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Measurement of ^{182}Hf with HI-13 AMS system

Jiuzi Qiu [a,b], Shan Jiang [a,*], Ming He [a], Xinyi Yin [a], Kejun Dong [a], Yongjing Guan [c], Yiwen Bao [a], Shaoyong Wu [a], Jian Yuan [a], Bingfan Yang [a]

[a] *Department of Nuclear Physics, China Institute of Atomic Energy, Beijing, 102413, China*
[b] *Chinese People's Armed Police Force Academy, Langfang, Hebei, 065000, China*
[c] *The College of Physics Science and Engineering Technology, Guangxi University, Nanning 530004, China*

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Abstract

^{182}Hf with half-life of about (8.90 ± 0.09) Ma is an extinct radionuclide and can only be produced by a supernova explosion in nature. ^{182}Hf is one of a few radionuclides in the million-year half-life range for tracing a possible supernova event in the vicinity of the Earth within the last 100 million years. This may be accomplished by finding measurable traces of live ^{182}Hf in suitable terrestrial archives. With accelerator mass spectrometry (AMS), an ultra-sensitive nuclear analytical technique, it is possible to detect minute amounts of ^{182}Hf. The detection method of ^{182}Hf with HI-13 AMS system at China Institute of Atomic Energy (CIAE) and the chemical procedures to reduce ^{182}W interference are presented.

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Keywords: ^{182}Hf; Accelerator mass spectrometry AMS; Supernova; Isotope ratio

1. Introduction

^{182}Hf is a long-lived radionuclide of particular interest in the study of supernova explosion events. ^{182}Hf is believed to be produced by *r*-process nucleosynthesis, but it can also be produced by a fast *s*-process in massive stars [1]. During a supernova explosion, a certain amount of ^{182}Hf could be injected into the surrounding interstellar medium (ISM). If such an event took place in the vicinity of the Earth within a few half-lives of ^{182}Hf, a signal should be detectable in appropriate archives. The fact that primordial ^{182}Hf had already decayed, together with supernova as the only known production source in nature, makes ^{182}Hf an ideal candidate as an indicator of a possible supernova explosion in the vicinity of the Earth within the last 100 million years. Recently, an indication for a nearby supernova explosion has been found through the detection of ^{60}Fe ($t_{1/2} =$ 1.6 Ma) in terror-manganese crusts [2]. But more measurements are needed. One advantage of ^{182}Hf compared to ^{60}Fe is the possibility to detect signals from older supernova events because of its longer half-life.

In any production scenario, live ^{182}Hf is expected to be present in the ISM as a result of recent nucleosynthesis. Gamma-ray detection of ^{182}Hf is not feasible due to its overall low activity. However, the deposition of ISM grains by accretion onto Earth could make direct detection of live ^{182}Hf possible in slow-accumulating reservoirs such as deep-sea sediments. With accelerator mass spectrometry it is possible to detect minute amounts of ^{182}Hf. ^{182}Hf detection by AMS was first presented by Christof Vockenhuber [3] at the Vienna Environmental Research Accelerator (VERA), a dedicated AMS facility based on a 3-MV tandem accelerator. Vockenhuber tried to direct ^{182}Hf in deep-sea sediment samples, but failed to obtain satisfactory results due mainly to the insufficient sensitivity and the interference from the isobaric nuclide ^{182}W.

* Corresponding author. Tel.: +86 10 69358335; fax: +86 10 69357787.
E-mail address: jiangs@ciae.ac.cn (S. Jiang).

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In this paper, a method for the detection of ^{182}Hf with a 13-MV tandem accelerator (HI-13) mass spectrometer and the chemical procedures to reduce W content are described.

2. Experimental

The two prerequisites for AMS measurement of ^{182}Hf are high mass resolution to reduce the interference from the stable neighboring isotopes, mainly ^{180}Hf and isobar separation to reduce the interference from the stable isobar ^{182}W. The AMS facility of CIAE could satisfy mass resolution for ^{182}Hf measurement by narrowing the image slits of injection magnet and analyzing magnet. If the width of the slit is reduced to ~2 mm, the mass resolution of our AMS facility can be increased to ~220, whereas the transmission is still above 80%. However the energy of less than 100 MeV available at the AMS facility of CIAE can not separate the stable isobar ^{182}W from ^{182}Hf in the final detector system. According to Vockenhuber [3], a ^{182}W suppression of about 6000 can be achieved by using sample material of HfF_4 and extracting negative ions of HfF_5^- from the ion source. Although the main interference to detection can be significantly reduced by using HfF_4 sample material and extracting HfF_5^- beam from ion source, we found that chemical separation is still necessary.

