1977

A heated uranium tetrafluoride target system to release non-rare gas fission products for the TRISTAN isotope separator

Ronald Lee Gill

Iowa State University

Recommended Citation

Gill, Ronald Lee, "A heated uranium tetrafluoride target system to release non-rare gas fission products for the TRISTAN isotope separator " (1977). *Retrospective Theses and Dissertations*. 7558.
https://lib.dr.iastate.edu/rtd/7558

INFORMATION TO USERS

This material was produced from a microfilm copy of the original document. While the most advanced technological means to photograph and reproduce this document have been used, the quality is heavily dependent upon the quality of the original submitted.

The following explanation of techniques is provided to help you understand markings or patterns which may appear on this reproduction.

1. The sign or "target" for pages apparently lacking from the document photographed is "Missing Page(s)". If it was possible to obtain the missing page(s) or section, they are spliced into the film along with adjacent pages. This may have necessitated cutting thru an image and duplicating adjacent pages to insure you complete continuity.

2. When an image on the film is obliterated with a large round black mark, it is an indication that the photographer suspected that the copy may have moved during exposure and thus cause a blurred image. You will find a good image of the page in the adjacent frame.

3. When a map, drawing or chart, etc., was part of the material being photographed the photographer followed a definite method in "sectioning" the material. It is customary to begin photoing at the upper left hand corner of a large sheet and to continue photoing from left to right in equal sections with a small overlap. If necessary, sectioning is continued again — beginning below the first row and continuing on until complete.

4. The majority of users indicate that the textual content is of greatest value, however, a somewhat higher quality reproduction could be made from "photographs" if essential to the understanding of the dissertation. Silver prints of "photographs" may be ordered at additional charge by writing the Order Department, giving the catalog number, title, author and specific pages you wish reproduced.

5. PLEASE NOTE: Some pages may have indistinct print. Filmed as received.

77-25,987

GILL, Ronald Lee, 1949-
A HEATED URANIUM TETRAFLUORIDE TARGET SYSTEM
TO RELEASE NON-RARE GAS FISSION PRODUCTS
FOR THE TRISTAN ISOTOPE SEPARATOR.

Iowa State University, Ph.D., 1977
Chemistry, nuclear

A heated uranium tetrafluoride target system

to release non-rare gas fission products

for the TRISTAN isotope separator

by

Ronald Lee Gill

A Dissertation Submitted to the

Graduate Faculty in Partial Fulfillment of

The Requirements for the Degree of

DOCTOR OF PHILOSOPHY

Department: Chemistry
Major: Physical Chemistry

Approved:

Signature was redacted for privacy.

In Charge of Major Work

Signature was redacted for privacy.

For the Major Department

Signature was redacted for privacy.

For the Graduate College

Iowa State University
Ames, Iowa

1977

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

I. INTRODUCTION

A. The TRISTAN Isotope Separator

The TRISTAN on-line isotope separator facility, located at the Ames Laboratory Research Reactor, began producing mass separated short-lived gaseous fission products in October 1966. It was the first such facility to become operational at a reactor. Since that time, the TRISTAN group has been involved in a systematic and detailed study of the nuclear properties of gaseous fission products as well as the development of the associated equipment and techniques necessary to support such a program. The primary thrust of the program is to construct nuclear decay schemes of mass separated neutron-rich nuclides produced by thermal neutron fission of ^{235}U. While gamma-ray spectroscopy is the principal tool used in these studies, the detector array also provides the capability of studing half-lives, beta-ray spectra, delayed neutron emission, internal conversion electron spectra and gamma-gamma angular correlations in an on-line configuration.

Since the facility went into operation, the decay schemes of about 38 nuclides have been determined for fission products in the mass ranges of 85 to 93 and 136 to 143. More than fifty publications have come out this work, with about eleven papers giving facility descriptions.

Even though the TRISTAN facility is adequately described elsewhere [1], a general description of the isotope separator

should be given here since this work concerns itself with the
development of a new target system and an interested reader
may need a convenient reference. The system layout prior to
July 1976 is illustrated in Figure 1.

The target is located at Face 6 of the Ames Lab Research
Reactor and utilizes the external neutron beam from that beam
tube. The thermal neutron flux at this port has been meas-
ured to be about $3x10^9$ neutrons/cm^2/sec. This represents
about 65% of the total neutron flux, the remainder being epi-
thermal neutrons. The target is an aluminum container with
provision for an outlet to the transfer line and an inlet for
xenon and krypton doped helium support gas that helps to
transport the fission products as well as sustain the plasma
in the ion source. Inside the target container there are
aluminum trays to contain the ^{235}U target material, original-
ly uranyl stearate. More than 10 grams of highly enriched
uranyl stearate may be put into a single target load. Fis-
sion products which can diffuse out of the target are trans-
ported to the ion source via a 2-m teflon lined neoprene tube
by the mechanism of molecular flow with some assistance from
the sweeping action of the support gas. Thus only gaseous
fission products can arrive at the ion source. This limits
the activities available for study to xenon, krypton and
their daughters, although a fresh uranyl stearate target will
emanate some bromine and iodine for a period of a few weeks,
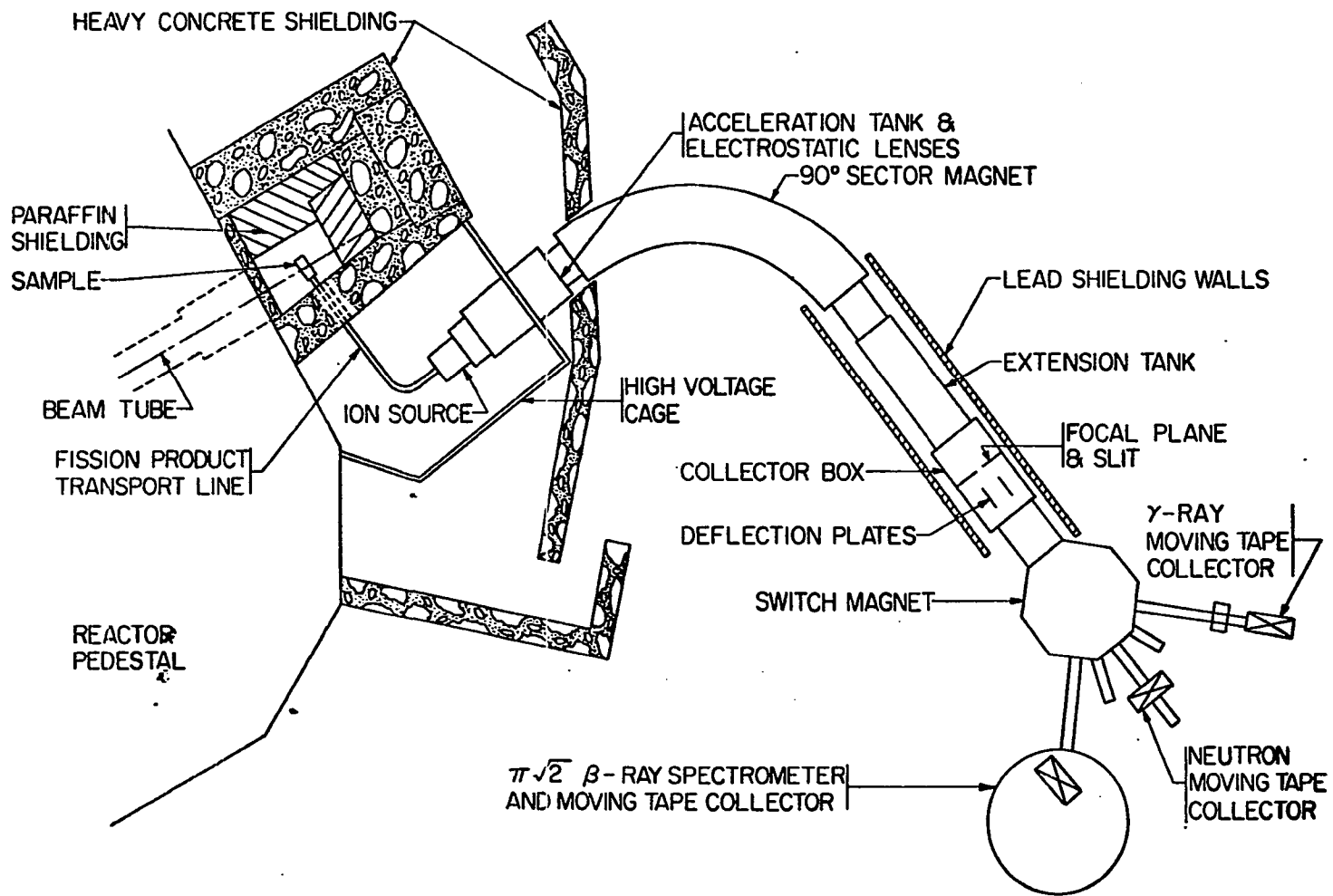
Figure 1. Layout of the TRISTAN isotope separator system.

at best.

