The evaluation of experimental data in fast range for n + $^{56}$Fe(n, inl)

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Abstract. Iron is one of the five materials selected for evaluation within the pilot international evaluation project CIELO. Analysis of experimental data for n + $^{56}$Fe reaction is the basis for constraining theoretical calculations and eventual creation of the evaluated file. The detail analysis was performed for inelastic cross sections of neutron induced reactions with $^{56}$Fe in the fast range up to 20 MeV where there are significant differences among the main evaluated libraries, mainly caused by the different inelastic scattering cross section measurements. Gamma-ray production cross sections provide a way to gain experimental information about the inelastic cross section. Large discrepancies between experimental data for the 847-keV gamma ray produced in the $^{56}$Fe(n, n'γ) reaction were analyzed. In addition, experimental data for elastic scattering cross section between 9.41 ∼ 11 MeV were used to deduce the inelastic cross section from the unitarity constrain.

1. Introduction

The Collaborative International Evaluated Library Organization (CIELO) [1], a pilot project (Subgroup 40) of the Working Party on International Nuclear Data Evaluation Co-operation (WPEC-SG40) under auspices of OECD’s Nuclear Energy Agency, focuses on a small number of the highest-priority isotopes ($^1$H, $^{16}$O, $^{56}$Fe, $^{235,238}$U and $^{239}$Pu) for reactor application. This paper describes efforts to resolve the discrepancies between the measurements and provide recommendation for important cross sections of $^{56}$Fe in the fast neutron energy range to serve as a base for theoretical calculations.

There is an amount of experimental data published for inelastic reactions which is the major part of nonelastic cross section for $^{56}$Fe and $^{nat}$Fe. But there are also large discrepancies among them even with the same measured method. These discrepancies also exist in the nuclear evaluated data libraries, such as ENDF/B-VII.1, JEFF-3.1, JENDL-4.0, CENDL-3.1 and ROSFOND. A recent measurement of neutron transmission through iron spheres, for quasi-monoenergetic neutrons ranging from about 6 to 11 MeV, suggested decreases of $^{56}$Fe(n, inl) cross section by 21%, 29%, and 35% at 6.2, 8.2, and 10.8 MeV respectively. Such large decrease of the inelastic cross section warrants an in-depth study of the existing experimental datasets. This is a subject of the present paper. The salient points of this work are summarized below:

- The experimental data were taken mainly from the EXFOR [2] library. Any information absent in the EXFOR Data File was taken from the original publications if available. The experimental data were also collected from the recent new measurements which were mostly obtained directly from the authors.
- We also used measurements on natural iron for the reactions for which either the difference between $^{56}$Fe and $^{nat}$Fe is known to be small or for which an accurate conversion from $^{nat}$Fe to $^{56}$Fe is possible using the cross sections for the minor isotopes.
- All experimental data were critically reviewed and corrected according to the newly recommended standard cross sections for monitor reactions and recommended decay data, if possible.
- Measurements of γ-ray production cross section for the transition from the first 2+ level (E = 847 keV) were also included because of the high-resolution experiments and accurate conversion procedures.
- The unitarity constrain has been used for the reactions and energy regions where reliable measurements exist for the competing reactions.

2. The analysis of experiment data

Inelastic scattering cross section on $^{56}$Fe is the major part of the absorption cross section in the energy range relevant to nuclear engineering. The uncertainty assessment performed by WPEC Subgroup 26 for innovative reactor systems shows that the knowledge of the inelastic scattering cross section of $^{56}$Fe should be improved to meet the target accuracy requirements. New measurements and evaluations have been requested in the range 0.5 ∼ 20 MeV to reduce the uncertainty down to 2 ∼ 10% depending on the energy and the reactor considered [3].

In addition to the scarce of experimental data around 14 MeV, there is no data available that could be used for constraining theoretical calculations in the fast energy...
region. In addition, the inelastic cross sections of different evaluated libraries do not agree with each other. This is a challenge for creation of the evaluated file.

