A new convention for the epithermal neutron spectrum for improving accuracy of resonance integrals

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Abstract
A new convention for the epithermal neutron spectrum component is formulated in this work, aimed at improving the accuracy of resonance integrals determination. The $(1 + \beta)/(\beta E + E^{1+\alpha})$ form here proposed, is an approximating function of the epithermal neutron spectrum based upon calculations performed by the state-of-art Monte Carlo code MVP-3. Bias effects on determination of resonance integrals, due to the application of the well-known and used so far approximating functions, such as $1/E, 1/E^{1+\alpha}$ as well as the new form $(1 + \beta)/(\beta E + E^{1+\alpha})$ are compared, where $E$ is the neutron energy, $\alpha$ a shape factor, and $\beta$ a new shape factor introduced in this work. The other bias effect is also investigated, which is caused by neglecting the position dependence of a neutron spectrum inside an irradiation capsule. To get a demonstration of the bias effects due to these assumptions, upon the determination of a neutron spectrum from a quantitative point of view in a practical case, the thermal neutron-capture cross section and resonance integral of $^{135}$Cs measured at a research reactor JRR-3 are re-evaluated. A superior property of the proposed new mathematical expression is discussed. The experimental method is proposed to determine the new shape factor $\beta$ by a combinational use of triple flux monitors ($^{197}$Au, $^{59}$Co, and $^{94}$Zr), and its analytical methodology is formulated.

1. Introduction
Neutron cross sections are the basic physical data for neutronics calculations in many apparatuses for fundamental nuclear physics research as well as applications in many fields. Since significant advancements on neutronics calculations have been achieved [1–3] by utilizing a Monte Carlo method [4] coupled to high performance computing systems, the accuracy of neutron cross sections is thus becoming a dominant factor determining the final results and their uncertainties of calculations. That being said, needs for improvements in the accuracy on neutron cross sections are expanding as also summarized by the OECD/NEA’s working group WPEC/SG-C [5].

To bridge the gap between the required and current accuracy in nuclear data, national or international research projects have been conducted worldwide [6–8]. As an example, WPEC/SG-41 tackled an accuracy improvement on neutron-capture cross section of $^{241}$Am by integrating the knowledge so far available on both differential and integral measurements, and pointed importance of identifying bias effects and correcting them before evaluating a recommended value [9].

The neutron activation method is one of the integral measurement approaches commonly available, which has been historically and widely utilized to get measurements on the capture cross sections for thermal neutrons as well as determination for resonance integrals [10–13]. Therefore, these cross sections in evaluated nuclear data sets such as BNL-325 [14] and Atlas [15] are basically influenced by the specific neutron activation measurement method of choice, as well as the analysis methods adopted. Recently, an identified bias effect has been examined in details in the case of $^{241}$Am [16, 17] about the thermal neutron-capture cross section measured by means of activation method, where a huge resonance does exist at energy of 0.3 eV, which is lower than the Cd
cut-off energy (i.e. about 0.5 eV). Although a substantial bias on resonance integrals was also pointed out in [17], the correction method developed in [17] is limited for determination of thermal neutron-capture cross sections only. In order to investigate bias effects on resonance integrals as well, the shape of neutron spectrum in the epithermal energy region is therefore important, as also expected by the resonance integral definition itself. In this paper, the bias effects, originating in the assumption on a neutron spectrum mainly in the epithermal energy region, is examined in detail.

The best known method about the neutron activation analysis is the one proposed by Westcott et al [18], where the neutron spectrum is approximated by the sum of Maxwellian and 1/E distributions. The energy-dependent function is expressed as follows:

\[
\phi(E) = \phi_{th}(E) + \phi_{epi}(E) = \phi_1 \cdot \frac{2}{\sqrt{\pi}} \cdot \frac{T}{T_0} \cdot \frac{E}{(kT)^2} \cdot e^{-E/kT} + \phi_2 \cdot \frac{\Delta(E)}{E},
\]

where \(E\) is the neutron energy, \(T\) the temperature of neutrons defined by the Maxwellian distribution in Kelvin (K), \(k\) the Boltzmann constant \((k \equiv 1.380 \times 10^{-23} \text{ J K}^{-1} = 8.617 \times 10^{-5} \text{ eV K}^{-1})\), and \(\Delta(E)\) is a so-called ‘joining’ function approximated by a step function that rises up at an energy point of about 5 kT. The fast neutron component is neglected in the Westcott convention and in this paper is discussed in the next section. The \(\phi_1\) and \(\phi_2\) are constants expressing intensities of thermal and epithermal neutron flux components. As a joining function, several forms have been proposed in the last decades, and their validity will be examined later in this paper.

Ryves [19] introduced a more sophisticated function, of \(1/E^{1+\alpha}\) kind to account for the epithermal neutron spectrum component with a shape factor \(\alpha\), which is expressed as:

\[
\phi_{epi}(E, \alpha) = \phi_2 \cdot \frac{\Delta(E)}{E} \cdot \left(\frac{E_{\text{ref}}}{E}\right)^\alpha.
\]

This approach has been investigated and utilized in application fields of neutron activation analysis (NAA) [20, 21]. However, such a \(1/E^{1+\alpha}\) form has not become popular in deducing resonance integrals as pointed out by Yücel et al [22].

In the neutron activation analysis, not only the spectrum shape but also its position dependence needs to be considered, as pointed in [11]. However, the difference of neutron flux levels measured between flux monitors and an irradiation sample has been assumed to be negligible in most cases or fully neglected.

In this paper these bias effects are evaluated by deducing neutron-capture cross sections, especially resonance integrals, from the quantitative point of view. In this regard, the neutron spectrum calculated by a continuous-energy Monte Carlo code MVP-3 [1] is used as a reference. Better approximating functions have thus been extracted for the joining energy range and the epithermal energy region, by using the reference spectrum. A new formulation for reaction rate calculations has thus been drawn by using the derived approximating functions: the new formula enables to analyze experimental data with reduced bias. In order to get demonstration on how the bias effects, originating in the assumptions on a neutron spectrum, have an impact in a practical case, the experimental data concerning the \(^{135}\text{Cs}\) nuclide irradiated at the JRR-3 have been re-evaluated. A method determining shape factors in the approximating function is here proposed and examined.

