AMS MEASUREMENT OF ^{59}Ni AT CHINA INSTITUTE OF ATOMIC ENERGY

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ABSTRACT. The long-lived radioisotope ^{59}Ni is of interest in various research fields including neutron dosimetry, radioactive waste management, and astrophysics. In order to achieve the sensitivity required for such applications, the technique of accelerator mass spectrometry (AMS) ^{59}Ni measurement has been developed at the AMS facility at China Institute of Atomic Energy (CIAE). Based on the ΔE-Q3D detection system in the CIAE AMS facility, the interference in ^{59}Ni counting from the isobar ^{59}Co has been reduced by a factor of 8×10^6. A series of laboratory reference samples and a blank sample were measured to check the performance of ^{59}Ni measurement. A detection sensitivity of about 5×10^{-13} (^{59}Ni/Ni) has been obtained.

INTRODUCTION

^{59}Ni, with its half-life of 76 ka (Nishiizumi et al. 1981), is of interest in applications related to low-level radioactive waste management (Mount et al. 1998), for monitoring cosmic radiation (Hasegawa et al. 1968), neutron dosimetry (Persson et al. 2000), and astrophysics (Rugel et al. 2007; Dillmann et al. 2010). However, due to its long half-life and pure electron-capture decay without gamma emission, it is very difficult to measure ^{59}Ni via a decay-counting method. Accelerator mass spectrometry (AMS) is an ultra-sensitive technique used for counting long-lived radioisotopes. The main problem in AMS measurements of ^{59}Ni, however, is the interference from the stable isobar ^{59}Co, which is difficult to remove by chemical separation methods to concentrations below 1 ppm in Ni samples. In order to achieve the sensitivity required for the above-mentioned applications, a different technique for reducing the interference of ^{59}Co was applied at AMS laboratories (Kutschera et al. 1993; Paul et al. 1993; McAninch et al. 1997; Wallner et al. 2007). The LLNL AMS group developed a method for identifying ^{59}Co and ^{59}Ni ions according to their different K X-ray energies (McAninch et al. 1997). However, due to the limited identification power and low X-ray detection efficiency, their sensitivity for ^{59}Ni/Ni was limited to about 10^{-11}. A full-stripping technique was used for reducing the ^{59}Co interference at the Argonne National Laboratory (Kutschera et al. 1993) utilizing a particle energy of 641 MeV. This technique requires very high ion energies, which are unreachable in most existing AMS systems. In another approach, a gas-filled magnet (GFM) combined with a multi-anode gas ionization chamber is used for eliminating the ^{59}Co interference that is based on a 14MV MP tandem accelerator (Wallner et al. 2007). ^{59}Ni and ^{59}Co ions are spatially separated in the GFM due to their different mean charge state when passing through the gas in the magnet. A gas ionization chamber is used to further distinguish ^{59}Co and ^{59}Ni ions. Based on this technique, a sensitivity for ^{59}Ni/Ni of $\sim 10^{-14}$ was reported.

Recently, a ΔE-Q3D detection system has been developed at CIAE for measurements of medium-mass (mass number around 60) nuclides (Li et al. 2010) that is based on the HI-13 tandem accelerator. Details of the AMS setup were described previously (Dong et al. 2011). The radionuclides ^{32}Si and ^{53}Mn have been measured by the CIAE AMS facility with the ΔE-Q3D detection system (Dong et al. 2011; Gong et al. 2011). In this paper, a method to measure ^{59}Ni by the CIAE AMS facility is presented.

