Effect of correlation between cross sections and angular distributions in nuclear data of ^{63}Cu on estimation of uncertainty of neutron penetration

Naoki Yamano [1*], *Tsunenori* Inakura [1], *Chikako* Ishizuka [1], and *Satoshi* Chiba [1]

[1] Tokyo Institute of Technology, Ookayama, Meguro, Tokyo 152-8550, Japan

Abstract. Uncertainty in neutron reaction rates after penetrating a 608mm-thick copper benchmark experiment performed by the FNS facility of JAEA was estimated based on two different kinds of the Total Monte Carlo methods under random sampling methodology. 500 random nuclear data files were generated for ^{63}Cu by the T6 code system with perturbing underlying model parameters. In the first method, these files were used directly to yield processed library preserving all the correlations among different physical quantities. In the second method, the random files populated by T6 were used but the angular distribution data were kept fixed to the non-perturbed nominal ones. It was found that the two methods gave the same neutron reaction rate after the 608 mm penetration of a copper, however uncertainty of the second method was larger than that of the first method. It shows that the correlation between total cross section and angular distribution of elastic scattering at 0 degree, which stems from Wick's inequality, affects uncertainty of the calculated neutron reaction rate. The result is consistent with the case of ^{28}Si on deep penetration of thick concrete problem previously reported by the authors. It could be concluded that the uncertainty obtained by using the covariance files given in the ENDF-6 format may not give correct results for the uncertainty of neutron penetration calculation.

1 Introduction

Accurate estimation of uncertainties in the neutron transport calculation caused by nuclear data through error propagation is quite important for Verification and Validation (V&V) of design and safety analysis of nuclear systems. Usually, such an error propagation is carried out through the covariance files given in nuclear data in the ENDF-6 format [1]. However, it is also recognized that the ENDF-6 format cannot represent all the correlation of nuclear data that might be present in different kind of physical quantities. For example, the uncertainty of the angular distribution of elastic scattering is able to represent in the ENDF-6 format (MF34, MT2), however it is only processed by that of the average cosine of the angular distribution in the nuclear data processing code, which may be reasonable for application in design of nuclear reactors since the energy of neutrons involved is usually small, so the elastic scattering is quite close to be isotropic in the center-of-mass framework. However, uncertainty of the angular distribution and correlation of it to those of other quantities such as total cross section might be significantly important for systems where effects of the angular anisotropy is more important. Correlation of uncertainties of such different quantities, in this case that of angular distribution of elastic scattering and that of total cross section, is not represented properly in the ENDF-6 format, hence this correlation is normally ignored. However, we know that there is a positive correlation between these two quantities described by Wick's inequality [2], $\sigma_{el}(0) \geq \left(\frac{k}{4\pi}\sigma_t\right)^2$, where $\sigma_{el}(0)$, σ_t and k denote elastic scattering cross section at 0 degree, total cross section and wave number of incident neutron, respectively. The optical theorem of the quantum scattering theory is the underlying physics of this correlation. We have shown that the correlation of different quantities of ^{28}Si becomes significant on the deep penetration of fission neutrons through a thick concrete [3].

It is the purpose of this study to evaluate the effects of the correlation between forward elastic scattering and total cross section on the penetration of D-T neutrons through a thick copper, via Total Monte Carlo (TMC) method [4].

2 Method

We have done the following two calculations to prepare random nuclear data files.

1. In the first method, we have calculated 500 random nuclear data files for ^{63}Cu, which is one of the major constituents in natural copper, by using T6 code system [5]. In this calculation, underlying model parameters for the resolved resonance and continuous region were

* Corresponding author: yamano@zc.iir.titech.ac.jp

perturbed with probability distributions determined from a Bayesian Monte Carlo (BMC) method [6], and 500 ENDF-formatted random files were generated. These random files were used for the subsequent neutron transport calculations. This process will be explained later in more details. By this process, we could obtain the variance of the neutron reaction rates at each point of the thick copper, especially after penetrating the whole copper, and we could assign uncertainty of the neutron transport calculation from the variance. In this calculation, all the correlations of quantities present in the nuclear cross section were taken into consideration especially those between the angular distribution of elastic scattering and the total cross section.

2. In the second one, which is similar to that of the 1st method but all the angular distribution data (MF4, MT2) were kept fixed to the non-perturbed nominal ones, hence it ignores the uncertainty of the angular distribution of elastic scattering cross sections only. The 500 random files keeping all the angular distribution data to be the same were used for the subsequent neutron transport calculations.

