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Determination of Trace Elements in Uranium by HPLC-ID-ICP-MS

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Determination of Trace Elements in Uranium by ID-HPLC-ICP-MS

1 Executive Summary

This report covers the FY 16 effort for the HPLC-ID-ICP-MS methodology 1) sub-method validation for the group I&II elements, 2) sub-method stood-up and validation for REE, 3) sub-method development for the transition element, and 4) completion of a comprehensive SOP for three families of elements.

2 Individual Project Status Reports

2.3 HPLC Methodology

- LANL.16.005
- POP: 4/1/16 – 3/31/17
- PI: Ning Xu, Lav Tandon

2.3.1 Final Report

Previously, in FY15, LANL stood-up the HPLC-ID-ICP-MS method for the separation and determination of group I&II elements in a uranium matrix [1]. We also completed the HR-ICP-MS method development for the measurement of rare earth elements (REE). The overarching accomplishment for FY16 is that we completed the NTNFC requested task of standing up the HPLC-ID-ICP-MS method for analyzing trace impurities in uranium matrix and further tested method during the Trace Elements in Uranium Benchmarking Study (U Benchmarking study).

We have evaluated three HPLC separation methods for group I&II, rare earth, and transition elements. Each method consists of a different combination of separation column, eluents, elution regime, and detection system. We have also developed 16 individual elemental IDMS methods for the HR-ICP-MS measurements. We trained HPLC and HR-ICP-MS instrument operators and generated a comprehensive SOP (submitted separately from this document) to cover the procedures of isotope dilution spiking, HPLC separation (three sub separation methods), HR-ICP-MS measurements, data reduction, and QA/QC compliance. This SOP was used as guidance for the 2017 U Benchmarking Study [2]

The HPLC-ID-ICP-MS method was validated via a certified reference material (CRM), the Canadian based uranium ore (CUP-2), for the elements of interest. Conceptually, using HPLC-ID-ICP-MS method to separate individual trace element and followed by analyzing them by the ID-ICP-MS method improves measurement uncertainties caused by the external calibration and the isobaric/polyatomic interferences. Our two-year study has demonstrated that the methodology indeed successfully increased the measurement accuracy and precision for many analytes compared to traditional external calibration ICP-MS methodology. In addition, dissolved uranium samples were able to be separated by the HPLC directly without the need for U matrix removal at U concentrations as high as 1000 µg/mL in a solution.

2.3.1.1 Group I & II elements

The HPLC-ID-ICP-MS method for the separation and determination of group I & II elements in U matrix has been detailed in the FY15 annual final report [1]. Although initial efforts showed that the chromatographic separation of Ba, Ca, K, Mg, and Sr was successful (**Figure 1**) without the need for removing U matrix prior to the HPLC separation, the mass spectrometry analyses of Ca, K, and Mg proved to be difficult for two reasons: 1) the presence of gaseous species (e.g. Ar, Kr, Xe, O_2, N_2, CO_2) in the ICP plasma resulting in polyatomic or isobaric interferences, and 2) the low abundance of available isotopes hindering the HR-ICP-MS measurement in the medium/high resolution mode.

Barium and Sr benefit from the HPLC-ID-ICP-MS method. The primary interferences for these two elements are Xe, Rb, and Kr, which can be either removed by the HPLC separation or discounted unless the analytes are at ultra-low concentrations in the sample. Magnesium (^{24}Mg) can also be measured using this method, but medium resolution mode must be used for the HR-ICP-MS measurement to reduce the interference caused by the $^{12}C_2^+$. Medium mode provides higher resolution, but at the cost of decreased sensitivity.

Calcium (^{40}Ca) suffers from the interference of $^{40}Ar^+$, which is the ICP carrier gas. Although other Ca isotopes such as ^{42}Ca, ^{43}Ca, ^{44}Ca, and ^{48}Ca are available, the abundance is too low (<4% of Ca) to be analyzed in medium resolution mode by HR-ICP-MS. Similarly, potassium (^{40}K and ^{41}K) is affected due to its proximity to the large $^{40}Ar^+$ peak and polyatomic molecules such $^{40}Ar^1H^+$.

