
3	Sodium Fluoride Sorbent Pellets	21
4	Magnesium Fluoride Sorbent Pellets	23

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

REMOVAL OF IMPURITIES FROM URANIUM HEXAFLUORIDE
BY SELECTIVE SORPTION TECHNIQUES:
PROGRESS REPORT FOR 1966

For several years, the U. S. Atomic Energy Commission has placed limits on the types and quantities of various impurities permitted in uranium hexafluoride returned to its gaseous diffusion complex. Recently, with the advent of toll enrichment and other contractual arrangements for uranium ownership and processing services, these specifications have been revised and extended to cover all uranium hexafluoride delivered to the AEC. These latest criteria, listed in the appendix, deal with both physical and chemical properties of returned materials; included are 1 ppm restrictions for the metallic impurities niobium, titanium, ruthenium, tantalum, and antimony.

A program is under way at the Oak Ridge Gaseous Diffusion Plant to develop methods for removing volatile impurities from uranium hexafluoride. Successful development of purification techniques could, perhaps, allow the Commission to relax some of the more rigorous specifications for materials returned for cascade use, thereby reducing costs of feed preparation, scrap recovery, spent fuel treatment, and other uranium processing operations. The primary emphasis at the ORGDP has been on the use of solid sorbents which can selectively collect impurities from a uranium hexafluoride gas stream while allowing the uranium itself to pass through the system. Because of equipment simplicity and ease of operation, the sorption process appears to be generally preferable to other separation schemes, e.g., distillation, for this dilute mixture application. Relatively small and inexpensive sorbent systems could be placed either in feed lines at diffusion cascade sites or in uranium hexafluoride product systems at various processing installations.

The specific goal of the current ORGDP impurities program is to find materials which are effective sorbents for impurities present in uranium hexafluoride in the ppm range but which do not retain significant quantities of uranium. The work, therefore, is directed toward obtaining basic engineering data on the impurity sorption process and evaluating the total potential of various solid sorbents under study. Factors being considered include not only the basic stripping capabilities of the sorbent materials but also their availability or ease of manufacture and the severity of operating and pretreatment conditions. The results of previous experiments in this field were detailed in an earlier report*. This work included scoping studies designed to screen various sorbent-impurity combinations for applicability, as well as some fluorination-sorption tests utilizing uranium oxide spiked with impurities.

* Brater, D. C., Kaufman, H. L., Pashley, J. H., and Smiley, S. H., Removal of Impurities from Uranium Hexafluoride by Selective Sorption Techniques, Union Carbide Corporation, Nuclear Division, Oak Ridge Gaseous Diffusion Plant, June 6, 1966 (K-1666).

Because the flame reactor and fluid bed fluorination approaches taken in the earlier experiments were of somewhat limited usefulness in terms of sorption data collection, the experimental procedure was altered so that a volatile impurity fluoride stream could be blended with a uranium hexafluoride stream and immediately passed through a sorbent trap. This is the experimental approach which has been continued during the current series of tests. It should be noted, however, that this technique, direct spiking of uranium hexafluoride with impurity fluorides, is still, in some respects, a less desirable approach than the flame reactor and fluid-bed procedures. This is because fluorination of contaminated oxides, duplicating the standard uranium hexafluoride production route, probably offers more accurate representations of the exact chemical forms of impurities present in off-specification material. Thus, some risk may be involved in drawing conclusions from the spiked fluoride experiments. Considering the materials studied in the sorption pilot-plant runs, this is particularly true for ruthenium, where compounds other than ruthenium pentafluoride have been suggested as possible oxide fluorination products. On balance, however, it is felt that direct impurity addition to uranium hexafluoride, in addition to facilitating data collection, is reasonable and adequate for niobium, titanium, tantalum, and antimony; even for ruthenium, further tests of this type but involving other chemical forms of the element should provide sufficient additional data to allow confident selection of a sorbent material and satisfactory operating conditions.

The results of runs made during the past year, dealing with niobium, titanium, ruthenium, tantalum, and antimony fluoride impurities, are presented in this report following discussions of equipment details, sorbent and impurity preparation, and operating procedures. To aid in interpreting the recent results, previously reported test summaries are also included.

SUMMARY

In the past year, essentially all of the work performed at the ORGDP on uranium hexafluoride impurities stripping was centered around engineering-scale flow tests. Basically, the test system is equipped to deliver a uranium hexafluoride feed, containing some trace quantity of volatile impurity, to a sorption trap. The 3-inch-diameter Monel traps used in the experiments are usually filled to a height of 36 inches with the test bed material, and sample taps are provided on the inlet and outlet sides.

Based on preliminary work and on results attained in related National Laboratory programs, magnesium fluoride and sodium fluoride were selected as the primary sorbents for the recent program. Sodium fluoride, prepared by heating sodium bifluoride pellets in nitrogen for 5 hours at 1000°F, must be operated at about 650°F to keep uranium retention at an acceptably low level. Magnesium fluoride, however, can be used at a much lower temperature, i.e., 250°F, with a comparably low uranium holdup. This bed material is obtained from the Paducah Gaseous Diffusion Plant; further treatment of the pellets is necessary to remove water prior to the sorption runs. Impurity fluorides used to spike the uranium

hexafluoride are produced by reacting the base metals with elemental fluorine.

In this report, the conditions for and the results of recent tests are presented in detail, and related data from previous experiments are included for comparison purposes.

Considerable success has been achieved in six sorption tests, two made during the past year, aimed at removing niobium pentafluoride from uranium hexafluoride using either magnesium fluoride or sodium fluoride pellets. In fact, impurity loadings have typically averaged as high as 5 to 7% of the total bed weight for both sorbents. Of the two materials, however, magnesium fluoride is probably more desirable because of its lower operating temperature and because caking of pellets near the trap inlet, where the niobium holdup was high, was experienced in tests with sodium fluoride. Although sodium fluoride does have a demonstrated high capacity for niobium, it appears that, under certain operating conditions and especially at high niobium concentrations, the performance of this material is sensitive to changes in trap temperature and flow.

Several runs have been completed using titanium tetrafluoride as the impurity and either magnesium fluoride or sodium fluoride pellets as the sorbent. Transport problems, inherent in the difficult behavior of the titanium tetrafluoride, were encountered in almost every test, and the results of a number of these runs were disregarded because of inadequate impurity feed, analytical discrepancies, and/or sorber contamination. It has been demonstrated, however, during routine uranium processing in ORGDP recovery facilities, that magnesium fluoride can be used effectively to reduce the titanium level of slightly contaminated material, containing 2 or 3 ppm, to below the current 1 ppm titanium specification. This information, coupled with the results of a successful flow test with magnesium fluoride at low concentrations (9.8 ppm) of titanium, indicates that there should be no difficulty in adequately cleaning feed contaminated to the 10 ppm level.

Sodium fluoride at 650°F appears to be a better sorbent for ruthenium pentafluoride than magnesium fluoride; however, there were problems encountered with both materials when attempts were made to analyze the sorbent bed after termination of the run. Apparently, both the ruthenium-magnesium fluoride and ruthenium-sodium fluoride complexes are fairly unstable and tend to decompose and desorb. In this case, an oxyfluoride or perhaps an oxide would be suspected as the volatile species, but identification of the desorbed material was not possible. An alternative explanation is the presence of at least a portion of the ruthenium feed in some chemical form other than the pentafluoride, e.g., the hexafluoride or an oxyfluoride, which may not be so strongly bound by the sorbents tested. In summary, 650°F sodium fluoride does seem adequate for stripping ruthenium pentafluoride, but because both the capacity of sodium fluoride for other ruthenium forms and the stability of its complex with the pentafluoride are somewhat doubtful, further tests in this area are planned.

Three sorption tests were made to determine the effectiveness of 250°F magnesium fluoride and 650°F sodium fluoride for removal of tantalum pentafluoride from uranium hexafluoride. Stripping of tantalum with both sorbents was completely satisfactory. Furthermore, the average bed retention with magnesium fluoride, 5.3%, was particularly good. While the test results with sodium fluoride did not allow a comparable evaluation of the holding capacity of this material, the magnesium fluoride would probably be favored in any event because of its lower operating temperature.

Three antimony pentafluoride sorption tests were also completed this year. On the basis of operating temperatures and trap inlet and outlet stream impurity concentrations, magnesium fluoride at 250°F appears to be a better sorbent for the antimony than sodium fluoride. Either a short testing time or the absence of a pronounced breakthrough precluded collection of saturation data with either material, however, so that additional work with this impurity is also planned.

Some further scoping tests for other effective sorbents were conducted concurrently with a few of the sorption pilot-plant flow tests to evaluate quantitatively the capacity and selectivity of new materials for sorbing particular impurities. On the basis of these preliminary checks, lithium fluoride appears to be an excellent niobium stripper, as does aluminum fluoride for tantalum and, perhaps, for antimony. Calcium fluoride seems to be moderately successful with tantalum pentafluoride but probably not so good as magnesium fluoride. Results with mixed sodium and lithium fluorides were not particularly good.

