

Research on Measurement of ^{182}Hf with HI-13 AMS System^{*}

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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. Therefore ^{182}Hf is an ideal candidate nuclide for the study on possible supernova explosions in the vicinity of the earth within the last 100 million years. In addition, ^{182}Hf is a long-lived radionuclide of particular interest for nuclear engineering. Accurate measurement of ultra-trace ^{182}Hf is very important for reactor design, studies on nuclear cosmo-physics and other fields. With an accelerator mass spectrometry (AMS), it is possible to detect the trace amounts of ^{182}Hf . In this paper, 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. The energy and TOF two-dimension spectra of ^{182}Hf and ^{183}W for blank and a series of standard samples have been obtained, respectively. The ^{182}W contribution to the counts in ^{182}Hf peak was corrected by ^{183}W normalization. The detection sensitivity of this AMS facility for $^{182}\text{Hf}/^{180}\text{Hf}$ ratio measurement is about 4.15×10^{-11} at present.

Key words ^{182}Hf , accelerator mass spectrometry, 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, 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 all primordial produced ^{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 ($t_{1/2}=1.6\text{Ma}$) in ferro-manganese crusts^[2]. But more measurements are needed for confirmation. One advantage of ^{182}Hf compared to ^{60}Fe is the possibility to detect signals from supernova events which happened long time ago because of its longer half-life.

In any production scenario, ^{182}Hf is expected to be present in the ISM as a result of recent nucleosynthesis. The γ -ray detection of ^{182}Hf is unfeasible due to its overall low activity. However, deposition of ISM grains by accretion onto the earth could make direct detection of ^{182}Hf possible in slow-accumulating reservoirs such as deep-sea sediments. With an accelerator mass spectrometry it is possible to detect the trace

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amounts of ^{182}Hf . The AMS detection method of ^{182}Hf was first presented by Christof Vockenhuber^[3] at the Vienna Environmental Research Accelerator (VERA), a dedicated AMS facility based on a 3MV tandem accelerator. The detection of ^{182}Hf in deep-sea sediment samples was performed by Christof Vockenhuber at VERA. Due to the insufficient sensitivity and the interference from the isobaric nuclide ^{182}W , his results were unsatisfactory.

In this work, the detection method of ^{182}Hf with a 13MV tandem accelerator (HI-13) mass spectrometry and the chemical procedures to reduce the isobaric interference from ^{182}W are presented.

2 Sample preparation

The stable neighboring isotopes and isobar ^{182}W are the main interferences for ^{182}Hf detection. So the two prerequisites for AMS measurement of ^{182}Hf are high mass resolution (for separation of stable neighboring isotopes, mainly ^{180}Hf) and isobar separation (because of the stable isobar ^{182}W). A mass resolution of about 220 of our AMS facility could satisfy the measurement of ^{182}Hf . But the energy of less than 100MeV available at the high-energy side of the AMS facility is insufficient to separate the stable isobar ^{182}W from ^{182}Hf in the final detector system. According to Christof Vockenhuber^[3], ^{182}W suppression of about 6000 can be achieved by using the sample material of HfF_4 and the extracting negative ions of HfF_5^- from an ion source. However, further removal of ^{182}W by chemical separation is still required for successful AMS determination of ^{182}Hf .

2.1 Preparation of HfF_4 samples

In this experiment, ^{182}Hf was produced through irradiating 50mg 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 was about $4.54 \times 10^{13} \text{ n} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$ 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 $^{182}\text{Hf}/^{180}\text{Hf}$ was then determined with a thermal ionization mass spectrometry (TIMS) to be $(1.628 \pm 0.011) \times 10^{-6}$. Standard samples with $^{182}\text{Hf}/^{180}\text{Hf}$ ratios of $(3.221 \pm 0.034) \times 10^{-8}$ and $(3.452 \pm 0.042) \times 10^{-10}$ were prepared using a series of dilution of the irradiated sample with non-irradiated enriched HfO_2 powder. Meanwhile the $^{182}\text{W}/^{183}\text{W}$ ratio in samples measured with TIMS was 1.78.

