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New Particle Working Standards for NWAL Particle Laboratory Calibration and Quality Control – LANL LG-SIMS Characterization and Evaluation

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1   Abstract

The LG-SIMS capability at Los Alamos National Laboratory (LANL) has a key role in evaluating the isotopic characteristics of particle materials generated by Savannah River National Laboratory (SRNL) and by Pacific Northwest Laboratory (PNNL) for the network of analytical laboratories (NWAL). Regarding the generation of new uranium working reference materials, the IAEA has provided specific isotope metrics that must be met in order for a material to be deemed a qualified working reference material. These metrics include (1) that materials must be isotopically homogeneous from particle to particle, with a limited number of outliers (evaluated via LG-SIMS isotope mapping of large particle populations); and (2) that LG-SIMS mean isotope compositions of particle datasets agree with bulk values determined by TIMS or MC-ICP-MS, and also agree with the compositions requested by the IAEA (which is evaluated by LG-SIMS high-precision single particle isotope analyses). For such evaluations the LANL LG-SIMS team has developed a counting statistics-based model that is used to determine the homogeneity (or lack thereof) of particle datasets, and can also be used to predict the level of analytical precision required to resolve two different endmember isotope compositions. Regarding the generation of working reference particles with mixed U and Pu, the LANL LG-SIMS team has been developing methods for their characterization, including techniques to minimize molecular hydride interferences, and evaluating intra-element and inter-element isotope homogeneity (or lack thereof) of particle materials. Challenges of producing mixed actinide working reference materials are discussed within the context of observed LG-SIMS isotope characteristics of mixed Pu-U test materials.

2   Introduction

Environmental sample characterization by the IAEA Network of Analytical Laboratories (NWAL) relies on the availability and measurement of reference materials with known and homogeneous isotope compositions. Such materials are used to calibrate analytical instruments for accurate isotope ratios of environmental sample unknowns, and they are also used for quality control (QC) and proficiency testing of NWAL laboratories. With the support of various IAEA member state support programs, recent advances allow for production of actinide bearing QC reference material particles, including hydrothermal synthesis (Trilaud et al., 2018; Pope et al., 2019), and aerosol-based generation (Neumeier et al., 2018; Kegler et al., 2021; Scott et al., 2021). The IAEA provides specific isotope compositions and metrics to qualify generated materials as fit for QC purposes. Specifically, materials must be isotopically homogeneous from particle to particle, with documentation there are a limited number of outlier particles. In addition, particles must also have IAEA-requested isotope compositions that agree with bulk analyses, and meet specified Zeta-score criteria. For production of actinide-bearing particle materials at Pacific Northwest National Laboratory

(PNNL) (hydrothermal synthesis) and at Savannah River National Laboratory (SRNL) (aerosol-based generation), Los Alamos National Laboratory (LANL) supports their efforts through evaluation of sample particles by large geometry secondary ion mass spectrometry (LG-SIMS) characterization. Here, we detail the LANL LG-SIMS protocols to evaluate isotope homogeneity of samples at the particle level, and ways to determine the ability to discriminate different endmember isotope compositions. For recent efforts to produce QC materials with mixed Pu and U (Wellons et al., 2023), methods to calculate and reduce molecular hydride interferences at U and Pu masses analyzed by LG-SIMS are provided here, and challenges of mixed Pu-U particle synthesis are discussed within the context of LG-SIMS data from test materials particles.