In addition to effective impurity removal and low uranium retention, other important considerations in choosing sorbents for plant use include not only availability but also preparation and pretreatment requirements. Both sodium fluoride and magnesium fluoride are particularly attractive in this respect, because of their relatively low cost and the straightforward pretreatment procedures associated with their use. Similarly, techniques for preparing aluminum fluoride samples for the scoping studies were found to be about equally direct, so that this material may also be promising for plant-scale applications.

As can be seen, the work to date has been concerned more with evaluating the potentials of the various sorbent materials than with collecting scale-up data, although both aspects have certainly been considered. It is expected that development of information leading to engineering designs will be emphasized further in future testing programs.

EXPERIMENTAL DETAILS

During the past year, essentially all of the work performed at the ORGNP on uranium hexafluoride impurities stripping was centered around engineering-scale sorption flow tests. The impurities considered were niobium pentafluoride, titanium tetrafluoride, ruthenium pentafluoride, tantalum pentafluoride, and antimony pentafluoride. Two sorbent materials,

namely, sodium fluoride operated at 650°F and magnesium fluoride at 250°F, received most of the attention, although additional screening tests for other effective sorbents were begun late in the year. In this section, a description of the equipment, preparation methods for the sorbents and impurities used, a summary of the testing procedure, and a discussion of the experimental results are presented in turn.

EQUIPMENT

A schematic flow diagram for the engineering-scale impurity sorption system is shown in figure 1. Basically, the system provides a means of preparing a uranium hexafluoride feed containing some trace quantity of volatile impurity and subsequently passing this contaminated stream through a sorption trap. The design of the system allows both flow control of all constituent gases and temperature control throughout the traps, as well as sample points in trap inlet and outlet gas lines.

Uranium hexafluoride, sublimed from a heated storage tank, is metered by an orifice and, subsequently, fed to the trapping system. The volatile impurities, stored as liquid or solid fluorides, are similarly fed as vapors from feed cylinders contained within a temperature-controlled, electrically heated oven. During the tests, a small nitrogen carrier flow is diverted into the impurity cylinder(s) to entrain the volatile fluorides and to inject them into the uranium hexafluoride feed line upstream of the sorber. In most instances, control of the carrier gas flow has proved to be a satisfactory method for controlling the impurity level.

After the impurity is introduced, the uranium hexafluoride stream is directed into a 3-inch-diameter trap filled to a height of 30 to 36 inches with a solid sorbent bed material. The sorption system currently employed contains two Monel sorption traps which can be operated either individually or in parallel. These traps are contained within an electrically heated oven. Sample taps are provided on the inlet and outlet sides of the sorbent traps; each sample is pulled from the system through a small cold trap which is connected to the sample line and is immersed in a trichlorethylene-carbon dioxide slush bath. Figure 2 shows a typical sample cylinder, made of nickel.

The uranium hexafluoride, after passing through the sorption system, is directed into a series of cold traps. These traps are maintained at a temperature of around minus 28 to minus 35°F in a refrigerated trichlorethylene bath. Here, all condensables, including the fluoride impurity, are desublimed and the off-gas, consisting of a mixture of nitrogen and fluorine, is then vented to atmosphere.

Figure 1

URANIUM HEXAFLUORIDE IMPURITY TRAPPING SYSTEM

Figure 2
URANIUM HEXAFLUORIDE SAMPLE CYLINDER

SORBENT PREPARATION

Based on preliminary scoping tests* and on results attained in related National Laboratory programs, magnesium fluoride and sodium fluoride were selected as the primary sorbents for a more extensive testing program. This decision stemmed from the excellent combined capability of the two materials to strip all five of the volatile impurities presently under consideration without sorbing significant quantities of uranium. Perhaps equally important, these two materials are readily available and relatively inexpensive. In conjunction with the regular testing program, other sorbent materials have also been prepared.

Sodium Fluoride

The sodium fluoride pellets used in the sorption tests were prepared by heating commercially available† sodium bifluoride pellets in a nitrogen atmosphere for 5 hours at 1000°F. The bifluoride pellets are approximately 1/8-inch by 1/8-inch right circular cylinders as shown in figure 3. The pellet surface area, as determined by nitrogen adsorption, ranges from 1 to 2 sq m/g. For example, in two recent pellet batches, the surface areas were found to be 1.86 and 1.68 sq m/g. A void fraction between 0.35 and 0.50 is common.

In another study aimed at defining conditions for uranium hexafluoride sorption on sodium fluoride‡, commercial sodium fluoride, rather than bifluoride, pellets were purchased directly. These pellets were prepared by the vendor using a two-step process consisting of compaction of powdered sodium bifluoride followed by heating in air at a temperature near 570°F. Prior to use, the pellets were fluorinated for 1 hour at 750°F. These pellets were reported to have a surface area of 0.856 sq m/g, an average pore radius of 0.791 micron, and a void fraction of 0.40.

Table I presents some results from still another uranium hexafluoride sorption program§ in which the effects of preparation temperature on the surface area, porosity, and hardness of sodium fluoride pellets were determined. As can be seen from the table, severe sintering of the sodium fluoride pellets resulted from treatment temperatures in excess of 1000°F. This decrease in pellet surface area and porosity renders the pellet less active as a solid sorber and reduces its loading capability.

* K-1666, loc. cit.

† Technical Grade No. NA-0101T-1/8, Harshaw Chemical Company.

‡ McNeese, L. E., An Experimental Study of Sorption of Uranium Hexafluoride by Sodium Fluoride Pellets and a Mathematic Analysis of Diffusion with Simultaneous Reaction, Union Carbide Corporation, Nuclear Division, Oak Ridge National Laboratory, November 29, 1963 (ORNL-3494).

§ Smiley, S. H., Brater, D. C., Pashley, J. H., ORGDP Fuel Reprocessing Studies Summary Progress Report Fiscal Year 1964 through Fiscal Year 1965, Union Carbide Corporation, Nuclear Division, Oak Ridge Gaseous Diffusion Plant, October 29, 1965 (K-1649).

Figure 3
SODIUM FLUORIDE SORBENT PELLETS

TABLE I

EFFECT OF PREPARATION TEMPERATURE ON THE PHYSICAL STRUCTURE
OF SODIUM FLUORIDE PELLETS*

Treatment Temperature, °F	Surface Area, sq m/g	Void Fraction	Stokes Hardness, division†
1000	2.149	0.406	1.0
1095	0.118	0.120	5.5
1200	0.061	0.104	6.5
1290	0.031	0.038	8.0
1400	0.060	0.097	8.0

* K-1649, loc. cit.

† Comparative scale.

An average of about 10 pounds of sodium fluoride pellets is required to charge one of the 3-inch-diameter sorption traps to a height of 36 inches. The nominal bulk density of the pellets is 66 lb/cu ft. With sodium fluoride, the sorption trap is maintained at 650°F, because at lower bed temperatures, say 120 to 250°F, approximately 1 pound of uranium hexafluoride per pound of sodium fluoride can be retained*. At 650°F, uranium pickup is on the order of 0.1% or less, which is tolerable.

Magnesium Fluoride

The magnesium fluoride pellets used in all of the sorption runs made to date were produced at the Paducah Gaseous Diffusion Plant by tumbling moistened magnesium fluoride powder. The resulting pellets are spheroidal and are 3/16 to 5/16 inch in diameter (figure 4). Initially, the as-prepared pellets contain 7 to 8% water by weight and, therefore, must be dried prior to the sorption runs. The usual drying procedure consists of heating the pellets to 400°F in nitrogen for at least 4 hours. Following the drying, the pellets are exposed in a fluorine atmosphere for 2 hours at 250°F. After this preparatory treatment, the pellets have a water content of less than 1% and a nominal 44 sq m/g surface area. Vacuum drying has also been employed on occasion. It is interesting to note that, after pretreatment in fluorine, the pellets were found to have surface areas much higher than usually attained with nitrogen drying. Two

* ORNL-3494, loc. cit.

Figure 4
MAGNESIUM FLUORIDE SORBENT PELLETS

surface area determinations after the vacuum treatment yielded values of 78.5 and 99.5 sq m/g.

When filled to a height of 36 inches, a sorbent trap holds between 8 and 9 pounds of magnesium fluoride pellets; the average bulk density is near 56 pounds per cubic foot. For the sorption runs with magnesium fluoride, the trap can be maintained at 250°F. At this temperature, uranium pick-up on the magnesium fluoride is insignificant, yet the sorbent is effective in removing impurities from the gas feed stream.