The ion source operates in the oscillating electron mode
[2]. The discharge chamber consists of a boron nitride (BN)
inlet plate and BN outlet insulators, a tantalum anode (typi-
cally operated at 40 volts), a tungsten filament (typically
operated at 40 amps), a molybdenum end cap with a 1.5-mm
diameter exit hole, a stainless steel end plate and a
tantalum heat shield. The support gas mentioned above is
necessary since the fission product emanations alone would be
insufficient to sustain a stable discharge. The xenon and
krypton provide mass markers which aid in determining when
the ion beam is properly focused. The ion source operates
best in the 1 to 3 utorr region, but can be operated above 15
utorr, as measured at the differential pump located below the
ion source.

The positive ions produced in the ion source are ex-
tracted through the exit hole by a conical electrode operated
at about 5 kV. The ions are then further accelerated by an
acceleration drift tube operated at 55 kV. Ion beam focusing
is accomplished by means of a focus lens with a spherical
electrostatic field (typically operated at 20 kV) and another
lens with a cylindrical field (typically operated at 10 kV)
to control the height of the beam. The vertical position of
the ion beam is controlled by a pair of deflection plates lo-
cated immediately before the separating magnet, with variable

potential from -300 to +300 V.

The separating magnet is a 90° sector magnet with a mean radius of 160-cm and a mass range of ±7% of the central mass at the focal plane.

The collector box, positioned after the separating magnet, selects the particular mass to be studied by means of a 4-mm slit in an aluminum plate in the focal plane of the magnet. A fluorescent screen may be lowered over the slit to observe the position and shape of the stable xenon and krypton mass markers. The shape of non-visible beams can be determined by use of a beam scanner. The beam profile is displayed on an oscilloscope and beams of less than 300 pA current may be accurately focused. Since many experiments must be performed over a long period of time, the beam position may tend to drift. A beam position stabilizer inside the collector box senses any drift by detecting an imbalance in beam current on two adjacent pins, separated by about 3-mm and applies a correction voltage to the acceleration voltage of the separator, thus shifting the beam position until the current on the pins balance.

After the desired mass is selected in the collector box, the beam enters the switch magnet where it is routed to one of four experimental areas. The magnet has five outlet ports at: 0°, ±22.5° and ±45°. One of the 22.5° ports is not currently in use.

One 45° port leads to a moving tape collector (MTC) where all the gamma-ray spectroscopy experiments are performed. The MTC contains about 2000-m of aluminized Mylar tape with a 0.008-mm Al layer and a 0.025-mm Mylar layer. The tape may be moved forward or backwards, at various speeds, either continuously or in discrete steps. Two stations along the tape are available for gamma-ray analysis of an activity. One is located upstream, at the point of beam deposit, and is generally used to obtain equilibrium spectra and enhanced parent spectra. The other is located downstream and, after a suitable delay, is used to obtain enhanced daughter spectra. Both stations can accommodate two Ge(Li) detectors (180° apart) for gamma-gamma coincidence experiments, and both may be fitted with 0.025-mm Mylar windows for low-energy spectra obtained with LEPS detectors. A windowless Si(Li) detector for measuring conversion electron spectra can also be positioned at either station. This detector may be moved into the MTC vacuum in close proximity to the tape itself.

A third detector station houses a plastic scintillator and is used for beta-ray spectrum measurements. This detector is constructed so that the tape passes through the scintillating material, providing a nearly 4-pi geometry.

A seven-detector system for gamma-gamma angular correlations may also be added to the MTC when such experiments are

desired. The system consists of one Ge(Li) detector and six NaI(Tl) detectors. A complete description of this detector system is given by Basinger et al. [3].

A sample deposit box is located on one of the 22.5° ports of the switch magnet. This box allows a collected mass to be removed from the isotope separator system for off-line analysis. This is particularly useful when a chemical separation is necessary to isolate a long-lived member of a decay chain and for preparing longer-lived gamma-ray standards for off-line use.

The switch magnet port at 0° has been used for delayed neutron emission studies. A ^{3}He detector and a small moving tape collector are available for experimentation at this port.

The second 45° switch magnet port is connected to a double-focusing beta-ray spectrometer. The resolution of the spectrometer is sufficient to yield accurate beta-ray spectra of short-lived nuclides, and to resolve the large number of conversion electron peaks exhibited in the decay of odd-A or odd-odd nuclei. A complete description of the spectrometer is given by Halbig et al. [4].

The overall quality of the separated beam delivered to any of the experimental stations is excellent. The only significant contamination found in a well-focused beam is due to formation of hydride ions. Formation of a hydride ion causes

interference from the decay of activities one mass unit lower than the mass desired. Typically this contamination is on the order of a few percent, but may be as high as ten percent. In one instance, for mass 94, the contamination from mass 93-hydride was estimated to be 85% of the total activity. However, this is a rather extreme case, since the fission yield of ^{94}Kr is only about 20% of that for ^{93}Kr, and transport losses are much larger since the half-life of ^{94}Kr is only about 16% of that for ^{93}Kr.

Possibly the major short-coming of the TRISTAN facility as originally used was, as mentioned earlier, the inability of the uranyl stearate target to release fission products other than xenon and krypton.

B. Statement of the Problem

Recently, the lack of non-rare gas fission products has become a critical factor in the future of the TRISTAN facility. From a scientific point of view, there is a real need to fill in the gaps in nuclear systematics, particularly on the neutron-rich side of stability if the hope of a consistent nuclear structure theory is to be realized. There are many nuclides that have not been studied, but presumably could lend themselves to detailed study if they could be transported from the target into the ion source. From a practical

standpoint, if the TRISTAN project is to continue, some alternatives to the rare gases must be found, since nearly all the rare gas study objectives have been achieved. Finally, the political atmosphere needs to be considered. Funding prospects for basic research in general, and the TRISTAN project in particular, are not what most scientists would consider adequate. It has become necessary to demonstrate an ability to do "frontier research" to be assured of favorable funding status. It seems reasonable, therefore, to attempt to develop a target system that can make the study of non-rare gas fission products the "frontier" for the TRISTAN facility to explore.

The development of non-rare gas capabilities proceeded along three paths: a He-jet transport system coupled to an ion source; an integrated target-ion source system that would place the ion source directly in the neutron beam; and the use of alternative target materials at higher temperatures where non-rare gas fission products may be volatile. A description of the progress of each of these approaches is given by Talbert et al. [5]. The He-jet was considered to be a long range project, but worth pursuing. The in-beam ion source was considered the most likely approach to succeed since it would emulate the already proven system at the Studsvik, Sweden on-line facility [6]. The heated target system was considered the quickest approach to test since it

would require minimum alteration to the existing isotope separator system. It also carried the hope of being able to control, to some extent, the chemical species volatilized by controlling the temperature. The heated target system is the topic of this thesis.

Uranium tetrafluoride was chosen as the target material. It has good thermal stability with only two significant decomposition reactions:

$$3UF_4 + O_2 \rightarrow UO_2 + 2UF_6 \qquad (1)$$

and

$$UF_4 + 2H_2O \rightarrow UO_2 + 4HF. \qquad (2)$$
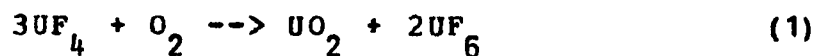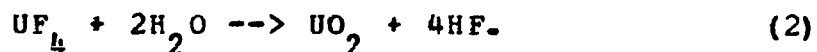
Reaction (1) should not occur below about 750°C and since the target will be under a good vacuum, there should be very little oxygen present to cause the reaction to proceed. Reaction (2) becomes spontaneous at about 250°C. While the vacuum will keep water vapor from the atmosphere from entering the target, extreme care must be taken to avoid the presence of water in the uranium tetrafluoride itself.

