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Neutron-induced inelastic γ-production cross sections on ^{58,60,64}Ni

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Abstract. This paper reports partial results of a (n, n'γ) measurement on nickel. The inelastic channel was measured using the Gamma Array for Inelastic Neutron Scattering (GAINS) spectrometer at the 100-m measurement cabin of the Geel Electron Linear Accelerator (GELINA) neutron source of the European Commission's Joint Research Centre (EC-JRC) in Geel, Belgium. Using γ spectroscopy, we were able to extract angle-integrated production cross sections for several γ rays but we report here only the results for the main transition in ^{58}Ni. We discuss however in detail the observed discrepancy between our data and other experiments (especially the work of Voss *et al.*). We also shortly comment on the quality of the neutron-target optical model potential in describing the inelastic data in this mass region. The calculations were performed using the TALYS 1.9 code in the default settings.

1 Introduction

The energy need of our society can not be overstated and it will very likely increase in the coming decades. Such needs must be mitigated in respect to the many consequences of the energy production sector, with pollution and global warming being the most acute. If one wishes a sustainable development for future generations, finding new, far less polluting, ways for producing energy is paramount for the present century. In this context, nuclear energy seems to be a feasible pillar on the medium to long term if the current nuclear technology is improved in terms of safety, nuclear waste, fuel availability, etc.

Generation IV nuclear reactors should solve many of these issues as they will be able to transmute the minor actinides from the fuel, thus shortening the time nuclear waste remains highly radioactive, while being fuelled by more abundant isotopes (for example ^{238}U or ^{232}Th, see Ref. [1] and the references therein). Another advantage of ^{238}U is given by the fact that the fission cross section on this isotope starts to become relevant only above 1-1.5 MeV incident neutron energy [2] which means that no neutron moderation is needed. Unfortunately, sensitivity studies for Generation IV nuclear facilities show that, for an optimal design, neutron inelastic data with very low uncertainty (typically below 5%) on, in particular, nickel isotopes is required [3]. Providing realiable experimental data with such low uncertainty is very challenging.

Nickel is one of the components of heat- and corrosion-resistant Fe-Cr-Ni steel, a frequently used structural material in all nuclear facilities. It has five stable isotopes: ^{58}Ni is the most abundant [68.077(6)%] followed by ^{60}Ni [26.223(5)%], ^{62}Ni [3.634(1)%], ^{61}Ni [1.139(4)%] and ^{64}Ni [0.926(6)%] [4]. This paper presents the results extracted for the main transition in ^{58}Ni (E_γ=1454.3 keV) while the data for the other transitions is reported in Ref. [5]. The uncertainty of our data for this transition ranges from around 3%, where the cross sections reaches its maximum value, up to 11% at very high incident energies. We also discuss in detail the possible reasons for the observed discrepancy between our data and the experiment of Voss *et al.* [6] (see Section 4).

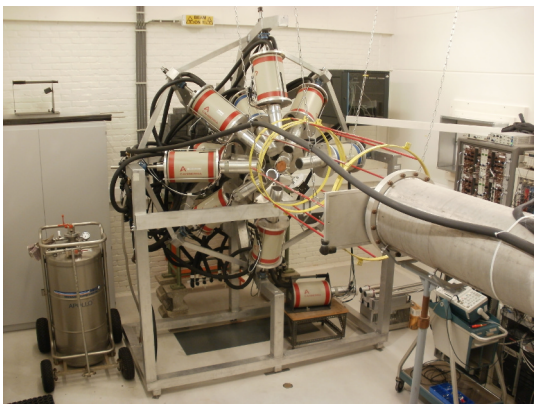

Figure 1. (Colour online) The GAINS spectrometer used during our (n, n'γ) measurement on nickel. It is currently operating at 100 m from the neutron source and consists of 12 large volume HPGe detectors placed at 110°, 125° and 150° relative to the incident neutron beam.