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SAMPLE PREPARATION

The ^{59}Ni sample was produced by a neutron-induced ^{58}Ni(n,γ)^{59}Ni reaction. In this work, 50.2 mg of high-purity natural Ni foil (99.994%) and 25.3 mg of Zr foil were irradiated with well-thermalized neutron flux from a reactor at CIAE. The neutron flux was determined by measuring the ^{95}Zr-emitting γ-ray with a HPGe detector and the well-known thermal neutron capture cross-section of ^{94}Zr(n,γ)^{95}Zr. The ^{59}Ni/Ni of the irradiated sample was determined to be $(2.06 \pm 0.09) \times 10^{-7}$ from the determined thermal neutron flux and the well-known thermal neutron capture cross-section value (4.13 ± 0.05b) of the ^{58}Ni(n,γ)^{59}Ni. After a cooling time of 1 month, the sample was used to prepare a series of laboratory reference samples by the following procedures. First, the irradiated sample was dissolved with 9N HCl. Subsequently, a series of reference solutions with different ratios of ^{59}Ni/Ni were prepared by stepwise dilution with accurate quantitative $NiCl_2$ solution. Ion exchange separation of cobalt and nickel is readily accomplished via chloride solution, which is based on the much stronger tendency of Co in forming chloroanions, i.e. $CoCl_3^-$ and $CoCl_4^-$, than that of Ni. Each solution was then placed onto an AG1 anion exchanger column. After passing the column, the eluate was collected and ammonia was added to the eluate until pH = 9 was reached and nickel was precipitated with the addition of dimethylglyoxime. This complex was centrifuged and ashed at 600 °C to produce NiO powder, which served as target material for the AMS measurement. A series of reference samples were prepared with ^{59}Ni/Ni ratios between 10^{-8} and 10^{-12}.

The chemical form of $NiCl_2$ was also tested as a target material. However, the maximum Ni^- beam current from a $NiCl_2$ target was 10 times lower than that of the conventional NiO target material. Furthermore, due to the hydroscopic character of $NiCl_2$, it easily forms NiH^-. $^{58}NiH^-$ with the same mass number of 59 will be injected together with $^{59}Ni^-$ into the tandem accelerator, thus causing background interference in the measurement of ^{59}Ni. NiO powder was therefore selected as the target material for our AMS measurements of ^{59}Ni.

ACCELERATOR MASS SPECTROMETRY OF ^{59}Ni

AMS System at CIAE

A dedicated ion injection system is used for AMS measurement. This injection system comprises a 40-sample NEC MC-SNICS ion source, a 90° spherical electrostatic analyzer, and a double-focusing 112° injection magnet. Two movable offset Faraday cups located at the injection magnet image side can be used for continuously measuring the beam current of the stable isotopes. The mass resolution of the injection system is in the range of M/M = 400–600, depending on beam optics and defining slits. A terminal voltage, up to 12MV, is produced in the middle of the accelerator tube. Carbon foils of thickness of 3 $\mu g/cm^2$ are used for stripping. The high-energy analysis system consists of a 90° double-focusing magnet followed by a switching magnet that transports the beam to different beam lines. Two beam lines are now usable for AMS experiments. Beam line 1 with a 15° electrostatic deflector is dedicated to AMS experiments. Beam line 2 with a Q3D magnet spectrometry is a multi-user beam tube. The AMS measurements of ^{59}Ni reported here were conducted with beam line 2.

Ion Beam Transport

The NiO powder was mixed with the same weight of pure Ag powder and pressed into Al target holders and loaded into the ion source. Ni^- ions were extracted from the ion source. After passing through the electrostatic analyzer and the injection magnet, $^{58}Ni^-$ and $^{60}Ni^-$ beams were inwardly and outwardly deflected relative to the $^{59}Ni^-$ trajectory, and the currents of $^{58}Ni^-$ and $^{60}Ni^-$ were measured by the 2 offset Faraday cups. $^{59}Ni^-$ ions were transported to the accelerator, which was set to a terminal voltage of 11.5 MV. The ions were accelerated to 11.5 MeV, and at the terminal they passed through a carbon stripper foil to produce Ni ions with positive charge states. A 90° double-focusing analyzing magnet was used to select $^{59}Ni^{12+}$ (and $^{59}Co^{12+}$) ions with a calculated energy of 149.5 MeV. None of these processes can separate ^{59}Ni and the stable isobar ^{59}Co. After passing a switching magnet, both the $^{59}Ni^{12+}$ ions and the isobar $^{59}Co^{12+}$ of the same energy were transported to the ΔE-Q3D detection system.

