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Evaluation of Neutron Capture Cross Sections and Covariances on ^{99}Tc and ^{129}I in the keV Energy Region

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Abstract. Neutron capture cross sections and covariances on radioactive ^{99}Tc and ^{129}I have been required for developing environmental load-reducing technology. Their evaluation was performed by using nuclear reaction calculation code CCONE and Baysian code KALMAN with data assumed on the basis of measured data. The obtained total and capture cross sections are in good agreement with the measured data. The resulting uncertainties of capture cross section were 12-18% and 20-29% for ^{99}Tc and ^{129}I, respectively, in the keV energy region.

1 Introduction

Neutron capture cross sections and covariances on long-lived fission products (LLFPs) as well as major and minor actinides have been required for development of environmental load-reducing technology. Neutron cross section data for the LLFPs of ^{99}Tc (half-life 0.21 Myr) and ^{129}I (15.7 Myr) are included in major evaluated libraries (e.g., JENDL-4.0 [1], ENDF/B-VII.1 [2] and JEFF-3.2 [3]). Their covariance data were, however, not included in JENDL-4.0 and JEFF-3.2, although they are already taken into account in ENDF/B-VII.1. The cumulative yields of radioactive ^{99}Tc and ^{129}I are 6.1-5.2% and 0.5-3.0%, respectively, in ^{235}U fission at thermal to fast neutron energies [4]. They have long lasting radiotoxicity due to long half-lives. Thus, it is well known that they are the candidates for nuclear transmutation by neutron capture reactions in, e.g., accelerator driven system. In the present work the evaluation of neutron capture cross sections and covariances for ^{99}Tc and ^{129}I was performed in the 1-10^3 keV energy region (hereafter referred to as keV energy region).

2 Evaluation Method

The evaluation of neutron cross sections was done by using nuclear reaction calculation code CCONE (version 0.8) [5]. The detailed methodology of cross section evaluation for ^{99}Tc can be found in Ref. [6]. For ^{129}I the coupled-channels optical model based on the rigid rotator model was adopted with the potential form of Kunieda et al. [7] for target nucleus. The coupled levels were taken from the Evaluated Nuclear Structure Data File (ENSDF) [8]. The deformation parameters fixed by fitting to measured data were relatively small (0.05 and 0.03 for ^{127,129}I, respectively), compared to neighboring even-even nuclides (0.12-0.18). Similar results were recently obtained by Shibata [9].

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The Hauser-Feshbach statistical model with width fluctuation correction was used with discrete level information retrieved from Reference Input Parameter Library (RIPL-3) [10]. The nuclear level density was adopted by the composite Gilbert-Cameron formula with Fermi-gas model [11]. The level density parameters were determined if s-wave resonance spacings were available from measurements. Otherwise, the systematic values of Ref. [11] were used. Generalized Lorentzian form was employed for gamma strength function of E1 radiation [12]. The M1 and E2 transitions for gamma-decays were also taken into account by the Lorentzian form [12].

Sensitivities of the parameters, related to the optical model potential (OMP) and reaction models, to the cross sections were also calculated by the CCONE code, and then, the sensitivity matrix **C** was generated from these sensitivities for covariance evaluation. The uncertainties for the potential depths of OMP were given to be 5%. The uncertainties for potential radius and diffuseness were 3% and 10%, respectively. The relatively large uncertainties (10-50%) were assigned for the deformation parameters, since their information was not directly obtained for odd isotopes. The uncertainties of level density parameter were chosen to be 3%, which was small relative to those of other parameters for the reaction models. Thus, the initial parameter covariance matrix was made from these parameter uncertainties without considering the correlation among the parameters.

The covariance evaluation was done by using Baysian code KALMAN [13], which is based on the generalized least-square approach. In this code the parameter covariance matrix was used as prior covariance matrix **X**. Experimental covariance matrix **V** was made by assuming data with errors at energy points. The data and energy were taken from the present cross section data. The data errors (σ) consisted of statistical (σ_{sta}) and systematic (σ_{sys}) components like $\sigma^2 = \sigma^2_{sta} + \sigma^2_{sys} = 2\,\sigma^2_{sta}$. The correlation among the data was prescribed as follows:

$$Corr\,(V_{i,j}) = \begin{cases} 1.0 & \text{if } i = j, \\ 0.5 & \text{if } i \neq j, \end{cases} \tag{1}$$