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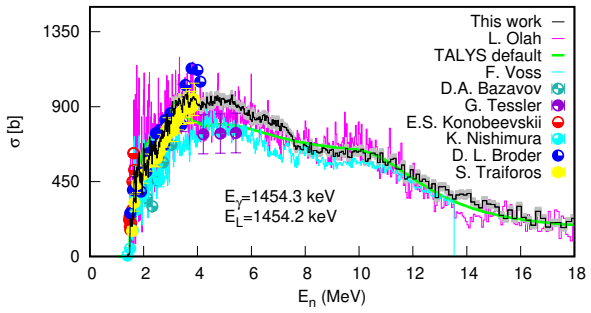

Figure 2. (Colour online) The inelastic angle-integrated γ-production cross section for the E_γ=1454.2-keV transition de-exciting the E_L=1454.2-keV level in ^{58}Ni measured in the present work. It is compared with other available experimental data (the associated referencing is given in the main text) and with TALYS 1.9 model calculations performed using the default settings of the code. The grey band represents the total uncertainty of our data. For readability reasons, we do not display here also the uncertainties associated to the Voss *et al.* and Olah *et al.* data.

2 Experimental setup

The (n, n'γ) measurement on nickel was performed using the GAINS spectrometer (see Fig. 1) and a neutron beam provided by the GELINA facility of the EC-JRC. In this type of experiments we employ γ spectroscopy and neutron time of flight techniques to determine the γ and neutron energies, respectively. The 100-m neutron flight path allows us to measure high resolution data and, due to the high efficiency of the large volume HPGe detectors, we can provide very low uncertainty cross section points. The detectors are read out by ACQIRIS DC440 digitizers with a sampling frequency of 420 MHz and an amplitude range of 12 bits. For data normalization we employed a fission chamber with ^{235}U deposits [7]. During the experiment, a very thick 2.661(21) mg/cm^2 natural nickel target with a diameter of 8.23(3) cm was irradiated for a total of around 15 days (after data reduction). Other experimental details can be found in Refs. [8–11].

3 Data analysis procedure

We refer the interested reader to Refs. [10, 12–15] for details regarding the data analysis procedure of our experiments at GAINS. Here we only mention that the primary extracted quantity is the differential cross section (that is, the γ-production cross section at those angles were we placed the detectors: 110° and 150°). For cross-checks we also extract the cross section at 125°. The 110° and 150° measurement angles are the nodes of the fourth order Legendre polynomials. This allows us to angle integrate the differential cross sections by making use of the Gaussian quadrature method [12, 13].

4 Results and discussion

The first and second excited levels in ^{58}Ni - part of the ground state rotational band - decay through relatively intense transitions, of 1454.2 and 1004.8 keV, which therefore collect a significant part of the inelastic strength. Detecting these two γ lines with very good statistics was very important during the experiment. We report here however only the cross section for the main transition in ^{58}Ni (displayed in Fig. 2 together with other previously published results). Among the previously measured data at other facilities, Voss *et al.* [6] reported the most extended data set with good incident energy resolution up to around 14 MeV. At a first glance, our data and those of Voss *et al.* agree very well in terms of shape but with a 20-30% difference in absolute values. One can notice that this difference actually becomes smaller at very high energies pointing to a combination of energy- and non-energy-dependent factors that might explain the discrepancy (possible candidates are discussed below). This discrepancy is consistently present for the other ^{58}Ni and even ^{60}Ni transitions (see Ref. [5]). The other values plotted in Fig. 2 are in agreement with our experimental data even though Bazavov *et al.* [16], Konobeevskii *et al.* [17], Nishimura *et al.* [18], Broder *et al.* [19] and Traiforos *et al.* [20] only reported cross section points close to the threshold region, below 4 MeV. The same figure also displays one more extended data set, measured at GELINA with very high resolution (200 m flight path) [21], which agrees well with our data.