The variances of the neutron reaction rates after complete penetration obtained by these two methods, that are the indicators of the uncertainty of neutron transport calculation, were compared, and we have elucidated the importance of the correlation of different quantities in nuclear data libraries.

The covariance of ^{63}Cu neutron cross sections, which is only evaluated for resonance parameters in ENDF/B-VIII.0 [7], was estimated using T6 as shown in the upper part of **Fig. 1**. The nominal parameters of the calculation were adjusted to reproduce neutron cross sections stored in ENDF/B-VIII.0. To estimate covariance matrices, firstly a distribution of parameters in the continuous region was calculated with the BMC method. Secondly, Monte Carlo calculation was performed based on the errors of resolved resonance parameters and the parameter distribution in the continuous region obtained from BMC as above. If the cross section errors in the resonance region were experimentally known, the uncertainties of the resonance parameters were carefully adjusted to reproduce those errors and all the resonance energies and widths were perturbed independently within the assigned errors. In the same way, all the parameters in the continuous region, which consist of more than 200 parameters including those of optical model potential, level densities, were perturbed as well as pre-equilibrium model parameters. All these perturbation calculations were conducted based on the parameter distributions which were obtained starting from a non-informative prior distribution using the BMC method, to produce 500 random files. Finally, a covariance file was generated by statistical processing of the results for these random files.

