

# RESEARCH ON MEASUREMENT OF $^{126}\text{Sn}$ BY AMS\*

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## Abstract

A new method was developed for AMS measurement of  $^{126}\text{Sn}$ . Major features of the method include the use of  $\text{SnF}_2$  as target material, the selection of  $\text{SnF}_3^-$  molecular ions for extraction form from the target, and the transmission of  $^{126}\text{Sn}$  beam current. A sensitivity of  $(1.92 \pm 1.13) \times 10^{-10}$  ( $^{126}\text{Sn}/\text{Sn}$ ) has been reached by measuring a blank sample.

## INTRODUCTION

$^{126}\text{Sn}$  is a long-lived beta emitting radionuclide with a half-life of  $(2.35 \pm 0.07) \times 10^5$  a [1]. Being a fission product, the main natural production mechanism of  $^{126}\text{Sn}$  is through spontaneous fission of  $^{238}\text{U}$  and the abundance ratio of the  $^{126}\text{Sn}/\text{Sn}$  in the earth's crust is less than  $10^{-14}$  [2]. Artificially produced  $^{126}\text{Sn}$  has entered our environment through nuclear weapons testing and released from reprocessing plants and may locally lead to strongly enhanced  $^{126}\text{Sn}$  concentrations. Although the fission yield of  $^{126}\text{Sn}$  is lower by two orders of magnitude as compared to the well-known fission nuclides such as  $^{90}\text{Sr}$  ( $T_{1/2}=29.1\text{a}$ ) and  $^{137}\text{Cs}$  ( $T_{1/2}=30.2\text{a}$ ), the longer half-life makes it a more nasty nuclide to the environment. Therefore,  $^{126}\text{Sn}$  is a very important nuclear to monitor the nuclear contamination and the ultra-high-sensitivity measurement of  $^{126}\text{Sn}$  in rocks or soil samples from the fission environment would provide very important information for nuclear safety inspection. However, the concentration of  $^{126}\text{Sn}$  in the environmental samples as well as fission products is very low (below the detection limit of the ordinary mass spectrometry) and the interferences from the molecular ions and isobar  $^{126}\text{Te}$  are very strong. AMS technology is probably the best method for highly sensitive measurement of  $^{126}\text{Sn}$  thanks to its many advantages [3, 4, 5, 6]. However, some difficulties, especially isobaric interference from  $^{126}\text{Te}$ , still pose obstacles in AMS measurement of ultratrace  $^{126}\text{Sn}$ .

The first AMS measurement of  $^{126}\text{Sn}$  was performed at the Argonne ATLAS facility [7]. They claimed that  $^{126}\text{Te}$  can be separated from  $^{126}\text{Sn}$  at high energies ( $>400$  MeV) using a gas-filled magnet and a gas ionization chamber. However, the transmission efficiency and the optical path of  $^{126}\text{Sn}$  were derived from  $^{107}\text{Ag}$  simulation and are not very accurate. So it does not allow absolute measurements of  $^{126}\text{Sn}/\text{Sn}$ . An absolute measurement of  $^{126}\text{Sn}/\text{Sn}$  in

samples extracted from spent fuel rods of a nuclear power plant has been carried out by P.Gartenmann at the Zurich AMS facility [8, 9]. The terminal voltage of the tandem they used was comparatively low and the  $^{126}\text{Te}$  background was very strong, So the results of their measurements,  $^{126}\text{Sn}/\text{Sn}=(9.23 \pm 0.87) \times 10^{-6}$ , have relatively large uncertainty. There were two other problems in the above AMS measurements – insufficient accuracy due to the lack of calibration standards; and insufficient sensitivity caused by strong isobaric interference from  $^{126}\text{Te}$  introduced mainly by the extraction of  $\text{Sn}^-$  from the ion source. In this paper, a new method for AMS measurement of  $^{126}\text{Sn}/\text{Sn}$  at Beijing HI-13 AMS system is presented. Major features of the method include the usage of  $\text{SnF}_2$  target and the extraction of  $\text{SnF}_3^-$  molecular negative ions. A sensitivity of  $(1.92 \pm 1.13) \times 10^{-10}$  ( $^{126}\text{Sn}/\text{Sn}$ ) has been reached by measuring a blank sample.