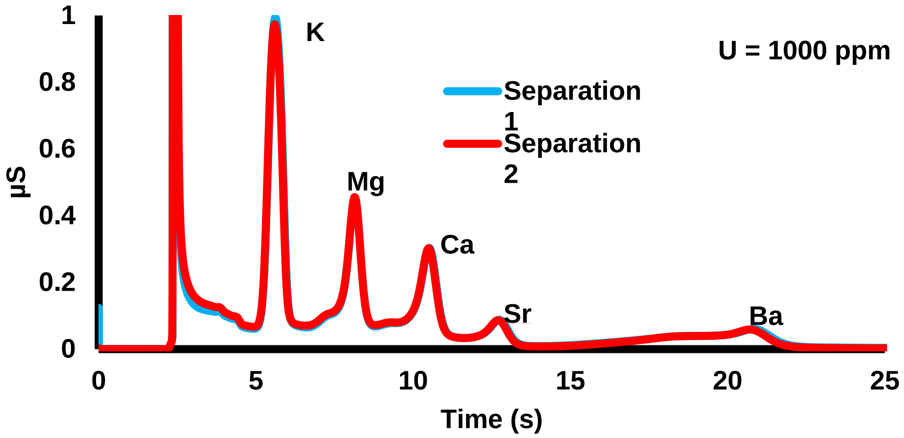

Figure 1. Chromatograph of separated group I & II elements in a uranium matrix. 10 µL injection at 0.3 µL/min

To develop the ID-ICP-MS method for the group I&II element measurement, a multi-element solution containing Ba, Mg, and Sr was prepared gravimetrically using NIST certified single element standards. This solution was mixed sequentially with ORNL single stable isotopes. The stable isotope concentration for each element was quantified by reverse-IDMS (rIDMS). The accuracy, when compared with the expected concentrations, and precision are listed in **Table 1**. It can be seen that the accuracy, expressed in the percent relative deviation (%RD) of <4% was achieved for all three elements with a %RSD of <3%.

Table 1. Measurement uncertainties for Group I&II elements (n=3)

	Reverse IDMS		NIST Mixture in U	
Element	% RD	% RSD	% RD	% RSD
Ba	-3.8	2.6	0.5	4.2
Mg	-2.0	1.1	9.9	3.1
Sr	0.4	0.9	2.8	3.6

To further test the method, we spiked the quantified ORNL stable isotope solution into a U matrix containing multi-element NIST standard solution (to mimic a U sample with trace impurity), and performed the separation in triplicate. Analyte fractions were collected and analyzed by HR-ICP-MS. The accuracies were 0.5% (4.2% RSD) and 2.8% (3.6% RSD) for Ba and Sr, respectively, which indicated that the methodology was sufficient for Ba and Sr. An elevated %RD (9.9%) and RSD (3.1%) were observed for Mg due to the reduced sensitivity in medium resolution mode.

The HPLC-ID-ICP-MS method for group II element separation was validated for Ba, Sr and Mg using the CUP-2 CRM. The results listed in **Table 2** are the average of the replicate HPLC separations. The measurement uncertainties were calculated to ISO GUM compliance at k=2. Both the accuracy and precision for Ba were at ~1% RD and RSD. Strontium yielded 7% RD (7% RSD), whereas Mg was at 18% RD (52% RSD)

Table 2. Measurements and uncertainties of Group II elements in CUP-2 (n=3)

Element	Certified value, µg/g of U	Uncertainty, µg/g of U	Measured value, µg/g of U	GUM uncertainty, µg/g of U	% RD	% RSD
Ba	133*	5.7	132	5	-1.4	0.9
Mg	3159*	148	3718	4103	18	52
Sr	67	2.3	72	15	7.3	7.0

* Reference values

Barium and Mg were further evaluated during the 2017 U Benchmarking study (**Table 3**). Barium achieved a similar high accuracy as that from the previous CUP-2 measurement with 0.01% difference (DF) for the two separations. High precision was also achieved for Mg (0.1% DF) with an accuracy of 90% RD. The reduced accuracy for Mg is due to the high background caused by its low first ionization potential and the $^{12}C_2^+$ interference. Strontium was not tested further as it was not a requested element for the U Benchmarking study. Nonetheless, the HPLC-ID-ICP-MS method provides reduced measurement uncertainties for Ba and Sr, as well as Mg.

Table 3. Measurements and uncertainties of Group II elements in CUP-2 during U Benchmarking study (n=2)

Element	Reference value, µg/g of U	Uncertainty, µg/g of U	Measured value, run1, µg/g of U	Measured value, run2, µg/g of U	% DF
Ba	133	5.7	133	133	0.01
Mg	3159	148	2802	2913	0.04

2.3.1.2 Rare earth elements (REE)

2.3.1.2.1 HPLC separation

The ORNL revised HPLC REE separation method requires a gradient-quad pump and four mobile phases: 1 M HCl, 1 M HNO_3, 0.4 M HIBA, and DI water, along with a post column reactor of 0.5 mM 4 – (2 – pyridylazo) resorcinol (PAR). A UV-vis detector was employed at the beginning of the method development to determine the analyte retention time so that analyte fractions can be collected within the appropriate time window. This detector was removed from the system later because the PAR reagent, being a purely organic solution, was detrimental to the subsequent ICP-MS sample introduction and measurement.

The schematic diagram of the process is illustrated in **Figure 2**. The separation (blue), with an overlaid illustration of the gradient profile (green and red), is depicted in **Figure 3**.

