Letter to the Editors

Application of Inductively Coupled Plasma Mass Spectrometry to the Determination of ^{99}Tc in Soil Samples[†]

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1. Introduction

Investigations on behaviors and effects of ^{99}Tc in the environment is essential to complete assessment of the environmental impact of the nuclear fuel cycle. The precise and accurate analytical technique, however, is still not sufficiently established. The radiochemical method of analysis followed by the direct activity measurement has the difficulties in complete separation of ^{99}Tc from the interfering ruthenium isotopes. Although neutron activation method has been developed to determine ^{99}Tc in environmental samples, there are still some problems that the measurement of short-lived ^{100}Tc (15.8 s) is difficult in the case of (n, γ) reaction, and that the detection limit (10^{-9} g) is rather poor in the measurement of $^{99\text{m}}\text{Tc}$ produced by (n, n') activation¹⁾.

Inductively coupled plasma mass spectrometry (ICP-MS) is a powerful analytical technique, and many advantages of it have been described in a number of studies²⁾. The main advantages

of ICP-MS are the simultaneous multi-element analysis for a large number of elements with very low detection limits, high precision and accuracy. Recently, many papers³⁾ have also reported on the application of ICP-MS to the analysis of various kind of samples for trace metals. In this paper, the authors described the approach of ICP-MS for determination of ^{99}Tc in soil samples.

2. Experimental

2.1 Apparatus

The ICP-MS apparatus used was a VG Isotopes (PlasmaQuad PQ 2 Plus) installed at Marubun Co., Tokyo. The nebulizer employed in the instrument was of the concentric type.

2.2 Reagents

Nitric acid used was of the super-analytical grade and stored in a Teflon bottle. Other reagents were of the analytical grade. The standard solutions for calibration were prepared by successive dilution of the stock solution (11 ng $^{99}\text{Tc}/\text{ml}$ in 0.1 M HNO_3) with 1 M HNO_3 . Deionized water with a resistivity greater than 12 M Ω /cm (Millipore) was used for the preparation of all sample solutions.

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2.3 Chemical separation

The air-dried soil sample taken at Okuetsu farm (Fukui Pref.) of 1 kg was sieved (<2 mm ϕ) and ashed at 450 °C for 6 h. The ashed sample was leached in 8 M HCl. The leached solution was filtered through a glass fiber filter (GA-100) on a Buchner funnel. The bulk of Fe in the filtrate was removed by isopropyl ether extraction. The aqueous layer was evaporated to dryness and dissolved in 1 M HNO₃. The solution was transferred in a separatory funnel and shaken with 5% TIOA-toluene. The technetium fraction was stripped with 1 M NaOH from the organic layer. The stripped solution was passed through an anion exchanger column (Dowex 1-X8, OH-form). The anion exchange column was washed with 1 M NaOH, 1 M HCl and 0.1 M HNO₃, successively. Finally, the technetium fraction was eluted with 8 M HNO₃. The eluate was made up to 100 ml with 8 M HNO₃. A 10 ml portion of the solution was introduced to ICP-MS measuring system.

2.4 Instrumental operation condition

Sample solutions were introduced to ICP-MS instrument by a concentric quartz nebulizer, with the solution uptake rate of 0.5 ml/min. The mass spectrometer was set to cover only the range of m/z 98.00–102.674. Data acquisition was carried out using a multi-channel analyzer (MCA). The MCA was set with a data acquisition memory group of 512 channels, a dwell time per channel of 80 μ s and 5800 separate sweeps. The measurements of the blank and each sample solution were respectively performed three times. Each sample solution was aspirated for the system equilibration for 0.5 min and then the mass spectra was recorded for 5 min.

3. Results and Discussion

The spectra for the standard solutions showed only the mass peak for ⁹⁹Tc without the interfering one. The peak of ⁹⁹Tc was clearly observed even in the case of the very low

Fig. 1 Calibration curve of ⁹⁹Tc.

Fig. 2 Mass spectrum of the soil sample.

concentration of 3.2 pg/ml. The calibration curve for ⁹⁹Tc showed a clear linearity in the range of the concentration from 3.2 pg/ml to 42 pg/ml as shown in Fig. 1. Figure 2 shows a mass spectrum for ⁹⁹Tc in the soil sample. The interfering peaks from molybdenum and ruthenium were also found in the mass spectra, showing that a higher degree of purification is required for the determination of ⁹⁹Tc by ICP-MS. By eliminating interference by molybdenum and ruthenium, ⁹⁹Tc in soil samples is expected to be determined by ICP-MS with high sensitivity, precision and accuracy.

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