Isobar Separation with ΔE-Q3D

The ΔE-Q3D detection system consists of a Q3D magnetic spectrometer with an absorber, which is used to absorb part of the ions' energy after they pass through it, at its entrance, and a 4-anode gas-ionization chamber with an entrance window of 70 mm × 40 mm at its focal plane. The Q3D magnetic spectrometer has an energy resolution of $\Delta E/E = 1.8 \times 10^{-4}$ and a dispersion of $\Delta X = 11.378$ cm/%/($\Delta P/P$) (Li et al. 1993).

For our ^{59}Ni measurements, a Si_3N_4 foil (Silicon Ltd./UK) with a thickness of 5 µm (made as a stack of 5 foils, each with a thickness of 1 µm) was used as an absorber. After $^{59}Ni^{12+}$ ions pass through the Si_3N_4 membrane, ions with charge states of 19^+, with a stripping probability of ~20%, were analyzed by the Q3D magnetic spectrometer. Due to the different energy losses of ^{59}Ni and ^{59}Co in the absorber, $^{59}Ni^{19+}$ and $^{59}Co^{19+}$ ions were produced with respective energies of 99.4 and 101.5 MeV, which were calculated with the SRIM-2000 program (Ziegler et al. 2000). This energy difference means that the magnetic rigidity of ^{59}Co was 1.05% higher than that of ^{59}Ni, corresponding to a peak distance of 120 mm between ^{59}Ni and ^{59}Co at the focal plane of the Q3D magnetic spectrometer.

A sample with a nominal isotope ratio $^{59}Ni/Ni = (8.53 \pm 0.51) \times 10^{-9}$ was used to check the separation between ^{59}Ni and ^{59}Co at the focal plane. The count rates of the ^{59}Ni and ^{59}Co ions as functions of the magnetic field of the Q3D were measured with the 4-anode gas ionization chamber. The results are shown in Figure 1. The optimum magnetic fields for ^{59}Ni and ^{59}Co are 0.4830T and 0.4890T, respectively. The optimum magnetic field for ^{59}Co is 1.24% higher than that for ^{59}Ni, corresponding to a peak distance of 140 mm between ^{59}Co and ^{59}Ni at the focal plane, which is a little higher than the calculated values with SRIM-2000 program. Setting the magnetic field of the Q3D to 0.4830T resulted in more than 70% of $^{59}Ni^{19+}$ events relative to all the $^{59}Ni^{19+}$ events at the focal plane recorded by the ionization chamber with much reduced interference from ^{59}Co. A suppression factor of about 500 for ^{59}Co was obtained.

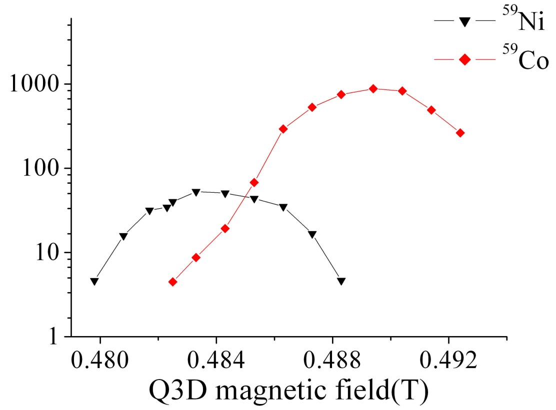

Figure 1 Count rates of ^{59}Ni and ^{59}Co at a given position of the focal plane as a function of the Q3D magnetic field.

^{59}Ni Determination with Gas Detector

Although most of the ^{59}Co was eliminated with the ΔE-Q3D technique, a small fraction of ^{59}Co ions can still enter the gas detector due to straggling and scattering effects. Further, the difference in their energy losses in the detector gas (Figure 2), was used to identify the number of ^{59}Ni ions with the 4-anode gas ionization chamber. Propane with a pressure of 25 mbar was used as detector gas of the ionization chamber. Five signals, i.e. 4 energy loss signals (E_1, E_2, E_3, E_4) from the anodes and a total energy signal (E_t) from the cathode, were used. A multiparameter data acquisition system was used to get the 5 signals. Figure 3 shows the spectra of E_1 versus E_3, and E_2 versus E_4 for a laboratory reference sample with a ^{59}Ni/Ni ratio of $(1.15 \pm 0.07) \times 10^{-9}$. The corresponding spectra for a blank sample are shown in Figure 4. It can be seen from these spectra that ^{59}Ni and ^{59}Co can be separated by the detector, although there is overlap between ^{59}Ni and ^{59}Co. In order to further remove the ^{59}Co background and obtain the ^{59}Ni counts, the multiparameter data gathered was analyzed off-line with appropriate gate coincidences on these 5 signals. About 85% of ^{59}Ni counts were accepted with appropriate gates on the 5 detector signals; at the same time, the ^{59}Co background was strongly reduced. According to blank sample measurements, more than 1.5×10^5 ^{59}Co counts are accumulated with only 10 counts remaining in the ^{59}Ni peak region after applying the same gates. The results show that a suppression factor of 1.5×10^4 for ^{59}Co can be achieved with the ionization chamber.

