

AMS measurement of In-situ produced cosmogenic ^{10}Be in Loess Quartz in Luochuan

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Abstract Loess is the most important and the most typical continental classic sediments of Quaternary. So the measurement of In-situ produced cosmogenic ^{10}Be is expected to be used for the Loess stratigraphic age. But the production rate of In-situ produced cosmogenic ^{10}Be in Loess quartz is very low. It requires very high sensitivity and accuracy of measurement. This work has measured the concentrations of ^{10}Be of two layers in Loess section of Luochuan including L8, L9 and the concentrations of ^{10}Be in (of) blank sample with the HI-13 AMS system at China Institute of Atomic Energy(CIAE), then calculated the content of ^{10}Be in loess quartz for each layer. The ratio of $^{10}\text{Be}/^9\text{Be}$ was found in the range of 10^{-13} . The background of $^{10}\text{Be}/^9\text{Be}$ was about 3×10^{-14} .

Key words Loess in Luochuan, In-situ produced cosmogenic ^{10}Be , AMS measurement

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1 Introduction

Studying of the In-situ produced cosmogenic nuclides, which are the long-lived radio nuclides produced by secondary cosmic ray particles has being developed rapidly in the middle of 1980s, especially ^{10}Be and ^{26}Al (Half-life are 1.5Ma ^[1] and 0.7Ma ^[2], respective.)^[3, 4]. Research and measurement of ^{10}Be concentration in natural medias, such as ice code, deep sea sediment, manganese nodules, soil^[5], adamic earth^[6], lassoos, lake sediment, has been caused more and more interest and regard^[7].

The In-situ produced cosmogenic nuclides is the nuclides made by universe radial particles come from the outer astospace (include proterozoic and secondary particles) hit the earth's surface matter (aerosphere or the earth's surface) occur the nuclear reaction. In-situ produced cosmogenic nuclides are some

long life radial nuclides (such as ^{10}Be , ^{26}Al)^[1] made by sub universe radial (mostly neutron and negative meson) which run through the aerosphere make reaction with some atom in loess quartz. Began in the mid-1980s, the "In-situ produced cosmogenic" nuclides research developed very rapidly, the ^{10}Be and ^{26}Al are the most in-depth study on the extent, and the range of application is wide. The measurement and research of ice code, deep sea sediment, manganese nodules, soil^[5], adamic earth^[6], lassoos, lake sediment, has been caused more and more interest and regard^[7]. But the work is not made more progress due to the limitation of sensitivity. The concentration of ^{10}Be in most real samples are about 10^{-12} ^[8-10], a fat lot can reach to 10^{-13} ^[11].

Loess is the most important and the most typical continental clastic sediments of Quaternary. It has the same source, almost the same initial dose and in

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which the quartz was well mixed in the desert. Furthermore, ^{10}Be and ^{26}Al are both in-situ produced cosmogenic nuclides which has almost the same geochemical behavior. The ratio of in-situ produced cosmogenic radionuclide ^{10}Be to ^{26}Al in loess quartz has great potential to evaluate the stratigraphic age of loess^[12]. As so far, it is the first time in the world to determine the buried age of the Chinese Loess using this method. But the productivity of in-situ produced cosmogenic ^{10}Be inside of the quartz crystal lattice is very low, just 10 at/a^[5]. It brings forward the demand of the high sensitivity and precision of the measurement. The CIAE-AMS group developed the method of the AMS measurement of ^{10}Be with the HI-13 tandem accelerator and improved the sensitivity. we also measured the concentration of in-situ produced cosmogenic ^{10}Be in two quartz samples which come from two grounds (L8 and L9) of loess in LuoChuan.

2 Experimental

The samples were made in the chemical laboratory in the Radiometry Center in CIAE, while the measurement of ^{10}Be was made with the CIAE HI-13 tandem accelerator mass Spectrometry facility.

2.1 Chemical agent and Material

The standard sample, SRM 4325, is purchased from National Standard and Technology Institute (NIST). It is in 1 mol/L HCl stroma, and the value of $n(^{10}\text{Be})/n(^9\text{Be})$ is 2.68×10^{-11} with the uncertainty of 5.1%. The total concentration of Be is 5.15 mg/g. The carrier solution was prepared by $\text{BeSO}_4 \cdot 4\text{H}_2\text{O}$ to about 1 mg $^{10}\text{Be}/\text{mL}$ in HCl stroma. $\text{BeSO}_4 \cdot 4\text{H}_2\text{O}$ was purchased from American Alfa Aesar company. Its purity quotient is 99.99% (metal core)(lot identification mark is 17K20). Other reagents such as HCl, HF and NH_4OH etc, are all in G. R grade. The plastic centrifugal tubes, the quartz crucibles and the teflon beakers were all dipped into the 6 mol/L HNO_3 over 12 hours to oxidize the impurities. Then they were cleaned repeatedly using deionized water and secondary deionized water.