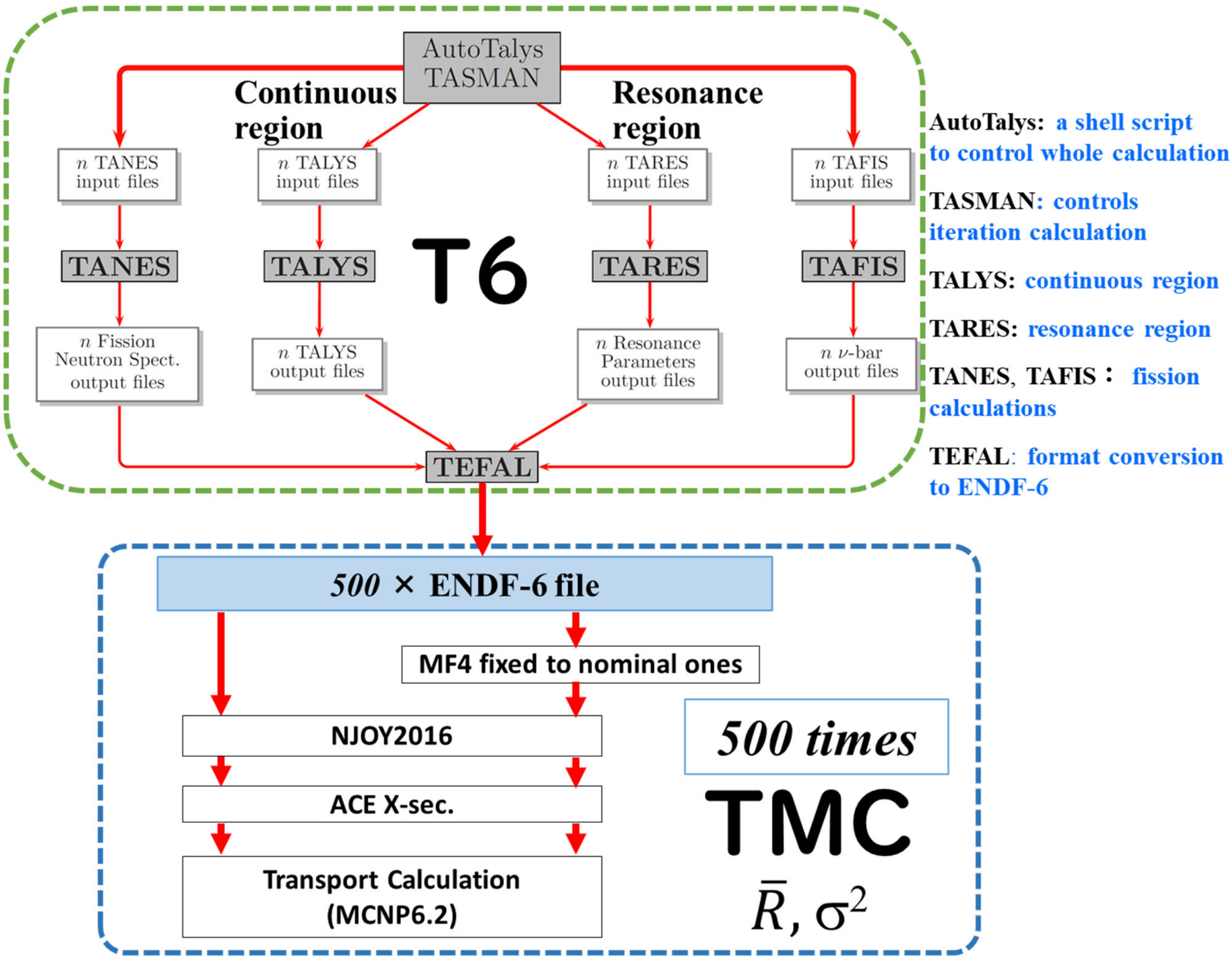


Fig. 1. Calculation scheme of cross section and associated covariance (T6, upper part), and the Total Monte Carlo (TMC) method (lower part). TANES and TAFIS in T6 were not used in the present work.

Figures 2 and **3** show the comparison of ^{63}Cu total cross sections and elastic cross sections between 500 random data calculated by T6 and ENDF/B-VIII.0, respectively. The cross sections in the resonance and the continuous regions calculated by T6 are consistent with those given in ENDF/B-VIII.0. The information on the correlation among the different reactions is used in the random files produced by T6.

To estimate the uncertainty of the neutron reaction rates, the TMC scheme was applied as shown in the lower part of Fig. 1. As it shows, a random file in the ENDF-6 format produced by T6 was processed using NJOY-2016 [8], and the ACE file was generated. When using NJOY, the total cross section is reconstructed from all the partial cross sections.

Then, a neutron transport calculation using MCNP6.2 [9] was performed to compute the 608 mm-thick copper benchmark experiment conducted by FNS of JAEA for which a D-T neutron source is located at 200 mm distance from the front surface of the copper slab [10]. The energy spectra of the D-T neutrons were also estimated by JAEA [10]. The two steps described above were repeated 500 times for each random file generated by T6, and the standard deviations of the calculated reaction rates were obtained by statistical processing using each of these 500 results. The computational model of the neutron transport calculation is shown in **Fig. 4** and the other conditions relating to the transport calculation are the same during repeated TMC runs, therefore, these uncertainties can be ignored.

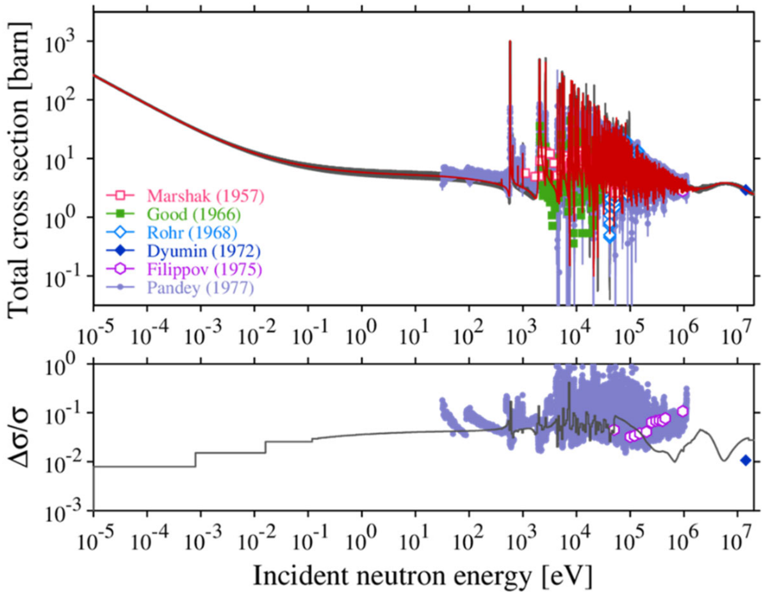


Fig. 2. ^{63}Cu neutron total cross sections estimated by T6 system (gray lines) compared with ENDF/B-VIII.0 (red line) and experimental data and the errors [12-17].

