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Direct, Uncorrected, Molecule-Free Analysis of ^{236}U from Uranium-Bearing Particles with NAUTILUS: A New Kind of Mass Spectrometer

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Abstract

We demonstrate use of the Naval Ultra-Trace Isotope Laboratory's Universal Spectrometer (NAUTILUS) at the U.S. Naval Research Laboratory (NRL) to measure ^{236}U directly from uranium-bearing particles free from molecular isobaric interferences. Particles with ^{235}U enrichments in the range of 0.32% to 3.28% and ^{236}U enrichments from no enrichment to 0.015% provided by the International Atomic Energy Agency (IAEA) were analyzed directly using the NAUTILUS. We report the experimental data here without correcting for molecular hydrides and/or applying any other background subtractions. The results from all samples agreed with the certified values within standard error save for the ^{236}U composition of the IRMM 023, which suffered from a combination of insufficient particle sizes and sub-µmol/mol ^{236}U concentrations. We were able, however, to directly measure as low as three µmol/mol of ^{236}U in individual particles regardless of the ^{235}U concentration. Our results are comparable with Large Geometry Secondary Ion Mass Spectrometry (LG-SIMS) and serve as baseline for a more comprehensive comparison between LG-SIMS and the NAUTILUS in the future. Moreover, we demonstrate the ability of the NAUTILUS to generate raster ion images with the same ease as traditional LG-SIMS instruments. By combining our ability to measure ^{236}U directly with raster ion imaging, we were able to detect a low intensity, small uranium-bearing particle in the presence of high molecular backgrounds for a non-ideal sample. This discovery could lead to more targeted and, therefore, less time intensive particle screening methodologies.

Keywords: NAUTILUS; nuclear Safeguards; Secondary Ion Mass Spectrometry (SIMS); Single-Stage Accelerator Mass Spectrometry (SSAMS); hydride correction; ion imaging

Introduction

Uranium-bearing particle analysis for international Safeguards traditionally employs a two-pronged approach. The first being Fission Track – Thermal Ionization Mass Spectrometry (FT-TIMS),[1] which excels in its ability to measure high precision uranium isotope ratios free from molecular isobaric interferences, but which suffers from extensive sample preparation requirements and relatively low throughput. The other being Large Geometry – Secondary Ion Mass Spectrometry (LG-SIMS),[2] which excels in its ability to measure uranium isotope ratios with much higher throughput and much less stringent sample preparation requirements than FT-TIMS, but which suffers from a number of molecular isobaric interferences that conspire to limits its overall precision and accuracy. The work presented herein is primarily concerned with improving high throughput uranium-bearing particle analysis (i.e. >5,000 particles per sample) and, therefore will draw a comparison only to standard LG-SIMS analysis. However, it is

important to note that FT-TIMS is a viable alternative to LG-SIMS when high precision is of greater value than high throughput.

The application of LG-SIMS to the international Safeguards community requires an ability to effectively screen for and detect uranium-bearing particles collected from Safeguards inspections.[3-10] A typical application of SIMS for nuclear Safeguards is to collect dust samples taken using cotton swipes at enrichment facilities and analyze them to ensure that the level of uranium enrichment remains within declared values. SIMS offers timely analyses of a statistically significant number of uranium-bearing particles, the ability to distinguish between uranium-bearing particles enriched or depleted in ^{235}U, and high selectivity for particles of interest within a vast background of environmental particles containing only natural uranium. Despite advancements in automation of the SIMS screening process, long analysis times are common and a single 1-inch round SIMS planchette can take upwards of 8-10 hours to fully screen. Factors that contribute to these long analysis times include the fact that uranium-bearing particles are measured one field-of-view (FoV) at a time and that there may be hundreds to thousands of FoVs per planchette, and that ^{234}U and ^{236}U are typically present in µmol/mol concentrations and require long counting times. In addition, particles of interest identified in the screening process require re-analysis in microprobe mode, which involves a set of different instrument analytical parameters. [11-13]

The most pressing challenge for SIMS, however, is that the $^{235}U^{1}H$ molecule complicates the measurement of ^{236}U. The presence of ^{236}U is characteristic of uranium reprocessed from spent nuclear fuel and is, therefore, of interest with respect to environmental sampling.[14, 15] High-resolution mass spectrometry techniques traditionally approach the challenge of molecular isobaric interferences by striving to increase the mass resolving power of the analysis. For example, $^{208}Pb^{28}Si$, $^{204}Pb^{16}O_2$, $^{235}U^{1}H$ and ^{236}U all of the same nominal mass at 236, but have different exact masses at 235.953, 235.963, 236.026 and 236.052 respectively. For instance, at a resolving power of 500, these masses are all overlapped and cannot be distinguished from one another. Increasing the resolving power to 3000 separates out $^{208}Pb^{28}Si$ and $^{204}Pb^{16}O_2$ leaving $^{235}U^{1}H$ and ^{236}U overlapping. It is much more difficult to separate out the hydride molecule from its adjacent isotope. In fact, it would take a resolving power of greater than 38,000 to distinguish between $^{235}U^{1}H$ and ^{236}U. Unfortunately, as the resolving power increases, the ion transmission decreases. Even under ideal conditions, the ion transmission of a CAMECA ims-1280 is essentially zero at the mass resolution required to separate $^{235}U^{1}H$ from ^{236}U. [16]