It is important to note that a 20-40% difference between GELINA data and those of Voss *et al.* was also observed in three other experiments that reported inelastic cross sections on ^{52}Cr [22], ^{238}U [23] and ^{56}Fe [24] (see Fig. 3). The Mihailescu *et al.* and Negret *et al.* experiments were performed at GELINA using the same fission chamber, a similar HPGe-based setup and data analysis procedure to the ones employed in the present work while Kerveno *et al.* made use of a different setup and fission chamber. Interestingly, the only other data set which agrees very well with Voss *et al.* is the one of Tessler *et al.* [25] even though the authors reported only 3 cross section points with relatively high uncertainties (see Fig. 2).

To understand these discrepancies, we list the most relevant characteristics of the Voss *et al.* and Tessler *et al.* experiments [6, 25]: a) relatively large ^{nat}Ni targets were used: ring-shaped with an inner diameter of 120 mm, outer diameter of 254 mm, and a 9.4 mm thickness (Voss *et al.*), and a cylinder with a diameter of 25.4 mm and a hight of 25.4 mm (Tessler *et al.*), b) the inelastic γ rays were detected using a single Ge(Li) detector placed at 125° or 55° (therefore, angle-integrated data was extracted by multiplying the differential cross sections with 4π), c) the neutron flux monitoring was performed using a calibrated proton recoil detector (scintillation counter), d) missing corrections for neutron multiple scattering, neutron beam attenuation and γ self-attenuation inside the target (except for the Tessler *et al.* data which was corrected for the latter effect).

Our data are normalized to the ^{235}U(n,f) standard cross section which is a different procedure than that employed by Refs. [6, 25]. The complexity of the flux measurement in the Tessler *et al.* and Voss *et al.* experiments could certainly generate part of the difference in absolute values seen in Fig. 2. Other important factors are given by the

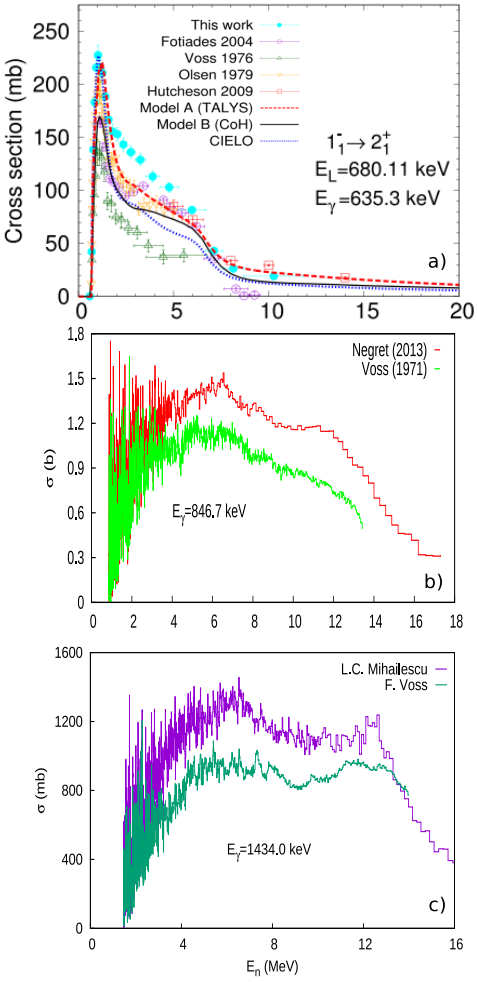

Figure 3. (Colour online) The neutron inelastic γ-production cross section of the 635.3-, 846.7- and 1434.0-keV transitions in ^{238}U (panel a), ^{56}Fe (panel b) and ^{52}Cr (panel c) measured by Kerveno *et al.* [23], Negret *et al.* [24], Voss *et al.* [6] and Mihailescu *et al.* [22]. The figure in panel a) was taken from Ref. [23] while the remaining data sets displayed here were taken from EXFOR [32].

mentioned missing corrections (see above). Ref. [6] states that the multiple scattering versus beam attenuation corrections (both around 10-20%) cancel each other while the γ self-attenuation correction should increase the reported cross sections by 20-25%.

