Determination of ^{79}Se in nuclear waste via AMS

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Among the long-lived radionuclides produced in nuclear reactors, ^{79}Se ($T_{1/2} = 1.24 \times 10^5$ y [1]) is of special interest for nuclear waste disposal, because it is very difficult to make reliable statements about its concentration. One cannot use vectorial fission nuclides such as ^{137}Cs for predicting the ^{79}Se activitiy, since it is produced by fission as well as by neutron activation. Also a measurement of the ^{79}Se activity is problematic because of its lo g half-life and its decay without γ-ray-emission. Additionally, the maximum β^- energy is only 151 keV.

For this reasons we attempted to measure ^{79}Se in nuclear waste samples with accelerator mass spectrometry, i.e. not by means of decay counting but by particle counting. Several factors, however, complicate AMS measurements of ^{79}Se compared to routinely measured AMS isotopes:

i) Bromium is a very volatile element which inevitably results in relatively high contaminations of the interfering stable isobar ^{79}Br in the samples.

ii) Bromium belongs to the halogens and therefore has a very high yield for the formation of negative ions in the sputtering process leading to an even higher ^{79}Br contamination in the beam.

iii) The relative difference of the nuclear charges of $^{79}_{34}$Se and $^{79}_{35}$Br is very low. Both the separation in a gas-filled magnet (see fig.1) and an energy loss detector are Z-dependent and for this reason less efficient.

iv) Both neighboring isotopes ^{78}Se and ^{80}Se are stable and are highly abundant (23.8 % and 49.6 %, respectively).

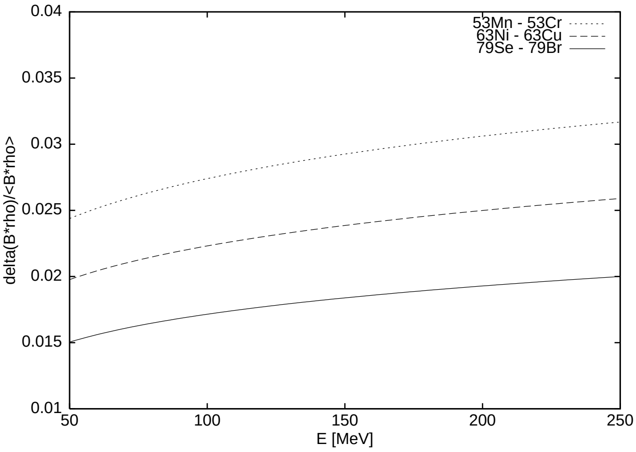

Fig. 1: Relative difference in magnetic rigidity for ^{79}Se and ^{79}Br in a gas-filled region (Calculation using the semi-empirical formula given in [2]). For comparison also the rigidity differences of the isobar pairs ^{53}Mn–^{53}Cr and ^{63}Ni–^{63}Cu are given.

Due to these complications only the Beijing AMS facility has published work on ^{79}Se up to now [3,1]. In ref. [3] no isobar separation was made at all. The tuning of the beam line was switched to $A = 81$ (^{81}Br) in order to subtract the ^{79}Br events from the signal at $A = 79$ (^{79}Se + ^{79}Br). In ref. [1] ^{79}Se and ^{79}Br were distinguished by their characteristic K_α X-rays emitted in a stopper foil. With this met od the sensitivity was improved to ^{79}Se/Se $= 3.6 \times 10^{-9}$.

Our attempt was a ^{79}Br rejection by means of the gas-filled magnet and the position sensitive ionization chamber with five energy loss sections (GAMS) [4] as already successfully used in measurements of ^{60}Fe [5], ^{63}Ni [6], or ^{53}Mn [7].

Due to the small spatial separation of ^{79}Se and ^{79}Br a fraction of ca. 0.4 % of the ^{79}Br ions entered the detector, even when a loss of about 50 % of the ^{79}Se ions was tolerated. To avoid counting rates above a few kHz only low beam intensities could be used. However, this disadvantage could be compensated by using a high charge state. For the 15+ state the stripping probability is only 1 % yielding both acceptable counting rates and high energies of 216 MeV which re advantageous for isobaric separation (see. fig. 1).

For a first test four samples of ion exchange resins from nuclear power plants were analyzed. After adding about 10 mg of stable selenium carrier selenium was extracted at the Radiochemical Institute. ^{79}Se/Se ratios between 1×10^{-9} and 1×10^{-7} were determined via AMS, corresponding to 8×10^{10} – 8×10^{12} ^{79}Se atoms, or activities between 14 and 1400 mBq.

For calibration a standard was produced by neutron activation of enriched ^{78}Se. The neutron flux was determined by the activity of ^{75}Se, produced via ^{74}Se(n,γ)^{75}Se.

The combination of gas-filled magnet and ionization chamber resulted in a ^{79}Br suppression of more than six orders of magnitude. Although there were ^{79}Br contaminations of 100 – 1000 ppm in the beam, the background level corresponded to ^{79}Se/Se ratios as low as a few times 10^{-10}.

Contaminations below 1 ppm [3] were already reached with a more elaborated sample preparation . Therefore, measurements of ^{79}Se at a 10^{-12} concentration level should become possible with GAMS. At this level isotopic background of ^{78}Se and ^{80}Se is no longer negligible, but can be easily suppressed by an additional time-of-flight measurement in front of the gas-filled magnet.

References

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