

STEPS TOWARD MEASUREMENT OF ^{135}Cs WITH AMS AT CIAE

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ABSTRACT. ^{135}Cs with a half-life of $T_{1/2} = 2.3 \times 10^6$ yr is an important nuclide in studies of the dispersal of nuclear material in the environment. Preliminary measurements using ^{133}Cs as a proxy for the long-lived ^{135}Cs , with accelerator mass spectrometry (AMS) have been developed at the China Institute of Atomic Energy (CIAE). In order to improve the sensitivity of ^{135}Cs AMS measurement, a new conducting material, Fe powder, was used in the experiment. According to the present results, the background level that can be obtained with blanks was $^{135}\text{Ba}/\text{Cs} \sim 1.83 \times 10^{-10}$ with the CIAE-AMS system. These measurements showed that the Fe was an inferior conducting medium because the interference of ^{135}Ba in Fe powder is 10 times higher than that in Ag powder.

KEYWORDS: ^{135}Cs , AMS, conducting material, background, measurement.

INTRODUCTION

The discharge of nuclear fuel material into the environment from a nuclear power plant requires a rapid and sensitive analysis method for detecting fission products in samples taken from the environment. The nuclide ^{135}Cs with a half-life of 2.3×10^6 yr is interesting because it plays an important role in many research fields, especially in nuclear environmental studies, due to its high fission yield and long half-life. In addition to formation by direct fission, it is produced as a daughter nuclide in the decays of ^{135}Xe and ^{135}I , so the concentration of ^{135}Cs increases during the operation of a reactor. Therefore, ^{135}Cs may be the appropriate tracer for the leakage of nuclear material from a reactor. However, ^{135}Cs is a pure β -decay nucleus and the lack of γ emission makes its detection by radiometric methods very difficult. Traditional mass spectrometry methods such as thermal ionization mass spectrometry (TIMS) (Lee et al. 1993) and inductively coupled plasma mass spectrometry (ICP-MS) (Zheng et al. 2014; Liezers et al. 2009) have been used for ^{135}Cs measurement. However, the interferences of stable molecular and atomic isobar ions strongly limit the detection sensitivity. Accelerator mass spectrometry (AMS) may be a promising method to detect ^{135}Cs (Eliades et al. 2013; Lachner et al. 2015).

In this paper, we report the experimental results for different samples of Cs, which were measured with the HI-13 tandem AMS facility at the China Institute of Atomic Energy (CIAE). In order to improve the sensitivity, a new conducting medium, Fe powder, was used for the first time in this experiment.

EXPERIMENT AND RESULTS

Ion Extraction

To improve detection sensitivity, interference should be removed or decreased. In AMS measurement, isobaric interferences can be removed or decreased by using suitable negative ions. ^{135}Ba is the main interference in the measurement of ^{135}Cs with AMS, however, it is very difficult to differentiate between ^{135}Cs and ^{135}Ba by using the ΔE -E method. For the purpose of finding the most favorable ions for ^{135}Cs measurements, which should maximize Cs ion currents and minimize currents of Ba, the beam current of stable isotope ^{133}Cs , such as Cs^- , CsF^- , CsF_2^- , and CsF_3^- were tested for ^{135}Cs AMS measurement. It should be noted that CsF_2^- was used for ^{135}Cs AMS measurement in the previous study by using PbF_2 as conducting material (Eliades et al. 2013;

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Table 1 Ion beam current of Cs⁻ from different samples.

Sample	Mass proportion	Extracted ion	Beam current value before experiment (nA)	Beam current value after experiment (nA)	Measuring time (min)
CsNO ₃ + Ag	1:2	Cs ⁻	4.0	3.0	100
CsNO ₃ + Fe	1:2	Cs ⁻	0.5	0.5	10
Ag		Cs ⁻	0.2	0.3	10
Fe		Cs ⁻	0.3	0.2	20

MacDonald et al. 2015a, 2015b; Zhao et al. 2016). Since the usual Cs sputtering would obscure the ¹³⁵Cs/¹³³Cs ratio of a sample, Rb sputtering was successfully applied and tested also for various other typical AMS elements. Cs⁻ currents of several 10 nA were extracted over hours from milligram amounts of Cs₂SO₄ material by Lachner et al. (2015). However, Cs⁻ has been selected as the suitable choice for the present ¹³⁵Cs AMS measurements based on our previous experimental results. The details of experiment can be found in Yin et al. (2015). The beam current was measured before and after the measurement. The ion beam current was relatively stable and long-lasting during the measurement time, as shown in Table 1.

