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Study of Chemical Changes in Uranium Oxyfluoride Particles

Progress Report November 2009 - July 2010

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This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344 and under the auspices of the European Commission, DG Joint Research Centre under Action Sheet 36. The analyzed samples were generated by the IRMM expressly for the purpose of this experiment and we have no evidence that fugitive emissions from a uranium enrichment facility would, or would not, appear similar to the analyzed samples.

Study of Chemical Changes in Uranium Oxyfluoride Particles

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LLNL-TR-

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Project definition

An important tool in the detection of undeclared nuclear activities is the analysis of environmental samples. Since the early 1990s, safeguards organizations such as the International Atomic Energy Agency (IAEA) and Euratom have been collecting environmental samples at nuclear facilities across the world [1]. The strength of environmental sampling lies in the observation that processes involving the treatment of nuclear material release small amounts of process material to the environment. This process material, together with an excess amount of dust, is collected by swiping surfaces with pieces of cotton cloth. The uranium-bearing particles collected in these swipes are measured for their uranium isotopic composition at the analytical labs of the IAEA and Euratom.

In the case of uranium enrichment facilities, the environmental samples often contain very small particles or particle agglomerates of uranium oxyfluoride (UO_2F_2). UO_2F_2 is a compound formed from the hydrolysis of uranium hexafluoride (UF_6), a gas used in large quantities for the enrichment of uranium. UF_6 is very reactive, and even a small release of this material in the presence of water gives rise to UO_2F_2 particulate material and HF. Somewhat surprisingly, environmental samples from enrichment facilities were also found to contain uranium-bearing particles without a measurable amount of fluorine [2]. This seemed to indicate UO_2F_2 loses its fluorine over time, or after exposure to certain environmental conditions. As both nuclear forensics and environmental sample analysis depend upon laboratory analysis of nuclear material that has often been exposed to the environment after it was produced, it is important to understand how those environmental conditions might have changed the chemical composition of the material over time, particularly for these chemically sensitive compounds.

Previous studies have already demonstrated that exposure to high temperature (350 °C), high relative humidity (> 70%) and ultraviolet light effectively accelerates the loss of fluorine in UO_2F_2 [3]. However, the exact conditions under which this happens and the molecular changes that accompany this decomposition are not well understood.

Expressly for the purpose of this study, we prepared two sets of uranium oxyfluoride particles at the Institute for Reference Materials and Measurements (EU-JRC-IRMM) in Belgium (first set was prepared in January 2009, second one in January 2010). Graphite planchets with uranium oxyfluoride particles were prepared from the hydrolysis of UF_6 . After shipment to LLNL, samples were either stored in a controlled environment at a specific temperature and humidity or in an inert atmosphere.

The morphology, chemical composition and relative amount of fluorine for individual particles and small particle agglomerates was determined by a range of complementary micro-analytical techniques including NanoSIMS, micro-Raman spectrometry (MRS), scanning (SEM) and transmission (TEM) electron microscopy with energy-dispersive X-ray spectrometry (EDX) or focused ion beam (FIB) technology.

This report summarizes our most recent findings obtained by NanoSIMS and MRS analysis. It also includes a summary of the 1-week visit of Dr. Elzbieta Stefaniak (IRMM) to LLNL as part of the Action Sheet agreement between EC-JRC-IRMM and DOE/LLNL. Additional spectroscopic and mass spectrometric measurements were carried out at Pacific Northwest National Laboratory, but are not included in this summary. It should also be noted that we have no evidence that fugitive emissions from a uranium enrichment facility would, or would not, appear similar to the analyzed samples.

This work has been performed under the auspices of the U.S. Department of Energy through a joint program between Lawrence Livermore National Laboratory (under contract DE-AC52-07NA27344) and the Pacific Northwest National Laboratory (under contract DE-AC05-76RL01830), and under the auspices of the European Commission, DG Joint Research Centre under Action Sheet 36.

Summary Activities November 2009 - July 2010

1. NanoSIMS analyses to determine the F-to-U ratio in single particles

The Cameca NanoSIMS secondary ion mass spectrometer at LLNL with its nanometer-scale spatial resolution was used to measure the relative amount of fluorine in uranium oxyfluoride particles. A primary beam of O^- ions was used to sputter a raster of typically $6\ \mu\text{m} \times 6\ \mu\text{m}$ centered around the particle (Fig. 3). The positively-charged secondary ions of fluorine, uranium and uranium oxide were collected by varying the magnetic field of the analyzer (peak jumping) (Table 1). The analysis was stopped when the fluorine count rate dropped to background values. The fluorine-to-uranium (F^+/U^+) ratio was calculated from the summed ion counts for fluorine and uranium, integrated over the total duration of the analysis.

EM 1	EM 2	EM 3	EM4	EM 5
	$^{19}\text{F}^+$			
				$^{238}\text{U}^+$
				$^{238}\text{U}^{16}\text{O}^+$

Table 1. Mass table showing the electron multiplier (EM) detectors selected for the collection of the ion species listed.

