Measurement of the half-life of ^{79}Se with accelerator mass spectrometry*

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Abstract: The accelerator mass spectrometry (AMS) is an effective method for the determination of the half-life of long-lived radionuclides. In this paper, we report a method for measurement of the half-life of ^{79}Se. The number of ^{79}Se atoms was determined from measured ^{79}Se/Se absolute ratios with the AMS system at the China Institute of Atomic Energy and the decay rate of ^{79}Se was determined by counting the emitted β-rays with a liquid scintillation spectrometer. The major improvements of our measurements include using the high abundance of an ^{79}Se sample which was cooled for many years to exclude the interference of short-lived nuclides, the extraction of SeO_2^- molecular ions, that results in a suppression of the ^{79}Br background by as much as about five orders of magnitude. Also, an AMS measurement of the absolute ratio of ^{79}Se/Se was developed to avoid systematic errors. The results show that ^{79}Se/Se is $(2.35\pm0.12)\times10^{-7}$ in the reference sample and the radioactivity of ^{79}Se is (1.24±0.05) Bq/g, so the half-life of ^{79}Se is $(2.78\pm0.18)\times10^{5}$ a.

Key words: ^{79}Se, half-life, AMS, attenuator

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1 Introduction

As a very important long-lived radionuclide, ^{79}Se is interesting in many research fields, such as the assessment of nuclear repository, nuclear waste disposal and management [1, 2] tracer techniques in vivo and astrophysics. Up to now, the half-life of ^{79}Se has been estimated to be in the range of 10^4–10^6a [3–6]. In 2002, Songsheng Jiang and Ming He measured the half-life of ^{79}Se with PX-AMS at China Institute of Atomic Energy (CIAE) [7]. The measured half-life of ^{79}Se was $(2.95\pm0.38)\times10^5$ a, which has been accepted by the U.S. National Nuclear Data Center as the reference half-life of ^{79}Se. However, a new value for ^{79}Se′s half-life ($(3.77\pm0.19)\times10^5$ a) was determined by means of inductively coupled plasma mass spectrometry (ICP-MS) and liquid scintillation counting (LSC) on a sample source isolated from a nuclear reprocessing solution [8] and some German scientists got another value($(3.27\pm0.08)\times10^5$ a) with a similar method [9]. In the study of Jiang and He, selecting CdSe as a sample of AMS and separating ^{79}Se from ^{79}Br with projectile X-rays detection (PXD) reduced the detection efficiency of ^{79}Se. At the same time, some systematic errors were introduced into the ratio of ^{79}Se/Se when using the Faraday cup to measure the stable isotopes. On the other hand, no instrumental system presently available in ICP-MS is able to resolve all interferences in samples. In the above-mentioned study, for example, they just used a selective chemical separation procedure and electro-thermal vaporisation (ETV) coupled with ICP-MS to improve the selectivity for measurement, the ^{79}Br background could not be measured directly, but was deduced from measured ^{81}Br ions. In this work, a new method for the AMS measurement of the half-life of ^{79}Se is developed at CIAE.

2 Determination of the absolute ratio of ^{79}Se/Se with AMS

2.1 Sampling

Our sample came from the purified selenium (enriched in ^{78}Se to 57%) which was irradiated with the high neutron flux from the heavy water research reactor at CIAE in 2000. The ^{79}Se/^{78}Se ratio of the irradiated sample was calculated with the software ORIGEN to be $(8.21\pm1.28)\times10^{-5}$. After a short cooling, the sample

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was prepared by stepwise dilution with high-pure selenium dioxide (purchased from the Alfa Aesar Company with 99.999% purity) for some other experiments and the value for the ^{79}Se/Se ratio in the diluted sample was about the level of 10^{-7}. Before our experiment, the sample(^{79}Se/Se$\sim10^{-7}$) had been cooled for more than a decade, and the interference of short-lived nuclides was negligible. For the stable beam current extraction of SeO_2^-, we used Ag_2SeO_3 as the target substance, and the conversion of Se to Ag_2SeO_3 was done by the same chemical process as our established method for measuring ^{79}Se with AMS [10].

2.2 AMS measurement

An AMS measurement was performed by the CIAE-AMS system. The sample of Ag_2SeO_3 was pressed into the standard NEC aluminum target holder and the mass ratio of Ag_2SeO_3 to Ag (conductive medium) turned out to be 1:30. SeO_2^- ions were extracted from an ion source, which resulted in an effective suppression of the ^{79}Br background by as much as five orders of magnitude compared to the injection of Se^-. Because the ratio of ^{79}Se/Se in the reference sample was about the level of 10^{-7}, ^{79}Br will no longer be an obstacle for this measuremet.