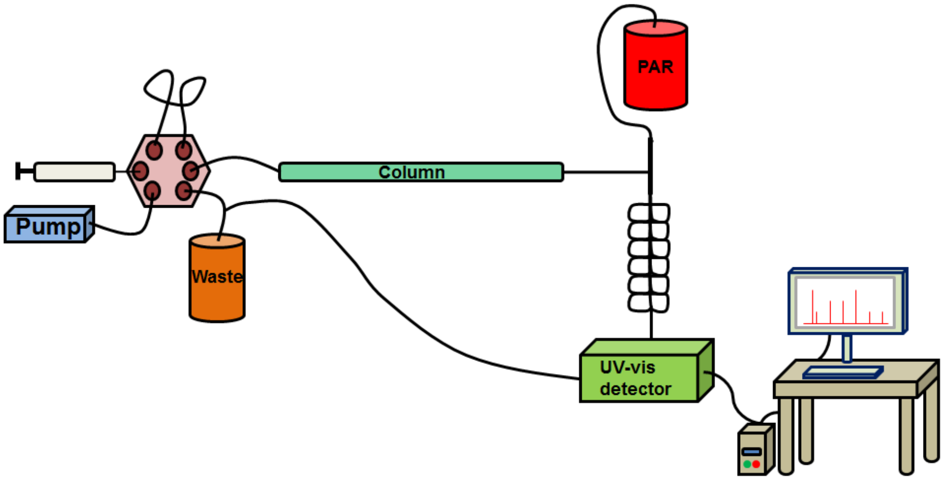

Figure 2. Illustration of the separation of rare earth elements with a CS5A column with detection by UV-vis spectroscopy

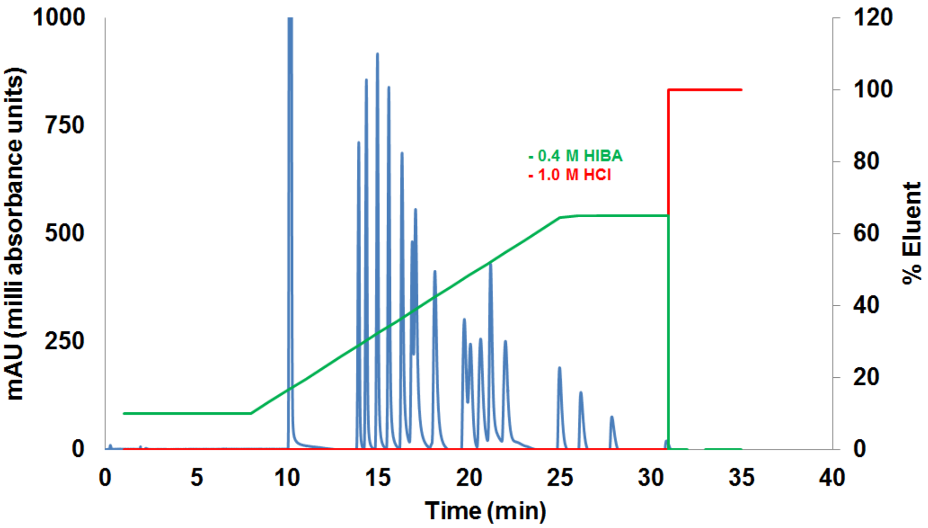

Figure 3. Chromatogram of a REE separation with an overlay of the solvent gradient

Figure 3 demonstrates the separation of 17 REEs in a multi-element standard. Though various eluent flow rates were explored, 1 mL/min yielded the highest separation efficiency. In addition, we have also evaluated the effect of U matrix on REE separation by HPLC. It can be seen from **Figure 4** that U matrix below 1000 µg/mL of U did not affect the REE separation. Elemental fractions were collected from the HPLC elution at a 30 sec interval. These fractions were analyzed by the HR-ICP-MS and the reconstructed chromatogram is shown in **Figure 5**. The rectangular regions in **Figure 5** show the time window a specific fraction collected and analyzed for each element by the HR-ICP-MS.

2.3.1.2.2 HR-ICP-MS analysis

2.3.1.2.2.1 Method development

To develop the ID-ICP-MS method for REE measurement, a multi-element solution containing eight REE, Lu, Yb, Er, Dy, Gd, Eu, Nd, and Ce, was prepared gravimetrically using NIST certified single element standards. This solution was mixed sequentially with ORNL single stable isotopes. The stable isotope concentration for each element was quantified by the rIDMS method. The accuracy and precision (n=3) was compared with the calculated concentration (**Table 4**). It can be seen that the percent relative deviation and reproducibility ranged from -2.6 to 0.5% RD and 0.4 to 3.0% RSD respectively.