Background Measurements

To measure the ¹³⁵Ba background level, CsNO₃ with different conducting media were measured with HI-13 tandem AMS system by extracting Cs⁻ ions. The ¹³⁵Ba background from the different conducting media, Fe powder (99.9%), and Ag powder (99.9%) were also measured by AMS. The system comprises an injection system with a 90° spherical electrostatic deflector in front of a double-focusing 112° injection magnet with mass resolution, M/ΔM = 400 to 800 (He et al. 2010). A high terminal voltage, up to 13 MV, is produced in the middle of the acceleration tube. The high-energy beam analysis system consists of a 90° double-focusing magnet followed by a switching magnet which transports the beam to the AMS beam line with a 15° electrostatic deflector and the surface barrier detector (SBD).

Different sample mixtures (as shown in Tables 1 and 2) were pressed into the standard NEC aluminum target holder. Cs⁻ ion was used for analysis extracted from the commercial CsNO₃ (99.99%) + Ag, CsNO₃ (99.99%) + Fe, Ag, and Fe samples. The ¹³³Cs⁻ ions were selected by the injection magnet, a terminal voltage of 7.6 MV was used for acceleration and the 8⁺ charge state was selected for Cs ions. Transmission parameters of all equipment were set for ¹³³Cs. Then the beam current of ¹³³Cs⁸⁺ was measured by the AMS Faraday cup. It should be noted that the previous measurement for CsNO₃+Ag sample (Yin et al. 2015) had been made using the 10⁺ charge state at 7.6 MV. Then the ¹³⁵Cs⁻ ions were selected by changing the injection magnet. Keeping other transmission parameters unchanged, only adjust the terminal voltage to 7.7 MV and change the electrostatic deflector voltage to V₁₃₃ × 135/133. The counts of ¹³⁵Ba were determined from the SBD detector, while the number of ¹³³Cs ions was calculated from the beam current (nA) of ¹³³Cs^{q+} in the AMS Faraday cup. Measuring time is 300 sec for each sample. The ¹³⁵Ba/¹³³Cs ratio can be calculated by the formula below:

$$R = \frac{N}{\frac{t}{q} \times n} \quad (1)$$

where N is the ¹³⁵Ba total counts, t is the measuring time, I is the beam current intensity (nA) in the AMS Faraday cup, q is the charge state of the ions, and $n = 6.25 \times 10^9$, the particle counts

per second for 1 nA beam current. The background of ^{135}Ba from different samples is shown in Table 2.

DISCUSSION

According to Middleton et al. (1989), the electron affinity of Ba is negative. This means that there should be no background of ^{135}Ba in the case of extracting Cs^- from the ion source. However, our results show that the ^{135}Ba background still can be detected in the case of extracting Cs^- although it is low. The representative energy spectrum of CsNO_3+Ag and CsNO_3+Fe samples detected with the SBD detector is shown in Figure 1. The two peaks of ^{118}Sn and ^{135}Ba are clearly visible. This implies that the Ba can be extracted as the form of Ba^- and the electron affinity of Ba should be larger than 0. The new data of electron affinity of Ba is about 0.14 eV (Petrinin et al. 1995), and our results support this data. Possible interfering ions were analyzed for ^{135}Cs AMS measurement using our own computer programs, and the results are shown in Table 3. The analysis results show that the rigidity of $^{118}\text{Sn}^{7+}$ is very similar to that of $^{135}\text{Cs}^{8+}$. So ^{118}Sn can be detected, but separated by the different energy (Figure 1).

Table 2 Background level of different samples.

