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Use of Samarium mass-independent isotope signatures in natural and transformed uranium samples: a promising tool in nuclear forensics

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Developing new tracers which constrain the origin and formation conditions of terrestrial samples is a major challenge in nuclear forensics. Here we explore the potential of mass-independent isotope anomalies to complement the insights provided by mass-dependent fractionation (MDF). Mass-independent signatures may provide additional clues for determining the provenance of samples of uranium ore and uranium ore concentrates, and ultimately help trace the U deposit from which they were extracted.

Samarium exhibits limited MDF signatures in natural samples. However, due to the properties of its isotopes, Sm is a particularly suitable candidate for observing a diverse range of mass-independent anomalies: (i) the Nuclear Field Shift Effect (NFSE) caused by differences in the mean charge radius of its isotopes; (ii) uranium fission, with several Sm isotopes (^{147}Sm, ^{149}Sm, ^{152}Sm, ^{154}Sm) being fission products and (iii) neutron capture by ^{149}Sm, which has one of the largest thermal neutron capture cross sections. Furthermore, the correction for instrumental mass bias, using the exponential law, leaves a residual mass-dependent component as the sample may follow other MDF laws, resulting in a fourth signature which is referred to as apparent MIF.

We determined the 7 stable isotopes of Sm in a variety of uranium ores and uranium ore concentrates using a Thermo-Ionisation Mass Spectrometer and deconvolved the isotope signature of each sample using the four components described above. While most samples' isotope signatures are dominated by neutron capture, other mass-independent processes can be identified in some samples. Thus the mass-independent Sm isotope signatures proves to be efficient for distinguishing the U deposits where the samples originated from, even in ore concentrates. Combined with the mass-independent signatures of other elements having different behaviors during ore formation, such as Mo, this tracer is a promising tool in nuclear forensics and could also be used as an environmental tracer in other fields of geochemistry.