The NAUTILUS at NRL represents an alternative methodology for removing molecular isobaric interferences.[17] The NAUTILUS is a combination of a small geometry SIMS analyzer and a Single Stage Accelerator Mass Spectrometry (SSAMS) detector. It works in much the same way as a traditional SIMS instrument. Primary ions sputter secondary ions into the mass spectrometer where they are energy and mass filtered. However, in the place where detection occurs by an electron multiplier (EM) or faraday cup (FC), the secondary ions pass into the SSAMS. The secondary ions exiting the SIMS accelerate by 300 keV and then enter the gas stripper tube. Here molecular dissociation with minimal scattering loss occurs as the higher energy secondary ions interact with gas (i.e. Ar) in the stripper tube. The amount of molecular dissociation is

exponentially proportional to gas flow in the stripper tube and is much higher than ion transmission loss though the rest of the SSAMS instrument. Previous work has shown that a 10^5 reduction in molecular ion signal is achievable with only 0.2 sccm (7.03×10^{15} atoms/cm^2) of Ar gas flow in the stripper tube, while only reducing transmission of atomic species by approximately 60 percent. [18, 19] In this way, the NAUTILUS nearly eliminates the contribution of $^{235}U^{1}H$ to the mass-to-charge (m/z) 236 mass channel and results in molecule-free analysis of uranium-bearing particles at a resolving power of only 500.

Although microprobe mode, which is a non-imaging mode, is usually required to determine the uranium isotopes present in any given particle, raster ion imaging is critical to perform the initial automated screening of the particle population.[4, 6, 7, 20] Therefore, it is important to implement raster ion imaging on the NAUTILUS this is comparable, at minimum, to what is available by traditional SIMS instrumentation. Because the front end of the NAUTILUS is a SIMS instrument, the mechanics of generating a raster ion image are identical. The primary ion beam follows a software determined raster pattern by applying voltage to the primary ion raster plates. Voltages applied to the secondary dynamic transfer plates compensate for variations in the location of the primary ions during the raster in order to maintain a consistent extraction of the secondary ions into the mass spectrometer. At this point, detection of the secondary ions either occurs at the EM at the end of the SIMS instrument or at the EM at the end of the SSAMS instrument. Synchronization of the primary ion raster pattern and the pulse train on the detection EM allows for the reconstruction of the raster ion image as ion intensity as a function of x and y image pixel position. Not only can we image at two separate locations (SIMS-EM and SSAMS-EM), we can electrostatically switch between the two, effectively producing On/Off raster ion images where the SIMS image is characteristic of a convolution of atoms and molecules and the SSAMS image is a truly molecule-free isotope image.

Experimental

The NAUTILUS combines a modified CAMECA ims-4f SIMS and a ±300 keV National Electrostatics Corporation (NEC) SSAMS with the ability to analyze singly charged ions up to mass 300. Unlike traditional tandem AMS relegated to measuring only negative ions, the SSAMS can accept either positive or negative ions from the SIMS. This is beneficial to SIMS as the majority of secondary ions observed for actinides are single-charge, positive ions. A complete description of the NAUTILUS has been previous reported.[18, 19, 21] Briefly, the secondary ions leave the sample at 4.5 keV of energy and are subsequently filtered by their energy and mass in the SIMS and then electrostatically deflected onto one of a suite of detectors including an EM, a FC, or a microchannel plate (MCP) beam imager. Alternatively, the ions deflect into the SSAMS after leaving the SIMS instrument. Here the secondary ions accelerate by 300 kV via 44 electrodes on a resistor chain from a high-voltage bias on the deck to Earth/SIMS ground. The accelerated secondary ions then focus into the center of a gas-stripping cell consisting of a number of concentric, differentially pumped cylinders, filled with Ar gas. The NAUTILUS then filters atomic ions by their mass and energy and analyzes them at the end of the SSAMS by either an EM or a MCP beam imager.

