Measurement of ^{79}Se by accelerator mass spectrometry using projectile X-ray technique

HE Ming, JIANG Shan, JIANG Song-Sheng, WU Shao-Yong,
Zhao Zhan-Xia, Dong Yue-An
(*China Institute of Atomic Energy, Beijing 102413*)

Abstract In order to carry out the measurement of heavy nuclei in accelerator mass spectrometry, characteristic X-rays of the incident projectile have been explored and used as a method for isobar discrimination. The projectile X-ray combined with AMS technique has been set up in China Institute of Atomic Energy. The measurement of ^{79}Se was performed by resorting to the projectile X-ray AMS technique and the detection sensitivity of ^{79}Se was improved more than 2 orders of magnitude. The detection limit was about 3.6×10^{-9} for ^{79}Se/Se.

Keywords Tracer ^{79}Se, AMS, Projectile X-ray

CLC numbers TL817^{+}.4, TL817^{+}.6, TL817^{+}.1

1 Introduction

In accelerator mass spectrometry (AMS) measurements of long-lived radioisotopes, stable isobars are the dominant background. For the radioisotopes of atomic number $Z \leq 20$, the isobars background can be eliminated by means of the different energy loss of isobar in matter due to the rate of energy loss is a function of Z. But with increasing atomic number, the energy straggling increases relative to the energy loss difference, so that isobar separation becomes progressively less effective. In order to separate the isobars with higher atomic numbers the ion energy has to be raised to higher values. However, for the energies accessible with larger tandem accelerator, the highest atomic number that can be separated is about $Z \approx 25 \sim 30$. The projectile X-ray is a new isobar rejection method, which does not rely on the energy loss, and the isobar discrimination capability is nearly independent of ion energy. The projectile ion are identified by the characteristic X-rays that emit when slowing down in target[1]. AMS combined with the projectile characteristic X-ray, which was referred as PX-AMS, can improve heavy nucleus detection sensitivity. So, some long-lived radionuclides (^{59}Ni, ^{79}Se, ^{93}Zr, ^{93}Mo, ^{99}Tc, ^{107}Pd) which were not able to be detected with required sensitivity before can be measured now with the method in principle[2]. ^{79}Se, as a tracer, is interesting in the study of selenium biochemistry and metabolism, and as a component of long-lived fission product for nuclear waste repository[3]. Especially in China it is more interesting to study the endemic disease (Kashan and kaschin-beck diseases) which related to Se.

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The values of published half-live of ^{79}Se are more than 2 orders of magnitude apart.[4] In order to carry out the measurement of ^{79}Se, the PX-AMS technique was used. For the first time the ^{79}Se was measured at CIAE with PX-AMS and the half-life of ^{79}Se has been obtained.

2 Elimination of the interference in the ^{79}Se measurement

The measurement of ^{79}Se was carried out by means of PX-AMS technique at China Institute of Atomic Energy. The schematic diagram of the PX-AMS system is shown in Fig.1. The measurement was performed in Charge State 9^+ at accelerator's terminal voltage of 8.05 MV. In the AMS measurement the interference come from their isotopes and isobars of interesting nuclei. The isotopic interference was eliminated by deflector and the isobaric interference was eliminate by its characteristic K_α ray.

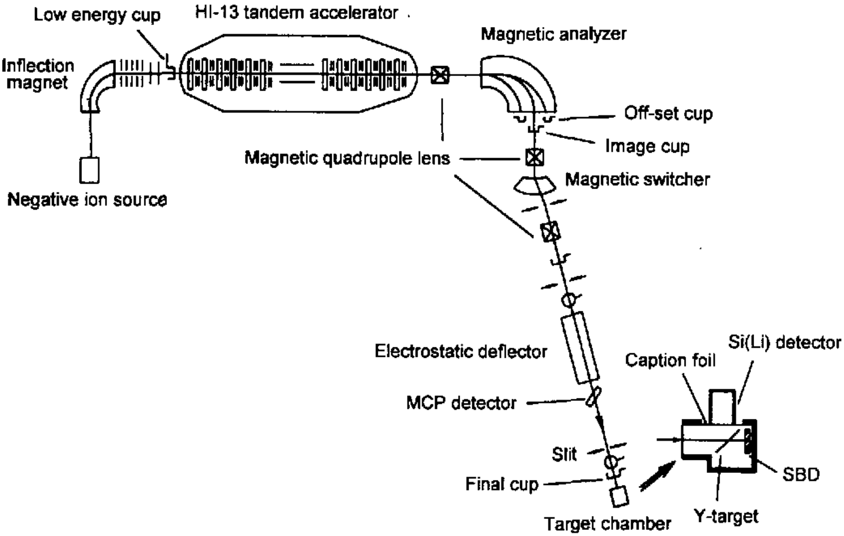

Fig.1 Schemiatic diagram of the PX-AMS system at the China Institute of Atomic Energy