A more fundamental reason for using uranium tetrafluoride may be found by noting the low boiling points of many fluorides that could be formed in the target. As the fission fragments thermalize in the target matrix, they should pick up fluorine atoms and take on the chemical properties of

their respective fluoride molecules at the target temperature and pressure. Table 1 lists the boiling points of halides of some elements produced in fission. A study of this table indicates that the fluorides are generally more volatile than the corresponding chlorides, bromides or iodides. Furthermore, based on a consideration of boiling points alone, As, Se, Br, Kr, Zr, Nb, Mo, Tc, Ru, Rh, Sn, Sb, Te, I and Xe could be expected to emanate from a uranium tetrafluoride target heated to about 800°C.

A series of articles by Weber et al. [8, 9, 10] describing the volatilization of various fission products from a uranium tetrafluoride matrix were found. They reported volatilizing Se, Kr, Zr, Nb, Mo, Tc, Ru, Sn, Sb, Te, I and Xe, in good agreement with what may be expected based on boiling point considerations. At temperatures of 800°C they reported yields of 70% to 95% with half-times between 5 and 15 seconds. Although the high yields were promising, the long half-times (time required for half of the total yield to be volatilized) would seem, at first glance, to eliminate the possibility of using this procedure to study activities with half-lives less than a few seconds. However, this was not expected to be a problem for two reasons. First, the target on the isotope separator is a dynamic system, that is fission products are continually being formed and volatilized, both from the bulk material and from surfaces, whereas in Weber's

Table 1. Boiling points of halides of elements produced in thermal neutron fission of ^{235}U.

| Element | Halide boiling point in °C[1] | | | |
	F	Cl	Br	I
As III	63	130	221	403
As V	-53			
Se IV	>100	288		
Se VI	-35			
Br I	-20	5	59	
Br III	135			
Br V	41			
Kr				
Rb I	1410	1390	1340	1300
Sr II	2490	1250	d[2]	d
Y III	>1387	1507		700
Zr IV	600s[3]	331s	357s	600d
Nb V	236	254	362	
Mo VI	35			
Tc VI	55			
Ru V	250			
Rh III	600s	800s		
Ag I	1159	1550	>1300d	1506
Cd II	1758	960	863	796
In III	>1200	300s	>436s	>210
Sn IV	705s	114	202	364
Sb III	319s	283	280	401
Sb V	150	79		401
Te IV	>97	380	>380	421d
Te VI	36			
I I		97	116d	184
I V	98			
I VII	4.5s			
Xe				
Cs I	1251	1290	1300	1280
Ba II	2137	1560	d	
La III		>1000		
Ce III	2300	1727		1397
Pr III	1700	1547		

[1]From reference [7].

[2]d=decomposes.

[3]s=sublimes.

experiments the samples were irradiated and allowed to decay for a day before heating. The separator would make good use of the recoil enhanced fast diffusion of fission products near the surface of the uranium tetrafluoride matrix, but in off-line experiments, like Weber's, this effect would not be seen. Secondly, an isotope separator does not require enormous yields from the target to provide adequate activities for study. Even if the total diffusion were slow, sufficient activity should be volatilized in a tenth-time (time required for one-tenth of the total yield to be volatilized) to make nuclear spectroscopy studies feasible.

At the onset of the project, the TRISTAN facility seemed to be the only isotope separator considering this type of target system. However, when the first on-line experiment was beginning, a parallel study by Burkard et al. [11] was brought to our attention. Although their results were negative, it seemed that the system developed and the TRISTAN isotope separator configuration was sufficiently different to warrant continued experimentation.

II. OFF-LINE EXPERIMENTS

A. Experimental Procedures

After deciding to use heated uranium tetrafluoride as a target material, it was necessary to perform some off-line experiments to determine the feasibility of such a system. Knowledge of the elements that can be volatilized, the temperatures required to volatilize them and any target design restrictions that might arise as a result of the properties of uranium tetrafluoride must be obtained in order to enhance the possibilities of an on-line success. These parameters were investigated by irradiating unenriched uranium tetrafluoride, sealed in quartz vials, in the Ames Lab Research Reactor for periods of one-half to one hour. The samples, consisting of less than 100 mg of uranium, were irradiated in a pneumatic rabbit system and allowed to decay for about 24 hours before handling. The samples were heated under vacuum, to simulate the conditions present in the isotope separator target, and any volatile elements were collected on a copper disk which was cooled with liquid nitrogen and located about 10-cm above the irradiated uranium tetrafluoride. A drawing of the apparatus used is shown in Figure 2. After the heating period, the copper disk was removed and the activity collected was counted with a Ge(Li) detector to determine what elements were volatilized. In this way the volatility of various fission products could be determined as a function
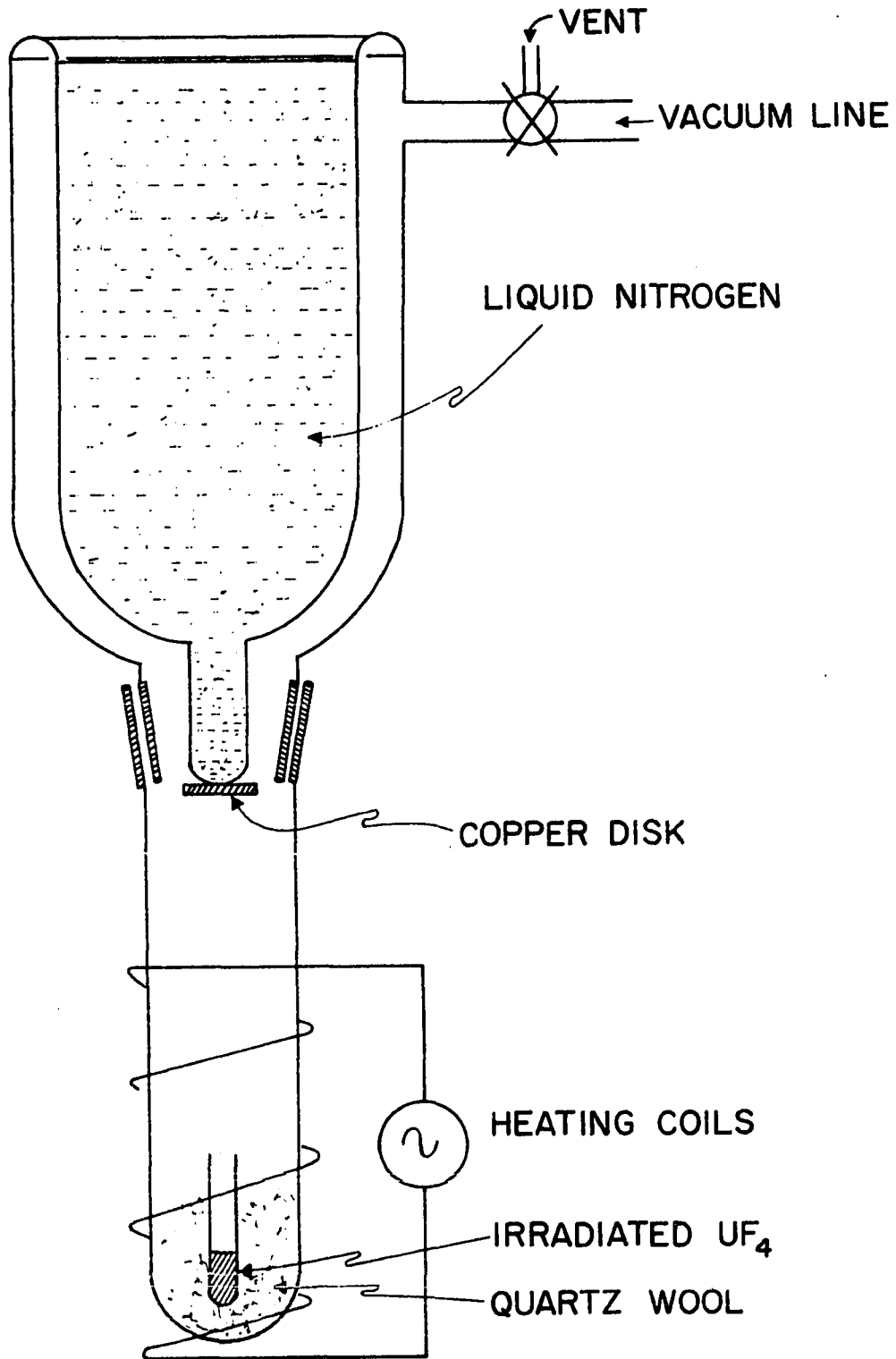
Figure 2. Apparatus for the off-line experiments.

of temperature and heating time. The experiments were designed to be qualitative since no information was available on the overall isotope separator efficiency. Without this information, no minimum required yield could be established for successful use of the target system and thus, on-line tests would be required as the ultimate criterion for success or failure.