About 10mg of the HfO_2 standard sample material was dissolved in 5mL 40% HF and 5mL 63% HNO_3 mixed solution. The solution was heated on a hot plate, and evaporated to about 2mL, another 5mL 40% HF and 5mL 63% HNO_3 was added and evaporated to about 1mL, then 2mL 40% HF and 2mL 63% HNO_3 solution was added and evaporated to near dryness. After that, 2mL 40% HF was added to dissolve the residue and then evaporated to dryness. At last the sample was roasted in oven for 2 hours at the temperature of 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 re-dissolved in 10mL 1mol/L HF solution for column separation. A 1mL sample solution was loaded onto an anion exchange column. The column was rinsed with 10mL of 1mol/L HF. Hf was then eluted by 30mL of 0.01mol/L HF-9 mol/L HCl, while W and Ta retained on the column. Tracer experiments showed that the average chemical yield of Hf was greater than 95%, the decontamination factor for W was about 80 and the contents of both W and Ta in the resulting sample were at 10×10^{-9} level. In order to further reduce the amount of ^{182}W , large amount of ^{186}W (enriched to 96.7%) was added in the sample and the above separation procedure was then repeated. In this way, the content of ^{182}W could be reduced to 10^{-9} level. The Hf sample purified with this procedure was transformed to HfF_4 powder again and it was satisfactory for keeping the isobaric interferences from AMS determination of ^{182}Hf .

3 Ion extraction

In the experiment, the sample material of HfF₄ was mixed with 1:1 silver powder and pressed firmly into Al-target holders of a 40 position MC-SNICS source. The silver powder was served as both electrical and thermal conductor.

The interest isotopes of Hf were sputtered by Cs⁺ as negatively charged HfF₅⁻ and extracted with about 15kV from the ion source. On the low-energy side, the beam was analyzed by means of a magnetic 90° deflector.

The combined efficiency of sputtering and ionization for HfF₅⁻ ions was measured to be about 3.4×10^{-3} with a target of known sample mass. The typical HfF₅⁻ beam current was about 150nA. The current for the whole lifetime of the target was collected and the amount of extracted ¹⁸⁰HfF₅⁻ ions was calculated.

4 Beam transport

The measurement of ¹⁸²Hf was performed with a 13MV tandem accelerator (HI-13) mass spectrometry 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 200MeV·amu and a 15° electrostatic deflector.

The value of terminal voltage was dictated by the maximum mass-energy product of the high-energy beam-transport system. For ¹⁸²Hf⁹⁺ ions, the maximum usable terminal voltage was 8.5MV, which corresponds to a final energy of 82.1MeV. In the terminal of the HI-13 tandem accelerator, a carbon foil of 3μg·cm⁻² in thickness was used as a stripper. On the high-energy side, ¹⁸²Hf⁹⁺ ions were analyzed.

At the beginning of the research, the ¹⁸²Hf beam transport was simulated with the sample material of ¹⁸⁰HfF₄ and extracting ions of ¹⁸⁰HfF₅⁻. Due to the significant scattering induced by carbon foil and Coulomb explosion, the beam current in high-energy side was too small to adjust for beam transport. In order to make the adjustment of beam transport easier and maximize ¹⁸⁰Hf⁹⁺ current for analysis, the sim-