instead of directly using statistical and systematic errors derived from measurements. One reason to make this assumption is that error given in experimental data is relatively small, but the dispersion of experimental data is much larger than the errors. The posterior covariance was calculated by

$$\mathbf{P} = (\mathbf{X}^{-1} + \mathbf{C}^T\mathbf{V}^{-1}\mathbf{C})^{-1} \tag{2}$$

$$= \mathbf{X} - \mathbf{X}\mathbf{C}^T(\mathbf{C}\mathbf{X}\mathbf{C}^T + \mathbf{V})^{-1}\mathbf{C}\mathbf{X}, \tag{3}$$

from the prior and experimental covariance matrices. Finally, the cross section covariance matrix was obtained by the usual equation $\mathbf{M} = \mathbf{C}\mathbf{P}\mathbf{C}^T$.

3 Results

3.1 ^{99}Tc

The evaluated results of total cross section and covariance are shown as a function of incident neutron energy in Figure 1. The gray shaded area represents the prior error which was given to cover the experimental data from uncertainties of the OMP parameters. The assumed errors at 5 energy points were shown by green bars. The red shaded area shows the posterior error obtained by the KALMAN calculation. The prior and posterior errors are also shown by blue dashed and solid lines at the right axis. The result of total cross section reasonably explains the experimental data of Foster Jr and Glasgow [14]. The posterior error was obtained to be 3-6% in the keV energy region.

Figure 2 shows the evaluated result of capture cross section. The evaluated errors are almost consistent with most of the measured data, except for Chou et al. [15] who derived the data slightly

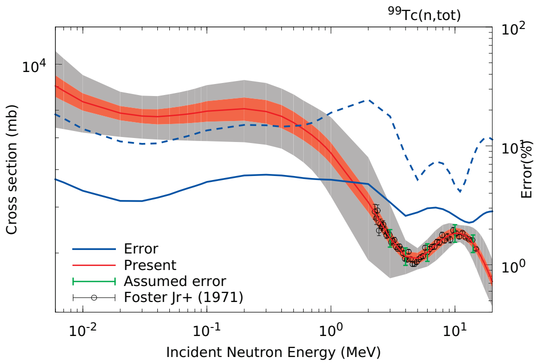

Figure 1. Evaluated results of total cross section and its uncertainty of ^{99}Tc.

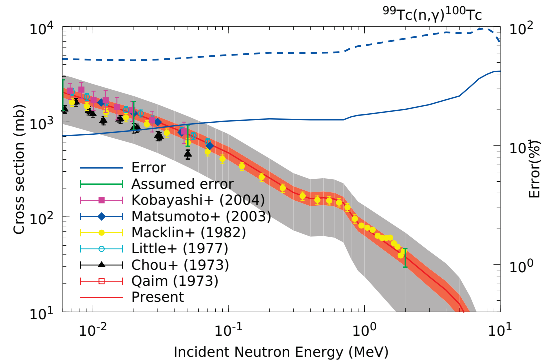

Figure 2. Evaluated results of capture cross section and its uncertainty of ^{99}Tc.

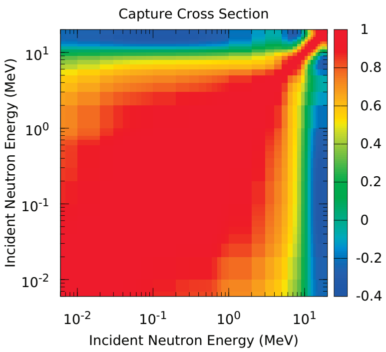

Figure 3. Correlation matrix for capture cross section of ^{99}Tc.

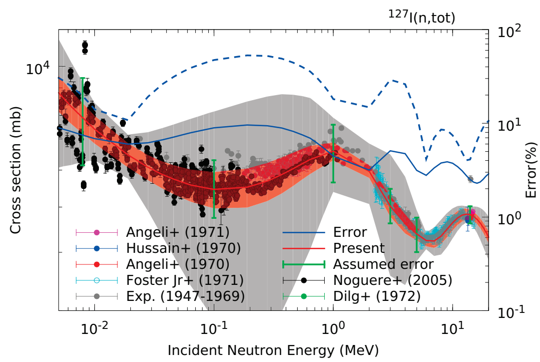

Figure 4. Evaluated results of total cross section and its uncertainty of ^{127}I.

smaller. The prior error was firstly assumed to be 50% or higher, but the posterior error was well constrained by the assumed errors and other information coming from measured capture gamma-ray spectra [16]. The evaluated result of uncertainty was 12-18% in the keV energy region.

