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Preparation and validation of Pu age dating materials for nuclear forensics

Introduction

Nuclear materials are subject to strict verification by the international nuclear safeguards. However, the IAEA's Incident and Trafficking Database records year by year cases where such a material is found out of regulatory control. Such an interception is typically followed by a comprehensive analysis of the material in order to provide hints on its intended use and potential origin. Nuclear forensics analysis provides information to the investigating authority to prevent malicious use, prosecute the perpetrators and to avoid further diversion [1-3].

Among the significant signatures in nuclear forensics, age dating (also known as the determination of the production date), was found to be a very valuable sample characteristic. The method is based on the radioactive decay of the parent radionuclide into the daughter nuclide: the measured parent-daughter ratio (also known as radio-chronometer) indicates the date of a specific production step (e.g. parent/daughter chemical separation step, metal casting), and it is colloquially referred to as the "age" of the material. This is a self-explanatory signature, i.e. it does not require a comparative dataset for the evaluation. The method is quite well established for uranium-bearing samples, but it has been less studied for plutonium materials [4] and would particularly benefit from a sound metrological basis.

Present Work

The present work describes a method for the preparation of plutonium age dating reference materials (similarly to the uranium IRMM-1000 certified reference material) and its independent validation by an international co-operation. The prepared samples can be used to validate experimental protocols for determining the production date of plutonium via the 234U/238Pu, 235U/239Pu, 236U/240Pu, and 241Am/241Pu chronometers. Reactor-grade plutonium was used as starting material and chemically purified using a dedicated method to ensure high Pu recovery, while maximizing U and Am separation efficiencies. The U and Am separation factors were determined by the addition of high amounts of 233U and 243Am spikes and their re-measurement in the final product. The prepared material is intended for quality control and assessment of method performance in nuclear forensics and safeguards [5].

In order to establish the proof-of-concept for a methodology for developing a Pu age dating reference material and to confirm our age dating results, several subsamples with various Pu concentrations and physical forms (solid and liquid) were sent to expert nuclear forensic laboratories (Lawrence Livermore National Laboratory and Los Alamos National Laboratory in the USA, AWE in the United Kingdom, CEA in France, FOI in Sweden and International Atomic Energy Agency laboratory in Seibersdorf). The co-operation allows detecting any problems and issues related to the preparation and tailoring the Pu age dating reference material to the laboratory needs.

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