## MEASUREMENTS OF $^{126}\text{Sn}$ WITH AMS BASED ON EXTRACTION OF MOLECULAR IONS

### Beam Current Extraction

The electron affinity of each element determines the negative ion yield from a sputter ion source and thus has an important role in any AMS measurements. Many elements or molecules can not form stable negative ions which is very helpful to effectively eliminate them when they are interference isobars of the nuclides of interest. There are two stable isobars for  $^{126}\text{Sn}$ , i.e.  $^{126}\text{Te}$  and  $^{126}\text{Xe}$ . Inert element Xe can not form a stable negative ion, therefore, the main interference for  $^{126}\text{Sn}$  AMS measurement is  $^{126}\text{Te}$ . We should find an appropriate target material and extracting corresponding molecular negative ions to depress the interference of Tellurium-126 as much as possible. So far,  $\text{Sn}^-$  atom ions were extracted from metal target in the measurement of  $^{126}\text{Sn}$  at some world-famous AMS laboratory [7,8]. By extracting  $\text{Sn}^-$  atom ions, large beam current (about 200nA) of  $\text{Sn}^-$  can be obtained. At the same time, however, a considerable beam current (about 2 $\mu\text{A}$ ) of  $\text{Te}^-$  will bring about a very strong background for  $^{126}\text{Sn}$  measurements. That is one of the main reasons that the sensitivities for AMS measurements of  $^{126}\text{Sn}$  were unsatisfactory in previous studies. In this work, six kinds of molecular negative ions of Sn ( $\text{Sn}^-$ ,  $\text{SnC}_2^-$ ,  $\text{SnO}^-$ ,  $\text{SnO}_2^-$ ,  $\text{SnF}_3^-$ ,  $\text{SnF}_5^-$ ), and correspondingly, the same ion forms of Te ( $\text{Te}^-$ ,  $\text{TeC}_2^-$ ,  $\text{TeO}^-$ ,  $\text{TeO}_2^-$ ,  $\text{TeF}_3^-$ ,  $\text{TeF}_5^-$ ) were tested. the extraction of

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either  $\text{SnF}_3^-$  or  $\text{SnC}_2^-$  molecular negative ions might effectively suppress the interference of Te, and the beam current of  $\text{SnF}_3^-$  is comparably larger (about 400 nA), while that of  $\text{TeF}_3^-$  is comparably weaker ( $<1$  nA). So, the extraction of  $\text{SnF}_3^-$  from ion source is the best choice for AMS measurement of  $^{126}\text{Sn}$ .

### Background

The  $^{126}\text{Te}$  background was measured with our AMS system by using a  $\text{SnF}_2$  commercial blank sample. A mixture of  $\text{SnF}_2$  and Ag powder with the mass ratio of 2:1 was pressed into aluminium cones, and then the target plate, on which all the aluminium cones were mounted, was baked at  $70^\circ\text{C}$  for 24 hours before it was put into the MC-SNICS (Multi-Cathode Source of Negative Ion by Cesium Sputtering) ion source.  $\text{SnF}_3^-$  negative ions were extracted from the cesium sputter source.  $^{126}\text{TeF}_3^-$  ions were selected by an injection magnet and injected into the accelerator. The charge state of  $11^+$  was selected by analyzing magnet after  $^{126}\text{Te}$  was stripped by a carbon foil stripper. The accelerator terminal voltage was chosen at 8.7 MV owing to the limitation of the electrostatic analyzer in our AMS system.  $^{126}\text{Te}$  counts were recorded by a multi-anode detector filled with 105 mbar P10 gas. As shown in Fig.1, a sensitivity of  $1.92 \times 10^{-10}$  can be reached, thanks to the effective suppression of  $^{126}\text{Te}$  interference by the extraction of  $\text{SnF}_3^-$  ions. The transmission of  $^{126}\text{Te}$  was previously simulated by the extraction and injection of  $^{124}\text{SnF}_3^-$ , and the acceleration of  $^{124}\text{Sn}^{11+}$  ions.