The International Atomic Energy Agency (IAEA) provided NRL with a number of uranium-bearing particle samples for evaluation and comparison to their CAMECA ims-1280 SIMS instrument. The samples originate from various chemical forms ranging from uranium oxide (U_3O_8), uranium hexafluoride (UF_6) and a uranium nitrate solution. It should be mentioned, however, that the UF_6 (IRMM CRM-023) standard was dissolved in a nitrate solution and then converted into uranium oxides in the process of producing particles. Furthermore, the uranium nitrate solution (IRMM CRM-183) standard underwent a similar conversion to uranium oxide particles prior to analysis. The U_3O_8 particle preparation consisted of a simple vacuum impaction of the solid particles onto a 1-inch diameter vitreous carbon planchette. The nitrate solutions were converted into monodispersed particles at the Forschungszentrum Juelich (Germany) using a vibration orifice aerosol generator (VOAG) passing through a drying column. The particle preparation then proceeded by vacuum impaction in a similar fashion as the other uranium-bearing samples. Additionally, the IAEA shared glass particle samples provided to them by The National Institute for Standards and Technology (NIST). The NIST sample preparation method involved directly depositing the glass powder with a swab by gently sprinkling the target surface. The U_3O_8 content in the glass is 10 wt. % with a remaining composition of 20 wt. % lithium dioxide (LiO_2), 16.5 wt. % boric oxide (B_2O_3), 3.5 wt. % aluminum oxide (Al_2O_3), 49.0 wt. % silicon dioxide (SiO_2) and 1 wt. % XO (where X is either barium, calcium or magnesium depending on the specific sample number). The isotopic composition of the NIST glass samples is known to be "near natural" with the ^{236}U composition of NIST-4 and NIST-5 known to be 5.6 ppm and 2.8 ppm respectively. The particles range in ^{235}U enrichment from 0.32% to 3.28% and in ^{236}U enrichment from no enrichment to 0.015%. Table-1 contains the full list of particle standards acquired from the IAEA. We estimate that the particles ranged in size from 0.8 – 5-µm in diameter, but have not confirmed this using Scanning Electron Microscopy (SEM). If this size range is correct, we expect ~$6x10^7$ to $4x10^8$ secondary uranium ions per particle assuming a 1% uranium ionization efficiency.[5] In practice, these numbers will be even lower due to isotopic variations and duty cycle considerations; however, it was common for us to observed count rates >10,000 counts per second for ^{238}U from an individual particle. NRL received the prepared planchettes shipped directly from the IAEA and analyzed them on the NAUTILUS without further manipulation.

NRL analyzed all particles using an identical procedure. We used NIST CRM 850 to calibrate the uranium isotope masses at ^{234}U, ^{235}U, ^{236}U, ^{238}U and $^{238}U^1H$ for both the SIMS and SSAMS sections of NAUTILUS. We used the optical microscope on the SIMS to locate individual particles for analysis coarsely and used the real-time MCP beam image for fine adjustment of the particle location. Analyses of individual particles used a 10 x 10-µm raster of a 1-nA, 10 keV O^- primary ion beam. Save for the use of O^- in place of O_2^+, this procedure is identical to that used by the SIMS analysis team at the IAEA. The analysis of particles continued until we counted a statistically relevant number of ions for each isotope or consumed the particle completely.

Additional SIMS parameters used for these measurements are as follows. On the primary side we used mass aperture one (750 µm) and primary aperture two (200 µm). On the secondary side we used field aperture one (1800 µm), contrast aperture one (400 µm), energy slit of ~100

eV, entrance slit of ~350 µm, exit slit of ~700 µm for a 2:1 ratio with the entrance slit and an imaged field of 75 µm. Since we are using dynamic transfer, no clipping of the secondary ion beam occurs even for the 250-µm raster for imaging, since the 75 µm-imaged field is larger than the primary beam size. The mass resolving power with these conditions is ~500. Without aberrations, the transmittance would be ~87.5%.

In order to demonstrate the utility of raster ion imaging on the NAUTILUS, we produced a particle dispersion on a single vitreous carbon planchette consisting of NIST CRM U500 particles along with crushed monazite particles. The monazite particles represent a source of natural uranium with no ^{236}U, but with a variety of other elements (i.e. rare earth elements and actinides). Previous analysis of this monazite (Smithsonian National Museum of Natural History, NMNH #R14013) by SIMS reveals a peak at nearly every mass making it challenging to measure uranium isotopes.[19] Production of the single planchette started with U500 particles drop dried from a liquid suspension and followed by a similar suspension containing crushed monazite powder in place of the U500 particles. Raster ion imaging of these overlapping particle populations occurred by using a 1-nA, 10 keV O^- primary ion beam, similar to the parameters for single particle analysis. Collected images used a raster pattern of 256 x 256 pixels over an area of 250 x 250 µm. We selected the isotopes ^{232}Th, ^{235}U, ^{236}U and ^{238}U for comparison between SIMS and NAUTILUS raster ion imaging. Interleaving these images electrostatically allowed for direct comparison within each run reducing the risk of biasing the image analysis toward any single outcome.

NIST certified values for U030a and U010

CRM	^{235}U/^{238}U	^{234}U %	^{235}U %	^{236}U %	^{238}U %
U010	0.010140	0.00541	1.0037	0.00681	98.984
U030a	0.031367	0.02778	3.0404	0.000599	96.9312

IRMM certified values for 023 and 183

IRMM	^{235}U/^{238}U	^{234}U %	^{235}U %	^{236}U %	^{238}U %
023	0.0338881	0.032827	3.2760	0.00001115	96.6911
183	0.0032157	0.0019688	0.32049	0.0147858	99.66276

Other uranium containing particles for QA/QC

	^{235}U/^{238}U	^{234}U %	^{235}U %	^{236}U %	^{238}U %
NIST-4	0.007436	0.00551	0.7381	0.00056	99.2559
NIST-5	0.007347	0.00543	0.7293	0.00028	99.2650
YC9081	0.007161	0.0055	0.711	N/A	99.284

Table-1: The certified values (where applicable) for the NIST CRM, IRMM and other uranium containing QA/QC standards used by the IAEA.

