Isolation and Purification of Uranium Isotopes for Measurement by Mass-Spectrometry ($^{233}$, $^{234}$, $^{235}$, $^{236}$, $^{238}$U) and Alpha-Spectrometry ($^{232}$U)

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Table of Contents

Summary ............................................................................................................... 01
1.0 Introduction .................................................................................................. 01
2.0 Uranium Isotope Measurement Techniques ............................................... 02
   2.1 Measurements of $^{234}$U, $^{235}$U, $^{236}$U, and $^{238}$U using Multi-
       Collector Inductively Coupled Plasma Spectrometry (MC ICP-
       MS)............................................................................................................ 02
   2.2 Measurements of $^{233}$U (and $^{236}$U) using Accelerator Mass
       Spectrometry (AMS)............................................................................ 03
   2.3 Measurements of $^{232}$U using Alpha-Spectrometry............................ 05
3.0 Materials and Methods................................................................................ 05
   3.1 MC ICP-MS measurements of $^{234}$U, $^{235}$U, $^{236}$U and $^{238}$U.......... 05
   3.2 Isolation and Purification of Uranium Isotopes for
       Measurement by AMS and Alpha-Spectrometry Techniques.......... 05
   3.3 Preparation of AMS Targets.............................................................. 07
   3.4 Electrodeposition of Uranium on Stainless Steel Plates.................. 08
4.0 Accuracy and Precision .............................................................................. 08
Acknowledgment............................................................................................... 09
References ....................................................................................................... 09

List of Tables

Table 1 Isotopes of uranium used in nuclear and/or environmental
applications........................................................................................................ 01
Table 2. List of the major alpha particle energies and abundances (%) of
uranium isotopes................................................................................................ 06
Table 3. Uranium isotopic measurements of reference solution (U010)
from the New Brunswick Laboratory (NBL)................................................... 10
Table 4 Uranium isotopic composition of LLNL Low Enriched Uranium ....... 11

List of Figures

Figure 1. Outline of the essential components of a Magnetic Sector, Multi-
Collector Inductively Coupled Plasma Mass Spectrometer............................ 02
Figure 2. Heavy element beam line at the Center for Accelerator Mass
Spectrometry (CAMS), Lawrence Livermore National Laboratory ............. 03
Figure 3a. Measured versus expected $^{233}$U content of standard AMS
targets............................................................................................................... 04
Figure 3b. Measured versus expected $^{236}$U content of standard AMS
targets............................................................................................................... 04
Abstract

This report describes a standardized methodology used by researchers from the Center for Accelerator Mass Spectrometry (CAMS) (Energy and Environment Directorate) and the Environmental Radiochemistry Group (Chemistry and Materials Science Directorate) at the Lawrence Livermore National Laboratory (LLNL) for the full isotopic analysis of uranium from solution. The methodology has largely been developed for use in characterizing the uranium composition of selected nuclear materials but may also be applicable to environmental studies and assessments of public, military or occupational exposures to uranium using in-vitro bioassay monitoring techniques. Uranium isotope concentrations and isotopic ratios are measured using a combination of Multi Collector Inductively Coupled Plasma Mass Spectrometry (MC ICP-MS), Accelerator Mass Spectrometry (AMS) and Alpha Spectrometry.

1.0 Introduction

The most abundant isotopes of uranium in nature (e.g., rocks, soil, surface waters) are uranium-234 (\(^{234}\text{U}\)), uranium-235 (\(^{235}\text{U}\)) and uranium-238 (\(^{238}\text{U}\)) and, to a lesser and more variable extent, uranium-236 (\(^{236}\text{U}\)). Uranium is also used as a source material for nuclear weapons, as fuel for nuclear reactors and in a number of other industrial applications. All the naturally occurring isotopes of uranium are long-lived and, with the exception of \(^{236}\text{U}\) at low relative isotopic abundance, can easily be measured with high precision and accuracy using mass spectrometric techniques (Table 1).