Samples prepared in 2009

A first batch of samples was prepared in January 2009 and samples were stored in either one of four environmental chambers at different temperature ($25\ ^\circ\text{C}$ or $40\ ^\circ\text{C}$), relative humidity ($<15\ \%$ or 76%) and lighting (diffuse light or dark) [4]. The NanoSIMS analyses on these samples showed a fairly large variation in the F^+/U^+ ratio between samples from the same batch (i.e. same release of UF_6), and even between particles from the same sample. Nevertheless, we came to the following conclusions for the F^+/U^+ ratio based on this first set of measurements:

- Value for fresh samples: $0.09 \pm 0.01 - 0.14 \pm 0.05$
- No significant difference between storage in diffuse light and in the dark (over a period of several months)
- No significant difference between storage in $24\ ^\circ\text{C}$ and $40\ ^\circ\text{C}$ (over a period of several months)
- Decrease by about 2 orders of magnitude after 3 months exposure to $77\ \%$ RH air
- Samples stored in dry air for a period of up to 9 months: F^+/U^+ ratio $\geq 0.05 \pm 0.01$
- Samples exposed to 30% or 45% relative humidity for at least 2 months showed a decrease in the F^+/U^+ ratio relative to the freshly-prepared samples, but given the high sample-to-sample variability, we need to measure the F/U ratio of each sample individually before and after this humidity exposure

Samples prepared in 2010

In January 2010, 127 fresh uranium oxyfluoride particle samples (120 graphite, 7 sapphire substrates) were prepared from the controlled hydrolysis of UF_6 ($^{235}\text{U} = 0.704\%$). All samples were stored in containers filled with argon until they were analyzed.

The 2009 measurements showed that the large variability within and between samples made it difficult to draw conclusions on the sample set as a whole. We therefore fine tuned the experiment and measured a set of particle samples prepared in January 2010 as soon as they were taken out of the argon-filled container, and re-measured the same samples after they had been exposed to certain environmental conditions. This approach eliminated the within-sample-variability, but did not exclude the variability between particles from the same sample (expressed in the standard deviation in *Figures 2* and *5*). NanoSIMS is an inherently destructive analysis technique, and every particle can only be measured once.

Measurements on a subset of the 2010 samples showed that the F^+/U^+ ratio was higher for the 2010 samples (0.21 ± 0.02 up to 0.77 ± 0.14) than for the samples prepared in 2009. The 2010 samples were stored in argon until they were analyzed, while the 2009 were stored in dry air, and this may be the reason why the ratio was higher. Both sample sets however, showed large variations in the F^+/U^+ ratio between samples from the same batch.

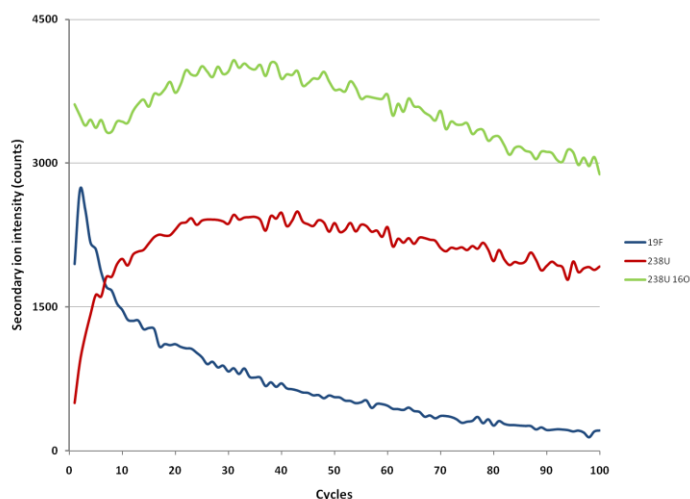


Fig 1: NanoSIMS depth profile showing the fluorine, uranium and uranium oxide secondary ions as a function of sputtering time (cycles) in a UO_2F_2 particle shortly after preparation at the IRMM in January 2010.

The depth profile in *Figure 1* shows the change in intensity for the uranium oxide, uranium and fluorine secondary ions as the particle is being sputtered by the primary ion beam. The particle analyzed was measured 2 months after its preparation at the IRMM in January 2010. Overall, the depth profiles of the freshly-prepared particles from 2010 looked very similar to those from 2009. The fluorine count rate started off at a value which was significantly higher than the uranium count rate, but reached its maximum before sputtering equilibrium was attained, and decreased from thereon. As expected, the uranium oxide count rate was much higher than the uranium count rate at the start of the analysis, but both secondary ion species showed a similar behavior after the first 10 cycles.

The 2009 measurements demonstrated that the exposure to high humidity most strongly reduced the F^+/U^+ ratio. We therefore exposed a subset of freshly-prepared particle samples to 70 % relative humidity for 18 and 117 hours respectively. Comparison before and after the 70 % relative humidity exposure showed that the F^+/U^+ ratio decreased from 0.65 to 0.26 (decrease by 60 %) and from 0.77 to 0.052 (decrease by 94 %) for the 18 and 117 hours experiment respectively. It should also be noted that the value for the F^+/U^+ ratio in the latter case was still at least a factor 6 higher than for the samples exposed to several months of high relative humidity (*Fig. 2*).

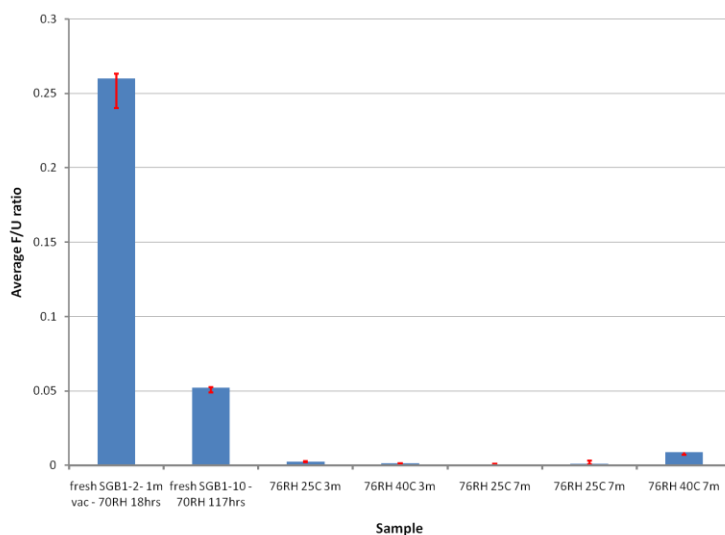


Fig 2: F^+/U^+ ratio for samples after exposure to high humidity (>70 %) for 18 hours (far left) to 7 months (far right). After 3 months or longer the fluorine count rates for these samples were so low that a correlation between the F^+/U^+ ratio and the exposure time became less evident.

