$\sum HfF_5^-$ current monitoring with off-axis Faraday cup in AMS measurement of ^{182}Hf at CIAE

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Abstract In order to reduce the systematic uncertainties caused by the sequential injection in AMS measurement of ^{182}Hf, a technique for instantaneous monitoring of off-axis $\sum HfF_5^-$ current was developed in the AMS laboratory at China Institute of Atomic Energy (CIAE). The stability and reliability of this method have been experimentally verified. As a result, the accuracy for AMS measurement of ^{182}Hf was significantly improved.

Key words ^{182}Hf, Off-axis Faraday cup, Accelerator mass spectrometry

1 Introduction

^{182}Hf, in half life of 8.90±0.09 million years, is an ideal candidate as an indicator of a possible supernova explosion in the vicinity of the solar system in the past several million years. This may be accomplished by finding measurable traces of ^{182}Hf on the Earth, which was first reported by C. Vockenhuber in 2004[1]. ^{182}Hf is also a long-lived radionuclide of particular interest for nuclear environment engineering as a suitable neutron flux monitor[2,3]. AMS (Accelerator mass spectrometry) is a promising method to detect minute amounts of ^{182}Hf. Successful AMS measurement of ultratrace ^{182}Hf can offer important experiences for measuring other heavy nuclides, and demonstrate the potential ability of AMS in search for super heavy elements. At China Institute of Atomic Energy (CIAE), AMS measurement of ^{182}Hf was first developed on the HI-13 tandem accelerator in 2006[4]. Since then, considerable efforts have been made to improve the measurement sensitivity towards its applications[5,6].

At present, a sensitivity of about 1×10^{-11} for $^{182}Hf/^{180}Hf$ has been achieved[6]. However, for samples with $^{182}Hf/^{180}Hf$ isotopic ratio at 10^{-10} or less, the measurement accuracy is still unable to satisfy the requirements of certain practical applications. One of the main obstacles is the systematic uncertainty caused by the beam current fluctuation when the traditional slow sequential injection method is used. To solve this problem, a technique for instantaneous monitoring of off-axis stable Hf isotope current was developed in the AMS laboratory at CIAE.

2 Technique

For dedicated AMS facilities, the sequential injection (namely fast cycling option[7]) of the ion beams of the high abundance reference isotope and the rare isotope of interest is generally employed. In this case, the strength of magnetic field for injection is fixed. By switching the pre-acceleration voltage, the sequential injection can be realized quickly. If the switching is cycled fast enough, the effect of fluctuations in the ion source output is minimized and the isotope ratio measurement accuracy ensured. For the AMS facility based on the HI-13 tandem accelerator at CIAE, however, the sequential injection had to be realized by alternative variation of the injection magnetic field at fixed pre-acceleration voltage. Consequently, only one isotope (either the abundant isotope or the rare one) could be measured at a time. Achieving good counting statistics for the rare isotope of interest (^{182}Hf) requests a relatively long measurement time, during which the

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fluctuations in the ion source output for the high abundance reference isotope ($^{180}HfF_5^-$) are very likely to occur and, unfortunately, unable to be monitored. The measurement accuracy is thus influenced greatly.

The use of a stable isotope current to monitor the ion source output, and for the normalization of isotope ratio measurement, is usually done at the high energy side of the AMS system. But this may be too costly to implement at the HI-13 tandem facility. Instead, we used a technique for monitoring the beam current of the high abundance reference isotope (^{180}Hf), based on simultaneous measurements of the ^{182}Hf count rate with a detector at the end of the AMS beam line and the stable Hf isotope current in an off-axis Faraday cup at the image plane of the injection magnet at the low energy side of accelerator.

This technique has been applied in several AMS laboratories in the world in recent years, either at the low energy side of accelerator[8,9] or at the high energy side[10]. Technical details and performances (stability and reliability) of the technique, however, have not been reported in cases when an overlapped current of several stable isotopes has to be used as an ion source output monitor. In this work, an in-depth investigation was performed to verify the effectiveness of this particular instantaneous monitoring technique and its contribution to the improvement of accuracy for AMS measurement of ^{182}Hf at CIAE.

The injection system dedicated for AMS at CIAE was set up at 2007[11]. At the image plane of the injection magnet, three Φ4-cm Faraday cups are mounted: a retractable on-axis Faraday cup (F3 Faraday cup), and two movable off-axis Faraday cups. In AMS measurement of ^{182}Hf, the left off-axis Faraday cup was used to monitor the overlapped current of several stable Hf isotopes (Fig.1). The width of slit at the image side of magnet is 1.6 mm for a high mass resolution. So, the F3 Faraday cup can be used to record the current of a particular stable Hf isotope or the mass scan spectrum for HfF_5^- (Fig.2).

In every experiment, the position of left off-axis Faraday cup should be optimized to get the best match with the injection system. In this experiment, the distance between the beam-line axis and the center position of the left off-axis Faraday cup is 2.8 cm.

