Uniform Deposition of uranium hexafluoride (UF₆): Standardized mass deposits and controlled isotopic ratios using a thermal fluorination method

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Abstract

We report a convenient method for the generation of volatile uranium hexafluoride (UF₆) from solid uranium oxides and other U compounds, followed by uniform deposition of low levels of UF₆ onto sampling coupons. Under laminar flow conditions, UF₆ is shown to interact with surfaces within a fixed reactor geometry to a highly predictable degree. We demonstrate the preparation of U deposits that range between approximately 0.01 and 500 ng·cm⁻². The data suggest the method can be extended to creating depositions at the sub-picogram·cm⁻² level. The isotopic composition of the deposits can be customized by selection of the U source materials and we demonstrate a layering technique whereby two U solids, each with a different isotopic composition, are employed to form successive layers of UF₆ on a surface. The result is an ultra-thin deposit that bears an isotopic signature that is a composite of the two U sources. The reported deposition method has direct application to the development of unique analytical standards for nuclear safeguards and forensics. Further, the method allows access to very low atomic or molecular coverages of surfaces.

Keywords

fluorination; UF₆; UO₂F₂; UO₂; nuclear safeguards; nuclear forensics

1.0 Introduction

Measurement and monitoring of U enrichment activities is of significant and growing international interest due to concerns over nuclear weapons proliferation. Environmental sampling and analysis is a fundamental tool of the International Atomic Energy Agency's (IAEA's) international nuclear safeguards treaty compliance and verification efforts [1]. Validated standards are essential for any accurate analytical process and are particularly important for measurements having nuclear security and international treaty implications. Generation of standards for U enrichment measurement validation is problematic due to the tendency of UF₆ to sublime and react with moisture and substrates. Standards generation has an additional challenge of making and handling quantitative mixed U isotope standards; the ratio of ²³⁵U to ²³⁸U, for instance, is a key metric for monitoring U enrichment activities. Present IAEA analytical protocols avoid the use of unstable and unavailable UF₆ standards [2]. However, the ability to generate U standards, particularly at low mass levels and with controllable isotopic ratios, would be very useful for analytical systems calibration and validation, analytical protocol development and evaluation, as well as supporting fundamental analytical, chemical, and physical studies of reactive chemical species at low mass coverages.

To begin to appreciate some of the controlling features of U deposition behavior, gaseous UF₆ was produced at mole concentrations ranging from an estimated 0.2 to 88 ppm in a carrier gas stream of ultra-high purity argon (Ar), by reaction of uranium dioxide (UO₂) with nitrogen trifluoride (NF₃), a mild fluorinating reagent [3-5]. The resulting UF₆/Ar stream was introduced into a flow geometry and deposited on sets of planar coupons along the transport path. The use of high specific activity ²³³U was

used to quantify the very low masses of U deposited on the coupons, and to confirm the uniformity of depositions between coupons placed within the gaseous transport path.

In this article, the authors first discuss a simple and unique UF_6 gas generation device coupled downstream to a deposition chamber that enables the creation of U deposits of controlled mass per unit area. Next, the authors demonstrate a process whereby the device can be applied to the preparation of U depositions that contain controlled isotopic signatures. In addition to nuclear safeguards and forensics quality assurance applications, the method has applications in the controlled transport and deposition of volatile species for fundamental and applied studies in areas such as corrosion and catalysis science.

2.0 Experimental

2.1 Reagents, standards, and materials

House stocks of 99.9% ^{233}U (as $^{233}\text{UO}_2$) and highly enriched uranium (HEU) (as $>90\%$ $^{235}\text{UO}_2$) were used for deposition studies. Ultra-high purity (UHP, 99.999%) Argon was purchased from Oxarc (Pasco, WA) and high purity NF_3 (99.99%) was purchased from Air Liquide (Houston, TX).

Gaseous UF_6 was generated and subsequently deposited on a variety of planar substrates (“coupons”) with surface areas of $\sim 3.8\text{ cm}^2$. These included disks of 316 stainless steel (SS), Teflon[®] (TN), cotton cloth (CL), and cellulose filter paper (FP). Additionally, coupons of high purity silicon wafers (SI) were cut into squares with the same approximate surface area.

2.2 Apparatus and method

A simultaneous thermogravimetric and differential thermal analyzer (TG/DTA) (EXSTAR6000, THASS GmbH, Friedberg, Germany) was used to obtain $\text{UO}_2 \rightarrow \text{UO}_2\text{F}_2 \rightarrow \text{UF}_6$ conversion rates as a function of temperature and NF_3 gas concentration. Additionally, the furnace from a modified Seiko Model 200 (RT Instruments, Woodland, CA) was used for the UF_6 deposition work (see Fig. 1). The