2. Calculations

2.1. Calculations of reference spectrum and its position dependence

In order to obtain the reference spectrum at the irradiation position where thermal neutron-capture cross sections and resonance integrals have been measured, the irradiation hole HR-1 (Hydraulic rabbit facility No. 1) in the JRR-3 (Japan Research Reactor No. 3) located at Tokai in Japan has been selected, and the neutron spectrum there available has been calculated by a Monte Carlo particle transport code MVP-3. Great care has been put in 3D geometry reconstruction in the input file of the reactor core, the irradiation hole selected, and the irradiation capsule as shown in figure 1. The fuel composition at the irradiation date has been calculated by using the burnup calculation code MVP-BURN [23] and the information incorporated in the input of the MVP-3 [24]. The temperature of the core region has been set to 334 K, while for the D_2O filled spectrum shifter volume surrounding it being 303 K, which approximated the averaged values in the core and moderator regions calculated with the condition of the 20 MW (thermal) reactor power when the experiment was performed. Since an irradiation sample and flux monitors cannot be set simultaneously at the same position in actual experimental conditions (see [25] for example), a bias will emerge due to a position dependence of the neutron flux inside an irradiation capsule. In order to get an assessment about such a bias effect from the quantitative point of view, neutron spectra inside the irradiation capsule have been calculated, as a function of a position \(X\) inside the capsule.
In figure 2(a) the calculated neutron spectra, expected at the different irradiation positions inside the capsule set in the HR-1 hole is shown. The dependence of the neutron spectra is shown for position coordinates \( X = (-10, 0) \) (i.e. the center position in the capsule), \((+10, 0)\), where the direction of \( X \) axis is sketched in figure 1. The unit of vertical axis of figure 2(a) is chosen to be a neutron flux per unit lethargy. Based upon such an axis unit setting, an epithermal neutron flux trend of form \( 1/E \) in equation (1) or \( \alpha = 0 \) in equation (2) has a constant value when plotted function of neutron energy. In figure 2(a), an apparent discrepancy from the constant expectation, that is, the decrease of neutron flux per unit lethargy as a function of energy, is shown at the epithermal region.

Figure 2(b) shows the ratios of neutron fluxes at the different positions. A plain difference on the neutron flux level, mainly epithermal region, is observed, while the difference for thermal neutron region around 0.01–0.1 eV being not so large. The irradiation position dependences of the integral of the thermal and epithermal neutrons are plotted in figure 3. Differences of the integrals of the neutron flux intensities are estimated to be about 0.14% and 0.8% per 1 mm shift in position for thermal and epithermal neutrons, respectively.

Position dependent neutron spectra from \( X = -10 \) mm to \(+10 \) mm calculated by 2 mm steps are summed up for improving the statistical uncertainty. This averaged spectrum is defined as the reference spectrum \( \phi_{\text{ref}}^{\text{E,ref}} \) which has been used for extracting the appropriate approximating functions. The epithermal neutron flux integrated over an energy region, ranging from 0.5 eV to 100 keV, is normalized to match the experimental value.

2.2. Effect of fast neutrons on activation

Contribution of fast neutrons (\( E > 100 \) keV) to the total reaction rate is evaluated by using the reference spectrum. Total and fast reaction rates are expressed by the equations,

\[
R_{\text{tot}} = \int_0^\infty \sigma(E) \cdot \phi_{\text{ref}}^{\text{E,ref}}(E) \cdot dE
\]

\[
R_{\text{fast}} = \int_{100 \text{ keV}}^\infty \sigma(E) \cdot \phi_{\text{ref}}^{\text{E,ref}}(E) \cdot dE
\]

The ratio of \( R_{\text{fast}} \) and \( R_{\text{tot}} \) is calculated using JENDL-4.0 nuclear data library [26] for several isotopes, and shown in table 1, along with their thermal neutron-capture cross sections \( \sigma_0 \), resonance integrals \( I_0 \), capture cross sections averaged over a Maxwellian spectrum peaking at a neutron energy of 30 keV \( \sigma_{30 \text{ keV}} \), and the ratios \( \sigma_{50 \text{ keV}} / \sigma_0 \). For comparison, those of ENDF/B-VIII.0 library [27] calculated by the nuclear data processing codes FRENDY [28] and PREPRO [29] are also tabulated. As shown, contributions of fast neutrons to total reaction rate are less than 0.01% except the case of \(^{94}\text{Zr} \) as being about 0.3%. As these examples show, the contributions of fast neutrons can be neglected in the cases of capture reactions if the accuracy less than about 0.3% is not required. It should be noted that a careful evaluation is required in the cases treating fission and threshold reactions, since these cross sections do not decrease as a function of neutron energy at fast neutron region in contrast to the capture reaction.
2.3. Approximating functions for joining and epithermal energy regions

In order to get an approximation of the epithermal neutron spectrum component, the equations (1) and (2) have been widely utilized. Several types of Δ(E) function forms in equations (1) and (2) have been proposed [18], which is reported as a 'cut-off' function in some [10] or a 'joining' function in other [17] and in this work, as well. In order to extract approximating functions for both the joining part and epithermal energy regions from the reference spectrum, a main Maxwellian distribution is subtracted by the reference spectrum. A thermal energy region below 0.125 eV (∼5k T) has been considered to get a fitting by a Maxwellian function for the Eth component, as reported in equation (1). The fitted Maxwellian curve is shown by a solid line, and the reference spectrum minus the Maxwellian component are plotted by open circles in figure 4(a). The latter spectrum is defined as 'the reference epithermal spectrum' hereafter. As shown, the reference spectrum is well fitted at thermal energy region with a Maxwellian function; a fitting gives a value of 27.28 meV for kT with an uncertainty as small as 0.02 meV, which corresponds to 316.5 (3) K. The digit in parentheses is the one-standard-deviation uncertainty in the last digits of the given value.

In figure 4(b), the fitting by a 1/E1+α form (blue solid line) can reproduce the spectrum much better than that by a 1/E form (dashed black line). The fitting function is expressed by:

$$\phi_{\text{cp}}(E) = \phi_{2} \cdot \frac{\Delta_{\phi}(E)}{E} \cdot \left(\frac{E_{\text{ref}}}{E}\right)^{\alpha},$$

where Δ2 is expressed as below:

$$\Delta_{\phi}(E) = \frac{1}{1 + \left(\frac{4.99 \text{ kT}}{E}\right)^{7}}.$$  

The fitted value of α is 0.0959 (15) and that of φ2 is 1.636 (12) × 10^{12} [1/sec/cm²].

Figure 2. (a). Neutron spectra inside an irradiation capsule inserted in the HR-1 irradiation hole of JRR-3 reactor. The position dependence of the neutron spectrum in the capsule is shown for the cases of X = −10 (Black), 0 (Red), +10 (Green) [mm], where X is the coordinate position inside the capsule on the axis connecting the core and the capsule as sketched in figure 1: the coordinate X at the center of the capsule is set as 0 [mm]. (b). The ratio of neutron spectra calculated at the position coordinates X = −10 and X = 0 positions (Black circles) and that between X = +10 and X = 0 positions (Green triangles). The red line showing the constant value equal to 1 is for eye guide.
The residuals divided by the fitted fluxes are plotted in the bottom of the figure 4(b) for $\Delta_{\gamma}(E) / \beta^{+} \alpha$ (blue circles). Some noticeable deviations are present in the short energy range at about 0.2–0.3 eV, and in a wide energy region, ranging from a few eV to 100 keV even if equation $(5)$ is used. The deviation at around 0.2–0.3 eV is expected to be reduced by allowing adjustment of the parameters in the joining function. However, the deviation observed in the wide energy range from a few eV to 100 keV cannot be reduced, if any new free parameter other than $\alpha$ is not allowed.