3   LG-SIMS Methods

The LG-SIMS at LANL is a Cameca IMS 1280 **(Figure 1)**. For analysis of actinides a duoplasmatron ion source generates a beam of O⁻ ions that are focused and accelerated (13 kV) through the primary column and ultimately strike the sample. This causes sputtering of secondary ions from the sample, which are focused and accelerated (10 kV extraction voltage) through the transfer column, electrostatic analyzer, coupling column, magnet, projection column, and finally to the detectors. Throughout the primary and secondary flight paths are deflectors, hexapoles, stigmators, apertures, and slits used to align the beam, limit aberrations, and to control the mass resolution (M/ΔM). Detection of signal employs multicollection, meaning the magnet is held at a fixed axial mass, and five electron multiplier (EM) detectors (Fig. 1: labeled as L2, L1, C, H1, H2) are positioned to collect signals from different masses simultaneously. Typical operating parameters include a transfer magnification area of 60 × 60 microns, an entrance slit width of 122 microns, a contrast aperture setting of 400 microns, a field aperture setting of 4000 to 6000 microns, an energy slit setting of 50 eV, and an exit slit width of 500 microns. These settings correspond
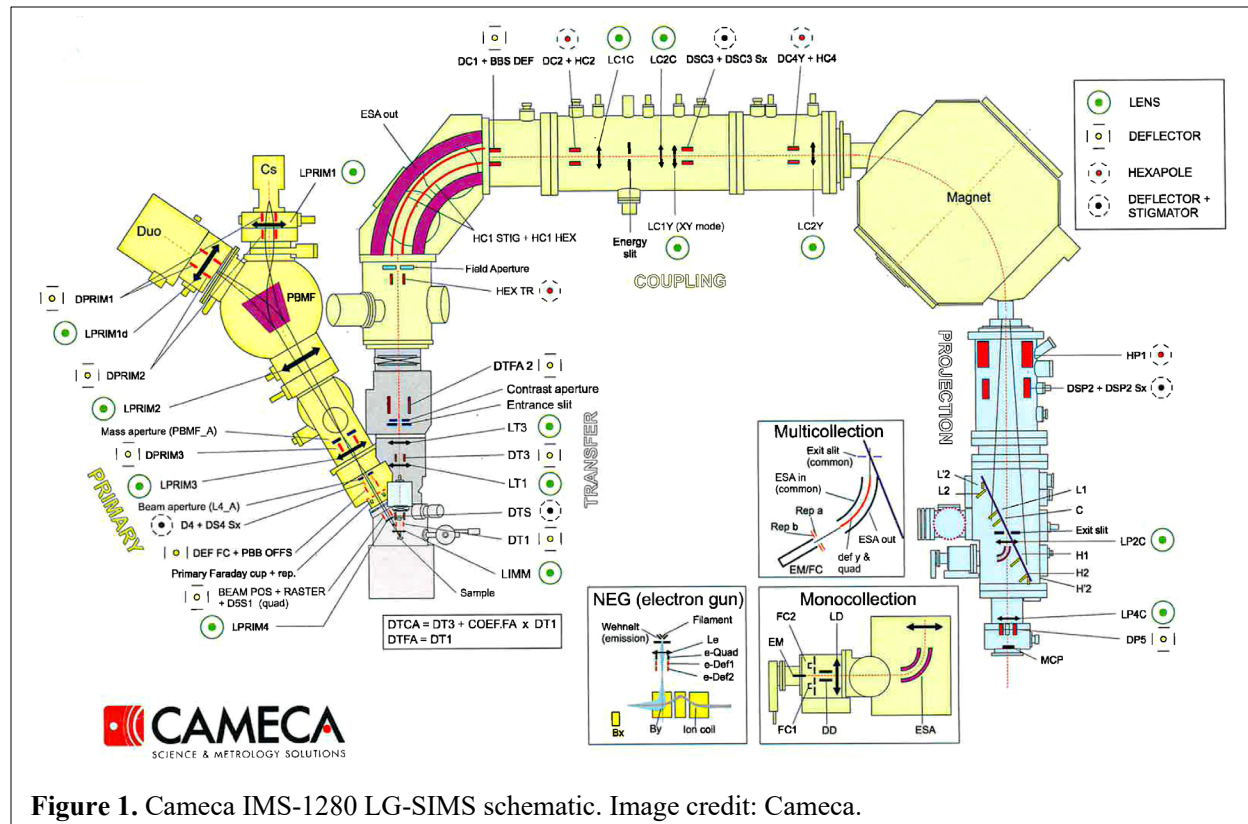

Figure 1. Cameca IMS-1280 LG-SIMS schematic. Image credit: Cameca.

to a mass resolution of ~1800. Depending on the primary ion beam raster size and particle coverage (further detailed below), the primary ion beam current is adjusted so that a signal of ~1e5 counts per second (cps) of the major isotope is achieved on its corresponding EM detector. For particles only containing uranium the detectors are positioned for the following masses: L2: 234; L1: 235; C: 236; H1: 238; H2: 239. At mass 239 $^{238}U^1H$ signal is used to estimate and subtract the contribution of $^{235}U^1H$ at mass 236 (because a prohibitively high mass resolution is required to separate signals of $^{235}U^1H$ and ^{236}U) under the assumption that $(^{238}U^1H/^{238}U)_{measured}$ is equivalent to $^{235}U^1H/^{235}U_{measured}$. This allows for determining a hydride-corrected ^{236}U signal (see also: Simons and Fassett, 2017). For analyses of particles with Pu and U, it is common that four of the five multicollectors are positioned to detect signal from masses 235, 238, 239, and 240 (e.g. typically the most abundant isotopes of Pu and U), with the fifth multicollector positioned for a desired minor isotope of Pu or U. At mass 241, ^{241}Pu and ^{241}Am are not resolved under the aforementioned operating conditions. Also, no mathematical estimate of hydride interference signal can be employed for simultaneous detection of U and Pu isotopes (e.g. $^{238}U^1H$ and ^{239}Pu, $^{239}Pu^1H$ and ^{240}Pu, $^{240}Pu^1H$ and $^{241}Pu + ^{241}Am$, $^{235}U^1H$ and ^{236}U) because there are too many unconstrained variables. However, the hydride abundance can be estimated from measurements of the substrate surrounding particles, and used to correct raw data, as detailed later.

Samples consist of micron-sized particles that are electrostatically adhered to 1-inch diameter circular substrates (commonly doped Si wafers or carbon planchets). There are several methods for dispersing particles onto wafer substrates, including electrostatic deposition of aerosols (e.g. Scott et al., 2021), deposition of liquid particle suspensions (e.g. Middendorp et al., 2017), and vacuum suction and impaction (e.g. Esaka et al., 2004).

