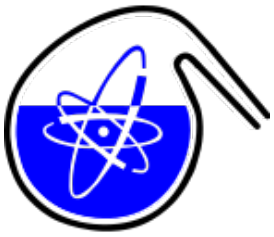

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Radiochemcial analysis of long-lived radionuclides using mass spectrometry: New progress in the methodology

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Human nuclear activity has produced a large amount of radionuclides, some of them was released to the environment, while most of them are remained in various radioactive waste to be deposited. Among these radionuclides, long-lived radionuclides such as 14C, 36Cl, 79Se, 99Tc, 126Sn, 236U, 237Np, and isotopes plutonium, neptunium, americium and curium are most important in view of waste depository, decommissioning, environmental radioactivity and tracer studies of environmental processes. In addition, natural process also produce many radionuclides, such as 14C, 10Be, 26Al, 36Cl, 129I, 210Pb, 226Ra, these radionuclides are also important for radiation risk, especially in geological dating and environmental process investigation.
Radiometric methods based on counting of radioactive decay is conventional methods for measurement of radionuclides, especially for short-lived radionuclides, but the sensitivity is not sufficient for measurement of long-lived radionuclides (> 10 ky) in low-level. Mass spectrometry typically used for measurement of stable isotopes by counting the number of atoms of nuclides is suitable for measurement of long-lived radionuclides. However, the interferences from the samples matrix, isobars, polyatomic ions, tailing of high-level stable isotopes with a m/z close to the analyte radionuclide are the major challenge in its application for the determination of long-lived radionuclides of low-level samples. In the past years, mass spectrometry, especially accelerator mass spectrometry (AMS) and ICP-MS analytical techniques has been developed very rapidly, which significantly benefit to the determination of long-lived radionuclides, and stimulated its application in many research fields.
In our laboratory, we have developed serious analytical methods for the determination of long-lived radionuclides in environmental and waste samples by using radiochemical separation for removal interferences and AMS and ICP-MS for their sensitive measurement. An overview of these methods will be presented, with a focus on the following methods. (1) A novel ICP-MS method for determination of plutonium isotopes (239Pu, 240Pu, 241Pu) by using sequential mass spectrometers and reaction/collision cell. A removal efficiency of uranium of up to 1E8 has been achieved by employing a mixed reaction gasses to sufficiently removal of uranium and significantly improve the sensitivity of plutonium. (2) A sensitive AMS measurement method for determination of ultra-low level of 129I and 236U with microgram carrier isotopes (127I and 238U, respectively). New radiochemical separation and target preparation techniques were developed enable to prepare suitable and effective targets. (3) A novel AMS measurement method for rapid directly determination of 14C and 129I in environmental samples without chemical separation.

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