modified instrument had a single sample arm that held both the reference and sample pans. The sample arm was made of nickel and was coated with a protective ceramic. A quartz furnace tube was closed over the sample arm and was isolated from ambient atmosphere by a large VCO O-ring fitting arrangement (Swagelok, Solon, OH). The sample, typically UO_2 powder, was added to a nickel sample pan as a solid or as a suspension in deionized water (1 to 50 μL volume addition). The furnace was closed over the sample and it was heated at $5^\circ\text{C}/\text{min}$ to 120°C for an hour while the water was removed under a purge of Ar. The furnace was then ramped to a desired temperature and held isothermally. The U sample was then exposed to 5% NF_3 in Ar. The 5% NF_3/Ar was routed into the furnace through a 1.58 mm diameter nickel tube to ~ 5 cm from the sample pan at a constant flow rate of $200\text{ cm}^3\cdot\text{min}^{-1}$. UF_6 generated by the thermo-fluorination reaction was carried from the furnace by this gas mixture. Temperatures below 500°C were used to avoid the formation of a precipitate of $(\text{NO})_2\text{SiF}_6$ [6] which would otherwise block the passage of gas at the exit of the quartz tube. UF_6 generated by the thermo-fluorination reaction was carried from the furnace by this gas mixture. Planar coupons were fixed to an aluminum foil tray with double-sided tape. The tray was placed inside the deposition chamber and the chamber was closed with 6.4 cm quick flanges for air and moisture-tight sealing of the flow path. The deposition chamber was a 5 cm diameter 316 stainless steel tube, with a length of about 17.1 cm (internal surface area $\sim 308\text{ cm}^2$). The deposition chamber was connected to the output of the furnace with stainless steel tubing about 15.2 cm in length and 1.9 cm in diameter. The length of the connections provided good thermal equilibration of emitted gases from the center of the furnace ($400\text{--}500^\circ\text{C}$) to the deposition chamber, which maintained an internal temperature of $\sim 20^\circ\text{C}$. The deposition chamber and connections to the furnace were not corroded by room temperature exposure to the fluorinating gas [3, 7] and were inexpensive to replace, as they eventually became contaminated with U. The gases were routed out of the deposition chamber to a water bubbler to control U emissions and then were routed to the plenum of the fume hood.

Following a UF₆ deposition experiment, the back flange of the deposition chamber was detached and the coupons removed. Upon removal from the chamber, the U deposit was exposed to ambient air, thus permitting hydrolysis reactions to occur. The coupons were counted directly by alpha spectroscopy (described later). In order to maximize the sensitivity of the measurement, high specific activity (SA) ²³³UO₂ (²³³U SA = 0.357 Bq·ng⁻¹) was utilized as the source U to provide for quantifiable measurements on the sample coupons down to ~10 pg·cm⁻² (~3.6 mBq·cm⁻²).

For each ²³³UO₂ fluorination experiment, samples were exposed at a given temperature to the 5% NF₃/Ar mixture for 1 h prior to their removal from the chamber for subsequent U deposit analysis. In several trials, new coupons were then added to the deposition chamber and the reaction was continued for an additional 5 h without refreshing the source ²³³UO₂. These experiments were run to understand the nature of residual U that might remain in or near the sample pan or on the furnace walls adjacent to the sample pan. Because 5% NF₃/Ar is a mild fluorinating reagent, U volatilization would require temperatures near 500° C for efficient UF₆ production to occur [12]. The experimental apparatus had a steeply diminishing temperature gradient between the sample pan in the center of the furnace (near 500°C) and the deposition chamber (20°C). Potential U residues in and around the sample pan may or may not have been pure UF₆ but plausibly could have included silica bearing or metal bearing uranium. It is therefore expected that secondary processes of thermal desorption of UF₆ contributed to the overall U deposit observed on the second set of sampling coupons.

The text below introduces the concept of “fast volatilization” of UF₆ as produced in several minutes, as compared to “slow volatilization” of UF₆ that might last over hours to days. The latter is used to describe volatilization behavior observed in the standard TG experiment wherein the mass loss and heat of reaction over time can be monitored. For the small U masses used in the majority of the experiments described here, a fast volatilization regime is described wherein mass detection was below the sensitivity of the analytical balance in the TG. The heat signature for the exothermic reaction of UO₂ to UF₆ was

generally too small to be of quantitative value for most of the U samples used in these experiments. However, the heat of reaction was recorded by the sample thermocouple in a few trials. The largest of these occurred for the higher U source masses ($>100\text{ }\mu\text{g}$) listed in Table 1 and qualitatively indicated that the majority of the U was volatilized in 10 minutes or less.

The duration of sample volatilization is important because it allows for an estimate of the mole concentration of UF_6 evolved in the NF_3/Ar carrier stream over a time interval, at a given flow rate. This in turn provides the perspective that UF_6 mole concentrations across all experiments were quite dilute ($<100\text{ ppm}$) in the carrier gas stream. For source masses under $100\text{ }\mu\text{g}$ we have calculated a range of probable UF_6 concentrations as volatilized between 1 and 10 minutes (Table 1).

2.3 Preparing deposits with isotopic mixtures of U

The experimental apparatus was used to demonstrate an isotope layering approach, whereby two sources of UO_2 (HEU and ^{233}U) would be sequentially deposited on a coupon to create a controlled isotopic mixture. First, several milligrams of HEU were placed in a sample pan that was then loaded into the furnace. A clean SS coupon was placed in the deposition chamber.

The HEU was then exposed to the NF_3/Ar gas flow and heated, under fast volatilization conditions (1 h), to provide a HEU deposit. Upon completion of the reaction, the coupon was removed from the deposition chamber, and the alpha energy spectrum (AES) of the HEU deposit was obtained by AEA. The ^{235}U alpha activity allowed the deposit mass and spectral resolution to be calculated. Next, the pan originally containing the HEU was used to receive an aqueous suspension of $^{233}\text{UO}_2$. The sample coupon holding the HEU deposit was returned to the deposition chamber. After evaporating the water from the pan at 120°C under Ar flow (as described previously), a fast volatilization (1 h) was performed. Once again, the sample coupon was removed from the deposition chamber and its AES was obtained. The

²³³UO₂ fast volatilization on the same coupon was repeated two additional times, and an AES was obtained after each coupon exposure.

2.4 System contamination control

The volatilization of ^{233,235}UO₂ resulted in contamination of the interior surfaces of the furnace, connecting conduit, and the deposition chamber. Contamination levels were proportional to the mass of U source material loaded into the sample pan. It proved relatively simple to remove the instrument contamination by washing the surfaces with dilute nitric acid and water, air-drying overnight, and then purging the instrument at 500° C with 30% NF₃. It became impractical to remove all contamination if picogram deposition levels were of interest. Prior to performing ultra-low depositions, the deposition chamber, the attachments to the furnace, the furnace tube, and the sample arm were replaced.

