Space- and Time-Dependent Neutron Thermalization in Graphite by Reaction-Ratio Method

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Space- and time-dependent neutron thermalization in a 230×190×190 cm³ graphite pile has been studied by considering the reaction ratios for Gd/¹⁰B, Sm/¹⁰B, Cd/¹⁰B, Gd/Sm, Gd/Cd and Sm/Cd as spectral indicators. The reaction ratios were measured at r = 10, 30, 50 and 70 cm from the fast neutron source placed at the center of the pile. The measurements were repeated at 2.175 µsec intervals in the time range up to ~600 µsec after the fast neutron burst. Theoretical time-dependent reaction ratios were obtained from the calculated space- and time-dependent thermal neutron energy spectra by solving numerically the neutron diffusion equation, using scattering kernels based on various phonon models for graphite. From comparisons of the measured and calculated time-dependent reaction ratios, the adequacy of the various phonon models for graphite is discussed. Also, proof is given of space-dependence shown by the time-dependent thermal neutron spectra; and the relationship between the space-dependence of the time-dependent neutron spectra and that of the peaking times of the reaction ratios is made clear.

KEYWORDS: space dependence, neutron thermalization, time dependence, graphite pile, reaction ratios, neutron diffusion equation, scattering kernel, phonon spectrum, thermal neutron spectrum, Young-Koppel model, Yoshimori-Kitano model, Page-Haywood model

I. INTRODUCTION

In recent years, the time-dependent thermal neutron energy spectrum in the crystalline moderator has received considerable attention in the field of neutron physics and reactor engineering, for its close relation to the neutron scattering parameters and the phonon frequency spectrum of the crystalline moderator.

Theoretically, the time-dependent thermal neutron energy spectra in graphite can be obtained by solving the Boltzmann equation in the diffusion approximation utilizing the time-step method and/or the expansion method making use of the time eigenfunctions, together with the scattering kernels based on theoretical phonon models. However, the theoretical studies undertaken so far are all based on the calculation of space-independent, time-dependent thermal neutron energy spectra in the infinite moderator, or on calculations in which the buckling dependence of the time-dependent neutron spectra in finite geometry is considered. Few examples are known where consideration has been given to the space-dependence of the time-dependent neutron thermalization. With regard to the phonon frequency spectrum for graphite, the Young-Koppel phonon spectrum derived from the Yoshimori-Kitano theoretical phonon model for graphite is commonly utilized to obtain the scattering kernels and other parameters related to thermal neutron scattering. However, this theoretical phonon model is not necessarily perfect. In order to assess the adequacy of the various phonon models, studies have been undertaken on the time-dependent thermal neutron energy spectra, and experiments in neutron wave propagation have also been conducted to this end.

On the one hand, time-dependent thermalization in graphite has been examined experimentally by many investigators. The studies reported so far are of two categories. Those of one category cover measurements of the time-dependent differential neutron spectra by means

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of the chopper time-of-flight spectrometer synchronized with a pulsed accelerator. The data obtained from such measurements are limited to the center of the graphite block. The other category\(^{(10)-(14)}\) comprises measurements of the time-dependent reaction rates on materials showing non-\(1/\nu\) energy response by means of the integral method. Most of the studies of the latter category\(^{(10)-(12)}\) have aimed mainly at measuring the neutron slowing-down times up to \(\sim 100 \mu\text{sec}\) following a fast neutron burst, and comparing the observed values with those calculated from the Williams formula\(^{(16)}\) based on the Parks phonon model for graphite. No satisfactory explanation has so far been given for the discrepancy of time-dependence found between the measured and calculated reaction rates in the time range of interest. Concerning the space-dependence of the time-dependent reaction rates, Takahashi & Sumita\(^{(11)}\), as well as Maekawa & Yamamuro\(^{(12)}\) have suggested the existence of spatial effect for neutron slowing-down times in graphite, but significant experimental evidence had yet to be established. What is more, no explicit explanation has been given so far on the relationship between the space-dependence of the time-dependent neutron energy spectra and that of the peaking times of the reaction rates.

