$^{90}Sr/^{88}Sr$ ratio analysis with extremely low noise: An application of energy filtered-TIMS

*Shigeyuki Wakaki[1], Jo Aoki[2], Ryoya Shimode[2], Katsuhiko Suzuki[3], Takashi Miyazaki[4], Jenny Roberts [5], Hauke Vollstaedt [5], Satoshi Sasaki[6], Yoshitaka Takagai[2,7]

1. Kochi Institute for Core Sample Research, JAMSTEC, 2. Faculty of Symbiotic Systems Science, Fukushima University, 3. Submarine Resources Research Center, JAMSTEC, 4. Volcanoes and Earth's Interior Research Center, JAMSTEC, 5. Thermo Fisher Scientific Bremen GmbH, 6. Thermo Fisher Scientific K.K, 7. Institute of Environmental Radioactivity, Fukushima University

Strontium-90 is a major anthropogenic radionuclide produced by fission of ^{235}U and ^{239}Pu. Because of the potential threat of long-term internal exposure, emission and subsequent diffusion of ^{90}Sr into local environment due to nuclear reactor accidents should be quantitively monitored. Conventional radiometric ^{90}Sr analysis requires relatively large sample size and long analysis time (e.g. 2 weeks), which are not adequate for urgent environmental survey. Recently, rapid and sensitive ^{90}Sr analysis methods were successively developed utilizing various state-of-the-art mass spectrometric techniques, such as ICP-MS (e.g. Takagai et al., 2014), ICP-MS/MS (Ohno et al., 2018), TIMS (Kavasi and Sahoo, 2019), AMS (Sasa et al., 2020) and ID-TE-TIMS (Ito et al., 2020).
In this study, we demonstrate a novel ^{90}Sr analysis method based on isotope ratio analysis using energy filtered-TIMS (Wakaki et al., 2022). Strontium isotope ratios were measured at Fukushima university using Triton Plus (Thermo Scientific). Stable Sr isotopes were collected with faraday cup detectors. ^{90}Sr were detected by a secondary electron multiplier mounted with a set of RPQ lenses, which is an energy filtering device effective for both high and low mass side peak tailing. Approximately 1 h of measurement time with ^{88}Sr ion beam of ca. 2.6×10^{-10} A requires sample amount of >100 ng of Sr.
Typical noise sources of mass spectrometric ^{90}Sr measurement are isobaric interreference of ^{90}Zr and peak tailing of the major ^{88}Sr ion. With multiple noise reduction schemes, including the use of RPQ lenses, we have succeeded to lower the noise level on ^{90}Sr to ca. 0.004cps level. Abundance sensitivity, defined as $^{90}Sr/^{88}Sr$, of our method was estimated from the analysis of the ^{90}Sr-free NIST SRM 987 reagent (blank sample) as $8.3 \pm 1.8 \times 10^{-12}$. Detection limit of $^{90}Sr/^{88}Sr$ ratio, defined as 3 σ of the blank analyses, was estimated as 2.7×10^{-12}. With a minimum sample amount of 100ng of Sr, this detection limit of the $^{90}Sr/^{88}Sr$ ratio corresponds to a ^{90}Sr activity of 0.0012 mBq. The ^{90}Sr detection limit of our method is superior to the previous mass spectrometric ^{90}Sr detection methods by two to three orders of magnitude. The accuracy of this method to measure the $^{90}Sr/^{88}Sr$ ratio on the 10^{-11} order was demonstrated by the analysis of biological samples from Fukushima and certified reference materials IAEA 156 and IAEA 330. High sensitivity and short measurement time of this method are suitable for urgent environmental survey and also for studying environmental diffusion of ^{90}Sr using size-limited samples.

References: Ito et al., 2020, Anal. Chem. 92, 16058–16065; Kavasi and Sahoo, 2019, Anal. Chem. 91, 2964–2969; Ohno et al., 2018, J. Anal. At. Spectrom. 33, 1081–1085; Sasa et al., 2020, J. Nucl. Sci. Tec. 58, 72-79; Takagai et al., 2014, Anal. Methods 6, 355–362; Wakaki et al., 2022, Sci. Rep. 12, 1151.

Keywords: Strontium-90, mass spectrometry, radionuclide, 90Sr/88Sr ratio