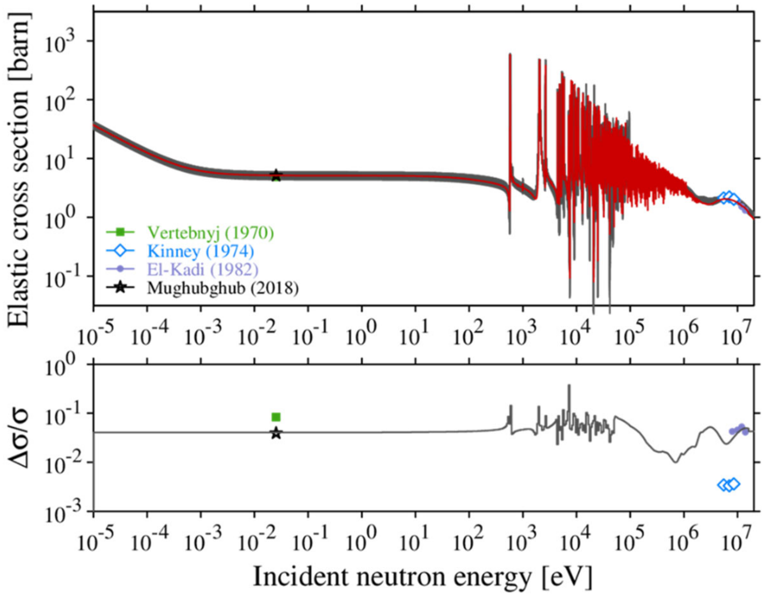


Fig. 3. ^{63}Cu neutron elastic cross sections estimated by T6 system (gray lines) compared with ENDF/B-VIII.0 (red line) and experimental data [18-21].

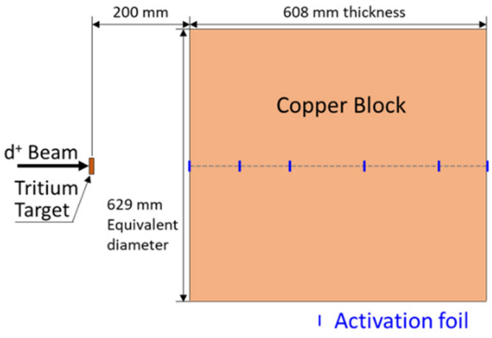


Fig. 4. Computational geometry model of FNS copper benchmark experiment.

3 Results and discussion

Neutron reaction rates for ^{58}Ni(n,2n)^{57}Ni reaction together with experimental data in the copper slab and their standard deviations are shown in in the lower panel of **Fig. 5**. The average reaction rates obtained with random files generated by T6 and those ignoring the perturbation of the angular distribution were equivalent to each other, and the calculated values were in good agreement with experimental data except for the 608 mm penetration. It was reported that scattered neutrons from concrete wall around the experimental room affected the experimental data [10]. However, the fractional standard deviation (fsd) which means the relative standard deviation to the reaction rate, as shown in the upper panels, was slightly different. The standard deviation of ^{58}Ni(n,2n)^{57}Ni reaction rate at the 608 mm copper transmission obtained by random files generated with ignoring the perturbation of the angular distribution was 18.4%, which must be compared to 14.3% obtained by those generated by T6.

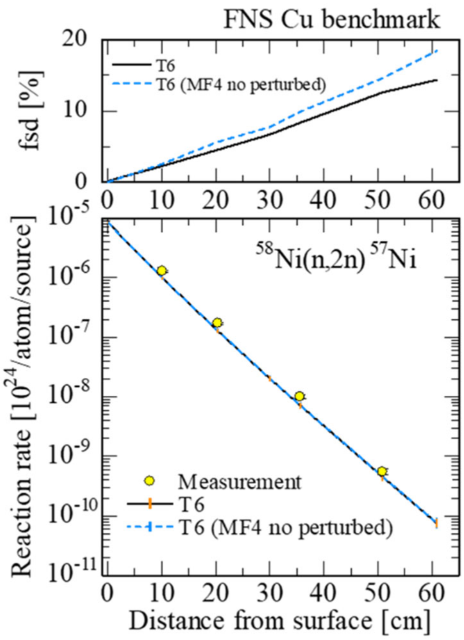


Fig. 5. Comparison of ^{58}Ni(n,2n)^{57}Ni reaction rate distribution and the standard deviation between T6 and the result ignoring perturbation of angular distribution.