Polley & Walker\(^{(13)}\) have determined the time-dependent reaction ratio between Cd and Hg as a spectral indicator at two spatial positions in graphite by means of measurements at \(60 \mu\text{sec}\) intervals in the time range of \(90-1,300 \mu\text{sec}\) after the neutron burst. This interval of \(60 \mu\text{sec}\); however, would appear too coarse for comparison of the measured data with calculated values for the purpose of evaluating the adequacy of theoretical models of neutron thermalization, when one considers the fact that the time-dependent Cd/Hg ratio is extremely sensitive to the neutron energy spectrum in the earlier stages of neutron thermalization. No inference has been made by the above authors on any space-dependence of the time-dependent neutron energy spectra and that of the peaking times of the reaction rates.

Based on the foregoing considerations, the time-dependent neutron thermalization has been studied experimentally and theoretically at various spatial positions \(r=10, 30, 50\) and \(70\) cm from a fast neutron source placed at the center of a graphite pile. Six time-dependent reaction ratios obtained from four reaction materials (Gd, Sm, Cd and one of \(1/\nu\) energy dependence) were used as the spectral indicators. Reaction ratios were adopted as subjects of measurement, since the ratio between two reaction rates cancels the time dependence of neutron intensity. The first three materials have resonance at different positions along the neutron energy spectrum in the thermal neutron energy region, so that the reaction ratios vary from position to position along the spectrum. Time-dependent reaction ratios were measured at \(2.175 \mu\text{sec}\) intervals up to \(\sim 600 \mu\text{sec}\) following the fast neutron burst. The theoretical time-dependent reaction ratios to be compared with the experimental data were obtained by solving numerically the space- and time-dependent neutron diffusion equation based on the four different types of phonon frequency spectrum models, \textit{i.e.}, those for graphite given by Young-Koppel\(^{(6)-(7)}\), Yoshimi-Kitano\(^{(8)}\) and Page-Haywood\(^{(16)}\), and the simplified model for solid proposed by Debye.

It will be seen from what follows that the reaction ratio technique should be useful for examining time-dependent thermalization and for assessing the phonon models for crystalline moderator. Also, proof is given of space-dependence shown by the time-dependent neutron energy spectra; and the relationship between the space-dependence of the time-dependent neutron energy spectra and that of the peaking times of the reaction ratios is made clear.

**II. Calculational**


The space- and time-dependent neutron diffusion equation was solved numerically to obtain the neutron energy spectra as a function of space and time in the graphite pile. This equation is

\[
\frac{1}{\nu} \frac{\partial \phi(r, E, t)}{\partial t} = D(E) \nabla^2 \phi(r, E, t) + \left( \sum_{\nu} s_\nu(E) \right) \phi(r, E, t) + \int_0^\infty \sum_{\nu} s_{\nu}(E' \rightarrow E) \phi(r, E', t) dE' + S(r, E, t) \delta(t),
\]

where \(\phi(r, E, t)\) is the neutron flux per unit...
energy interval at position $r$ and time $t$, $\Sigma_a(E)$, $\Sigma_{in}(E)$ and $\Sigma_{ir}(E'\rightarrow E)$ are the macroscopic absorption cross section, the macroscopic total inelastic scattering cross section and the inelastic scattering kernel respectively, $D(E) = 1/(3\Sigma_{tr}(E))$ is the diffusion coefficient ($\Sigma_{tr}(E)$: Macroscopic transport cross section, where $\Sigma_{tr}(E) = (\Sigma_{in}(E) + \Sigma_{ir}(E')) \cdot (1 - \bar{\mu}(E))$, $\bar{\mu}$: Average cosine of scattering angle in laboratory system) and $S(r, E, t) \delta(t)$ is the source energy spectrum. Then, $\Sigma_{in}(E'\rightarrow E)$, $\Sigma_{ir}(E)$ and $\Sigma_{tr}(E)$ were obtained from various theoretical phonon frequency distributions for graphite by making use of the THRUSH code\(^{(17)}\) based on the incoherent approximation. Exception was made for the case of the transport cross section at the neutron energy of 0.001 eV, where, in consideration of the Bragg cut-off effect, $0.1 \cdot \Sigma_{fr}$ (Macroscopic free-atom scattering cross section) was used as the value of $\Sigma_{tr}$. The phonon frequency distributions used as input data for the THRUSH code are shown in Fig. 1, together with the effective temperatures of graphite crystal obtained from each phonon spectrum.

