

Detection of the ultra trace Isotope U-236 using High Resolution Resonance Ionisation Mass Spectrometry

S.Raeder¹, S. Fies¹, K.Wendt¹, N. Trautmann² and J.V. Kratz²

¹ WA LARISSA, Institute of physics, Johannes Gutenberg-University, D-55099 Mainz

² Institute of nuclear chemistry, Johannes Gutenberg-University, D-55099 Mainz

The detection of the ultra trace isotope ²³⁶U in uranium containing samples provides information about origin and history of the contamination. The long living radionuclide ²³⁶U is produced from ²³⁵U by neutron capture and the low natural neutron fluxes results in natural isotope ratios $\frac{^{236}\text{U}}{^{238}\text{U}} < 10^{-10}$. A significant enhancement of this isotope indicates a neutron exposition and therefore an anthropogenic origin of the sample. The determination of the $\frac{^{236}\text{U}}{^{238}\text{U}}$ – ratios by selective mass spectrometry in a region with anthropogenic uranium contamination allows for the study of the migration behaviour of elemental uranium species in the environment.

The method of High Resolution Resonance Ionization Mass Spectrometry (HR-RIMS) combines a selective laser-ionization with the mass resolution of commercial mass spectrometers. Due to the electron structure of the atomic shell it is possible to ionize evaporated uranium atoms isotopes selective using narrow bandwidth cw-lasers. For ²³⁶U an optical three step excitation and ionization scheme is applied and a quadrupol mass filter is used for mass selection an suppression of surface ions. The method competes in abundance sensitivity with AMS.

First analytical measurements with synthetic samples demonstrate a selectivity $> 10^8$ and an efficiency $> 5 \cdot 10^{-7}$ for the whole system. In a linear arrangement of ion optic and quadrupol mass filter the selectivity is limited by background from neutral uranium atoms at a level of 10^{-7} compared to resonant laser ions. The detection system was thus upgraded by a 90°-deflector to separate the laser ions from the remaining neutral atoms resulting in a corresponding reduction of the background. Measurements of certified samples and first measurements of environmental samples are foreseen. Furthermore the development of a direct sample injection, which allows a coupling of the RIMS-technique to chromatographic and electrophoretic methods, will be discussed. The status of the development and concepts for improvement of the system will be presented as well as the foreseen application.