Figure 3 shows the resulting correlation matrix of capture cross section. The strong and positive correlation is found below 10 MeV. This is because the positive change of parameters such as level density parameter and normalization factor of gamma strength functions for the compound nucleus, makes the cross section uniformly increase in this region as seen in the figure on parameter sensitivities for ^{129}I below.

3.2 ^{129}I

In contrast to ^{99}Tc, the limited number of experiments was made for radioactive ^{129}I. Therefore, the neutron cross sections of ^{129}I were evaluated, together with those of stable ^{127}I, because many experimental efforts have been done for ^{127}I. The present evaluation of total cross sections was based on data recently measured by Noguere et al. [17, 18]. The result for ^{127}I is illustrated in Figure 4. The adopted OMP parameters have mass-number dependence, but the potential differences coming

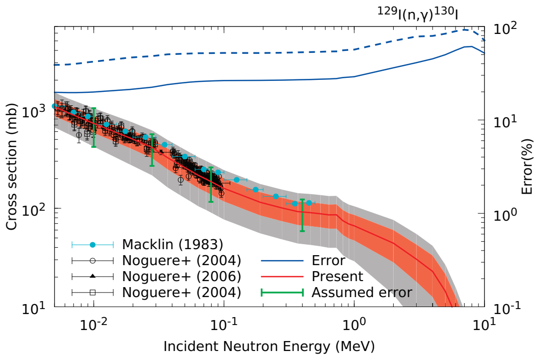

Figure 5. Evaluated results of capture cross section and its uncertainty of ^{129}I.

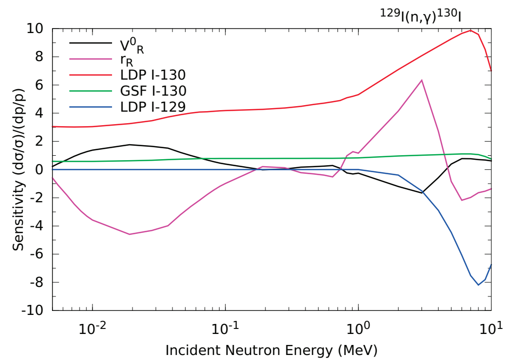

Figure 6. Sensitivities of model parameters to capture cross section for ^{129}I.

from the mass-number difference are not so large that the total cross sections show almost similar characteristics. The present OMP parameters reasonably explain the measured data of both ^{127,129}I. For covariance evaluation the representative points were assumed to encompass all measured data in the energy range of 5 keV to 20 MeV for ^{127}I, whereas measured data for ^{129}I were provided only below the neutron energies of 60 keV [17]. Thus, the overall uncertainty evaluation comes from the assumed data of ^{127}I. The uncertainty of 5-10% for ^{129}I in the keV energy region was derived by the present method of covariance evaluation, although individual errors of measured data are typically much smaller. This result can be found from Figure 4. Figure 5 shows the evaluated capture cross section on ^{129}I. The data of Macklin [19] are marginally consistent with those of Noguere et al. [17, 20]. The present evaluation takes into account both the measured data and gives the errors of 20-29% in the keV energy region.

The sensitivities of the parameters for the OMP and reaction models to the capture cross section on ^{129}I are represented in Figure 6, which were calculated by the CCONE code. The capture cross section is very sensitive to the level density parameter of compound nucleus ^{130}I (LDP I-130). The negative sensitivity for level density parameter of target ^{129}I (LDP I-129) is found above 1MeV. This is due to the contribution of inelastic scattering cross section. The gamma strength function of ^{130}I (GSF I-130) has a weak, but positive sensitivity to the cross section. The sensitivities for OMP parameters (e.g., constant leading term V_R^0 of potential depth and radius r_R in the real volume term) are changed to be positive and negative with changing neutron energy.

4 Summary

The evaluation of neutron capture cross sections and covariances on well-known LLFPs of ^{99}Tc and ^{129}I was performed by using CCONE-KALMAN code system, in order to recognize the present uncertainties of capture cross sections which are needed to develop environmental load-reducing technology. The measured data of stable ^{127}I were used for the better cross section and uncertainty evaluations of ^{129}I due to the lack of experimental information. The calculated data for ^{99}Tc and ^{129}I well reproduce the measured data of total and capture cross sections. The obtained uncertainties of capture cross section resulted in 12-18% for ^{99}Tc and 20-29% for ^{129}I in the keV energy region. The capture cross sections of the evaluated libraries (JENDL-4.0, ENDF/B-VII.1 and JEFF-3.2) are within the present uncertainties.