Sample	Mass proportion	Extracted ion	Terminal voltage VT (MV)	Charge state	Count rate (M = 135)	$^{135}\text{Ba}/\text{Cs}$
$\text{CsNO}_3 + \text{Ag}$	1:2	Cs^-	7.6	8^+	0.130 ± 0.013	$(1.8 \pm 0.2) \times 10^{-10}$
$\text{CsNO}_3 + \text{Fe}$	1:2	Cs^-	7.6	8^+	0.170 ± 0.010	$(1.4 \pm 0.1) \times 10^{-9}$
Ag		Cs^-	7.6	8^+	0.007 ± 0.005	$(1.5 \pm 1.0) \times 10^{-10}$
Fe		Cs^-	7.6	8^+	0.060 ± 0.012	$(1.3 \pm 0.3) \times 10^{-9}$

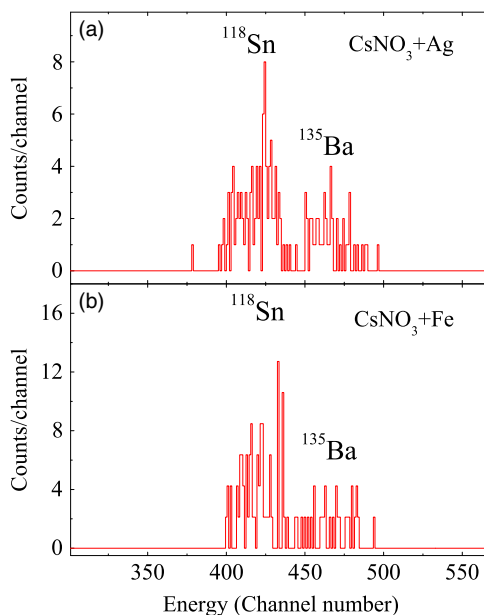


Figure 1 Representative energy spectra of $\text{CsNO}_3 + \text{Ag}$ and $\text{CsNO}_3 + \text{Fe}$ samples.

Table 3 Possible interfering ions for ^{135}Cs AMS measurement.

M	q	E (MeV)	ME/q ²	E/q
67	4	34.28685	143.5762	8.571713
84	5	42.84388	143.9555	8.568777
101	6	51.40092	144.2082	8.566821
118	7	59.95796	144.3886	8.565423
135	8	68.51500	144.5238	8.564375

The importance of the conducting medium in AMS measurement cannot be neglected. In our previous experiments (Yin et al. 2015), Ag powder was used as the conducting medium. In order to affirm whether the Ag powder is the appropriate conducting medium or not, a different conducting medium, Fe powder, was used in the present experiment. As shown in Table 1, for the sample of CsNO_3 with Ag, the beam current of Cs^- is little higher than that from the CsNO_3 with Fe. For the sample of Ag, the beam current of Cs^- is almost equal to that of Fe. It can be seen from Table 2 that the background of ^{135}Ba from CsNO_3 mixed with Ag is ~ 10 times lower than that from CsNO_3 mixed with Fe. Furthermore, the background of ^{135}Ba from sample Ag is also ~ 10 times lower than that from sample Fe. Therefore, the results indicate that the conducting medium Ag powder is better than Fe powder in our AMS measurement, as shown in Tables 1 and 2.

In addition, the sensitivity of 1.8×10^{-10} ($^{135}\text{Ba}/\text{Cs}$) can be obtained from the present experiment, which is slightly worse than that of previous result (Yin et al. 2015). The likely reason for this is that, in case of the Cs^- ion with charge state 10^+ , an interference peak from the scattering of some ions with similar energy cannot be completely separated from the peak of ^{135}Ba .

SUMMARY

The AMS measurement method of ^{135}Cs has been studied at CIAE. The new conducting medium Fe powder was used for the first time in this experiment. For the sample of CsNO_3 with Ag, the beam current of Cs^- is slightly higher than that of CsNO_3 with Fe. The background of ^{135}Ba from the sample mixed with Ag is ~ 10 times lower than that from the sample mixed with Fe. The results indicate that the conducting medium Ag powder is better than Fe powder in AMS measurement for ^{135}Cs . The ^{135}Ba background level is about 1.8×10^{-10} ($^{135}\text{Ba}/\text{Cs}$). However, more investigations in the future, such as the sample of Cs_2SO_4 , may be useful to improve the sensitivity of AMS measurement. Additional measurements are needed in order to deal with the problem of cross contamination. We note that some recent works (MacDonald et al. 2015a, 2015b; Zhao et al. 2016) have carried out detailed experiments on this issue.

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