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Natural Abundance (Atom %)</th>
<th>Half-life (years)</th>
<th>Origin</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{232}\text{U})</td>
<td>-</td>
<td>68.9</td>
<td>(^{233}\text{U}) (n,2n); (^{232}\text{Th}) (α,4n); product of (^{232}\text{Pa}) or (^{236}\text{Pu}) decay</td>
</tr>
<tr>
<td>(^{233}\text{U})</td>
<td>-</td>
<td>(1.592 \times 10^5)</td>
<td>Product of (^{233}\text{Pa}) decay</td>
</tr>
<tr>
<td>(^{234}\text{U})</td>
<td>0.005</td>
<td>(2.455 \times 10^7)</td>
<td>Daughter product of the (^{238}\text{U}) decay chain</td>
</tr>
<tr>
<td>(^{235}\text{U})</td>
<td>0.72</td>
<td>(7.038 \times 10^8)</td>
<td>primordial</td>
</tr>
<tr>
<td>(^{236}\text{U})</td>
<td>ca. (1-1000 \times 10^{-14})</td>
<td>(2.342 \times 10^5)</td>
<td>(^{235}\text{U}) (n,y); product of (^{236}\text{Np}) or (^{240}\text{Pu}) decay</td>
</tr>
<tr>
<td>(^{238}\text{U})</td>
<td>99.27</td>
<td>(4.468 \times 10^9)</td>
<td>primordial</td>
</tr>
</tbody>
</table>
At Livermore, very precise and accurate measurements of the high abundance isotopes of uranium in solution (in the 10’s of ng total U ml\(^{-1}\) range) are routinely performed by Multi-Collector Inductively Coupled Plasma Spectrometry (MC ICP-MS). No additional sample treatment is usually required for solutions prepared from relatively pure nuclear materials other than sample dilution and direct aspiration into the instrument. Low-level detection and measurement of the minor abundance (predominantly manmade) isotopes of uranium such as uranium-232 (\(^{232}\text{U}\)), uranium-233 (\(^{233}\text{U}\)) and, in some cases \(^{236}\text{U}\), usually requires some form of pre-concentration and removal of potential isobaric and/or radiometric interferences. In this latter case, isotopes of uranium are separated and purified from solution using standardized techniques based on ion exchange chromatography.

2.0 Uranium Isotope Measurement Techniques

Uranium isotope concentrations and isotopic ratios are measured using a combination of Multi Collector Inductively Coupled Plasma Mass Spectrometry (MC ICP-MS) \([^{234}\text{U},^{235}\text{U},^{(236}\text{U})\) and \(^{238}\text{U}\)], Accelerator Mass Spectrometry (AMS) \([^{233}\text{U},^{236}\text{U}]) and Alpha Spectrometry (\(^{232}\text{U}\)).

2.1 Measurements of \(^{234}\text{U},^{235}\text{U}, (^{236}\text{U})\) and \(^{238}\text{U}\) using Multi-Collector Inductively Coupled Plasma Spectrometry (MC ICP-MS).

A schematic diagram of a typical MC ICP-MS system with a motorized detector positioning system is shown in Fig.1.

Figure 1. Outline of the essential components of a Magnetic Sector, Multi-Collector Inductively Coupled Plasma Mass Spectrometer.
The Environmental Chemistry Group within Chemistry and Materials Science Directorate at Livermore utilizes an IsoProbe MC ICP-MS (GV Instruments, UK) for a wide range of isotopic measurements of solutions, including isotopic analyses of uranium. This magnetic sector, mass spectrometry system allows for the simultaneous measurement of different ion mass beams and is ideally suited to making very precise and accurate measurements of uranium isotopes in solution. The total efficiency of the mass spectrometer, as defined by the number of ions detected relative to the total atoms present in the sample, is greater than 1%. The analyzer array in the collector block contains 9 faraday detectors and 8 pulse counting detectors. The pulse counters include 6 channeltrons which may be positioned for simultaneous multi-isotope ion collection, one fixed high-mass channeltron and a Daly detector. Ions along the axial beam are filtered by a wide aperture retarding potential (WARP) filter and measured on the Daly detectors for high abundance sensitivity applications (e.g., low-level measurements of $^{236}$U in presence of large quantities of $^{235}$U). All of the faraday cups may be positioned for simultaneous multi-collection of higher ion currents. Simultaneous multi-collection allows isotope ratios to be measured with high precision because the fluctuations in ion beam intensity are seen by each detector at the same time.

