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Uranium isotopic analysis for the nuclear industry

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Keywords

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Introduction

This application note describes the analytical performance of the Thermo Scientific™ Neptune XT™ Multicollector ICP-MS™ for measuring uranium isotopes from low enriched uranium. The methodology follows a standard test protocol that is used within nuclear industry.

ASTM C 1477 – 08 (ASTM International 2009) describes an analytical methodology for isotopic measurements of low enriched uranium, natural uranium and depleted uranium. This protocol is currently used within uranium enrichment plants to determine the ^{235}U content of feed and product materials. Uranium solutions, obtained from the hydrolysis of UF_6, are measured using multicollector inductively-coupled plasma mass spectrometry (MC-ICP-MS). The data are corrected for blanks, hydride on ^{236}U and then for instrumental bias effects by measurement of a certified reference material (CRM). The protocol is optimized for productivity, whilst achieving 'fit-for purpose' relative uncertainties of better than 0.1% for ^{235}U. ^{234}U and ^{236}U are measured with lower precision (better than 1%), as the minor isotopes are reported for information purposes only. Thirty samples and quality controls can be measured within 8 hours; the sequence and data corrections are fully automated. The analytical performance of the methodology is dependent on the stability of the instrument (mass bias and ion counter yield).

Instrumentation

A Neptune XT MC-ICP-MS was equipped with a central SEM and RPQ. The RPQ lens can improve abundance sensitivity by a factor of ten (*m/z* 237.05 / ^{238}U was measured as 0.35 ppm). A Compact Discrete Dynode (CDD) dynode type SEM was used for measurement of ^{234}U; this detector type has similar performance characteristics to the full-sized SEM.

The major isotopes ^{235}U and ^{238}U were measured on Faraday cups. The same system is capable of measuring high-precision isotope ratios from across the periodic table, including Li, Fe, Sr, Nd, Hf and Pb.

A Cetac ASX-112 FR autosampler was used to collect sample solutions from a rack of up to 42 vials, with an additional rack available for standards, quality control and tune solutions. Blanks were measured directly from the flushed rinse station.

In wet plasma with standard cones and PFA spray chamber the sensitivity was 44 V/ppm, with less than 70 ng uranium consumed per analysis.

Outline of methodology

This test methodology follows the key protocols from ASTM C 1477 – 08. The samples were diluted to 100 ng/g uranium in a high purity 2 wt.% HNO_3 matrix. The instrument was ready within 1 hour of igniting the plasma.

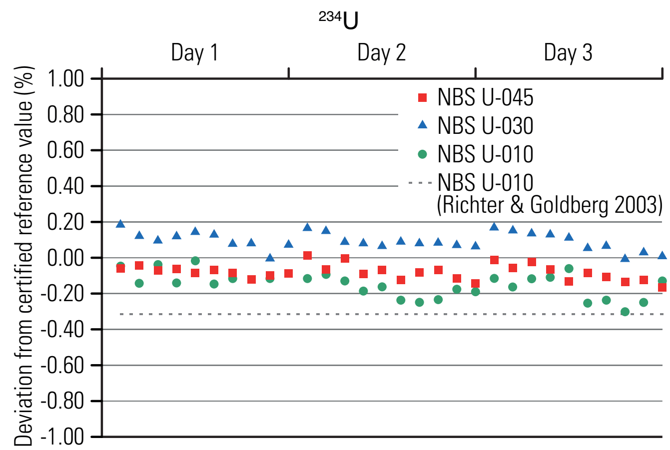
Test protocol

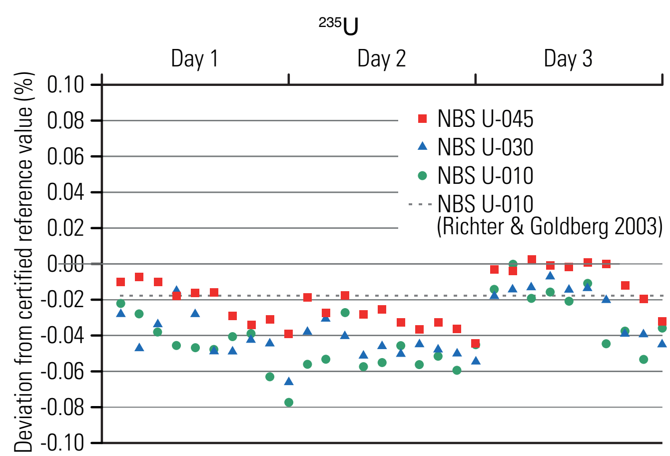
The ‘samples’ in this test protocol were the certified reference materials: NBS U-045, U-030 and U-010 (New Brunswick Laboratory, Argonne, IL, USA). The standard was IRMM-187 (Institute for Reference Materials and Measurements, Geel, Belgium); this was measured only once at the start of each sequence. On each of 3 consecutive days, a test sequence was run with 30 ‘samples’ (alternating ten measurements of each of NBS U-045, U-030 and U-010). The plasma was automatically switched off after each test sequence.