The fractional standard deviation of ^{93}Nb(n,2n)^{92m}Nb reaction rate at the 608 mm copper transmission obtained by random files generated by those ignoring the perturbation of the angular distribution was 18.6%, whereas 14.5% obtained by those generated by T6 as shown in **Fig. 6**.

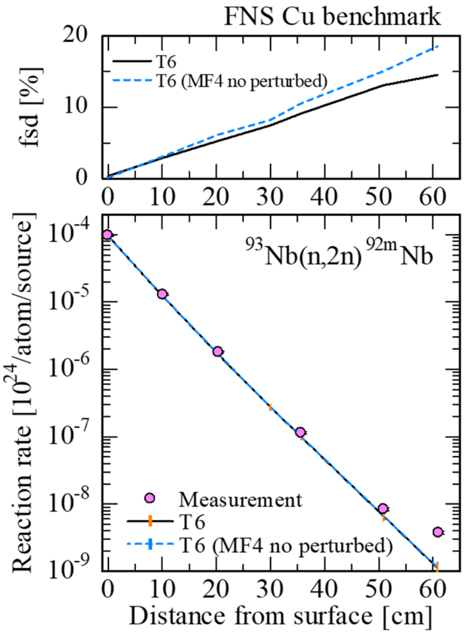


Fig. 6. Comparison of ^{93}Nb(n,2n)^{92m}Nb reaction rate distribution and the standard deviation between T6 and the result ignoring perturbation of angular distribution.

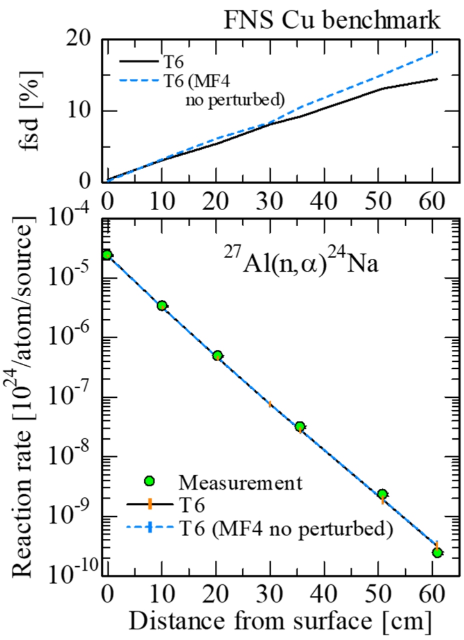


Fig. 7. Comparison of ^{27}Al(n,α)^{24}Na reaction rate distribution and the standard deviation between T6 and the result ignoring perturbation of angular distribution.

The fractional standard deviation of $^{27}Al(n,\alpha)^{24}Na$ reaction rate at the 608 mm copper transmission obtained by random files generated by those ignoring the perturbation of the angular distribution was 18.3%, whereas 14.5% obtained by those generated by T6 as shown in **Fig. 7**.

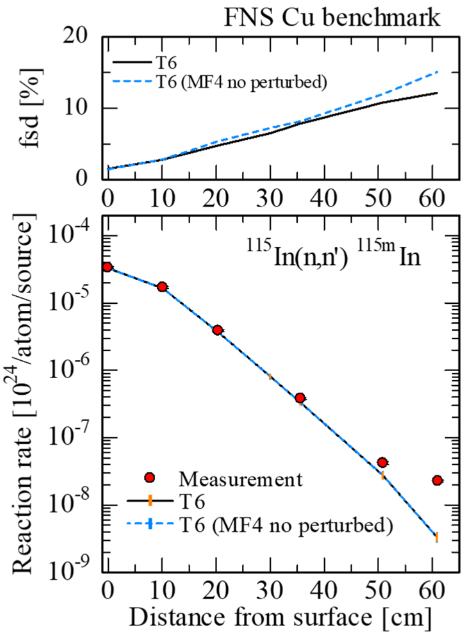


Fig. 8. Comparison of $^{115}In(n,n')^{115m}In$ reaction rate distribution and the standard deviation between T6 and the result ignoring perturbation of angular distribution.

The fractional standard deviation of $^{115}In(n,n')^{115m}In$ reaction rate at the 608 mm copper transmission obtained by random files generated by those ignoring the perturbation of the angular distribution was 15.1%, whereas 12.2% obtained by those generated by T6 as shown in **Fig. 8**.