To check - as a rough estimate - if this is indeed the case, we calculated the γ-ray attenuation in nickel and got a 21-28% absorption (for E_γ = 1.5 MeV and an *effective* target thickness of 5-7 mm). A 21-28% increase of the Voss *et al.* cross section values shown in Fig. 2 due to γ self-attenuation corrections would greatly improve the agreement with our data.

Figure 2 also compares our data with theoretical calculations provided by the TALYS 1.9-default code [26]. The theoretical curve is able to describe well our cross section values, except for the underestimation in the 3-8 MeV region. The same disagreement can also be observed for the secondary transitions in ^{58}Ni, and even for ^{60}Ni (see Ref. [5]). This indicates, among others, the rather poor quality of the neutron-target optical model potential in this mass region. Other causes for this disagreement relate to other reaction ingredients that TALYS 1.9 calls during the calculation: nuclear structure information, modeling of the pre-equilibrium emission, the role played by the deformation within direct/coupled channels calculations, etc.

The authors of Ref. [27] argue the impact of nuclear structure information imported from RIPL [28] by reaction codes, showcasing the relevant effects of the branching ratio inaccuracies on the predicted cross sections (we calculate level and total inelastic cross sections making use of available level scheme information on the target nucleus). Therefore, such inaccuracies or missing structure information (leading the reaction codes to make unphysical assumptions regarding decay paths and branching ratios) generate both a larger total uncertainty and a poor theoretical description of the experimental cross sections. In Ref. [23] Kerveno *et al.* studied the impact of the pre-equilibrium emission on the neutron inelastic cross sections. The authors showed for ^{238}U that the default value for the spin cut-off parameter employed by the exciton model yields poor agreement with experimental data. After fixing the value of this parameter using more microscopical approaches for the pre-equilibrium reactions, the resulting inelastic cross sections displayed a considerable improvement across transitions in describing experimental data. Similarly, the authors of Refs. [29, 30] showed that a proper description of energy-averaged compound nucleus cross sections in the presence of a direct component (strongly coupled reaction channels) using the Engelbrecht-Weidenmüller (EW) transformation [31] leads both to an increased inelastic cross section - helpful in the ^{58}Ni case - and also to a lowering of the elastic channel enhancement typically induced by the width fluctuation corrections (which are relevant out of the Ericson regime, in the low energy region, where isolated or weakly overlapping CN resonances occur).

These studies were indeed mainly dedicated to actinides [23, 27, 29, 30]. However, the well-known collectivity of the ground state rotational band in ^{58}Ni, requiring coupled channels calculations for a proper description, likely makes such effects relevant in this case as well. A more in-depth study of all these issues impacting the modeling of the inelastic channel our group measures will be left however for future work.

5 Conclusions

Using the GAINS spectrometer at the GELINA neutron source, we measured the neutron inelastic channel on several nickel isotopes: ^{58}Ni, ^{60}Ni and ^{64}Ni. We reported here however high resolution, low uncertainty data only for the main transition in ^{58}Ni with E_γ=1454.3 keV. The uncertainty of our data for this transition ranges from around 3%, where the cross sections reaches its maximum value, to 11% at very high incident energies. Our results compare very well with previous experiments except for the data of Voss *et al.* [6] and Tessler *et al.* [25] which display a similar shape but different absolute values. This is also the case for three other nuclei: ^{52}Cr [22], ^{238}U [23] and ^{56}Fe [24].

We discuss in detail these discrepancies by comparing the experimental techniques and data analysis procedures in all the mentioned experiments. We conclude that, most likely, the cause for the discrepant values is given by the neutron flux normalisation (proton recoil detector versus fission chamber) and the lack of γ self-attenuation corrections in the Voss *et al.* experiment. We also comment on the rather poor quality of the default neutron-target optical model potential implemented in TALYS 1.9 for ^{58}Ni and ^{60}Ni, and on possible ways to improve these theoretical predictions.

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