ulation transportation was divided into three steps. First, because the ¹⁸⁰HfO₂⁻ current was much larger than ¹⁸⁰HfF₅⁻ at low-energy side, the sample material of ¹⁸⁰HfO₂ was used and ¹⁸⁰HfO₂⁻ ions were extracted from the ion source to simulate ¹⁸⁰Hf⁹⁺ beam transport of the sample material of ¹⁸⁰HfF₄ and ¹⁸⁰HfF₅⁻ ions. In the simulation transport, ¹⁸⁰Hf⁹⁺ ions had the same energy with the ¹⁸²Hf⁹⁺ ions, and the parameters of ions optics system were tuned for the optimum state of beam transport. Secondly, to hold the line, the sample material of ¹⁸⁰HfF₄ was used and the ¹⁸⁰HfF₅⁻ ions were extracted to transport the ¹⁸⁰Hf⁹⁺ beam with the same energy as ¹⁸²Hf⁹⁺ beam, and the parameters of ions optics system were finely tuned for the optimum state of ¹⁸⁰Hf beam transport. Thirdly, the sample material of ¹⁸⁰HfF₄ was used and the ¹⁸⁰HfF₅⁻ ions were extracted to simulate the ¹⁸²Hf beam transport of the sample material of ¹⁸²HfF₄ and ¹⁸²HfF₅⁻ ions. In the third step, the ¹⁸⁰Hf⁹⁺ ions had the same momentum as ¹⁸²Hf⁹⁺ ions, and the parameters of electronic and magnetic elements after analyzing magnet were further tuned for optimal state of ¹⁸⁰Hf⁹⁺ beam transport. The research showed that the three-step method of beam simulation transport makes the adjustment much easier.

The transmission efficiency from low-energy side Faraday-cup to detector was measured to be about 5.0×10^{-3} .

5 Ion detection

The ¹⁸²Hf ions were detected by a golden-silicon surface barrier detector (SBD) for energy determination, and a time of flight (TOF) detector for isotope identification. The flight length of the TOF detector was 2m. The energy detector with energy resolution of about 4% was used to separate the nuclides having large mass difference and different charge states with ¹⁸²Hf. The TOF detector was used to distinguish the adjacent nuclides with the same charge state, such as Hf isotopes which have small difference in energy but large difference in time of flight with ¹⁸²Hf. The start detector of TOF was a micro-channel plate (MCP) detector with a carbon foil of 10μg·cm⁻² in thickness, and the stop detector was a SBD (meanwhile used as

energy detector). The time resolution of TOF detector was better than 0.6ns, and the difference of time of flight between ^{180}Hf and ^{182}Hf was about 2.4ns. Therefore, the TOF detector was sufficient to distinguish ^{182}Hf from ^{180}Hf . The detector efficiency was measured to be about 80%. The overall efficiency for ^{182}Hf measurement with the AMS facility was about 1.36×10^{-5} .

6 Results

The energy and time of flight two-dimension spectra of ^{182}Hf (shown in Fig. 1) and ^{183}W have been obtained, respectively. The stable Tungsten isotope of ^{183}W was measured for an accurate subtraction of the ^{182}W contribution to the mass 182 events with the known $^{182}\text{W}/^{183}\text{W}$ isotope ratio of 1.78 measured by TIMS. The energy and TOF two-dimension spec-

tra of ^{182}Hf and ^{183}W for blank sample and a series of standard samples have been measured respectively, and then the ratios of $^{182}\text{Hf}/^{180}\text{Hf}$ were determined relative to the reference value of $(1.628 \pm 0.011) \times 10^{-6}$ for the irradiated sample. The results were shown in Table 1. The detection limit of $^{182}\text{Hf}/^{180}\text{Hf}$ ratio was obtained to be 4.15×10^{-11} for our AMS facility.

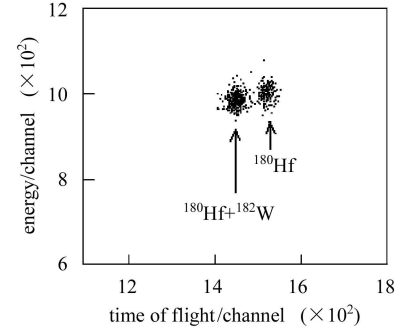


Fig. 1. Two-dimension Spectrum of ^{182}Hf .