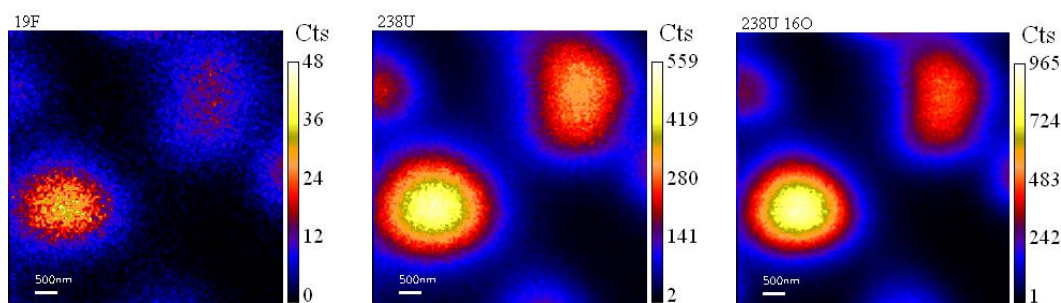


Fig 3: 6 μm x 6 μm ion images (F^+ , U^+ , UO^+) of UO_2F_2 particles exposed to 117 hours of 70 % relative humidity.

Particles that were heated to 200 °C for 19 hours still contained significant amounts of fluorine ($F^+/U^+ = 0.11 \pm 0.05$), while heating to 350 °C removed most of the fluorine. This was later confirmed by micro-Raman spectroscopy (*Section 4*), where a uranyl peak was detected around 863 cm^{-1} (characteristic of UO_2F_2) for the particles heated to 200 °C, while the spectrum of the sample that was heat-treated at 350 °C resembled that of U_3O_8 .

Using a focused ion beam (FIB) on a specialized scanning electron microscope, we made multiple cross-sections of a single particle (coated with a Pt layer for easy manipulation) to determine if the inner structure of a particle that was heated to 350 °C was visibly different from those not exposed to high temperature. Comparison of the cross sections of the heat-treated and freshly-prepared particle did not show any significant differences in terms of the presence of small voids (*Fig. 4*).

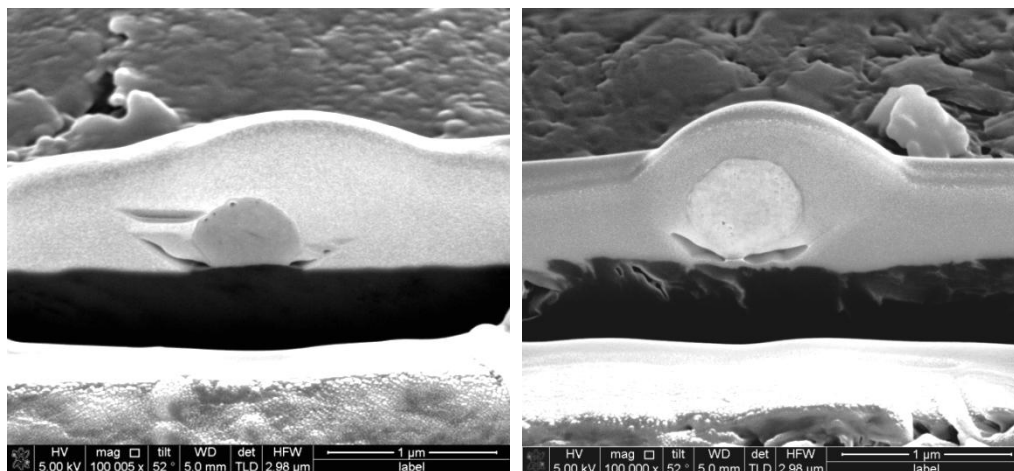


Fig 4: FIB cross-sections of a Pt-coated particle that was freshly-prepared (left) and one that was heated to 350 °C (right).

As a summary, the plot in *Figure 5* shows the average F^+/U^+ ratio (4-7 particles/sample) for all samples analyzed.