The procedure of simulation transport of  $^{126}\text{Sn}$  ( $^{126}\text{Te}$ ) is expounded as follow: Firstly, the sample material of  $^{124}\text{SnF}_2$  was used and  $^{124}\text{SnF}_3^-$  ions were extracted from ion source to simulate  $^{126}\text{Sn}^{11+}$  beam transport of sample material of  $^{126}\text{SnF}_2$ . In the simulation transport, the voltage of the terminal was set at 8.70 MV to make  $^{124}\text{Sn}^{11+}$  ions having the same energy as that of  $^{126}\text{Sn}^{11+}$  ions. The electric parameters of ions optics system were tuned for the optimum state of beam transport. Secondly, the voltage of the terminal was raise to 8.84 MV to make,  $^{124}\text{Sn}^{11+}$  ions having the same momentum as that of  $^{126}\text{Sn}^{11+}$  ions and the parameters of magnetic elements after analyzing magnet were further tuned for optimal statue of  $^{124}\text{Sn}^{11+}$  beam transport. The transmission efficiency from low-energy side Faraday-cup to detector was measured to be about  $4.0 \times 10^{-3}$  for  $^{124}\text{SnF}_2$  sample and  $^{124}\text{Sn}^{11+}$  ions. Thirdly, the voltage of the terminal was set back to 8.7 MV and the sample material of  $^{126}\text{SnF}_2$  was used and  $^{126}\text{SnF}_3^-$  ions were extracted from ion source and recorded by the multi-anode detector after passing through injection magnet, accelerator, analyzing magnet, electrostatic analyzer, and a  $7 \text{ mm} \times 7 \text{ mm}$  slit.  $^{126}\text{Sn}/\text{Sn}$  value would be determined by alternant measurements of  $^{124}\text{SnF}_2$  current at low energy side and the count rate of  $^{126}\text{Sn}$  on the detector at high energy side. The sensitivity of  $^{126}\text{Sn}/\text{Sn}$  is  $(1.92 \pm 1.13) \times 10^{-10}$ , given by the background from the measurement of a commercial  $\text{SnO}_2$  sample.

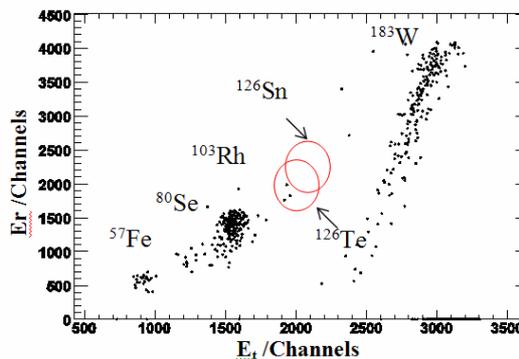


Figure 1: Two-dimensional spectra of a blank.

## CONCLUSIONS AND DISCUSSION

By experiments on extracting multiform negative molecular ions,  $\text{SnF}_3^-$  was found to be the most suitable form of ions in terms of maximizing the beam current of  $\text{SnF}_3^-$  while minimizing that of  $\text{TeF}_3^-$ . In addition,  $\text{SnF}_2$  was found to be the most suitable chemical form of target samples and the procedures for the preparation of  $\text{SnF}_2$  was developed. Finally, the method for measuring  $^{126}\text{Sn}$  by AMS was established, including transmission of  $^{126}\text{Sn}$  beam current, detection of  $^{126}\text{Sn}$  particles and the sensitivity determination by the measurement of blank sample. The sensitivity for  $^{126}\text{Sn}/\text{Sn}$  was  $(1.92 \pm 1.13) \times 10^{-10}$ , 2-3 orders of magnitude better than those reported in previous studies on AMS measurement of  $^{126}\text{Sn}$  [7, 8].

There are some interference peaks of different masses in the two-dimensional spectra (as can be seen from Figure 1), because some ions with the same values of  $\text{ME}/q^2$  and  $M/q$  as  $^{126}\text{Sn}^{11+}$  can pass through the analysis magnet and the electrostatic deflector and be recorded by the multi-anode detector. Thanks to the high resolution of the detector, these peaks do not interfere with the measurement of  $^{126}\text{Sn}^{11+}$  peak.

There is still room for further improvement. First, chemical procedures of removing Te, such as the ones reported by Catow [10] and Zhang [11], can be added into the preparation of samples. Second, special detection method, such as gas filled magnet combined with time of flight and projectile X-ray detection, can be considered. In addition, the provision of  $^{126}\text{Sn}$  samples with high concentration will make it possible to obtain more accurate standard samples determined by TIMS or ICPMS. These and other improvements are being implemented or considered in our lab for the AMS measurement of  $^{126}\text{Sn}$  at much lower levels.

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