Improving Uranium Sensitivity of TIMS using Graphite Powder Deposition on Rhenium Filaments

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In this study, we present a newly developed technique to improve thermal ionization mass spectrometry (TIMS) sensitivity at ultra-trace levels of uranium by depositing graphite powder onto rhenium filaments at high temperatures. A two-fold enhancement in uranium ion ($^{238}U^{+}$) intensity was observed when this technique was applied. The TIMS uranium ion intensity was monitored to investigate the evaporation behavior, ionization of uranium under graphite deposition conditions, and concentration dependence of graphite deposition. Isotopic analysis of uranium at ultra-trace concentrations was then performed to confirm the validity of our technique, given that our analysis agreed with certified values.

I. INTRODUCTION

The concentration of nuclear materials analyzed for nuclear safeguards is typically at ultra-trace levels. Swipe sample analysis, which is performed by the network of analytical laboratories (NWAL), associated with the International Atomic Energy Agency (IAEA), requires only a few nanograms (or less) of uranium for chemical analysis.[1] Although thermal ionization mass spectrometry (TIMS) offers high sensitivity, as well as excellent accuracy and precision for isotopic analysis, improving the sensitivity is essential to ensure the reliability of swipe samples analysis.

Studies towards improving TIMS sensitivity have focused mainly on the carburization of TIMS filaments, which refers to the pre-treatment of filaments by applying carbon using carbon-containing gases or solutions.[2-3] Carburization manipulates the electron work function of filament materials, resulting in enhanced ionization efficiency based on the Langmuir-Saha equation. Another possible effect of carburization is the chemical transformation of loaded samples. Uranium is changed into uranium carbide, which is easily evaporated upon heating and ionized as U+ at high efficiency. However, carburization using carbon-containing gases or solutions can be complicated, and includes baking filaments under a benzene atmosphere, which requires a special device.

In this study, graphite powder was used as a carbon source. Graphite powder is superior to other carbon sources, given its ease of use within the carburization process. A certified reference material of uranium at ultra-trace levels was used to optimize the carburization technique, and to investigate the effects of graphite carburization.

II. EXPERIMENTAL

A graphite powder suspension (approximately 2 μL) was added to filaments on which the uranium reference sample (~10 pg of U030; National Bureau of Standards, USA) was loaded. A thermal ionization mass spectrometer (TRITON Plus; Thermo Fisher Scientific, Bremen, Germany) was used to monitor the maximum 238U+ intensity. Determination of isotopic uranium ratios (n(234U)/n(238U), n(235U)/n(238U), and n(236U)/n(238U)) was performed using simultaneous measurement and continuous heating methods.[4-5]

III. RESULTS

The preliminary study determined that an optimum concentration of graphite powder suspension was prepared by vigorously mixing 0.005 g of graphite powder with 0.5 mL of 2 M nitric acid. A two-fold enhancement in the uranium ion intensity was observed using graphite powder carburization, as shown in Fig. 1. Isotopic uranium ratios at ultra-trace levels (~ 10 pg) were determined by TIMS using the carburized condition; these concentrations agreed well with certified values, demonstrating the validity of the approach.

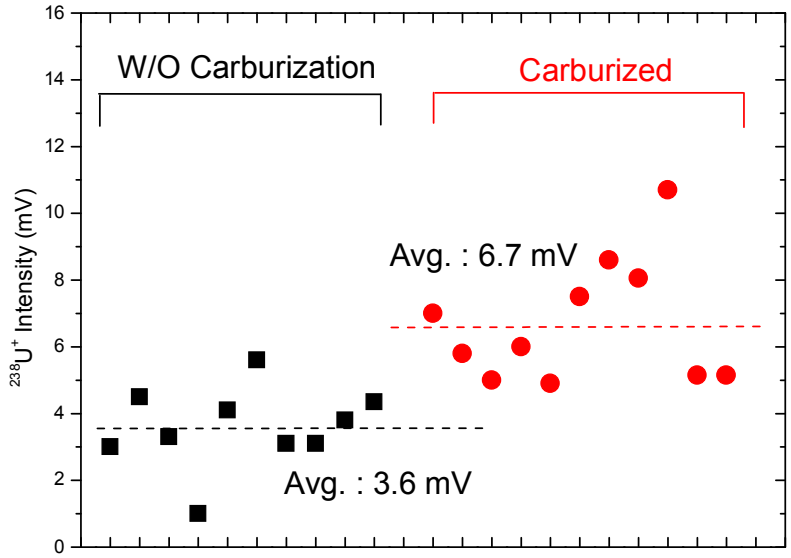

Fig.1. Effect of carburization using graphite powder

II. CONCLUSIONS

Carburization using graphite powder improved the uranium ion intensity of TIMS by a factor of two. Good agreement between measured uranium isotopic ratios and corresponding certified values shows that the graphite powder carburization technique is valid, with reasonable accuracy and precision down to ~10 pg of uranium.

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