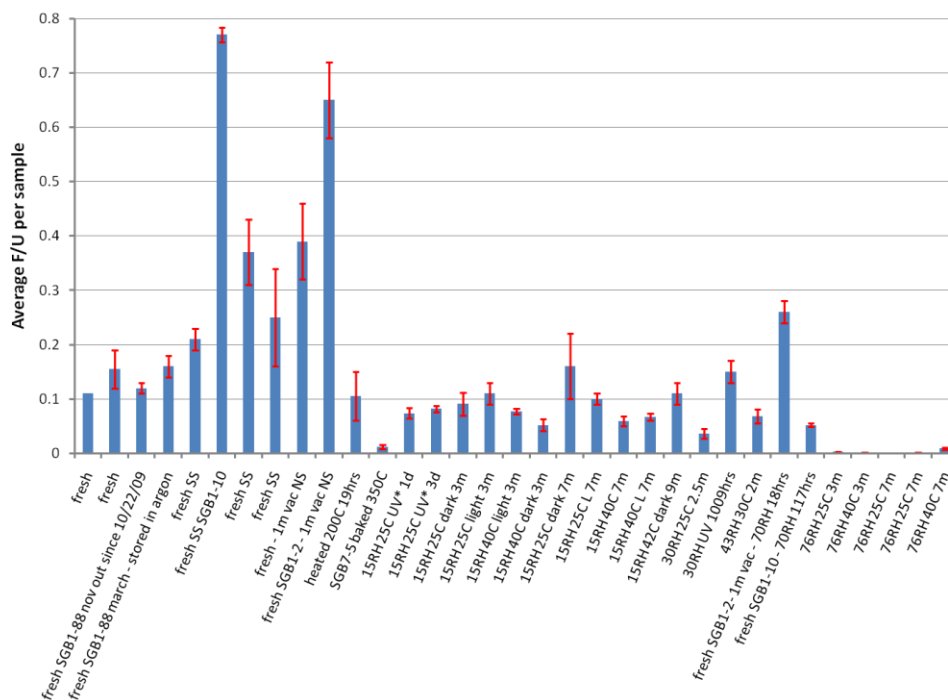


Fig 5: Overview of the F^+/U^+ ratios of all uranium oxyfluoride particle samples measured by NanoSIMS.

2. Improvements on sample preparation

The ion images in *Figure 3* show several areas of high uranium and fluorine counts. Even though the F^+/U^+ ratio could be calculated for these areas, the sample preparation can be improved on the following points:

1. For the majority of the particle samples analyzed, the number of particles per area was quite high. As can be seen from *Figure 3*, a high particle load makes it difficult to analyze individual particles. This was also part of the feedback IRMM received after its first particle measurement evaluation program (NUSIMEP-6) [5]. In order for environmental sampling (using SIMS) to be effective, the particle distribution should probably not exceed 1-2 particles per $25 \mu\text{m}^2$.
2. The ideal particle sample has a very narrow particle size distribution, with little or no agglomeration.
 - By looking at the ion images, small particle agglomerates are difficult to distinguish from larger single particles. The difference in morphology however, may complicate the interpretation of the depth profiles and the F^+/U^+ ratio
 - The more uniform the particle morphology, the smaller the ion beam sputtering effects on the ion count rates, and on the F^+/U^+ ratio
 - Larger (single) particles are preferred for SIMS analysis (higher count rates), but in practice particles produced from UF_6 hydrolysis typically do not exceed $1 \mu\text{m}$

To overcome the issue of high particle load, a Kleindiek MM3A-EM micro-manipulator was installed at LLNL. This is a tool that allows for nanometer-scale, remote-controlled movements of a tungsten tip inside an electron microscope. With the micro-manipulator we can pick up individual particles through electrostatic attraction and transfer them to a specific location on a blank planchet. Uranium oxyfluoride particles were successfully transferred from their original planchet to a blank planchet (*Fig. 6*) and NanoSIMS analysis on these transferred particles will follow in the next weeks.

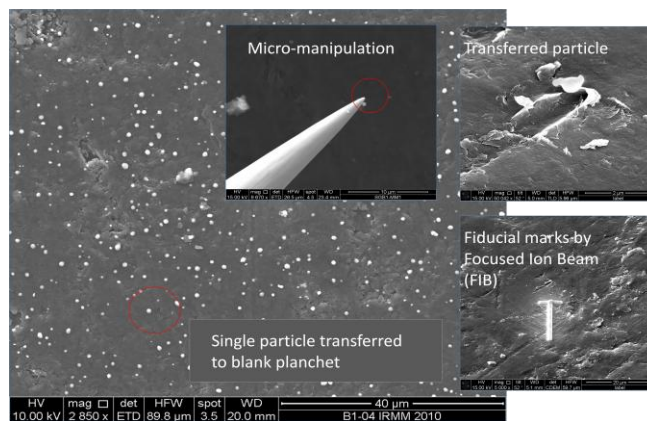


Fig 6: Particle transfer using a micro-manipulator and FIB markings.

3. Visit of Dr. E. Stefaniak to LLNL - April 12-16, 2010 (DOE-JRC Action Sheet 36)

Pursuant to the Arrangement between European Commission DG Joint Research Centre (EC-JRC) and the Department of Energy (DOE) to continue cooperation on research, development, testing, and evaluation of technology, equipment, and procedures in order to improve nuclear material control, accountancy, verification, physical protection, and advanced containment and surveillance technologies for international safeguards, dated 1 September 2008, the IRMM and LLNL have established cooperation in a program on the Study of Chemical Changes in Uranium Oxyfluoride Particles. This technical collaboration is performed under the so-called IRMM-LLNL Action Sheet 36.

Exchange visits of LLNL and IRMM scientists on the preparation and analysis of uranium oxyfluoride particles are part of the Action Sheet 36. It is within this context that Dr. Elzbieta Stefaniak (IRMM) visited LLNL for a period of 5 days. During Dr. Stefaniak's visit to the lab we demonstrated the analysis of uranium oxyfluoride particles produced at the IRMM using both the NanoSIMS and the SIMS 3f, hereby focusing on two specific programmatic responsibilities stated in the Action Sheet:

- LLNL will analyze the uranium oxyfluoride particles at defined time intervals to study the effects of time, humidity, temperature and lighting on the rate of fluorine loss
- LLNL will investigate the particle morphology, the density of particles on the graphite planchet and the presence or absence of a uranium film covering the planchet surface

The latter was verified by means of a Resistive Anode Encoder (RAE) on the SIMS 3f, which allows for the imaging of a 150 μm diameter area on the planchet's surface. Images of the uranium secondary ions recorded with the RAE detector after 5 and 900 seconds of sputtering are shown in *Figure 7*. The number of particles was very high for this area, which made it difficult to resolve individual particles. The particles appeared somewhat more clearly after 900 seconds of sputtering, but it remained inconclusive whether there was a film of uranium covering the surface.

