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Ion source development for ultratrace detection of uranium and thorium ☆

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ABSTRACT

Efficient ion sources are needed for detecting ultratrace U and Th impurities in a copper matrix by mass spectrometry techniques such as accelerator mass spectrometry (AMS). Two positive ion sources, a hot-cavity surface ionization source and a resonant ionization laser ion source, are evaluated in terms of ionization efficiencies for generating ion beams of U and Th. The performances of the ion sources are characterized using uranyl nitrate and thorium nitrate sample materials with sample sizes between 20 and 40 μg of U or Th. For the surface ion source, the dominant ion beams observed are UO^+ or ThO^+ and ionization efficiencies of 2–4% have been obtained with W and Re cavities. With the laser ion source, three-step resonant photoionization of U atoms has been studied and only atomic U ions are observed. An ionization efficiency of about 9% has been demonstrated. The performances of both ion sources are expected to be further improved.

1. Introduction

Ultrasensitive analytical techniques are required for qualitative and quantitative analysis of the unwanted impurities in the materials constructing the detectors for ultra low-background experiments. A specific example is the Majorana Demonstrator (MJD) experiment [1] designed to search for neutrinoless double-beta-decay of the isotope ^{76}Ge. The MJD is a 40-kg detector array made from enriched ^{76}Ge and natural germanium and is being built to demonstrate the feasibility of a future one-ton-scale experiment. A specific goal of MJD is to achieve the ultra-low backgrounds, about *one count per ton-year in the region of interest*, required for the larger experiment. In order to achieve these backgrounds, the MJD detector is constructed and operated deep underground to avoid background from cosmic rays and the detector mounts and cryostats are made from ultra-pure copper electroformed and machined underground. Monte Carlo simulations indicate that uranium (U) and thorium (Th) impurities in this copper are expected to be the dominant sources of background and must be below the levels of a few 10^{-14} by weight for ^{238}U and ^{232}Th.

We have proposed to use accelerator mass spectrometry (AMS) to evaluate the U and Th impurities in the ultra-pure copper materials for MJD. AMS is presently the most sensitive and well established technique for trace elemental and isotope analysis, in comparison with other mass spectrometry techniques such as inductively coupled plasma mass spectrometry (ICP-MS), resonant ionization mass spectrometry (RIMS), and thermal ionization mass spectrometry (TIMS). It has been previously used to detect rare actinide isotopes with detection limits of 10^{-11}–10^{-12} isotope abundance ratios [2]. A major factor that limits the AMS sensitivity is the low ion-source efficiency, which is important for trace analysis. Cs-sputter negative ion sources are typically used for AMS and their ionization efficiencies are reported to be 0.01–0.1% for U [3–6].

This work is motivated by the need for more efficient ion sources for AMS analysis of U and Th impurities in a copper matrix at the trace levels required for the MJD experiment. Our approach is to

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use highly efficient positive ion sources which promise much higher ionization efficiencies for U and Th than Cs-sputter negative sources. This approach takes advantage of the available expertise and infrastructure of the former Holifield Radioactive Ion Beam Facility (HRIBF) [7] at the Oak Ridge National Laboratory (ORNL) where various positive ion sources were developed for the generation of ion beams of exotic nuclei. The positive ions were converted to negative ions by charge exchange and then sent to the 25-MV Tandem accelerator for acceleration to the desired energies for subsequent nuclear studies. The infrastructure at HRIBF is unique for AMS applications. The 25-MV tandem accelerator has the highest terminal voltage in the world. Charge stripping can be accomplished with thin carbon foils or a dilute gas in the terminal followed by a 180° magnet for charge-state selection. A number of ion sources are available to provide negative ion beams, including Cs-sputter negative ion sources and positive ionization sources in combination with a charge exchange cell. The capability of the HRIBF infrastructure for AMS application has been proven in a recent work [8] where fully stripped ^{36}Cl ions were obtained with the 25-MV tandem, thus effectively removing the ^{36}S isobars and pushing the detection limit of ^{36}Cl/Cl to a few times 10^{-16}. More information about the AMS system at HRIBF can be found in Ref. [9].

Two positive-ion sources are considered: a hot-cavity surface ionization source (HCSIS) and a hot-cavity resonant ionization laser ion source (RILIS). High temperature cavity ion sources have been used for more than 30 years at isotope separator on-line (ISOL) facilities for producing radioactive ion beams of elements of relatively low ionization potentials [10–12]. More recently, cavity-type surface ionization sources [13–18] are employed for TIMS systems for the purpose of improving the ionization efficiency and thus detection limits of low-level isotope analyses of elements such as U, Th, and Pu for which the conventional filament-type thermal ionization sources have very poor efficiencies. Measured ionization efficiencies of 3–8.5% for U [13,14,16,18] and 1–3% for Th [13,15] have been reported for those sources. The RILIS is developed for the production of isobarically pure radioactive ion beams (RIBs) at the ISOL facilities. It has been used, or demonstrated the ability, to provide ion beams of more than 40 elements in the periodic table with ionization efficiencies up to 40% [19,20]. An important advantage of the RILIS is its high selectivity for suppressing unwanted interfering ions. The two positive sources have been evaluated for their efficiencies in generating ion beams of U and Th. The design features and operating principles of the sources, the experimental setup, and the experimental results will be presented.