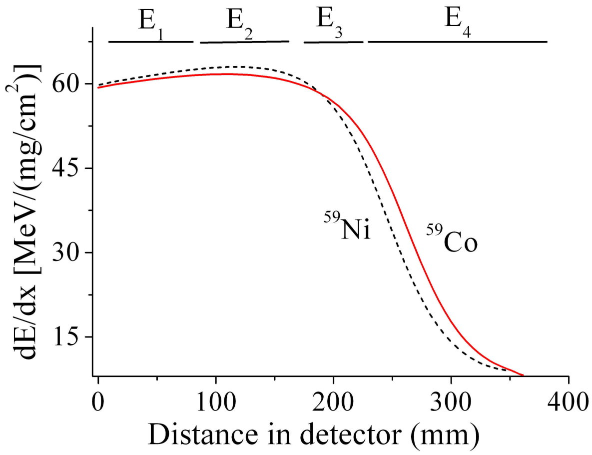

Figure 2 Calculated energy losses of ^{59}Ni and ^{59}Co versus ion position in the detector as determined by the SRIM-2000 program for a particle energy of 89 MeV entering the detector gas after considering the energy loss in the 2.0-µm Mylar detector window.

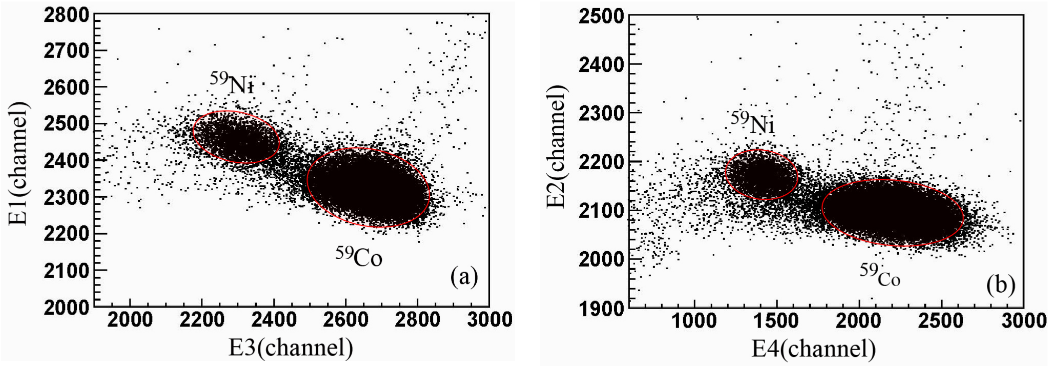

Figure 3 Two-dimensional spectra of E_1 versus E_3 (a) and E_2 versus E_4 (b) for a laboratory reference sample with ^{59}Ni/Ni = 1.15×10^{-9}.

