1–13 Rapid Analysis for Long Half-Life ^{99}Tc

—ICP-MS Analysis with Solid-Phase Extraction and Gas-Phase Reaction—

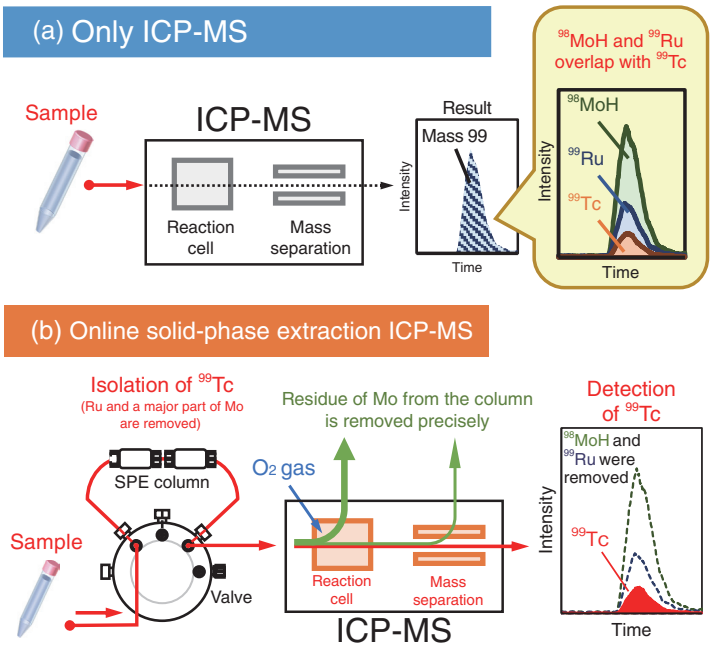

Fig.1-25 (a) Typical inductively coupled plasma–mass spectrometry (ICP-MS) analysis and (b) proposed online solid-phase extraction ICP-MS

The precise quantification of ^{99}Tc is prevented because of overlapping with the same mass of ^{99}Ru and ^{98}MoH. The proposed method involves the separation of ^{99}Ru and ^{98}MoH to determine ^{99}Tc correctly.

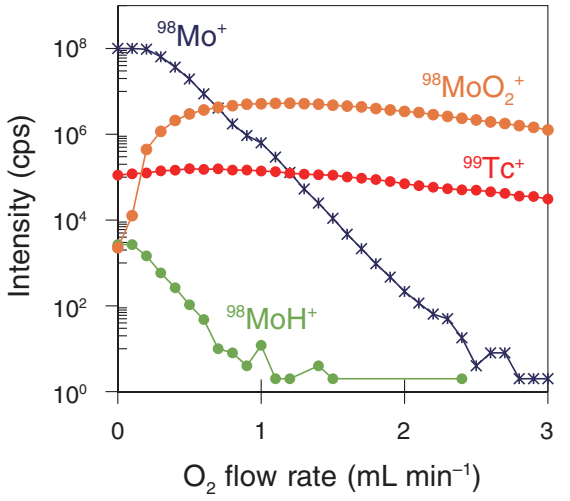

Fig.1-26 Gas-phase reaction of oxygen and elements (^{98}Mo and ^{99}Tc) in the reaction cell of ICP-MS

^{98}Mo and ^{98}MoH decreases logarithmically and are converted $^{98}MoO_2$ with increasing O_2 content, whereas the ^{99}Tc remains constant. O_2 reaction enables mass discrimination, and this method supports the precise quantification of ^{99}Tc in samples containing enriched ^{98}Mo.

Technetium-99 (^{99}Tc) is an artificial radionuclide with a long half-life of 210,000 years and a pure beta emitter. ^{99}Tc is produced as one of the fission products and in yields equal to the yields of cesium-137 and strontium-90 (approximately 6%) from nuclear material such as uranium-235. Specific seaweeds accumulate ^{99}Tc, although they transport it widely in the marine environment. The concentration of ^{99}Tc in seaweeds and the annual discharge amounts in nuclear-reprocessing plants in foreign countries were reported to have a reasonable correlation. Similarly, it is expected that ^{99}Tc will become one of the important tracers for monitoring the transport of radionuclides around the sea in Japan. However, there is no official analytical method for ^{99}Tc in Japan. In this study, we developed an analytical method to understand ^{99}Tc transport in the environment.

The radioanalytical method used for ^{99}Tc (beta counting) needs the separation of all other beta nuclides–this pretreatment process is complicated and time-consuming. The analysis is concentrated on a large sample volume to detect an ultratrace level of ^{99}Tc because of the low sensitivity compared with mass spectrometry. In this study, an analytical method using inductively coupled plasma–mass spectrometry (ICP-MS) to measure ^{99}Tc with a small sample volume was developed. ICP-MS enables rapid and highly sensitive determination of specific mass numbers (in this case, mass number 99 is selected). However, the quantification of ^{99}Tc with typical ICP-MS analysis is prevented by coexisting signal-overlapping materials, namely, ruthenium-99 (^{99}Ru) and ^{98}MoH (created from molybdenum-98 (^{98}Mo) and hydrogen in the equipment), which have the same mass number as ^{99}Tc (Fig.1-25(a)). Therefore, we developed an analytical system equipped with multistage separation with a solid-phase extraction (SPE) column packed with a selective adsorption resin for ^{99}Tc, and we adopted a gas-phase reaction with oxygen in the reaction cell of ICP-MS for the effective separation of the interfering materials (Fig.1-25(b)). The SPE column eliminated ^{99}Ru and ^{98}Mo, and their concentrations decreased to values lower than 1/4000 and 1/3000, respectively. However, the concentrations of ^{99}Ru and ^{98}Mo are 400 times and 1.6 billion times higher than that of ^{99}Tc in the Japan Sea. Thus, the required removal rate of ^{98}Mo was not achieved. Then, the gas-phase reaction with oxygen in the reaction cell of ICP-MS discriminated precisely between ^{99}Tc and ^{98}Mo by converting ^{98}Mo and ^{98}MoH to $^{98}MoO_2$ (mass number: 130) via an oxidative reaction, while ^{99}Tc remained nonreactive (Fig.1-26). The synergistic effect of SPE and gas-phase reaction enables analysis of samples containing high concentration of ^{98}Mo, which has a concentration a trillion higher than that of ^{99}Tc.

We performed measurements for a seawater reference material (certified value: 159-250 mBq/L) using this proposed method, and the obtained values (200.1 ± 9.6 mBq/L) were in agreement with those of the reference material. The online SPE ICP-MS contributes to wide-area research of ^{99}Tc in the environment owing to the high-throughput analysis (analytical time: within 30 min per sample).

(Makoto Matsueda)

Reference

Matsueda, M. et al., Online Solid-Phase Extraction-Inductively Coupled Plasma-Quadrupole Mass Spectrometry with Oxygen Dynamic Reaction for Quantification of Technetium-99, ACS Omega, vol.6, issue 29, 2021, p.19281–19290.