Two types of analysis are employed to characterize a sample (**Figure 2**): (1) using a large primary ion beam raster (typically 150 µm^2 to 250 µm^2) per analysis to map particles, which allows for collecting large datasets that are representative of the sample; and (2) high-precision single particle analysis using a small primary ion beam raster (5 µm^2 to 10 µm^2). Particle mapping data are lower in precision because the signal per analysis is split amongst the particles within the mapped region **(e.g. Figure 2a)**, but they are advantageous because the large populations allow for evaluating the particle-to-particle isotope homogeneity of a sample, and the proportion of outliers that may be present. Care must be taken during mapping to ensure the raster size is not significantly larger than the magnification area, to prevent shadowing effects at the edges of maps. Single particle data have comparatively higher precision because
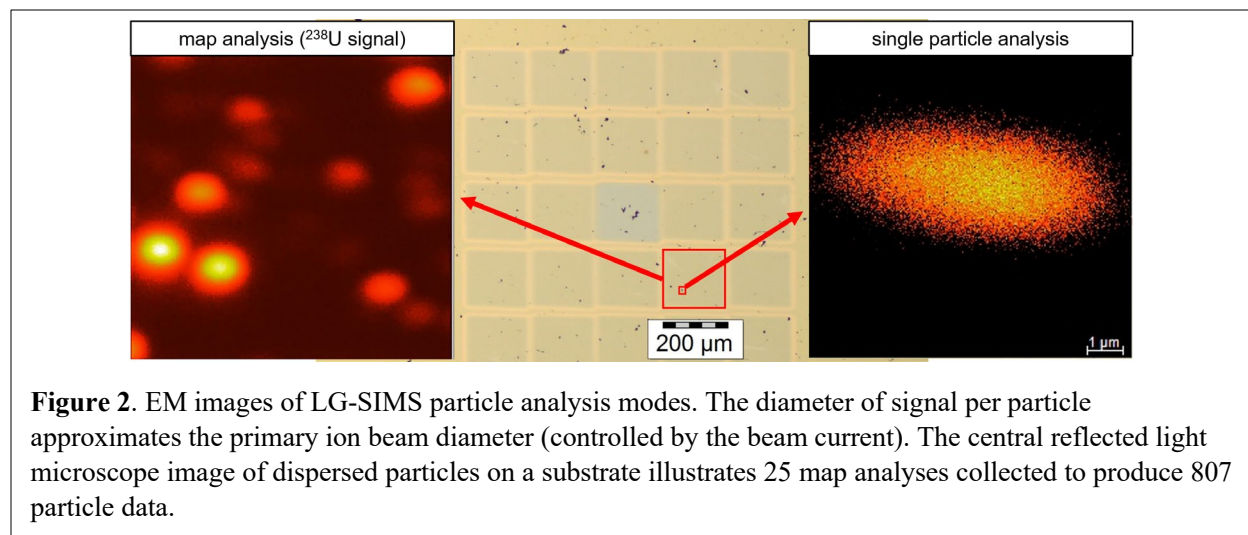
Figure 2. EM images of LG-SIMS particle analysis modes. The diameter of signal per particle approximates the primary ion beam diameter (controlled by the beam current). The central reflected light microscope image of dispersed particles on a substrate illustrates 25 map analyses collected to produce 807 particle data.

all signal from the analysis is attributed to only one particle (**e.g. Figure 2b**). This means that, although fewer single particle data are typically collected within a given time frame, they better constrain the isotope composition of a sample. As such, Zeta-scores of single particle data are used to evaluate fit-for purpose of particle isotope compositions. Also, single particle data best resolve materials with different isotope compositions, as discussed later. Map analyses are typically 4 minutes in duration (plus a one-to-five-minute sputter cleaning step) with a 5 nA to 20 nA primary ion beam current (depending on the raster size and the particle coverage per area). Depending on the particle coverage, a few to tens of map analyses are collected in order to build up hundreds to thousands of particle data. Cameca's Automated Particle Measurement (APM) software is used to process isotope ratios of identified particles, with uncertainties reported as counting statistics. Single particle analyses are typically 2 minutes in duration (plus a 30 second sputter cleaning step) using a 500 pA to 1 nA primary ion beam current. Each analysis is split into ten blocks of 12 seconds, and uncertainties are reported as the expanded standard error of the block data. Typically 10 to 50 single particle analyses are collected per sample.