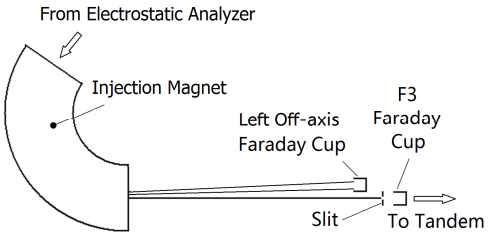

Fig.1 Schematic setup for instantaneous monitoring of beam current for abundant reference nuclide in the measurement of ^{182}Hf. (F3 Faraday cup is temporarily mounted for the measurement of $^{180}HfF_5^-$ current I_{F3} and I_{F3}/I_{oc} only).

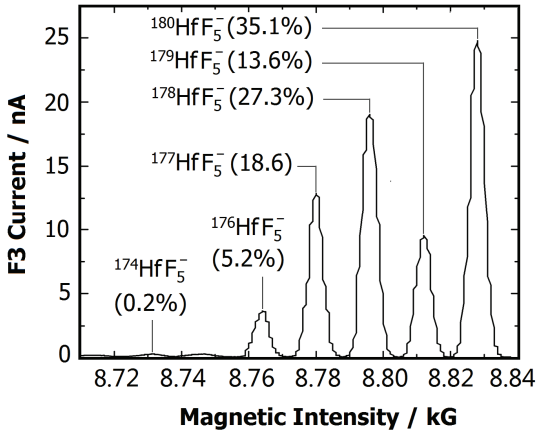

Fig.2 The mass scan spectrum for HfF_5^- obtained in F3 Faraday cup. The relative intensities are very close to their natural isotopic abundances, indicating the lack of contaminating beams in the mass range covering HfF_5^- of all Hf isotopes.

In the AMS measurement of ^{182}Hf, Al-target holders were used for HfF_4 samples (the target material). There is no evident of reactions between Al and F, even saving them together for a long time. But it was found that using Cu-target holders, the background of M=182 was about 100 times higher than that of using Al holders, which is unfavorable for the ^{182}Hf AMS. Therefore, in every experiment, pure HfF_4 powder was pressed firmly into Al-target holders of a 40 position MC-SNICS source directly, in an effort to achieve a larger suppression of background.

From the target material, HfF_5^- ions were extracted. After pre-acceleration, the ions were analyzed by an electrostatic analyzer and an injection magnet. The ions with certain magnetic rigidity were selected and passed through the slit at the image side of the injection magnet (Fig.1), where most of the unwanted isotopes were rejected. The mass spectrum for HfF_5^- scanned on the on-axis F3 Faraday cup is shown in Fig.2.

As shown in Fig.1, when $^{182}HfF_5^-$ ions were selected by the slit, HfF_5^- ions containing Hf isotopes

other than ^{182}Hf (i.e., $^{180}HfF_5^-$, $^{179}HfF_5^-$, $^{178}HfF_5^-$, etc.) were reflected into, and recorded by, the left off-axis Faraday cup, because of their relatively smaller magnetic rigidities. In this way, the counts of ^{182}Hf measured at the end of the AMS beam line and the $\sum HfF_5^-$ current or integrated charges can be obtained simultaneously. It would be better to record current of a particular stable isotope, such as $^{180}HfF_5^-$, for the ion source output monitoring. However, the spatial dispersion on the image plane of the injection magnet is not large enough for this option to be readily implemented for the heavy masses of Hf isotopes. As an alternative, an overlapped current from several stable Hf isotopes measured in the left off-axis Faraday cup was used as a proxy for $^{180}HfF_5^-$. The validity and reliability of this substitution were experimentally verified.

$^{180}HfF_5^-$ beam current (I_{F3}) was measured on F3 Faraday cup by setting the magnetic field corresponding to its magnetic rigidity. Next, the $\sum HfF_5^-$ current (I_{oc}) was measured in the left off-axis Faraday cup immediately by switching the magnetic field corresponding to the magnetic rigidity of $^{182}HfF_5^-$. The switching was cycled fast enough to avoid the uncertainty from the fluctuation of the ion source output. Thus, the ratio of $^{180}HfF_5^-$ current (I_{F3}) to off-axis $\sum HfF_5^-$ current (I_{oc}) was repeatedly determined by multiple alternate measurements. The experimental data of I_{F3}, I_{oc}, and the calculated ratio of I_{F3}/I_{oc} are listed in Table 1.

Table 1 The off-axis $\sum HfF_5^-$ current (I_{oc}), $^{180}HfF_5^-$ current (I_{F3}) and I_{F3}/I_{oc}. Averaged ratio of I_{F3}/I_{oc} % = 35.5±1.4.

I_{oc}/nA	I_{F3} /nA	I_{F3}/I_{oc} / %
13.6	5.1	0.375
55.8	20.0	0.358
78.0	26.0	0.333
10.26	3.6	0.351
82.0	31.0	0.378
60.0	21.0	0.350
185.0	65.0	0.351
70.0	25.0	0.357
19.0	6.4	0.337
55.0	19.7	0.358

The averaged ratio of I_{F3}/I_{oc} is 35.5 with a relative standard deviation of only 3.9% for a variation range of more than an order of magnitude in $^{180}HfF_5^-$ current (from 5.1 to 65 nA), indicating $^{180}HfF_5^-$ current (I_{F3}) can be indirectly monitored by measuring I_{oc} at the left off-axis Faraday cup, while measuring ^{182}Hf at the end of the AMS beam line.