A Cetac Aridus desolvating nebulizer coupled with a Cetac ASX-100 autosampler is used for sample introduction. Data collection, including sample intake equilibration, takes about 15 minutes per sample. Mass bias correction factors are determined from periodic analyses of natural uranium standards (NIST SRM 4321C). The relative detector gain factor used to correct the Daly signal for comparison with the Faraday output system is determined using the same set of standards. The NBL uranium standard, U010, along with other internal standard uranium isotopic stock solutions is also analyzed routinely for quality assurance purposes.

### 2.2 Measurements of $^{233}$U (and $^{236}$U) using Accelerator Mass Spectrometry (AMS)

The systems configuration for the heavy beam line at CAMS is shown in Fig. 2.

**Figure 2.** Heavy element beam line at the Center for Accelerator Mass Spectrometry (CAMS), Lawrence Livermore National Laboratory.
One of the most sensitive techniques available for routine measurements of $^{233}$U (and $^{236}$U) in the presence of high abundance isotopes of uranium is Accelerator Mass Spectrometry (AMS). The lower limit of detection for AMS measurements of $^{233}$U at CAMS is around $10^6$ atoms. AMS sensitivity is primarily determined by the number of $^{233}$U ions counted during the measurement cycle and, consequently, on the number $^{233}$U atoms present in the sample target as well as the background count rate and the data acquisition time. However, there are practical limitations on sample loading depending on the inherent isotopic composition of the sample and limitations on the dynamic measurement range of the AMS data acquisition system. The AMS systems configuration at CAMS allows for quasi-continuous measurements of ions of isotopes of interest (e.g., $^{233}$U) compared with that of a reference isotope (e.g., $^{236}$U) over a dynamic measurement range of about 5 orders of magnitude, the upper limit being controlled by the count rate capacity of the detector system. We have determined that the maximum loading capacity of targets is equivalent to $\sim 10^{11}$ atoms of the reference isotope. Above this limit, increases in the detector count rate and associated dead-time corrections for the reference isotope become unacceptably high to provide reliable measurements of the isotopes of interest. For analysis of uranium solutions prepared from nuclear materials, MC ICP-MS measurements are used to determine the bulk isotopic composition of the material prior to the preparation of AMS sample targets. A verified quantity of uranium can then be processed into targets for AMS measurements ($^{233}$U or $^{236}$U) or, for $^{232}$U measurements, electroplated on stainless steel (S/S) discs and counted by alpha spectrometry.

![Figure 3a](image1.png)  
**Figure 3a.** Measured versus expected $^{233}$U content of standard AMS targets.  

![Figure 3b](image2.png)  
**Figure 3b.** Measured versus expected $^{236}$U content of standard AMS targets.
2.3 Measurements of $^{232}\text{U}$ using Alpha-Spectrometry

$^{232}\text{U}$ is an alpha emitter with a relatively short half-life (69 years) and is usually present in uranium source materials at very low concentrations (e.g., $^{232}\text{U}/^{238}\text{U} < 10^{-10}$). For alpha-spectrometric measurements, uranium isotopes are isolated and purified by double ion-exchange and up to 60 $\mu$g of total uranium electroplated onto polished stainless steel plates. The source plates are then counted on dedicated low-background Si surface barrier detectors for a maximum of 14 days. The detectors are coupled to an Octete™ PC alpha spectrometry system.

The initial sample purification step is usually performed on prepacked UTEVA anion exchange columns supplied by Eichrom® Technologies Inc, USA. The UTEVA resin is composed of dipentylpentane phosphonate adsorbed on an inert polymeric support and has a very high affinity for uranium (VI). After the UTEVA column separation, uranium isotopes are further purified on AG®1-X8 (BioRad Laboratories, USA) to help remove final traces of polonium-210 ($^{210}\text{Po}$) and thorium-228 ($^{228}\text{Th}$). Internal references solutions and reagent blanks are prepared and analyzed with each batch of samples. For low-level measurements, alpha source plates should ideally be stored for several months and re-counted to check for in-growth of characteristic daughter products.