Other Sorbents

In a search for even more effective sorbent materials, scoping tests were conducted this year simultaneously with some of the regular runs. Small samples of the test materials, lithium fluoride-sodium fluoride composite, lithium fluoride, aluminum fluoride, and calcium fluoride were placed in separate perforated containers and were suspended above the sorbent beds. The primary interest in these initial tests was in the actual test sorbent capacity and selectivity for the particular impurity. More definitive sorption rate data, important for fixing trap residence time requirements, will be collected for the more promising materials in future experiments.

Lithium fluoride was prepared by fluorinating fused lithium carbonate and breaking the product into small irregular pieces. In one determination, the resulting surface area was found to be 6.72 sq m/g. By chemical analysis, the fluoride content of the material was determined to be 66.9% (as compared to 73.25% theoretical), with a lithium content of 22.0%. The principal remaining element was probably oxygen. Two batches of a sodium-lithium fluoride complex sorbent were made in the same manner used to prepare the lithium fluoride. In the first sample, 25 grams of lithium carbonate was dry blended with 75 grams of sodium fluoride, and the mixture was fused at 1000°F and then fluorinated. The resulting chunks had a surface area of 2.18 sq m/g. By chemical analysis, the fluoride content was 42.5%, and the lithium content was 4.0%. The second sample batch was composed initially of 75% sodium carbonate and 25% lithium carbonate. The final material had a surface area of 4.81 sq m/g, and the chemical analysis showed a fluoride content of 48.8% and a lithium content of 3.8%.

Calcium fluoride and aluminum fluoride samples were prepared by direct fluorination of 6-mesh calcium sulfate (Drierite)* and activated alumina spheres, respectively. The resulting surface areas were 249.7 sq m/g for the calcium fluoride and 55.2 sq m/g for the aluminum fluoride.

IMPURITY PREPARATION

Selected physical properties of the fluoride impurities used in the recent sorption tests are listed in table II. These compounds are all produced

* Product of the W. A. Hammond Drierite Company.

locally by direct fluorination in a 4-inch-diameter, 4-foot-long, nickel muffle furnace. Batches of the starting materials, which are normally the base metals*, are loaded into nickel pans, placed inside the reactor, and treated with undiluted commercial grade fluorine. Fluorination temperatures are different for the preparation of the various fluoride impurities but are below 1000°F. Fluorine rates are less than 0.5 lb/hr. The volatilized fluorides exiting from the muffle system are trapped directly by condensation or desublimation in the feed cylinders used in the sorption tests.

TABLE II
SELECTED PHYSICAL PROPERTIES OF TEST IMPURITIES*

Compound	Molecular Weight	Melting Point, °F	Boiling Point, °F
Niobium pentafluoride	187.90	ca. 162	457
Titanium tetrafluoride	123.89	> 750	543 (sublimes)
Ruthenium pentafluoride	196.06	214	482
Tantalum pentafluoride	275.94	206	445
Antimony pentafluoride	216.74	44.6	301

* Weast, R. C., et al, Handbook of Chemistry and Physics, 45th Edition, The Chemical Rubber Company, Cleveland, Ohio (1964).

TESTING PROCEDURE

The main experimental effort of each sorption run involves collecting representative samples of the contaminated uranium hexafluoride feed stream entering and leaving the sorption trap at specified time intervals from the start of the test. At the completion of each test, incremental sections of the sorption bed are also sampled to provide loading data.

Upon charging the sorbent trap with pellets, pretreating, and placing the impurity feed cylinder in the feed oven, the various sections of the apparatus are brought to temperature, and the gas flows are established. Typical flow conditions for the tests are 20 lb/hr uranium hexafluoride,

* Fluorination of niobium oxide to form niobium pentafluoride has also been done successfully.

1.5 lb/hr fluorine, and 0.2 scfm nitrogen diluent. The superficial velocity corresponding to these conditions varies with operating temperature but is near 0.5 ft/sec.

Once operation of the sorption system has begun, samples of condensables entering and leaving the sorber are withdrawn periodically, generally every 2 to 5 hours. Approximately 5 minutes is required to remove a 40- to 50-gram sample, and from 20 to 30 samples are usually taken during a test. The total amount of uranium hexafluoride fed during a run is determined from a cumulative record of cold trap inventory, allowing, of course, for the small amount of material removed from the system as samples.

After completion of a run, the sorption system is purged with nitrogen for several hours, and the contents of the sorption trap are emptied pneumatically. This method of removing the sorbent pellets may be somewhat objectionable in terms of material losses, but no other method short of complete disassembly of the apparatus can be used conveniently with the present system. Incremental samples of the bed are removed from the trap and are individually analyzed after ball-milling to establish an impurity sorption profile and to determine uranium pickup.

TEST RESULTS

Sorption test conditions and experimental results for runs conducted during 1966 are presented in the following sections. For comparison purposes, discussions of the recent work are preceded by summaries of previously reported data from earlier tests*.

Sorption of Niobium Pentafluoride

Considerable success has been achieved in six tests (two made during the last year) aimed at removing niobium pentafluoride from uranium hexafluoride by selective sorption using either magnesium fluoride or sodium fluoride pellets. Of the two materials, however, magnesium fluoride is probably the more desirable because it can be operated with low uranium holdup at a considerably lower temperature than can sodium fluoride; e.g., 250 versus 650°F. Also, there were some trap plugging problems at high niobium concentrations experienced in the sodium fluoride tests. A summary of the data obtained in these tests is presented as table III.

Previous Work. Two sorption runs were made using sodium fluoride pellets as the sorbent bed material. For the first test, which lasted 26.9 hours, the system was operated with a uranium hexafluoride feed rate of 21.0 lb/hr and a fluorine rate of 1.5 lb/hr. The sorbent trap was charged with 13 pounds of pellets and was maintained at a temperature of 750°F. A total of 565 pounds of uranium hexafluoride was fed through the trap. Analyses of condensables from the sorber inlet and outlet streams showed

* K-1666, loc. cit.

TABLE III

SELECTIVE SORPTION OF NIOBIUM PENTAFLUORIDE FROM URANIUM HEXAFLUORIDE

	Run Number					
	I-2	I-3	I-4	I-6*	I-28	I-29
Reference	†	†	†	†	‡	‡
Run Duration, hr	26.9	41.7	83.7	58.8	47.4	80.3
Sorbent	NaF	NaF	MgF ₂	MgF ₂	MgF ₂	MgF ₂
Sorbent Temperature, °F	750	750	255	250	250	250
Uranium Hexafluoride Rate, lb/hr	21.0	17.4	19.2	20.0	21.7	22.9
Average Inlet Niobium Concentration, ppm	409	345	294	455	287	465
Average Outlet Niobium Concentration Before Breakpoint, ppm	0.5	1.2	0.8	1.6	< 1	7.8
Time Until Breakpoint, hr	not reached	not reached	not reached	12.5	23	25
Average Outlet Niobium Concentration After Breakpoint, ppm	-	-	-	218	32	171
Average Impurity Loading on Bed, weight percent metal	5.8	7.2	5.2	5.5	2.80	4.7§
Uranium Holdup on Bed, weight percent of total uranium fed	0.00136	0.000795	0.00133	0.000145	0.00198	0.00250

27

* Twelve-inch rather than 36-inch-deep sorbent bed used for this run.

† K-1666, loc. cit.

‡ Present work.

§ This value estimated using graphical techniques.

that the uranium hexafluoride feed contained an average of 409 ppm niobium*, while the outlet contained about 0.5 ppm niobium during the entire run. Breakthrough of the niobium did not occur. The pressure drop across the sodium fluoride trap increased from 0.5 to 0.8 psi over the first 23.7 hours of operation. The trap continued to plug and had to be removed from service after 26.9 hours of operation. Inspection of the trap revealed a hard cake at the trap inlet (for about the first 2 inches of the bed), but the remainder of the pellets was free-flowing. Chemical analysis of the gray cake material showed niobium and uranium concentrations of 15.9 and 0.2%, respectively. In the first 4 inches of the sorbent bed, the sodium fluoride pellets held 10.0% niobium; while in the last 12 inches, the niobium concentration was less than 35 ppm niobium. Uranium holdup on the bed was insignificant.

For the second sorption test using sodium fluoride, the system was operated for 41.7 hours at a uranium hexafluoride feed rate of 17.4 lb/hr and a fluorine flow of 1.5 lb/hr. The sorption trap was filled with 13.0 pounds of pellets and was again held at a temperature of 750°F. Analyses of samples taken from the uranium hexafluoride feed stream showed an average inlet niobium concentration of 345 ppm. Until the completion of the run, the sorber outlet contained an average 1.2 ppm niobium. This run was also terminated because of plugging, as was experienced with the previous test. At the start of the sorption run, the sodium fluoride trap pressure drop was 0.46 psi, but after 22 hours of operation, this value increased to 0.72 psi. When the test was finally ended, the trap pressure drop had risen to 9.5 psi. Inspection of the trap bed material revealed a hard cake in the first 2 inches of the bed, as before. Analyses of bed segments showed an average 11.4% niobium and 276 ppm uranium in the first 4 inches (inlet side) of the bed and less than 300 ppm niobium and 100 ppm uranium for the last 12-inch increment.