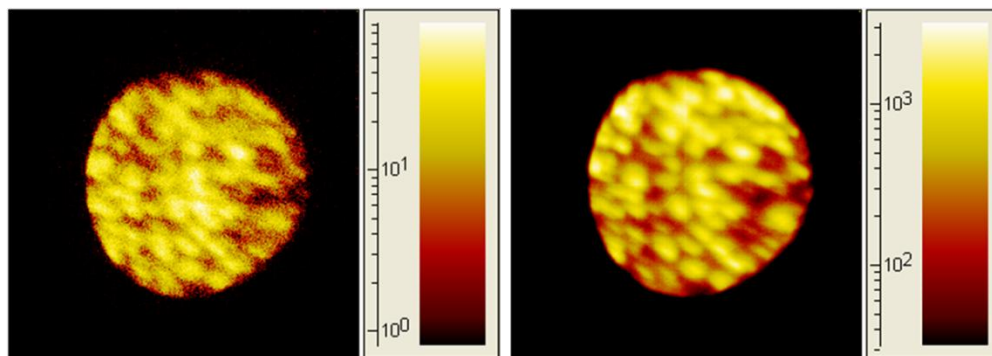


Fig 7: 150 μm RAE images of $^{238}\text{U}^+$ secondary ions collected after 5 seconds (left) and 900 seconds (right) of sputtering.

The brighter the color in the images, the more secondary ions were collected at that position.

These images show that the number of particles per surface area was very high for this sample, and that most of the uranium particles were still present after 900 seconds of sputtering.

One of the ongoing nuclear safeguards projects at the IRMM with respect to environmental sampling is the production of new certified test samples for NUSIMEP inter-laboratory comparisons. The last NUSIMEP campaign, NUSIMEP-6, was concluded in 2008 and involved the isotopic analysis of uranium oxyfluoride particles using secondary ion mass spectrometry [5]. The feedback provided by the participating labs (including LLNL) allowed the IRMM to further improve the sample preparation process. For the next NUSIMEP campaign, the IRMM plans to use uranium oxyfluoride particle samples with two different enrichment levels. In this context, a stainless steel sample containing uranium oxyfluoride particles with an enrichment of 2 % and 25 % ^{235}U respectively was sent to LLNL for initial testing. SEM imaging showed a fairly low particle density (# particles/area) for this sample (*Fig. 8*). Nine particles were located and measured for their uranium isotopic composition using NanoSIMS.

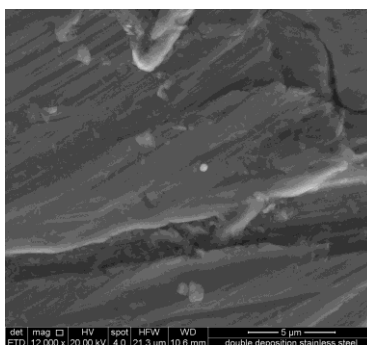


Fig 8: SEM image of a uranium oxyfluoride particle (white dot) on a stainless steel substrate.

The plot shown in *Figure 9* summarizing the $^{235}\text{U}/^{238}\text{U}$ ratio vs. $^{235}\text{UO}/^{238}\text{UO}$ ratio for the 9 particles analyzed indicated that no significant mixing occurred between the two different enrichment on this stainless steel substrate. Additional measurements on (graphite) planchets with a higher particle load will have to confirm this conclusion.

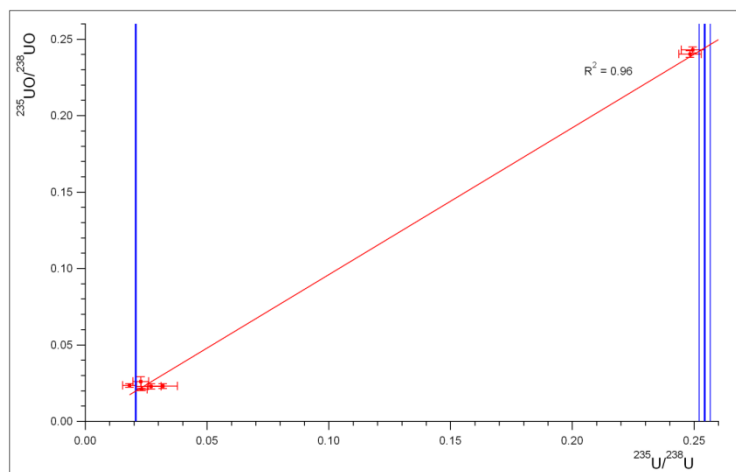


Fig 9: $^{235}\text{U}/^{238}\text{U}$ ratio plotted vs. $^{235}\text{UO}/^{238}\text{UO}$ ratio for 9 different particles showing 2 distinct uranium enrichment levels. Vertical lines indicate the certified isotopic composition of the two UF_6 materials used for this mixed deposition.

In addition to this experimental work, Dr. Stefaniak gave a presentation on the work performed at the IRMM and her project on ‘The Development of Uranium Reference Particles’ in particular. She also received a general lab tour.