To further test the method, we spiked the quantified ORNL stable isotope solution into a U matrix containing multi-element NIST standard solution and performed the separation in triplicate. Fractions were collected every 1.5 min and analyzed by HR-ICP-MS (**Table 4**). It can be seen from Table 4 that the %RD for all REE ranged from -0.4 to 0.6% except for Gd (2.6%), and the precisions were <1% RSD for all REE with the exception of Eu, (2.3%). For Dy, Eu, Gd, Nd, and Yb, two studies (n=6) were conducted over the course of two months (between July 2016 and September 2016). The fact that the accuracy and precision remained relatively unchanged over this two month period reiterates the consistency of the reduced measurement uncertainty for REE using the ID-HPLC-ICP-MS method.

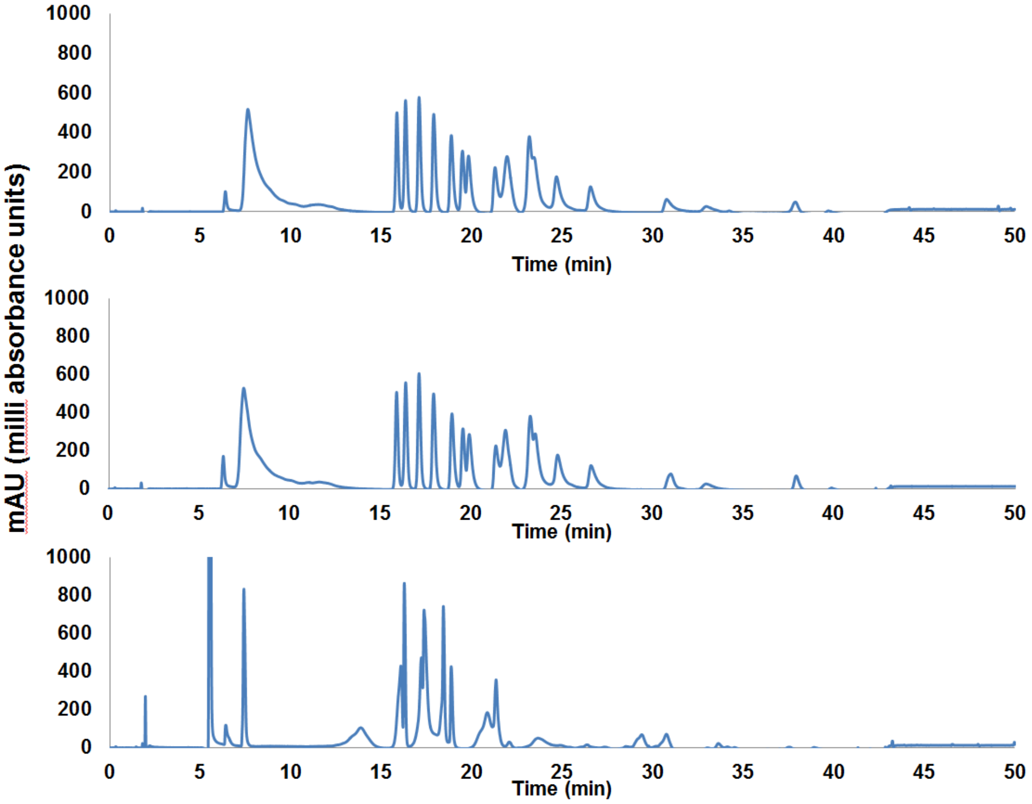

Figure 4. Chromatograms of a REE standard in various uranium matrices (500 µg/mL U – top, 1,000 µg/mL U – middle, and 5,000 µg/mL U – bottom)

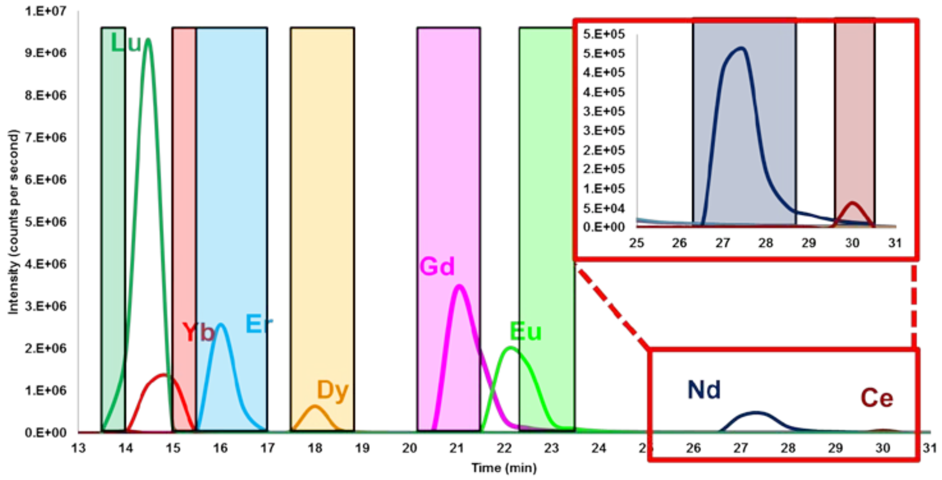

Figure 5. Eight REE chromatogram separated by HPLC and detected by HR-ICP-MS

Table 4. Measurement uncertainties for REE (n=6)