Although sodium fluoride does have a demonstrated high capacity for niobium, it appears that, under certain operating conditions and especially at high niobium concentrations, the performance of this material is sensitive to changes in temperature and flow. Also, it should be noted that the test temperatures were 100°F higher than used in later runs with sodium fluoride. Therefore, some of the problems experienced at the higher temperatures might be ameliorated by operation at 650°F.

Two sorption tests with niobium pentafluoride and magnesium fluoride were made previously. In the first of these tests, the sorber was operated for 83.7 hours with a uranium hexafluoride rate of 19.2 lb/hr for a total of 1,607 pounds of uranium hexafluoride fed. The sorption trap contained 8.37 pounds of magnesium fluoride, and a sorbent temperature of 255°F was maintained. The pressure drop across the traps remained constant over the entire operating period. Analyses of condensables entering and leaving the sorber showed the average inlet and outlet niobium concentrations

* The concentrations of impurities in uranium hexafluoride and on sorbent materials are reported on a metal-to-uranium and metal-to-sorbent weight basis unless otherwise specified.

to be 294 and less than 0.8 ppm, respectively, with no breakthrough of the niobium indicated. Sorbent bed analyses showed a loading of 5.21% niobium and 0.17% uranium. The uranium holdup value represents only 0.00133% of the total uranium passed through the trap.

The second niobium test with magnesium fluoride was designed to push the system to the breakthrough point. For this test, lasting 58.8 hours, the system was operated with a uranium hexafluoride rate of 20.0 lb/hr and a fluorine rate of 1.5 lb/hr. The sorbent trap was charged to a height of about 12 inches with 2.875 pounds of magnesium fluoride pellets. The niobium concentration in the trap inlet stream averaged 455 ppm. Analyses of condensables leaving the sorber showed that breakthrough of the niobium occurred after about 12.5 hours of operation, at which point, the niobium content increased from 0.4 ppm to 315 ppm. After breakthrough, the niobium concentration in the outlet stream averaged 218 ppm for the remaining 40 hours of the test. It is interesting to note that the sorbent continued to remove over half of the niobium from the uranium hexafluoride feed even after breakthrough. This phenomenon, which is characteristic of the sorption systems presently under consideration, can probably be explained by a sorption mechanism similar to a catalysis mechanism and appears to be the result of combined diffusion and reaction rate effects. Analyses of the sorbent bed showed that the magnesium fluoride pellets held averages of 5.48% niobium and 0.04% uranium. Again, the uranium retention was negligible.

Recent Test Results. Two sorption runs were made in the latest series with niobium pentafluoride as the impurity and magnesium fluoride as the sorbent. The test results are presented in tables IV and V. The first of these runs was an attempt to remove simultaneously two different impurities, niobium and tantalum, from a uranium hexafluoride stream. In this test, the system was operated for 47.4 hours with a uranium hexafluoride feed rate of about 21.7 lb/hr, a fluorine rate of 1.5 lb/hr, and a nitrogen diluent flow of 0.23 scfm. The sorbent trap was filled with 7.94 pounds of magnesium fluoride pellets and was maintained at 250°F. A total of 1,031 pounds of uranium hexafluoride, containing an estimated 0.192 pound of niobium and 0.0234 pound of tantalum, was passed through the trap. Analyses of samples taken at the trap inlet showed average niobium and tantalum concentrations of 287 and 31 ppm, respectively. Analyses of samples from the trap outlet stream, given in table IV, indicated excellent removal of both niobium and tantalum, with niobium being reduced to less than 1 ppm for the first 23 hours of operation, after which time, breakthrough occurred. The tantalum was reduced to less than 1 ppm for the entirety of the run. Analysis of the bed showed a total of 0.216 pound of niobium (2.80% of the final total bed weight), 0.0297 pound of tantalum, and 0.014 pound of uranium adsorbed in the trap. Only moderate loading of the niobium on the bed was achieved. The uranium holdup represented a typically negligible 0.0020% of that passed through the system.

The second recent niobium run was an extended test lasting 80.3 hours and was designed to define the sorption breakthrough curve more completely. In this test, the system was operated with a uranium hexafluoride feed

TABLE IV

SIMULTANEOUS SORPTION OF NIOBIUM PENTAFLUORIDE AND TANTALUM PENTAFLUORIDE
BY 250°F MAGNESIUM FLUORIDE PELLETS

Run Number	On-Stream Time, hr	Concentrations, ppm				Bed Segment, in.	Sorbent Analysis		
		Niobium		Tantalum			Niobium, %	Tantalum, %	Uranium, %
		Inlet	Outlet	Inlet	Outlet				
I-28	3.2	204	0.4	24	< 1	Inlet 6	3.63	0.51	0.05
	7.2	249	0.4	30	< 0.5	Second 6	3.78	0.41	0.17
	15.4	266	0.4	-	< 0.5	Third 6	3.19	0.33	0.15
	19.4	287	1.2	-	< 0.5	Fourth 6	2.47	0.27	0.19
	23.4	305	1.0	12	< 0.5	Fifth 6	1.78	0.27	0.28
	25.4	236	18	24	< 1	Sixth 6	1.18	0.63	0.13
	27.4	332	23	23	< 1				
	29.4	231	33	19	< 1				
	31.4	265	31	33	< 1				
	33.4	274	35	27	< 1				
	37.4	324	41	22	< 1				
	39.4	380	33	44	< 1				
	43.4	286	27	46	< 1				
	46.9	324	35	29	< 1				

TABLE V

SORPTION OF NIOBIUM PENTAFLUORIDE BY 250°F MAGNESIUM FLUORIDE PELLETS

Run Number	On-Stream Time, hr	Niobium Concentrations, ppm		Sorbent Analysis		
		Inlet	Outlet	Bed Segment, in.	Niobium, %	Uranium, %
I-29	1.5	488	< 0.5	Inlet 6	2.80	0.53
	5.5	141	1.5	Second 6	2.07	0.45
	10.5	68	8.0	Third 6	1.80	0.23
	15.5	233	16.0	Fourth 6	1.69	0.41
	20.5	294	43.0	Fifth 6	2.62	0.66
	25.5	468	18			
	30.3	529	138			
	35.3	541	148			
	40.3	378	44			
	45.3	407	65			
	50.3	850	283			
	55.3	867	250			
	60.3	600	247			
	70.3	510	153			
	80.3	607	213			

rate of 22.9 lb/hr, a fluorine rate of 1.5 lb/hr, and a nitrogen flow of 0.23 scfm. The sorbent trap was charged with 8.62 pounds of magnesium fluoride pellets and was maintained at 250°F. A total of 1,843 pounds of uranium hexafluoride, containing 0.581 pound of niobium, was fed through the sorber. By analyses of condensables entering and leaving the magnesium fluoride trap, summarizing data shown in table V, the inlet niobium concentration was found to average 465 ppm, with a trap outlet concentration of about 7.8 ppm prior to breakthrough. Breakthrough occurred after about 25 hours of operation, after which, the trap outlet stream averaged 171 ppm niobium for the remainder of the test. Determination of pellet bed loading by chemical analysis of the magnesium fluoride showed a total of 0.190 pound of niobium (2.15% average) and 0.0393 pound uranium (0.44% average) sorbed on the magnesium fluoride. Compared with previous runs, the niobium holdup again appeared to be lower by a factor of two or more. On the other hand, a material balance can be made by graphically integrating the sorber inlet and outlet stream niobium concentrations over the length of the run and subtracting the two values. Using this procedure, which has proved to be generally reliable for estimating bed loading*, the niobium holdup was calculated to be 0.453 pound, or 4.7% of the final bed weight. This estimate of bed loading gives a much better material balance and is also in substantial agreement with results from earlier tests. Perhaps some of the niobium-magnesium fluoride complex (identified by other workers as magnesium hexafluoronioibat[†]) was lost in sampling or analytical operations. This loss, if, of course, real instead of merely an indication of analytical and/or sampling uncertainties, possibly could have been in the form of powdered complex sloughed from the surface of the pellets.

In summary, 250°F magnesium fluoride seems to be completely adequate for removal of niobium, even when it is present in high concentrations. Sodium fluoride is also an effective sorber but, in fact, tends to overload and form cakes (if there is a substantial amount of niobium in the feed), at least at the 750°F operating temperatures employed in the tests made to date.

