

Determination of plutonium in environmental samples using AMS

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A reliable and sensitive method for the determination of plutonium in environmental samples by accelerator mass spectrometry (AMS) is presented. It allows, in particular, to investigate the different sources of environmental plutonium, such as from atmospheric weapons testing, nuclear power production, reprocessing of nuclear fuel and accidents in nuclear facilities. The procedure consists of the basic steps: sample dissolution, radiochemical separation of the plutonium isotopes, and their measurement by AMS and alpha spectrometry.

AMS was especially chosen in addition to alpha spectrometry, because it allows the determination of ²³⁹Pu/²⁴⁰Pu isotopic ratios as well as detecting ²⁴¹Pu [1]. By combining AMS with alpha spectrometry, all relevant plutonium isotopes can be measured. Thereby a reliable distinction of different sources of plutonium in the environment is possible. Further advantages of this technique are short analysis times and high sensitivity. The detection limits of AMS are much lower than those of alpha spectrometry are.

The demands for sample preparation of both, alpha spectrometry and AMS, are met by extraction chromatography. Special attention in the radiochemical separation scheme is paid to remove impurities of ²³⁸U, which disturb the ²³⁹Pu determination via AMS. Best results for radiochemical separation and uranium decontamination are achieved by using TEVA® resin columns, manufactured by Eichrom Technologies [2].

The presented analytical method was especially designed for the analysis of plutonium in soils and in water. Appropriate acid leaching and precipitation techniques were chosen for the two matrices, respectively. The radiochemical yield was determined by addition of ²⁴²Pu as a tracer.

In the sample solution, the oxidation state of the plutonium isotopes is adjusted to +IV with NaNO₂. The prepared solution is loaded on a TEVA® resin, which contains an aliphatic quaternary amine. Several steps of purification are carried out prior to the elution of plutonium with 5 ml diluted hydroxylamine (*c* = 0.1%). The volumes and flow rates of the eluates were varied to optimize the separation and speed of the method.

After the radiochemical separation, the plutonium is electrolytically deposited on a stainless steel disc and measured by alpha spectrometry to determine ²³⁸Pu, which cannot be detected by AMS. After that, the plutonium is leached from the disc with 10 ml 3 M HNO₃ and converted to targets suitable for the measurement by AMS. Thus, this procedure enables the measurement of the same sample with two different analytical methods.

Reference materials such as IAEA-381 (Irish Sea Water), IAEA-375 (soil from Brjansk, Russia) and IAEA-6 (Austrian soil) were analyzed to validate this method.

Samples from the highly contaminated areas near Chernobyl (Ukraine), which were earlier analyzed [3], were re-investigated using the new procedure involving alpha spectrometry and AMS. Good correspondence of the results, obtained by both methods was observed.

The instrumental effort of AMS is much higher than for alpha spectrometry, but the low detection limits, the possibility to distinguish the isotopes ²³⁹Pu and ²⁴⁰Pu and to receive information about ²⁴¹Pu makes efforts worthwhile. This is in particular true since the plutonium AMS measurements were performed with a newly developed compact AMS machine, at a terminal voltage of 300 kV [4].

The measured ²³⁹Pu/²⁴⁰Pu isotopic ratios of the analyzed samples provide information on the origin of the plutonium. Thus, the plutonium emitted in the environment from Chernobyl accident can be distinguished from the plutonium of the global fallout or other sources. In addition, the differentiation of plutonium fractions offers the possibility to study the dispersion and mobility of plutonium in the environment.

- [1] L. Wacker et al., Nucl. Instr. and Meth. B 2005, 240, 452–457.
- [2] Eichrom Technologies: Americium, Plutonium, Thorium, Curium and Uranium in Water (with Vacuum Box system). Rev. 1.0 (ACW16). 2006.
- [3] S. Hippler, Strahlenexposition durch anthropogene Actinoide in der nördlichen Ukraine, PhD Thesis, University Hannover 2006.
- [4] L.K. Fifield, H.-A. Synal, M. Suter, Nucl. Instr. and Meth. B 2004, 223-224, 802.