Reference materials of a similar matrix and isotope composition are analyzed with unknowns to determine the instrument bias, defined as $X_{raw}/X_{reference}$, where X is the isotope ratio of interest. The instrument bias can then be used to correct raw isotope ratios from unknowns to make them accurate. Several uranium particle certified reference materials (CRMs) exist, allowing for robust instrument bias corrections. However, mixed U/Pu particle CRMs currently do not exist. Instead, working reference materials of mixed U and Pu, for which U and Pu intra- and inter- element ratios have been determined by bulk mass spectrometry techniques can be used to determine the LG-SIMS instrument bias, as well as the element Pu/U relative sensitivity factor (RSF), defined as the $(U/Pu)_{raw}/(U/Pu)_{reference}$. Note that in the absence of bias correcting data, the homogeneity of datasets can still be meaningfully evaluated, albeit with systematic inaccuracy.
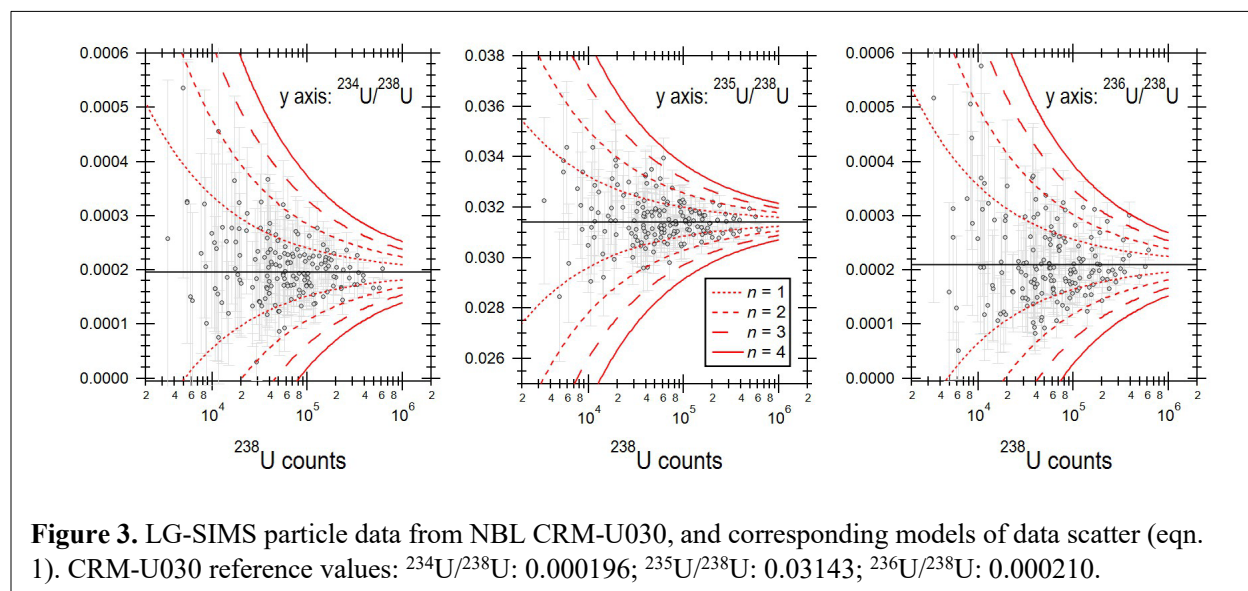
4   Discussion

4.1   Evaluating isotope homogeneity of particle datasets.

Assuming CRMs are isotopically homogeneous at the particle level (they are certified only at the bulk level), their corresponding LG-SIMS particle data can be used to model the expected scatter of isotope ratios as a function of counts per particle. This is illustrated in **Figure 3**, showing an example LG-SIMS mapped particle dataset from New Brunswick Laboratory (NBL) CRM U030 U_3O_8 material. Notably, the particle data are equivalently scattered on either side of the average isotope ratio of the dataset. Also of importance is that particle data with more counts exhibit less scatter about that average, while those with fewer counts exhibit a greater extent of scatter about the average (**e.g. Figure 3a**). This observed behavior can be modeled using counting statistics, expressed as:

$$avg.\ isotope\ ratio \pm \left(avg.\ isotope\ ratio \times n \times \sqrt{\frac{1}{N(avg)counts} + \frac{1}{D(avg)counts}} \right) \qquad (1)$$