$^{79}SeO_2^-$ ions were then selected by the injection system and injected into the accelerator. The negative molecular ions were accelerated by the tandem terminal voltage (7.95 MV). Stripping foil (3 μg/cm^2) was employed to break up the molecular ions and to produce atomic ions with high charge states. The resulting positively charged ions were further accelerated by the same terminal voltage. A 90° double focusing high energy analyzing magnet was used to select $^{79}Se^{11+}$ with an energy of 93.2 MeV. After a switching magnet, the $^{79}Se^{11+}$ was transported to the AMS beam line, then the $^{79}Se^{11+}$ was selected by using a 15° electrostatic deflector and finally detected with a fully depleted gold silicon surface-barrier detector (FGSBD). The ^{79}Se optical guidance in the accelerator was simulated with a ^{78}Se beam with the same magnetic rigidity as ^{79}Se. The difference of the transmission efficiency in the AMS system between these two isotopes is mainly rooted in the stripping probability of foil. For the correction of transmission, ^{80}Se was selected to be another pilot of the ^{79}Se transmission.

Usually, the beam intensity of ^{78}Se and ^{80}Se are measured with a Faraday cup, and the count of ^{79}Se is measured with the FGSBD. This will cause a larger system error for the ratio of ^{79}Se/Se by using the different detection system [11]. One straightforward method for the determination of the absolute ratio is to measure the counts of ^{79}Se, ^{78}Se and ^{80}Se with the same FGSBD. However, due to the very strong beam intensity of the ^{78}Se and ^{80}Se, the detector can not be used to measure them directly. In order to reduce the counts to an accepted high counting rate for the detector, we chose the mass ratio of Ag_2SeO_3 to Ag, the tandem terminal voltage and the selection of charge states as above. We also installed two attenuators in the CIAE-AMS system. This was done to bring the intensity of the beam to a value which is compatible with the detector's maximum counting rate. The attenuator consists of a perforated steel sheet. The holes are uniformly distributed on the sheet. The diameter of each hole is 50 μm and the distance between the holes is 1 mm. By locating the two attenuators at a distance of 5 m from each other and also separating them by one optic element, their effects are statistically independent and can be multiplied as a whole. One attenuator was installed just before the 15° electrostatic deflector. Another attenuator was installed 10 cm before the FGSBD. At these locations, due to the defining apertures and slits, the ion optics remain nearly unchanged for the different beam tunings.

Two pilot beams, ^{78}Se and ^{80}Se, were used for checking the influence of these attenuators. In the first step, the two attenuators were retracted and the relevant settings for the optimal transmission efficiency (from the Faraday cup in the low energy terminal to the target Faraday cup) for the ^{78}Se and ^{80}Se pilot beams were respectively obtained (Table 1).

Table 1. Transmission efficiency of ^{78}Se and ^{80}Se in the CIAE-AMS system.

parameters	value	
isotopes	^{78}Se	^{80}Se
implanting ions	$^{78}SeO_2^-$	$^{80}SeO_2^-$
beam intensity in the low energy terminal(LET)	11 nA	24 nA
tandem terminal voltage	8.05 MV	7.85 MV
charge states	11+	11+
beam intensity in the target	0.24 nA	0.41 nA
stripping efficiency	2.96%	2.43%
the total transmission efficiency from the LET to the target	0.198%	0.155%

We define the transmission efficiency of ^{78}Se from the LET to the target as follows:

$$Te(^{78}\text{Se})=\frac{\text{Beam intensity of }^{78}\text{Se in the target}}{\text{Beam intensity of }^{78}\text{Se in the LET}}\div 11\times 100\%. \quad (1)$$

Where 11 is the charge states of ^{78}Se and the transmission efficiency of $Te(^{78}\text{Se})$=0.198% was obtained. We also measured the total transmission efficiency of ^{80}Se with the same method and $Te(^{80}\text{Se})$=0.155% was obtained. The total transmission efficiency can be described as follows:

$$Te(\text{Se})=\eta\text{Se}_{\text{strip}}\times\eta\text{Se}_{\text{Rtran}}, \quad (2)$$