The standard deviation of the latter, which provides for uncertainties of the angular distributions and energy distributions of secondary neutrons as well as cross sections, is smaller than the former which includes only uncertainties of the cross sections primarily. The other threshold reactions showed similar trends.

Neutron reaction rates for (n,γ) reactions in the copper slab and their standard deviations are also shown in **Figs. 9** and **10**. The fractional standard deviations for (n,γ) reactions showed different trends compared with the threshold reactions. The standard deviations for the (n,γ) reactions were almost the same between the two methods.

The random files shown as "MF4 no perturbed" were created that the angular distribution data (MF4, MT2) were kept fixed to the non-perturbed nominal ones where the perturbation of the elastic scattering angular distribution was ignored, so its correlation to the total cross section was also ignored. On the other hand, the random files generated by T6 includes perturbations of the angular distribution of secondary neutrons and correlation to other quantities.

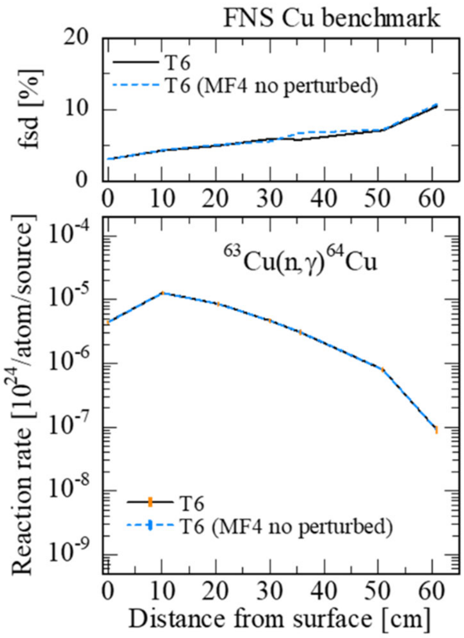


Fig. 9. Comparison of $^{63}Cu(n,\gamma)^{64}Cu$ reaction rate distribution and the standard deviation between T6 and the result ignoring perturbation of angular distribution.

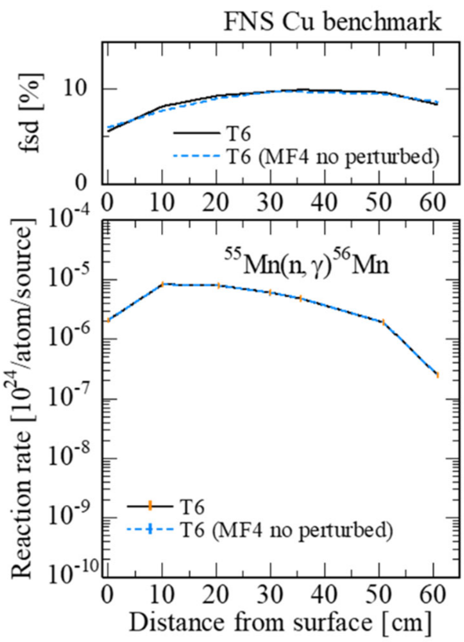
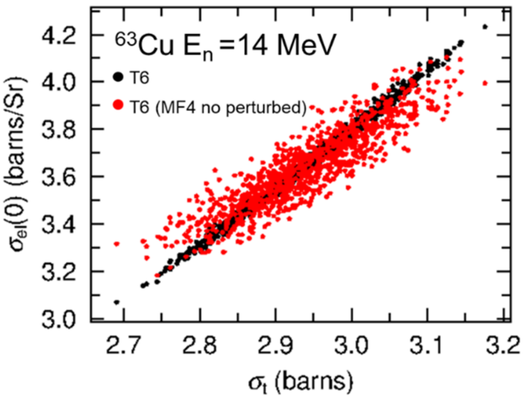


Fig. 10. Comparison of $^{55}Mn(n,\gamma)^{56}Mn$ reaction rate distribution and the standard deviation between T6 and the result ignoring perturbation of angular distribution.