Sorption of Titanium Tetrafluoride

Several runs have been completed using titanium tetrafluoride as the impurity and either magnesium fluoride or sodium fluoride as the sorbent. Transport problems inherent in the difficult behavior of the titanium tetrafluoride were encountered in almost every test, and the results of

* For example, in the two-impurity test, the graphical technique predicted a loading of 2.30% niobium and 0.35% tantalum, compared with 2.80% and 0.39%, respectively, found analytically.

† Prusakov, V. N., Ezhov, V. K., Lebedev, O. G., and Popov, V. K., "Some Aspects of Nuclear Fuels Reprocessing by Fluorination Method", I. V. Kurchatov Atomic Energy Institute, Moscow, USSR. Presented at the XXXVI International Congress on Industrial Chemistry, Brussels (September, 1966).

a number of these runs, including those made last year, had to be disregarded because of inadequate impurity feed, analytical discrepancies, and/or sorber contamination. A summary of the more meaningful data obtained in past work with titanium is given in table VI, which also includes unreported results from an earlier sorption experiment.

Previous Work. Two sorption runs, one lasting 50.9 hours and the other 17.2 hours, were made with titanium tetrafluoride using 650°F sodium fluoride pellets as the sorbent. In the 50.9-hour test, the system was operated with a uranium hexafluoride feed rate of 19.9 lb/hr and a nitrogen flow rate of 0.26 scfm. The sorbent trap was charged with 9.75 pounds of pellets. A total of 1,013 pounds of uranium hexafluoride, containing an estimated 0.218 pound of titanium, was fed through the bed. Analyses indicated an average inlet titanium concentration of 317 ppm, while the concentration at the outlet averaged 116 ppm, with apparent breakthrough within the first few hours of the run*. The sorbent bed was found to contain 0.203 pound of titanium, a value which agrees fairly well with the estimate of the total quantity fed and with the inlet and outlet stream analyses. Bed analyses also showed an average uranium holdup on the order of 0.003%, which represented only a trace of the total uranium fed.

The results of the second titanium run using sodium fluoride as the sorbent were very erratic. In fact, sorber outlet titanium concentrations were found to be as high or higher than inlet concentrations. There was a considerable quantity of titanium indicated by analyses to be present in the uranium hexafluoride feed stream even before the impurity cylinder was valved in, so that the sorption system apparently was contaminated with material which had deposited in the feed system during previous tests.

Eight titanium tetrafluoride sorption runs have been completed using magnesium fluoride as the solid sorbent. At the beginning of this work, feeding problems were encountered, and the first five runs were completed with less than 10 ppm. titanium present in the feed material.

After the impurity feed apparatus was modified for the titanium system, these problems were eased considerably, and a sixth test with magnesium fluoride was made. In this test, which lasted 10.1 hours, the sorber system was operated with a nominal uranium hexafluoride rate of 20.0 lb/hr and a nitrogen flow rate of 0.26 scfm. Based on analyses of sorber inlet samples, a total of 0.0204 pound of titanium tetrafluoride was fed with the uranium hexafluoride through the bed. The results are confusing because outlet samples from the trap indicated that essentially no titanium was retained by the sorbent. Bed analyses, on the other hand, showed an average titanium holdup of 1.14%. This discrepancy was probably a result of the previously-noted transport problems associated with titanium tetrafluoride handling. The difficulties in obtaining representative inlet and outlet sorber samples become particularly magnified with concentrated titanium tetrafluoride systems, since it has been observed that titanium

* The outlet titanium concentration was 15 ppm after 1.9 hours of operation and 141 ppm after 3.9 hours.

TABLE VI

SELECTIVE SORPTION OF TITANIUM TETRAFLUORIDE FROM URANIUM HEXAFLUORIDE

	Run Number				
	I-17	I-8	I-14	I-16	I-18
Reference	*	†	*	*	*
Run Duration, hr	50.9	23.25	10.1	18.1	42.0
Sorbent	NaF	MgF_2	MgF_2	MgF_2	MgF_2
Sorbent Temperature, °F	650	255	250	250	250
Uranium Hexafluoride Rate, lb/hr	19.9	20.2	20.0	22.7	20.5
Average Inlet Titanium Concentration, ppm	317	9.8	58	68	158
Average Outlet Titanium Concentration Before Breakpoint, ppm	15	0.6	58	61	128
Time Until Breakpoint, hr	1.9	not reached	not apparent	not apparent	not apparent
Average Outlet Titanium Concentration After Breakpoint, ppm	115	-	-	-	-
Average Impurity Loading on Bed, weight percent metal	2.06	-	1.14	4.64	1.6
Uranium Holdup on Bed, weight percent of total uranium fed	0.00004	-	0.0110	0.00926	0.0371

* K-1666, loc cit.

† Unreported data from previous test.

tetrafluoride vapor tends to deposit on confining solid surfaces, such as feed and sample lines.

The seventh titanium tetrafluoride-magnesium fluoride test lasted 18.1 hours. The sorption system was operated with a uranium hexafluoride feed rate of 22.7 lb/hr and a nitrogen flow rate of 0.26 scfm. The trap was filled with 7.81 pounds of magnesium fluoride and was maintained at 250°F. As before, analyses of condensables entering and leaving the sorber did not agree with bed loading values obtained by pellet analyses after completion of the run. The inlet and outlet feed streams averaged 68.5 and 61.0 ppm titanium, respectively; however, the sorbent bed contained an average of 4.64% titanium (0.36 pound titanium) and 0.33% uranium. Conflicting with the bed analyses, the sorber inlet and outlet stream data predicted only 0.0189 pound of titanium fed and only 0.0050 pound retained. Again, there apparently was difficulty in obtaining representative gas stream samples.

The results of the eighth test with titanium tetrafluoride conformed to this general pattern. For this test, which lasted 42.0 hours, a uranium hexafluoride feed rate of 20.5 lb/hr and a nitrogen flow rate of 0.23 scfm were maintained. The sorbent trap was loaded with 7.77 pounds of magnesium fluoride. Analyses of material entering and leaving the sorber indicated 0.092 pound of titanium metal fed and only 0.018 pound held on the bed. Analyses of the sorbent pellets, however, showed average concentrations of 1.6% titanium (equivalent to 0.140 pound titanium) and 2.78% uranium, thereby indicating retention of most of the titanium fed. The uranium holdup corresponded to a uranium loss of 0.037%.

Other Data. One additional test, conducted as a part of the previous testing program, is of particular interest now. In this test, the system was operated for 23.25 hours with a uranium hexafluoride feed rate of 20.2 lb/hr, a fluorine rate of 1.5 lb/hr, and a nitrogen pick-up flow of 4 scfm. The sorbent trap was charged to a height of 12 inches with 2.93 pounds of magnesium fluoride pellets and was maintained at 255°F. A total of 467 pounds of uranium hexafluoride, containing an average 9.8 ppm titanium, was passed through the trap. As shown in table VII, analyses of condensables from the trap outlet showed the average titanium concentration to be 0.6 ppm during the entire run. Considering the fact that the titanium concentration in the uranium hexafluoride feed actually approximated the contamination level expected in most of the cascade returns which do not meet specifications, the stripping action during the test was excellent. Bed analyses were not made, however, and loading could not be checked.

TABLE VII

SORPTION OF TITANIUM TETRAFLUORIDE
BY 250°F MAGNESIUM FLUORIDE PELLETS

Run Number	On-Stream Time, hr	Titanium Concentrations, ppm	
		Inlet	Outlet
I-8	0.75	9	0.8
	3.0	31	0.6
	7.0	6	0.4
	11.0	6	0.4
	15.0	7	0.6
	19.0	6	0.6
	23.0	4	0.8

It has been demonstrated during routine uranium processing in ORGDP recovery facilities that magnesium fluoride can be used effectively to reduce the titanium level of slightly contaminated material (containing 2 or 3 ppm) to below the current 1 ppm titanium specification. This information, coupled both with the results of the flow test with magnesium fluoride at low concentrations (9.8 ppm) of titanium and with the fact that this sorbent has been found to retain significant amounts of titanium, indicates that magnesium fluoride is probably adequate for cleaning feed contaminated to the 10 ppm level. Additional tests with this impurity will be made.

Sorption of Ruthenium Pentafluoride

A summary of results obtained to date for ruthenium pentafluoride sorption is contained in table VIII. Sodium fluoride at 650°F appears to be a better sorbent for ruthenium pentafluoride than magnesium fluoride at 250°F; however, there were problems encountered in both cases when attempts were made to analyze the sorbent bed after termination of the run. Apparently, both the ruthenium-magnesium and ruthenium-sodium fluoride complexes (identified as magnesium hexafluororuthenate and sodium hexafluororuthenate*, respectively) are fairly unstable and tend to decompose and

* Prusakov, V. N., et al, loc cit.