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References

[1] K. Shibata, O. Iwamoto, T. Nakagawa, N. Iwamoto, A. Ichihara, S. Kunieda, S. Chiba, K. Furutaka, N. Otuka, T. Ohsawa, T. Murata, H. Matsunobu, A. Zukeran, S. Kamada, J. Katakura, J. Nucl. Sci. Technol. **48**, 1 (2010)

[2] M.B. Chadwick, M. Herman, P. Oblozinský, M.E. Dunn, Y. Danon, A.C. Kahler, D.L. Smith, B. Pritychenko, G. Arbanas, R. Arcilla, R. Brewer, D.A. Brown, R. Capote, A.D. Carlson, Y.S. Cho, H. Derrien, K. Guber, G.M. Hale, S. Hoblit, S. Holloway, T.D. Johnson, T. Kawano, B.C. Kiedrowski, H. Kim, S. Kunieda, N.M. Larson, L. Leal, J.P. Lestone, R.C. Little, E.A. McCutchan, R.E. MacFarlane, M. MacInnes, C.M. Mattoon, R.D. McKnight, S.F. Mughabghab, G.P.A. Nobre, G. Palmiotti, A. Palumbo, M.T. Pigni, V.G. Pronyaev, R.O. Sayer, A.A. Sonzogni, N.C. Summers, P. Talou, I.J. Thompson, A. Trkov, R.L. Vogt, S.C. van der Marck, A. Wallner, M.C. White, D. Wiarda, P.G. Young, Nucl. Data Sheets, **112**, 2887 (2011)

[3] OECD Nuclear Energy Agency Data Bank.

[4] J. Katakura, JAEA-Data/Code 2011-025 (Japan Atomic Energy Agency) (2012)

[5] O. Iwamoto, J. Nucl. Sci. Technol. **44**, 687 (2007)

[6] N. Iwamoto, J. Nucl. Sci. Technol. **49**, 244 (2012)

[7] S. Kunieda, S. Chiba, K. Shibata, A. Ichihara, E.Sh. Sukhovitskii, J. Nucl. Sci. Technol. **44**, 838 (2007)

[8] Evaluated Nuclear Structure Data File. Available from http://www.nndc.bnl.gov/ensdf/

[9] K. Shibata, J. Nucl. Sci. Technol. **52**,1174 (2015)

[10] R. Capote, M. Herman, P. Oblozinsky, P.G. Young, S. Goriely, T. Belgya, A.V. Ignatyuk, A.J. Koning, S. Hilaire, V.A. Plujko, M. Avrigeanu, O. Bersillon, M.B. Chadwick, T. Fukahori, Z. Ge, Y. Han, S. Kailas, J. Kopecky, V.M. Maslov, G. Reffo, M. Sin, E.Sh. Soukhovitskii, P. Talou, Nucl. Data Sheets, **110**, 3107 (2009)

[11] A. Mengoni, Y. Nakajima, J. Nucl. Sci. Technol. **31**,151 (1994)

[12] J. Kopecky, M. Uhl, Phys. Rev. **C41**, 1941 (1990)

[13] T. Kawano, K. Shibata, JAERI-Data/Code 97-037 (Japan Atomic Energy Agency) (1997) [in Japanease]

[14] D.G. Foster Jr., D.W. Glasgow, Phys. Rev. **C3**, 576 (1971)

[15] J.-C. Chou, H. Werle, J. Nucl. Energy, **27**, 811 (1973)

[16] T. Matsumoto, M. Igashira, T. Ohsaki, J. Nucl. Sci. Technol. **40**, 61 (2003)

[17] G. Noguere, A. Brusegan, A. Leprêtre, N. Herault, O. Bouland, G. Rudolf, *Int. Conf. on Nucl. Data for Sci. and Technol., Santa Fe 2004 Vol.2* (AIP, New York, 2005) 1462

[18] G. Noguere, A. Brusegan, A. Leprêtre, N. Herault, O. Bouland, G. Rudolf, Private communication (2005)

[19] R.L. Macklin, Nucl. Sci. Eng. **85**, 350 (1983)

[20] G. Noguere, O. Bouland, A. Brusegan, P. Schillebeeckx, P. Siegler, A. Leprêtre, N. Herault, G. Rudolf, Phys. Rev. **C74**, 054602 (2006)