Trkov et al $[30]$ proposed some years ago the same formula $1 / \beta^{+} \alpha$ in which the parameter $\alpha$ is allowed to be energy-dependent as expressed by the mathematical relation $\alpha(E) = \alpha_0 + \alpha_1 \times \ln(E) + \alpha_2 \times \ln^2(E)$. However, it is anticipated here that is difficult to determine three parameters from the limited experimental values of reaction rates or reaction rate ratios. In order to get an as practical as possible fitting operation for an energy region from a few eV to 100 keV, trial functional forms allowing only one additional free parameter have been investigated instead. The additional parameter is added to the denominator in order to allow for a function shape degree of freedom. This is expressed by the $1/(\beta^{+} \alpha)$ form function. For normalization purpose, the

![Figure 3. The irradiation position dependences of thermal ($E < 0.5$ eV) and epithermal ($E > 0.5$ eV) neutron fluxes inside the irradiation capsule, as a function of position coordinate $X$. Epithermal neutron flux (open circles) is multiplied by a factor 7 to compare position dependence with thermal neutron flux (closed circles) in the same scale.]

| Isotopes | Library | $\sigma_0$ [b] | g-factor | $I_0$ [b] | $\sigma_{20 \text{ keV}}$ [mb] | $\sigma_{50 \text{ keV}} / \sigma_0$ | $R_{\text{fast}} / R_{\text{tot}}$ |
|----------|---------|----------------|----------|---------|-----------------|----------------|-----------------|
| $^{197}\text{Au}$ | Atlas | 98.65 (9) | 1.0086 | 1550 (28) | 599 (6) | 0.61% | 0.0026% |
| | JENDL | 98.65 | 1.005 | 1571 | 608.9 | 0.62% |
| | ENDF/B | 98.70 | 1.006 | 1545 | 618.3 | 0.63% |
| $^{59}\text{Co}$ | Atlas | 37.18 (6) | 1.004 | 75.8 (20) | 38 (3) | 0.10% | 0.0005% |
| | JENDL | 37.21 | 1.001 | 75.85 | 39.18 | 0.11% |
| | ENDF/B | 37.18 | 1.001 | 75.80 | 34.26 | 0.09% |
| $^{94}\text{Zr}$ | Atlas | 0.0511 (17) | 1.004 | 0.28 (1) | 26.4 (10) | 51.7% | 0.3356% |
| | JENDL | 0.0507 | 1.001 | 0.287 | 26.12 | 51.5% |
| | ENDF/B | 0.0500 | 1.001 | 0.314 | 27.73 | 55.3% |
| $^{135}\text{Cs}$ | Atlas | 8.3 (3) | 0.990 | 38.1 (26) | 169 (11) | 2.04% | 0.0095% |
| | JENDL | 8.30 | 1.000 | 53.5 | 162.9 | 1.96% |
| | ENDF/B | 8.66 | 0.998 | 50.8 | 200.9 | 2.32% |
| $^{241}\text{Am}$ | Atlas | 720 (20) | 1.0021 | 1425 (100) | 2164 | 0.30% | 0.0016% |
| | JENDL | 684.3 | 1.0105 | 1588 | 2394 | 0.35% |
| | ENDF/B | 684.3 | 1.0105 | 1591 | 2467 | 0.36% |
factor $(1 + \beta)$ is multiplied in the numerator in order to get the condition $(1 + \beta)/(\beta E + E^{1+\alpha}) = 1$ when $E = 1$ [eV]. The $\beta$ dependence of the new $(1 + \beta)/(\beta E + E^{1+\alpha})$ function is plotted in figure 4(e) and, for comparison, the $\alpha_1$ dependence of the $1/E^{1+\alpha_1} \times \text{IntE}^\beta$ form function is also plotted in figure 4(f). The parameter $\alpha_1$ has been set to a fixed value of zero in order to limit the number of free fitting parameters. As shown in figure 4(e) the parameter $\beta$ affects the neutron spectrum shape nearly equally in a wide energy range, while the parameter $\alpha_1$ is more sensitive to high energy region as being the logarithmic coefficient for the neutron energy $E$. The resulted best fitted spectra have at last been calculated by using the set of adjusted parameters $(\alpha, \beta) = (0.221, 4.21)$, in the case of the $(1 + \beta)/(\beta E + E^{1+\alpha})$ form function and the $(\alpha_0, \alpha_1) = (0.0373, 0.00564)$ in the case of the $1/E^{1+\alpha_0+\alpha_1} \times \text{IntE}^\beta$ form function under the restriction of $\alpha_2 = 0$. This paper therefore focuses on the utilization of the new $1/(\beta E + E^{1+\alpha})$ form function, and comparison with the alternative $1/E^{1+\alpha_0+\alpha_1} \times \text{IntE}^\beta$ function is discussed as well.

In figure 4(b), the fitting by the $(1 + \beta)/(\beta E + E^{1+\alpha})$ form epithermal spectrum multiplied by the $\Delta_4^{\text{Free}}$ is also shown by solid line; the resulting fitting expression is expressed as below.

$$\phi_{\text{epi}}(E) = \phi_2 \frac{\Delta_4^{\text{Free}}}{E} \cdot \frac{1 + \beta}{\beta + E^\beta},$$

where the function $\Delta_4^{\text{Free}}$ is expressed by introducing four additional free parameters $m_1, m_2, \mu_1, \mu_2$ instead of $\Delta_4^{\text{Orig}}$ given in [18], as reported below.

$$\Delta_4^{\text{Free}}(E, T, \mu_1, \mu_2, m_1, m_2) = \frac{1}{1 - \frac{0.26}{1 + \frac{T}{\mu_1}}} + \left(\frac{\mu_1 E}{E}\right)^{m_1}.$$  

The new shape factor $\beta$ is introduced in this work with the aim to achieve a superior fitting. The fitted values for $\phi_2, \alpha, \beta, m_1, m_2, \mu_1, \mu_2$ are 1.466 (16) $\times$ 10^{-15}, 0.221 (11), 4.21 (64), 1.93 (18), 5.88 (28), 26.1 (29), 5.06 (4),
Table 2. Fitted parameters for six mathematical forms of approximating functions considered in this work. The $m_1$, $m_2$, $\mu_1$, $\mu_2$ parameters are given values for 1–4 function forms, while those for 5 and 6 function forms are instead treated as free parameters. Uncertainties obtained by the fitting are shown inside parentheses.