Results and discussion

The particle samples acquired by NRL from the IAEA represent a variety of U-bearing standards for quality assurance (QA) and quality control (QC) purposes. The NIST CRM standards demonstrate a measurement capability over a range of ^{235}U enrichments (that overlaps with those typically encountered by IAEA inspectors). The IRMM standards, although analyzed in their final oxidized form, represent an ability to analyze particles derived from alternative sources of uranium. We used the NIST-4 and NIST-5 glass particles to evaluate our ability to detect ppm levels of ^{236}U. For all of these samples, we postulate the same core question: Can the NAUTILUS efficiently dissociate the ^{235}U^{1}H interference on ^{236}U for samples of interest to the international Safeguards community?

Figure-1 shows the cumulative data for all of the runs (3-4 particles per sample) of all of the following samples (yellowcake [YC], CRM U010, CRM 030a, IRMM 183 and IRMM 023). The plots show the uranium isotope ratios of ^{234}U/^{238}U vs. ^{235}U/^{238}U in Figure-1a and ^{236}U/^{238}U vs. ^{235}U/^{238}U in Figure-1b. Dashed lines indicate the locations of the certified reference values where applicable. The IRMM 023 certified reference value for ^{236}U of approximately 100 nmol/mol is located essentially at the zero mark in Figure-1b. Error bars represent the standard error for each individual particle measurement (many of the error bars are within the graph symbols and are not visible). These errors result primarily from counting statistics and scale with the square root of the number of counts obtained for each particle analysis. We can see that all of the measured values agree well with the certified values within error save for the IRMM 023, which has a bit more scatter than the other sample materials. A lack of ion counts drives the uncertainty in these measurements. The IRMM 023 particles proved relatively small compared to the other samples and an individual particle is often completely exhausted before

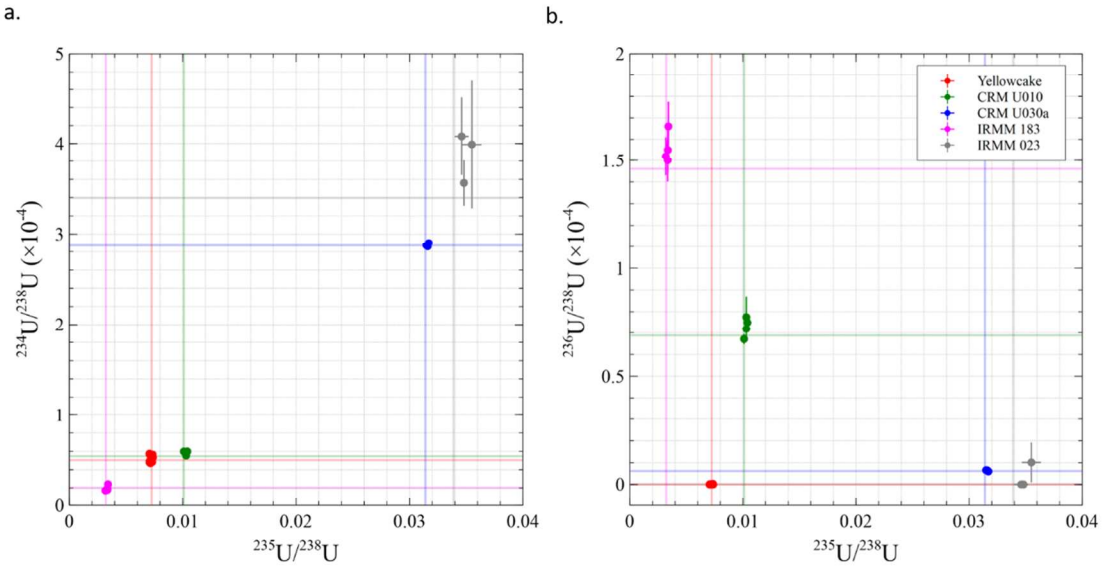

Figure-1: Plots show the uranium isotope ratios of ^{234}U/^{238}U (a) and ^{236}U/^{238}U (b) vs. ^{235}U/^{238}U for QA/QC samples. The dashed lines indicate the location of the certified reference values.

obtaining an adequate number of ion counts for a high precision measurement.

It is important to mention, that the ^{236}U values reported for these samples remain uncorrected. This is in contrast to traditional LG-SIMS measurements that require a hydride correction that relies on an accurate measurement of the ^{235}U, ^{236}U, ^{238}U and $^{238}U^{1}H$ signals. Although previously demonstrated to be effective over a wide range of ^{235}U enrichments, the hydride correction is limited in cases where the ^{236}U signal is present at trace levels and the ^{235}U enrichment is relatively high. Particles containing less than approximately one µmol/mol of ^{236}U could not be corrected using traditional methods.[5] Additionally, the hydride correction would not be effective for samples containing both ^{238}U and ^{239}Pu, as the nominal mass channel at m/z 236 would contain $^{235}U^{1}H$ and ^{236}U and the nominal mass channel at m/z 239 would contain $^{238}U^{1}H$ and ^{239}Pu.[22] We contest that for specific cases; only a direct ^{236}U measurement using the NAUTILUS would provide adequate results.