The *avg. isotope ratio* is that of a dataset or any desired isotope ratio to be modeled. $N_{(avg)counts}$ and $D_{(avg)counts}$ correspond to all possible count combinations of the numerator and denominator of the *avg. isotope ratio* or any desired isotope ratio to be modeled. The term *n* approximates a Gaussian distribution, probability = erf($n/\sqrt{2}$), meaning values of 1, 2, and 3, and 4 yield correspond to approximately 68.2%, 95.5%, 99.7% and 99.99% of data representing a homogeneous material falling within the model bounds, as shown in **Figure 3**. Values of *n* between 3 and 4 are suggested to best predict the expected scatter of data for a given isotope ratio. Overall the model provides a robust metric to evaluate if a given material is
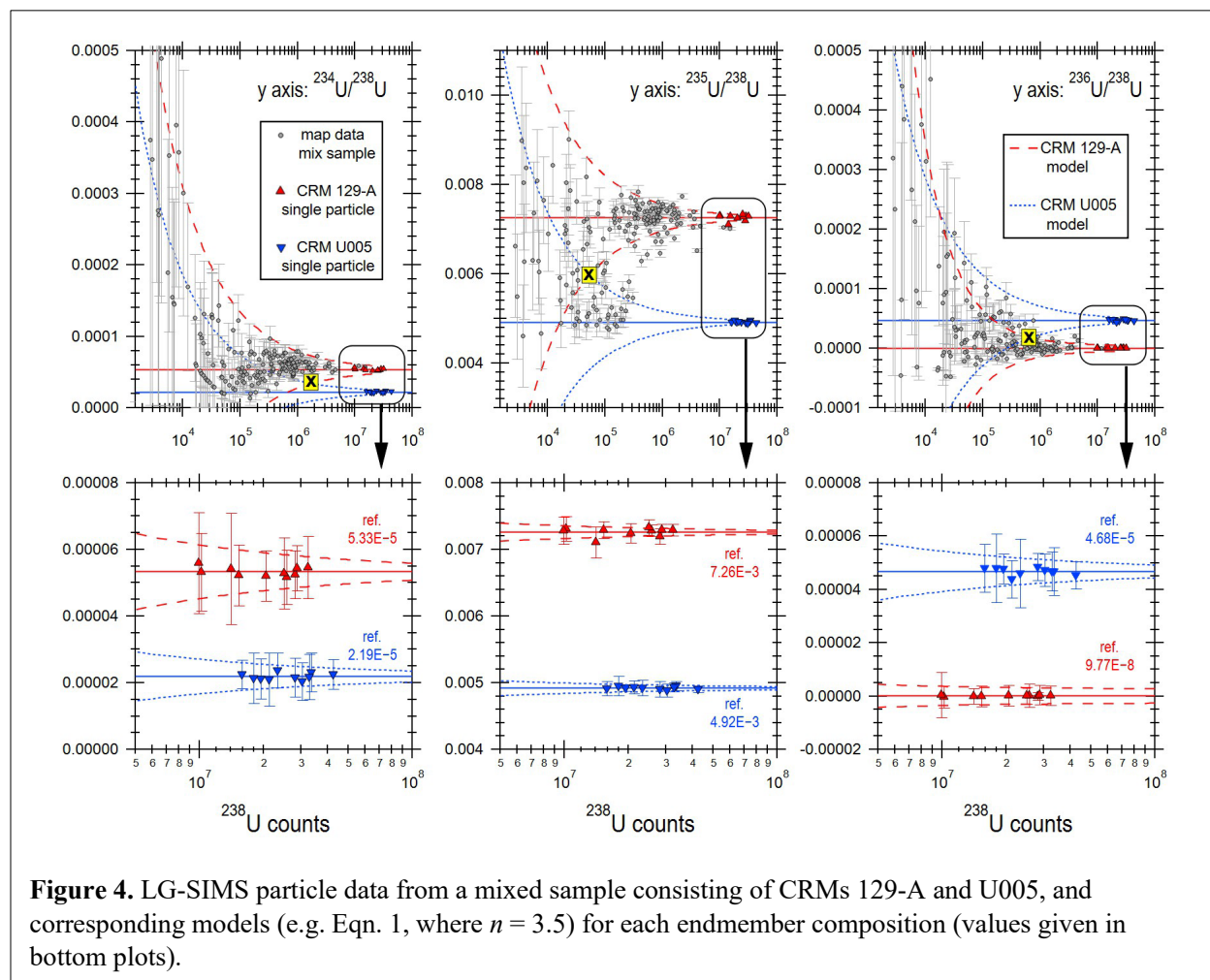
isotopically homogeneous at the particle level. Specifically, fit-for-purpose evaluation of working reference materials by the IAEA stipulates only a limited proportion of particles can be outliers compared to the model.

Figure 3. LG-SIMS particle data from NBL CRM-U030, and corresponding models of data scatter (eqn. 1). CRM-U030 reference values: $^{234}U/^{238}U$: 0.000196; $^{235}U/^{238}U$: 0.03143; $^{236}U/^{238}U$: 0.000210.

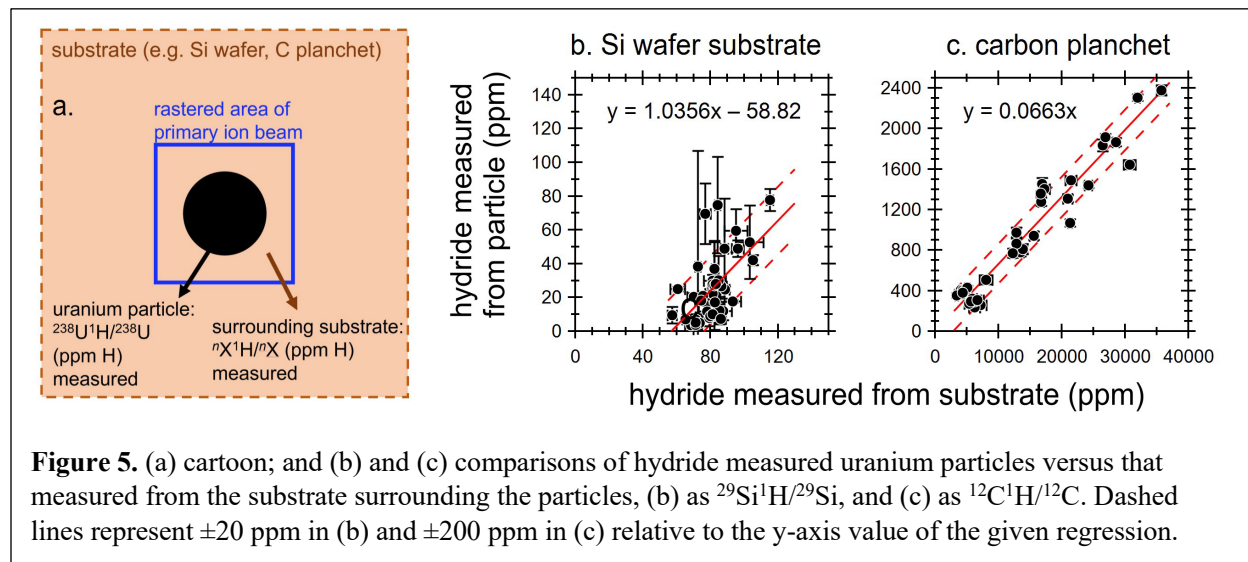## 4.2 Determining the precision needed to resolve different endmembers.