2.5 Alpha energy analysis (AEA)

The mass and isotopic composition of U deposited on the coupons within the deposition chamber was determined by AEA using an Alpha Duo[®] 2-channel alpha spectrometer running on MAESTRO[®] alpha analysis software (v. 6.08) (ORTEC, Oak Ridge, TN). Sample coupons were positioned ~1 cm below the passivated implanted planar silicon diode detector, and were counted in vacuum for up to 24 h. Detection efficiency (19.8±0.1%) was determined by an electroplated ²³⁹Pu/²⁴¹Am/²⁴⁴Cm calibration source prepared by the National Institute of Standards and Technology (NIST). Energy calibration of the spectrometer was likewise performed using the NIST standard, as the peak maxima channels for the three electrodeposited actinides were correlated to their primary alpha energy emissions. Convolved alpha energy spectra of U isotopic mixtures were deconvolved using PeakFit automated peak separation analysis software (v. 4.12, Systat Software, Inc., San Jose, CA), using the “AutoFit Peaks I Residuals” method.

The mass (ng) of each isotope deposited on the 3.8 cm² coupons (*M*) was calculated by Eq. 1,

$$M = A_{\alpha} \cdot (I_{\alpha} \cdot SA \cdot E_d)^{-1} \quad (1)$$

where A_{α} is the integrated area of the alpha peak (cps), I_{α} is the intensity fraction of the isotope's alpha emission (obtained from the National Nuclear Data Center [8]), E_d is the detection efficiency of the alpha spectrometer (cps/Bq), and SA is the specific activity of the isotope (Bq/ng) according to Eq. 2:

$$SA = \lambda \cdot A_v \cdot u^{-1} \quad (2)$$

where λ is the decay constant of the radionuclide (s⁻¹), A_v is Avogadro's constant (6.022x10¹⁴·nmole⁻¹), and u is the molar mass of the isotope (ng/nmole) [9].

3.0 Results and discussion

3.1 Two approaches to uniform and low mass UF₆ deposition

Two methods for depositing very small amounts of UF₆ with known isotopics on a variety of planar materials (coupons) were developed. The first used “slow volatilization” of U with an extended deposition period over several hours or days and the second approach used a “fast volatilization” condition whereby deposition occurred within about 1 h. The general experimental process is shown as a flow diagram in Fig. 2, with a graphical representation of the apparatus shown in Fig. 1.

Rates of gas solid reactions, in this case the fluorination and volatilization of U solids, can vary extensively depending upon the source material's morphology, particle size and shape, and process history [10, 11]. However, TG data can be used to quickly bracket the rate of fluorination and volatilization for a given material. Once the fluorination behavior for a particular material is determined by the TG measurement, the rate of mass loss (UF₆ volatilization) can be controlled by either the reaction temperature or the NF₃ concentration to produce consistent small mass depositions of UF₆ downstream from the point of generation.

3.1.1 Slow UF₆ volatilization

The reaction of 5, 10, and 30% NF₃ in Ar carrier gas with UO₂ to produce UF₆ near 450°C is shown in Fig. 3 (Left). The reaction kinetics with varied NF₃ concentration provides a convenient visualization of how a low pressure release of UF₆ from the instrument can be accomplished. The reaction proceeds in two steps:

The reaction of solid UO₂ with NF₃ produces an initial mass increase as UO₂ is converted to nonvolatile UO₂F₂(s) at a rate $d[\text{UO}_2\text{F}_2(s)]/dt$ (Eq. 3). Continued exposure of the UO₂F₂ to NF₃ results in mass loss as UF₆(g) is formed in the second step (Eq. 4), and is volatilized from the sample pan at an observed rate on the TG (labeled as $d[\text{UF}_6]/dt$ in Fig. 3 (Left)).

A region we refer to as the “flat top”, where $-d[\text{UO}_2\text{F}_2(s)]/dt = d[\text{UF}_6(g)]/dt \sim 0$,¹ is a general feature of the reaction of NF₃ with UO₂, NpO₂, and PuO₂ [5]. Research conducted at PNNL appears to indicate that the “flat top” formation caused by NF₃ is a characteristic shared across other actinide dioxides [4, 5, 12].

As shown in Fig. 3 (Left), the duration of the flat top region can be reduced or extended by a change in NF₃ concentration, or by varying furnace temperature. Careful examination of the flat top regions in the figure indicates that the sample masses approach the theoretical maximum conversion (~14% mass increase) to UO₂F₂. The rate of UO₂F₂ mass change can be made quite slow, but is actually never quite zero (i.e., a “leak” of UF₆(g) occurs in the flat top region). In other words, the formation of UF₆ (Eq. 4) competes with the formation of the non-volatile UO₂F₂ (Eq. 3). The disappearance of UO₂F₂ is much faster with the use of stronger fluorinating reagents where a flat top region cannot be observed [13]. Even

¹ The region where the mass change recorded by the TG microbalance becomes virtually imperceptible over time.

during the relatively rapid conversion of the dioxide to UO₂F₂(s) using 450°C 30% NF₃/Ar (trace “c” in Fig. 3), non-discernable volatilization of the hexafluoride does occur. These small releases of UF₆(g) were below the mass sensitivity of the TG instrument ($\pm 3 \mu\text{g}$) and were not appreciable to us until higher the specific activity ²³³U isotope was used to track the behavior at very low mass levels via post-reaction analysis of the sample coupons by AEA.