B. Results of the Off-line Experiments

The off-line experiments resulted in the volatilization of Zr, Nb, Mo, Tc, Ru, Sb, Te, I and Xe isotopes from the heated uranium tetrafluoride and their collection on the cooled copper disk. Table 2 gives the minimum temperatures at which isotopes of each element could be observed consistently. The minimum detectable activity was about 100 total counts, above background, in a photopeak. Typically the disk was counted for 30 minutes. The results in Table 2 agree well with what was expected on the basis of boiling points listed in Table 1. On the basis of chemical behavior and the correlation with boiling point data, it seems reasonable that As, Se, Br, Kr and Sn can also be volatilized. Fission product isotopes of these elements were not observed since the half-lives are so short that they decayed away before the experiment was performed.

Table 2. Approximate temperature required to volatilize
certain fission products from uranium tetrafluoride.

Element	Temperature °C	Element	Temperature °C
Zr	700	Sb	250
Nb	400	Te	150
Mo	400	I	150
Tc	300	Xe	150
Pu	400		

Although the experiments were not intended to be quantitative, by normalizing yields at each temperature to the yield of a volatile element such as xenon and correcting for the amount of uranium tetrafluoride irradiated, length of decay and counting periods and the length of the irradiation, some indication of the yield versus temperature could be obtained. As expected, the yield had a threshold at some minimum temperature and rose quickly to a maximum yield at some higher temperature. Figure 3 shows the results of this treatment for ^{132}Te and ^{135}Xe. The other elements volatilized showed similar behavior. In some cases the yields were low or the errors large enough that correction of the data gave meaningless results. The temperatures required to volatilize half the total yield are given in Table 3 for each element that yielded acceptable results. All the data used were from samples that were heated for one hour at a constant

Table 3. Temperatures required to volatilize half of the
total yield from irradiated uranium tetrafluoride.

Element	Half-Temperature °C	Element	Half-Temperature °C
Mo	540	Te	730
Tc	480	I	400
Sb	420	Xe	260

temperature.

The results of the experiments to determine the elements
that could be volatilized and the temperature dependence of
the yields were essentially in agreement with the work per-
formed by Weber et al. [10]. However, several potential
problems were indicated by these studies.

Even trace quantities of water were sufficient to con-
vert the uranium tetrafluoride to uranium dioxide by means of
reaction (2). In particular, experiments showed that there
was sufficient water in the helium normally used as support
gas to completely convert the uranium tetrafluoride to urani-
um dioxide in a relatively short time at elevated tempera-
tures. Further experimentation showed that if the helium was
first dried by passing it through a suitable drying agent,
such as magnesium perchlorate or molecular sieves, the prob-
lem could be eliminated. The simplest way of doing this
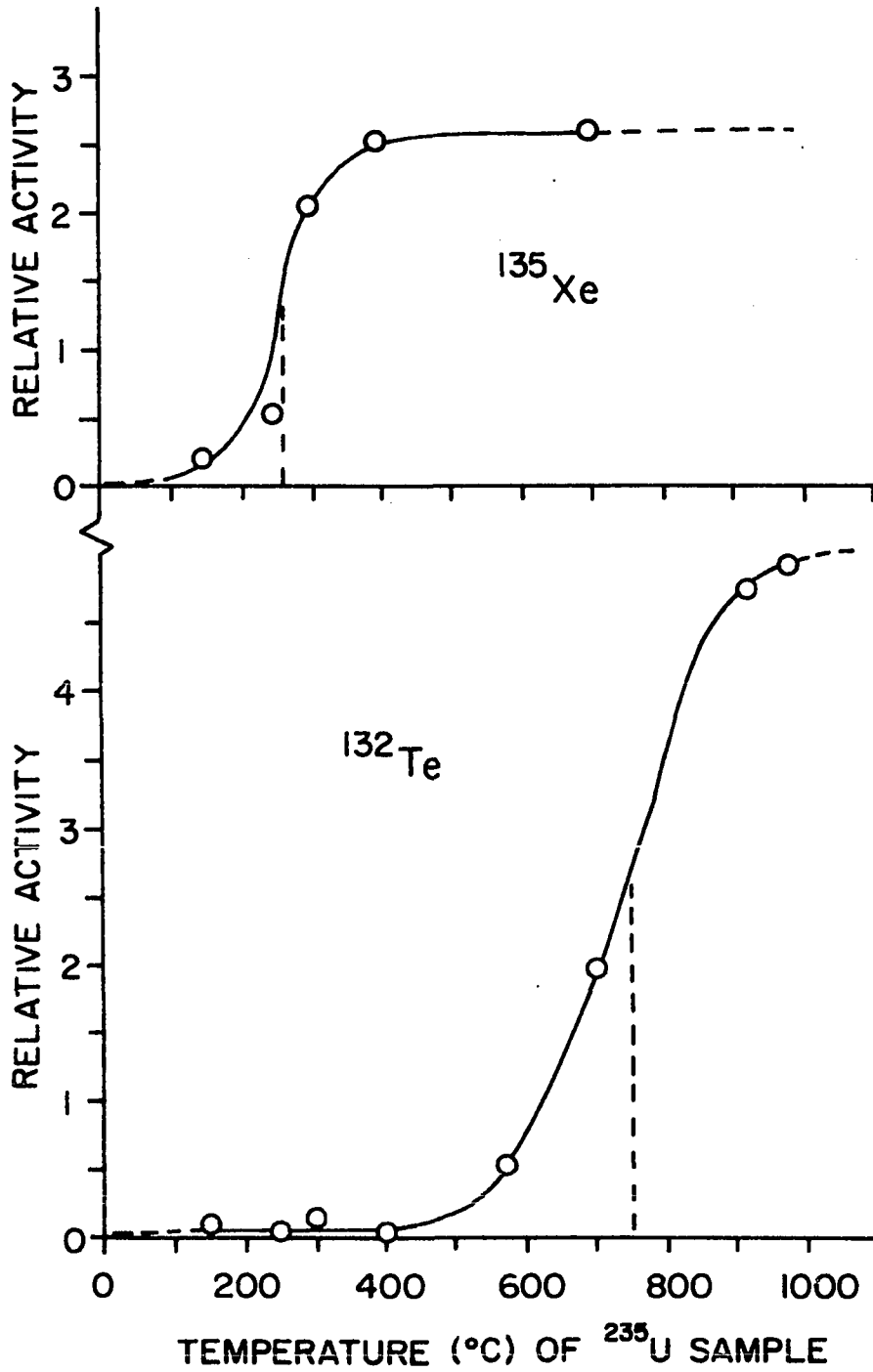would be to place a drying tube in the support gas line of

Figure 3.   Temperature dependence of the yields for
^{132}Te and ^{135}Xe.

the isotope separator so that the helium could be dried before entering the target.

Another problem was recognized while trying to solve the oxidation problem. Uranium tetrafluoride is a very finely divided powder and the addition of even a small amount of support gas is sufficient to blow it off any shelves or trays that are expected to contain it. Even simple evacuation of the target container was observed to cause the loss of nearly all the target material. Thus some means of mounting the uranium tetrafluoride was necessary to prevent it from becoming airborne. The use of graphite cloth and graphite cement[1] provided a solution to the problem. A slurry of uranium tetrafluoride and the graphite cement was made and absorbed onto a section of the cloth. This was slowly dried at about 100°C, to prevent oxidation due to water in the atmosphere, and formed a rigid, porous material that could be impregnated with more than 100 mg UF_4/cm^2 with minimal losses during evacuation or addition of support gas. Irradiation and heating of a section of the impregnated cloth showed that the yields were greatly increased. The gross counting rates were increased by more than a factor of ten, with particular enhancement observed for the more volatile elements such as

_____

[1] Union Carbide Corporation, Carbon Products Division, "UCAR" Cement Grade C-34.

iodine and xenon.

