

Measurement and validation of the isotopic composition in inhomogeneous samples by LA-MC-ICP-MS

Introduction

Uranium-bearing materials are strictly controlled by the international nuclear safeguards system. However, if such materials get out of regulatory control and subsequently confiscated, a comprehensive characterization is required to assess the hazard, to determine the intended use and the potential origin. Nuclear forensics focuses on the analysis of these intercepted nuclear or other radioactive materials to provide information for the investigating authority to avoid the diversion and subsequent malicious use [1-3].

Most of the nuclear forensic samples contain visible or non-visible inhomogeneity, which can be exploited for potential nuclear forensic conclusions. One of the available micro-analytical techniques for such measurements is laser ablation inductively coupled plasma mass spectrometry with simultaneous detection (LA-ICP-MS) [4,5]. As LA uses a focused laser beam scaled down to a few micro-meters, LA-ICP-MS analysis can reveal sample inhomogeneity in the material in question and is able to measure the spatial distribution of the isotopic composition. However, the precise measurement can be hindered by instrumental parameters (e.g. laser beam size or scan speed) or by the sample characteristics (e.g. grain size or differences in U isotope enrichment) [6].

Present Work

The present work examines the nuclear forensic value of U isotopic inhomogeneity for by LA-ICP-MS measurements. To study the important parameters, a synthetic sample was prepared by the mixing of two solid certified standard materials (SRM U-010 and SRM U-030) to mimic an inhomogeneous U sample. By a rough LA measurement using a line scan or a 2-D area (map) on the sample surface, the points-of-interest in the inhomogeneous sample can be found and chosen (localization). After finding the required sample position, a more precise measurement can be performed on the selected small locations. The procedure allows the accurate analysis of the isotopic composition at the relevant spots and the proper identification of the end-products (i.e. the constituting starting materials). The developed LA-ICP-MS method was also compared with the large-geometry secondary ion mass spectrometry.

[1] Mayer K, Wallenius M, Varga Z (2013) Nuclear forensic science: Correlating measurable material parameters to the history of nuclear material. *Chemical Reviews* 113, 884-900

[2] Kristo MJ, Gaffney AM, Marks N, Knight K, Cassata WS, Hutcheon ID (2016) Nuclear Forensic Science: Analysis of Nuclear Material out of Regulatory Control, *Annual Review of Earth and Planetary Sciences* 44, 555-579

[3] Varga Z, Wallenius M, Krachler M, Rauff-Nisthar N, Fongaro L, Knott A, Nicholl A, Mayer K (2021) Trends and Perspectives in Nuclear Forensic Science. *TrAC Trends in Analytical Chemistry* (accepted)

[4] Krachler M, Wallenius M, Nicholl A, Mayer K (2020) Spatially-resolved uranium isotopic analysis of contaminated scrap metal using laser ablation multi-collector ICP-MS, *RSC Advances* 10 16629–16636

[5] Jovanovic SV, Kell T (2021) Nuclear forensic analysis with laser ablation inductively coupled plasma mass spectrometry in CMX-6. *Journal of Radioanalytical Nuclear Chemistry* 329 319–326

[6] Varga Z, Wallenius M, Nicholl A, Mayer K (2020) Assessment of uranium inhomogeneity and isotope imaging for nuclear forensics, *Spectrochimica Acta - Part B Atomic Spectroscopy* 171, 105920

Primary authors: Dr VARGA, Zsolt (European Commission Joint Research Centre Institute for Transuranium Elements); Dr WALLENIOUS, Maria (European Commission, Joint Research Centre, Institute for Transuranium Elements); Mr NICHOLL, Adrian (European Commission, Joint Research Centre); Dr MAYER, Klaus (European Commission - Joint Research Centre, Institute for Transuranium Elements)

Presenter: Dr VARGA, Zsolt (European Commission Joint Research Centre Institute for Transuranium Elements)

Session Classification: Oral Session #7 – Signature Research on Isotopic Signatures and Age-Dating

Track Classification: 1. Nuclear Forensics Capability Building: Initiation and Sustainability: 1.2 New Technologies, R&D and Signature Research in Nuclear Forensics