In the present calculation, the graphite pile was assumed to be a sphere with a radius of 120 cm. This assumption may be justified since the spatial distribution of the neutron flux is nearly spherically symmetric with respect to the fast neutron source positioned at the center of the pile. Therefore, in Eq. (1), $P^2 = \partial^2/\partial r^2 + 2/r \cdot \partial/\partial r$, and the boundary conditions for Eq. (1) become (i) $(\partial \phi/\partial r)_{r=0} = 0$, and (ii) $\phi(120, E, t) = 0$, for all values of $E$ and $t$.

In this paper, the initial stage of thermalization was taken as the neutron slowing-down time corresponding to the Cd-resonance. First of all, the neutron energy spectrum in this stage of thermalization must be determined, since it is required as a source term for subsequent calculation of the space- and time-dependent thermal neutron spectra. The computational procedure for this purpose is as follows. The energy mesh was divided into 45 groups in the energy range of 0.005$\sim$2.5 eV. The source spectrum $S(E)$ was calculated from the Williams formula (Eqs. (16), (17), (33), and (30 in Ref. (15)) relevant to the slowing-down time corresponding to the In-resonance. This time was estimated to be 25$\mu$s from the result of the measurements which will be described in Chap. III. The spatial distribution of neutron flux at the peaking time of In-resonance was obtained from measurements of the induced radioactivity present in In foils covered with Cd filters placed at intervals of 10 cm in the pile. A time step of 0.5$\mu$s was chosen, and the range of radial coordinate (0$\sim$120 cm) was covered at spatial intervals of 10 cm to cover the range of the radial coordinate (0$\sim$120 cm). In this case the Young-Koppel phonon model was used as a phonon frequency spectrum, considering the fact that in the epithermal energy region the time-dependent neutron spectra do not depend so much upon the phonon spectrum of the moderating medium as in the thermal energy region.

By taking as source spectra the neutron energy spectra obtained above at the peaking time of the Cd-resonance, the space- and time-dependent thermal neutron spectra were calculated on the basis of the various phonon models. The spatial distribution of the source spectrum at the peaking time of Cd was experimentally obtained both from measurements of the difference between the induced radioactivity of In foils covered with Gd filters of two different thicknesses (0.1 and 0.05 mm) placed at intervals of 10 cm, and from the measured reaction rates at the peaking time of Cd-resonance (as described in Chap. III). The energy mesh was divided into 38 groups in the energy range of 0.001$\sim$0.44 eV; 10 cm was taken as spatial interval; and as time interval the choice was 2$\mu$s for the time range below 300$\mu$s, 5$\mu$s in the time range of 300$\sim$800$\mu$s, and
10 μsec in the range above 800 μsec.

2. Calculations of Time-dependent Reaction Rates and Reaction Ratios

The theoretical space- and time-dependent reaction rates or reaction ratios to be compared with the experimentally determined values were obtained by calculating the integral of the product of the calculated space- and time-dependent thermal neutron spectra and the effective reaction cross sections of materials for the neutron energy of interest. The reaction cross-section data of the materials used were taken from BNL-325(18), ANL-7387(19) and KFK-1080(20).

In this case the self-shielding factors at different energies for each of the foils used were calculated under the assumption that the angular distribution of neutrons incident on the foil was isotropic. The error in the calculated reaction rates arising from the above assumption for particular foils used in the experiment was estimated to be <1.5% for Gd, <0.5% for Sm and negligible for Cd, considering the anisotropy due to the presence of the cavity hole for the neutron detector.

III. Experimental

A schematic diagram of the experimental arrangement is shown in Fig. 2.