Fits to the curves similar to those in Fig. 3 (trace “c”) have been reported and indicate that NF₃-U solid reaction rates are a convolution of chemical and diffusional components [11]. For instance, the UO₂ conversion shown in Fig. 3 (trace “a”) for 1, 5, 10, and 30 h exposures should all produce about zero UF₆ according to $-d[\text{UO}_2\text{F}_2(s)]/dt = d[\text{UF}_6(g)]/dt \sim 0$, except for the existence of the indeterminate leaks. These small leaks, amounting to micrograms of U, appear to result from volatilization of very small particles in the U sample. As the smallest of particles are reacted away, a very low release of U follows along the flattop because of the diffusional nature of the UO₂→UO₂F₂→UF₆ reaction. This continues until the flat top region is breached and determinate mass changes are engaged ($d[\text{UF}_6(g)]/dt > 0$).

For a fixed NF₃ concentration and temperature, the concentrations of ²³³UF₆(g) volatilized in the flat top regions deposit on the transport walls, the deposition chamber, and sample coupons placed therein. Consequently, a practical use of the flat top region, for the actinide dioxides in particular [5], is that uniform mass depositions can be achieved by using the good rate control it provides. However, the time required to produce deposits can be quite long. We refer to the approach of using the flat-top region to provide a source of UF₆ as “slow volatilization”.

Table 2 shows the effects of temperature and exposure time on the deposition quantity and reproducibility of UF₆ on sample coupons positioned in the deposition chamber. For all depositions shown in Table 2, 5% NF₃ was reacted with ²³³UO₂. Experiments 1 and 2 were performed under slow volatilization conditions. The different coupon materials, as listed in Table 2, were used across the experimental set to assess if a change in deposition quantity with coupon type could be observed. For

deposition masses within the range of those listed in Table 2, the coupon material did not appear to impact the deposition process.

Experiment 1 in Table 2 is an example of a very slow volatilization of UF₆. The ²³³UO₂ source mass was 263 μg (231 μg ²³³U) and the coupons were SS. The furnace temperature was set at 454° C to cause a UF₆ release in the flat top region (similar to trace “a” in Fig. 3). The amount deposited in the 1 h run on three SS coupons did not differ significantly from that observed in the subsequent 5 h run because of the low UF₆ production and release rate along the flattop region. The effect was imposed by the low reaction temperature and NF₃ concentration at which even the smallest particles in the sample were slowly reacted. However, a decrease in sample-to-sample variability in the deposition mass was observed for the subsequent 5 h exposure (13% vs. 42% relative standard deviation for the 5 h and 1 h exposures, respectively).

Experiment 2 was performed at a higher temperature (478° C) but lower mass of source ²³³UO₂. At this temperature and 5% NF₃, a shorter duration flat top and faster UF₆ release than that shown in trace “c” of Fig. 3 resulted. After a 1 h exposure, the total U deposited on the coupons was significantly greater than the first experiment (3.6±0.7 vs. 1.2±0.7 ng·cm⁻²) because of the ~20°C temperature increase, even though the source mass was about 3.5x lower. The subsequent 5 h exposure on fresh coupons resulted in a ~12x smaller deposited mass (0.3±0.1 ng·cm⁻²). This occurred because the higher temperature more quickly purged or volatilized smaller particles from the source mass during the initial 1 h exposure than was observed in experiment 1. This odd result in which longer exposure time (5 h) led to reduction in deposit mass, indicates that only a residual U mass is left at the source after the initial volatilization (1h) is complete. The residual U is still a viable source of reproducible deposition. The idea was tested extensively below.

3.1.2 Fast UF₆ volatilization

It was observed that if the source mass that produced the volatile fluoride was 100's of micrograms and lower, then trace UF₆ deposition could be approached in a more practical, (i.e., faster) manner. For low source masses of U solids, downstream deposition of UF₆ essentially became independent of the rate of volatilization. As the amount of available material volatilized per unit time tended toward zero, very uniform and trace depositions were achieved. This "fast volatilization" process is shown in Fig. 3, Right; it illustrates the complete volatilization of several milligrams of UO₂ over ~10 min by thermogravimetric analysis. If this quantity of source material can be volatilized in 10 min, it is highly probable that the microgram quantities of material used in the fast volatilization experiments (Table 1, experiments 3-9) react at and below this time scale. This fast volatilization behavior has been used not only for fluorination of UO₂, but for other forms of U as well (e.g., UO₂(NO₃)₂, UO₂F₂, UF₄, etc.) [4].

Stable and controlled deposition of ²³³UF₆ was thus achieved by fast volatilization of low source mass ²³³UO₂. Experiments 3 through 9 in Table 2 were run between 485° and 495° C and for 1 or 5 h durations. These experiments were run to determine preferred operating conditions that enable rapid and effective generation of uniform U depositions at selected concentrations. At this increased temperature range, much faster rates of UF₆ volatilization occurred, with >95% of the U source material volatilized in 10 min or less. Experiments 3 and 4 (485° and 488° C) used 158 and 114 µg ²³³U, respectively. Low variability in deposited mass resulted across these coupons, with relative standard deviations of 8% and 1% for experiments 3 and 4, respectively. Experiments 5 through 7 were also performed at 488° C, but the source mass of ²³³U was stepped down from 41.1 to 26.6 to 1.2 µg. The resulting deposited ²³³U mass decreased across these three experiments (down to 0.07±0.02 ng·cm⁻² for experiment 7), but the precision also decreased with diminishing source ²³³U mass (7% for experiment 5; ~30% for experiments 6 and 7).