The model given in equation 1 can be used to estimate the level of precision (counts) required to discriminate isotope ratios of two different endmembers. This is illustrated in **Figure 4**, using an *n* value of 3.5 in equation 1, and showing LG-SIMS map and single particle data collected from a sample consisting of mixed NBL CRMs 129-A (natural uranium) and U005 (depleted uranium). For each endmember isotope ratio, models of expected data scatter are constructed, and there are specific ^{238}U count abundances (x-axis) where each of the modeled isotope ratios cross (**Figure 4**, boxed x symbols in top plots); greater versus fewer counts relative the crossover point correspond to endmembers being discriminated versus the lack thereof, respectively. For example, for CRMs 129-A and U005, the respective model crossover points predict at least 1.8 million, 60 thousand, and 700 thousand counts of ^{238}U, respectively, are required to discriminate each CRM in terms of their $^{234}U/^{238}U$, $^{235}U/^{238}U$, and $^{236}U/^{238}U$ ratios, respectively. If only considering the mapped dataset in **Figure 4**, many of the particles have sufficient counts to resolve the CRMs by their $^{235}U/^{238}U$ ratios, but their $^{234}U/^{238}U$ and $^{236}U/^{238}U$ ratios are not resolved; note that a small proportion of the mapped data plot between the models (most clearly seen in the top plot of the $^{235}U/^{238}U$ data), which represent agglomerates of CRM 129-A and U005 particles that led to intermediate isotope compositions (verified by scanning electron microscopy imaging). However, re-analysis of a subset of particles identified as CRMs 129-A and U005 by their mapped $^{235}U/^{238}U$ data (e.g. falling within their respective model bounds), by the single particle method (e.g. **Figure 2**) allows for discriminating the CRMs by all their isotope ratios, because this mode of analysis generates more counts (**Figure 4**, bottom panels). Overall, the example shown in **Figure 4** demonstrates the utility of the model to predict the level of precision required to discriminate two (or more) endmember isotope sources. This is very useful, not only for LG-SIMS, but for any analysis method where signal is generated as counts. Foremost, the model can be used to determine if endmembers can be resolved within reasonable instrument and sample constraints; if so, then it also informs how one

might set up an analysis routine (detector count rates, analysis durations, etc.) to achieve the necessary analytical precision.

Figure 4. LG-SIMS particle data from a mixed sample consisting of CRMs 129-A and U005, and corresponding models (e.g. Eqn. 1, where $n = 3.5$) for each endmember composition (values given in bottom plots).