	Reverse IDMS		*NIST Mixture in U*	
Element	% RD	% RSD	% RD	% RSD
Dy	0.5	0.5	-0.2	0.9
Er	-0.7	0.4	-0.4	0.5*
Eu	-1.1	3.0	-0.3	2.3
Gd	-2.6	1.5	-2.6	1.0
Lu	-0.4	1.1	-0.2	1.1*
Nd	1.1	0.5	0.6	0.7
Yb	-2.2	2.9	-0.4	0.5

* n=3

Cerium is not suitable for this method due to a small HPLC peak, indicative of a poor separation yield, and an isobaric interference with ^{136}Ce (enriched isotope) by ^{136}Xe. Even operating the ICP-MS at the medium resolution mode, the two peaks are unresolvable. Cerium can only be analyzed by using the HR-ICP-MS in high-resolution mode. However, that requires a higher concentration of Ce in the sample, which is not within the scope (trace impurities) of this study.

2.3.1.2.2.2 Method validation

The REE separation and determination method was validated using the CUP-2. Although CUP-2 is a certified reference material, it is not certified for rare earth elements. Consequently, we elected to compare the measured REE values to the long-term cumulative results provided by LLNL, one of the laboratories that participated in the round robin study for CUP-2 characterization. A detailed sample preparation procedure has been described in the Trace Elements in Uranium Benchmarking Study report [2]. An aliquot of the dissolved CUP-2 solution was spiked with Dy, Er, Eu, Gd, Lu, Nd, and Yb stable isotopes (Provided by ORNL) which were quantified by the rIDMS method. All dissolutions/dilutions were performed gravimetrically.

An initial separation “screen” was performed (**Figure 6**) in order to determine the retention time profile of REE (**Figure 6a**) and examine the effect of the uranium matrix (**Figure 6b**) on the REE separation and analysis. **Figure 6b** illustrates that uranium matrix was eluted out of the column from the beginning to 17 min, co-eluting with Lu, Yb, and Er.

Method accuracy was evaluated based on the comparison with LLNL values obtained by the traditional ICP-MS method (without HPLC separation). Two dissolutions were performed months apart. **Table 5** is the averaged results from triplicate separations. Accuracy is evaluated based on the comparison with LLNL values obtained by the traditional ICP-MS method (without HPLC separation). All REE are consistent with LLNL values ranging from -2.5 - 9.5% RD and 0.1 - 6.1% RSD.

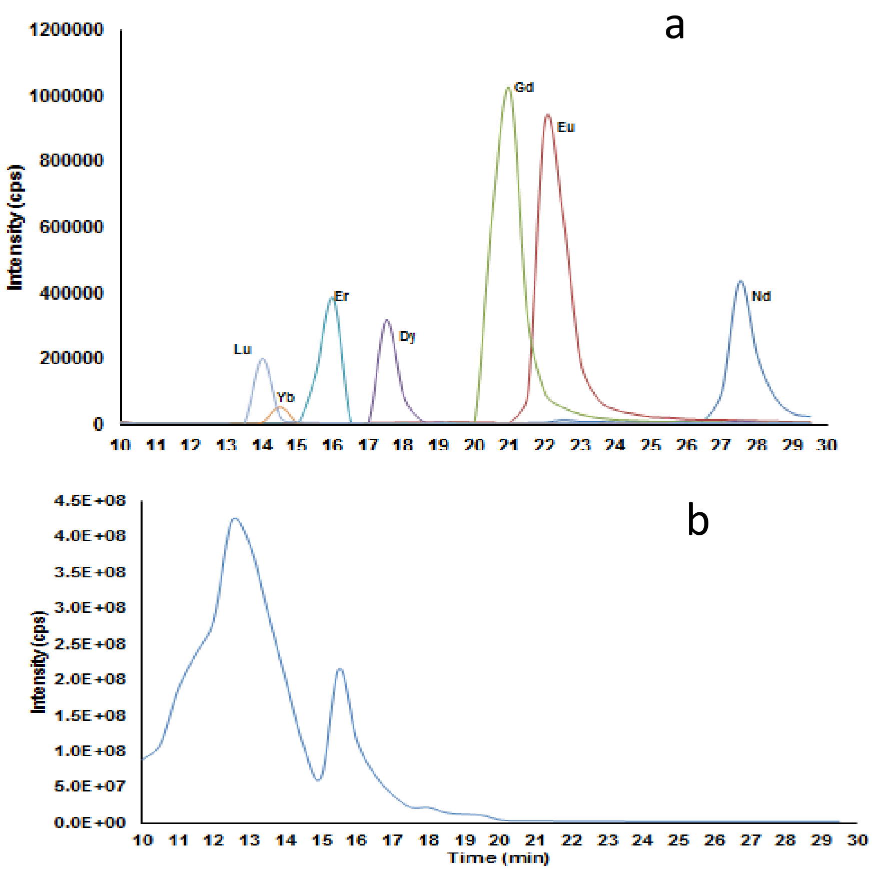

Figure 6. Chromatogram elemental fraction to examine the elution time of (a) each REE element and (b) uranium matrix