Figure-2 shows the cumulative data for all of the runs (3-7 particles per samples) of all the following samples (YC, NIST-5, NIST-6 and CRM U030a). The plots show the uranium isotope ratios of $^{236}U/^{238}U$ vs. $^{235}U/^{238}U$. Figure-2a compares the first three samples (YC, NIST-5 and NIST-6) and indicates that the known values of three µmol/mol and six µmol/mol for NIST-6 and NIST-5 respectively agreed with the NAUTILUS results. As a reference, we included the YC particle data to show our uncorrected, ^{236}U background. Our count rate for m/z 236 was essentially zero over the course of measuring samples with no ^{236}U. Figure-2b compares all of the data in Figure-2a, but adds in the data for the CRM U030a particles. Here, a direct comparison between CRM U030a and NIST-5 can be made where each contains approximately six µmol/mol of ^{236}U, but where the CRM U030a contains approximately four times the amount of ^{235}U than the NIST-5 particles. Direct ^{236}U measurement using NAUTILUS are unaffected by the amount of ^{235}U over a dynamic range of approximately 10^5. This means that if we assume a hydride contribution of approximately 1:1000 $^{235}U^{1}H$ counts relative to ^{235}U, a direct ^{236}U measurement at six µmol/mole would be possible even for highly enriched uranium-bearing particles. Arguably, a more interesting contention is that given enough atoms, a direct, sub-100 nmol/mol ^{236}U measurement on a 3% ^{235}U enriched particle is possible. We intend to test this hypothesis in the future by obtaining larger IRMM 023 particles with a certified ^{236}U composition of 111.5 nmol/mol and a ^{235}U composition of 3.28 %. A number of other studies have focused on particle analysis of relatively pure materials under ideal conditions (i.e. U_3O_8 particle vacuum impaction onto vitreous carbon planchettes). However, our interests extend to systems that are more complicated. To that end, Figure-3 shows several images of CRM U500 particles dispersed on a background of monazite particles. We focused on four main isotopes to image (^{232}Th, ^{235}U, ^{236}U and ^{238}U); however, as mentioned above, analysis of the monazite bulk material has revealed that there is a peak at nearly every mass. Each pane of Figure-3 compares the SIMS image taken prior to molecular dissociation and the molecule-free SSAMS image taken

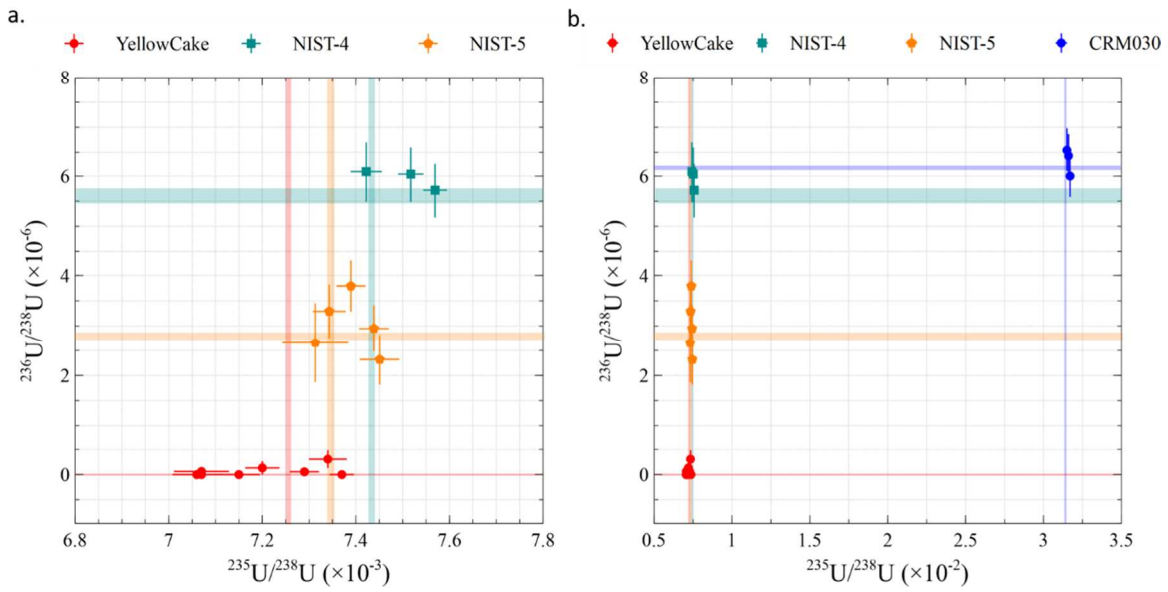

Figure-2: Plots show the uranium isotope ratios of $^{236}U/^{238}U$ vs. $^{235}U/^{238}U$ for YC, NIST-5 and NIST-6 (a) and for an additional sample CRM U030a (b). The dashed lines indicate the location of the known values for the ^{236}U concentrations.

at the end station of the NAUTILUS. Overall, the ^{232}Th, ^{235}U and ^{238}U images in Figure-3a, b and d, respectively, look comparable, save for a greatly reduced background for the SSAMS images compared to the SIMS ones. This is, primarily, because most of the CRM U500 particles are large and have high intensities. This makes them easily distinguished from the background, at least visually. The one exception to this observation is the ^{236}U signals shown in Figure-3c. Arrows point to three particles of interest in this pair of images. Although only represented by a handful of pixels, the SSAMS image differentiates the three distinct particles, whereby the SIMS image is only capable of identifying two particles. The SIMS image backgrounds for the m/z of 236 are simply too high for the lowest intensity particle.