| Function ID | 1 $\Delta_1(E)/E$ | 2 $\Delta_2(E)/E^{1+\alpha}$ | 3 $\Delta_{\text{Orig}}(E)/E^{1+\alpha}$ | 4 $\Delta_{\text{Orig}}(E)1 + \beta/\beta + E^\alpha$ | 5 $\Delta_{\text{Free}}(E)1 + \beta/\beta + E^\alpha$ | 6 $\Delta_{\text{Free}}(E)1/E^{\ln(E+\alpha)+\ln(E)}$ |
|-------------|------------------|-------------------------------|---------------------------------|---------------------------------|---------------------------------|---------------------------------|
| $\phi_2 \times 10^{-12} [1/cm^2/s]$ | 1.093 (39) | 1.637 (12) | 1.465 (16) | 1.469 (12) | 1.466 (16) | 1.470 (17) |
| $\alpha$ or $\alpha_2$ | — | 0.0995 (15) | 0.0780 (26) | 0.244 (14) | 0.221 (11) | 0.0396 (48) |
| $\beta$ or $\alpha_1$ | — | — | — | 5.44 (89) | 4.21 (64) | 0.0054 (4) |
| $m_1$ | — | — | 5.00$^a$ | 5.00$^a$ | 1.93 (18) | 2.03 (20) |
| $m_2$ | 7.00$^a$ | 7.00$^a$ | 7.00$^a$ | 7.00$^a$ | 5.88 (28) | 5.92 (30) |
| $\mu_1$ | — | — | 16.4$^a$ | 16.4$^a$ | 26.1 (29) | 25.7 (30) |
| $\mu_2$ | 4.95$^a$ | 4.95$^a$ | 4.75$^a$ | 4.75$^a$ | 5.06 (4) | 5.05 (5) |

$^a$ Fixed value.
respectively. The values of \( m_0, m_1, \mu_1, \mu_2 \) differ much from the original fixed values 5.00, 7.00, 16.4, 4.75 given in [18], respectively. The free parameters \( m_2 \) and \( \mu_2 \) determine the sharpness and position of the rising edge shown in figure 4(b) at around 0.13 eV. The \( m_1 \) and \( \mu_1 \) instead determine the falling edge sharpness and the position of the bump shown in figure 4(b) at around 0.2−0.3 eV. The original values of \( m_1, m_2, \mu_1, \mu_2 \) make a too sharp bump at around 0.2−0.3 eV, and is not suitable to reproduce the \( \phi_{\text{ep}}^{\text{cal}}(E) \). Since the parameters \( m_1, m_2, \mu_1, \mu_2 \) are insensitive to the shape of neutron spectrum above about 2 eV, the gross shape of epithermal neutron spectrum is mainly determined by the shape parameters \( \alpha \) and \( \beta \). The resultant joining functions are compared in figure 4(c). The fitted values of \( \phi_2, \alpha, \beta \) obtained by fitting the \( \phi_{\text{ep}}^{\text{ref}}(E) \) with each combination of approximating functions are summarized in table 2.

The reference spectrum available within a Cd capsule has been calculated by the MVP code in the same manner followed for the calculation of \( \phi_{\text{ep}}^{\text{ref}}(E) \), which is shown by closed circles in figure 4(d). The \( \phi_{\text{ep}}^{\text{ref}}(E) \) spectrum multiplied by an attenuation factor by a Cd capsule is also plotted in the figure: triangle symbols are calculations with the effective Cd thickness of 1.4 mm, and square with that of 2.2 mm. In the calculation of the attenuation factor a scattering cross section is neglected and the capture cross section is approximated by that of \( ^{113}\text{Cd} \) multiplied by its isotopic abundance ratio 0.1222, since this cross section is dominant in all stable Cd isotopes. The equation for transmission is expressed by

\[
\text{Tr}(E) = e^{-\sigma_{\text{cap}}(E)n_{\text{Cd}}}^{\ell}
\]

where \( n_{\text{Cd}} \) is an atomic density of \( ^{113}\text{Cd} \), \( \sigma_{\text{cap}}(E) \) the capture cross section of \( ^{113}\text{Cd} \), and \( \ell \) the adjusted effective Cd thickness. The size and thickness of the Cd capsule given in [25] are reproduced in the geometry input of the MVP code.

### 2.4. Reaction rate expression using the generalized approximating functions

Based upon the aforementioned considerations, in the following the most general expression for reaction rate determination is formulated, by taking into account the neutron flux approximating functions discussed in the previous section.

By inserting equation (7) as \( \phi_{\text{ep}} \) into equation (1), the reaction rate \( R \) in equation (3) is expressed as

\[
R = \int_0^\infty \sigma(E) \cdot (\phi_{\text{th}}(E) + \phi_{\text{ep}}(E)) \cdot dE
\]

\[
= \phi_1 \cdot \int_0^\infty \sigma(E) \cdot \left( \frac{2}{\sqrt{\pi}} \cdot \sqrt{\frac{T}{T_0}} \cdot \frac{E}{(kT)^2} \cdot e^{-\frac{E}{kT}} \right) \cdot dE
\]

\[
+ \phi_2 \cdot \int_0^\infty \sigma(E) \cdot \left( \frac{\Delta_{\text{free}}^{\text{cd}}(E, T, \mu_1, m_1, \mu_2, m_2)}{E} \cdot 1 + \frac{\beta}{\beta + E^{\alpha}} \right) \cdot dE
\]

\[
= \phi_1 \cdot g_{w}(T) \cdot \sigma_0 + \phi_2 \cdot \int_0^\infty \sigma(E) \cdot \left( \frac{\Delta_{\text{free}}^{\text{cd}}(E, T, \mu_1, m_1, \mu_2, m_2)}{E} \cdot \frac{1 + \beta}{\beta + E^{\alpha}} \right) \cdot dE,
\]

where \( g_{w}(T) \) is the Westcott factor depending only on neutron temperature \( T \). If we define a quantity \( H_{\text{join}} \) by the equation

\[
H_{\text{join}}(T, \alpha, \beta, \mu_1, m_1, \mu_2, m_2) = \int_0^\infty \sigma(E) \cdot \left( \frac{\Delta_{\text{free}}^{\text{cd}}(E, T, \mu_1, m_1, \mu_2, m_2)}{E} \cdot \frac{1 + \beta}{\beta + E^{\alpha}} \right) \cdot dE,
\]

the reaction rate \( R \) can be expressed as

\[
R = \phi_1 \cdot g_{w}(T) \cdot \sigma_0 + \phi_2 \cdot H_{\text{join}}(T, \alpha, \beta, \mu_1, m_1, \mu_2, m_2).
\]

The quantity \( H_{\text{join}} \) reflects the shape of an epithermal neutron spectrum. As shown in figure 4(c), it corresponds not to a resonance integral with a Cd cut-off energy of about 0.55 eV, but to a resonance integral with a cut-off energy of about 5kT, which is determined by the joining function \( \Delta^{\text{free}}_{\text{cd}}(E, T, \mu_1, m_1, \mu_2, m_2) \). This formula is also different from the Høgdahl convention [31], where an epithermal neutron component in an energy region below 0.55 eV is assumed to be neglected.

A reaction rate above 0.55 eV may be experimentally measured by means of the activation technique by irradiating a sample within a Cd capsule. The reaction rate \( R' \) within a Cd capsule is expressed by

\[
R' = \phi_2 \cdot H^{\text{cd}}(T, \alpha, \beta, \mu_1, m_1, \mu_2, m_2).
\]
3. Discussions on the bias effects

3.1. Biases due to assumption on the shape of neutron spectrum

In order to evaluate biases due to assumptions on neutron spectrum, reaction rates within a Cd capsule, $R'$ (fitted approximating functions), are compared with $R$ (MVP). For the calculations of the $R'$ (fitted approximating function), the fitted parameters reported in table 2 are used. Table 4 shows results of calculated $R'$ for $^{197}$Au, $^{59}$Co, $^{94}$Zr, $^{135}$Cs and $^{241}$Am isotopes. The absolute value of the $R'$ (MVP) is normalized to 32.90 $[10^{-10}/s]$ for $^{59}$Co to match with experimental data reported in section 4 for allowing direct comparison.