$^{232}\text{U}$ has a well defined alpha energy decay peaks at 5.32 and 5.362 MeV providing good spectral resolution from the more abundant isotopes of uranium. However, high-energy straggling effects will produce spurious counts in the $^{232}\text{U}$ region-of-interest at high mass alpha plate loadings (Table 2). As a trade off between spectral resolution and $^{232}\text{U}$ detection sensitivity, the alpha plate loadings are typically limited to about 150-200 $\mu$g of total uranium.

3.0 Materials and Methods

Uranium isotopes in solution are routinely handled in disposable acid washed Teflon® or polyethylene vials. Chemicals are prepared new from ultra-pure reagent stocks and <18 Megohm high-purity deionized water. The use of glass is avoided wherever possible.

3.1 Measurements of $^{234}\text{U}$, $^{235}\text{U}$, ($^{236}\text{U}$) and $^{238}\text{U}$ using Multi-Collector Inductively Coupled Plasma Spectrometry (MC ICP-MS)

MC ICP-MS measurements of $^{234}\text{U}$, $^{235}\text{U}$, ($^{236}\text{U}$) and $^{238}\text{U}$ are usually performed on acid dilutions of the primary stock solutions as received or prepared by total dissolution of solids. A subaliquot containing an estimated (or known) amount of uranium is transferred to a 30 mL Teflon® vial, to which a small amount of concentrated HNO$_3$ is added. The sample is then taken to dryness and rehydrated in 1 mL of a 2% ultra-pure HNO$_3$ solution. The solution is then transferred with repeated washings to an acid-washed polypropylene conical tube using a plastic syringe outfitted with a 0.45 micrometer syringe filter. The filtered sample containing 3 mL of 2% ultra-pure HNO$_3$ is now ready for analysis.
Table 2. List of the major alpha particle energies and abundances (%) of uranium isotopes.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Alpha Particle Energy (Mev)</th>
<th>Alpha Particle Abundance (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{232}\text{U}$</td>
<td>5.320</td>
<td>68</td>
</tr>
<tr>
<td></td>
<td>5.263</td>
<td>32</td>
</tr>
<tr>
<td>$^{233}\text{U}$</td>
<td>4.821</td>
<td>83.4</td>
</tr>
<tr>
<td></td>
<td>4.778</td>
<td>14.6</td>
</tr>
<tr>
<td></td>
<td>4.722</td>
<td>1.5</td>
</tr>
<tr>
<td>$^{234}\text{U}$</td>
<td>4.773</td>
<td>72</td>
</tr>
<tr>
<td></td>
<td>4.722</td>
<td>28.0</td>
</tr>
<tr>
<td>$^{235}\text{U}$</td>
<td>4.597</td>
<td>4.6</td>
</tr>
<tr>
<td></td>
<td>4.556</td>
<td>3.7</td>
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<td></td>
<td>4.502</td>
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<td></td>
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<td></td>
<td>4.396</td>
<td>57</td>
</tr>
<tr>
<td></td>
<td>4.366</td>
<td>18</td>
</tr>
<tr>
<td></td>
<td>4.344</td>
<td>1.5</td>
</tr>
<tr>
<td></td>
<td>4.323</td>
<td>3</td>
</tr>
<tr>
<td></td>
<td>4.216</td>
<td>5.7</td>
</tr>
<tr>
<td>$^{236}\text{U}$</td>
<td>4.493</td>
<td>74</td>
</tr>
<tr>
<td></td>
<td>4.443</td>
<td>26</td>
</tr>
<tr>
<td>$^{238}\text{U}$</td>
<td>4.195</td>
<td>77</td>
</tr>
<tr>
<td></td>
<td>4.147</td>
<td>23</td>
</tr>
</tbody>
</table>