Taking a closer look, Figure-4 zooms in on the three particles of interest. Analysis of this region commenced by alternating SIMS m/z 236 measurements in Figure-4b and d with SSAMS m/z 236 measurements in Figure-4a and c. We split these analyses into the first three iterations in Figure-4a and b and the last three iterations in Figure-4c and d and discovered that we completely consumed the small, less intense CRM U500 particle in the process. This observation is present in the SSAMS images comparing Figure-4a and c where there are originally three distinct particles, but only two remain after a few image cycles. This finding is in contrast to the SIMS images comparing Figure-4b and d where there are only two observable particles in either image. A closer examination of a sub-region of the images indicated by the dashed box in Figure-4d (we interrogated the region for each image, although have only shown it in a single image for demonstration purposes) reveals the reason for the discrepancy

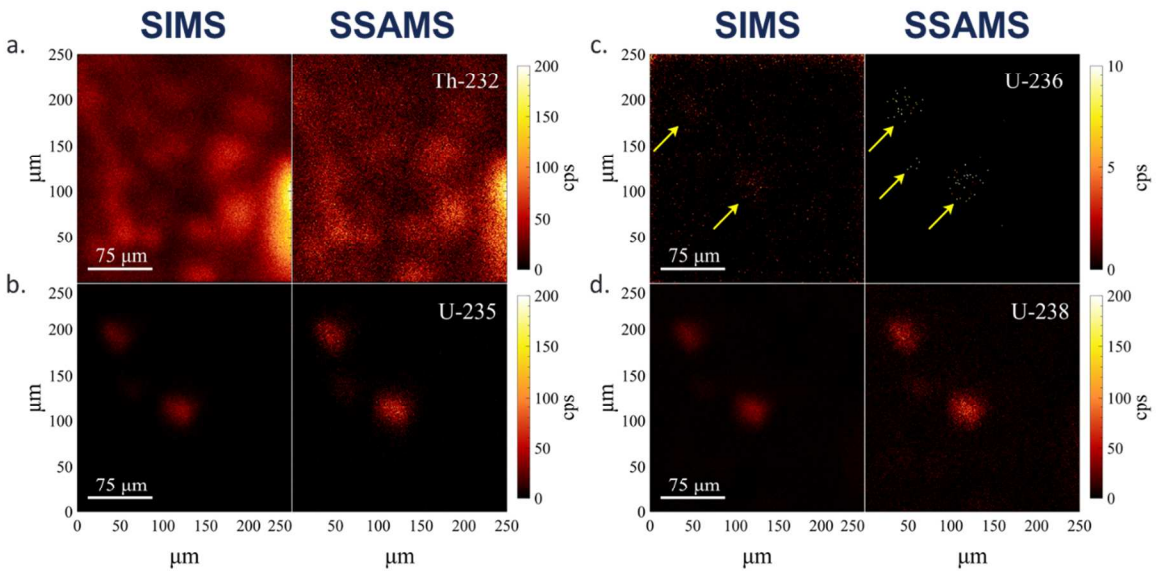

Figure-3: Plots show the comparison between SIMS and SSAMS images for ^{232}Th, ^{235}U, ^{236}U and ^{238}U for a, b, c and d respectively. The sample is NIST CRM U500 particles in the presence of monazite particles. The yellow arrows indicate particle identified by m/z 236.

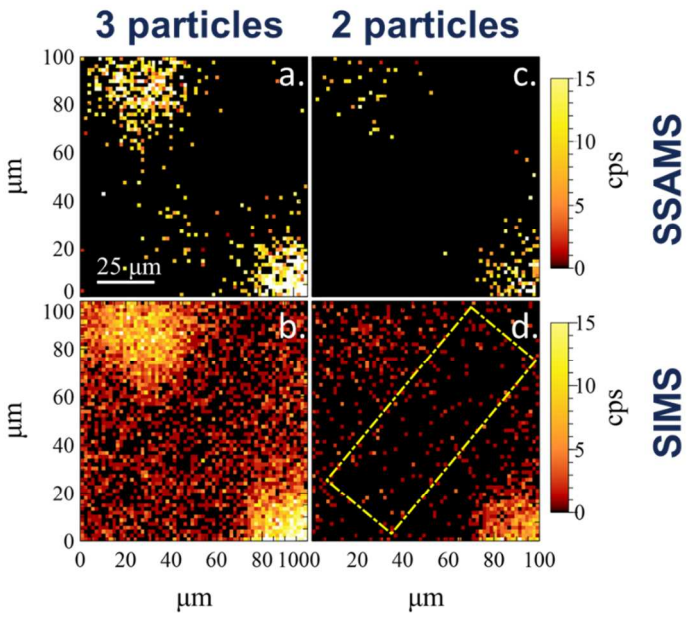

Figure-4: Plots show the comparison between SIMS (b, d) and SSAMS (a, c) images for ^{236}U for the first three (a, b) and last three (c, d) image iterations. The dashed yellow box shows the region of interest for the two-dimension data present in Figure-5.