The results of $R$ and $R'$ are also tabulated. The $R$ and $R'$ are determined mainly by a Maxwellian component and partly by a component in an energy region of about 0.1–1 eV. As shown, the value of $R$–$R'$ calculated by any fitting function reproduces that by the reference spectrum within 0.5%, except for the form 1 function applied to $^{241}$Am: this is the special case where a huge resonance does exist at 0.3 eV as studied in $^{59}$Co to match with experimental data reported in section 4 for allowing direct comparison.
Table 4. Reaction rates $R$ (without the Cd shield), $R'$ (with the Cd shield), and its difference $R-R'$ for five different isotopes ($^{197}$Au, $^{59}$Co, $^{94}$Zr, $^{135}$Cs, $^{241}$Am) calculated by using the reference spectrum and the five different forms of the approximating function considered. Are also reported comparison with experimental data by Katoh et al [25] for $^{197}$Au, $^{59}$Co, $^{135}$Cs isotopes.

| Isotopes | Rates [$10^{-10}$/s] | Function Type | ID | Ref. | 1 | 2 | 3 | 4 | 5 | 6 | Exp |
|----------|----------------------|----------------|-----|------|---|---|---|---|---|---|-----|
| $^{197}$Au | $R$ | 105.1 | 101.3 | 106.1 | 104.6 | 105.6 | 105.6 | 105.5 | 100.3 | (2) |
| $R'$ | 19.63 | 16.00 | 20.41 | 18.82 | 19.87 | 19.86 | 19.85 | — | 20.0 | (4) |
| $R'/R$ (MVP) | 1 | 0.815 | 1.040 | 0.959 | 1.012 | 1.011 | 1.011 | — | — | — |
| $R-R'$ | 85.48 | 85.33 | 85.70 | 85.80 | 85.74 | 85.70 | 85.69 | 80.3 | (4) | — |
| $^{59}$Co | R | 32.90 | 32.72 | 32.86 | 32.87 | 32.90 | 32.88 | 32.88 | 32.9 | (6) |
| $R'$ | 0.788 | 0.760 | 0.779 | 0.749 | 0.800 | 0.800 | 0.797 | 0.825 | (15) | — |
| $R'/R$ (MVP) | 1 | 0.965 | 0.989 | 0.951 | 1.016 | 1.016 | — | — | — | — |
| $R-R'$ | 32.11 | 31.96 | 32.08 | 32.12 | 32.10 | 32.08 | 32.08 | 32.1 | (6) | — |
| $^{94}$Zr | R | 0.0456 | 0.0464 | 0.0457 | 0.0458 | 0.0456 | 0.0456 | — | — | — |
| $R'$ | 0.0019 | 0.0029 | 0.0020 | 0.0020 | 0.0019 | 0.0019 | 0.0019 | — | — | — |
| $R'/R$ (MVP) | 1 | 1.540 | 1.045 | 1.080 | 0.986 | 1.002 | — | 1.001 | — | — |
| $R-R'$ | 0.0437 | 0.0435 | 0.0437 | 0.0437 | 0.0437 | 0.0437 | 0.0437 | — | — | — |
| $^{135}$Cs | R | 7.72 | 7.67 | 7.70 | 7.69 | 7.73 | 7.72 | 7.72 | 7.55 | (3) |
| $R'$ | 0.556 | 0.542 | 0.550 | 0.529 | 0.569 | 0.564 | 0.562 | 0.464 | (1) | — |
| $R'/R$ (MVP) | 1 | 0.975 | 0.989 | 0.951 | 1.024 | 1.015 | 1.011 | — | — | — |
| $R-R'$ | 7.16 | 7.13 | 7.15 | 7.16 | 7.16 | 7.15 | 7.15 | 7.09 | (3) | — |
| $^{241}$Am | R | 643.4 | 623.8 | 642.0 | 642.3 | 642.3 | 642.3 | 642.1 | 642.9 | — |
| $R'$ | 18.2 | 13.2 | 18.4 | 17.3 | 17.3 | 18.3 | 18.3 | — | — | — |
| $R'/R$ (MVP) | 1 | 0.724 | 1.013 | 0.938 | 0.950 | 1.007 | 1.004 | — | — | — |
| $R-R'$ | 625.2 | 610.7 | 626.7 | 625.0 | 625.0 | 624.7 | 624.7 | 624.6 | — | — |

Note: Absolute values of reaction rates in table 3 are normalized as the reaction rate of $^{59}$Co being $32.9 \times 10^{-10}$/s for the function type 5.

Energy mesh of $\Delta u = 0.1133$ (250 group constants) is used for calculations of reaction rates using the reference spectra and five kinds of fitted functions.
to be considered in deducing resonance integrals. For examples, the ratio for $^{197}$Au is 20% smaller than that for $^{135}$Cs. If $^{197}$Au is used as a flux monitor to measure the resonance integral of $^{135}$Cs, the bias of about 20% needs to be considered. On the other hand, if a $^{59}$Co is used as the flux monitor, the bias will be as small as 1%. As demonstrated here, the detailed study of the neutron spectrum in advance of activation experiments will help to select a suitable flux monitor.

In case of function of the form 2 ($1/\sqrt{E + 1}$ in an epithermal region), the range of the ratios is limited within 0.99 to 1.04. It is 0.94 to 1.08 for function of the form 3, and 0.95 to 1.02 for function of the form 4. It should be noted that the difference of about 5% exists between the ratios of $^{197}$Au and $^{59}$Co in the case of function of the form 2, although both are commonly used as standards for neutron activation measurements. In these cases, a bias of about 5% need to be considered.

On the other hand, the function of the form 5 introduced in this paper significantly reduces the range: from 1.002 to 1.016. A bias need to be considered in such a case, which is much reduced compared to the function of the form 1 to 4. The difference between the ratios of $^{135}$Cs and $^{59}$Co is low as 0.1%. If a $^{59}$Co is used as the flux monitor to measure the resonance integral for $^{135}$Cs, the bias due to the assumption of the approximating function of the form 5 will be expected as low as 0.1%. The function of the form 6, that is, $\Delta R(E)/E^{1/2} \ln(E)$ also reduces the range very well: from 1.004 to 1.011.

The difference of $R'$ on $^{241}$Am is as high as 5% between the form 4 and the form 5. This difference can be explained by the huge resonance located at about 0.3 eV in the capture cross section of $^{241}$Am. The difference of the form 4 and the form 5 is significant at the energy region of about 0.2–0.5 eV as shown in figure 4(c). If the uncertainty of resonance integrals is required to be less than 1%, it is thus to be considered the form 5 or 6 as the approximating function. In addition to $^{241}$Am, there are several isotopes, whose resonances exist in this region, such as $^{239,241}$Pu, $^{237}$Np, $^{242}$Am, $^{231,232}$Pa, $^{251}$Cf, $^{151}$Eu, $^{151}$Sm, $^{167}$Er. For these isotopes, the bias pointed here will have a value to be investigated.