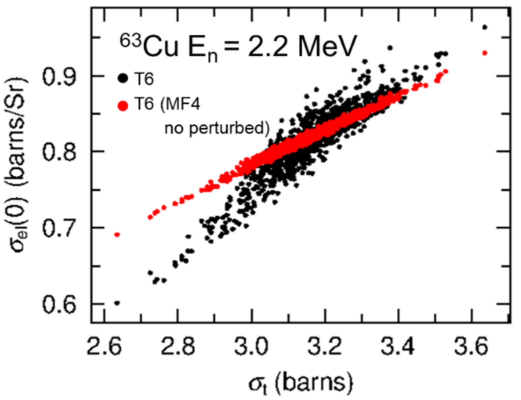
The statistical error of estimated uncertainty (fsd) for the TMC method (N=500) was estimated to be about 3% based on the χ-distribution [11] for the $^{58}Ni(n,2n)^{57}Ni$ reaction rate at the 608 mm transmission.

Figures 11(a) and **(b)** show the correlations between the total cross section and the elastic scattering cross section at 0 degree in the 1,000 random files generated by T6 at the incident neutron energy of 14 MeV and 2.2 MeV, respectively.

In these figures, red points indicate that the angular distribution data (MF4, MT2) were kept fixed to the non-perturbed nominal ones. It is clear that there is a positive correlation between the total cross section and the forward elastic scattering cross section, which is consistent with Wick's inequality derived from the optical theorem.



(a) Incident neutron energy is 14 MeV.



(b) Incident neutron energy is 2.2 MeV.

Fig. 11. Correlation between ^{63}Cu total cross section and elastic scattering cross section at 0 degree of 1,000 random files generated by T6.

This correlation gives rise to the cancellation of the variance of neutron threshold reaction rates of the penetrated neutrons in the following manner: in general, neutron transmission decreases as the total cross section increases. However, the forward scattering cross section, which is positively correlated with the total cross section, works to increase neutron transmission. Therefore, the forward elastic scattering cross section changes so as to cancel the change in neutron transmission caused by the change in the total cross section caused by the perturbation of the parameter. That is, the standard deviation of neutron threshold reaction rates were reduced when the correlation of the differential elastic scattering cross section to the total cross section was considered properly. This result is consistent with the ^{28}Si uncertainty on deep penetration of thick concrete problem previously reported by the authors [3]. Therefore, it was found that 1) uncertainty of the angular distribution data must be properly considered, 2) its correlation to other quantities like total cross section must also be considered properly, and 3) if the items 1) and 2) were ignored, the uncertainty of the neutron threshold reaction rates are overestimated. These conclusions give an important warning to the interpretation of the neutron transport calculations which were thoroughly based on the ENDF-formatted files, where items 1) and 2) are not fulfilled in general. It has to be noticed at the same time that these conclusions are applicable to cases where the anisotropy of neutron scattering is highly important for the results like the neutron penetration problems considered in this work.

4 Conclusion

The effect of uncertainty in nuclear data on the neutron penetration of copper slab was estimated with the TMC method based on nuclear data generated by T6. The average threshold reaction rates obtained with random files generated by T6 and those ignoring the correlation between the angular distribution of elastic scattering and the total cross sections were equivalent. However, the standard deviation of ^{58}Ni(n,2n)^{57}Ni reaction rate obtained with those ignoring the correlation between the angular distribution of elastic scattering and the total cross sections at the 608 mm copper transmission was 18.4%, which is larger than 14.3% obtained with T6 data. The cause of difference was attributed to that the correction of the angular distribution of elastic scattering and total cross section could not be considered in calculations with the ENDF-6 format. It has been concluded that the correlation of angular distribution of secondary neutrons to other quantities, especially to total cross section, needs to be dealt with correctly in the neutron deep penetration where angular anisotropy is important.

References

1. A. Trkov, M. Herman, D.A. Brown, (Eds). ENDF-6 Formats Manual, Data Formats and Procedures for the Evaluated Nuclear Data File ENDF/B-VI, ENDF/B-VII and ENDF/B-VIII. Brookhaven National Laboratory (US); 2018. (BNL report BNL-203218-2018-INRE).
2. G.C. Wick. A Theorem on Cross Sections. Phys. Rev. **75**(9), (1949) 1459.
3. N. Yamano, T. Inakura, C. Ishizuka, S. Chiba. Crucial importance of correlation between cross sections and angular distributions in nuclear data of ^{28}Si on estimation of uncertainty of neutron dose penetrating a thick concrete. J. Nucl. Sci. Technol. **59**(5), (2022) 641-646.
4. A.J. Koning, D. Rochman. Towards sustainable nuclear energy: Putting nuclear physics to work. Annals of Nuclear Energy. **35**(11), (2008) 2024-2030. DOI: 10.1016/j.anucene.2008.06.004.
5. A.J. Koning, D. Rochman. J.-Ch. Sublet, et al. TENDL: Complete Nuclear Data Library for

Innovative Nuclear Science and Technology. Nucl. Data Sheets. **155**, (2019) 1-55.