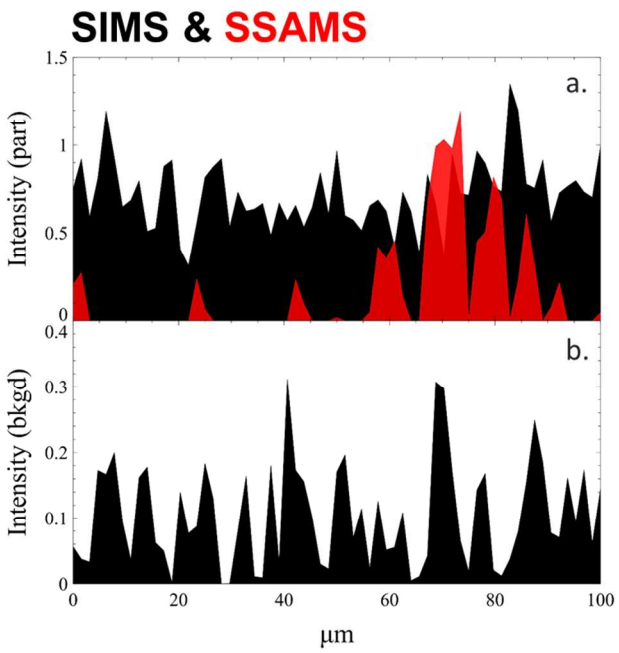

Figure-5: Plots show the line scan comparison between SIMS (black) and SSAMS (red) from Figure-4 first three (a) and last three (b) image iterations. There is no SSAMS signal in the last three (b).

between the SIMS and SSAMS images. Figure-5 shows the comparison of these sub-regions SIMS vs. SSAMS for the first three image cycles in Figure-5a and for the last three image cycles in Figure-5b. The two-dimensional data in Figure-5 represents the sum of all of the counts along the y-axis of the sub-region rectangle plotted versus the x-axis position in order to form a thick line scan of the area between the two more intense CRM U500 particles. As previously mentioned, the SSAMS backgrounds are essentially zero for the mass channel at m/z 236 allowing for the detection of the low intensity CRM U500 particle. SIMS, on the other hand has higher backgrounds that, in this case, obscure the low intensity particle allowing for the detection of only two particles. While we are aware that these data represent only one unidentified particle in one FoV, extrapolation of these data to the total scan area of a single vitreous carbon planchette with well over a thousand such FoVs could conservatively result in at least tens of unidentified particles. Additionally, the unidentified particles would be disproportionately less intense and, therefore, smaller than the average population of identified particles introducing a significant size selection bias into the SIMS only data.

Conclusions

We have demonstrated the utility of the NAUTILUS for the direct, uncorrected measurement of ^{236}U in concentrations as low as a three µmol/mol from IAEA QA/QC standards ranging in ^{235}U enrichment from 0.32% to 3.28%. We have shown that single particle analysis by NAUTILUS is no more complicated and only slightly more time consuming (primarily due to mono-collection vs. multi-collection detector variations) than traditional LG-SIMS analysis and, therefore, is compatible with the already existing IAEA particle analysis protocols. Our ability to directly

measure ^{236}U is unaffected by the amount of ^{235}U present in the sample. Moreover, where the LG-SIMS community has largely focused in the literature on investigating the hydride correction in the ideal case (pure oxide samples well separated on a clean substrate), we have considered two more probing questions. What if the sample has multiple actinides (i.e. U and Pu) and/or the sample matrix is complex with many more opportunities for interfering species? What if an analysis calls for particle distributions not well separated from each other and not sufficiently well removed from a more complex background material? Future analysis directly comparing our results with those of LG-SIMS will be critical to answering these questions in a detailed and comprehensive manner.

As required for automated screening of particle distributions by LG-SIMS, we have demonstrated that the NAUTILUS is capable of raster ion imaging comparable to traditional LG-SIMS methods. Direct analysis of the ^{236}U present in CRM U500 particles in a field of monazite particles illustrates our ability to detect low intensity, small particles free from background signals. This opens the door for advanced screening methodologies. For example, instead of screening for ^{234}U, ^{235}U, ^{236}U, ^{238}U and $^{238}U^{1}H$ as is typical for LG-SIMS analyses, one could simply screen for ^{236}U with the NAUTILUS. This method would likely allow for the use of faster scanning speeds, therefore, decreasing the total time to screen the total area of a vitreous carbon planchette. Knowing that nearly every pixel is a positive particle identification, leads to a much more targeted screening approach than is currently available by traditional LG-SIMS. Additionally, we have demonstrated the ability of the NAUTILUS to detect low intensity, small particles that are within the background signal of comparable SIMS measurements. While we are aware that LG-SIMS will not likely encounter this challenge for pure oxide particles on a clean substrate, we contend that the capabilities of the NAUTILUS may supplant traditional LG-SIMS when the need to analyze non-ideal samples arises.

