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Study of the ^{60}Ni(n,2n)^{59}Ni reaction from threshold to 20 MeV and the half-life of ^{59}Ni

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Abstract. The production of long-lived ^{59}Ni as activation product is of concern for a fusion environment. Its main production is the (n,2n) reaction of 14-MeV neutrons on stable ^{60}Ni with some additional contributions from neutron capture reactions on ^{58}Ni via lower energy neutrons. Information for the production of ^{59}Ni via the (n,2n) reaction is strongly discordant or completely missing. Ni metal samples were available to us which had previously been irradiated with neutrons at energies of 17 and 19 MeV. After the irradiation the amount of produced ^{59}Ni was measured via accelerator mass spectrometry utilizing the 14-MV tandem accelerator of Maier-Leibnitz Laboratory in Munich. A second important issue is the half-life of ^{59}Ni: The accepted value is $t_{1/2}$ = (76 ± 5) kyr which is substantially lower than the value of (108 ± 13) kyr which was obtained a few years later in an independent measurement. The later value is based on relative measurements using the thermal ^{54}Fe neutron capture cross section. A new measurement of this cross section is underway at the VERA laboratory in Vienna applying AMS for the quantification of ^{55}Fe. The new data will allow to generate a more precise value for the half-life of ^{59}Ni compared to the previous one.

1 Introduction

The production of long-lived radionuclides as activation products is of concern for a fusion environment since they may lead to significant long-term waste disposal [1–4]. For such nuclides production cross-sections, total induced activities and the knowledge of their decay pattern are key parameters for safety and design analyses. One of those radionuclides is ^{59}Ni with a half-life of 76,000 years. Its main production is the (n,2n) reaction on stable ^{60}Ni with some additional contributions from neutron capture reactions on ^{58}Ni via lower energy neutrons. Experimental data for the production of ^{59}Ni via the (n,2n) reaction are strongly discordant or completely missing. Two previous measurements exist for a neutron energy of 14.8 MeV. However, their results disagree by a factor of four (refs. [5] and [6,7]). Such a discrepancy is clearly far from the required accuracy needed for activation calculations in fusion reactor design technology. No experimental data exist so far for neutron energies, both lower than and higher than 14.8 MeV. A recent evaluation of neutron induced cross sections on Ni isotopes [8] in the relevant neutron energy range from threshold to 20 MeV favours the higher value for the ^{59}Ni(n,2n)^{60}Ni reaction [6] and indicates a saturation of the excitation function between 17 and 19 MeV.

We have irradiated several Ni metal samples with neutrons with an energy of 17 and 19 MeV. After the irradiation the amount of produced ^{59}Ni nuclei in the samples has been measured via accelerator mass spectrometry (AMS) utilizing the GAMS setup of the Maier-Leibnitz Laboratory (MLL) in Munich [9]. AMS represents a very sensitive method to study isotopic abundances of radionuclides at minute levels for some specific elements. Utilizing AMS, the content of these rare isotopes is counted directly by sputtering the sample material and analyzing the particles of interest, which allows the identification of the nuclide of interest with a particle detector.

A second important issue is the half-life of ^{59}Ni: The accepted value is $t_{1/2}$ = (76 ± 5) kyr and is substantially lower than the value of 108 kyr which was obtained a few years later in an independent measurement [7]. The later value is based on relative measurements using the thermal neutron capture cross sections of ^{58}Ni and ^{54}Fe. A new measurement of the ^{54}Fe(n,γ) cross section is underway at the VERA (Vienna Environmental Research Accelerator) laboratory applying AMS for the quantification of ^{55}Fe. Together with the significantly improved knowledge on the thermal ^{58}Ni(n,γ) cross section, the expected new result for the ^{54}Fe(n,γ)^{55}Fe reaction will allow to generate a more precise value for the half-life of ^{59}Ni compared to that reported in [7].

2 Neutron irradiation

The neutron irradiations were performed at the University of Tübingen using a single-ended 3-MV van-de-Graaff accelerator. 17 and 19 MeV neutrons were generated via the (d,t) reaction when bombarding the Ti-T target with 1.2 and 2.6 MeV deuterons, respectively. Actually, these Ni samples served in a previous experiment as fluence monitors, when irradiating Al samples for the measurement of the ^{27}Al(n,2n)^{26}Al cross sections at 17 and 19 MeV [3,10]. The width of the neutron energy distributions were calculated from the energy loss of the deuterium in the Ti-T target, and from kinematics from all possible emission angles of the produced neutrons.

Natural nickel samples with a thickness of 0.125 mm and a diameter of 10 mm had been irradiated. The distance between the neutron-producing target and the Ni foil was 14 mm

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Table 1. Main parameters for the neutron irradiation of the Ni samples used for the ^{60}Ni(n,2n)^{59}Ni measurement (see also [4]). For the fluence determination the total production of ^{57}Co was used. Cross section data for this reaction were taken from Tagesen et al. [8].