where $\eta\text{Se}_{\text{strip}}$ is the stripping efficiency of Se and $\eta\text{Se}_{\text{Rtran}}$ is the revised transmission efficiency of Se. By using the date in Table 1, the $\eta^{78}\text{Se}_{\text{Rtran}}$=6.69% and $\eta^{80}\text{Se}_{\text{Rtran}}$=6.38% were obtained, which means the total transmission efficiency of Se is mainly effected by stripping efficiency. The total transmission efficiency for ^{79}Se over the whole AMS system could be obtained by interpolation between the $\eta^{78}\text{Se}_{\text{Rtran}}$ and $\eta^{80}\text{Se}_{\text{Rtran}}$. The total transmission efficiency of $Te(^{79}\text{Se})$=0.175% was obtained (Eq. (3))

$$Te(^{79}\text{Se})=\eta^{79}\text{Se}_{\text{strip}}\times\frac{\eta^{78}\text{Se}_{\text{Rtran}}+\eta^{80}\text{Se}_{\text{Rtran}}}{2}, \quad (3)$$

where $\eta^{79}\text{Se}_{\text{strip}}$=2.68% was measured by WINBEAM 1.3.0. After that, the two attenuators were inserted, the ^{78}Se and ^{80}Se were measured with the FGSBD, and the mean value of $^{80}\text{Se}/^{78}\text{Se}$=1.65±0.05 was obtained (Fig. 1). After the correction of the total transmission efficiency:

$$^{80}\text{Se}/^{78}\text{Se}=(^{80}\text{Se}/^{78}\text{Se})_{\text{target}}\times Te(^{78}\text{Se})\div Te(^{80}\text{Se}), \quad (4)$$

the revised ratio of $^{80}\text{Se}/^{78}\text{Se}$=2.11±0.06 was obtained. As we know, the natural abundance ratio of $^{80}\text{Se}/^{78}\text{Se}$ is 2.09, which means that the two attenuators did not cause the difference of ion beam transport for ^{78}Se and ^{80}Se. Our method of absolute determination with AMS is reasonable.

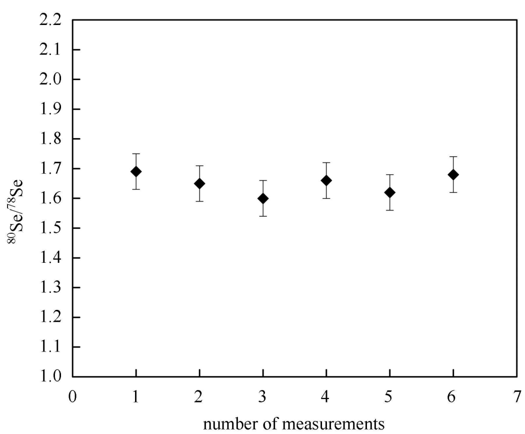

Fig. 1. The measured value of $^{80}\text{Se}/^{78}\text{Se}$ in the sample.

The attenuation coefficient of the attenuator was determined as follows:

$$Ac(^{78}\text{Se})=\frac{N_1(^{78}\text{Se})}{N_2(^{78}\text{Se})}, \quad (5)$$

where $N_1(^{78}\text{Se})$ and $N_2(^{78}\text{Se})$ are the counts of ^{78}Se at the condition of retracting and inserting the attenuator, respectively. We repeated the measurement of $N_1(^{78}\text{Se})$ and $N_2(^{78}\text{Se})$ for 6 times in each of the beam intensities, which was chosen in the LET (there are 5 different beam intensities at the same terminal voltage). The results are shown in Fig. 2. An attenuation coefficient of 108.0±1.0 was obtained.

After the system was checked with ^{78}Se and ^{80}Se, the ^{79}Se was measured with the FGSBD when the two attenuators were being retracted, and then the ^{78}Se was measured with the same detector after inserting the two attenuators. The ratio of the counting rate for ^{79}Se and ^{78}Se was obtained. The results are shown in Fig. 3. The mean value of $^{79}\text{Se}/^{78}\text{Se}$ is $(1.02\pm0.04)\times10^{-2}$. After considering the attenuation coefficient, total transmission efficiency, and natural abundance of ^{78}Se,

$$^{79}\text{Se}/\text{Se}=\frac{(^{79}\text{Se}/\text{Se})_{\text{target}}}{(108.0\pm1.0)^2}\times 23.78\%\times Te(^{78}\text{Se})\div Te(^{79}\text{Se}), \quad (6)$$

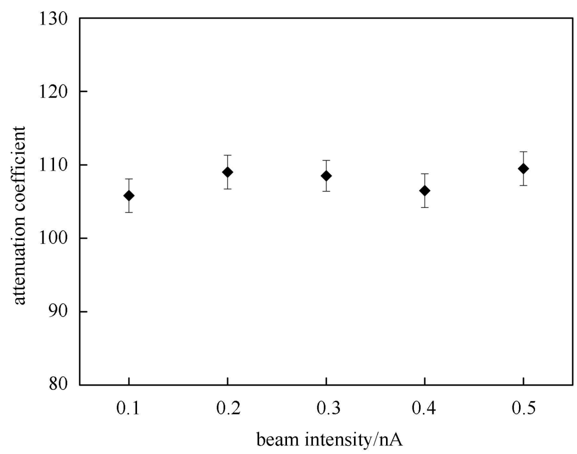

Fig. 2. The attenuation coefficient of the attenuator.