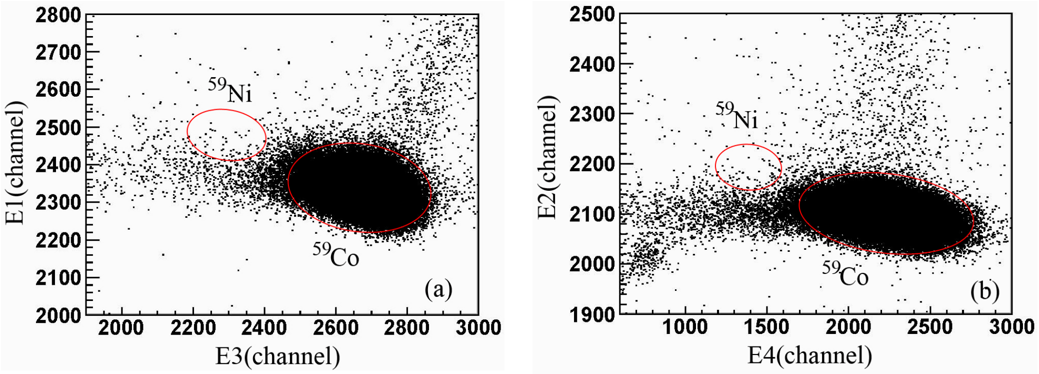

Figure 4 Two-dimensional spectra of E_1 versus E_3 (a) and E_2 versus E_4 (b) for a blank sample

RESULTS AND DISCUSSION

A series of laboratory reference samples and a blank sample with the same chemical treatment were measured with this method. There is no fast switching system in the CIAE-AMS system. In order to monitor the beam current during ^{59}Ni measurement, simultaneous measurement of the beam current of ^{60}Ni$^-$ or ^{58}Ni$^-$ with the offset Faraday cups is used. The ratio of ^{60}Ni$^-$/^{58}Ni$^-$ measured with the offset Faraday cup agreed with the natural abundance ratio. This means there was no interference in the beam current measurement of ^{60}Ni$^-$ and ^{58}Ni$^-$ with the offset Faraday cup. By this method, the beam current of ^{60}Ni$^-$ was measured simultaneously during ^{59}Ni measurement with the 4-anode gas ionization chamber. These 2 values combined with the AMS system transmission and detection efficiency were used for ^{59}Ni/Ni ratio determination. The sample with a ^{59}Ni/Ni ratio of $(8.53 \pm 0.51) \times 10^{-9}$ was used as the standard sample. The standard sample and other samples were measured in turn for normalizing the AMS system transmission and detection efficiency. The typical time for each measurement was 5 min, but 30 min for the blank sample. These measurements were repeated 5 times. After normalizing by the standard sample value, the ^{59}Ni/Ni ratios of other samples including the blank sample were obtained. The measured ^{59}Ni/Ni ratio vs. nominal values for the series of laboratory reference samples are shown in Figure 5. The values for blank samples are also shown in

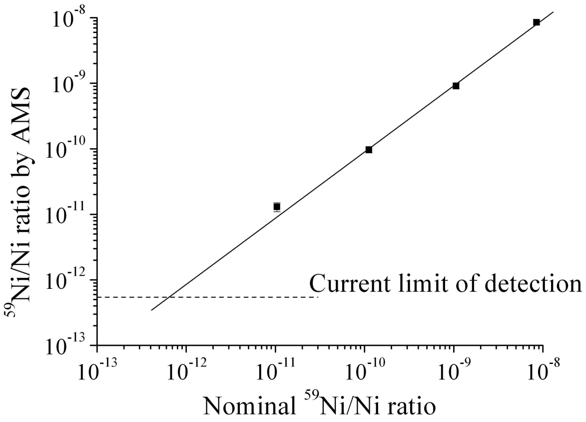

Figure 5 AMS measured ^{59}Ni/Ni ratio vs. nominal values for a series of laboratory reference samples and a blank sample. The dashed line shows the measured value of the blank sample.

Figure 5. The measured ratios are consistent with the nominal values within 12% with the exception of the sample with a ^{59}Ni/Ni ratio of $(1.10 \pm 0.09) \times 10^{-11}$. For this sample, the measured value is 25% higher than its nominal value. The discrepancy could be due to ^{59}Ni contamination introduced in the sample preparation, cross-talk in the ion source, or unexpected background from ^{59}Co in the detector. This will be checked later. A background level of about 5×10^{-13} for ^{59}Ni/Ni was obtained with the blank sample.

To summarize, a ^{59}Ni measurement method has been preliminarily developed at the AMS facility at CIAE with a current sensitivity of $^{59}\text{Ni/Ni} = 5 \times 10^{-13}$. More sophisticated chemical procedures for removing Co and higher ion energy for improving ion identification with gas detector will be explored in order to further improve the measurement sensitivity of ^{59}Ni.

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