4. Micro-Raman Spectroscopy for molecular fingerprinting

The initial Raman experiments described in our previous progress report [4] indicated that uranium oxyfluoride particle samples prepared under different conditions could be distinguished based on their Raman spectra. The experiments also showed that the power applied to the sample should be kept below a certain threshold in order to avoid the conversion of UO_2F_2 to U_3O_8 .

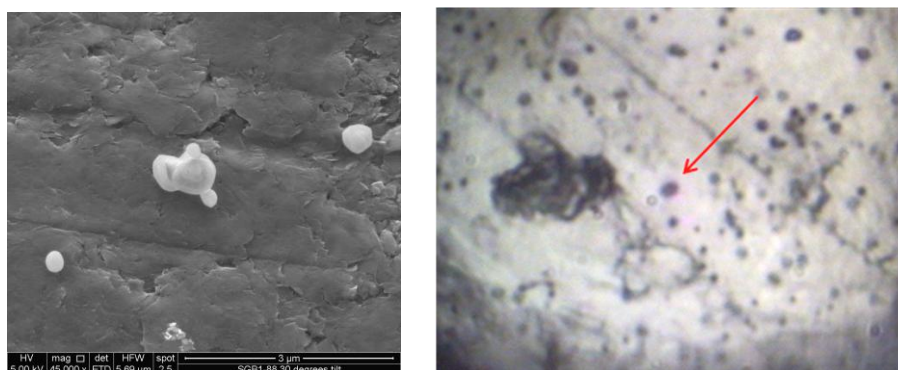


Fig 10: SEM image (left) vs. optical microscope image on the Raman instrument (right) (different particles).

In 2010, we expanded the sample set for the Raman measurements. Samples exposed to specific environmental conditions were measured at LLNL (Figure 11 shows a schematic of the system). All measurements were carried out with laser light of wavelength 632.8 nm (HeNe). The small size of the particles required a small focused spot size to provide adequate spatial resolution for the Raman measurements (*Fig. 10*). Laser light was focused onto the sample with a 100× objective lens estimated to produce a focal spot of approximately 1 μm in diameter (*Fig. 11*). The incident power was minimized to not more than 2.1 mW.

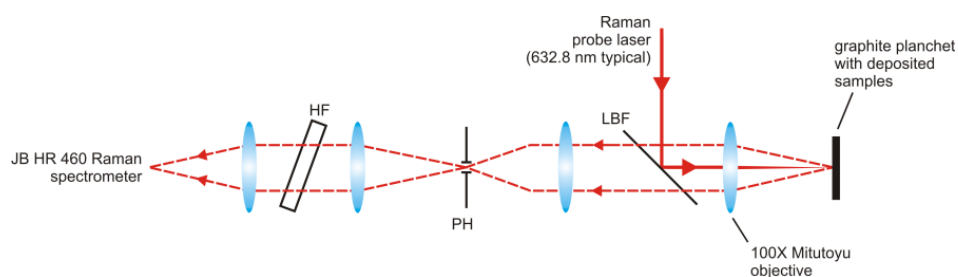


Fig 11: Schematic of the Raman system at LLNL.

The Raman spectrum from a sample stored in argon since its preparation at the IRMM was used as a baseline for comparison with samples exposed to specific environmental conditions. Storage in an inert atmosphere is assumed to preserve the particles' chemical and molecular structure. The spectrum of this 'non-exposed' baseline sample was in general agreement with the spectrum of hydrated UO_2F_2 described in literature [6]: the uranyl (UO_2)²⁺ symmetric stretching frequency estimated to be around 863 cm^{-1} was the most prominent peak in the spectrum, with a shoulder towards the lower frequency side (*Fig. 12*).

At lower Raman shifts (not shown), a peak around 180 cm^{-1} was detected, attributed to the U-O bend, and other broad and fairly weak features appeared in the $100\text{-}300\text{ cm}^{-1}$ and $500\text{-}600\text{ cm}^{-1}$ range.

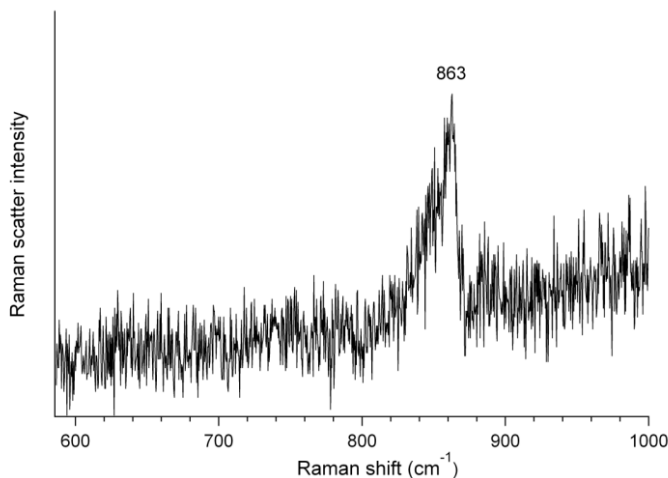


Fig 12: Raman spectrum of a ‘baseline’ sample, stored in argon since its preparation at the IRMM.

The spectrum of this baseline sample was then compared to samples that were either exposed to high humidity, ultraviolet light, high temperatures or ambient air for longer periods of time. Previous studies indicated that humidity may be the main factor accelerating the loss of fluorine in uranium oxyfluoride particles formed from UF_6 hydrolysis. We therefore exposed different particle samples to air at 30 % and 70 % relative humidity for 3 months and 5 days respectively, and measured their Raman spectrum.