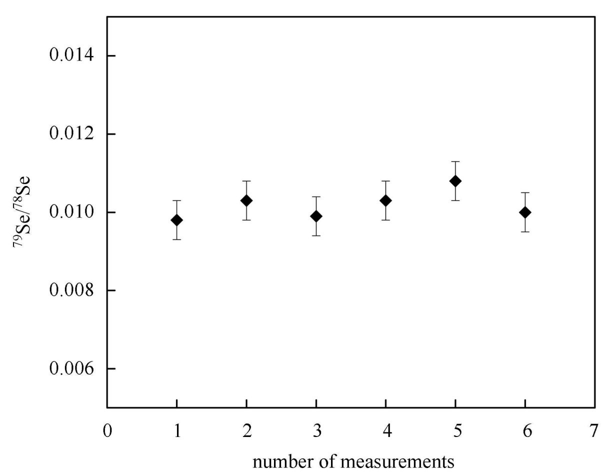

Fig. 3. The measured value of $^{79}\text{Se}/^{78}\text{Se}$ in the sample.

where 23.78% is the natural abundance of ^{78}Se. Introducing errors of the natural abundance of ^{78}Se, the total transmission efficiency of ^{79}Se and ^{78}Se. The absolute ratio of ^{79}Se/Se= $(2.35\pm0.12)\times10^{-7}$ in the sample was obtained. We also measured the concentration of total selenium in the sample with atomic absorption spectrometry and the result is 8764 μg/ml.

3 Determination of the decay of ^{79}Se

^{79}Se decays by β-emission with a maximum energy of 150.7 keV and no γ-emission. The decay rates of ^{79}Se in the samples were determined by counting emitted β-rays with an A317001 liquid scintillation spectrometer at CIAE. The quenching and counting efficiency were checked by using a standard solution of ^{14}C, the decaying β-rays of which have almost the same energy (156 keV) as that of ^{79}Se. In view of the low ^{79}Se activity, a background correction was performed with a blank sample (solution of H_2SeO_3), which was prepared with the same chemical procedure as the ^{79}Se sample, but with no ^{79}Se added. We used a 1.03209 g blank sample and a 0.96122 g reference sample (^{79}Se/Se=$(2.35\pm0.12)\times10^{-7}$, the same sample as we measured above) for the measurement. The room temperature was 24 ℃, relative humidity was 50%, and the time of detection was 600 minutes. The uncertainty of the determination of decay rates was about 2.4%, mainly coming from counting efficiency calibrated with a standard ^{14}C solution. Considering the recovery efficiency of ^{79}Se for making the Ag_2SeO_3 sample and statistical uncertainty, the uncertainty of the decay rate for the Ag_2SeO_3 sample was 4%. The decay rate of ^{79}Se in the sample was (1.24±0.05) Bq/g.

4 Results and discussion

Considering the total amount of selenium in the sample and combining the decay rate with the ratio of ^{79}Se/Se, the half life of ^{79}Se can be deduced by the relationship:

$$\mathrm{d}N/\mathrm{d}t=-\lambda N(t), \tag{7}$$

where $\mathrm{d}N/\mathrm{d}t$ is the decay rate, $N(t)$ is the number of ^{79}Se at the time t, $\lambda=\ln2/T_{1/2}$ is the decay constant, and $T_{1/2}$ is the half-life. The half-life of ^{79}Se is deduced to be $(2.78\pm0.18)\times10^5$ a and the relative standard deviation is 6.5%. This result is similar with the value of $(2.95\pm0.38)\times10^5$ a (Jiang and He et al., 2002). Compared with the previous method, the uncertainty from the ^{79}Br isobaric background and systematic errors of different detectors is negligible and the detection efficiency of FGSBD is better than PXD. So, the relative standard deviation of the half-life of ^{79}Se is reduced from 12.9% to 6.5% in our measurement. In view of the general difficulty to know all systematic uncertainties correctly, we want to do an independent half-life measurement in the future.

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