1. Pulsed Neutron Generator

The pulsed neutron source used was a TO-SHIBA 200 kV neutron generator which produced 14 MeV neutrons using the T(d, n)4He reaction. The generator was equipped with both pre- and post-acceleration pulsing systems to yield an off-current to on-current ratio as low as 10⁻⁵. The pulse width used was either 1 μsec or 10 μsec, and a pulse repetition frequency of 34 Hz was used throughout the experiments. The beam duct in the target assembly installed inside the graphite pile has an external diameter of 4 cm, and is made of pure Al to minimize thermal neutron absorption.

2. Graphite Pile

The graphite pile, composed of graphite blocks of reactor grade, with a gross average density of 1.74 g/cm³, is 230×190×190 cm³ (longitudinal*).

* Direction of the beam duct
dimension being 230 cm), and is shielded on all sides by 1 cm thick powdered boric oxide sealed in Al containers to prevent re-entry of low energy room-return neutrons. A tritium target was placed at the center of the pile to permit analyses under the assumption of spherically symmetric geometry. A neutron detector was placed at various positions from the target along the longitudinal axis to ensure measurements to be performed in a nearly isotropic neutron field. The experimental hole for the neutron detector has a cavity of 7 cm diameter.

3. Neutron Detectors

In order to test the adequacy of the calculated neutron spectrum at the initial stage of the thermalization, foils of In 0.05 mm thick, of Er 0.25 mm thick, of Cd 14 µm thick and of Lu 0.25 mm thick, as well as Eu powder, were used for the measurements of neutron slowing-down time in the energy range 0.1~1 eV. Similarly, for the measurements of the space- and time-dependent thermalization, use was made of foils of Gd 4 mm thick, of Sm 25 µm thick and of Cd 14 µm thick. The dimensions of these foils were approximately 4x4 cm² and the purity was 99.9%. The neutron-induced-reaction rates in the foil were determined with a 0.5 mm thick plastic scintillator through detection of the electrons converted in the foil by the low energy prompt g-rays, except in the cases of the In foil and Eu powder, where a thin NaI scintillator was used. The foil was positioned close to the scintillator. The advantage of detecting the converted electrons by a thin plastic scintillator over the direct detection of the prompt g-rays by a NaI scintillator is that it can provide a better signal-to-background ratio in measurements of the time-dependent reaction rates in the foil, thus making it possible to obtain a greater accuracy of the experimental data. For the detector of 1/ν energy response, a BF₃ proportional counter (Mitsubishi ND8523-60) was employed. Amplification of the signals from these detectors was obtained by high-speed current-mode pulse amplifiers, which offered the advantage of assuring better time resolution. The output signals were fed to the time analyzer through a pulse height discriminator.

4. Time Analysis

For time analysis in this series of measurements, use was made of a fast 256-channel time analyzer(21) embodying a high-speed plated-wire memory with 175 nsec cycle time. An important feature of this analyzer is that it operates in multiscaler mode, and dispenses with the counting-loss correction for the dead time due to the memory cycle, often required when a time-of-flight mode time analyzer is used. The channel widths used in the experiments were 0.5, 1.175 and 2.175 µsec. The background signals prevailing after a sufficiently long time had elapsed following a burst of fast neutrons were stored in the last 256th channel, and throughout the experiments a standard interval of 25.6 msec was made to elapse between the end of the preceding channel and the rise of the subsequent background channel. The time analyzer was triggered by pulses synchronized with the drive pulses for the post-acceleration pulsing system. The starting point in the time axis—i.e. the instant of neutron burst—was chosen to correspond to the midpoint of the burst, which was determined by observing the shape of the response registered by the plastic scintillator to recoil protons.