Use of the γ-ray cross section to infer the inelastic was applied as early as 1956 by Day [4]. Due to the specific nuclear structure properties of the $^{56}\text{Fe}$ nucleus observed in many experiments with γ-rays transition measurements, the production of the 847-keV γ-ray amounts to 0.93 ± 1.00 of the total inelastic cross section with an energy dependence that can be deduced from known decay schemes and experimental data of partial γ-ray production. In 1992, the energy dependence of the coefficient

$$ R \equiv \frac{\sigma_{\text{total inelastic}}}{\sigma_{847 \text{ keV}}} \quad (1) $$

was given by Vonach [5] relying on the γ-transition scheme and the γ-yield measurements [6-8]. Where $\sigma_{847\text{keV}}$ means 847-keV γ-line production cross section and $\sigma_{\text{total inelastic}}$ means the total inelastic cross section. This R value was utilized in this work to convert the high accurate measurements of 847-keV γ-line production cross section to the total inelastic cross section.

More than 40 measurements for the (n,n'γ) cross section at various incident neutron energies have been performed since 1970's but the consistency between these measurements is generally poor. Below about 2 MeV, Perey et al. [9] performed a measurement with the white neutron source at Oak Ridge Electron Linear Accelerator (ORELA) facility by using a hydrogen-free carbon fluoride liquid scintillator to detect the γ rays in a 4π geometry. In 1976, Smith et al. [10] measured the excitation function of the 847-keV γ-ray and its angular distributions at the Argonne National Laboratory Fast Neutron Generator (the neutron energy range was 0.93–2.03 MeV). In 1990’s, the neutron energy range was extended. Dickens et al. [11] measured at ORELA in 1991 γ-rays for incident neutron energies between 0.8 and 41 MeV by using a HPGe detector at 125 degrees. In 2004, more accurate gamma-ray cross-sections was obtained by Nelson et al. [12] with the Germanium Array for Neutron-Induced Excitations (GANEI) at Weapons Neutron Research Facility (WNR) at LANSCHE [13]. Spallation neutrons from the target travel a well-collimated 20.34-m flight path to the GANEI sample. In addition to absolute cross-section measurements performed from 4 MeV to 20 MeV in 1994 (most at 125 degrees for natFe), measurements relative to the $^{54}\text{Cr}$ (n,γ) 1434-keV γ-ray were made to reduce corrections for neutron multiple scattering as well as corrections for γ-ray attenuation in the sample at 14.5 MeV. The contributions of $^{57}\text{Fe}(n,2n\gamma)$ reaction was subtracted in this results and data for $^{56}\text{Fe}(n,n'\gamma)$ were obtained by correcting natural iron results for isotopic abundance of $^{56}\text{Fe}$ (91.8%). The results from absolute and relative measurement agree well with each other and the accuracy is typically 5 ± 10%. In addition, 34 experimental results were compared in this work and it has been deduced that some of the previous measurements were wrong.

A new measurement was performed with the white neutron source at Geel Linear Accelerator (GELINA) of IRMM by Negret et al. [14] in 2014. The Gamma Array for Inelastic Neutron Scattering (GAINS) spectrometer was used. Eight high-efficiency HPGe detectors were placed at 110° and 150° with respect to the incoming beam and the distance between detectors and sample was 119 mm and 154 mm respectively. Multiple checks were performed to ensure consistency of the results for gamma production cross sections and total inelastic cross sections.

Another new measurements were performed by Beyer et al. in 2014 [15] for $^{56}\text{Fe}(n,\gamma')$ cross section in the energy range from 0.8 to 9.6 MeV by using the photoneutron source nELBE at the superconducting electron accelerator ELBE of the Helmholtz-Zentrum Dresden-Rossendorf (HZDR) in Germany. The γ-ray spectra were measured with a high-purity germanium detector which was positioned at a distance of 20 cm from the target and at an angle of 125° relative to the beam direction.