Table 5. Measurements and Uncertainties of REE in CUP-2 (n=9)

Element	Certified value, µg/g of U	Uncertainty, µg/g of U	Measured value, µg/g of U	GUM uncertainty, µg/g of U	% RD	% RSD
Dy	22.5	0.8	22.5	0.2	-0.1	0.5
Er	10.5	0.4	10.3	0.6	-2.5	1.2
Eu	0.88	0.03	0.96	0.2	8.7	0.1
Gd	17.8	0.6	18.6	1.7	4.7	0.4
Lu	1.03	0.05	1.02*	0.1	-1.3	6.1
Nd	24.0	0.8	26.2	5.0	9.5	0.5
Yb	8.3	0.3	8.0	0.7	-3.7	2.6

*n=6

Although there are only three data points, the fact that six of the seven elements obtained less than 3 % RSD demonstrates that HPLC-ID-ICP-MS is an adequate tool to minimize measurement uncertainty.

One important aspect of the IDMS method is to determine the appropriate amount of stable isotope spike. Ideally, IDMS requires a concentration that results in an isotopic ratio in the mixture that is between the spike and natural ratios. In the case of Lu, with a concentration of only 1 µg/g of U in CUP-2, consequently matching the spike amount is vital, as the $^{175/176}$Lu ratio in the ORNL spike is also low. Our initial spike volume resulted in a large error because the $^{175/176}$Lu in the mixture was too close to that in the enriched isotopes. One should also bear in mind that the "accuracy" comparisons here are made against LLNL non-certified values; therefore, the error percentage values reflect only the *differences* in the results obtained from the ID-HPLC-ICP-MS method vs. the LLNL traditional (no HPLC separation) method.

The validated method was applied to the REE elements in the U Benchmarking study [2]. There was good agreement between the multiple runs in different days for each of the three U oxide test samples, and among the four replicate sub samples for U ocxide. In general, there was <5% RDS for elements at higher concentration (>5 µg/g of U_3O_8) and ~ 20% when the concentrations are close to the detection limits.

Table 6. Measurements and uncertainties of REE in CUP-2 during U Benchmarking study (n=2)

Element	Reference value, µg/g of U	Uncertainty, µg/g of U	Measured value, run1, µg/g of U	Measured value, run2, µg/g of U	% DF
Dy	22.5	0.8	24.1	26.1	0.08
Er	10.5	0.4	12.0	11.9	0.01
Eu	0.88	0.03	1.0	1.1	0.08
Gd	17.8	0.6	20.2	19.9	0.01
Lu	1.03	0.05	1.02	1.08	0.05
Nd	24.0	0.8	26.8	27.4	0.02
Yb	8.3	0.3	9.2	10.0	0.09

REE in CUP-2 was examined again during the U Benchmarking study as a quality control (**Table 6**). The percent differences between the two separations for all elements are <0.1% indicating a good reproducibility of the for sample process. The measured REE values were somewhat higher than that obtained months ago (**Table 5**). A tight 90-day testing period prohibits us from re-evaluating the causes and re-analyzing the CUP-2 sample. Although the 90-day testing period seems ample, it is actually insufficient for applying the labor-intensive HPLC-ID-ICP-MS method to the evaluation of 16 elements extensively. For instance, under ideal conditions, the preparation of three test samples and QC samples (duplicate dissolutions, pre-screening for concentration range, calculating and preparation for stable isotope spike, isotope dilution preparation) takes 7 working days. The HPLC separation for one family of elements (pre-screening for retention time window, duplicate separations for each dissolved sample and QC samples) takes 7 days. Three families of elements will take 26 days because the HPLC system has to be re-configured and flashed in between sub-methods. The isotope measurement for each analyte fraction by HR-ICP-MS has to strictly follow the required sequence (reagent blank, NIST standard, enriched spike, non-spiked sample, preparation blank and spiked sample) and takes 30 days for all fractions. With only 77 working days for the 90-day study, the added the task days (10 days) for data reduction and report generation easily exceeded the requirement (83 days). In reality, one has to take into consideration the re-runs, instrument downtime, and other unforeseen facility difficulties. As a result, the completion of the HPLC section of the 90-day Benchmarking study was only possible with extra non-working days by multiple analysts.