Finally, it was anticipated from the beginning that transport line losses might be significant. To test this, a 2-m section of 1.27-cm outside diameter stainless steel tubing was wrapped with heating tape and insulation. An irradiated piece of impregnated graphite cloth was placed at one end and heated. The volatile fission products were collected at the other end of the tube. After the heating period, the collected fission products were observed to correspond with those expected for heating uranium tetrafluoride to the temperatures involved. While there were, of course, losses along the tube, they were not as large as had been feared and indeed the gross activity was comparable to that expected in other experiments. Thus transport from the target to the ion source was confirmed for a uniformly heated transfer line.

C. Conclusions

Uranium tetrafluoride absorbed onto graphite cloth was found to be a good target material. It was very stable at high temperatures, provided the system was free of both water and oxygen, with minimal losses of target material. A wide variety of non-rare gas fission products could be released with good yield at high temperatures, in the range of 400 to

800°C and the indications were that they could be transported the required distance from the target to the ion source. Therefore, based on the successes of the off-line experiments, the next phase of the project, experimentation in an on-line configuration, could be justified.

III. THE FIRST ON-LINE EXPERIMENT

A. Experimental Procedure

The primary purpose of the first on-line heated target system was to observe non-rare gas fission products. Testing the target design for actual in-use reliability, its ability to achieve the necessary temperatures and to get some idea of the efficiency of this target system relative to the ambient temperature uranyl stearate targets normally used, were secondary purposes.

To obtain a reference point for relative yield considerations, a survey of some of the available masses was made using a large Ge(Li) detector. The survey was first made with a uranyl stearate target and consisted of accumulating a gamma spectrum at each mass where sufficient activity could be produced to warrant data taking. The procedure was then repeated with the heated uranium tetrafluoride target at various temperatures. Mass chains with no rare gas member were given special emphasis in the heated target surveys, since observation of activity at one of those masses would give definitive evidence of non-rare gas production. Iodine and tellurium activities were carefully searched for since they were expected to be among the fission product elements easiest to volatilize. The relative intensities of gamma-rays from the decay of ^{138}Xe were used as an internal standard (since ^{138}Xe is available from the isotope separator

system) for calculating the relative efficiency of the Ge(Li) detector used in the surveys.

B. Apparatus

The target container built for the first on-line experiment is shown in a cut-away view in Figure 4. The target was constructed of stainless steel, with thin walls facing the neutron beam to reduce neutron losses. An outer vacuum jacket, pumped separately from the isotope separator vacuum, served to distribute the heat evenly over the inner can which contained the uranium tetrafluoride. The vacuum jacket pumping line also served as a passageway for a thermocouple to monitor the temperature of the inner can.

A tube through the vacuum jacket and into the inner can allowed the helium support gas to enter the target. The target material was arranged on graphite cloth shelves as shown in Figure 4. In the first on-line experiment, the target was loaded with 4 g of ^{235}U as uranium tetrafluoride.

The back flange of the target was welded on after the uranium tetrafluoride was in place. The target was heated by a heating coil mounted on the brackets provided on the back flange. The target heater could be controlled independently.

A tube at the top of the target allowed volatile fission products to escape the inner can, enter the transfer line and

Vacuum jacket

Stainless steel flange

Connector for transfer line

Graphite shelves for UF$_4$

0.03" ss "window"

Bracket to support furnace

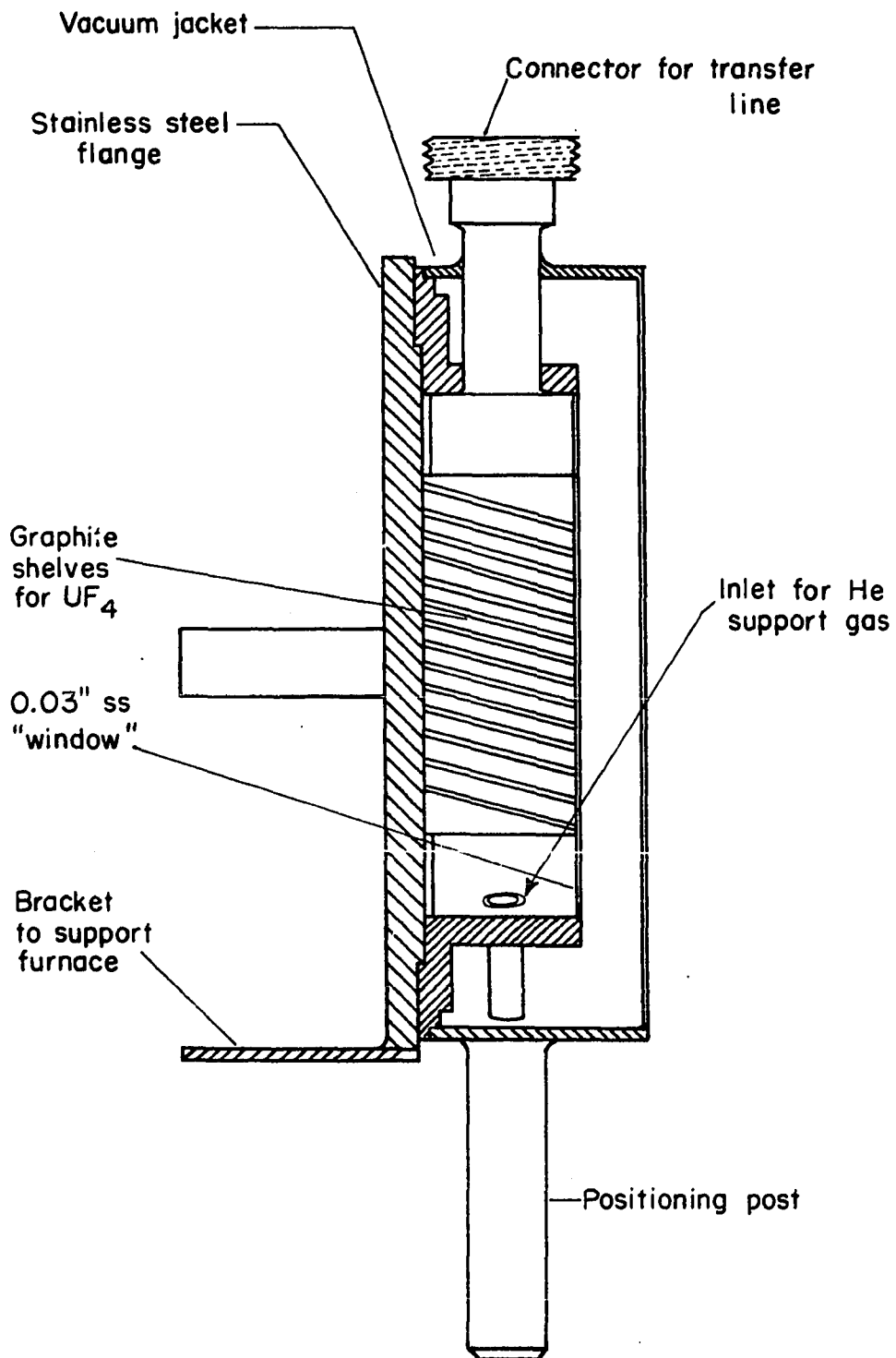Inlet for He support gas

Positioning post

Figure 4. Target container for the first on-line experiment.

migrate to the ion source. The transfer line was a 2-m long stainless steel tube with a 1.27-cm outside diameter and a 0.089-cm wall thickness. The tube was heated by wrapping it with heating tape and layers of asbestos tape for thermal insulation. The heat on each half of the transfer line, the half nearest the ion source and the half nearest the target, could be controlled independently. In this way a temperature gradient could be maintained, if desired.