The spectrum of a sample exposed for 3 months to air at room temperature and 30 % relative humidity showed the uranyl peak at the same position as in the baseline spectrum, i.e. around 863 cm^{-1} . In addition however, a less intense but distinct band appeared around 843 cm^{-1} (*Fig. 13*). The close proximity of both peaks may imply that the band at 843 cm^{-1} was also produced by the $(\text{UO}_2)^{2+}$ stretching frequency, but in a different hydration state.

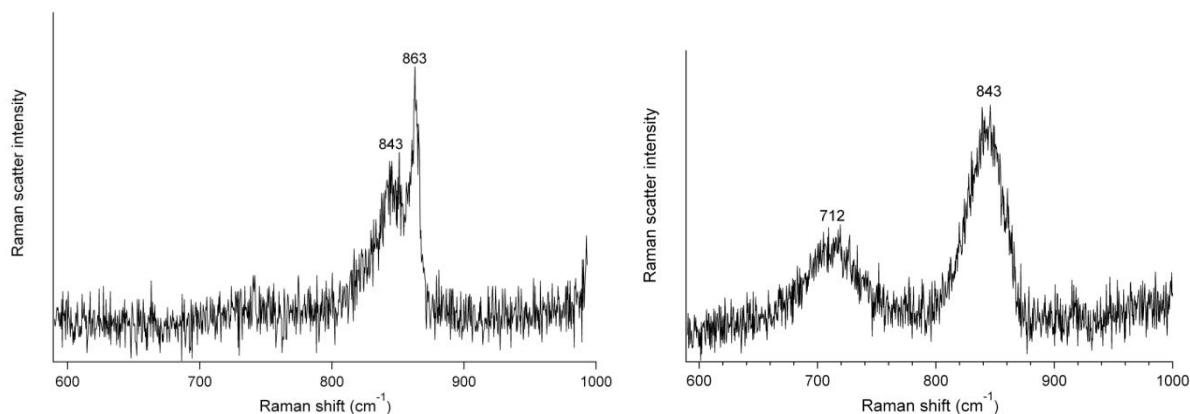


Fig 13: Raman spectrum of a sample stored in 30 % rel. humidity for 3 months (left) and 70 % rel. humidity for 5 days (right). Exposure to high humidity generally resulted in peak broadening and shifts towards lower frequencies.

After an exposure to 70 % relative humidity for 5 days (117 hours), the initial uranyl peak around 863 cm^{-1} could no longer be resolved. The band around 843 cm^{-1} had become the most distinct feature in the spectrum (*Fig. 13*). These measurements suggest that the uranyl peak which was detected around 863 cm^{-1} in the baseline spectrum, broadened and shifted towards lower frequencies due to the absorption of water.

Somewhat surprisingly, the spectrum of a sample stored in ambient air (in a fume hood) at moderate humidity for over 5 years still showed a distinct uranyl peak at 867 cm^{-1} (*Fig. 14*). The most intense peak in its spectrum however, was detected around 830 cm^{-1} . The origin of the latter Raman shift has not been identified.

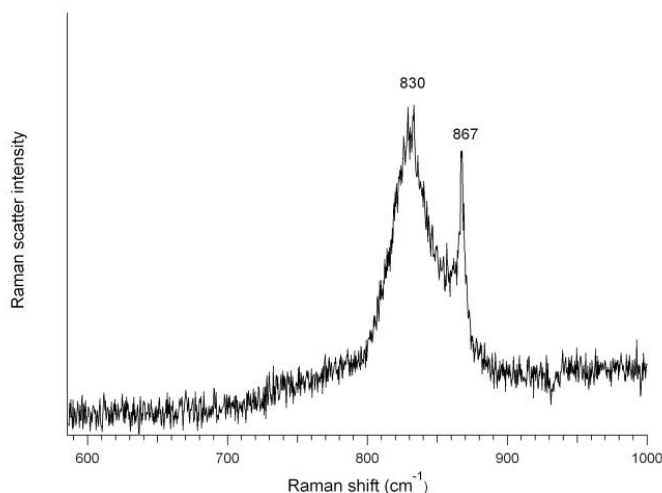


Fig 14: The Raman spectrum of a sample stored in ambient air for more than 5 years still showed a uranyl peak around 867 cm^{-1} .

A summary of the different exposure conditions and their respective Raman bands is given in Table 2.

Exposure	Duration	Raman bands (cm^{-1})
argon atmosphere	2 months	863(s)
30 % rel. humidity	3 months	843(m) + 863(s)
70 % rel. humidity	5 days	712(m) + 843(s)
200 °C open air oven	19 hours	863(s)
UV light	42 days	715(m) + 863(s)
ambient air	5 years	830(s) + 865(m)

Table 2 : Overview of the Raman bands detected for different exposure conditions. The intensity of each of the bands is indicated as strong (s), medium (m) or weak (w).

These measurements demonstrated that even though the decomposition of uranium oxyfluoride happens very slowly, micro-Raman spectroscopy is able to distinguish subtle differences in uranium oxyfluoride particles depending on the conditions to which they were exposed. This so-called ‘molecular fingerprinting’ of single particles and small particle agglomerates is considered to be a valuable complement to the information obtained from more quantitative techniques such as secondary ion mass spectrometry.

Conclusions & Outlook

The experiments carried out between November 2009 and July 2010 confirmed that a combination of complementary analytical techniques is the best approach to characterize uranium particles produced from UF_6 hydrolysis.