2.2 Preparation of Samples

Loess samples were taken from the loess section ($35^\circ 45'\text{N}$, $109^\circ 25'\text{E}$) in Luochuan in Shanxi province.

Quartz grains were extracted from the loess samples in the State Key Laboratory of Earthquake Dynamics in Institute of Geology in China Earthquake Administration (CEA). After being crushed (dried at 40°C before if moist) and sifted to about 60~140 item., the carbonate, organic matter and feldspar were removed by HCl, H_2O_2 and HF successively. Finally, they are dried at 40°C in the oven after bathing to acetone. Pure quartz grasins were obtained after infrared ray inspection of feldspar.

The steps of chemistry flow to extract in-situ ^{10}Be from the quartz and the preparation of ^{10}Be targets for AMS measurement are described as following: atmosphere produced ^{10}Be was firstly removed with HF to insure the exact determination of in-situ produced cosmogenic ^{10}Be whose concentration was 10^2 or 10^3 times lower than the atmosphere produced ^{10}Be . ^9Be carrier (1 mg/mL) was added into the samples and then dissolved the quartz completely by HF. BeCl_2 and AlCl_3 were extracted with the cation-exchange resin, then precipitated with ammonia to get the $\text{Be}(\text{OH})_2$ and $\text{Al}(\text{OH})_3$ by adjusting the PH value of the solution to about 8.8 and 5.1 separately. After laying out and purification, $\text{Be}(\text{OH})_2$ and $\text{Al}(\text{OH})_3$ were centrifuged by 3000 runs/min 10 min. Discarded the solution, transferred the precipitates to the quartz crucible by anhydrous ethanol, dry the hydroxides in room temperature or below 30°C in the oven, then burn the $\text{Be}(\text{OH})_2$ in the furnace at 1000°C for about 30 min to obtain BeO, while $\text{Al}(\text{OH})_3$ at 930°C for the same time to obtain Al_2O_3 . After cooling, the oxidides were put into 2ml centrifugal tubes with airproof. Before measurement, mixed the prepared BeO and pure Nb powder in definitely proportion and pressed the samples into the aluminum target pricks(with the diameter of 1 mm and the depth of 4 mm). Al_2O_3 was mixed with Ag powder with 1:1 in proportion and then pressed into high pure copper target pricks(size is the same as former). All of the target pricks were cased into the target plate that holds 40 target pricks. The target plate was then roasted at 30°C for several hours and then cased into MC-SNICS target negative source for AMS measurement.

2.3 The AMS Measurement of ^{10}Be

2.3.1 The beam current of BeO^-

The beam current is one of the key points for AMS measurement. The measurement sensitivity and pre-

cision can be improved when the beam current increased. Now, large increase of the BeO^- injection current is obtained. This is due to change of the compound pressed together with BeO , Nb instead of Ag. As shown in Fig. 1(a), the beam current is 3 time higher improved when the Nb is used as mixture instead of Ag. The Fig. 1(a) also show the suitable ratio of BeO to Nb is 1:2 by weight. Moreover, as it's shown in Fig. 1(b), the beam current is increase steadily when the Nb is used, whereas the beam current is unstable when Ag is used as mixture. Now, the Nb is used to mix with BeO with ratio of 2:1 by weight and normally the beam current is about 1.5 μA .

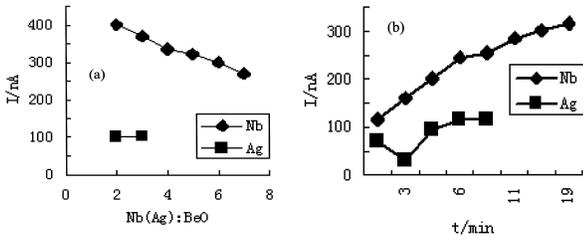


Fig. 1. The infection of electric medium to the beam. (a) different electric medium; (b) different electric medium to the stability of the beam.

2.3.2 Simultaneous measurement

The ratio of the radioisotopes to the stable isotope is the final result for AMS measurement. So, the beam current of stable isotope and the counts of rare radioisotopes are the important values in AMS measurement. In order to get higher precision, the sequential injection method or the simultaneous measurement method has been developed in almost all the AMS system. In our AMS system, the simultaneous measurement method has been developed.