C. Results of the First On-line Experiment

When the target was first heated, there was an enormous amount of outgassing and operation of the isotope separator became impossible for several days, until the pressure decreased to operable ranges. Throughout the runs, the ion source pressure never really stabilized to a desirable, low pressure. The pressure was always erratic, due to continual outgassing, and this made operation of the isotope separator very difficult. At one point, the acceleration voltage, normally adjusted to 55 kV, could not be raised above 35 kV without the occurrence of arcing. This, along with the erratic ion source operation, made the ion beam focusing and isotope separator stability very poor, requiring frequent retuning and reducing the overall efficiency of the separator system.

Because of the poor operating characteristics, the mass surveys were difficult to do and it was expected that the results would show large efficiency variations due to the variation of operating parameters encountered during the data taking process. In addition to the difficulties mentioned above, the target and transfer line never reached the desired temperature range of 600 to 800°C. The highest temperature reached was 400°C which was high enough to volatilize, in good yield, only the most volatile of the elements desired. As the experiment proceeded, waste heat from the target ignited some Plexiglas used to insulate the acceleration voltage from the ground potential shielding, causing significant damage and curtailing the experiment before any corrective measures could be undertaken.

Throughout the surveys, no non-rare gas activities were observed, other than a barely detectable amount of [136]I observed in the unheated survey. However, gamma-ray spectra from the ambient temperature survey showed that there was no formation of hydrides, as mentioned earlier. The spectra from the heated surveys did show significant hydride formation. This was probably related to the outgassing from the target. Residual solvent, used in preparing the uranium tetrafluoride slurry, could have provided the hydrogen atoms for hydride formation to occur.

Table 4. Gaseous activity yields for uranium tetrafluoride
target relative to ^{138}Xe, 258.3-keV transition
unsaturated yield of 1000 at ambient temperature.

Mass	Temperature °C	Relative yield at saturation	% hydride at saturation
90	ambient	1080 ± 50	-
91	"	830 ± 50	-
89	400	340 ± 10	45 ± 4
90	"	530 ± 30	4.0 ± 0.5
91	"	120 ± 10	3.6 ± 0.4
138	ambient	3120 ± 110	-
139	"	1040 ± 60	-
141	"	34 ± 2	-
137	400	2400 ± 200	-
138	"	1470 ± 50	3.0 ± 0.3
139	"	1020 ± 60	0.24 ± 0.04
140	"	330 ± 10	8.2 ± 0.9
141	"	35 ± 2	12.6 ± 1.6
^{136}I	ambient	0.32 ± 0.03	-

The results of the surveys at ambient temperature and
for the heated target are given in Table 4. The relative
yields are given as calculated at saturation activity for
each mass to remove the half-life dependence of the results.
The krypton masses, 89 to 91 in Table 4, show a decrease in
yield for the heated target, whereas the xenon masses, 137 to
141 in Table 4, show little change, except for mass 138,
which shows a decrease. The data have not been corrected for
fission yield. Doing so does not show any discernible trend,
which may be attributed to the poor operating characteristics

of this system. The ^{136}I observed at ambient temperature was not observed at elevated temperatures.

Table 5 compares the relative yields for the uranium tetrafluoride target with those for the uranyl stearate target. The ratio given in the final column gives the number of grams of ^{235}U, as uranium tetrafluoride, required to give a yield equivalent to one gram of ^{235}U, as uranyl stearate. The percent hydride contamination is given to illustrate the improvement obtained by the ambient temperature uranium tetrafluoride over uranyl stearate.

D. Conclusions

The most serious problem to be corrected was the poor thermal efficiency of the target. Because of the large heat loss from the target, the temperature did not reach the desired range and fission products would not be volatilized from the target. The heat loss also caused the fire. Thus, it was necessary to redesign the target to reduce heat losses so that higher temperatures could be attained and dangerous waste heat could be avoided.

It was also deemed wise to avoid the use of Plexiglas as insulating material, thus eliminating the fire hazard inherent in this material. Pyrex or quartz glass can be used as effective high voltage insulation, even though it may be

Table 5. Comparison of uranium tetrafluoride activity at saturation with uranyl stearate for gaseous fission products, relative to ^{138}Xe, 258.3-keV transition unsaturated yield of 1000 for uranium tetrafluoride.

	Uranium tetrafluoride 3 g ^{235}U		Uranyl stearate 10 g ^{235}U		
Mass	Relative yield at saturation	% hydride at saturation	Relative yield at saturation	% hydride at saturation	Ratio of USt to UF$_4$
137[1]	2400 ± 200	none	15800 ± 1400	none	2.0
138	3120 ± 110	none	81700 ± 2500	0.15 ± 0.06	7.9
139	1040 ± 60	none	155000 ± 8000	0.71 ± 0.12	45.
140[1]	330 ± 10	8.2 ± 0.9	8200 ± 300	5.7 ± 1.7	7.5
141	34 ± 2	none	200 ± 10	22 ± 4	1.8
88	No UF$_4$ data for ^{88}Kr		9500 ± 600	none	-
89[1]	340 ± 10	45 ± 4	21800 ± 700	33.3 ± 2.3	19.
90	1081 ± 50	none	14000 ± 600	10.9 ± 0.8	3.9
91	830 ± 50	none	3050 ± 140	4.1 ± 0.4	1.1

[1]Uranium tetrafluoride data indicated were taken at 400°C.

expensive and fragile.

Another serious problem may have been transfer line losses. The temperature of the transfer line never reached the desired level. To correct this, higher wattage heating tape should be used and more layers of asbestos, as insulation, should be wrapped around the transfer line. The amount of asbestos insulation used in the first on-line experiment was limited by the diameter of the hole through the shielding wall. This hole should be enlarged for future experiments so

that more asbestos insulation can be wrapped over the trans-
fer line. The heating tapes will have to be more carefully
wrapped over the transfer line to eliminate some cool spots,
such as places where the line was bent, which were noticed
during the experiment.

Finally, since the uranium tetrafluoride target produced
no hydride contamination, it was decided to use ambient tem-
perature uranium tetrafluoride targets, in place of uranyl
stearate, as the normal target material. The rare gas yields
were high enough for continued rare gas studies when targets
containing 25 g of ^{235}U loaded onto 25 graphite cloth disks
were used. With these targets, very little hydride contamin-
ation was observed, there was significant yield of even low
fission yield xenon and krypton masses, reasonable yields of
iodine and some bromine could be obtained.

IV. THE SECOND ON-LINE EXPERIMENT

A. Experimental Procedure

The purpose of this experiment was to produce usable quantities of non-rare gas fission products and to demonstrate that the difficulties experienced in the first on-line experiment had been overcome. The reliability of the new system was under test more than its efficiency. Uranium tetrafluoride had already been shown to be a suitable target material during the period between the first and second on-line experiments.

The mass surveys proceeded much like those in the first experiment, except that the 4-pi plastic scintillator, located inside the MTC vacuum, was used to determine the yields, and gamma-ray spectra were taken to make positive identification of the nuclides present. Again, special emphasis was placed on surveying mass chains with no rare gas member to give dramatic evidence of non-rare gas production. The available masses were surveyed before heating, when the target was at temperature and again after it had cooled off, to indicate any degradation of the target material.

B. Apparatus

The Plexiglas insulation was replaced with a 30-cm diameter Pyrex cylinder and the enlarged hole through the

shielding was fitted with a 15-cm diameter quartz tube. There was a hole in the Pyrex cylinder to allow neutrons access to the target, since Pyrex contains a substantial quantity of boron. To reduce further the fire hazard, a large blower was used to exhaust warm air from the shielding cavity.

The new target is shown in Figure 5. The bend in the transfer line was made a part of the target for ease of assembly. The pumps for the vacuum jacket were placed on the other side of the shielding wall, since space inside the shielding is at a premium. The transfer line, vacuum jacket pumping line, water cooling, electrical power and thermocouples all had to come through the shielding wall via the quartz tube.

In this target, the entire target chamber was surrounded by vacuum to insulate it thermally from the outer can. The target chamber was welded shut after 15 g of highly enriched uranium tetrafluoride was loaded into it. The can was heated with a 0.46-mm diameter tungsten wire which was electrically insulated by wrapping it around ceramic posts on the target container's flange. Tantalum foil, 0.13-mm thick, served as a heat reflector.

