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Comprehensive characterization of Single Hot particles: from isotopic composition to bioavailability

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When on April 26, 1986 the reactor of the Chernobyl nuclear power plant exploded, nuclear fuel was in part released as microscopic solid particles. Structurally intact hot particles pose a risk to humans mainly by the (unlikely) path of inhalation. However, over the decades, weathering may lead to considerable release of radionuclides that subsequently can enter the human food chain. Since hot particles differ strongly with respect to morphology, chemical composition and stability, investigations need to be performed on single particles, rather than on bulk samples.
We separate and extract the micrometer sized particles from the environmental matrix in a scanning electron microscope (SEM) equipped with a micro manipulator. Subsequently, the particles are fixed by SEM glue on tungsten needles made in-house. Element composition is imaged by EDX-measurements, and radioactive isotopes are detected by gamma spectrometry. To selectively image the elemental and isotopic composition at below 100 nm spatial resolution, we combine secondary-ion mass spectrometry with resonant laser ionization. Identification of actinide elements and fission products is possible at ultra-trace levels, including Am-242m and Cm. Isobaric interferences, particularly U-238/Pu-238 and Pu-241/Am-241, are suppressed by up to five orders of magnitude.
In contrast to most mass-spectrometric techniques, only negligible mass is consumed, leaving the particle intact for further studies. Selected individual particles were sequentially leached in ammonium acetate solution, hydrochloric acid, oxalic acid, nitric acid and aqua regia. The solutions are then measured via gamma spectrometry and ICP-MS to analyze their uranium content and the amounts of leached fission and breeding products. Fuel particles from the Chernobyl accident are usually classified according to their appearance, dissolution rates and oxidation state: the further oxidized and brittle UO_{2+Z}
, non-oxidized UO_2 and zirconium merged bearing U_X -Zr_Y-O_Z particles, of which we investigated at least one particle each.
The first leaching steps did not attack the structure of the particles, and only small fractions of Am-241 and Cs-137 were leached under mild conditions, indicating a low bioavailability. Leaching behaviors were mostly in line with the predictions based on the visually assigned classes and oxidation states. From these data, conclusions on potential risks to human health and environment are drawn. It also demonstrates the potential of a technique that is applicable to almost all elements and opens up new scientific applications beyond the nuclear field.

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