For experiments 5 and 7, sets of coupons were exposed for 1 h and then the coupons were replaced with a second set that were exposed for 5 h, without refreshing the source U. Whereas the 1 h exposure

for experiment 5 resulted in a deposited mass of $3.0 \pm 0.2 \text{ ng} \cdot \text{cm}^{-2}$, the subsequent 5 h exposure resulted in a ~30 fold drop in ²³³UF₆ deposition, as less ²³³U was available in the furnace region following the initial 1 h reaction. A similar observation occurred with experiment 7. The ²³³U deposition quantities dropped from $0.07 \pm 0.02 \text{ ng} \cdot \text{cm}^{-2}$ for a 1 h exposure to $0.01 \pm 0.00 \text{ ng} \cdot \text{cm}^{-2}$ for the subsequent 5 h exposure. The second coupon set, placed in the deposition chamber at the end of the first hour, received very little UF₆ because most of the U source material was volatilized during the first hour. The sequential coupon exposure technique described here clearly provides a path to ultra-low UF₆ depositions.

Experiments 8 and 9 were run at the hottest furnace temperatures (~490° C) and highest ²³³UO₂ masses of the experimental set. At these temperatures, most of the source material would be expected to volatilize within 10 min, despite the quantities of ²³³UO₂ present. While the coupon set from experiment 8 was exposed within the deposition chamber for 1 h, the experiment 9 coupon set was exposed for 5 h. Evaluation of the data reveals one aspect worth noting: UF₆ deposits formed within the deposition chamber appear to be stable over time. Experiment 9 had 1.9x greater ²³³U source mass than experiment 8; correspondingly, the resulting mean ²³³UF₆ deposit mass ratio was calculated at $2.0(\pm 0.3)$ times greater for experiment 9 vs. 8. This seems indicative that the UF₆ deposit following a fast volatilization is quite stable, despite up to 5 h continuous exposure to nominally room temperature 5% NF₃/Ar gas flow. Otherwise, the proportionality between the ²³³UF₆ deposits and the source ²³³UO₂ masses would not have been maintained.

Finally, we contrast the fast volatilization experiments 3 and 8 (that used similar source masses) to the slow volatilization technique demonstrated in experiment 1. The deposition masses in the latter were a factor of ~10 lower because of the strong temperature dependence on the rate of UF₆ release. As a result, only a small fraction of the total U available was volatilized at this lower temperature. The quantity of UF₆ deposited on the sampling coupons in experiment 1 is a reflection of the partial pressure of UF₆ volatilized in the flat top region along which we have approximated the UF₆ release to be zero. The fact

that the deposit did occur, and is in line with deposition mass data taken under fast volatilization conditions, suggests that similar partial pressures overall were accessed for both approaches. The deposited mass in experiment 1 was the lowest of all the 1 h exposures in Table 2 except for experiments 6 and 7, which used 26.6 and 1.2 μg U source material, respectively. The fast volatilization of the majority of a 1.2 μg U sample (experiment 7) conservatively took less than 10 min. The maximum UF₆ concentration would have likely been <2 ppm in the NF₃/Ar carrier stream (Table 1). Consequently, we see that the leaks of UF₆ that occur before and along the flat top regions in Fig. 3 (traces a, b, and c) have become not so indeterminate and in retrospect can be a reproducible method of UF₆ delivery.

Compilation of the fast volatilization experimental data revealed a strong correlation between U source mass and U deposits within the deposition chamber. In Fig. 4a, the ²³³U source mass is plotted against the deposition mass (corrected to the surface area of the coupon) using the 1 h coupon exposure data for temperatures between 478-495° C (fast volatilization experiments 3 through 9 in Table 2). Experiment 1 is not used in the plot; because of the low furnace temperature only a fraction of the U sample was volatilized. Experiment 2 used a lower temperature as well, but virtually the entire U sample was consumed in the first hour. Therefore, experiment 2 data is included in Fig. 4a. Experiment 9 had a 5 h duration, but given the similarity of conditions to experiment 8 (whereby the sample pan was empty after 1 h), this data was included as well. Regression analysis of the data set indicates that ~65 pg ²³³U·cm⁻² was deposited for every microgram of originating ²³³U in the sample pan. Based on this deposition rate, and deposition chamber's internal surface area of ~308 cm², it can be estimated that only ~2% of the ²³³U source mass is captured within the deposition chamber (and coupon) surfaces. The remaining mass remains in the sample pan and furnace / conduit walls or migrates out of the chamber's exit portal.

The linearity of the data shown in Fig. 4a highlights the predictability that the geometry of the apparatus imposed on the deposition, so long as fast volatilization conditions are met. It also suggests

that an expanded range of mass depositions are possible with either lower or higher source masses of U. For uniform depositions to occur using larger source masses, the rate of formation and volatilization of UF_6 should be minimized to maintain a dilute release condition (i.e., as per Table 1). Otherwise, we hypothesize that aggregation of the target gas molecules would contradict the non-interaction condition required to produce the uniform depositions described here. The data also suggests that if sub-picogram depositions were targeted using ^{233}U , sub-nanogram source masses would be required. Alternately, as shown in experiments 5 and 7 in Table 2, sets of new coupons could be added after a 5 h period, then after a 10 h period, and so on, to achieve progressively lower deposition masses.

The use of other U source materials, such as UO_3 , U_3O_8 , $\text{UO}_2(\text{NO}_3)_2$ were investigated, and the results of the fast volatilization of microgram scale source masses of these varied U solids did not change the deposition characteristics. At the appropriate temperature, NF_3 will convert any form of U to UF_6 [4]. The source material can be added to the sample pan as a solution or a suspension. However, aqueous suspensions require drying prior to NF_3 exposure. When adding dissolved U salts to the sample pan (e.g., $\text{UO}_2(\text{NO}_3)_2$), calcination to UO_3 , U_3O_8 , or other thermally stable form should first be performed at the appropriate temperature.