4.3 Estimating and reducing molecular hydride abundances for mixed Pu-U particle LG-SIMS analysis.

A challenge for LG-SIMS mixed Pu and U isotope analysis is that molecular hydrides at mass 236 (^{236}U and $^{235}U^{1}H$), and all Pu masses (^{X}Pu and $^{X-1}[U,Pu]^{1}H$) cannot be mathematically constrained. However, it is possible to estimate hydride backgrounds through measurements of the substrate surrounding particles. This is illustrated in **Figure 5a** and hereafter described for a single particle analysis routine (hydride form the substrate can also be measured from mapped areas, but is not detailed here). After aiming onto a particle using actinide signal, but before a Pu and U isotope analysis (see LG-SIMS Methods), the magnet is switched to detect ^{n}X and $^{n}X^{1}H$ signal from the substrate with a peak hopping analysis using one of the multicollector EMs; the hydrogen background in ppm can then be calculated from the $^{n}X^{1}H/^{n}X$ ratio. For silicon wafers the $^{29}Si^{1}H$ and ^{29}Si signals work well because they are comparable to those of the actinide hydride signals from the particle (as detailed below). A mass resolution of at least 3500 is required to separate ^{29}Si from $^{28}Si^{1}H$ signal, which is achieved with the following settings: contrast aperture: 150 microns; entrance slit: 80 microns; field aperture: 1300 microns, energy slit: 50 eV; exit slit: 250 microns
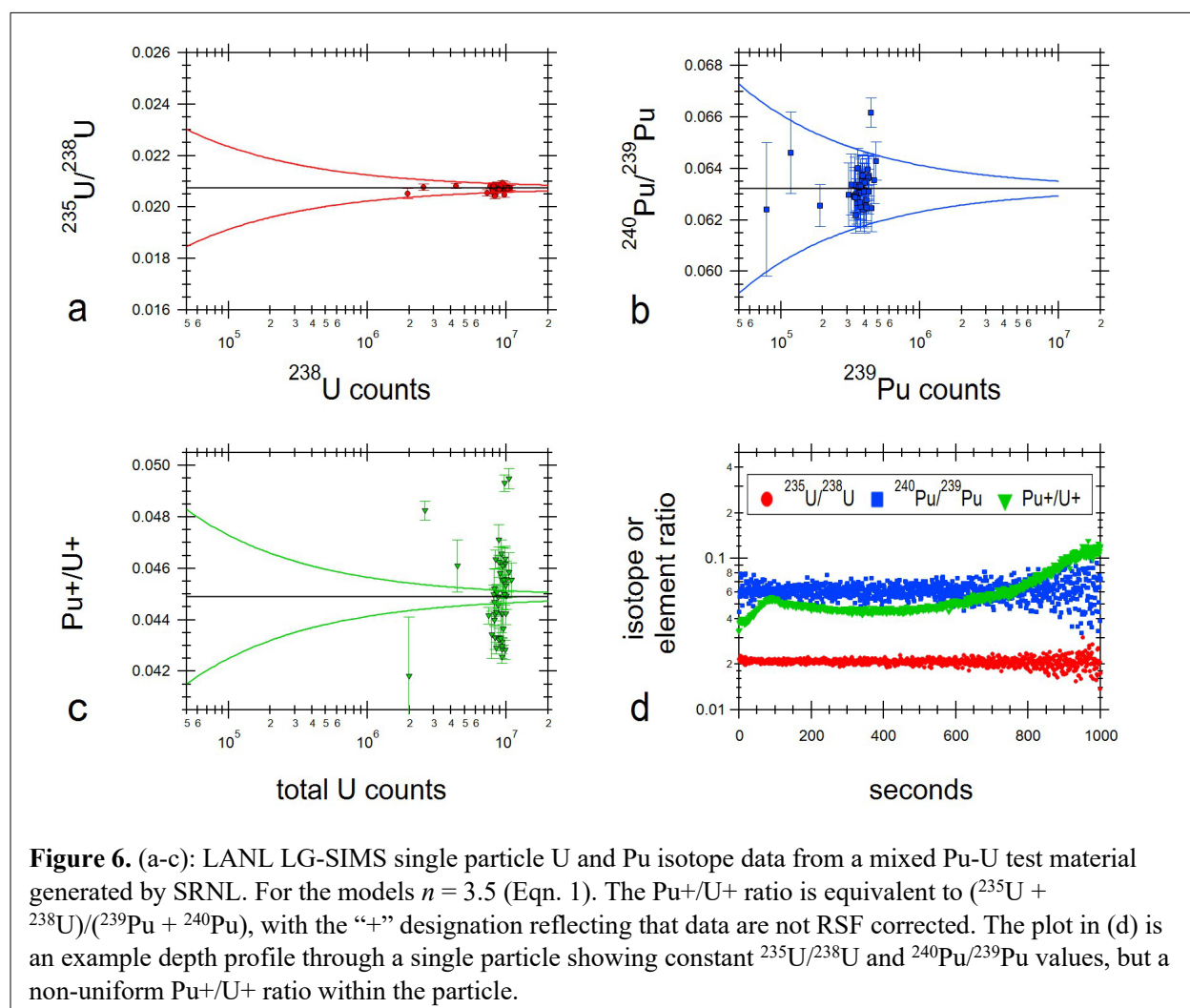
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Figure 5. (a) cartoon; and (b) and (c) comparisons of hydride measured uranium particles versus that measured from the substrate surrounding the particles, (b) as $^{29}Si^1H/^{29}Si$, and (c) as $^{12}C^1H/^{12}C$. Dashed lines represent ±20 ppm in (b) and ±200 ppm in (c) relative to the y-axis value of the given regression.

(multicollection position 2). For carbon planchets the hydride is measured as the $^{12}C^1H/^{12}C$ ratio. A mass resolution of 3500 is needed to separate $^{12}C^1H$ and ^{13}C, and is achieved with the same settings as those for $^{29}Si^1H/^{29}Si$ analysis, except that the contrast aperture is 400 microns, and the field aperture is 3000 microns. These settings produce signal that is comparable to that of the actinide hydride signals measured from the particle. For each substrate type an analysis of 155 seconds in employed, consisting of five cycles of the following: (1) a 3 second measurement of ^nX signal followed by a magnet peak hop and a 4 second magnet settling time; and (2) a 20 second measurement of $^nX^1H$ signal followed by a magnet peak hop and a 4 second magnet settling time. The $^nX^1H/^nX$ uncertainty of a single analysis is calculated from the expanded standard error of the cycling data. Prior to the substrate analyses, a 2-minute sputter cleaning step is applied using a 50 micron primary beam raster and a 30 nA beam current.

To test the accuracy of measured hydride backgrounds substrates, their data can be compared to hydride backgrounds measured from particles only with uranium (as $^{238}U^1H/^{238}U$). This is shown in **Figures 5b and 5c**, for particles on Si wafer substrates and carbon planchets, respectively. Uranium particle data come from test materials provided by SRNL that were electrostatically deposited onto substrates, and they also come from CRMs deposited onto substrates via pipetted methanol suspensions. From **Figures 5b and 5c** there are linear relationships between the hydride background measured from the substrate versus the hydride measured from the particle. For Si wafer substrates the data produce a slope of approximately 1, with the hydride measured from the particle being about 60 ppm lower than that measured from the surrounding substrate **(Figure 5b)**. In **Figure 5c** the hydride measured from particles is only 0.0663 times that of the hydride background measured from the surrounding carbon planchet. However, the most important finding is that the hydride background from particles on Si wafer substrates is less than 100 ppm. At such low hydride backgrounds the effect of molecular hydride interferences is negligible for most Pu and U intra-element and inter-element isotope ratios. As such, the use of Si wafer substrates, sample preparation using electrostatic particle deposition and/or pipetted methanol particle suspensions, and the analysis routine described here allows for LG-SIMS to make proper isotope ratio measurements of particles with mixed Pu and U. Carbon planchets generate higher hydride backgrounds (hundreds to thousands of ppm), which, if uncorrected, can cause inaccuracies of mixed Pu-U particle data by LG-SIMS. Overall, the differences of hydride backgrounds from Si wafer and carbon planchet substrates shown in **Figure 5** is similar to previous findings by Simons and Fassett (2017).