TABLE VIII
SELECTIVE SORPTION OF RUTHENIUM PENTAFLUORIDE
FROM URANIUM HEXAFLUORIDE

	Run Number	
	I-15	I-27
Reference	*	†
Run Duration, hr	64	8.5
Sorbent	NaF	MgF ₂
Sorbent Temperature, °F	650	250
Uranium Hexafluoride Rate, lb/hr	21.0	23.8
Average Inlet Ruthenium Concentration, ppm	251	204
Average Outlet Ruthenium Concentration Before Breakpoint, ppm	2	23.4
Time Until Breakpoint, hr	21	not apparent
Average Outlet Ruthenium Concentration After Breakpoint, ppm	72	-
Average Impurity Loading on Bed, weight percent metal	1.79(1.85‡)	0.33‡
Uranium Holdup on Beds, weight percent of total uranium fed	0.0070	0.0725

* K-1666, loc cit.

† Present work.

‡ Values obtained using graphical techniques.

desorb when exposed to room air. An oxyfluoride or perhaps even an oxide would therefore be suspected as the volatile species, but identification of the desorbed material was not possible. An alternative explanation is the presence of at least some of the ruthenium feed in some chemical form other than the pentafluoride, e.g., the hexafluoride or an oxyfluoride, which may not be so strongly bound by the sorbents tested.

Previous Work. A sodium fluoride pellet trap was used for the first sorption test with ruthenium pentafluoride. The sorber was operated for 64 hours with a uranium hexafluoride rate of 21.0 lb/hr, a fluorine rate of 1.5 lb/hr, and a nitrogen flow of 0.26 scfm. The trap was filled with 9.88 pounds of sodium fluoride pellets and was maintained at 650°F. About 1,350 pounds of uranium hexafluoride (with an estimated 0.23 pound ruthenium) was passed through the trap. Analyses of condensables entering and leaving the sorber showed an average inlet ruthenium concentration of 251 ppm and an average outlet ruthenium concentration of less than 2 ppm before breakthrough, which occurred after 21 hours of operation. For the remainder of the test, the sorber outlet averaged 72 ppm ruthenium. Inspection of the sorbent bed revealed a considerable amount of fines, indicating excessive surface degradation of the sodium fluoride pellets. Chemical analyses of the sorbent bed showed average ruthenium and uranium contents of 1.79 and 0.65%, respectively. It appeared that some of the ruthenium was desorbed from the bed prior to analysis, in which case, the 1.79% value, corresponding to 0.177 pound ruthenium, would be low. However, a graphical estimate, based on inlet and outlet stream ruthenium compositions, placed the sorption at 0.183 pound ruthenium, thereby indicating only a slight loss of the trapped material upon exposure to air.

Recent Test Results. The second sorption test with ruthenium, summarized in table IX, was made using 250°F magnesium fluoride pellets. For this test, the system was operated for 9.3 hours with a uranium hexafluoride feed rate of 23.8 lb/hr, a fluorine rate of 1.5 lb/hr, and a nitrogen flow of 0.23 scfm. The sorbent trap was charged with 7.88 pounds of pellets. A total of 220 pounds of uranium hexafluoride, containing approximately 0.028 pound of ruthenium, was passed through the trap. Analyses of condensables entering and leaving the sorber showed average ruthenium concentrations of 204 and 23.4 ppm, respectively, in the uranium hexafluoride. The outlet concentration is an order of magnitude higher than in the comparable test with sodium fluoride. Using graphical methods to estimate bed loading, 0.0259 pound of ruthenium was predicted for an overall bed average of 0.33%. Two 6-inch bed increments near the trap inlet and outlet were analyzed and were found to contain 0.95 and 0.42% ruthenium, respectively. Desorption of material from the pellets after their removal from the trapping system was again noted, but because of the lower overall loading, this release appeared to be less vigorous than that noticed with sodium fluoride.

TABLE IX

SORPTION OF RUTHENIUM PENTAFLUORIDE
BY 250°F MAGNESIUM FLUORIDE PELLETS

Run Number	On-Stream Time, hr	Ruthenium Concentrations, ppm	
		Inlet	Outlet
I-27	0.5	330	43
	1.5	204	19
	2.5	279	22
	3.5	244	27
	4.5	227	19
	5.5	200	15
	6.5	168	19
	7.5	119	16
	8.5	65	31

In summary, 650°F sodium fluoride seems adequate for stripping ruthenium pentafluoride and, in terms of ruthenium loading, appears to be preferable to 250°F magnesium fluoride. As was the case with other impurities, uranium losses with both materials were found to be negligible. Both the capacity of sodium fluoride for other ruthenium forms and the stability of its complex with the pentafluoride are, however, somewhat doubtful, and further tests in this area are already under way.

Sorption of Tantalum Pentafluoride

Three tests were conducted to determine the effectiveness of both 250°F magnesium fluoride and 650°F sodium fluoride for sorbing tantalum pentafluoride. The test results are summarized in table X. In the first experiment, the sorption system was operated for 19.9 hours with a uranium hexafluoride feed rate of 21.6 lb/hr, a fluorine rate of 1.5 lb/hr., and a nitrogen flow of 0.23 scfm. The sorbent trap was filled with 10.44 pounds of sodium fluoride pellets and was maintained at 650°F. A total of 430 pounds of uranium hexafluoride, containing about 0.0111 pound of tantalum, was passed through the trap, and the average inlet tantalum concentration was 39 ppm. Analyses of samples taken from the gas stream leaving the sorber, table XI, showed the tantalum concentration at that

TABLE X

SELECTIVE SORPTION OF TANTALUM PENTAFLUORIDE
FROM URANIUM HEXAFLUORIDE

	Run Number		
	I-23	I-28	I-30
Reference	*	*	*
Run Duration, hr	19.9	47.4	75
Sorbent	NaF	MgF ₂	MgF ₂
Sorbent Temperature, °F	650	250	250
Uranium Hexafluoride Rate, lb/hr	21.6	21.7	18.6
Average Inlet Tantalum Concen- tration, ppm	39	31	541
Average Outlet Tantalum Concen- tration Before Breakpoint, ppm	0.5	< 1	1.0
Time Until Breakpoint, hr	not reached	not reached	not reached
Average Outlet Tantalum Concen- tration After Breakpoint, ppm	-	-	-
Average Impurity Loading on Bed, weight percent metal	0.15	0.40	5.0
Uranium Holdup on Bed, weight percent of total uranium fed	0.00032	0.00321	0.035

* Present work.

TABLE XI

SORPTION OF TANTALUM PENTAFLUORIDE BY 650°F SODIUM FLUORIDE PELLETS

Run Number	On-Stream Time, hr	Tantalum Concentrations, ppm		Sorbent Analysis		
		Inlet	Outlet	Bed Segment, in.	Tantalum, ppm	Uranium, ppm
I-23	0.5	66	< 0.5	Inlet 6	13,300	330
	2.5	78	< 0.5	Second 6	225	100
	5.5	42	< 0.5	Third 6	22	150
	6.5	59	< 0.5	Fourth 6	25	34
	9.5	35	< 0.5	Fifth 6	11	66
	13.5	10	< 0.5	Sixth 6	420	102
	15.5	18	< 0.5			
	17.5	25	< 0.5			
	19.5	28	< 0.5			

point to be less than 0.5 ppm for the entire length of the run. A total of 0.0109 pound tantalum, or most of that fed, was retained by the trap. Removal of tantalum was excellent during the run, but since breakthrough was not attained, saturation data cannot be reported. Uranium retention was negligible.

The second sorption test with tantalum has already been discussed, with analytical details given in table IV. The test was made using magnesium fluoride as the sorbent and a two-component impurity feed consisting of niobium and tantalum. As noted in the earlier discussion, excellent stripping of the tantalum was achieved; however, again, breakthrough was not achieved, and saturation data were not obtained.

The third tantalum run was an extended test, using 250°F magnesium fluoride as the solid sorbent, planned to define the sorption breakthrough curve and to obtain saturation data. Analytical values are given in table XII. For this test, which lasted 75 hours, a uranium hexafluoride feed rate of 18.6 lb/hr, a fluorine rate of 1.5 lb/hr, and a nitrogen flow of 0.23 scfm were maintained. The sorbent trap was charged with 8.83 pounds of magnesium fluoride pellets. A total of 1,392 pounds of uranium hexafluoride, containing about 0.504 pound of tantalum, was passed through the trap. Analyses of trap inlet samples showed an average tantalum concentration of 541 ppm. The outlet tantalum concentration averaged less than 1 ppm for the length of the run. The sorbent bed held 0.503 pound of tantalum for an average bed concentration of 5.0% tantalum. Both stripping and loading of tantalum appeared excellent, especially considering the fact that breakthrough never occurred. Uranium holdup on the pellet bed was exceptionally high, averaging 3.25% of the final bed weight. Since the magnesium fluoride pellets had been sufficiently dried, excessive moisture was apparently not responsible for the unusual uranium retention. The surface area of the sorbent pellets was found to be 78.5 sq m/g instead of the usual 44 sq m/g, however, and this observation might explain the behavior. It should be noted that, even though higher than in other tests using magnesium fluoride at 250°F, the amount of uranium held on the bed represented only 0.035% of the total amount of uranium passed through the system.