3.2 Isolation and purification of uranium isotopes for measurement by AMS and alpha-spectrometry

A subaliquot containing an estimated (or known) amount of uranium is transferred to a 30 mL Teflon® vial, to which a small amount of concentrated HNO$_3$ is added. The sample is then taken to dryness and rehydrated in 10-15 mL of a 2.5 M HNO$_3$-1M purified aluminum nitrate [Al(NO$_3$)$_3$] solution. The sample is then passed through a UTEVA (50-100 μm mesh) column which is conditioned with 6 mL of 3 M HNO$_3$. The transfer is completed by rinsing the sample vial with a further two 1 mL aliquots of 3 M HNO$_3$ and the resin washed with an additional four 5 mL aliquots (20 mL total) of 3 M HNO$_3$ taking care to rinse down the walls of the funnel and column stem. The column resin is then washed with four 5 mL aliquots (20 mL total) of a 5 M hydrochloric acid–0.05 M oxalic acid solution and the uranium isotopes eluted using 20 mL of 0.02M H$_2$SO$_4$. The uranium fraction is collected in a 30 mL Teflon® vial. A plastic watch glass is placed on top of the vial and the sample slowly evaporated to dryness. Care is taken not to bake the dried residue on the inside of the vial.

The dried eluant from the UTEVA column is further purified by anion exchange using an AG® 1-X8 column preconditioned with 20 mL of 10 M HCl.
The dry sample (in the 30 mL Teflon® vial) is rehydrated with 3-5 mL of 10 M HCl and loaded onto the column resin. The sample container and ion-exchange column are rinsed with an additional four 5 mL aliquots (20 mL total) of 10 M HCl taking care to ensure to rinse down the walls of the funnel and column stem. Additional rinse steps are included for those samples known to contain high concentrations of $^{210}$Po. The resin is then washed with a small amount (1-2 mL) of 10 M HCl and uranium isotopes eluted with 20 mL of 0.5M HCl. Again, a plastic watch glass is placed on top of the vial and the sample slowly evaporated to dryness. Care is taken not to bake the dried residue on the inside of the vial.

Process blanks, sample replicates, and laboratory control standards are treated in exactly the same manner and analyzed with each batch of samples processed.

3.3 Preparation of AMS Targets

Rehydrated subaliquots from the double column cleanup procedure are ready for processing into AMS targets. For each sample, 0.3 g of a 1 mg g$^{-1}$ iron (Fe) solution is first dispensed into a separate 15 mL acid-washed polypropylene conical bottom disposable centrifuge tube. The sample residue is rehydrated using 2 mL of 3 M HNO$_3$ solution and analytically transferred to the 15 mL centrifuge tube (containing the 0.3 mg of Fe) using repeated (4 x) washings with 2 mL of HNO$_3$ solution. The final volume of sample solution at this stage of sample preparation should be approximately 10 mL. Uranium isotopes are then precipitated from solution on iron hydroxide [Fe(OH)$_3$] by addition of 2–4 mL of a 1:1 ammonium hydroxide deionized water solution (NH$_4$OH·xH$_2$O) and the sample allowed to stand overnight. The sample Fe precipitate containing the uranium isotopes is then recovered by centrifugation, the resulting sample pellet carefully washed with 2 mL of ultra-pure water (the residual NH$_4$OH is sufficient to keep the solution basic) and the material transferred to a quartz crucible. The Fe sample solution is slowly taken to dryness by placing the crucible under an infra-red heat lamp. After drying, the crucible is capped and transferred to a high-temperature oven and baked 800°C. The crucible is removed from the oven and allowed to cool, approximately 3 mg of niobium metal added and the mixture homogenized by mixing using the end of a #60 gauge drill stem. The dry sample powder is then transferred to a clean-labeled aluminum AMS sample holder or cathode drilled to 0.006″ deep with a #60 gauge drill bit, and pounded into a target until the material is compact (i.e., until the surface has a shiny, metallic luster). Each sample cathode is stored in a separate labeled holder until analysis by AMS.