5. Background Subtraction

The background counts of captured γ-rays in this series of experiments are due to the prompt γ-rays from carbon nuclei in graphite and from the detector vessel provided with a photomultiplier, as well as to the induced activities of the sample foil or of the detector vessel. The prompt γ-rays intensity varies with elapse of time after the neutron burst while the induced activities are almost time-independent. The time-dependent backgrounds were therefore measured with the sample foil removed, but with the other instrumental conditions unaltered. On the other hand, the time-independent backgrounds due to induced activities of the foil were determined from the difference in count between that stored in the background channel—obtained with the foil inserted—and that obtained with the foil removed, the latter count being included in the time-dependent background counts mentioned above. The overall background counts were estimated as the sum of these two kinds of background. In obtaining the background counts, the measured data were normalized with reference to the monitor counts obtained with the two monitor systems shown in Fig. 2.
IV. RESULTS AND DISCUSSIONS

1. Calculated Space- and Time-dependent Neutron Spectra

Figure 3 shows, as an example, the time-dependent neutron energy spectra at r=30 cm, calculated on the basis of the Young-Koppel phonon spectrum. In this figure, the initial energy spectrum at 25 μsec after fast neutron burst was calculated from the Williams formula. The fineness of energy mesh was chosen separately for the periods prior to and subsequent to the peaking time of Cd-resonance (80 μsec after burst), as described in Chap. II.

The space dependence of the calculated time-dependent thermal neutron energy spectra is shown in Fig. 4, together with the space-independent spectra obtained by neglecting the 1st term of the right-hand side in Eq. (1). Figure 5 presents the deviations of the space-dependent spectra from those obtained by disregarding their space dependence. It is irrefutably proved from Figs. 4 and 5 that the time-dependent thermal neutron energy spectrum is space-dependent in the pile: At a given instant after neutron burst, the thermal neutron spectra become harder with increasing distance from the fast neutron source placed at the center of the pile. This can be understood when it is considered that the neutrons on the whole diffuse outward with time, and that, due to thermal elastic scattering, a part of the neutrons in the outer portions of the pile are those possessing an energy spectrum corresponding to that relevant to the inner portions at an earlier instant. This result is also consistent with the fact that the transport mean free paths of
neutrons are slightly longer in the higher energy region due to the tendency toward forward scattering of the neutrons in the laboratory system and also because the higher energy neutrons diffuse faster toward the outer portions of the pile. Figures 4 and 5 indicate that, as time elapses after neutron burst, neutrons of $\sim 0.001\text{eV}$ become increasingly dominant in regions remote from the center, on account of the large transport mean free path of neutrons with energy below the Bragg cut-off.

In order to test whether or not the calculated neutron energy spectra at the Cd peaking time based on the Young-Koppel phonon model was adequate as source neutron spectra, a comparison was made between the calculated and measured peaking times of the reaction rates of the resonance materials Er, Eu, Cd and Lu, in reference to the measured peaking time of 25 $\mu\text{sec}$ for In-resonance. Since the space-dependence of the peaking times of the reaction rates should be negligible in the time range considered, the comparison was performed only for $r=30\text{cm}$ in the pile. The result is shown in Table 1. From this table, it may be reasonably concluded that the neutron energy spectra at the Cd peaking time calculated on the basis of the Young-Koppel phonon spectrum can validly be used as the source energy spectra in calculating neutron thermalization.

Table 1 Comparison between measured and calculated peaking times of reaction rates of different resonance materials

<table>
<thead>
<tr>
<th>Material</th>
<th>Resonance energy (eV)</th>
<th>Measured values ($\mu\text{sec}$)</th>
<th>Calculated values ($\mu\text{sec}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>In</td>
<td>1.46</td>
<td>$25.0 \pm 1.0$</td>
<td>25.0</td>
</tr>
<tr>
<td>Er</td>
<td>0.58</td>
<td>$41.0 \pm 2.0$</td>
<td>42.0</td>
</tr>
<tr>
<td>Eu</td>
<td>0.32</td>
<td>$44.5 \pm 2.0$</td>
<td>42.0</td>
</tr>
<tr>
<td>Cd</td>
<td>0.18</td>
<td>$79.9 \pm 2.0$</td>
<td>76.5</td>
</tr>
<tr>
<td>Lu</td>
<td>0.14</td>
<td>$94.5 \pm 4.0$</td>
<td>91.5</td>
</tr>
</tbody>
</table>

2. Time-dependent Reaction Rate for Material of $1/\nu$ Energy Response

The theoretical time-dependent reaction rates for the material of $1/\nu$ energy dependence are almost insensitive to the phonon spectrum but depend on the initial spatial distribution of the neutron flux in the pile at the time of Cd peaking. Figure 6 shows the initial spatial distributions of the neutron flux measured by the two different methods mentioned in Chap. II.

