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^{36}Cl-AMS measurements with 3-MV tandem accelerator

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Abstract ^{36}Cl is one of the most interesting nuclides in accelerator mass spectrometry (AMS) measurements. The application of ^{36}Cl has been widely applied in various fields. All most all of ^{36}Cl AMS measurements at natural isotopic concentrations have yet been performed at tandem accelerator with 5 MV or higher terminal voltage. The measure improvement of ^{36}Cl and other medium heavy isotopes performed at 3 MV in AMS facilities is one of the hottest topics in AMS measurements. In order to increase the suppression factor of ^{36}S, the energy loss straggling and angular straggling of ^{36}Cl and ^{36}S ions in various counter gases (P10, isobutane and propane) were investigated. Some groundwater samples were measured with energy of 32 MeV, and the results were in good agreement with the result obtained with ion energy of 72 MeV. The results indicate that the approximate detection limit of ^{36}Cl in 3 MV AMS facility is ^{36}Cl/Cl=1×10^{-14}, and the uncertainty is 30% when the sample with isotopic ration ^{36}Cl/Cl≈10^{-13}.

Key words Accelerator mass spectrometry, Isobaric identification, ^{36}Cl, Sensitivity

1 Introduction

There is only one long-lived radionuclide of chlorine, ^{36}Cl ($T_{1/2}$= 301 ka). Natural sources of ^{36}Cl including production exist in the atmosphere and in the lithosphere. The applications of ^{36}Cl cover a wide range in the earth sciences and environmental sciences. The most common applications of natural ^{36}Cl include determinating the exposure ages and erosion rates of rocks, dating old groundwater and estimating groundwater residence times, and reconstructing the geomagnetic dipole record in ice cores[1-5]. The application of anthropogenic ^{36}Cl is involved in evaluating locations for nuclear waste storage facilities, and evaluating radiological conditions near nuclear facilities. The sensitivity of AMS is mainly limited by the interferences from isobars, isotopes and other backgrounds. The source of interferences has been discussed in details and a program has been designed for experimental spectra in AMS measurement in our previously work[6].

In the case of the ^{36}Cl AMS measurement, suppression of the stable isobar ^{36}S is performed based on the chemical removal during sample preparation and the different energy loss in matter. Almost all of the ^{36}Cl AMS measurements at natural isotopic concentrations have yet been performed at tandem accelerator with 5 MV or more terminal voltage. The measure improvement of the ^{36}Cl and other medium heavy isotopes carried out in 3 MV AMS facilities is one of the most important trends in AMS measurement. Several methods have been developed to identify ^{36}Cl and ^{36}S at tandem accelerator with 3 MV or less terminal voltage. A gas-filled time-of-flight method has been built and developed to improve the ability of isobaric identification in AMS measurements at the China Institute of Atomic Energy (CIAE)[7-9], and our recent experiment results suggested that ^{36}Cl at natural concentrations which without high precisely required, maybe performed at tandem accelerators with 3-MV terminal voltage. A ΔTOF method was built for the measurement of the environmental ^{36}Cl samples at the Vienna Environmental Research Accelerator (VERA), but the detection limit of isotopic ratio ^{36}Cl/Cl is

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higher than 10^{-14}[10]. An enormous step forward in the detector development was the use of very thin and homogeneous silicon nitride foils as entrance windows, which reduce the energy loss and energy straggling when the ion passing through the windows[11,12]. An excellent performance of the ionization chamber designed at ETH Zurich is achieved by a silicon nitride entrance and exit windows, a small detector volume and a silicon strip detector. Based on the ionization chamber, more measurements on real exposure dating samples in the range of ^{36}Cl/Cl=3×10^{-14} to 10^{-11} have been performed at VERA. A new technology called Isobar Separator for Anions has been developed for chemical filtration of isobars at low energy, before tandem accelerator, at the University of Toronto[13]. This paper described our investigation of ^{36}S suppression factor with detection system and the measurement of water samples.