2.3.1.3 Transition elements (TE)

2.3.1.2.1 HPLC separation

The transition elements, Cd, Cu, Fe, Ni, Ti, and Zn, were separated on a DIONEX Ionpac CS5A chromatography column (and CG5 guard column) via a gradient introduction of a mobile phase consisted of MetPac pyridine-2, 6-dicarboxylic acid (PDCA) eluent, 66 mM potassium hydroxide, 74 mM formic acid, and 5.6 mM potassium sulfate. A detailed operation procedure was included in Appendix A. A 250 µL sample loop was employed to introduce the sample to the column. Initially, we followed the ORNL protocol to apply a 0.5 mL/min flow rate for separation. This flow rate resulted in a poor separation for all five elements (**Figure 7a**). By further exploring the various flow rates, it was determined that the 1.2 mL/min provided the optimal separation (**Figure 7b**).

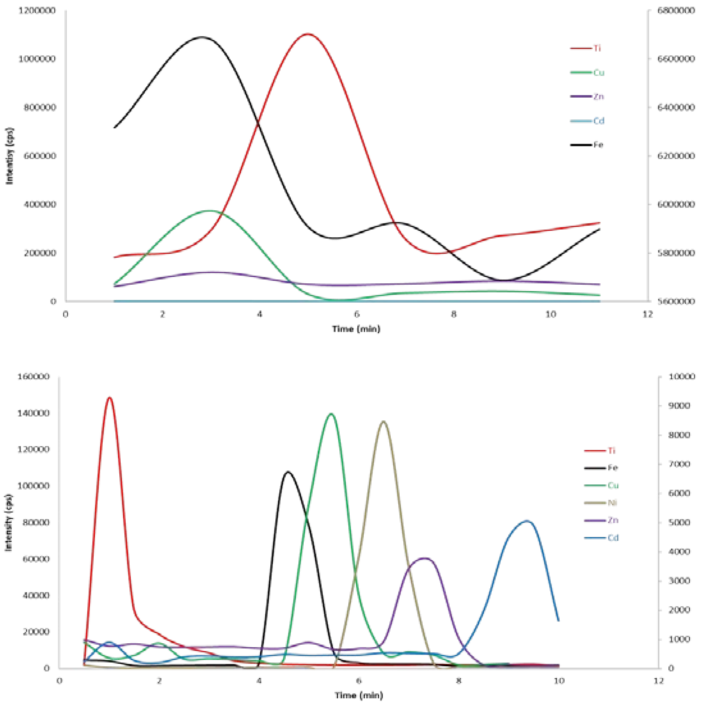

Figure 7. Separation of transition elements at a) 0.5 mL min^{-1}, top, and b) 1.2 mL min^{-1}, bottom, flow rate.

2.3.1.2.1.1 ID-ICP-MS determination

The enriched isotope spike solution containing Cd, Cu, Fe, Ni, Ti . Zn were prepared from the materials received from ORNL in either in an oxide or metal form. Unlike other analytes, the complete dissolution of Ti metal was only achieved when HF was added into the 8 M HNO_3 during heating. Like the previous two families of elements, the dissolved enriched isotope solutions were mixed with NIST certified single element standards one at a time gravimetrically, to form a mixture that contains 10 µg/g of each element.

The rIDMS was performed to determine the concentration of the enriched spikes solution (**Table 7**). It is seen in Table 7 that Cd, Cu and Fe were close to the expected concentrations while Ni and Ti were 36 and 51% lower, respectively. Zn was not included because the NIST standard was not available. It was speculated that the low concentrations of Ni and Ti were due to either incomplete dissolution or post-digestion precipitation.

Table 7. Measurement uncertainties for TE (n=3)

	Reverse IDMS		*NIST Mixture in U*	
Element	% RD	% RSD	% RD	% RSD
Cd	10	9.2	0.5	2.1
Cu	7.6	1.3	30	6.3
Fe	0.5	5.3	29	73
Ni	-36	2.4	8.3	16
Ti	-51	0.4	6.2	0.3

In order to confirm our speculation, the enriched isotope solutions were spiked one by one into a mixture of NIST certified element solutions and separated by HPLC. The results from the HR-ICP-MS analyses failed either the accuracy or the precision expectations on all TE except for Cd. We have attempted to characterize the enriched spike solutions further using the CUP-2 reference material. The results are presented in **Table 8** (n= 3). As can be seen in **Table 8**, both Ni and Ti are more agreeable than in the previous test, indicating that the dissolutions for Ni and Ti were indeed successful. CUP-2 was analyzed again during the Benchmarking study. This time, only Fe and Ti were detected and the error was ~20 %.

