

Selective Ultratrace Determination of Uranium Isotopes in the Environment

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The determination of the isotopic signature of uranium traces found in the environment is of crucial importance for the detection of undeclared nuclear waste and/or anthropogenic contaminations. The isotope of interest is ²³⁶U, which is produced by neutron capture from ²³⁵U. In the environment neutron fluxes are very low and thus the natural abundance of ²³⁶U compared to ²³⁸U is well below 10⁻¹⁰ (1). By contrast, the ²³⁶U concentration in spent nuclear fuel can reach up to several percent due to the high neutron flux in nuclear reactors (2). So the uranium isotope ²³⁶U is a sensitive tracer for anthropogenic uranium and can provide further information on migration and speciation behaviour of man made nuclear waste in the environment.

In a close collaboration between Mainz University and PNNL, Richland, High Resolution Ionization Mass spectrometry (HR-RIMS) is developed to precisely measure the isotopic composition in low level uranium samples. For this purpose, the initial sample is evaporated from a graphite furnace into vacuum to form a well collimated atomic beam. Neutral uranium atoms are resonantly excited by precisely tuned laser light by multi-step optical excitation steps along bound atomic states up to final ionization. Photo ions generated are guided through a quadrupole mass filter for background reduction and further enhancement of the isotopic selectivity. Finally uranium ions are quantitatively detected by a channeltron detector.

The first and most important step for highly selective ultra trace analysis of uranium using a HR-RIMS system is the identification and characterization of a suitable ionization scheme. Optical transitions between atomic levels in an appropriate excitation scheme must have narrow natural line width to provide high optical selectivity as well as sufficient transitions strength for efficiency. In that case saturation with moderate laser power is possible. Extensive laser spectroscopy on uranium intermediate levels were done by P.G. Schumann and B.A. Bushaw (3,4) and lead to a suitable excitation scheme using laser wavelengths of 415 nm, 829 nm and 722 nm.

For first tests of analytical measurements samples with known amount of about 10¹⁷ uranium atoms were inserted on zirconium foil and heated up to ~2500° C in a graphite furnace. A total efficiency of 3· 10⁻⁷ and isotopic ratios ²³⁶U/²³⁸U down to 10⁻⁸ have been demonstrated so far. Analytical measurements were performed on a synthetic dilution series in the isotopic range of ²³⁶U/²³⁸U ratios from 10⁻³ down to 10⁻⁸. These measurements confirm the linearity of the system over the full accessible range. The final characterization concerning accuracy and reproducibility will be done using certified samples from IRMM (Institute of reference material and measurements, Geel, Belgium).

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(2) G. Kirchner and C. Noack, *Nucl. Safety*, 29 (1988) 1

(3) P.G. Schumann, B.A. Bushaw and K.D.A. Wendt, *Spectrochim. Acta Part B*, 60/11 (2005) 1402-1411.

(4) B.A. Bushaw, S.L. Ziegler, S. Raeder, K. Wendt, *Spectrochim. Acta B*, submitted 2007.