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143Nd/144Nd Ratio –a Powerful Signature for Origin Assessment of Natural Uranium Products

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Since the early 1990s illegal possession, transfer and other unauthorised acts involving nuclear materials have taken place. In order to identify the hazard, intended use and origin of the illicitly trafficked nuclear materials, various analytical methods using including radiometry, mass spectrometry and electron microscopy have been developed for nuclear forensics during the last years/decades. Among the characteristic parameters that can be determined by the above-mentioned methods, the concentration and isotopic composition certain impurities of uranium materials have been found highly indicative of the feed material, production facility and its location as well as the last chemical processing date of the nuclear material (1).

Up to now the isotopic patterns of O, Pb, Sr, S, Th, and U have been investigated (2,3). Also the pattern of the rare-earth elements (REE) has been studied and found characteristic to the geological formation of the uranium deposit and their nuclear products. Due to the similarity in chemical properties, the relative abundances of REE remain mainly unaltered through the processing in the front-end of the nuclear fuel cycle (4). Besides the REE pattern, the 143Nd/144Nd isotope ratio is widely used in geology for chronometry and provenance measurements. The 143Nd amount varies in nature due to the 147Sm decay (T1/2=1.06 * 1011 a) to 143Nd. As 144Nd is neither radioactive nor radiogenic, the number of 144Nd amounts does not change with time due to radioactive decay, making it suitable as a reference isotope. Therefore, the 143Nd/144Nd ratio is indicative of the geological condition, hence together with the other characteristics of the material in question, the Nd isotope ratio can be a promising signature for nuclear forensics. The measurement of 143Nd/144Nd isotope ratio in uranium samples is, however, an analytically challenging task, as the concentration of Nd is very low in nuclear samples (< ppb level) due to the effective purification processes. Therefore, pre-concentration of REE prior the measurement by inductively coupled plasma mass spectrometry (ICP-MS) is required (5).

In the present paper the possibility of 143Nd/144Nd isotope ratio for provenance assessment in nuclear forensics is evaluated. For this purpose a novel procedure has been developed for the determination of the 143Nd/144Nd isotope ratio and the Nd/Sm ratio in various uranium-bearing materials, such as uranium ores, ore concentrates (yellow cakes) and high-purity uranium oxides (UO3, UO2). The procedure comprises pre-concentration of REE from the uranium matrix, Nd/Sm separation and further concentration by extraction chromatography. Subsequently, the REE were measured by double focusing inductively coupled plasma mass spectrometry (ICP-MS) and the Nd isotope ratio was determined by multi-collector inductively coupled plasma mass spectrometry (MC-ICP-MS). The results of uranium sample analysis of world-wide origin are shown and correlations between the determined 143Nd/144Nd ratio, Nd/Sm ratio and deposit type are presented.

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