	Ni-1	Ni-4
Neutron energy (MeV)	17.0 ± 0.23	19.0 ± 0.27
Neutron fluence (10^{12}n cm^{-2})	1.48 ± 0.07	1.38 ± 0.06
^{58}Ni $\rightarrow$ ^{57}Co (mbarn)[1]	864 ± 35	904 ± 33

irradiated, and the samples were positioned at 0° relative to the incoming d^{+}-beam. The range of neutron energies over the full sample area was about 250 keV (FWHM) (see table 1). For the fluence determination the ^{58}Ni(n,np+pn+d)^{57}Co reaction was chosen and was measured directly via the induced activity in the samples. ^{57}Co is also produced through the ^{58}Ni(n,2n)^{57}Ni reaction with consecutive decay of ^{57}Ni. This contribution was taken into account as well. The original sample arrangement was a stack of foils in the succession of Ni-Al-Ni-Al-Ni. The time history of the relative flux was monitored with a scintillation counter positioned under 108° relative to the incoming d^{+}beam in a distance of about 3 m. Pulse shape discrimination was used for the neutron-γ discrimination. For the determination of the activity of the irradiated Ni samples the 122 keV γ-line was measured with a HP-Ge-diode. Self absorption of photons in the Ni foil was negligible.

For the 17-MeV datapoint an irradiation time of four days was necessary to produce a neutron fluence of 1.48×10^{12} n · cm^{-2}. About 8 days of irradiation were necessary to obtain a fluence of 1.38×10^{12} n.cm^{-2} for the 19-MeV datapoint. The irradiation geometry was chosen in such a way, that the production of ^{59}Ni via (n,γ) on ^{58}Ni from thermalized neutrons was negligible (see ref. [11] for a more detailed estimation of the production of low-energy neutrons for typical irradiation geometries).

3 AMS measurements

3.1 AMS setup in Munich

^{59}Ni is a radionuclide with a half-life too-long to be suitable for an off-line decay counting. Therefore, we have measured the number of produced ^{59}Ni nuclei using the technique of AMS at the Maier-Leibnitz-Laboratory in Munich which is based on a 14 MV tandem accelerator [9]. The combination of a large accelerator like the MP tandem available at the Munich laboratory – hence providing high particle energies – and a dedicated particle detection system featuring a time-of-flight system, a gas-filled magnet and a multi-anode ionization chamber fulfills the requirement of a sensitive ^{59}Ni detection.

Ni^{-}-ions are extracted from a Cs sputter source. They pass on the low-energy side a 90° analyzing magnet and a 18° electrostatic deflection unit before entering the MP tandem with typical energies of 184 keV. A terminal voltage between 12 and 13 MeV was chosen for our AMS measurements. The positively charged ions pass a Wien filter providing an isotope selectivity and in case of Ni, with a double focusing 90° analyzing magnet the 12^{+} charge state was selected. The time-of-flight system consists of thin foils plus two micro channel-plates with a flight path of 3.5 m. Depending on the charge state selected, ions with typical energies of 150 to 200 MeV enter then a gas-filled magnet. Within the magnet chamber a spatial separation of isobars with different atomic number Z is achieved. This effect is the result of the Z dependence of an ion's mean charge state after multiple collisions with the gas atoms in the magnet, which leads to a different radius in the magnet. The interfering isobars can be blocked from entering the final particle detector [12]. Therewith, the particle count rate for the isobar registered with the detector can be lowered to a feasible level. In our case a position sensitive multi-delta-E ionization chamber was used for extracting the differential energy loss information in the segmented anodes (see ref. [9]).

With AMS the basic results are atom ratios of the radioisotope relative to a stable isotope by measuring the number of radionuclides relative to the current (measured with a Faraday cup in front of the detector system) of a stable isotope. Both, the count rate of the radionuclide and currents of the stable ions are measured sequentially by adjusting ion optical elements and the terminal voltage accordingly to the different masses. The isotope ratios of unknown samples are then compared with the ratios obtained from standard sample of known ^{59}Ni concentration and from non-irradiated background samples. Depending on the concentration of isobaric ^{59}Co in the sputter material, a detection limit in the range of a few times 10^{-14} for the isotope ratio ^{59}Ni/Ni was found applying this AMS setup [13]. The typical reproducibility of the AMS measurements in Munich is of the order of 5 to 10% [3].