Fig. 4b shows a scanning electron microscopy (SEM) image of uranium deposited on a stainless steel coupon. The image is not representative of the deposits discussed above because the quantity of uranium on the coupon is 1000x greater than highest deposit discussed in Table 2. The inset on the image also allows view of some of the structure common to these higher levels of uranium deposition. It is possible to image changes in the morphology of these structures with variable atmospheric condition and change in deposition surface. However, at the lower levels of deposition in Table 2 the structures became increasingly tenuous and challenging to image at all.

3.2 U Deposits with varying isotope compositions

The U volatilization and deposition process described above provides an approach to prepare mixed and variable isotopic ratio standards. As previously noted, the ratio of ²³⁵U to ²³⁸U is a key metric for monitoring U enrichment activities. Generation of standards with controlled U isotopic ratios is important for nuclear safeguard analytical efforts. Additionally, generation of standards with other actinides and metals would be useful in supporting safeguards as well as post-detonation debris analysis R&D.

In addition to controlled mass deposits of UF₆, the described system enables the controlled variation of isotopic ratios within those deposits. Two methods of producing mixed isotopics were investigated. The first method requires the combination of two isotopically unique sources into the sample pan, followed by volatilization of the mixture. This is accomplished via the addition of two isotopically unique U solid sources (heterogeneous addition), or via the addition of two solubilized sources of U (homogeneous addition). The second method involves a layered deposition via sequential source material volatilizations. In other words, a second isotope or isotope mixture would be layered atop a coupon that had already received a deposition from an isotopically distinct source. For confirmation of the resulting standard set's isotopic fingerprint, several coupons would be run in parallel, and the deposition mixture would be confirmed by splitting the resulting coupon set, followed by radiometric or mass spectrometric determination of one or more of the coupons within the set.

The second method described above is demonstrated herein. A source of HEU (as UO₂) was added to a sample pan placed on the sample arm of the furnace. Because of the ~4500x lower SA of ²³⁵U vs. ²³⁸U, several milligrams of HEU source material needed to be added to the sample pan in order for the resulting deposition to be quantifiable by alpha spectrometry. Then, a clean sample coupon was placed in the deposition chamber. The HEU was exposed to the NF₃/Ar gas flow and heated to provide a HEU deposition. Upon completion of the reaction, the coupon was removed from the deposition chamber, and

the AES of the HEU deposit was obtained by AEA. This original AES can be seen in Fig. 5 (solid black line). The spectrum shows the characteristic alpha signal for ²³⁵U (predominant peaks being 4.36 MeV (18.9%) and 4.40 MeV (57.7%) plus others), along with two ²³⁴U alpha peaks (4.72 MeV (28.4%) and 4.77 MeV (71.4%)). The integrated area of the small ²³⁵U alpha peak was initially used to estimate that the coupon received a deposit of ~1.7 μg ²³⁵U (~440 ng·cm⁻²). Little, if any, alpha energy attenuation was observed from this deposition. Based on the 4.77 MeV primary ²³⁴U alpha peak, the full width at half maximum (FWHM) was calculated to be ≤32 keV. This is in close agreement with the FWHM observed using a NIST traceable electrodeposited AEA calibration source that gave a FWHM of 28±3 keV, as would be expected from the small mass of material deposited on the surface.

The HEU loaded coupon was then returned to the deposition chamber, and the sample pan was spiked with an aqueous suspension of ²³³UO₂ particles. The fluorination process was repeated using a fast volatilization condition in order to create the first layer of ²³³U on the original HEU deposit. A subsequent AES (Fig. 5, blue dotted line) shows the emergence of the ²³³U alpha peak (4.82 MeV, 84.3%) above the 4.77 MeV alpha emission of the original ²³⁴U peak. The same process was repeated twice more to create layers 2 (purple solid line) and 3 (red dotted line). Whereas the ²³³U alpha signal increased with each added layer, the original HEU alpha signals appeared to remain constant.

Peak separation analysis software was employed to deconvolve the four composite alpha spectra (Fig. 5) so that each component of the spectra could be quantified, and the peak resolution for the top and bottom U layers could be determined. Fig. 6a provides peak height normalized AES for individually deposited sources of the HEU and ²³³U oxide materials deposited in the manner described above. The sum of the two normalized spectra is also shown for the condition whereby the primary ²³⁴U and ²³³U alpha peaks are of equal height. Fig. 6b shows the results of the deconvolution process for the discrete HEU and ²³³U sources. The peak analysis software identified the two primary ²³⁴U alpha emissions (234

#1 = 4.72 MeV; 234 #2 = 4.77 MeV) and three of the primary ²³³U alpha emissions (233 #1 = 4.73 MeV (1.6%); 233 #2 = 4.78 MeV (13.2%); 233 #3 = 4.82 MeV (84.3%)).

Deconvolution of the four complex alpha spectra shown in Fig. 5 was performed, using the deconvolutions in Fig. 6b as a guide. Once this was completed, the peak resolution of the major ²³⁴U and ²³³U peaks (via FWHM) was calculated as being between 26 and 31 keV. This is in close agreement to the peak resolution observed with the electroplated source, and it was concluded that the resolution of the “top” ²³³U layer was instrumentally indistinguishable from the “bottom” HEU layer (i.e., no significant self-absorption of ²³⁵, ²³⁴U alpha emissions was observed).

The deconvolved alpha peak areas were subsequently utilized to determine the mass of each U isotope on the sequentially layered coupon. This was accomplished by considering the calculated peak areas, AEA system detection efficiency, alpha emission intensities [8], and U isotope specific activities, as per Eq 1. Table 3 provides a listing of the determined U isotope masses deposited per unit area of the coupon. The table shows that the calculated ²³⁵U and ²³⁴U depositions remained constant throughout the sequential ²³³U depositions, with the isotopes measuring at 470±34 and 2.34±0.32 ng·cm⁻², respectively. The mass of ²³³U is shown to increase from 0.38 to 1.05 to 1.54 ng·cm⁻² with each successive deposition layer from the ²³³UO₂ source. The minute quantity of ²³³U (0.09 ng·cm⁻²) found in the original HEU deposition was likely from contamination of system components from earlier ²³³U studies.