Stripping of tantalum with both sodium fluoride and magnesium fluoride was completely satisfactory. Furthermore, the bed retention with magnesium fluoride was particularly good. While the test results with sodium fluoride did not allow a comparable evaluation of the holding capacity of this material, magnesium fluoride would probably be favored in any event because of its lower operating temperature.

Sorption of Antimony Pentafluoride

Three sorption tests, two using magnesium fluoride and the other using sodium fluoride as the solid sorbents, were completed using antimony pentafluoride as the system impurity. These tests are summarized in table XIII, while more detailed information is presented in tables XIV, XV, and XVI. In the first test, operation continued for 24.6 hours with a uranium hexafluoride feed rate of 23 lb/hr, a fluorine rate of

TABLE XII

SORPTION OF TANTALUM PENTAFLUORIDE BY 250°F MAGNESIUM FLUORIDE PELLETS

Run Number	On-Stream Time, hr	Tantalum Concentrations, ppm		Sorbent Analysis		
		Inlet	Outlet	Bed Segment, in.	Tantalum, %	Uranium, %
I-30	1.0	65	0.7	First 6	8.05	4.51
	6.0	77	< 0.5	Second 6	6.78	4.45
	11.0	80	< 0.5	Third 6	6.19	3.57
	16.0	68	< 0.5	Fourth 6	5.71	4.80
	21.0	67	< 0.5	Fifth 6	1.40	1.02
	26.0	64	< 0.5	Sixth 6	1.27	1.15
	31.0	645	< 0.5			
	36.0	507	0.5			
	41.0	331	< 0.5			
	46.0	569	3			
	51.0	563	< 1			
	61.0	435	< 1			
	71.0	639	< 1			
	75.0	628	< 1			

TABLE XIII

SELECTIVE SORPTION OF ANTIMONY PENTAFLUORIDE
FROM URANIUM HEXAFLUORIDE

	Run Number		
	I-20	I-24	I-31
Reference	*	*	*
Run Duration, hr	24.6	20.6	80.7
Sorbent	MgF ₂	NaF	MgF ₂
Sorbent Temperature, °F	250	650	250
Uranium Hexafluoride Rate, lb/hr	23.0	23.0	22.5
Average Inlet Antimony Concentration, ppm	72	17	17.1
Average Outlet Antimony Concentration Before Breakpoint, ppm	2.0	7.8	< 1
Time Until Breakpoint, hr	not reached	not apparent	40†
Average Outlet Antimony Concentration After Breakpoint, ppm	4.5	-	2
Average Impurity Loading on Bed, weight percent metal	0.49	0.043	0.21
Uranium Holdup on Bed, weight percent of total uranium fed	0.0151	0.0015	0.014

* Present work.

† Although an increase in the trap outlet stream antimony concentration, i.e., a break, was observed after 40 hours, the sorbent continued to perform well, and the average outlet concentration was held to 2 ppm antimony for the rest of the run.

TABLE XIV

SORPTION OF ANTIMONY PENTAFLUORIDE BY 250°F MAGNESIUM FLUORIDE PELLETS

Run Number	On-Stream Time, hr.	Antimony Concentrations, ppm		Sorbent Analysis		
		Inlet	Outlet	Bed Segment, in.	Antimony, %	Uranium, %
I-20	0.5	34	0.9	Inlet 6	1.49	0.60
	2.5	27	1.8	Second 6	1.17	0.29
	4.5	34	0.8	Third 6	0.38	0.17
	6.5	44	1.4	Fourth 6	0.12	1.44
	8.5	207	1.1	Fifth 6	0.04	1.47
	10.5	144	1.1	Sixth 6	0.63	0.30
	12.5	124	1.8			
	14.5	71	1.4			
	16.5	39	1.2			
	18.5	39	1.1			
	20.5	76	6.0			
	22.5	41	1.4			
	24.5	40	6.0			

TABLE XV

SORPTION OF ANTIMONY PENTAFLUORIDE BY 650°F SODIUM FLUORIDE PELLETS

Run Number	On-Stream Time, hr	Antimony Concentrations, ppm		Sorbent Analyses		
		Inlet	Outlet	Bed Segment, in.	Antimony, %	Uranium, ppm
I-24	0.5	67	9	Inlet 6	1.42	810
	2.6	42	16	Second 6	0.39	440
	4.6	18	4	Third 6	0.03	330
	6.6	9	1.6	Fourth 6	115 ppm	260
	8.6	4	11	Fifth 6	111 ppm	500
	10.6	5	18	Sixth 6	108 ppm	-
	12.6	21	5			
	14.6	1.6	5			
	16.6	4	7			
	18.6	15	4			
	20.6	11	-			

TABLE XVI

SORPTION OF ANTIMONY PENTAFLUORIDE BY 250°F MAGNESIUM FLUORIDE PELLETS

Run Number	On-Stream Time, hr	Antimony Concentrations, ppm		Sorbent Analyses		
		Inlet	Outlet	Bed Segment, in.	Antimony, %	Uranium, %
I-31	1.0	39	< 1	Inlet 6	1.04	0.47
	5.0	24	< 1	Second 6	0.87	0.75
	10.0	30	< 1	Third 6	0.92	1.13
	15.0	40	< 1	Fourth 6	0.16	3.00
	20.0	23	< 1	Fifth 6	0.20	4.88
	25.0	15	< 1	Sixth 6	39 ppm.	5.48
	30.0	14	< 1			
	35.0	17	< 1			
	40.0	11	< 1			
	45.0	11	1.5			
	50.0	9	3.0			
	60.0	8	3.0			
	70.0	18	2.0			
	80.0	5	3.0			

1.5 lb/hr, and a nitrogen flow of 0.23 scfm. The sorbent trap was charged with 8.1 pounds of magnesium fluoride held at a temperature of 250°F during the run. A total of 566 pounds of uranium hexafluoride, with approximately 0.027 pound of antimony, was fed through the system, and the average antimony concentration was 72 ppm in the feed stream. As shown in table XIV, analyses of material withdrawn at the sorber outlet showed that excellent stripping of the antimony was achieved, with the antimony concentration in this stream averaging 2.0 ppm for the first 18.5 hours of the run and 4.5 ppm for the remainder of the test. A total of about 0.026 pound of antimony was held on the bed. Uranium holdup was 0.015% of the total amount fed.

Sodium fluoride, at 650°F, was used as the sorbent material in the second experiment. The system was operated for 20.6 hours with a uranium hexafluoride rate of 23 lb/hr, a fluorine rate of 1.5 lb/hr, and a nitrogen flow rate of 0.23 scfm. The trap was loaded with 10.44 pounds of sodium fluoride, and a total of 476 pounds of uranium hexafluoride, containing 0.00547 pound of antimony, was fed. The average inlet antimony concentration was 17 ppm on a metal basis. Analyses of outlet condensables, listed in table XV, showed a relatively high average 7.8 ppm antimony leaving the sorber, with no distinct breakpoint. A total of 0.00298 pound of antimony was held in the trap. Only a negligible quantity of uranium was retained.

The third antimony run was an extended run lasting 80.7 hours. In this test, the system was operated with a uranium hexafluoride feed rate of 22.5 lb/hr, a fluorine rate of 1.5 lb/hr, and a nitrogen flow rate of 0.23 scfm. The sorbent trap was charged with 8.25 pounds of magnesium fluoride pellets and was maintained at 250°F. During the run, a total of 1,820 pounds of uranium hexafluoride, containing 0.0208 pound of antimony, was passed through the trap. Analyses, summarized in table XVI, showed an average 17.1 ppm antimony in the uranium hexafluoride feed. For the first 40.0 hours of operation, the sorber outlet stream averaged less than 1 ppm antimony and even for the remainder of the run, the average outlet value was only 2 ppm, thus confirming excellent stripping of this impurity. The antimony holdup was 0.0180 pound, while the uranium sorption amounted to 0.014% of the total uranium fed through the bed. While the uranium retention was tolerably low, it was considerably higher than usually found with 250°F magnesium fluoride. A possible explanation is that, for this run, as with the last tantalum test, the magnesium fluoride pellets had a higher than usual surface area, 99.5 sq m/g.