### 3.2. A bias due to position dependence of neutron flux

As shown in figure 3, an intensity of an epithermal neutron flux changes about 0.8% per 1 mm. For example, in a case that a position difference between a sample and a flux monitor is 4 mm, about 3% bias needs to be considered in determining a resonance integral deduced from $R'$. On the other hand, a bias in $R - R'$, which is almost proportional to a thermal neutron-capture cross section, is limited to about 0.6% even if there is the same position difference of 4 mm.
By setting more than two flux monitors for each isotope placed at symmetric positions, sandwiching a sample and averaging reaction rates of these flux monitors, the bias due to the position dependence is cancelled: amounts of flux monitors are assumed enough small not to induce any effect on the neutron flux at a sample position. This position dependence needs to be considered in planning experiments including choice and setting of sample and averaging reaction rates of these amounts of monitors. Based on such an approach, the two ratios $R_{\alpha}^{(197\text{Au})}/R_{\alpha}^{(94\text{Zr})}$ and $R_{\beta}^{(197\text{Au})}/R_{\beta}^{(98\text{Mo})}$ are calculated by

$$
\left( \frac{197\text{Au}}{197\text{Au}'} \right)/\left( \frac{94\text{Zr}}{94\text{Zr}'} \right) \quad \text{and} \quad \left( \frac{197\text{Au}}{197\text{Au}'} \right)/\left( \frac{98\text{Mo}}{98\text{Mo}'} \right)
$$

The absolute values of $R_{\alpha}^{(197\text{Au})}/R_{\alpha}^{(94\text{Zr})}$ are normalized to unity at $\alpha = 0$. The normalization factors are $1/21$, $1/227$, and $1/3515$, respectively.

By setting more than two flux monitors for each isotope placed at symmetric positions, sandwiching a sample and averaging reaction rates of these flux monitors, the bias due to the position dependence is cancelled: amounts of flux monitors are assumed enough small not to induce any effect on the neutron flux at a sample position. This position dependence needs to be considered in planning experiments including choice and setting of flux monitors. The use of a mixed alloy such as a Zr–Au–Lu $^{32}$ will be useful in planning a set of plural multi-flux monitors inside an irradiation capsule. Nevertheless, a difference of fluxes at a target sample and a flux monitor positions needs to be considered. The use of a homogeneously mixed alloy including both a target sample and flux monitors may decrease the bias due to position dependence, if atomic and isotopic ratios in the mixed alloy can be determined with a required accuracy.

3.3. Determination of an $\alpha$-shape factor

In this section, the relation between an $\alpha$-shape factor and reaction rate ratios between $^{197}\text{Au}$ and $^{59}\text{Co}$ within a Cd capsule is examined. Using this relation, one can estimate an $\alpha$-shape factor in an experiment if $R_{\alpha}^{(197\text{Au})}$ and $R_{\alpha}^{(94\text{Zr})}$ are given. As an example, in case of experiments by Katoh et al $^{[25]}$ in which the ratio $R_{\alpha}^{(197\text{Au})}/R_{\alpha}^{(98\text{Mo})}$ is given as 24.2 (7), one can roughly estimate the alpha shape factor as 0.060 (13).

Yücel et al derived an $\alpha$-shape factor from the ratio of the $^{197}\text{Au}$ to $^{98}\text{Mo}$ reaction rates in a Cd capsule and claimed that the $\alpha$-shape factor can be determined experimentally. Figure 6 shows that the $R_{\alpha}^{(197\text{Au})}/R_{\alpha}^{(98\text{Mo})}$ can be used to determine the $\alpha$-shape factor, as well as the combinational use of $^{197}\text{Au}$ and $^{98}\text{Mo}$. If $^{94}\text{Zr}$ is used as a second monitor in combination with $^{197}\text{Au}$ and its $R_{\alpha}^{(197\text{Au})}$ is measured with the same uncertainty, the $\alpha$-shape factor will be determined with higher accuracy since the sensitivity of the ratio on $\alpha$-shape factor is much higher than those of other combinations as shown in figure 6.

3.4. Determination of both $\alpha$ and $\beta$-shape factors

This section describes the method determining both $\alpha$ and $\beta$-shape factors by a combinational use of the triple flux monitors, $^{197}\text{Au}$, $^{59}\text{Co}$, and $^{94}\text{Zr}$. We assume reaction rates $R_{\alpha}$ in a Cd capsule are available experimentally for all monitors. Based on such an approach, the two ratios $R_{\alpha}^{(197\text{Au})}/R_{\alpha}^{(98\text{Mo})}$ and $R_{\beta}^{(94\text{Zr})}/R_{\beta}^{(98\text{Mo})}$ are deduced, which are defined here as $a_1 \pm \delta a_1$ and $a_2 \pm \delta a_2$, respectively. On the other hand, for each set of $\alpha$ and $\beta$, these ratios are calculated by $H_{\alpha}^{\text{Cd}}(\alpha, \beta; 197\text{Au})/H_{\alpha}^{\text{Cd}}(\alpha, \beta; 59\text{Co})$ and $H_{\beta}^{\text{Cd}}(\alpha, \beta; 94\text{Zr})/H_{\beta}^{\text{Cd}}(\alpha, \beta; 98\text{Mo})$ according to equation (13), which are defined here as $f_1(\alpha, \beta)$ and $f_2(\alpha, \beta)$, respectively. Parameters $T$, $\mu_1$, $\mu_2$, $m_1$, and $m_2$ in equation (13) are omitted for short expressions. The joint probability of the set of $\alpha$ and $\beta$ occurs is expressed by the normal distribution function $W(\alpha, \beta)$, which is defined by

Figure 6. The relation between the ratio $R_{\alpha}^{(197\text{Au})}/R_{\alpha}^{(94\text{Zr})}$ and an alpha shape factor where $M = 98\text{Zr}$ (solid line), $98\text{Mo}$ (dashed line), and $94\text{Zr}$ (dotted line). The absolute values of $R_{\alpha}^{(197\text{Au})}/R_{\alpha}^{(94\text{Zr})}$ are deduced from Figure 6.
For example, in a case that \( \tilde{f}_1(\alpha, \beta) = 24.7 \pm 0.247 \) and \( \tilde{f}_2(\alpha, \beta) = 0.00250 \pm 0.000025 \) are obtained experimentally, the contour plot of the \( \Delta \chi^2 = 1 \) is obtained by numerical calculations as shown in figure 7(a). From the maximum point of the plot, the best estimates of the set of \( \alpha \) and \( \beta \) values, \((\alpha_{fit}, \beta_{fit})\), are deduced. The contour \( W = e^{-1/2} (\geq 0.6) \) is equivalent with that of \( \Delta \chi^2 = 1 \) in equation (16). From the contour \( W = e^{-1/2} \) projects onto the \( \alpha \) and \( \beta \) axes, the one standard deviations \( \delta \alpha \) and \( \delta \beta \) of \( \alpha \) and \( \beta \) are deduced, respectively [33]. The integrated joint probability for the parameter values within the contour \( \Delta \chi^2 = 1 \) equals 39%. In the contour plot, there is a strong correlation between the two parameters. The correlation coefficient can be obtained from the contour \( \Delta \chi^2 = 1 \): the contour intersects the \( \Delta \alpha \) (=\( \alpha - \alpha_{fit} \)) axis at the value \( \delta \alpha \sqrt{1 - \rho^2} \) and likewise for \( \Delta \beta (=\beta - \beta_{fit}) \) as shown in figure 7(b). The correlation coefficient can be used in the evaluation of the effect of the shape parameters on reaction rates. The correlation coefficient \( \rho \) should be used.