3.2 ^{59}Ni AMS measurements

Ni^{-} ions were produced in the ion source with typical currents of the order of μA. The tandem was operated at a terminal voltage of 12.58 MV and the charge state 12^{+} was selected. Background signals in the spectra due to tails of the stable isobar ^{59}Co were sufficiently low. Nonetheless, in order to reduce further the natural Co concentration in the nickel samples, these foils were chemically treated prior to the AMS measurement: the irradiated foils were dissolved in 10N HCl and purified by anion exchange chromatography with DOWEX AG1 resin. In a subsequent step ammonia was added until pH 9 and nickel was precipitated with dimetylglyoxim. This complex was centrifuged and washed with pure water, then ashed to nickel(II)oxid which served as sputter material. Due to this procedure the count rate of the isobar ^{59}Co in the particle detector can be reduced significantly.

Four independent AMS measurements have been performed in order to check possible systematic uncertainties, also associated with such close-to-background experiments. Standard samples served as reference material for the final isotope ratios, they were produced via the irradiation of natural nickel powder with thermal neutrons. Its isotope ratio was calculated from activity measurements relative using the thermal neutron cross section value of ^{64}Ni ($\sigma_{\gamma} = 1.52 \pm 0.03$ b, see also [13]) and taking into account the recently remeasured thermal cross-section value of ^{58}Ni ($\sigma_{\gamma} = 4.13 \pm 0.05$ b

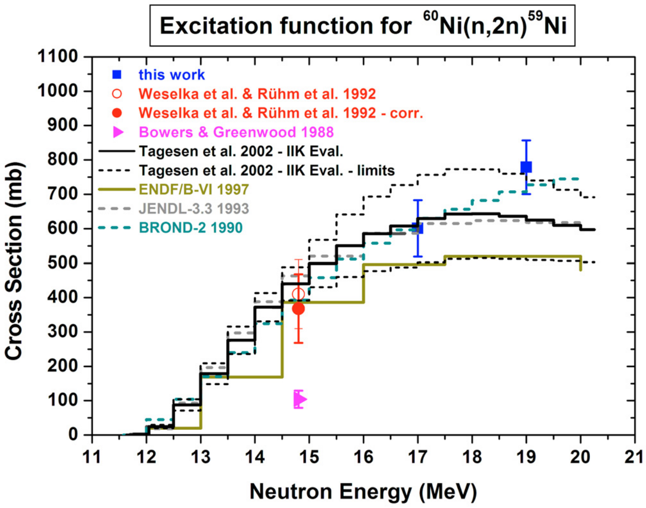

Fig. 1. Comparison of the new AMS data at 17 and 19 MeV (squares) with published data. The symbols indicate the experimental data, lines depict evaluations of the ^{60}Ni(n,2n) reaction.

[14]). The ^{59}Ni/Ni isotope ratio of that standard material was determined to (9.1 ± 0.4) × 10^{-11}.

4 Results for the ^{60}Ni(n, 2n)^{59}Ni cross section at 17 and 19 MeV

Figure 1 shows the new results for the ^{60}Ni(n,2n)^{59}Ni cross section in comparison with published data. The two experimental datapoints at 14.8 MeV are plotted as triangle (Bowers and Greenwood [5]) and open circle (Weselka et al. and Rühm et al. [6,7]). The filled circle is a renormalized value of [7], taking into account the new data for the thermal cross-section value of the ^{58}Ni(n,γ)^{59}Ni reaction [14], which served in the original work of Rühm et al. as a reference reaction. Also plotted are in figure 1 evaluations for the energy range from threshold (E_{th} = 11.58 MeV) to 20 MeV. The latest evaluation performed by Tagesen et al. [8] (noted as IIK evaluation) gives an uncertainty band. It indicates a largely constant cross-section value of about 600 mbarn in the neutron energy range from approx. 16 MeV to 20 MeV with an uncertainty of 100 to 150 mbarn.

JENDL-3.3 is very similar to the IIK evaluation, while the BROND-2 evaluation shows no tendency of saturation in the shape of the excitation function. ENDF/B-VI shows a similar trend compared to the IIK evaluation, however, lies significantly lower, 100 to 150 mbarn compared to the other evaluations. In general, all evaluations strongly support the experimental value of [6,7]. From those results the lower experimental value seems to be ruled out.

Our new data (see table 2) fit well to the evaluations and support the experimental value at 14.8 MeV as well. We are within the limits given by the IIK evaluation, At 17 MeV neutron energy the new experimental value is exactly as found for the IIK evaluation. At 19 MeV the experimental value is at the upper limit (1σ) of the IIK evaluation and, in addition, it suggests a still increasing excitation function within the energy range between 17 and 19 MeV.

Table 2. Two Ni samples were measured with AMS (Ni-1 and Ni4) using the GAMS setup in Munich. The new cross section data are given in the second line.