The sample introduction system on the AMS may contain as many as 64 targets on a rotating wheel. Of these targets, a series of background measurements are routinely performed on samples prepared from the target matrix materials (i.e., Fe and Nb) along with standard targets that also contain various mixtures of natural uranium, and spike concentrations of National Institute of Standards and Technology (NIST) traceable $^{233}$U and $^{236}$U standard solutions purchased from either the New Brunswick Laboratory (NBL) (Argonne, IL) or the Isotope Products Laboratories (IPL) (Valencia, CA). In preparing standard AMS targets, an effort is made
to mimic the uranium composition of test samples to more appropriately assess the effects of measurement backgrounds, mass bias, abundance sensitivity and/or the dynamic measurement range on the quality and reliability of the analyses.

3.4 Electrodeposition of Uranium on Stainless Steel Plates

Subaliquots from the double column cleanup procedure are also used in preparing electroplated sources for alpha-spectrometric measurements of $^{232}$U ($^{232}$U/$^{238}$U). The electrodeposition apparatus consists of a Teflon® base assembly and cover plate, a platinium (Pt) wire anode, and a plating cell consisting of a 30 mL polyethylene vial. The base assembly screws down on a polished S/S alpha discs (or plates) which also serve as the cathode. A suitably sized sample aliquot (nominally containing about 60 μg of uranium) is transferred to a 30 mL Teflon® vial, to which a small amount of concentrated HNO$_3$ is added and the sample slowly evaporated to dryness. Approximately 1 mL of concentrated sulfuric acid (H$_2$SO$_4$) is added to the Teflon® vial and the sample heated on a hot plate until evolution of white fumes. The sample is then cooled to room temperature, and the walls of the vial carefully rinsed down with 2 mL of 1 M H$_2$SO$_4$. Two drops of a 0.1% methyl red indicator solution are added to the Teflon® vial and the contents of the vial analytically transferred into an electrodeposition cell using three additional 2 mL rinses of 1 M H$_2$SO$_4$. The electrolyte solution is then adjusted to pH ≤6.2 (yellow endpoint) by dropwise addition of concentrated ammonia solution (NH$_4$OH) with constant swirling and back titrated to pH ≤4.4 (red endpoint) by dropwise addition of 1 M H$_2$SO$_4$. Two drops of 1 M H$_2$SO$_4$ are added in excess. At this stage, the electrodeposition cell is fully assembled by attaching the platinum anode to the Teflon® cover plate and the power turned on. Samples are electroplated at 1 mA for approximately 60 minutes. Prior to turning off the power, a small quantity of concentrated NH$_4$OH (app. 1 ml) is introduced into the cell (from the vent orifice on the Teflon® cover plate) and the current allowed to flow for an additional 60 seconds. The electrodeposition apparatus is then disassembled and the sample solution discarded. The S/S disc is then rinsed consecutively with a 1:1000 solution of NH$_4$OH·high purity deionized water (0.5 mL of NH$_4$OH in 500 mL of high purity water) and acetone, and allowed to air dry on clean absorbent paper. For low-level $^{232}$U measurements the S/S alpha discs are flamed to incandescent red in order to remove traces of $^{210}$Po by volatilization. The alpha discs are stored in separate labeled polycarbonate holders until analysis by alpha-spectrometry. The time between sample isolation and purification, and counting is minimized to reduce the number of counts (and applicable corrections) in the sample spectrum from ingrown of $^{232}$U daughter products.

4.0 Accuracy and Precision

The MC ICP-MS instrument is routinely calibrated with certified uranium isotopic standards available from the New Brunswick Laboratory (NBL). Table 3 contains the results of quality control (instrument setup and
calibration) analyses performed at different times on NBL U010, a 10% 235U LEU isotopic standard. The precision for 234U/238U, 235U/238U and 236U/238U atom ratio measurements of this material was 0.09%, 0.04% and 0.08%, respectively. Multiple measurements of the same solution (N=5, for the measurements tabulated) yielded improved precision by factors of 2 to 3 fold. The respective measurement bias for each set of isotope ratio measurements was -0.4%, 0.01% and 0.77%, respectively.

An example set of data on the uranium isotopic composition of selected LLNL reference solutions is shown in Table 4. These data were derived from a combination of MC ICP-MS, AMS and alpha spectrometric detection and measurement using the standardized preparative methods outlined in this report.