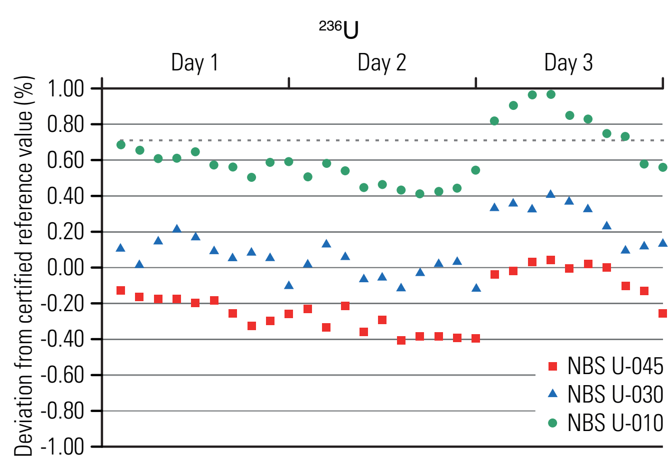
Results

The data for each of the standards and isotopes are plotted in Figures 1–3, and statistics are summarized in Table 1. It can be seen that for each isotope the scatter of measured data is small and within the range of uncertainty of the certified reference materials.

More recent and precise reference values from Richter & Goldberg (2003) are plotted for comparison, for each isotope the measured data plot closer to these lines than to the zero of the certified reference value. The data show the stability of the instrument over 8 hours, with respect to mass bias and ion counter yields. The analytical performance is well within that required for application in the nuclear fuel industry.







Figures 1–3. The scatter of data for the measurements is small compared to the target precisions, and compared to the uncertainty of the certified reference values (the certified reference uncertainty for NBS U-010 approximately covers the full plot area in each graph).

Table 1. Mean and external reproducibility for each measured standard, relative standard deviation (1σ RSD) from 3 consecutive analytical sessions (n=30 for each standard).

		^{234}U (wt.%)	^{235}U (wt.%)	^{236}U (wt.%)
NSB U-045	mean	0.03806%	4.4590%	0.02738%
	RSD	0.04%	0.01%	0.14%
NB US -030	mean	0.01871%	3.0076%	0.02026%
	RSD	0.05%	0.02%	0.15%
NBS U-010	mean	0.00531%	0.9908%	0.00680%
	RSD	0.07%	0.02%	0.16%

Conclusions

The data show that the Neptune XT is a MC-ICP-MS that is easily capable of measuring low enriched uranium materials in accordance with the ASTM C 1477 – 08 protocol, and that it is highly stable with respect to mass bias and ion counter yield. The Neptune XT is the only MC-ICP-MS supported by a global network of dedicated service engineers, vital for continuous industry critical operations.

Optional configurations

The Neptune XT MC-ICP-MS can be configured with a variety of options so that sample quantities from fg to µg can be analyzed.

- The *Jet Interface* option offers exceptional ICP-MS sensitivity, allowing small samples to be measured on Faraday cups, or the smallest samples to be measured on ion counters with significantly improved counting statistics.
- Optional collector configurations allow ^{238}U and ^{235}U to be measured on either Faraday cups or ion counters, allowing the smallest samples to be analyzed.
- The dual RPQ option allows both ^{236}U and ^{234}U to be measured with improved abundance sensitivity. This enables precise quantification of ^{234}U in the presence of intense ^{235}U beams.
- Using a mix of 10^{11}, 10^{12} and 10^{13} ohm amplifiers, it is possible to measure the minor isotopes on Faraday cups, avoiding the uncertainties from ion counter yield measurements and yield drift. The Thermo Scientific™ 10^{13} Ohm Amplifier Technology™ offers improved signal to noise ratios for small ion beam intensities, and can be connected to any Faraday cup via a unique relay matrix.
- The Neptune XT MC-ICP-MS can be adapted for glove box or fume hood containment where required.

Figure 4. Neptune XT Multicollector ICP-MS.

Related products

The Thermo Scientific™ Triton XT™ is a multi-collector thermal ionization mass spectrometer (TIMS) that is well established within the nuclear industry. It can optionally be equipped with the same ion counting configurations that are available on the Neptune XT MC-ICP-MS. The Triton XT TIMS now offers capabilities for the 'modified total evaporation' methodology, for improved minor isotope analysis.

Two high-precision analytical instruments cater for the needs of uranium isotope analysis within the nuclear industry, all of which are supported by a global network of dedicated service engineers.

Figure 5. Triton XT Thermal Ionization MS.

References

1. Richter, S. Goldberg, S.A. 2003. *Int. J. Mass Spectrom.* Vol. 229, pp. 181 – 197.
2. ASTM International 2009. *ASTM C1477 - 08 Standard Test Method for Isotopic Abundance Analysis of Uranium Hexafluoride and Uranyl Nitrate Solutions by Multi-Collector, Inductively Coupled Plasma-Mass Spectrometry.* ASTM International, West Conshohocken, PA, USA.

Find out more at **thermofisher.com/MC-ICP-MS**

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