The flange on the outer can was sealed with a water cooled O-ring. To reduce heat transfer from the inner chamber to the outer can, all the contacts between them were
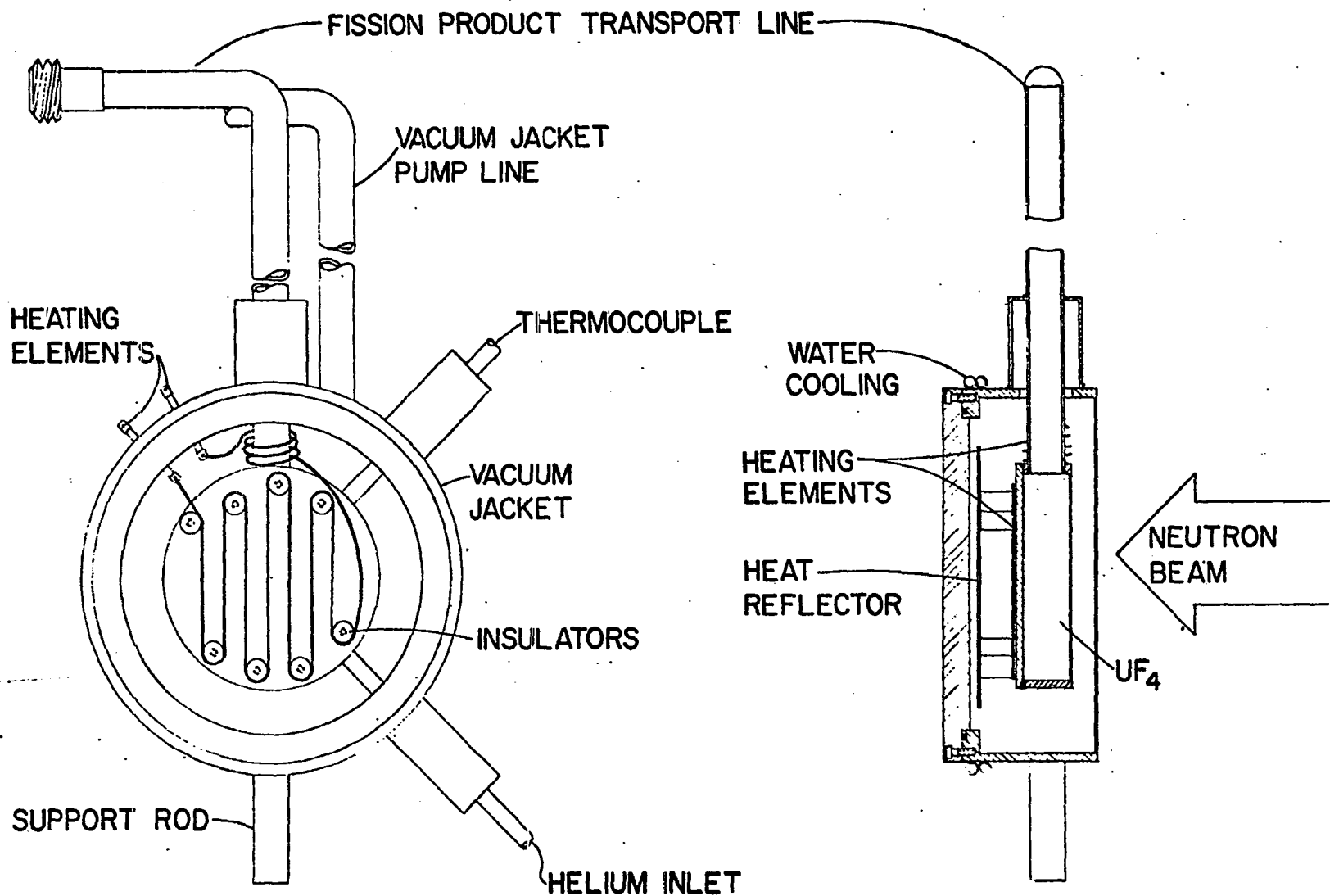
Figure 5. Target container for the second on-line experiment.

made via thin stainless steel protrusions which established a thermal gradient.

The target was tested extensively off-line before being loaded with uranium tetrafluoride. It was determined to be leak-free, 800°C. could be reached with relative ease, the tungsten filament had a long life time if the vacuum jacket was pumped effectively, and even at the highest internal temperatures the outer jacket did not become significantly warm. Thus the target seemed to meet all the requirements necessary to be considered worthy of on-line operation.

C. Results of the Second On-line Experiment

After the ambient temperature survey was made, the target heater was turned on and the high outgassing observed in the first experiment was again observed. The pressure came down into the operating range after several days of heating. Each time the temperature was increased, there was some additional outgassing which would cease after a few hours.

Throughout the heated surveys the transfer line was maintained at about 800°C, while the target temperature was varied from 500 to greater than 800°C. During the heated surveys, the isotope separator itself functioned normally with none of the arcing problems experienced in the first ex-

periment.

In the survey made before heating, bromine and iodine
were observed with good yield. However, as Table 6 shows,
the yield decreased as the temperature was increased and did
not return after the heat was removed. The gamma-ray spectra
collected for the bromine and iodine masses were examined
carefully for evidence of selenium and tellurium, but none
was found. No evidence of any other non-rare gas fission
product was observed at any of the masses surveyed.

The yields tended to decrease upon heating, showed a
maximum at higher temperatures, then decreased after the heat
was removed. It should be noted that the survey made at
500°C may not be characteristic of optimum performance since
the ion source pressure was higher than for the other runs
due to incompleted outgassing. Hence, the 500°C survey
yields are expected to be lower than they could have been,
had the outgassing been completed.

Other, less complete, surveys were made with tempera-
tures exceeding 800°C. In these surveys only non-rare gas
activities were searched for, using gamma-ray spectroscopy
for identification. As stated above, none were found. The
possibility of formation of molecular ions, with single or
multiple charges and single or multiple fluorine atoms, was
explored, but no such species were observed for either rare
gas or non-rare gas activities.

Table 6. Yields from surveys with a 15 g $^{235}UF_4$ target, for the second on-line experiment.

| Nuclide | CPS x 10^5 at saturation[1] | | | |
	Before heating	500°C	650°C	After heating
^{86}Br	0.155	0.006	0.009	-
^{87}Br	0.277	0.001	0.016	-
^{88}Br	0.283	0.001	0.022	-
^{87}Kr	114.	11.0	3.78	41.1
^{88}Kr	72.	8.5	177.	44.6
^{89}Kr	50.3	5.54	16.2	40.8
^{90}Kr	299.	4.23	21.2	67.
^{91}Kr	7.1	2.08	51.9	36.0
^{92}Kr	4.13	0.75	9.8	2.58
^{93}Kr	0.499	0.425	1.29	0.188
^{94}Kr	0.009 ± 1.3%	0.018	0.047	-
^{135}I	130.	0.008	0.034 ± 4.3%	0.002
^{136}I	0.296	0.001	0.052	0.003
^{135}Xe	1.35	2.67	0.045 ± 4.3%	3.85
^{137}Xe	132.	29.7	592.	176.
^{138}Xe	157.	73.	470.	117.
^{139}Xe	28.8	20.5	90.	57.4
^{140}Xe	67.	19.7	89.	16.2
^{141}Xe	3.03	3.06	2.75	0.170
^{142}Xe	0.323	0.269	0.480	0.017
^{143}Xe	0.012	0.017	0.018	-
^{144}Xe	0.001 ± 16.6%	0.002 ± 7.5%	-	-

[1]Standard deviations are less than 1%, except where noted.

In all the surveys the hydride contamination was very low. There was no detectable hydride in the ambient temperature surveys both before and after heating. There was some observable hydride in the heated surveys, presumably associated with the outgassing, as it was worse for the lower tem-

perature surveys where the outgassing was incomplete. For
the worst case the hydride content, as determined by the
amount of ^{90}Kr activity in the ^{91}Kr beam, was about 1%. This
is still significantly lower than that observed with the ura-
nyl stearate target, as reported in Table 5.

Since the system failed to produce any non-rare gas fis-
sion products, the question of whether or not any got into
the ion source was explored. Three days after the surveys
were completed, the system was dismantled and samples of ac-
tivity found at various positions in the transfer line and
ion source were counted with a Ge(Li) detector. The result-
ing gamma-ray spectra were analyzed to see if any non-rare
gas fission products could be observed. The long-lived ac-
tivities observed and their positions are indicated in Table
7.

