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Measurement of Sulphur Isotopic Ratio for the Nuclear Forensic Investigation of Uranium Ore Concentrates (Yellow cakes)

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As an answer to the illicit trafficking of nuclear materials in the 1990s a new scientific topic has emerged, commonly referred to now as nuclear forensics (1). The aim of the nuclear forensic investigations is to identify the hazard and origin of the confiscated or found nuclear materials and ultimately strengthen security measures and prevent nuclear terrorism thereafter. Over the last few years several signatures of nuclear materials have been investigated and developed to establish the links between the measurable parameters of the unknown material in question and the source of the nuclear materials. These measurable parameters or signatures include e.g. elemental or anionic impurities, isotopic composition, structural analysis, morphology and age determination. This complex dataset can give information about the source of uranium ore or feed materials, process and the production facility. Uranium ore concentrate (commonly known as yellow cake) has a special role among the investigated nuclear materials, as it is the first purified industrial product of nuclear fuel fabrication, and thus it is highly useful to identify the source and propagation of various applicable signatures.

The sulphur isotope abundance shows relatively high variation in nature due to the large relative mass difference between its isotopes, the variety of chemical forms and the widespread occurrences in nature (2). Typically, natural materials with oxidized sulphur have d34S values between +5 ‰ and +25 ‰, while for materials with reduced sulphur it ranges between -5 ‰ and +15 ‰. The sulphur isotope ratio in uranium ore deposits is also reported to exhibit large variation, such as in the sandstone-type uranium deposits of the Colorado Plateau and Wyoming (-20.5 to -17.8 ‰) or in the uranium roll-type deposit in South Texas, USA (-25 to -40 ‰). However, as the sulphur content in the nuclear material derives not only from the feedstock (ore), but is also introduced into the process stream as process chemical (e.g. as H2SO4 with an approximate d34S value of -5 to +15 ‰), its contribution to the final d34S value in the product has to be considered. Therefore, it is expected that sulphur isotopic composition can be indicative both for the process (chemicals used) and the ore type depending on the hydrometallurgical production route.

In order to investigate if the sulphur isotopic composition is a meaningful signature in nuclear forensics, a novel method has been developed and validated for the measurement of 34S/32S isotope ratio in uranium ore concentrates (yellow cakes) (3). The developed ion exchange separation method effectively separates and preconcentrates sulphate from uranium and the possibly interfering matrix components, such as cations. The measurement was performed by multi-collector inductively coupled plasma mass spectrometry.

Determination of 34S/32S ratio in uranium ore concentrates of world-wide origin showed significant differences between the samples (3). This variation can be exploited to differentiate samples of different origin, for instance to verify or exclude an assumed origin. Moreover, as the 34S/32S ratio can be indicative of the feed ore used for the production in several instances, the uranium ore deposit type can be identified, which can make this signature highly valuable to provide clue on the provenance of unknown nuclear materials, thus trace it back to its source. Further studies are on-going to reveal the dominant source of sulphur in the uranium ore concentrate (ore vs. process chemicals), to investigate the sulphur isotope ratio in the course of the industrial processes and to find further correlations between the d34S value in the ore concentrate and the deposit type (geolocation).

References

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