Using high spatial resolution secondary ion mass spectrometry (**NanoSIMS**) we calculated the average F^+/U^+ ratio (4-7 particles per sample) for freshly-prepared samples and samples exposed to specific environmental conditions. The NanoSIMS measurements in 2009 demonstrated that there was a fairly large variability between samples and even between particles in a sample. The F^+/U^+ ratio was significantly reduced however, after a 3 month exposure to high relative humidity (> 70 %). For particle samples stored in dry air for up to 9 months, the F^+/U^+ ratio did not decrease below 0.05 ± 0.01 .

In 2010, we prepared a new set of samples in collaboration with the IRMM (Action Sheet 36). The samples were stored in an argon atmosphere until they were analyzed by NanoSIMS. The F^+/U^+ ratio of a subset of the 2010 samples ranged from 0.21 ± 0.02 to 0.77 ± 0.14 , which was higher than what was measured for the 2009 samples.

To quantify the decrease in the relative amount of fluorine, we measured the samples before and after exposure to specific environmental conditions. For a sample that was exposed to 70 % relative humidity for just 5 days, the F^+/U^+ ratio decreased by 94 %. Heating the particles in an open air oven at 200 °C, resulted in a F^+/U^+ ratio of 0.11 ± 0.05 . At 350 °C however, the F^+ count rate was reduced to background. These results were consistent with the **micro-Raman** measurements using a 632.8 nm HeNe laser focused down to a 1 μm spot size: the spectrum of the sample heated to 200 °C showed a clear UO_2^{2+} peak around 863 cm^{-1} , characteristic of UO_2F_2 , while the spectrum of the 350 °C sample resembled that of U_3O_8 . Their particle morphology and inner structure, as verified by **SEM-FIB**, was not visibly different however.

Samples exposed to different levels could also be distinguished based on their Raman spectra: at 30 % relative humidity, an additional peak appeared in the spectrum around 843 cm^{-1} . This peak became very broad and intense for particles that were exposed to 70 % relative humidity for 5 days, while the peak at 863 cm^{-1} disappeared from the spectrum.

Peak broadening and shifts towards lower frequencies were found to be characteristic of samples exposed to high relative humidity.

Finally, a sample that was stored in a fume hood at the IRMM for over 5 years was examined, and its spectrum did not resemble any of the samples we have measured so far.

These measurements demonstrated that even though the decomposition of uranium oxyfluoride happens very slowly, micro-Raman spectroscopy can distinguish subtle differences in uranium oxyfluoride particles depending on the humidity and temperature conditions to which they were exposed. This so-called ‘molecular fingerprinting’ of single particles and small particle agglomerates is considered to be a valuable complement to the information obtained from more quantitative techniques such as secondary ion mass spectrometry.

For the next months, we plan to continue this study on the chemical changes in uranium oxyfluoride particles in close collaboration with the IRMM and PNNL. The samples prepared in 2010 will (continue to) be exposed to various levels of temperature, humidity and ultraviolet light. Through the use of a micromanipulator and focused ion beam we hope to reduce the number of particles per planchet, facilitate particle relocation and homogenize the particle morphology for micro-Raman and NanoSIMS analyses. A vacuum impactor for particle transfer has been installed and will be tested in the coming weeks.

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Attended conferences, publications and research visits (Dec 09 - July 10)

Publications

Kips, R., J. Crowhurst, M. J. Kristo, I. D. Hutcheon, E. Stefaniak, “*Micro-Raman Spectroscopy of Uranyl Difluoride*,” *Journal of Nuclear Materials*, (2010), in preparation

Kips, R., J. Crowhurst, M. J. Kristo, I. D. Hutcheon, E. Stefaniak, “*Micro-Raman Spectroscopy of Uranium Oxyfluoride Particulate Material for Nuclear Safeguards*,” *Proceedings of the 51th Annual Meeting of the Institute for Nuclear Materials Management*

Kips, R., M. J. Kristo, I. D. Hutcheon, K. B. Olsen, J. Amonette, Z. Wang, D. Gerlach, T. Johnson, E. Stefaniak, “*Measuring Fluorine in Uranium Oxyfluoride Particles using Secondary Ion Mass Spectrometry for Nuclear Forensics*,” *Radiochimica Acta*, (2010), in publication, *Proceedings of the 4th International Conference on the Asia-Pacific Symposium on Radiochemistry*

Attended conferences

51th Annual Meeting of the Institute for Nuclear Materials Management (INMM)
July 11-15, 2010, Baltimore, MD

4th International Conference on the Asia-Pacific Symposium on Radiochemistry (APSORC)
November 29 - December 4, 2009, Napa, CA

Research Visits

Visit E. Stefaniak (IRMM) to Lawrence Livermore National Laboratory - April 12-16, 2010
Summary of visit included in Section 3 (pp. 8-9)

Visit R. Kips to Pacific Northwest National Laboratory - December 15-17, 2009
Tour of the CLIFS, Raman, XRD and SIMS facilities
Presentation on status of work
Technical Discussions

Planned conferences

IAEA Symposium on International Safeguards: Preparing for Future Verification Challenges
November 1-5, 2010, Vienna, Austria
Abstract accepted for oral presentation: “*Investigating Chemical and Molecular Changes in Uranium Oxyfluoride Particles using NanoSIMS and Micro-Raman Spectroscopy*”

International Chemical Congress of Pacific Basin Societies (PACIFICHEM)
December 15-20, 2010, Honolulu, HI
Abstract accepted for oral presentation: “*Combined NanoSIMS and micro-Raman characterization of uranium oxyfluoride particles for nuclear safeguards*”