For clarity, consider the following hypothetical scenario. There are two particle populations with >10,000 particles each, they have been enriched to the same $^{235}U/^{238}U$ ratio and, therefore, cannot be discriminated based on their major uranium isotope composition alone. These particle populations differ in the fact that one population was enriched from fuel that had not been reprocessed (i.e. no ^{236}U) and the other from fuel that had been processed (i.e. containing ^{236}U). If we simply wanted to answer the question, “was this uranium-bearing particle population derived from fuel that had been reprocessed,” we could use the aforementioned particle searching methodology for ^{236}U with the NAUTILUS to get a fast, yes or no determination without the need to make high precision measurements of all of the uranium isotopes. In this scenario, if we observe ^{236}U in the first, fast-pass scan, we then revisit the individual particles for a more through high precision measurement. For this type of “real-world” application, the NAUTILUS would be well suited to add value to the already formidable combination of FT-TIMS and LG-SIMS.

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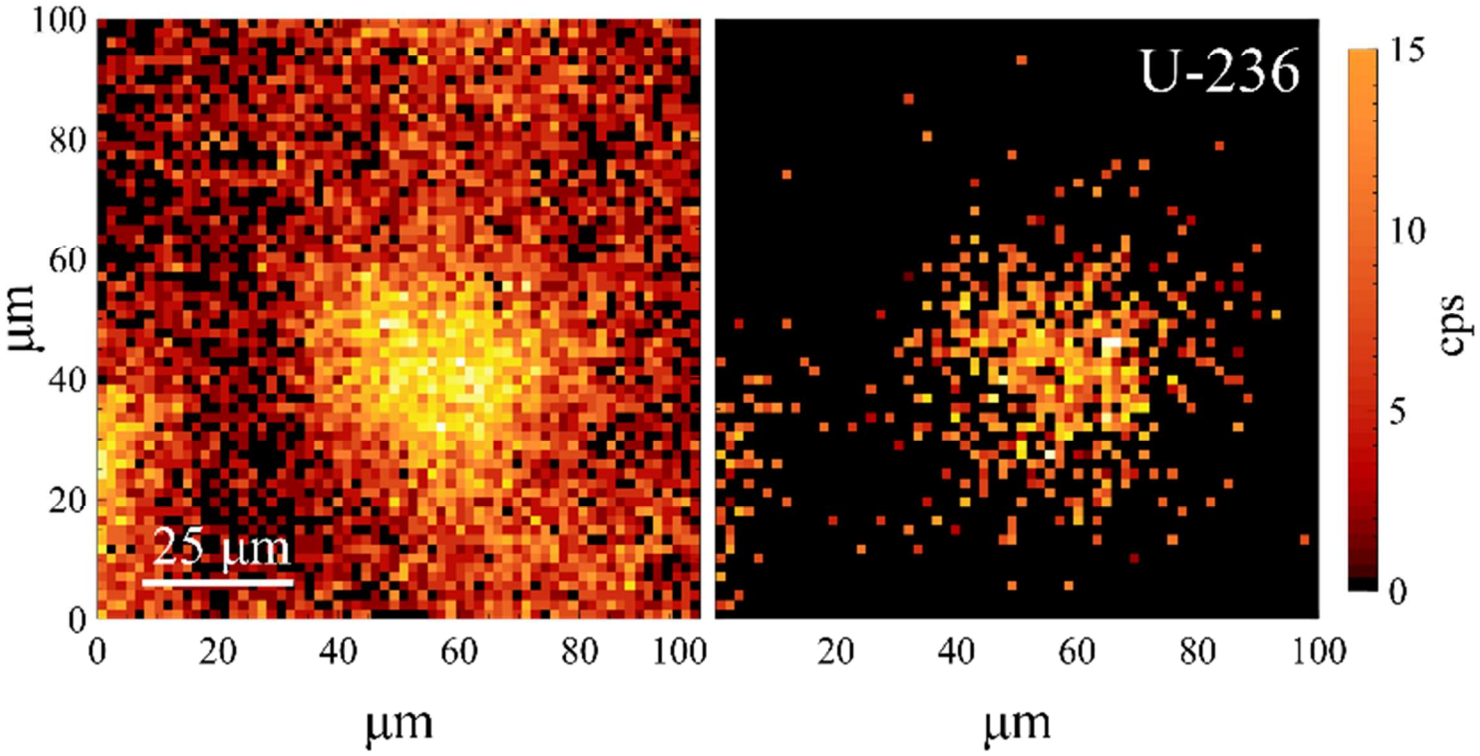

The Naval Ultra-Trace Isotope Laboratory's Universal Spectrometer (NAUTILUS) can measure ^{236}U directly from uranium-bearing particles free from molecular isobaric interferences.