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Production, Analysis, and Application of Microparticulate Uranium Oxide-based Reference Materials

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Abstract

A special session was organized in the framework of the INMM & ESARDA – Joint Annual Meeting, 2023 representing the current status and future perspective of recent developments and activities regarding the production, analyses and application of microparticle reference materials in nuclear safeguards and nuclear forensics. The session was aimed at identifying future requirements and needs to sustainably support a robust quality control system for international safeguards and nuclear forensics efforts.

Introduction

The analysis of individual micrometric and submicrometric uranium-oxide based particles collected by International Atomic Energy Agency (IAEA) safeguards inspectors on swipe samples during in-field verification inspections requires well-defined microparticulate reference materials for quality control measures. The IAEA is building up new capacities to the worldwide Network of qualified Analytical Laboratories (NWAL) for the provision of well-designed and characterized microparticle reference materials suitable for the verification of analytical data, the daily calibration performance of analytical instruments, method development and evaluation as well as for the conduction of frequently performed proficiency tests.

Summary of Special Session

Eleven applications for oral presentations were submitted for the special session on production, analyses, and application of microparticle reference materials. The session was split into two parts. The first part mainly addressed recent achievements regarding the production of microparticulate reference materials for IAEA's NWAL. The second part was dedicated to fundamental science aspects suitable for the development of novel reference particles for the NWAL, to the optimization of the design of reference microparticles as well as to recent advancements on the development of analytical techniques applying these particles.

Production and Application of Reference Particles

The production of uranium-oxide based microparticulate reference materials for the QC system of the IAEA was presented using two different procedures. The laboratories at Savannah River National Laboratory (SRNL) and Forschungszentrum Jülich GmbH (FZJ) are producing the microparticles using an aerosol-based method [1-3] while the Pacific Northwest National Laboratory (PNNL) is utilizing a wet-chemical, hydrothermal synthesis method [4]. Both methods allow for the provision of well-defined and qualitatively high-level microparticulate reference materials. The aerosol-based method at FZJ was already optimized for the provision of two batches officially certified reference materials according to the isotopic composition the amount uranium per particle for one batch. Very recently one additional batch of highly enriched uranium (50% ^{235}U enrichment) microparticles [2] was provided to the IAEA. The aerosol-based method that is already established at SRNL [1] and currently under implementation at FZJ [3] is designed to produce large quantities in the µg to mg range of microparticles. This range of yield was very recently applied to develop a method for the successful production of surrogate environmental swipes [5] which is a very important test material for the IAEA's NWAL.

For the application as reference materials in safeguards and nuclear forensics a sound understanding regarding of the elemental and isotopic composition, crystal structure and homogeneity of the microparticles is essentially required. The synthesis and characterization on the atomic level is of particular importance for the development of doped reference materials. For instance, synthesis parameters such as pH and temperature significantly govern the crystal growth and therefore, the morphology [4] and crystal structure [4,6] of the particles. Additionally, it was presented that the aerosol-based method at SRNL [7] can be transferred to the production of molybdenum-oxide and tungsten-oxide microspheres. Regarding the development of composite reference particles a systematic study on bulk model materials demonstrated the structural incorporation of Th into uranium oxide structure (UO_3 and U_3O_8) which are relevant for the aerosol-based process to produce reference materials for age-dating application [6]. The outcome of these complementary studies will provide important insight into the particles' properties, e.g. the homogeneity, which significantly influence the data evaluation of frequently performed mass spectrometric (MS) measurements, for instance using LG-SIMS (Large Geometry Secondary Ion Mass Spectrometry) and support the development and optimization new analytical methods for particle analysis [8, 9].

Characterization of Microparticles

The analysis of microparticulate materials is very crucial for the application of microparticle reference materials in nuclear safeguards and nuclear forensics. The analysis of isotopic composition of single microparticles is routinely performed utilizing LG-SIMS measurements within the NWAL. Well-established measurement protocols for the LG-SIMS exist to detect major (^{238}U and ^{235}U) and minor (^{234}U and ^{236}U) isotopes and even of traces of 'foreign' elements in the microparticles, such as Pu [10] with very high precision. Additionally, the primary ion beam (O^-, O^{2-}, O^{3-}), sample and substrate (e.g. Si, carbon) interaction has to be considered to impact usefully yields, precision and isotope correction factors [10, 11]. Although very well-established, the LG-SIMS measurements are extremely time-consuming since the sample preparation includes locating relevant microparticles on the cotton swipes and subsequent their removing and analyzing. The microextraction ICP-MS technique is currently under investigation at the Oak Ridge National Laboratory (ORNL) and is a very promising alternative for the quick sampling of particles from the cotton swipes and measuring, e.g. the U and Pu isotope ratios [9].

Besides the isotopic composition determined by advanced mass-spectrometric methods the morphological description of the particles (size, geometry, etc.), identification of major and minor elemental composition as well as the chemical phases (molecular composition and crystal structure of compounds) is currently investigated by the CEA [8]. By coupling an SEM/EDS system with a µ-Raman spectrometer this information can be collected simultaneously and provide important information about the internal and particle-to-particle elemental homogeneity. Additionally, potential heterogeneities can be directly assigned to specific phenomena, such as morphological and/or phase changes.

Conclusions

The special session 'Production, Analysis, and Application of Microparticulate Uranium Oxide-based Reference Materials' was well-attended with very lively discussions between particle producers, analysts, and end-users. As such, it stimulated the discussion as well as networking within this community to identify future requirements and needs to sustainably support a robust quality control system for international safeguards and nuclear forensic efforts. From the huge interest (number of contributions and attendees) and positive feedback to this special session the organisation of a follow-up special session at the INMM Annual Meeting 2024 is recommendable.

Acknowledgements

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