\[
W(\alpha, \beta) = e^{-\frac{(\Delta \alpha)^2}{2}} \times e^{-\frac{(\Delta \beta)^2}{2}} = e^{-\frac{\chi^2}{2}}. \tag{16}
\]

Figure 7. (a) A contour plot of the normal distribution function \( W(\alpha, \beta) \). (b) A contour plot of \( \Delta \chi^2 = 1 \): the contour projects onto the \( \Delta \alpha \) and \( \Delta \beta \) axes gives the values of the standard deviation \( \delta \alpha \) and \( \delta \beta \). The contour intersects each axis at a fraction \( \sqrt{1 - \rho^2} \) of the corresponding standard deviation.
together with \( \delta \alpha \) and \( \delta \beta \) in the evaluation of the uncertainties of \( H^{CD} \) and \( H^{\text{Join}} \), which propagate to the uncertainties of resonance integral and thermal neutron-capture cross section through the equations (20)–(22) formulated in section 4.1.

4. A refined formula for activation analysis and its application

4.1. Formula for activation analysis

In this section, a formula is given for activation analysis. Reaction rates \( R \) and \( R' \) are expressed as below using \( H^{CD} \) and \( H^{\text{Join}} \) defined in section 2.4.

\[
\frac{R}{g_w(T) \cdot \sigma_0} = \phi_1 + \phi_2 \cdot \frac{H^{\text{Join}}}{g_w(T) \cdot \sigma_0}
\]

(17)

\[
R' = \phi_2 \cdot H^{CD}
\]

(18)

Using equation (18), the \( H^{CD}(X) \) for a target isotope \( X \) is expressed by using the \( H^{CD}(m) \) of a monitor isotope, and experimental reaction rates \( R' \) as below

\[
H^{CD}(X) = \frac{R'(X)}{R'(m) \cdot \frac{H^{CD}(m)}{H^{CD}(X) \cdot \frac{I_0(X) \cdot k(X)}{I_0(X) \cdot k(X) \cdot \frac{R'(X)}{R'(m) \cdot \frac{H^{CD}(m)}}}}}
\]

By dividing \( H^{CD}(X) \) with \( H^{CD}(m) \), the correction factor \( k(X) \) is obtained. Then, the resonance integral is deduced by the next equation.

\[
R'(X) = \frac{I_0(X) \cdot k(X)}{I_0(X) \cdot k(X) \cdot \frac{R'(X)}{R'(m) \cdot \frac{H^{CD}(m)}}}
\]

(20)

The \( g_w(T) \cdot \sigma_0 \) can be derived by two ways: (i) a combination of \( R \) and \( R' \) for one kind of isotope monitor, (ii) a combination of \( R \) for two kind of isotopes. In the first case, it is expressed as below

\[
g_w(T, X) \cdot \sigma_0(X) = \frac{R(X) - R'(X)}{R(m) - R'(m) \cdot \frac{H^{\text{Join}}(X) \cdot \frac{I_0(X)}{I_0(X) \cdot \frac{R'(X)}{R'(m)}}}}}
\]

(21)

In the second case (for example, a combinational use of Au and Co as flux monitors), it is expressed as below

\[
g_w(T, X) \cdot \sigma_0(X) = \frac{R(X) - R'(X)}{R(m) - R'(m) \cdot \frac{H^{\text{Join}}(X) \cdot \frac{I_0(X)}{I_0(X) \cdot \frac{R'(X)}{R'(m)}}}}}
\]

(22)

4.2. Revision of the \( ^{135}\text{Cs} \) experimental data

The \( \sigma_0 \) and \( I_0 \) for \( ^{135}\text{Cs} \) measured by Katoh et al [25] are revised by applying the formula given in section 4.1. By utilizing six types of approximation functions, the \( \sigma_0 \) and \( I_0 \) are obtained. The results are summarized in table 5.

The resonance integral for \( ^{135}\text{Cs} \) relative to that for \( ^{197}\text{Au} \) fluctuates so much by changing an approximating function. On the other hand, it is almost constant in the case \( ^{59}\text{Co} \) flux monitor. This tendency is explained by the discussion in section 3.1 that small bias effect is expected in the case \( ^{59}\text{Co} \) flux monitor. It is reasonable to choose 43 (3) b, determined to be relative to that for \( ^{59}\text{Co} \), as the revised resonance integral for \( ^{135}\text{Cs} \). A 3% uncertainty due to position dependence of neutron flux is smaller than the experimental uncertainty 7% given in Katoh’s paper. Therefore, 7% uncertainty is adapted for the uncertainty of the revised value.

Regarding the thermal neutron-capture cross section, the revised value changes negligibly by a small amount, even if any approximating function is selected. This means that the traditional Westcott convention, as well as the developed method in this paper, is quite accurate for deducing thermal neutron-capture cross sections. From Katoh’s reaction rates, about 6% discrepancy of \( \sigma_0(\text{Cs}) \) is deduced between values determined relative to \( ^{197}\text{Au} \) and \( ^{59}\text{Co} \) monitors, which slightly exceeds experimental uncertainty of about 3%. If \( R(\text{Au}) \) is increased by 3% and \( R(\text{Co}) \) is decreased by the same 3% for example, both values deduced relative to \( ^{197}\text{Au} \) and \( ^{59}\text{Co} \) monitors give the same value 8.3 b, which agrees with Katoh’s value 8.3 (3) b within their uncertainties. However, there is a possibility of other unrecognized experimental bias effect; it will have a value to re-examine this difference experimentally.
Table 5. Revised capture cross sections for $^{135}$Cs for six types of approximating functions.