2 Experimental

Preparation of AgCl from groundwater samples was carried out at CIAE. In order to reduce or eliminate the isobaric interference by ^{36}S in the ^{36}Cl AMS measurements, a sulfur reduction process was included in the sample preparation scheme. Following the preparation scheme established by Tsukuba University but slightly modified procedures[14]. About 500-mL ground water was heated on a hot plate (70°C) to concentrate it to 200 mL. All of the samples were filter through 0.2 μm membrane filter. A certain amount of ^{35}Cl-enriched carrier (2 mg NaCl) was added to each sample to determine the natural Cl by isotope dilution. Fig.1 shows the sample preparation procedure. The main process of sulfur eliminating in the circle is marked with dotted lines as shown in Fig.1. Finally, the AgCl was washed three times with deionized water and twice with 99.5% ethanol with ultrasonic wave, rather than $BaSO_4$ precipitation method to eliminate the sulfur contamination in the product. After that, the AgCl was dried in the oven at 130°C for 3 h.

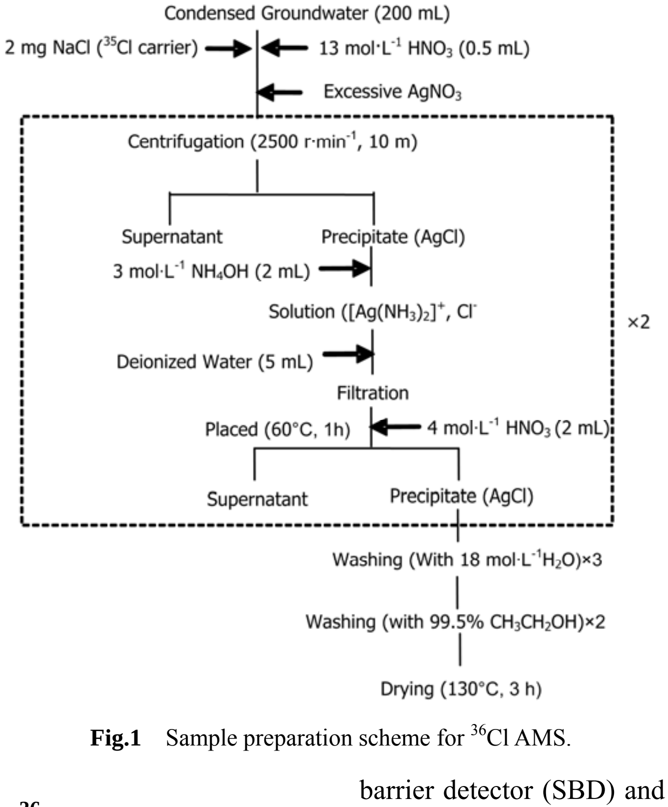

Fig.1 Sample preparation scheme for ^{36}Cl AMS.

3 Detection setup and ^{36}S suppression

The structure of our current GF-TOF detector system consists of a micro-channel plate (MCP), a suface barrier detector (SBD) and a chamber. MCP and SBD detector are used to provide start and stop timing signals, respectively. The chamber is served as an $\Delta E-E$ detector. The compact MCP detector with a carbon foil of 7 μg·cm^{-2} in thickness is based on two electrostatic mirrors with fast MCP detectors, which provide a time resolution of a few hundred picoseconds for ions. Fig.2 shows a schematic diagram of our current chamber. It consists of a split-anode ionization chamber, with a 10×10 mm^2 silicon nitride entrance window (100 nm thickness from silson ltd., Northampton, UK), and a SBD detector with a 26 mm diameter which is located at the back of the chamber. The anode consists of two separated anode named ΔE_1 and ΔE_2 with lengths of 300 and 100 mm, respectively. The best separation was achieved by adjusting the gas pressure such, that the optimum residual energy is about 1/5 of the incidence energy (namely that the ions have lost about 4/5 of their initial energy when they pass through gas). This result is good agreement with the result of C.Vockenhuber[10].

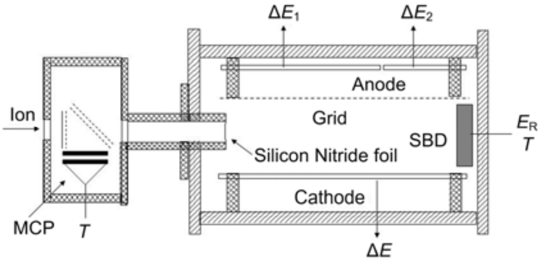

Fig.2 Schematic of our current chamber for ^{36}Cl providing two independent energy loss signals, one residual energy signals.