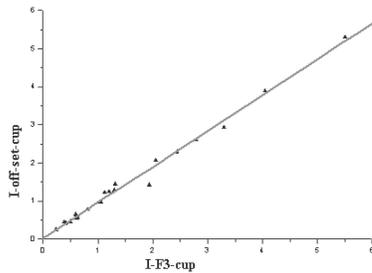


Fig. 2. The curve of beam comparison. I-off-set cup: The average current measured by off-set Faraday cup; I-F3 cup: The measured current of No. 3 central Faraday cup.

When the sample is bombarded by Cs^+ , the $^{10}\text{BeO}^-$ and $^9\text{BeO}^-$ will produced and extracted form

the ion source simultaneously, both of them are pre-accelerated to 110 keV, and pass through the 90° electrostatic deflector. After the injection magnet, the $^{10}\text{BeO}^-$ and $^9\text{BeO}^-$ will be separated. At the image point of the injection magnet, the distance between the $^{10}\text{BeO}^-$ and $^9\text{BeO}^-$ is 60 mm. So, the beam current of $^9\text{BeO}^-$ is simultaneously measured by a off-set faraday cup during detector counting the ^{10}Be . This method needs the high stability of the AMS system. A standard sample was used to check the stability of the AMS system. By using a standard sample with ration of $^{10}\text{Be}/^9\text{Be}=2.68\times 10^{-11}$, the measurement result shown that the reproducibility is around 3%

The measured results in Fig. 2 show the excellent linear relationship between measured values. The gradient of the line is about 1. It can be concluded that it can get high precision by well monitoring the change of beam current with the simultaneous measurement equipment.

2.3.3 The AMS measurement of ^{10}Be

BeO^- beams are exacted from the ion source and accelerated up to 110 keV towards the 90° electrostatic deflector. After energy selection, the BeO^- is transported to the 112° injection magnet, Then the $^{10}\text{Be}^{16}\text{O}^-$ is transport to the accelerator, at the same time, the $^9\text{Be}^{16}\text{O}^-$ beam current is measured with the off-set faraday cup. For the higher ion energy and higher transmission, ^{10}Be measurement have been carried out at a terminal energy of 8.4 MV, the BeO^- molecules are dissociated at the carbon stripper foil. The 90° analyzing magnet, switch magnet and 15° electrostatic deflector, which is set to select the charge state $3+$ (55% yield, ions with a final energy of 28.4 MeV), suppress any interfering ions, with the exception of ^{10}B . This isobar has a magnetic rigidity and energy irresolvable from those of ^{10}Be .

The isobar ^{10}B is a limiting factor in ^{10}Be measurement. Several dE/dx techniques are used to separate ^{10}Be from $^{10}\text{B}^{[13]}$. Since ^{10}B has a higher stopping power than ^{10}Be it can be completely stopped in an absorber placed in front of the ^{10}Be detector. This passive absorber technique has a higher efficiency at higher energies. So, In order to remove and identify the ^{10}B interference, an absorber and a multi-anode gas ionization chamber is used in our lab. According to calculation, the ranges of ^{10}Be and ^{10}B with energy of 28.4 MeV in Ni are 21.5 μm and 15 μm , respectively. So, 17 μm Ni absorber is used as the

entrance window of ionization chamber. Due to the energy, range straggling and especially the foil thickness inhomogeneous, parts of ^{10}B can enter the detector. Then the multi-anode chamber with 150 mbar P10 gas is used to identify ^{10}Be and ^{10}B . In our experimental, the total energy E_t , and energy loss signals (ΔE_1 and ΔE_2) are used to identify ^{10}Be and ^{10}B . A two-dimensional spectra ($E_t - \Delta E_1$) of standard sample is shown in fig.3. The fig shows that it is clear to identify ^{10}Be and ^{10}B . The spectra also show that there is another background ^7Be , this background is due to the $^1\text{H} (^{10}\text{B}, \alpha)^7\text{Be}$ reaction with hydrogen in the absorber entrance foil and the gas. Using the two-dimensional spectra it is also clear to identify ^{10}Be and ^7Be . The ^7Be background can be greatly reduced when the foil is kept clean and heated more than 3 hour at 200 °C before install in the detector. The background of ^{10}Be measurement has been investigated. In our experiment, three signals (ΔE_1 , ΔE_2 and E_t) are used to identify ^{10}Be , by gating on the three signals, the background form the ^{10}B and ^7Be can be identify very clearly. The measurement of blank sample showed that the background was around $n(^{10}\text{Be})/n(^9\text{Be})=1 \times 10^{-14}$.

