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Preparation and Validation of an Uranium Age Dating Reference Material

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If nuclear materials are diverted and afterwards interdicted, detailed investigation is required to identify the possible origin, intended use and hazard related to the material. Such analysis, which is now commonly referred to as nuclear forensics, involve the comprehensive physical, chemical and isotopic measurements (e.g. physical dimensions, crystal structure, radioactive and stable chemical impurities, classical forensic analysis) as well as the interpretation of these measured parameters [1-3]. Based on this complex information, the assumed origin of the material can be verified or for an unknown material the provenance can be identified with high reliability.

Numerous characteristics (so-called signatures) of the material can be used for such purpose, such as the isotopic composition of U, Pb or Sr, elemental impurities, trace-level radionuclide content, crystal structure or anionic residues. Besides these parameters the elapsed time (commonly referred to as the "age" of the material) since the last chemical purification of the material can also be measured for radioactive (nuclear) materials. This unique possibility is based on exploiting the presence and decay of the long-lived radionuclide (usually uranium or plutonium as major component in case of nuclear materials): in the course of the production the radionuclide is chemically purified from the impurities, including also its radioactive decay products. After production, the radioactive progenies start to grow-in again in the material. Assuming that the parent-daughter separation was complete, the elapsed time since the last separation, thus the production date, can be calculated according to the decay equations after the measurement of the parent-daughter ratio in the sample. This age value enables either to identify the origin of the unknown sample or to verify the source of the feed material. In contrast to most other characteristics used in nuclear forensics, the production date of the material is a predictive signature, thus it does not require comparison samples for origin assessment. This feature makes the production date one of the most prominent signatures for attribution.

However, in order to put the obtained results on a more solid scientific or legally defensible foundation, dedicated reference materials are required. In consequence, an emerging need for such materials has been recently expressed by the community involved in national or international nuclear security programs.

Our major objective was the preparation and validation of a uranium-based reference material, which can be applied for the validation of age measurements based on the 230Th/234U chronometer. The material was prepared from high-purity uranium solutions with various uranium enrichments by completely separating the thorium decay product [4]. By this means, the production date is very precisely known (with an uncertainty of less than about 5 hours). In contrast to other methods of producing age dating reference materials , this approach does not require measuring the age of the final material and thus deriving a certified production date, because, if all conditions are fulfilled (completeness of separation, long-term stability, closed system), the 230Th present in the material will solely depend on the radioactive decay laws. Therefore, the material prepared can be used as a primary standard for age dating of uranium materials.

The aim of the present collaboration is to prove the applicability of this methodology for the preparation of a uranium age dating reference material by the independent measurement of expert laboratories. Since the validation requires the measurement of the 230Th decay product at very low level from the freshly separated material, state-of-the-art instruments and well-established techniques are required. Though the primary objective of this joint collaboration is to prove that the concept of preparing such age dating material is feasible, it also enables identification of the best methodologies for age dating. The availability of age dating reference materials will help validate current and future age dating protocols, leading to a more robust source of nuclear

forensic signatures and a legally defensible basis for the use of age dating results in nuclear forensics investigations. Validation of these methods will increase their relevance and applicability as part of the tool-kit available for nuclear forensics investigations.

References

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