| ID   | Function Type   | Monitor | $\Delta_1(E)$ ($\delta_{\text{Fl}}$) | $\Delta_1(E)$ ($\delta_{\text{Fl}}$) | $\Delta_1^{\text{Exp}}(E)$ | $\Delta_1(1 + \beta)$ ($\beta < E$) | $\Delta_1^{\text{Exp}}(E)$ | $\Delta_1(1 + \beta)$ ($\beta < E$) | $\Delta_1^{\text{Exp}}(E)$ | Exp Katoh et al [25] |
|------|-----------------|---------|----------------|----------------|----------------|-------------------------------|----------------|-------------------------------|----------------|---------------------|
| $\delta_{\text{Fl}}$ | Au & Co | 1 | 37 | 46 | 44 | 43 | 44 | 44 | 38 | 38 (3) |
| | Co | 2 | 42 | 43 | 43 | 42 | 43 | 43 | 43 |
| $\delta_{\text{Fl}}$ | Au | 3 | 8.8 | 8.8 | 8.8 | 8.8 | 8.8 | 8.8 | 8.8 | 8.3 (3) |
| | Co | 4 | 8.2 | 8.3 | 8.3 | 8.3 | 8.2 | 8.2 | 8.2 |
| | Au & Co | 6 | 8.2 | 8.1 | 8.2 | 8.2 | 8.2 | 8.2 | 8.2 |

Table 6. The resonance integral of $^{135}$Cs.

| Reference year | This work 2020 | Katoh et al 1997 | Baerg et al 1958 | Nakamura et al 2019 | JENDL 4.0 2011 | Atlas 6th 2018 |
|----------------|----------------|-----------------|----------------|----------------|-----------------|----------------|
| $\delta_{\text{Fl}}$ (Au) | 43 (3) | 38 (3) | 62 (2) | 42 (3) | 53 | 38 (3) |

* The resonance integral from 0.55 eV.

In contrast to the formula for the two-flux monitor method in [12, 13], the analysis method formulated here gives the thermal neutron-capture cross section in three ways as shown in the bottom of table 5. These independent values will be effectively used to check consistence of measurements. It should be noticed that the two-flux monitor method was utilized to analyze experiments utilizing a Cd filter with open-ended cylinder shape as described in the experiments reported in [34]. In the case of a fully-closed Cd filter, it is not necessary to use the two-flux monitor method.

Recently, Nakamura et al measured the resonance integral of $^{135}$Cs at the research reactor KUR using a Gd filter [35]. Their result is 42 (3) b after correcting from their original value 45 (3) b the contribution in the energy range between 0.133 and 0.55 eV. This value is consistent with the revised value 43 (3) b in this study, as compared in table 6. However, it should be noted that their resonance integral is derived based on the traditional Westcott convention by using a combination of Au and Co flux monitors. In order to evaluate the correction factor, the neutron spectrum in the KUR irradiation position needs to be calculated using Monte Carlo simulations with detailed geometries and compositions.

The evaluated value of Atlas 6th edition [15] seems to be based on Katoh’s data. Therefore, the value should be increased to be consistent with the revised value. The evaluated value of JENDL 4.0 seems to be about 23% overestimation. The data by Baerg et al [36] was deduced relative to the resonance integral of $^{59}$Co. They used 48.6 b for that of $^{59}$Co available at the time [37], which is about 2/3 of the current evaluated data 75.85 b. After correcting the resonance integral of $^{59}$Co, their value increases to 96 b. There is an apparent discrepancy between the revised Katoh’s value and Baerg’s value. Therefore, it would not be reasonable to calculate an averaged value from these contradicted data. According to our analysis, we recommend the revised value based on Katoh’s data [25].

5. Conclusions

A new convention here proposed for the epithermal neutron spectrum component, that is, $(1 + \beta)/((\beta E + E^1 + \alpha))$ form function, with two shape factors $\alpha$ and $\beta$ is formulated aimed at improving accuracy of resonance integrals. Bias effects on determination of resonance integrals, due to the approximating functions traditionally used, having mathematical forms $1/E$, $1/E^{1/\alpha}$, as well as the new type $(1 + \beta)/((\beta E + E^1 + \alpha))$ are at last compared. The possible bias due to a $1/E$ type assumption is shown to be up to about 50% on determination of the resonance integral. The bias is shown to be reduced to the level of about 5% by introducing a $1/E^{1/\alpha}$ form function. On the other hand, by utilizing the new $(1 + \beta)/((\beta E + E^1 + \alpha))$ form function here proposed, the bias is shown to be further limited to as low as about 1%. It is also shown that the $1/E^{1/\alpha + \alpha + \alpha(1 - \ln E)}$ type function in combination with the joining function $\Delta_1^{\text{Exp}}(E)$ is effective to decrease the bias, as well.

The effectiveness of using the dual flux monitors technique, $^{197}$Au and $^{59}$Co, is discussed to determine an $\alpha$-shape factor. Use of triple flux monitors, $^{197}$Au, $^{59}$Co, and $^{94}$Zr, is proposed to determine both an $\alpha$- and a $\beta$-shape factors. A bias effect, due to neglecting position dependence of neutron flux, is also examined; a possible bias up to about 3% is deduced, based upon the simulation in the case of JRR-3 research reactor, and several experimental techniques to reduce the bias effect are discussed.

---

**Table 5**. Revised capture cross sections for $^{135}$Cs for six types of approximating functions.

| ID   | Function Type   | Monitor | $\Delta_1(E)$ | $\Delta_1(E)$ | $\Delta_1^{\text{Exp}}(E)$ | $\Delta_1(1 + \beta)$ | $\Delta_1^{\text{Exp}}(E)$ | $\Delta_1(1 + \beta)$ | $\Delta_1^{\text{Exp}}(E)$ | Exp Katoh et al [25] |
|------|-----------------|---------|----------------|----------------|----------------|----------------------------|----------------|----------------------------|----------------|---------------------|
| $\delta_{\text{Fl}}$ | Au & Co | 1 | 37 | 46 | 44 | 43 | 44 | 44 | 38 | 38 (3) |
| | Co | 2 | 42 | 43 | 43 | 42 | 43 | 43 | 43 |
| $\delta_{\text{Fl}}$ | Au | 3 | 8.8 | 8.8 | 8.8 | 8.8 | 8.8 | 8.8 | 8.8 | 8.3 (3) |
| | Co | 4 | 8.2 | 8.3 | 8.3 | 8.3 | 8.2 | 8.2 | 8.2 |
| | Au & Co | 6 | 8.2 | 8.1 | 8.2 | 8.2 | 8.2 | 8.2 | 8.2 |

**Table 6**. The resonance integral of $^{135}$Cs.

| Reference year | This work 2020 | Katoh et al 1997 | Baerg et al 1958 | Nakamura et al 2019 | JENDL 4.0 2011 | Atlas 6th 2018 |
|----------------|----------------|-----------------|----------------|----------------|-----------------|----------------|
| $\delta_{\text{Fl}}$ (Au) | 43 (3) | 38 (3) | 62 (2) | 42 (3) | 53 | 38 (3) |
At last, a general formula for activation analysis is given, which enables to utilize the developed method. In order to get a demonstration of the discussed method and related impacts of the bias effects, the thermal neutron-capture cross section and resonance integral of $^{135}$Cs measured at a research reactor JRR-3 have been re-evaluated. The Katoh’s resonance integral data [23] is revised as 43 (3) b, which is 13% larger than the original value. The proposed method is expected to be widely applied to re-evaluate experimental data for resonance integral determination, and to plan an experiment aiming accuracy less than 1% for resonance integrals.

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