In this work, the measurements for 847-keV γ-ray production cross section were renormalized to the standard cross sections, the isotopic abundance, and converted into inelastic cross section according applying the R factor as mentioned before. Figure 1(a) displays the re-normalized inelastic cross section comparison of Perey and Dickens of ORNL, Smith of ANL, Beyer of HZDR and Negret of IRMM below 2 MeV. The data of Perey and Negret agree well with each other and the result of Negret has higher resolution than the previous measurements. Although it is difficult to directly compare various experiments at low neutron energies when the resolution is very different, Negret’s result is in good agreement with the recent experiment from Beyer after a proper re-binning. In the energy range of 6 ~ 11 MeV, however, it can be seen from Fig. 1(b) that four sets of data are largely discrepant. The data given by Dickens are lower by 10% than Negret’s data and lower by 20% than Nelson’s result. In Beyer’s results, one datum at ~8 MeV is consistent with Negret’s data but another two values at ~11 MeV agree well with those of Nelson. It is difficult to resolve the discrepancy as it results from the complex experimental process and different experimental facilities. In our past similar measurements, it was found that the cascade decay seriously affects the detection efficiency when the distance between sample and detector becomes short. The cascade decay effect may cause underestimation of the results. A measurement using a $^{137}\text{Cs}$ source as a reference to check and find the efficiency correction factor (due to cascade decay) as a function of distance between sample and detector could help to solve this issue.

3. The analysis of the unitarity constrain among all reactions

Further studies based on the unitarity constrain among all reactions were used to check the inelastic cross section in the energy region of 9.41~11 MeV. In the neutron energy range below 20 MeV, total cross section can be obtained by

$$ \sigma_{(a,\text{tot})} = \sigma_{(a,el)} + \sigma_{(a,\gamma)} + \sigma_{(a,\text{int})} + \sigma_{(a,p)} + \sigma_{(a,\alpha)} + \sigma_{(a,\text{2n})} + \sigma_{(a,\text{np})} \quad (2) $$

As the reaction-thresholds of $^{56}\text{Fe}(n,2n)$ and $^{56}\text{Fe}(n,\text{np})$ are higher than 11 MeV and the capture cross section is less than a few mb and can be neglected entirely between 9.41~11 MeV, the inelastic scattering cross section was derived as:

$$ \sigma_{(a,\text{int})} = \sigma_{(a,\text{tot})} - \sigma_{(a,el)} - \sigma_{(a,p)} - \sigma_{(a,\alpha)} \quad (3) $$
Actually, the total cross section is in general known with an accuracy of about 1\(\sim\)3%. The careful evaluated (n,p) excitation function for the incident neutron in the interval 9 \(\sim\) 10 MeV and 10 \(\sim\) 11 MeV were given with uncertainty 3.09% and 2.87%, respectively by Zolotarev [16] in 2002 which were included in IRDFFv1.05. The maximum cross section for (n,\(\alpha\)) in the above energy region is below 30 mb. It is obvious that (n,\(\alpha\)) cross section could be derived if there were accurately measured neutron elastic-scattering cross section in this region.\textsuperscript{nat}Fe(n,el) cross section measurement between 9.41 \(\sim\) 15.2 MeV were measured in 1994 at PTB by Schmidt [17] with uncertainty less than 3%. The four red points in Fig. 1(b) are the results which were obtained according to Eq. (2). Red points overlap with the data of Nelson and within 4%. Above 11 MeV, all sets are consistent. In the 14 MeV energy region (13.5 \(\sim\) 14.8 MeV), a highly accurate evaluated result at 14 MeV, performed by Vonach at the IRK, was also included. This value was taken as a single data point replacing all experimental data in that energy range. Figure 1(b) shows that this value (blue point at 14 MeV) agrees well with Nelson.

The black line is the evaluated result of this work which is based on the Negret’s data below 6 MeV and Nelson’s results up to 20 MeV.

4. Conclusions

In order to obtain a more accurate description of the cross section data within our best knowledge, the existing old and new experimental data were collected and carefully analyzed. The new recommended data for inelastic cross sections on \(^{56}\text{Fe}\) were performed in the fast neutron energy range. Various methods were applied to understand discrepancies in the inelastic scattering cross sections, especially in the energy range between 9 \(\sim\) 11 MeV. Such improved experimental data will provide more reliable constraints for theoretical calculations within the CIELO project.

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References