The failure of the method to quantify transition elements is two-fold. The first is in the HPLC separation: elution peaks shifted dramatically, i.e., the elemental retention varied from sample to sample, which made it difficult to “catch” the target fraction. Even multiple “screen” runs were executed prior to the separation and prolonged column washes were performed. This effect was most prominent for iron. Constant peak shifting is indicative of the incompatibility of the column with the transition elements under the current eluent and gradient profile provided by the ORNL protocol. Another difficulty stems from the interference during the ICP-MS measurement. For instance, ^{56}Fe has a sevier polyatomic interference by ArO (56 m/z) in the plasma, therefore, it is difficult to measure the $^{57/56}$Fe ratio. Because the ORNL stable isotope is ^{57}Fe, one must select ^{56}Fe for ratio measurement due to the low abundance of ^{58}Fe in the sample.

Table 8. Measurements and uncertainties of TE in CUP-2 (n=3)

Element	Certified value, µg/g of U	Uncertainty, µg/g of U	Measured value, µg/g of U	GUM uncertainty, µg/g of U	% RD	%RSD
Cu	28*	1	28	2	-2.2	3.5
Fe	4291	95	4200	419	-2.1	4.5
Ni	40	2	32	N/A	-20	N/A
Ti	262	7	217	75	-17	1.4
Zn	48*	20	53	N/A	11	N/A

* Reference values

2.3.1.4 Summary

Like any analytical chemistry method, HPLC-ID-ICP-MS is not universal, therefore, can only be applied to a select group of elements. It is the conclusion from this study that this methodology is suitable for improving measurement uncertainties for the rare earth elements (Dy, Eu, Er, Gd, Lu, Nd, and Yb) and Group II elements (Ba, Sr, and Mg), but not suitable for elements such as Ca, Ce, K. This method can only be applicable to transition elements if further method development on both HPLC separation and ICP-MS measurement could be performed. The method validation using a complex U matrix has led to the following conclusions:

1. For the seven REE elements, the respective accuracy and precisions are <3% RD and <3% RSD, for measuring the NIST certified standard solution and <10% error and <7% RSD, respectively, for measuring the CUP-2. The HPLC separations are proven to be robust and the HR-ICP-MS analyses are free from polyatomic interferences, allowing the measurements to be performed in low-resolution mode. Ce was the only element that is not suitable for this method due to the poor HPLC separation yield and the isobaric interference by Xe in the plasma.
2. The measurement accuracy for Ba is <2% RD for both the NIST standard solution and CUP-2 with <5% RSD. Sr can be measured with <8% RD and <7% RSD in both the NIST standard solution and CUP-2. Mg can only be measured by this method if the concentration is high enough (>500 µg/U) at an accuracy of 10 to 20% error.
3. The rest of the group I&II elements, such as K, Rb, Ca, are not suitable for the ID-HPLC-ICP-MS method due to the analytical difficulty caused by polyatomic interferences during the ICP-MS measurement. Transition elements are also not suitable under the current method conditions due to the unreliable separation, certain polyatomic interferences, and low analyte isotopic abundance.

4. The method detection limits for each analyte were calculated at a signal to noise ratio equal 3 using the process blank at the isotope masses of interest (Appendix A).

   There are 14 high priority elements on the U Benchmarking list, and Er and Nd are the only two that are deemed suitable for the HPLC-ID-ICP-MS method. Others are either mono isotopic, or were not studied in the ORNL transferred methods. There are 11 moderate priority elements, Dy and Mg are the two that can be validated. Out of the 11 low priority elements, 5 of them, Ba, Eu, Gd, Lu, and Yb, were validated using CUP-2. While the methodology is indeed advantageous for improving the measurement uncertainty, the prolonged analysis time prohibits a thorough evaluation of all the analytes of interest, especially the transition elements, during a 90-day study.

2.3.1.5 References

1. "Determination of trace elements is uranium and plutonium by HPLC-ID-ICP-MS", Benjamin Manard, Benjamin Byerly, Ning Xu, Lav Tandon, Los Alamos National Laboratory Report LA-UR-16-22162
2. "Trace Elements in uranium benchmarking study", Benjamin Manard, Ernest M Wylie, II, Ning Xu, Dennis P. Montoya, Dara Aragon, Michael Schappert, Michael S. Rearick, Kattathu J. Mathew, Ann R. Schake, Lav Tandon, Los Alamos National Laboratory Report LA-UR-17-20350

Appendix A

HPLC-ID-ICP-MS Method Analyte Detection Limits, µg/g of U

Analyte	DL
Dy	0.001
Lu	0.007
Yb	0.008
Gd	0.01
Eu	0.02
Er	0.03
Nd	0.08
Ba	0.2
Sr	0.6
Fe	1
Cu	1
Ni	1
Ti	4
Zn	6
Ca	40
Mg	300