High Precision TIMS Measurement of U030 with a Minimized Background Effect

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1. Introduction

Highly precise analysis of nuclear materials in swipe and environmental samples plays essential roles in monitoring undeclared nuclear activities. A thermal ionization mass spectrometry (TIMS) [1] combined with isotope dilution mass spectrometry (IDMS) technique was used to evaluate uranium samples in trace level. The preliminary experiments for minimizing the backgrounds from rhenium filaments showed that the degassing procedure under 3.5A for 1 hour is in the most effective condition. Additionally, the backgrounds from the process blanks, de-ionized water and nitric acid used to dilute samples, were measured to be negligible under the operating conditions. The quantification of U030 sample with IRMM-040a spike sample agreed with the certified value.

2. Methods and Results

2.1 Mass spectrometry

The measurement was performed with TIMS (TRITON, Thermo Co.) schematically described in Fig. 1. The differential pumping system consisting of a turbo-molecular pump and two ion-getter pumps maintained the ultra-high pressure of the system. In the thermal ionization source the double filament system evaporated, and then, immediately ionized the sample by extremely high temperature (~1800℃). The ions were analyzed by a magnetic sector followed by being transduced into electrical current by a combination of faraday cups and a secondary electron multiplier (SEM).

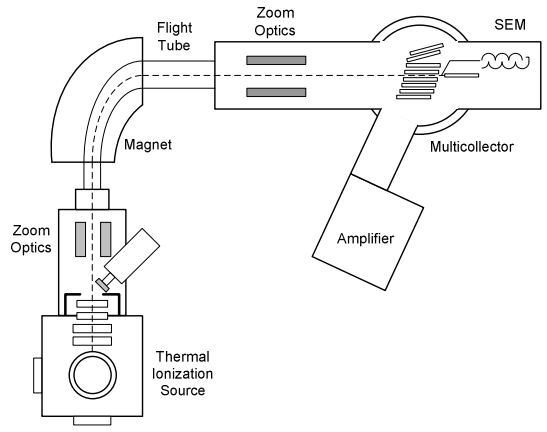

Fig. 1. The schematic diagram of TRITON TIMS

2.2 Degassing Procedure

The filaments prepared with zone-refined rhenium ribbon (0.04 mm and 0.7 mm in thickness and width, respectively, Thermo Co.) were degassed under a high vacuum condition (~10^{-7} mbar) at different heating currents (3.5A and 4.5A) and duration times (0 and 60 min.) [2]. The surface images of the filaments obtained by an optical microscope (LEICA, DC 100) showed no significant differences in appearance before and after the degassing procedure.

We measured the backgrounds from the filaments at the extreme condition with 3.5A for the evaporation filament current (E.F.). As shown in Fig. 2, the degassing procedure reduced the backgrounds from the filaments by an order of magnitude. Although the higher current (4.5A) eliminated more backgrounds, 3.5A was determined to be the most efficient heating condition considering the relatively small effect from the increment by 1.0A. Regarding the degassing time, 60 min duration was most effective in reducing the backgrounds.

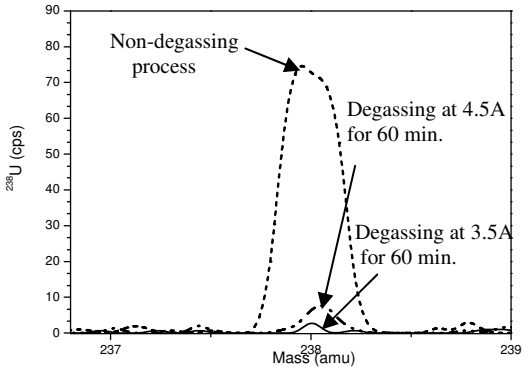

Fig. 2. The SEM scanning of ^{238}U signal depending on the degassing current and time

2.3 Determination of ^{238}U in a U030 sample

We measured the backgrounds from the filaments and the process blanks prior to the quantification of U030 reference sample. Although considerable amounts of the backgrounds were measured under the extreme filament heating condition described in the previous section, no noticeable backgrounds were observed under

the actual operating condition (E.F.=1.0A) with the detection sensitivity of the TIMS employed in this study.

The ^{238}U content in a U030 (NIST) sample was determined by IDMS with diluted IRMM-040a as a spike using the following equation [3, 4];

$$c(^{238}U, x) = \frac{R_y - R_b}{R_b - R_x} \cdot \frac{1}{R_y} \cdot \frac{m_y}{m_x} \cdot c(^{233}U, y) \quad (1)$$

where,
R_b : amount ratio n(^{233}U)/n(^{238}U) in the blend
R_x : amount ratio n(^{233}U)/n(^{238}U) in the sample
R_y : amount ratio n(^{233}U)/n(^{238}U) in the spike
m_x : mass of the sample
m_y : mass of the spike
$c(^{238}U,x)$: amount content of ^{238}U kg^{-1} in the sample
$c(^{233}U,y)$: amount content of ^{233}U kg^{-1} in the spike

In addition, the determination of isotopic ratios was carried out for the U030 reference sample. The estimated values of ^{238}U and the isotopic ratios in the sample were consistent with the certified value from NIST.

3. Conclusions

We varied the heating currents and duration times for the degassing procedure to minimize the backgrounds from the filaments. We observed that degassing the filaments at 3.5 A for 60 min reduced the backgrounds significantly. IDMS with IRMM-040a spike resulted in a reasonable agreement with the certified values for the U030 sample. Based on the current measurement, the TIMS system employed in this study is considered to be adequate to analyze nuclear materials in swipe and environmental samples with high precision.

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