As the rate of $n(^{10}\text{Be})/n(^9\text{Be})$ in the loess samples is very low, every sample's measure time is more than 1hour. Each sample is circular measured more than twice, then requested average value of gained $n(^{10}\text{Be})/n(^9\text{Be})$.

3 Result and discussion

A two-dimensional spectra ($E_t - \Delta E_1$) of standard sample is shown in Fig. 3(a). The figure shows that it is clear to identify ^{10}Be and ^{10}B . The spectra also show that there is another background ^7Be , this background is due to the $^1\text{H} (^{10}\text{B}, \alpha)^7\text{Be}$ reaction with hydrogen in the absorber entrance foil and the gas. Using the two-dimensional spectra, it is also clear to identify ^{10}Be and ^7Be . The background ^7Be can be greatly reduced when the foil is kept clean and heated more than 3 hours at 200 °C before install in the detector. The background of ^{10}Be measurement has been investigated. The two-dimensional ($E_t - \Delta E_1$) spectra of blank sample is also shown in Fig. 3(b). In our experimental, three signals (ΔE_1 , ΔE_2 and total energy E_t) are used to identify ^{10}Be , by gating on

the three signals, the background from ^{10}B and ^7Be can be identified very clearly. The measurement of blank sample showed that the background was around $n(^{10}\text{Be})/n(^9\text{Be}) = 2.9 \times 10^{-14}$.

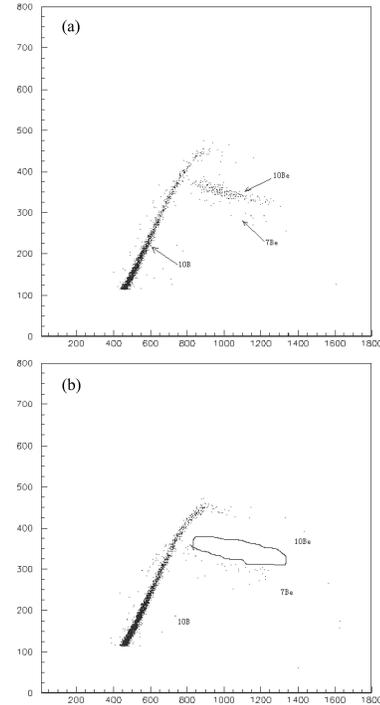


Fig. 3. Two-dimensional energy spectra ($E_t - \Delta E_1$) of BeO samples. (a) $n(^{10}\text{Be})/n(^9\text{Be}) = 2.68 \times 10^{-11}$ standard sample; (b) blank sample.

The results of AMS measurement of quartz samples are listed in table 1. Data analyses include calculated $n(^{10}\text{Be})/n(^9\text{Be})$ and its uncertainty, all the result revised the mass fractionation (namely take the metrical result of actual samples as the standard sample result unitary management).

The experiment took simultaneity measurement greatly improved the metrical precision, the ratio $n(^{10}\text{Be})/n(^9\text{Be})$ relatively standard deviation of samples in Table 1 far off less than relatively statistical error of ^{10}Be counts, the uncertainly of metrical result mainly come from the statistical error of ^{10}Be counts.

Table 1. AMS measurement results of loess samples.

| Sample | Unitary value of $n(^{10}\text{Be})/n(^9\text{Be})$ | Content of ^{10}Be in quartz(atom/gram) |
|--------|---|--|
| L8 | $(2.52 \pm 0.60) \times 10^{-13}$ | $(8.40 \pm 2.02) \times 10^6$ |
| L9 | $(1.73 \pm 0.42) \times 10^{-13}$ | $(5.77 \pm 1.37) \times 10^6$ |

take SRM 4325 as unitary consult.

4 Conclusions and expectation

This work has measured the concentrations of ^{10}Be of four formations in Loess section of Luochuan including L8, L9 and the concentrations of ^{10}Be in (of) blank sample, then calculated the content of ^{10}Be in loess quartz for each formation. It has set a base to calculate the buried age for each formation, confirm Loess stratigraphic age and realize the absolute dating for the older classic sediment.

We haven't finished the measurements of ^{26}Al because of the limitation of the beam time. The value of

$n(^{26}\text{Al})/n(^{27}\text{Al})$ is intending to be 10^{-14} or less. The concentrations of ^{26}Al in Loess section of Luochuan are to be measured not far in the future.

As the improved of accelerator mass spectrum and the accumulated of metrical data, the investigation extent of In-situ produced cosmogenic will embed continually and the range of application also will extend more. It can be expatiated that in come-at-able future, the measurement of In-situ produced cosmogenic accelerator mass spectrum will became an important tool in the earth's surface physical geography action and landform physiognomy research of Quaternary.

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