6. A.J. Koning Bayesian Monte Carlo method for nuclear data evaluation. Eur. Phys. J. A. **51**, (2015) 184. DOI: 10.1140/epja/i2015-15184-x. See also P.J. Griffin, IEEE Transactions on Nuclear Science. 66(1). Sandia National Laboratory (US); 2018. (Sandia NL report SAND-2018-11821J).
7. D.A. Brown, M.B. Chadwick, R. Capote, A.C. Kahler, A.Trkov, et al. ENDF/B-VIII.0: The 8th Major Release of the Nuclear Reaction Data Library with CIELO-project Cross Sections, New Standards and Thermal Scattering Data. Nucl. Data Sheets. **148** (2018) 1-142.
8. R.E. MacFarlane, A.C. Kahler. Methods for Processing ENDF/B-VII with NJOY. Nucl. Data Sheets. **111** (2010) 2739-2890.
9. C.J. Werner (Ed.). MCNP® USER'S MANUAL Code Version 6.2. Los Alamos National Laboratory (US); 2017. (LANL report LA-UR-17-29981).
10. F. Maekawa, Y. Oyama, C. Konno, Y. Ikeda, K. Kosako, H. Maekawa. Benchmark Experiment on a Copper Slab Assembly Bombarded by D-T Neutrons. Japan Atomic Energy Agency (Japan); 1994. (JAEA report JAERI-M 94-038).
11. B. Foad, A. Yamamoto, T. Endo. Uncertainty and regression analysis of the MSLB accident in PWR based on unscented transformation and low rank approximation. Ann. Nucl. Energy. **143** (2020) 107493.
12. H. Marshak, H.W. Newson. Neutron Resonances in the keV Region: Odd-Intermediate Nuclei. Phys. Rev. **106** (1957) 110.
13. W.M. Good, D. Paya, R. Wagner, T. Tamura. Total Cross Sections for Kilovolt Neutrons of Even-Odd Nuclei in the Region of the 3s Strength-Function Resonances. Phys. Rev. **151** (1966) 912.
14. G. Rohr, K.N. Mueller. The Total Cross-Sections of Cu-63 and Cu-65 in keV Region. Report from Euratom-countries + Euratom to EANDC, **89** (1968) 1.
15. A.I. Dyumin, D.M. Kaminker, G.N. Popova, V.A. Smolin. Total Neutron Cross Sections for Titanium, Chromium, and Copper Isotopes. Izv. Rossiiskoi Akademii Nauk, Ser. Fiz. **36** (1972) 852.
16. V.V. Filippov. Measurement of Capture and Total Cross Sections for Intermediate and Fast Neutrons by Energy Correlation Methods. 3.All Union Conf. on Neutron Phys. Vol. 2 (1975) 53, Kiev, 9-13 June 1975.
17. M.S. Pandey, J.B. Garg, J.A. Harvey. Neutron total cross sections and resonance parameters of ^{63}Cu and ^{65}Cu. Phys. Rev. C. **15** (1977) 600.
18. V.P. Vertebnyj, M.F. Vlasov, N.L. Gnidak, et al. Neutron Cross-Sections of Isotopes used as Absorbers in Atomic Reactors. Nuclear Data for Reactors Conf. Vol.1 (1970) 651, Helsinki, 1970.
19. W.E. Kinney, F.G. Perey. ^{63}Cu and ^{65}Cu neutron elastic and inelastic scattering cross sections from 5.50 to 8.50 MeV. Oak Ridge National Laboratory (US); 1974. (ORNL report ORNL-4908).
20. S.M. El-Kadi, C.E. Nelson, F.O. Purser, et al. Elastic and inelastic scattering of neutrons from ^{54,56}Fe and ^{63,65}Cu: (I). Measurements from 8 to 14 MeV and a spherical optical model analysis. Nuclear Physics, Section A. **390** (1982) 509-540.
21. S.F. Mughabghab. Atlas of neutron resonances. 6th ed. (2018) Elsevier Science; New York (NY). ISBN-9780444637697.