References


Wong K.M., T. Jokela, V.E. Noshkin (1994). Radiochemical procedures for Analysis of Pu, Am, Cs, and Sr in water, soil, sediments and biota samples. Lawrence Livermore National Laboratory, UCRP-ID-116497, 19pp + appendixes

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Table 3. Uranium isotopic measurements of reference solution (U010) from the New Brunswick Laboratory (NBL).

<table>
<thead>
<tr>
<th>Quality Control Sample</th>
<th>Multicollector Inductively Coupled Plasma Mass Spectometry</th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>234U/238U</td>
<td>235U/238U</td>
<td>236U/238U</td>
</tr>
<tr>
<td>U010-1</td>
<td>0.000054410± 0.00000005</td>
<td>0.0101290± 0.00000070</td>
<td>0.00006935± 0.00000008</td>
</tr>
<tr>
<td>U010-2</td>
<td>0.000054460± 0.00000006</td>
<td>0.0101360± 0.00000070</td>
<td>0.00006928± 0.00000006</td>
</tr>
<tr>
<td>U010-3</td>
<td>0.000054420± 0.00000005</td>
<td>0.0101420± 0.00000020</td>
<td>0.00006922± 0.00000006</td>
</tr>
<tr>
<td>U010-4</td>
<td>0.000054440± 0.00000006</td>
<td>0.0101440± 0.00000060</td>
<td>0.00006940± 0.00000007</td>
</tr>
<tr>
<td>U010-5</td>
<td>0.000054370± 0.00000005</td>
<td>0.0101450± 0.00000045</td>
<td>0.00006943± 0.00000006</td>
</tr>
<tr>
<td>Mean &amp; standard deviation (N=5)</td>
<td>0.000054433± 0.00000002</td>
<td>0.0101415± 0.00000016</td>
<td>0.00006933± 0.00000003</td>
</tr>
<tr>
<td>Certified value (U010)</td>
<td>0.00005466± 0.00000056</td>
<td>0.01014± 0.000010</td>
<td>0.0000688± 0.00000078</td>
</tr>
<tr>
<td>NEW NBL value</td>
<td>(0.00005442± 0.00000007)</td>
<td>(0.01014± 0.00000010)</td>
<td>(0.00006929± 0.00000012)</td>
</tr>
<tr>
<td>Measurement Bias c/w NBL value</td>
<td>-0.40%</td>
<td>0.01%</td>
<td>0.77%</td>
</tr>
</tbody>
</table>
Table 4  Uranium isotopic composition of LLNL Low Enriched Uranium.

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Multi-Collector Inductively Coupled Plasma Mass Spectrometry</th>
<th>Accelerator Mass Spectrometry</th>
<th>Alpha Spectrometry</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$^{234}\text{U}^{238}\text{U}$</td>
<td>$^{235}\text{U}^{238}\text{U}$</td>
<td>$^{236}\text{U}^{238}\text{U}$</td>
</tr>
<tr>
<td>LLNL 3A</td>
<td>0.0004260 ± 0.0000006</td>
<td>0.04661 ± 0.00003</td>
<td>0.000155 ± 0.000005</td>
</tr>
<tr>
<td>LLNL 4A</td>
<td>0.0003133 ± 0.0000006</td>
<td>0.04661 ± 0.00003</td>
<td>0.000035 ± 0.000006</td>
</tr>
<tr>
<td>LLNL 10A</td>
<td>0.0003199 ± 0.0000005</td>
<td>0.04664 ± 0.00003</td>
<td>0.000032 ± 0.000006</td>
</tr>
<tr>
<td>LLNL 14A</td>
<td>0.0004136 ± 0.0000001</td>
<td>0.04666 ± 0.00001</td>
<td>0.000180 ± 0.000001</td>
</tr>
</tbody>
</table>

$^a$ Isotope ratio data shown for the error-weighted average and standard deviation of 5 separate analyses

$^b$ Error-Weighted average and standard deviation of 5 separate sample analyses

$^c$ Error-weighted average and standard deviation of 2 separate sample analyses