Table 1. The ratios of $^{182}\text{Hf}/^{180}\text{Hf}$ measured for blank and standard samples.

sample	$(^{182}\text{Hf}+^{182}\text{W})/^{180}\text{Hf}$	$^{183}\text{W}/^{180}\text{Hf}$	$^{182}\text{W}/^{180}\text{Hf}$	$^{182}\text{Hf}/^{180}\text{Hf}$
blank	$(4.15 \pm 2.95) \times 10^{-11}$			
3.45×10^{-10}	$(4.85 \pm 0.69) \times 10^{-10}$	$(1.48 \pm 0.47) \times 10^{-10}$	$(2.25 \pm 0.71) \times 10^{-10}$	$(2.60 \pm 0.86) \times 10^{-10}$
3.22×10^{-8}	$(3.37 \pm 0.34) \times 10^{-8}$	$(4.70 \pm 1.42) \times 10^{-10}$	$(7.14 \pm 6.87) \times 10^{-10}$	$(3.30 \pm 0.35) \times 10^{-8}$
1.63×10^{-6}	$(1.63 \pm 0.01) \times 10^{-6}$	$(2.37 \pm 0.58) \times 10^{-10}$	$(3.60 \pm 3.57) \times 10^{-10}$	$(1.63 \pm 0.01) \times 10^{-6}$

7 Summary

The detection limit of ^{182}Hf depends on the ^{182}W content in the sample material and the background from neighboring isotopes, mainly ^{180}Hf . Chemical separation is a direct method to remove the W isotopes from the sample material. In addition, if we know the $^{182}\text{W}/^{183}\text{W}$ ratio in the samples, we can correct the ^{182}W interference by measuring the ^{183}W . In this work, the main difficulty of the AMS measurement of ^{182}Hf is the intense beam of isotope ^{180}Hf . Although most of the ^{180}Hf are suppressed by the injection magnet and the analyzing magnet, a small fraction of this intense beam can interfere with the ^{182}Hf measurement. The main reason for this leakage of interfering ions is charge changing processes induced by the residual gas within the elements. Angular scattering on the residual gas, electrodes, slits or vacuum

chamber walls can also allow the interfering ions to be detected. The scattering cross-sections are in the order of 10^{-20} b compared to the cross sections of 10^{-16} — 10^{-15} b for charge changing^[5]. However, in the detector of TOF, the angular scattering on the carbon foils is serious. The leakage can be reduced by simply adding more filter elements and using thinner carbon foils. Each additional filter element can provide leakage suppression for several orders of magnitude. A new ion injector with mass resolution of larger than 380 is being constructed, and the thinner carbon foils from ACF-Metals Arizona Carbon Foil Co. Inc. are expected to be mounted for suppressing the interference from the neighboring isotopes in our AMS at CIAE.

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用 HI-13 串列加速器质谱装置测量 ^{182}Hf 的研究*

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摘要 ^{182}Hf 的半衰期为 $(8.90\pm 0.09)\text{Ma}$, 是一个接近灭绝的放射性核素. 超新星爆炸是自然界中已知的惟一能产生 ^{182}Hf 的途径. 因此 ^{182}Hf 是研究近 1 亿年来在地球附近可能发生的超新星事件的理想核素. 另外, ^{182}Hf 是核工程中特别感兴趣的一个长寿命放射性核素. 精确测量超痕量的 ^{182}Hf 对反应堆的设计和核天体物理学以及其他研究领域都是非常重要的. 用加速器质谱有可能实现对超低含量 ^{182}Hf 的测量. 在中国原子能科学研究院的 HI-13 加速器质谱装置上对 ^{182}Hf 的测量方法以及样品的化学去钨方法进行了研究, 分别得到了空白样品以及系列标准样品的 ^{182}Hf 和 ^{183}W 的能量-飞行时间双维谱. ^{182}W 对 ^{182}Hf 计数的贡献是通过测量 ^{183}W 的计数归一扣除的. 目前本工作对 ^{182}Hf 的测量灵敏度为 4.15×10^{-11} ($^{182}\text{Hf}/^{180}\text{Hf}$ 比值).

关键词 ^{182}Hf 加速器质谱 超新星 同位素比值

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