3.3 Stability of U Deposits on Surfaces

The stability of the originally deposited HEU layer observed during the series of subsequent ²³³UF₆ layering treatments indicates that the deposit was firmly attached to the coupon surface, regardless of the coupon type used. As previously described, the initial temperature ramp under Ar flow provided continuous removal of moisture and atmospheric reactants (e.g., O₂, CO₂) from the UO₂ source material and the chamber as a whole. This assured the exclusive production of UF₆ once the fluorinating reagent

was added to the gas stream. As UF₆ has a high vapor pressure (~11.7 kPa at ~21° C [14, 15]) in a dry environment, it would be expected to sublime from the coupon over time if it remained UF₆. Yet, the AEA-determined mass of the HEU deposition layer remained constant throughout the subsequent multi-step ²³³U layering process. In addition to several NF₃/Ar gas stream exposures in the deposition chamber, the deposit was exposed to an estimated cumulative 80 h of atmosphere and 60 h under vacuum within the alpha spectrometer.

Since no specific effort was made to protect the U deposits on the coupons from adventitious moisture upon their removal from the deposition chamber, hydrolysis to non-volatile UO₂F₂ (via Eq. 5) certainly occurred to some degree, thus providing a source of UF₆ alteration [16, 17].

There are currently no methods for chemical speciation at the level of uranium deposition we have presented. In fact (at the deposition levels discussed) we have had little success with even imaging these deposits with state of the art microscopes available to us. It may be stated here that metal fluorides can be produced along with lower valence (and non-volatile) uranium fluorides (e.g., UF₄, U₂F₉, UF₅) by reactions of metals and their oxides with UF₆ [18]. For instance, UF₄ and UO₂F₂ have been observed around breaches in UF₆ cylinders with attendant iron fluoride products [19]. Consequently, the UF₆ might partially or completely react with the surface of a SS coupon. Additionally, UF₆ will react with the silicon coupons to yield UF₄ and SiF₄, and similarly with the nitrocellulose filter paper to yield UF₄ and CF₄ [20]. No surface reaction should have occurred on the Teflon coupons.

The mechanism(s) that promotes the film stability on the different coupon types is not completely understood at present. Regardless, the stability of the deposits appear to indicate that films prepared in the manner described are robust and could conceivably be stored and/or shipped to end users without fear of film degradation.

4.0 Conclusions

An experimental apparatus and operating conditions to produce low mass depositions of UF₆ with specified isotopic ratios that have the potential to be used as isotopic standards has been developed and demonstrated. The approach and system are simple, using a furnace to heat NF₃ and U to produce gaseous UF₆ which is transported to a room temperature chamber containing target coupons where the U deposits. By controlling the original U source mass, furnace temperature, fluorinating gas concentration and exposure time, the amount of deposited U can be reproducibly controlled. Uranium-233 deposits ranging between 0.01 and 27 ng·cm⁻², as well as a much higher mass deposit of 470±34 ng HEU·cm⁻², were produced within the experimental apparatus. Combined, this represents a demonstrated U mass deposition range of ~50,000x. Additionally, the deposition method can be used to prepare successive layers of U isotopics in order to create unique isotope signatures on coupons. The deposits have been demonstrated to be stable toward changes in atmospheric and vacuum conditions.

Extension of the method to controlled UF₆ depositions at and below picogram levels should be possible, but quantitative validation of depositions at these ultra-low mass levels would require careful application of high sensitivity mass techniques. Alternatively, it would require the use of AEA measurements coupled with U isotopes of higher specific activity than ²³³U (e.g., ²³²U; SA = 828 Bq·ng⁻¹).

Analysts involved in the measurement of perturbed U isotopic ratios or other signatures may be able to employ deposits such as those described as quality control standards; these may have application in the field of nuclear safeguards verification and forensics. The method has additional application to fundamental studies regarding ultra-low mass depositional processes and in the controlled deposition of other elements amenable to conversion into volatile fluoride species.

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Figure Captions

Fig. 1. Experimental apparatus for deposition of UF_6 onto coupons. The U source material is added to a pan at the end of the sample arm. An influent mixture of NF_3 and Ar converts UO_2 to UF_6 (white molecules). The UF_6 is transported through a conduit by the Ar carrier gas flow (green atoms). Next, the $\text{UF}_6/\text{NF}_3/\text{Ar}$ stream enters the deposition chamber. UF_6 is deposited on the surfaces of the deposition chamber. This includes deposition onto the sample coupons placed on a sample tray (tray not shown). Residual UF_6 flows from the deposition chamber and is captured by a water trap.

Fig. 2. General process flow diagram for the generation and deposition of UF_6 onto sample coupons placed downstream of the furnace apparatus.

Fig. 3. TG scans of the rate of UF_6 production as a function of time with NF_3 . (Left) Slow volatilization with 5, 10, and 30% NF_3 at 450°C show conversion of UO_2 initially to UO_2F_2 and then to UF_6 . (Right) Fast volatilization of UO_2 to $\text{UF}_{6(g)}$. Between 0 and 30 min, temperature was heated at $20^\circ\text{C}\cdot\text{min}^{-1}$ and held isothermal at 488°C , 5% NF_3 turned on at 488°C .

Fig. 4. (a) Deposition of ^{233}U as $^{233}\text{UF}_6$ (corrected to coupon surface area) as a function of ^{233}U source mass (as $^{233}\text{UO}_2$). The legend indicates the experiments listed in Table 2. Linear regression analysis provides the line $y = 0.0646x - 0.1684$ ($R^2 = 0.9969$). (b) An SEM image of uranium deposited on a stainless steel coupon. The quantity of uranium on the coupon is approximately a factor of 1000 greater than highest deposit discussed in Table 2. The inset on the image also allows view of some of the structure common to these higher levels of deposition.