2.1 Elimination of isotopic interference

The characteristic X-ray energy depends on atomic number, the isotopes ^{78}Se and ^{80}Se with the same K_α energy as ^{79}Se. So, in order to eliminate isobar interference by using the projectile X-rays, these isotopes interference must be eliminated first. The chemical form of CdSe was used as sample material for increasing Se^- current and reducing the isotopes interference. ^{78}Se and ^{80}Se are the main interference in the measurement of ^{79}Se. ^{78}Se and ^{80}Se, which have the same magnetic rigidity as ^{79}Se after magnetic analyzer, have energy higher and lower 1.013% than that of ^{79}Se, respectively. The electrostatic deflector was used to eliminate the isotopic interference. When the ions pass through the deflector, the ^{80}Se (^{78}Se) beam spots is about 12 mm lower (higher) than the ^{79}Se at a slit (4 mm in vertical direction and 7 mm in horizontal direction). In order to separate the isotopic interference, the beam size is focus into less than 4 mm in the vertical direction at the slit. Fig.2 shows the isotopic interference are eliminated by adjusting the deflector voltage. The corresponding isotopic background is about ^{79}Se/Se$\approx 10^{-10}$ after the deflector.

2.2 Target

When bombarding a target with ions, one observes characteristic X-rays from the ions and target elements. The yields of the projectile X-rays have strong correlation with targets and ions energy. When there is a match between the K shell level of projectile and that of target, there is a strong resonance in the K shell vacancy production for the incident ions, Kubo *et al* studied the situation and explained it by resorting to the molecular orbitals[5]. When ion bombards a solid targets, a good target is an element with an atomic number that is slight higher than that of the interesting projectile. This is because there is a match of the inner shell binding energy between the projectile and the target (multiple collisions lead to outer-shell excitation which cause the projectile inner shell binding energy increase[6]). In this way, the X-ray yield from the projectile is at its maximum and the count rate from the target is quite low. In the measurement of ^{79}Se, the Y target was selected because Se K_α ray yield is at its maximum.

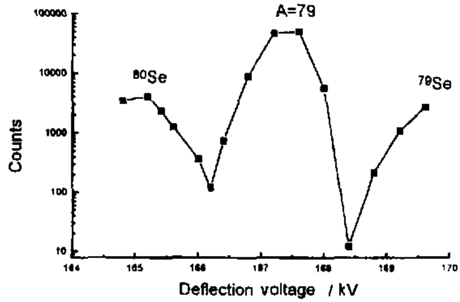

Fig.2 Isotopic interference in the measurement of ^{79}Se

2.3 Elimination of isobaric interference

After the ions pass through the slit, the isotopic interference is considered to be negligible. Only the ^{79}Se and its isobar ^{79}Br collide with Y target. Their characteristic K_α rays are emitted and measured by a Si(Li) detector. The difference of Se K_α ray

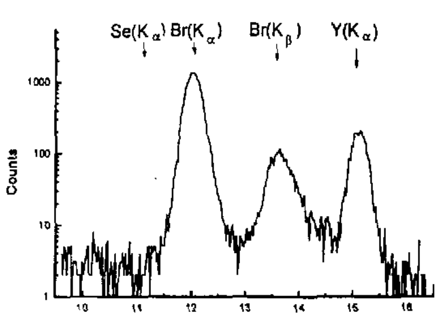

Fig.3 X-ray spectrum for a ^{79}Se blank sample

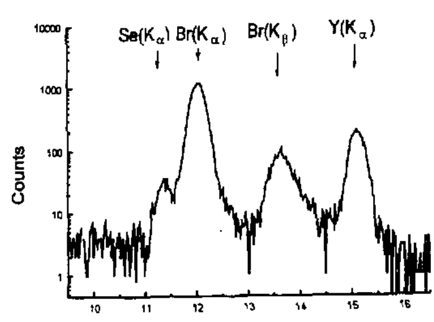

Fig.4 Measured X-ray spectrum for a ^{79}Se sample

(11.222 keV) and Br K_α ray (11.924 keV) is big enough to be separated by the Si(Li) detector (energy resolution (FWHM)=174 eV for 5.9 keV X-ray). Fig.3 is the X-ray spectrum of a ^{79}Se blank sample, Fig.4 is the X-ray spectrum of a ^{79}Se sample. The Y peak originated from the excitation of the target foil. The X-rays energy is higher compared to the X-rays of a single ionized atom[7] Fig.3 shows that a fraction of Br K_α counts extend through the Se region as a low energy tail. The suppression factor (ratio of Br counts in the Br region to Br counts in the Se region) in $\pm 1\sigma$ is about 250 and the background corresponding to ^{79}Se/Se ratio of 3.6×10^{-9}. The sensitivity of the ^{79}Se measurement was improved more than 2 orders of magnitude when the projectile X-ray method combined with AMS technique.