To further increase the suppression factor of ^{36}S, the energy loss straggling and angular straggling of ^{36}Cl and ^{36}S ions in various counter gases, for example, P10 (argon plus 10% methone) and isobutane, as well as propane were investigated. The energy and angular straggling of the ^{36}S ions passing through 100 nm thick silicon nitride entrance window and 30 cm thick different gas layer with a certain pressure were estimated with the SRIM-2008 code[15]. Fig.3 shows the energy straggling of residual energy of ^{36}S ions passing through the silicon nitride entrance window and gas layer. The differences of residual energy in three cases are almost the same, the mean value are all 5.75 ± 0.01 MeV, but the energy straggling (FWHM) in the entrance window and gas layer would be 0.31 MeV for P10, and 0.26 MeV for isobutane and propane.

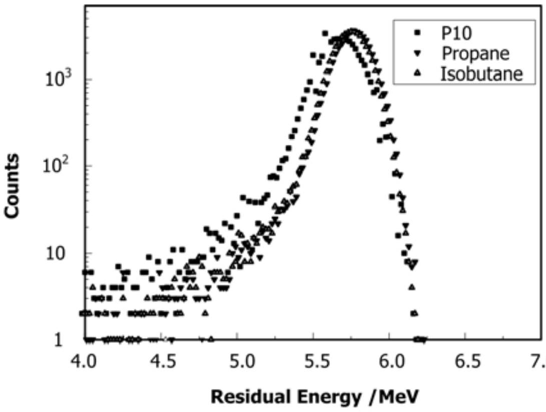

Fig.3 Calculated spectra of residual energy of ^{36}S ions with a SRIM simulation.

According to the active area of SBD detector, a large number of ions were simulated and shown that the detector efficiency of ^{36}S ions is about 80% for using P10 gas, and close to 95% for using isobutane and propane gas. However, if the effect of carbon foil of MCP detector was considered, the dettector efficiency of ions is reduced to less than 70% for using P10 gas. Based on the calculated results and previously experimental results, the propane gas was selected to identify ^{36}Cl and ^{36}S. Several groundwater samples collected from Beijing (B6, B12 and B24) and Tianjin (T4, T8 and T10) were measured again at energy of 32 MeV, which had been measured previously with energy of 72 MeV. Since the sample was not prepared specialized in this experiment, most of the material in the cathodes were exhausted during the measurement with a energy of 72 MeV，the currents of ^{35}Cl ions in lower energy site (before accelerating) ranged from 200 to 300 nA. This means the current is about one order of magnitude less than the general values. According to the current, two groundwater samples (B6 and B24) were analyzed. B6 and B24 samples were taken at a depth of 3159 and 2950 m in Beijing area. The measuring time for one target was 30 min. The results of ^{36}Cl measurements with different incidence energy (32 and 72 MeV) on two groundwater samples are given in Table 1.

At an energy of 32 MeV, two different runs on the same blank sample yield suppression factors of 1×10^4. Samples prepared by above chemical protocols from analytically pure NaCl yield a ^{36}S/^{35}Cl about 5×10^{-10}.

Table 1 Results of groundwater samples

Sample	Depth / m	32 MeV	72 MeV
		^{36}Cl/Cl	^{36}Cl/Cl
B6	3159	$(1.35\pm0.41)\times10^{-12}$	$(1.41\pm0.14)\times10^{-12}$
B24	2950	$(3.17\pm1.10)\times10^{-13}$	$(4.35\pm0.44)\times10^{-13}$

4 Conclusion

The energy and angular straggling of ^{36}S ions in various detector gases were analyzed, and several groundwater samples were measured at the energy of 32 MeV, which closed to energy limited by the 3 MV tandem AMS. For the ion energy of 32 MeV, a suppression factor of 1×10^4 was obtained using the current detector. For this measurement, the uncertainty of our results is up to 30%, the main contribution is probably due to the low and unstable beam current. However, the abundance sensitivity for ^{36}Cl is 1×10^{-14} with the energy of 32 MeV. Fortunately, the isotopic ratios of the groundwater samples were high enough to the detection limited. According to our results and other reported previous method, neither detection method nor chemical removal in sample preparation appear capable of measuring ^{36}Cl/Cl to 10^{-15} or lower in 3 MV AMS facility. However, if the Isobar Separator for Anions technique (sea Ref.[13]) can be well performed in small AMS systems, it should be a positive answer.

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