Development of Analytical Methods for Measurement of Radioactive Elements

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Introduction

As the consequence of the nuclear accident due to The Tohoku Earthquake and tsunamis, a vast area of Fukushima was contaminated by radioactive materials releasing from Fukushima Daiichi Nuclear Power Plant (1F). Technical support for environmental restoration of Fukushima and decommissioning of 1F are the most important mission of Japan Atomic Energy Agency (JAEA). In this presentation, we outline Okuma Analysis and Research Center and our mission on technical development for analyzing rubble and wastes from the decommissioning of 1F. Analytical methods are being developed at the center for determining difficult-to-measure nuclides.

Outline of Okuma Analysis and Research Center

The Okuma Analysis and Research Center has been established near the 1F site including an administrative building and two laboratories. Presently, the administrative building is in operation and the first laboratory (Laboratory-1) is under construction. The laboratory-1 will analyze low and middle level rubble and wastes by basically radiometric methods. Other alternative methods are being developed.

Isotopes for determination are listed in table 1. Gamma spectrometer using an HPGe detector will be used for seven isotopes, alpha spectrometer for mainly actinide isotopes and beta counter instruments, such as liquid scintillation counter, for beta measurement. Since beta counting instruments are not enough selectivity to measure each isotope concentration based on difference of its beta energy, chemical purification process is necessary before instrumental measurement.

Table 1: Isotopes of interest and analytical methods

Type of decay	Analytical instrument (candidate)	Nuclide (Half-life in year; Energy in MeV)
Beta	Liquid Scintillation counter or Low background gas flow counter	H-3(12.3; 0.019), C-14(5.7e3; 0.16), Cl-36(3.0e5; 0.71), Ni-63(100; 0.067), Se-79(3.0e5; 0.15), Sr-90(29; 0.55), Zr-93(1.5e6; 0.061), Tc-99(2.1e5, 0.29), Pd-107(6.5e6; 0.033), Sn-126(2.3e5; 0.25), 129-I(1.57e7; 0.154), Cs-135(2.3e5; 0.27), Sm151(90; 0.077), Pu-241(14; 0.021)
	Gamma Spectrometer	Co-60(5.3; 1.2, 1.3), Nb-94(2.0e4; 0.71,0.87), Sn-126(2.3e5; 0.088), Cs-137(30; 0.66), Eu-152(14; 0.344), Eu-154(8.6; 0.12), Am-241(4.3e2; 0.060)
Alpha	Alpha spectrometer	U-233(1.6e5; 4.8), U-234(2.5e5; 4.8), U-235(7.0e8; 4.4), U-236(2.3e7; 4.5), U-238(4.5e9; 4.2), Np-237(2.1e6; 4.8), Pu-238(88; 5.5), Pu(239(2.4e4; 5.2), Pu-240(6.6e3; 5.2), Pu-242(3.8e5; 4.9), Am-241(4.3e2; 5.5),Am-243(7.4e3; 5.3), Cm-244(18; 5.8), Cm-245(8.4e3; 5.4), Cm-246(4.8e3; 5.4)
Electron Capture	Low Energy g-ray/X-ray spectrometer	Ca-41(1.0e5), Ni-59(1.0e5), Mo-93(4.0e3)

ICP-QQQ-MS And Automated Sample Preparation System.

Inductively Coupled Plasma Mass Spectrometry (ICP-MS) is one of the most powerful analytical instruments with high sensitivity and short analysis time. Suitable analytical methods utilizing inductively coupled plasma triple quadrupole mass spectrometer (ICP-QQQ-MS) are being developed at our laboratory. The developed techniques are aimed at difficult-to-measure nuclides with long half-life. The inherent challenge of isobaric interference in a regular ICP-MS is expected to be suppressed using two tandem mass filters and a collision reaction cell that are equipped in an Agilent 8900 ICP-QQQ-MS. We are investigating optimal conditions to separate inferring ion pairs such as (Ni-59, Co-59), (Ni-63, Cu-63), (Se-79, Br-79), (Sr-90, Zr-90, Y-90), (Zr-93, Nb-93, Mo-93), (Pd-107, Ag-107), (I-129, Xe-129) and (Cs-135, Ba-135). Some preliminary results were achieved. Optimal conditions for the measurements are being further investigated.

Analysis of Short Half-Life Nuclides by Automated Sample Preparation System

To simplify and automate measurement instruments, we are developing an automated sample preparation system. The system is a combination of all equipment that performs basic steps of sample treatment including sample digestion, filtration, solid phase separation, etc. The system is expected to minimize the manipulation of human operators so that minimizing radiation exposure and variations of analytical results associated with one’s manipulations.

Since ICP-MS is not sufficiently sensitive for the determination of short half-life nuclides, radiometric method should be applied to the measurement. Before radiometric measurement, nuclides of interest, for example Ni-63 (half-life of 100 years) and Sr-90 (half-life of 29 years), must be separated and purified with a long and complicated separation/purification process. We planned automated system to deal with the separations for a precise and safe measurement, even if increasing number of analytical samples.

The sample preparation automation has been developed using robotic system. The robots can resemble one’s manipulations to perform the same analytical manuals. Various automated analysis instruments are designedly assembled in one unit, which is aimed at one specific separation scheme such as for Sr purification. Recently, fully automated analysis using microchips is available; what man should do is just setting/injecting samples. These systems are suitable for routine sample preparation of similar samples, and for where preparation process should be fixed. However, at Okuma analysis research center, analysis process will be adjusted to the sample condition after the operation, because, sampling plan should be decided the decommissioning processing steps. Therefore, the automated pre-treatment system should be flexible in design to respond the change of analytical process.

Acknowledgments
This presentation includes results obtained under the entrusted research program of the Agency for Natural Resources and Energy, the Ministry of Economy, Trade and Industry of Japan to the International Research Institute for Nuclear Decommissioning of which JAEA is a member.