Performance of Other Sorbents

Some scoping tests for other effective sorbent materials were conducted concurrently with a few of the regular runs in order to evaluate quantitatively the capacity and selectivity of new materials for the various impurities. Results of these recent tests are presented in table XVII. Separate wire mesh containers holding 41 grams of sodium fluoride-19% lithium fluoride pellets, 22 grams of sodium fluoride-22.8% lithium fluoride pellets, and 16 grams of lithium fluoride pellets were placed

TABLE XVII
ANALYSES OF SORBENT SAMPLES USED IN SCOPING TESTS

Test Sorbent	Temperature, °F	Run Number	Impurity	Impurity Concentration, weight percent on test sorbent	Reference Impurity Concentration, weight percent on magnesium fluoride bed	Uranium Concentration, weight percent on test sorbent
NaF-19% LiF	250	I-29	NbF ₅	0.32	4.7*	0.69
NaF-22.8% LiF	250	I-29	NbF ₅	0.70	4.7*	21.44
LiF	250	I-29	NbF ₅	10.92	4.7*	0.07
CaF ₂	250	I-30	TaF ₅	1.63	4.98(8.05†)	0.09
AlF ₃	250	I-30	TaF ₅	10.03	4.98(8.05†)	1.53
AlF ₃	250	I-31	SbF ₅	1.12	0.32(1.04†)	0.07

* Graphical estimate of average loading.

† Highest values for pellets near the trap inlet.

above the regular 250°F magnesium fluoride bed in test I-29, which employed niobium pentafluoride as the impurity. Analyses of the pellets after completion of the run indicated an excellent capability of lithium fluoride at 250°F to sorb niobium pentafluoride selectively from uranium hexafluoride; the niobium concentration on the sample pellets of lithium fluoride averaged 10.92%, with a very small uranium holdup and no caking. The results for the other two test sorbent materials were not good, and furthermore, the uranium retention values were confusing.

In test I-30, where tantalum pentafluoride was used as the impurity, 54 grams of aluminum fluoride and 114 grams of calcium fluoride were placed above the regular magnesium fluoride bed, which was held at 250°F. Chemical analysis showed tantalum concentrations of 10.03 and 1.63% on the aluminum fluoride and calcium fluoride pellets, respectively. Since the uranium holdup on the aluminum fluoride was satisfactory, this material demonstrated an exceptionally good overall potential as a sorbent material for tantalum. Even the calcium fluoride, although somewhat overshadowed by the performance of the aluminum fluoride sample, performed well enough to warrant further testing.

In test I-31, made using antimony pentafluoride, 86 grams of aluminum fluoride was placed in the trap. After completion of the run, the sample pellets were found to contain 1.12% antimony and a low 0.07% uranium. Considering the magnesium fluoride behavior in the same test, the performance of aluminum fluoride again looked extremely encouraging.

CONCLUSIONS AND RECOMMENDATIONS

In general, encouraging definitive uranium purification results have been obtained in sorption tests using uranium hexafluoride-impurity fluoride gas mixtures. Based on test data, it appears that the combination of magnesium fluoride and sodium fluoride sorbent traps, operated in a series arrangement, should prove capable of meeting cascade return specifications for niobium, tantalum, titanium, antimony, and ruthenium. In a plant purification system with backup traps installed in series with the primary traps to allow efficient sorbent utilization, average bed loadings for these impurities of 5% or higher could probably be achieved without difficulty. Furthermore, the magnesium fluoride and sodium fluoride sorbents are readily available and are easy to prepare.

There are, however, possible limitations which have come to light on the basis of present test information. First, because it was difficult to collect meaningful titanium data with inlet concentrations greater than about 10 ppm, conclusions regarding magnesium fluoride performance for titanium at higher concentrations are not yet possible. Also, a trapping method has been demonstrated only for the ruthenium form, presumed to be the pentafluoride, produced by direct fluorination of the metal. No sorption work has yet been done, however, starting with oxides, where fluorination might yield significant quantities of other ruthenium compounds, such as oxyfluorides. Within the bounds of these restrictions,

it appears that present AEC specifications can be met for the five impurities studied.

Flow testing of magnesium fluoride, sodium fluoride, and other sorbent materials to collect impurity saturation values and other design information is continuing. Already, aluminum fluoride has demonstrated excellent capacities for some of the impurities of interest, and this material will be evaluated further in more extensive tests. Special emphasis is also being placed on the element ruthenium because experience with some fuel reprocessing schemes has shown that removal of this element from uranium product streams is especially difficult. As the need arises, attention will be given to other impurities, such as bromine, for which current specifications might prove difficult to meet in view of new uranium processing and recovery schemes now being considered*.

Another significant facet of the ORGDP impurities program concerns the theoretical aspects of the sorption process. It is hoped that a useful model of the removal mechanism can be developed so that the experimental results can be correlated and extrapolated to assist in confident selection of near-optimum sorption parameters for plant systems. To aid in this area, some small-scale tests elucidating the effects of sorbent pretreatment conditions on average pore sizes and other physical properties are scheduled.

Finally, the importance of using large quantities of as-received off-specification materials in future sorbent tests should be emphasized. Such work is considered to be highly desirable, since it would eliminate the need for guessing at impurity chemical forms and concentrations. Additionally, operational fluorination tests using fluid beds and flame reactors could serve to establish the fates of the various impurities during uranium hexafluoride production operations and to guide the sizing of a removal system. In the earlier ORGDP work with spiked oxide, several questions were raised; the conditions for impurity volatilization from the oxide, the significance of impurity retention by bed materials, the severity of holdup on filters and in other equipment items, the extent of deposition on piping surfaces, and even the approach to equilibrium transport conditions in the tests were not established. By conducting sorption tests with actual returns, it is anticipated that many of these questions could be answered satisfactorily so that the whole problem of uranium hexafluoride impurity removal could be more sharply defined.

* In addition to established aqueous methods for processing irradiated nuclear fuel, other reprocessing techniques involving fluorination with both elemental fluorine and such interhalogens as bromine pentafluoride and chlorine trifluoride are now being considered in large-scale development programs in the United States and abroad.

ACKNOWLEDGEMENT

The authors would like to express their gratitude to several persons at the Oak Ridge Gaseous Diffusion Plant for their considerable help. Other members of the Engineering Development Department (to which the authors belong) assisted substantially. S. H. Smiley, the Department Superintendent, and J. H. Pashley, the Department's Chief Engineer, provided much of the overall project direction and made significant contributions to the technical aspects of the program.

It is also a pleasure to acknowledge the efforts of R. B. Farrar, J. T. Mottern, and R. J. Wertz, of the Works Laboratory. Their analyses of process stream and sorbent bed samples provided essential test data.

APPENDIX

PRESENT SPECIFICATIONS FOR URANIUM HEXAFLUORIDE
DELIVERED TO THE U. S. ATOMIC ENERGY COMMISSION

The latest (effective July 1, 1967) AEC specifications for uranium hexafluoride are listed in table XVIII. This table appeared as table IV on page 16584 of the Federal Register, Vol. 31, No. 250, Wednesday, December 28, 1966.

TABLE XVIII
SPECIFICATIONS FOR URANIUM HEXAFLUORIDE
DELIVERED TO THE AEC

Item*	Numerical Value
Maximum vapor pressure of filled container at 200°F in pounds per square inch, absolute	75
Minimum weight percent of uranium hexafluoride in material	99.5
Maximum mole percent of hydrocarbons, chlorocarbons, and partially substituted halohydrocarbons	0.01
Maximum number of parts of elements indicated per million parts of total uranium:	
Antimony	1
Bromine	5
Chlorine	100
Niobium	1
Phosphorus	50
Ruthenium	1
Silicon	100
Tantalum	1
Titanium	1
Total of elements forming nonvolatile fluorides (having a vapor pressure of 1 atmosphere or less at 300°C), e.g., aluminum, barium, bismuth, cadmium, calcium, chromium, copper, iron, lead, lithium, magnesium, manganese, nickel, potassium, silver, sodium, strontium, thorium tin, zinc, and zirconium	300
Maximum number of parts of elements or isotopes indicated per million parts of uranium-235:	
Chromium	1,500
Molybdenum	200
Tungsten	200
Vanadium	200
Uranium-233	500
Uranium-232	0.110
Maximum thermal neutron absorption of total impurity elements as equivalent parts of boron per million parts of total uranium	8
Maximum total of gamma activity due to fission products and uranium-237 as percent of gamma activity of aged natural uranium and as measured in a high-pressure ionization chamber (Drawing D-AWM-8796 of Nuclear Division, Union Carbide Corporation)	20

TABLE XVIII
(Continued)SPECIFICATIONS FOR URANIUM HEXAFLUORIDE
DELIVERED TO THE AEC

Item*	Numerical Value
Maximum beta activity due to fission products as percent of beta activity of aged natural uranium	10
Maximum number of parts of plutonium per billion parts of total uranium	1
Maximum alpha activity from all transuranic elements in disintegrations per minute per gram of total uranium . . .	1,500
* All specification analyses on uranium hexafluoride shall be performed on samples removed in the liquid state from each cylinder while its contents are liquid and homogeneous.	