	Ni-1	Ni-4
^{59}Ni/^{60}Ni ratio (10^{-12} at/at)	0.89 ± 0.11	1.08 ± 0.10
^{60}Ni(n,2n)^{59}Ni (mbarn)	601 ± 82	779 ± 78

The uncertainty of the new experimental data is 14 and 10%, respectively, and is largely dominated by the AMS isotope ratio measurement. The rather low ^{59}Ni/Ni isotope ratios of about 2×10^{-13} (i.e., about 1×10^{-12} for the ^{59}Ni/^{60}Ni isotope ratio) measured for the irradiated Ni samples are approx. a factor of 10 higher than the isotope ratio found for non-irradiated Ni blank samples. It is planned to perform additional neutron irradiations with samples enriched in ^{60}Ni over the whole energy range from threshold to 20 MeV. Enriched samples will result in an increased number of produced ^{59}Ni nuclei and being not sensitive to the production of ^{59}Ni via thermal neutron capture on ^{58}Ni.

5 Half-life of ^{59}Ni

The recommended value for the half-life of ^{59}Ni is $t_{1/2}$ = (76 ± 5) kyr [15]. Triggered by the unexpected low cross-section value for the ^{60}Ni(n,2n) reaction found by [5], also the half-life of ^{59}Ni was under debate since the value for the cross section at 14.8 MeV [5] was obtained via an activity measurement on an highly irradiated Ni sample. However, that low (n,2n) value was not confirmed by the experiment of [6,7], rather it confirmed the expectations from model calculations (see section 4). As a by-product of [7], a half-life measurement of ^{59}Ni was performed. The value measured, $t_{1/2}$ = (108 ± 13) kyr, was found to be substantially higher than the recommended value [15]. Using the newest available data for cross sections and K_α yield, this value [7] can be corrected to (97 ± 9) kyr, however, which is still substantially higher than the recommended value [13]. This higher value is based on relative measurements using the thermal ^{54}Fe neutron capture cross section producing ^{55}Fe, while Nishiizumi et al. [15] measured the half-life independent of neutron capture cross sections.

Recently, we could demonstrate that the AMS setup at the VERA laboratory allows precise measurements of ^{55}Fe [16]. Both, the relatively long half-life of ^{55}Fe ($t_{1/2}$ = 2.75 years) and the beta-decay mode of ^{55}Fe, which proceeds without γ-ray emission, makes AMS the favourable measurement technique. We have recently irradiated several Fe samples with thermal neutrons produced at the TRIGA Mark II reactor of the Atominstitut in Vienna. In addition, ^{55}Fe reference samples have been produced in Munich via the ^{58}Ni(p,α)^{55}Co reaction. ^{55}Co decays to ^{55}Fe with a half-life of 17.54 h. Combining the well-known half-life of ^{55}Co and the precisely measured activity of ^{55}Co allows to calculate the amount of the decay daughter ^{55}Fe with high precision. A similar experiment is

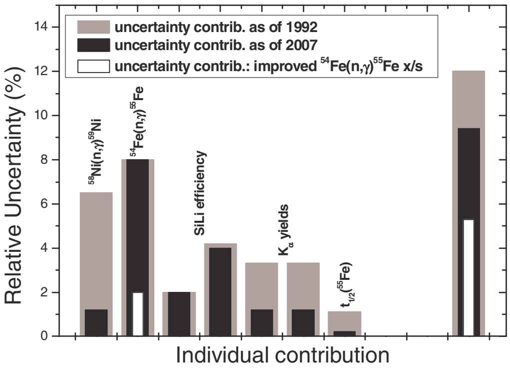

Fig. 2. Different contributions to the total uncertainty for the half-life measurement of ^{59}Ni [7]. The grey bars show the original values as of 1992. The black bars give the relative values with improved knowledge as of 2007. The white bars is the expected uncertainty contribution from the new measurement of the thermal ^{54}Fe(n,γ)^{55}Fe cross section. The total uncertainty is plotted as the bar on the right side.

being performed at the VERA laboratory irradiating enriched ^{54}Fe with protons; therewith ^{55}Co is produced as well. In both cases a direct production of ^{55}Fe can be excluded. The irradiated samples will be used as reference samples for the AMS measurement and relative to those, the ^{54}Fe(n,γ)^{55}Fe thermal neutron cross-section value is now being determined at the VERA laboratory. The new data are expected to be precise at a level of about 2%.

A new value of the ^{58}Ni(n,γ)^{59}Ni thermal cross section was published recently [14] with $\sigma_{n,\gamma} = 4.13 \pm 0.05$ barn, significantly lower and much more precise than the previously adopted value of 4.6 barn. In combination with the improved knowledge on the thermal neutron cross-section value for the ^{58}Ni(n,γ)^{59}Ni reaction, the uncertainty of the half-life value published by [7] should be reduced significantly. The various contributions to the uncertainty of the ^{59}Ni half-life are plotted in figure 2. It is expected that the new data, will allow to generate a more precise value for the half-life of ^{59}Ni compared to the previous one. And, this will allow to check if there still exists a discrepancy to the adopted value for the ^{59}Ni half-life.

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