The transport line inside the ion source, referred to in
Table 7, is a stainless steel tube, about 10-cm in length,
which is inside the vacuum housing of the ion source. For
practical reasons, this section could not be heated indepen-
dently and hence was heated only by conduction from the rest
of the transport line and from the ion source filament. This
tube represented a cool spot in the transfer line and it
picked up the greatest variety of fission products. Tellur-
ium and iodine, which were the most volatile non-gaseous fis-
sion products, were found everywhere in the system.

Table 7. Non-gaseous fission product activities found in the isotope separator system after the second on-line experiment.

Nuclide	Transport line near ion source	Transport line into ion source	Transport line near target	Ion source End cap	Anode
^{132}Te	□	□	□	□	□
^{131}I	□	□	□	□	□
^{99}Mo		□			
^{95}Zr		□	□		

[1]Other nuclides have half-lives too short to be observed.

The transport line near the target contained a brown viscous material which resembled a concentrated form of the solvent used in preparing the uranium tetrafluoride and graphite cement slurry. Apparently this was the cause of much of the outgassing experienced.

D. Conclusions

Again, no non-rare gas fission products, other than bromine and iodine, were observed after mass separation. However, examination of the dismantled system showed that non-gaseous fission products were being volatilized from the target and some were getting into the ion source. Their absence after mass separation suggests that an insufficient amount of fission product activity was getting into and

coming out of the ion source, or the ion source was not
efficiently breaking up the fluoride molecules, thus
spreading the yield over a large number of masses represent-
ing many possible molecular ions.

Insufficient yield arriving at the ion source was proba-
bly due to cool spots along the transfer line, particularly,
as indicated in Table 7, in the unheated section inside the
ion source. The solvent found in the transfer line could
also have impeded the transportation of fission products from
the target to the ion source. Insufficient yield getting out
of the ion source could be a function of the ion source con-
struction. Once an ionized atom strikes a surface, it must
diffuse off that surface in order to be extracted from the
ion source. If the surface does not allow easy diffusion,
the ion may remain trapped inside the ion source. Both boron
nitride and tantalum, the major construction materials in the
ionization cavity, are considered to be materials from which
diffusion is difficult.

Since Burkard et al. [11] did observe some zirconium
fluoride molecular ions in their experiments, the formation
of these complex ions may seem like a reasonable explanation
for the lack of success. However, this may be discounted by
two considerations. First, a very large atomic fluorine beam
was observed. The beam current exceeded 30 uA and indicates
ionization of large quantities of fluorine or fluoride mole-

cules. Some fluorine could come from radiation damage to the uranium tetrafluoride target from the neutron beam. Secondly, on the basis of observations from reference [11], these complex molecular ions were searched for exhaustively, but none were found. This is an even stronger justification for concluding that the ion source was efficiently breaking up any fluoride molecules entering it.

Thus, the failure to observe non-rare gas fission products can be assessed to losses of activity along the transport line and onto the surfaces inside the ion source.

V. DISCUSSION

A. Reasons for Failure

The general failure of the heated uranium tetrafluoride target system can be blamed on design restrictions imposed by the isotope separator configuration at the time of the experiments.

A major difficulty was imposed by the long transfer line required to transport volatile activities to the ion source. The length made it difficult to heat the line uniformly, and the two 90° bends in the line compounded the problem even further. Uniformly wrapping heating tape around the tube at the bends is nearly impossible, so that cool spots can be expected to form at these positions. Even if the transfer line could be heated uniformly, the losses due to adsorption onto the stainless steel surfaces could become a significant portion of the yield.

Some more problems arise due to the ion source design. As mentioned before, it was not practical to heat independently the tube from the ion source base to the ionization cavity, resulting in a cool area where volatile fission products could, and did, condense. Another major concern in the ion source design is the retention of fission products on the materials being used inside the ion source, particularly boron nitride and tantalum. Fission products adhering to the anode, end cap, or other portions of the ion source may be

unable to diffuse into the ion source plasma region, thus reducing the yield.

B. Suggestions for Improvement

The primary improvement that could be made is to shorten the transfer line. A very short, preferably straight, transfer line should greatly reduce the loss of activity before it reaches the ion source. Such a transfer line could be heated evenly by passing a high current through it if it was constructed of a thin metal. Thinness is mentioned only in that thinner metals usually have a higher resistance and hence a higher wattage could be achieved with less current than a thicker tube of the same metal. This would eliminate the problem of maintaining uniformity of temperature encountered with heating tapes. It would also allow the transfer line to be heated all the way into the ion source anode cavity. To avoid oxidation of the material used for the transfer line, a vacuum jacket would need to be built around the transfer line. The jacket would also serve as thermal insulation and could be water cooled, if necessary.

The problems caused by the ion source design could be reduced by replacing the reactive boron nitride insulators with quartz and the tantalum anode with graphite. Diffusion from hot graphite is relatively easy, and the reactive sur-

faces in the ion source could be eliminated. The design and operation of a graphite ion source differs considerably from that of the tantalum constructed "McConnell" ion source, but would not present any real difficulty since the earliest type of ion source used on TRISTAN was of graphite construction.

The biggest drawback of these suggestions is that they require a drastic change in the isotope separator configuration. The ion source would have to be moved much closer to the neutron beam. While this would require a lot of time, effort, planning and money, these things have already been invested in the ion source relocation project, or the in-beam ion source system. The system is discussed by Talbert et al. [5] and involves an ion optics system to bring the ion beam from the ion source to the proper plane and height of the separating magnet. The ion source used is a graphite ion source, with the target on the anode. The system has the flexibility to allow the ion source to be raised slightly above the neutron beam so that a rare gas target could be used if desired. Thus it is possible to have a very short transfer line. The plans for a graphite ion source exist, although they would require considerable alteration for use with the heated target system in mind.

At the time of writing, the in-beam ion source system has been operable for 9 months and has produced successfully non-rare gas fission products, many of which could not have

been produced by a successful heated uranium tetrafluoride target system. Therefore, there is not, at this time, a pressing need to implement these ideas for a new heated target system.

VI. LITERATURE CITED

1. J. R. McConnell and W. L. Talbert, Jr., Nuclear Instruments and Methods 128, 227(1975).

2. O. Almen and K. O. Nielsen, Nuclear Instruments and Methods 1, 302(1957).

3. G. J. Basinger, W. C. Schick, Jr. and W. L. Talbert, Jr., Nuclear Instruments and Methods 124, 381(1975).

4. J. K. Halbig, F. K. Wohn and W. L. Talbert, Jr., Review of Scientific Instruments 45, 789(1974).

5. W. L. Talbert, Jr., F. K. Wohn, J. C. Pacer, A. R. Landin, R. L. Gill, M. A. Cullison, G. A. Sheppard, K. A. Burke, K. L. Malaby and A. F. Voigt, Nuclear Instruments and Methods 139, 257(1976).

6. S. Borg, I. Bergstrom, G. B. Holm, B. Fydberg, L. E. DeGeer, G. Rudstam, B. Grapengiesser, E. Lund and L. Westgaard, Nuclear Instruments and Methods 91, 109(1971).

7. "Handbook of Chemistry and Physics", 49-th Edition, The Chemical Rubber Company, Cleveland, Ohio (1969).

8. M. Weber, N. Trautmann and G. Herrmann, Radiochemistry and Radioanalytical Letters 6, 73(1971).

9. M. Weber, N. Trautmann, H. Menke, G. Herrmann and N. Kaffrell, Inorganic and Nuclear Chemistry Letters 9, 519(1973).

10. M. Weber, N. Trautmann, H. Menke, G. Herrmann and N. Kaffrell, Radiochimica Acta 19, 106(1973).

11. K. H. Burkard, W. Dumanski, R. Kirchner and E. Roeckl, Gesellschaft fur Schwerionenforschung Darmstadt, Report GSI-PB-2-74, 1974.

VII. ACKNOWLEDGMENTS

draft of my thesis and for her, that is pretty good. Barbara put up with many weekends without me and did my share of the housework many times over. In spite of this she seldom complained and always had encouragement to offer.