Figure 7 compares the time-dependent counting rates measured by BF$_3$ proportional counter with the results calculated on the Young-Koppel phonon model with use made of the initial spatial distributions of the neutron flux shown in Fig. 6. It is seen that a steeper gradient of the initial spatial distribution of the neutron flux used in the calculation is conducive to a more sensitive...
time-dependence of the reaction rate for the material of $1/\nu$ dependence, in the time range under consideration.

3. Measured Time-dependent Reaction Rates for Cd, Sm and Gd Foils

In Fig. 8 is plotted an example the raw data of the time-dependent reaction rates of the $4\mu$m thick Gd-, $25\mu$m thick Sm- and $14\mu$m thick Cd-foils measured at $2.175\mu$s intervals at $r=10\text{ cm}$ in the pile. The height of the plots include the background counts, which also indicated separately by curves drawn at the bottom of the figure. The shapes of the curves for the measured time-dependent reaction rates at other spatial positions are similar in trend to those shown in Fig. 8, but with increasing distance from pile center, the reaction rates tend to accelerate their domination.

Fig. 8 Time-dependent reaction rates of $4\mu$m Gd-, $25\mu$m Sm- and $14\mu$m Cd-foils measured at $2.175\mu$s intervals, at $r=10\text{ cm}$ super-imposed on overall background counts obtained.

Fig. 9 Comparison between measured and calculated time-dependent reaction ratios for $\text{Cd}/^{10}\text{B}$, $\text{Sm}/^{10}\text{B}$ and $\text{Gd}/^{10}\text{B}$ at various spatial positions.

Error bars indicate standard deviations in experimental curve obtained by least squares fitting. $80\mu$s has been adopted for the Cd peaking time in the calculated curves, to correspond to the measured value. Reaction ratios are normalized to unity at maximum.
over the background with elapse of time after a burst. This tendency can be understood from the time-dependence of the neutron intensity, illustrated in Fig. 7.

4. Time-dependent Reaction Ratios for Cd/10B, Sm/10B and Gd/10B

Comparisons between the calculated and measured time-dependent reaction ratios for Cd/10B, Sm/10B and Gd/10B are shown in Figs. 9 for each spatial position in the pile. As described in Chap. II, the calculated neutron energy spectra at the theoretical Cd peaking time were used as the source spectra for the calculation of thermalization. The calculated Cd peaking time itself, has a small discrepancy with the measured data, as shown in Table 1. In Figs. 9, therefore, to permit consistent comparison of the time dependence shown by the measured and calculated reaction ratios following Cd peaking, the Cd peaking time in the calculated curves was fitted to the measured value of 80 μsec. With the same view, both calculated and measured reaction ratios presented in the ordinate of these figures are normalized to unity at maximum value. For comparison of the experimental data with the theoretical curves, the experimental data are fitted to a 10th order polynomial by least squares method. The margins of error marked on the experimental curves are based on the standard deviations of the observed values obtained in the least squares fitting. Figures 9 reveal that the measured time-dependent reaction ratios for Cd/10B, Sm/10B and Gd/10B are in better agreement with the calculated results based on the phonon models for graphite given by Page-Haywood and by Yoshimori-Kitano than that by Young-Koppel. The Debye simple model, adopted in combination with the Debye temperature of 0.18 eV, is apparently not valid for explaining the experimental results, although it yields an effective temperature for graphite crystal fairly close to that obtained from other phonon models, as seen in Fig. 1. This implies that the time-dependent neutron thermalization depends to significant extent upon the shape of the phonon frequency distribution, and cannot be related simply to the effective temperature of the moderator crystal, which is an averaged quantity dependent on the phonon frequency distribution.