3 Determination of ^{79}Se atoms in samples

In order to get the ^{79}Se atomic number from the counts of ^{79}Se K_α peak, it is necessary to determine the ^{79}Se K_α detection efficiency. The efficiency is related to the solid angle, the intrinsic detection efficiency for ^{79}Se K_α ray, ion energy and target thickness. For getting the ^{79}Se atoms in sample, it is necessary to measure the ^{79}Se/Se ratio of the sample.

3.1 Detection efficiency for ^{79}Se K_α rays

^{79}Se K_α ray detection efficiency can not be measured directly due to isobaric interference. Isotopes ^{78}Se and ^{80}Se were used. As mentioned above, a deflector can eliminate the isotopic interference. On the other hand, the deflector can choice ^{78}Se and ^{80}Se by adjusting the deflector voltage. When ^{80}Se (^{78}Se) bombards a Y target, the K_α counts of ^{80}Se (^{78}Se) are measured with a Si(Li) detector at 90° with respect to the incident beam (for avoiding Doppler broadening and bremsstrahlung background caused by the deflector). The counts of ^{80}Se (^{78}Se) are measured by the surface barrier detector directly after the Y target is removed. Then we obtained the ^{80}Se(^{78}Se) detection efficiency. The detection efficiency of ^{79}Se can be calculated from the detection efficiency of ^{78}Se and ^{80}Se by an interpolation. In this experiment, the ion energy is 80.5 MeV, the thickness of Y target is 3.8 mg/cm^2, the distance between the target and the Be window of Si(Li) detector is about 5 mm. The overall detection efficiency for the system was measured to be $(7\pm0.4)\times10^{-4}$ for K_α counts per incident ^{79}Se. The uncertainty is from the statistics error and reproducibility in the measurement of Se K_α peak counts and Se counts.

3.2 Measurement of ^{79}Se/Se ratio

For the measurement of ^{79}Se/Se ratio, the ^{80}Se current was measured with a final Faraday cup (behind the slit and in front of the Y target), the ^{79}Se was detected by K_α ray, and the atoms number of ^{79}Se was calcualted from ^{79}Se K_α counts and ^{79}Se K_α detection efficiency. The ion transfer efficiency from the final Faraday cup to the surface barrier detector (ϕ12 mm) was consider to be 100% within 10% uncertainty. From ^{80}Se current and the ^{79}Se counts we obtained the ^{79}Se/^{80}Se ratio. The ratios of ^{79}Se/Se in the samples were deduced after a correction for stripping probability of these isotopes and ^{80}Se isotopic abundance. The ^{79}Se atomic number of samples was calculated from the ratio of $^{79}Se/Se$ by using the relation: $N_{79} = M\,r\,N_0/A$, where M is the weight of CdSe sample, r is the ratio of $^{79}Se/Se$, A is the CdSe mole weight, N_0 is the Avogadro's number.

4 Preliminary measurement of ^{79}Se half-life

The half-life of ^{79}Se has been measurement with AMS technique[4]. In this experiment, isobar ^{79}Br was deduced from measurement of ^{81}Br. Now the ^{79}Br interference can be eliminated and ^{79}Se can be measured by the PX-AMS directly. The half-life of ^{79}Se was re-measured. Two ^{79}Se samples have been used.

From the measured decay rate, dn/dt, and the number of ^{79}Se atoms, N, the half-life($T_{1/2}$) can be deduced using the relation of dn/dt=-(ln2)·$N/T_{1/2}$. From our preliminary experimental results we get the half-life of ^{79}Se. The half-life is about $(1.24\pm0.19)\times10^5$a. The data for determination of the half-life are given in Table 1. The error is due to the systematic uncertainty, the uncertainty of ^{79}Se X-ray detection efficiency and the reproducibility in reading ^{80}Se current and ^{79}Se K_α counts, respectively.

Table 1 Data for determination of the ^{79}Se half-life

Sample No.	$(^{79}Se/Se)/10^{-8}$	N_{79}/atoms·(mgCdSe)$^{-1}$	(dn/dt)/(dis/min)·(mgCdSe)$^{-1}$	$T_{1/2}$/a
1	3.88±0.57	$(1.22\pm0.18)\times10^{11}$	1.32±0.07	$(1.22\pm0.19)\times10^5$
2	4.02±0.59	$(1.26\pm0.18)\times10^{11}$	1.31± 0.07	$(1.27\pm0.19)\times10^5$
Mean value				$(1.24\pm0.19)\times10^5$

5 Conclusions

From these experiments, it can be seen that combining projectile X-ray detection with AMS technique provides an effective method for detecting heavy nuclei at relatively low ion-energy. The isobaric interference and the isotopic interference currently limit the ^{79}Se detection sensitivity. A time of flight detector set in the exit of AMS beamline will allow rejection of the isotopic interference. The isobaric interference could also be reduced by combining PX-AMS with a SBD detector placed behind a relatively thin Y target. The SBD could be made from a residual energy E_R detector. Exploring chemical separation technique to further lower the Br background can also reduce the isobaric interference. Then the sensitivity of ^{79}Se detection will be $^{79}Se/Se\sim10^{-10}-10^{-11}$.

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