Fig. 5. Three successive deposition layers of ^{233}U atop a coupon initially prepared with a deposition of HEU (solid black line). Resolved and partially resolved alpha peaks are labeled with the source isotope.

Fig. 6. (a) Overlaid convolved alpha energy spectra of enriched $^{234}, ^{235}\text{U}$ and ^{233}U sources, normalized to the peak heights of the primary alpha emissions: 4.77 MeV (71.4%) ^{234}U and 4.82 MeV (84.3%) ^{233}U . Solid line is the sum of the two spectra. (b) Deconvolution of alpha energy spectra for ^{234}U and ^{233}U alpha emissions showing multiple peaks for each isotope.

Table 1. Source mass of U (as ²³³UO₂) and estimated UF₆(g) mole concentration ranges under fast volatilization conditions, whereby conversion of UO₂ to UF₆ occurs within a range of 1-10 min. Flow rate of 5% NF₃/Ar mixture is 200 cm³·min⁻¹. Total pressure = $P_{Ar} + P_{NF_3} + P_U \approx 1.01 \times 10^5$ Pa.

Exp't	Mass ²³³ U, μg ^a	UF ₆ mole concentration, ppm	
		1 min Volatilization ^b	10 min Volatilization ^c
2 ^d	65.8	88.4	8.8
3	158.3	---	21.2
4	114.3	---	15.3
5	41.1	55.2	5.5
6	26.6	35.7	3.6
7	1.2	1.7	0.2
8	237	---	31.8
9	448	---	60.2

a. Mass of ²³³U in ²³³UO₂

b. 200 cm³ swept volume

c. 2000 cm³ swept volume

d. Due to its lower reaction temperature, this experiment was considered a slow volatilization, but UF₆ evolution was sufficiently complete within a short timescale to consider it herein.

Table 2. Deposition of gaseous ²³³U onto sets of sample coupons from ²³³UO₂ source material reacted by thermo-fluorination. Furnace temperature, ²³³UO₂ source mass, and coupon type were varied.

Exp't	Mass ²³³ U, μg ^a	Temp, °C	Time, h	Deposited Mass on Replicate Coupons, ng·cm ⁻² b, c						Ave, ng·cm ⁻²	± 1σ
				1	2	3	4	5	6		
1	231 ^d	454	1	1.8 ^{SS}	0.8 ^{SS}	0.9 ^{SS}				1.2	0.5
		456	5	0.8 ^{SS}	0.9 ^{SS}	0.7 ^{SS}				0.8	0.1
2	65.8 ^d	478	1	3.3 ^{FP}	3.7 ^{FP}	4.5 ^{FP}	2.8 ^{FP}			3.6	0.7
		478	5	0.3 ^{FP}	0.4 ^{FP}	0.3 ^{FP}				0.3	0.1
3	158.3	485	1	10.3 ^{SS}	9.4 ^{SI}	8.8 ^{SI}				9.5	0.8
4	114.3	488	1	7.2 ^{SS}	7.5 ^{SS}	7.2 ^{SS}				7.3	0.1
5	41.1 ^d	488	1	2.8 ^{FP}	3.3 ^{FP}	3.0 ^{FP}				3.0	0.2
		488	5	0.12 ^{FP}	0.11 ^{FP}	0.11 ^{FP}				0.11	0.01
6	26.6	488	1	1.3 ^{FP}	2.4 ^{FP}	1.8 ^{TN}	2.6 ^{TN}			2.0	0.6
7	1.2 ^d	488	1	0.06 ^{SI}	0.05 ^{SI}	0.09 ^{SS}				0.07	0.02
		488	5	0.01 ^{SS}	0.01 ^{SS}	0.01 ^{SS}				0.01	0.00
8	237	490	1	10.3 ^{SS}	16.4 ^{SS}	16.0 ^{SS}	10.0 ^{SS}			13.2	3.5
9	448	495	5	34.4 ^{TN}	28.5 ^{TN}	26.1 ^{TN}	22.5 ^{CL}	26.0 ^{CL}	22.0 ^{SS}	26.6	4.5

a. Mass of ²³³U in ²³³UO₂b. Surface area of the coupons was ~3.8 cm²

c. Superscripted labels indicate coupon type: Stainless steel (SS); filter paper (FP); silicon (SI); cloth (CL); Teflon (TN).

d. Sequential experiments. Coupons removed following first 1 h deposition; replacement coupons added and then exposed for an additional 5 h.

Table 3. Calculated U deposition masses per cm² for an originating source of HEU and three successive deposited layers of ²³³U. U isotope masses were calculated from the four alpha energy spectra (Fig. 5) following alpha peak deconvolution.

Deposition Sequence	Sample ID	U Isotope Mass Conc., ng·cm ⁻²		
		²³⁵ U a	²³⁴ U b, c	²³³ U b, d
1	HEU	440	2.34	0.09
2	²³³ U Layer 1	453	2.37	0.38
3	²³³ U Layer 2	469	2.28	1.05
4	²³³ U Layer 3	518	2.37	1.54
	Average	470	2.34	---
	± 1σ	34	0.32	---

- a. Alpha peak areas taken directly from AES
- b. Alpha peak areas obtained by deconvolved AES
- c. Values based on the mean of deconvolved 4.72 MeV (28.4%) and 4.77 MeV (71.4%) ²³⁴U alpha emissions
- d. Values based on the 4.82 MeV (84.3%) ²³³U alpha emission only

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

UF₆ formation, transport & deposition on planar surfaces