2.1. Preparation of samples

In this experiment, ^{182}Hf was produced by irradiating 50-mg HfO_2, enriched in ^{180}Hf to 98.3%, with the high neutron flux of the heavy water research reactor at CIAE for eighteen days in December 2002. The reactor neutron flux is about 4.54×10^{13} n cm^{-2} s^{-1} at the sample irradiation site. In the reactor, ^{180}Hf may capture a neutron to produce ^{181}Hf, and the produced ^{181}Hf may capture a second neutron to produce ^{182}Hf. After a cooling time of 920 days, the sample was purified with chemical procedures to reduce W, the ratio of $^{182}Hf/^{180}Hf$ was $(1.628 \pm 0.011) \times 10^{-6}$ determined with a thermal ionization mass spectrometry (TIMS). Standard samples with $^{182}Hf/^{180}Hf$ ratios of $(3.03 \pm 0.03) \times 10^{-8}$ and $(3.00 \pm 0.03) \times 10^{-10}$ were prepared using a series dilution of the irradiated sample with non-irradiated enriched HfO_2 powder. Meanwhile the $^{182}W/^{183}W$ ratio in samples measured with TIMS was 1.78.

Approximately 10 mg of the HfO_2 standard sample material was dissolved in a 5-ml 40% HF and 5-ml 63% HNO_3 mixed solution. The solution was heated on a hot plate, and evaporated to about 2ml, another 5-ml 40% HF and 5-ml 63% HNO_3 was added and evaporated to approximately 1ml, then 2-ml 40% HF and 2-ml 63% HNO_3 was added and evaporated to near dryness. After that, 2-ml 40% HF was added to dissolve the residue and was then evaporated to dryness. Finally the sample was roasted in oven for 2 h at 120 °C to obtain desiccated HfF_4 powder. The blank sample material of HfF_4 powder was prepared using non-irradiated enriched HfO_2 with the same chemical procedures as for the standard sample.

2.2. Column separation procedure

The HfF_4 samples prepared above were respectively redissolved in 10-ml, 1-M HF solution for column separation. A 1-ml sample solution was loaded onto an anion exchange column. The column was rinsed with 10 ml of 1-M HF. Hf was then eluted by 30 ml of 0.01-M HF 9-M HCl, while W and Ta retained on the column. Tracer experiments showed that the average chemical yield of Hf was greater than 95%, and the decontamination factors for W and Ta were larger than 1000. The Hf sample purified with this procedure was transformed to HfF_4 powder again and it was satisfactory for eliminating isobaric interferences for AMS determination of ^{182}Hf.

2.3. Measurement of sputter and ionization yield

Sample material of HfF_4 was mixed with 1:1 w/w silver powder and pressed firmly into Al-target holders of the 40 position MC-SNICS source. The silver powder was served as both an electrical and thermal conductor.

The Hf isotopes of interest were sputtered by Cs^+ as negatively charged HfF_5 and extracted with about 15 kV from the ion source. On the low-energy side, the beam was analyzed by means of a 90° magnetic deflector.

The sputter and ionization yield for HfF_5^- ions was measured to be about 3.4×10^{-3} with a target of known sample mass. The typical HfF_5^- beam current was about 150 nA. The current for the whole lifetime of the target was collected and the amount of extracted $^{180}HfF_5^-$ ions was calculated.

2.4. Simulation transport of ^{182}Hf beam

The measurement of ^{182}Hf was performed with a 13-MV tandem accelerator (HI-13) mass spectrometer at CIAE [4]. On the high-energy side, the beam was analyzed by means of a 90° analyzing magnet with a mass-energy product of 200-MeV amu and a 17° electrostatic deflector.

The value of terminal voltage was dictated by the maximum mass-energy product of the high-energy beam-transport system. For $^{182}Hf^{9+}$ ions, the maximum usable terminal voltage was 8.5 MV, which corresponds to a final energy of 82.1 MeV. In the terminal of the HI-13 tandem accelerator, a carbon foil of 3 μg cm^{-2} thickness was used as a stripper. At the high-energy side, $^{182}Hf^{9+}$ ions were analyzed.

At the beginning of the research, the ^{182}Hf beam transport was simulated with sample material of $^{180}HfF_4$ and extracting ions of $^{180}HfF_5^-$. Due to the significant scattering induced by the carbon foil and Coulomb explosion,the beam current in high-energy side was too small to tune for beam transport. In order to make the adjustment of beam transport easier and maximize $^{180}Hf^{9+}$ current for