4.4 Example LG-SIMS data from synthesized mixed Pu-U test materials.

In preparation for generating mixed Pu-U working reference materials for the IAEA (e.g. Wellons et al., 2023), SRNL has produced several test materials to evaluate the synthesis process. **Figure 6** shows LG-SIMS single particle isotope data from a SRNL-generated test material consisting of a CRM U020-A feedstock (^{235}U/^{238}U: 0.0207), and a Pu feedstock with a ^{240}Pu/^{239}Pu ratio of 0.0636; these feedstocks were mixed to generate a nominal Pu/U ratio of 0.0309. The LG-SIMS data show homogeneity in ^{235}U/^{238}U and ^{240}Pu/^{239}Pu, and the averages agree with the nominal values (LG-SIMS average and 2SD: ^{235}U/^{238}U: 0.0207 ± 0.0002; ^{240}Pu/^{239}Pu: 0.0632 ± 0.0014) (**Figures 6a and 6b**). In contrast, the Pu/U (= $^{235+238}$U/$^{239+240}$Pu) values exhibit scatter beyond the predicted model bounds, and the average (0.0449 ± 0.0033 [2SD]) does not match the nominal Pu/U of 0.0309; however, part of this discrepancy is due to the LG-SIMS Pu/U RSF. As such, the y-axis in Figure 6c is denoted as Pu+/U+ in recognition that the data are not RSF-corrected. In order to better understand the LG-SIMS measured Pu/U average and RSF, 20 particles were removed from the Si wafer substrate with a computer-controlled micromanipulator, and were binned and analyzed by isotope dilution mass spectrometry (IDMS) at LANL. This generated a Pu/U of 0.0161 ± 0.0008. Using the IDMS data as the (Pu/U)$_{actual}$ of the material, and when combined with the LG-SIMS (Pu+/U+)$_{measured}$ value of 0.0449, the calculated LG-SIMS Pu/U RSF is 2.8. This RSF

Figure 6. (a-c): LANL LG-SIMS single particle U and Pu isotope data from a mixed Pu-U test material generated by SRNL. For the models $n = 3.5$ (Eqn. 1). The Pu+/U+ ratio is equivalent to (^{235}U + ^{238}U)/(^{239}Pu + ^{240}Pu), with the "+" designation reflecting that data are not RSF corrected. The plot in (d) is an example depth profile through a single particle showing constant ^{235}U/^{238}U and ^{240}Pu/^{239}Pu values, but a non-uniform Pu+/U+ ratio within the particle.

is similar to the Pu/U RSF value of 2.4 determined by SIMS measurements from Tamborini et al. (2002). Collectively, the LANL IDMS Pu/U and the LG-SIMS Pu/U RSF indicate that a lower-than-intended abundance of Pu was incorporated into the particles for this particular synthesis run at SRNL. This information was conveyed to SRNL and modifications to their synthesis process has led to production of subsequent test materials where Pu abundances more closely match the intended values. Regarding the greater LG-SIMS Pu/U data scatter relative to the model prediction, it is hypothesized this represents variable mixing of Pu and U within the synthesis liquid material and/or within each individual particle. For example, LG-SIMS depth profiles through single particles show that the Pu/U values are not constant (e.g. **Figure 6d**). However, some of this profiling behavior could also be due to crystallographic transitions in the Si wafer due to oxygen implantation from the primary ion beam (e.g. Sharp et al., 2016; Groopman et al., 2022). Overall, solving these issues will be important for meeting IAEA qualifications as fit-for-purpose mixed actinide particle reference materials.

4   Conclusions

Regarding the generation of new particle working reference materials for the IAEA, LG-SIMS particle isotope characterization is vital for evaluating if the products are fit-for-purpose. Specifically, LG-SIMS allows for determining if materials are isotopically homogeneous from particle-to-particle through mapping analyses and can also provide isotope compositions of materials via high-precision single particle analyses. For LG-SIMS analyses of particles with mixed Pu-U, challenges related to molecular hydride interferences can be accounted for by independently measuring hydride backgrounds from the substrates surrounding particles. By doing so, it has been found that particles affixed to Si wafer substrates by electrostatic deposition and via liquid particle suspensions, and employing a sputter cleaning routine, lead to negligible hydride interferences on affected U and Pu signals. For efforts to generate of new QC particles with mixed Pu and U, LG-SIMS characterization of test materials has revealed challenges related to Pu/U homogeneity from particle-to-particle, and differences between the actual versus intended Pu/U ratios of samples. This information serves as critical feedback to particle production laboratories, such that modifications to the process are made that will ultimately produce improved materials that meet fit-for-purpose as QC materials for the IAEA. Overall the methods described here for evaluating isotope homogeneity of particle datasets are applicable not only to QC materials, but to environmental materials, and not only to LG-SIMS, but for any analytical method where signal is generated as counts.

5   Acknowledgements

6   References

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