2. Hot-cavity surface ionization source

Atoms or molecules impinging on a hot metal surface may be emitted as atoms or ions, depending on the work function, Φ, of the metal surface and the first ionization potential, IP, of the atom or molecule. The degree of ionization, α, is given by the Saha-Langmuir equation:

$$\alpha = \frac{n_i}{n_0} = \frac{g_i}{g_0}\exp\left(\frac{\Phi - IP}{kT}\right). \tag{1}$$

where n_i and n_0 are the concentrations of the ions and neutrals leaving the surface and g_i and g_0 are the statistical weights of the ionic and atomic states, respectively, k is the Boltzmann's constant, and T is the temperature of the surface. The surface ionization efficiency, β, is defined as,

$$\beta = \frac{n_i}{n_i + n_0} = \frac{\alpha}{1+\alpha}. \tag{2}$$

From Eqs. (1) and (2) it is evident that high ionization efficiencies can be obtained for high work-function materials and low ionization-potential species such as the alkali metals. For the elements such as U (IP = 6.194 eV) and Th (IP = 6.307 eV) for which $IP > \Phi$, the surface ionization process is much less efficient. For example, the efficiency of ionizing U and Th on a tungsten surface (Φ = 4.54 eV) at T = 3000 K is only about 0.13% and 0.09%, respectively.

The degree of ionization predicted by Eq. (1) assumes that the ions are extracted as soon as they are formed. It has been observed that the efficiency of a surface ionization source can be substantially increased by the use of a high-temperature cavity with a small extraction hole [21,22]. For instance, ionization efficiencies of 4–39% were measured [23] for U with tantalum, tungsten, and rhenium cavities at about 2900 K. The mechanisms for the enhancement have been extensively studied [10,12,15,21–26] and will not be presented here in detail. Briefly, the neutral species undergo significantly higher number of wall collisions with the hot-cavity configuration and thus have a much larger probability to be ionized. In addition, a thermal plasma, consisting of surface ionized positive ions and thermionic electrons emitted from the hot walls, could form inside the hot-cavity. The plasma may reach near thermodynamic equilibrium with the cavity wall. Due to the high thermionic electron density at the wall, a negative plasma sheath potential with respect to the wall is created so that the positive ions can be confined to the plasma, preventing ion-wall collisions and thus ion losses. Therefore, the 'thermal' ionization efficiency can be much higher than that of surface ionization.

2.1. Description of the source

A cross sectional side view of the HCSIS assembly is displayed in Fig. 1, which shows the principal components: the hot-cavity ionizer, the sample tube, the graphite housing, and the extraction-electrode of the ion source. The hot-cavity ionizer is a cylindrical cavity of 3-mm inner diameter (ID), 30-mm long and 1-mm wall thickness. High work function and refractory materials such as Ta, W, and Re are tested as the ionizer material. The ionizer is connected to a closed-end Ta tube of 8.5-mm inner diameter and about 100 mm long, in which samples are placed at selected locations depending on the sample materials. The ionizer and the sample tube are heated resistively by passing a current through the tubular structure and can be heated to temperatures exceeding 3000 K in the ionizer. The sample materials in the sample tube are heated along with the sample tube and the ionizer. The volatile species effuse from the sample tube into the ionizer where they are ionized by surface ionization. The ions are extracted by the conical electrode.

2.2. Experimental results

The ion source was evaluated at the on-line test facility (OLTF) [7]. Positive ions extracted from the ion source were accelerated to about 40 keV energies and focused into a 90° dipole magnet with a resolving power of ~2000 for mass separation. The mass-selected ion beam current was measured with a Faraday cup after the magnet. The total ion current extracted from the ion source was also monitored with a Faraday cup before the dipole magnet.

The sample materials were made from 1000 ppm U or Th atomic absorption (AA) standard solutions, which contain U and Th in a nitric acid matrix in the form of uranyl nitrate and thorium nitrate, respectively. Small samples containing about 40 μg of U or Th atoms were used to evaluate the source. To make a sample, the selected 40 μL AA solution was dried on a thin Ti foil (0.0005 inch in thickness and about 5 × 6 mm in size) and then wrapped in the foil. The sample was placed in the sample tube of the source and heated when the ionizer and sample tube are heated. The sample tube was on average several hundred degrees colder than the ionizer. Ideally, the temperatures of the sample and the ionizer should

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