In Fig.3, the off-axis $\sum HfF_5^-$ current scanning spectrum shows that a fairly wide flat top around the magnetic strength corresponding to $^{182}HF_5^-$ (8.860 kG in this experiment). So, despite small variations in magnetic intensity during the ^{182}Hf measurement, monitoring with sufficiently reliable off-axis current of $\sum HfF_5^-$ can be guaranteed. Therefore, it is verified that this is a stable and reliable technique.

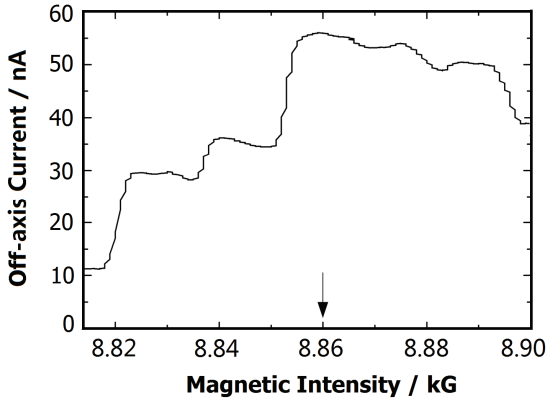

Fig.3 Off-axis current vs magnetic strength. There is a flat part at the vicinity of 8.860 kG (corresponding to the magnetic rigidity of $^{182}HfF_5^-$).

3 Results

In order to verify the contribution of this technique to improving the measurement accuracy of $^{182}Hf/^{180}Hf$, laboratory standard samples with $^{182}Hf/^{180}Hf$ isotopic ratio in the order of 10^{-10} were measured. In the experiment, the magnetic field of injection magnet was set corresponding to the magnetic rigidity of $^{182}HfF_5^-$. The counts of ^{182}Hf were recorded at the end of the AMS beam line, while the integrated charges of off-axis $\sum HfF_5^-$ current with time were obtained by the left off-axis Faraday cup simultaneously. In addition, the transmission of $^{180}HfF_5^-$ ions from low energy side of accelerator to the end of the AMS beam line was measured. Detailed description of the experimental method can be found in Ref.[6]. Thus the ratio of $^{182}Hf/^{180}Hf$ was calculated for a single measurement.

Table 2 shows that, by using this instantaneous monitoring technique, the relative deviation (between measured value and standard one) is 9%, rather than

24.6% with traditional sequential injection method, while the total relative uncertainties are 15.7% and 33.0% for instantaneous monitoring and sequential injection method, respectively. The improvement in measurement accuracy is evident.

Table 2 Comparison of measurement accuracies by instantaneous monitoring technique and sequential injection method.

Monitoring method	Standard $^{182}Hf/^{180}Hf$ ratio	Measured $^{182}Hf/^{80}Hf$ ratio	Relative uncertainty / %	Deviation / %
Instantaneous (this work)	1.33×10^{-10}	$(1.21\pm 0.19)\times10^{-10}$	15.7	9.0
Sequential injection [3]	3.45×10^{-10}	$(2.60\pm0.86)\times10^{-10}$	33.0	24.6

4 Discussion and Conclusion

A technique has been developed for instantaneous monitoring off-axis $\sum HfF_5^-$ current of the abundant Hf isotopes in AMS measurement of ^{182}Hf at CIAE, for improving accuracy of the isotope ratio measurement.

This technique measures the ^{182}Hf count rate at the end of the AMS beam line, while monitors the stable Hf isotope current in an off-axis Faraday cup at the image plane of the injection magnet at the low energy side, where contaminating species mixed in the beams of interest. However, experiment indicates that the mass spectra from HfF_4 samples are generally quite clean in the mass range covering HfF_5^- of all Hf isotopes. And the beam current measured in the left off-axis Faraday cup, $\sum HfF_5^-$, is proportional to $^{180}HfF_5^-$ current over a wide range of beam intensity with an uncertainty of less than 3.9%. This allows monitoring $^{180}HfF_5^-$current at the image side of injection magnet while measuring ^{182}Hf at the end of the AMS beam line. The technique improves accuracy of $^{182}Hf/^{180}Hf$ measurement, which is verified using laboratory standard samples with $^{182}Hf/^{180}Hf$ isotopic ratio in the order of 10^{-10}. Compared with those by using traditional sequential injection method, substantial reduction in measurement uncertainty can be achieved with this technique.

However, the measurement accuracy needs to be further improved to satisfy the requirements of certain practical applications. An additional measure to be taken in our laboratory in the near future is the use of samples (HfF_4 or NH_4HfF_5) prepared by multiphase synthesis[12] to increase the current of HfF_5^-, which is important for obtaining high count rate of ^{182}Hf, and to shorten the alternate measurement cycle between standard and unknown samples, thereby further reducing the systematic uncertainties associated with beam fluctuations.

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