Table 2 shows a comparison of the measured peaking times of the reaction ratios for In/10B, Cd/10B and Sm/10B with the calculated results based on various phonon models for graphite. The experimental peaking times of the reaction ratios were determined by finding out which value best reproduced the peaking times obtained by least squares fitting with various time ranges and various orders of polynomial. From Table 2, it is clear that the peaking times of the reaction ratios become longer as the distance from the fast neutron source is increased. This is due to the increasing hardness shown by the time-dependent thermal neutron spectrum, with increas-

<table>
<thead>
<tr>
<th>Position</th>
<th>Burst to Cd/10B (μsec)</th>
<th>Burst to In/10B (μsec)</th>
<th>Burst to Sm/10B (μsec)</th>
<th>Calculated values</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>135.7</td>
<td>55.5</td>
<td>80.5</td>
<td>55.2</td>
</tr>
<tr>
<td>30</td>
<td>138.1</td>
<td>53.4</td>
<td>78.4</td>
<td>59.7</td>
</tr>
<tr>
<td>50</td>
<td>141.6</td>
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<td>79.4</td>
<td>62.2</td>
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<tr>
<td>70</td>
<td>144.3</td>
<td>56.4</td>
<td>81.4</td>
<td>62.9</td>
</tr>
</tbody>
</table>

Table 2 Comparison between measured and calculated peaking times and peak-to-peak times of reaction ratios for In/10B, Cd/10B and Sm/10B
It should also be noted that the theoretical peaking times depend on the phonon models used in the calculations. The measured peak-to-peak times for different reaction ratios are in good agreement with the calculated results based on the Yoshimori-Kitano model and on the Page-Haywood model. This is consistent with the results given in Figs. 9.

5. Time-dependent Reaction Ratios for Gd/Sm, Gd/Cd and Sm/Cd

It is expected that the space-dependence of the time-dependent thermal neutron energy spectra affects the time-dependent reaction ratios for Gd/Sm, Gd/Cd and Sm/Cd, and this induces the space-dependence of these time-dependent reaction ratios. Figure 10 (a) shows the space-dependences of the measured time-dependent reaction ratios for Gd/Sm and Gd/Cd, while that for Sm/Cd is shown in Fig. 10(b). In Figs. 10(a), (b) all the data are normalized to the Cd/10B peaking time of 80 μsec of the initial stage of thermalization. The solid curves were obtained from the quotient between the 10th order polynomials of the experimental curves in Figs. 9. It is obvious from Fig. 10(a) that, in the time range up to ~600 μsec after a burst, the time-dependent thermal neutron spectra are spatially dependent, and that the space dependence is accentuated with elapse of time. This is consistent with the theoretical results shown in Figs. 4 and 5. On the other hand, no space dependence is discernible in the case of the measured time-dependent reaction ratio for Sm/Cd, as shown in Fig. 10(b). This result may be explained by considering the fact that the reaction ratio for Sm/Cd is sensitive to the neutron energy spectra only in the transient time range between the Cd and the Sm peaking times i.e. 80~150 μsec after a burst, and that in this early stage of thermalization there is smaller space dependence of the time-dependent neutron spectra.

Figures 11 present comparisons between the measured and calculated reaction ratios for Gd/Sm and Gd/Cd at various spatial positions in the pile. The measured time-dependent reaction ratio for Gd/Sm agrees with the calculated results based on the three different types of phonon models for graphite. With regard to the Gd/Cd ratio, the measured time-dependent ratio fits the calculated results based on the Yoshimori-Kitano model and the Page-Haywood model better than that based on the Young-Koppel model, except in the case of r=70 cm where the discrepancy between the measured and calculated reaction ratios is seen to have become quite significant. This discrepancy may be attributed to errors involved in the measured reaction rate for Cd; the counting rate is low at r=70 cm.

As described in the previous chapter, the measurements of the time-dependent reaction rates were carried out under the condition of a
neutron burst width of 10 μsec. This width of the burst is large enough to bring about a slight change to the shape of the neutron energy spectrum at the Cd peaking time, which, in turn, gives rise to small deviations in the calculated time-dependent reaction ratio for Sm/Cd, while the influence brought by the burst width on the time-dependent reaction ratios for Gd/Cd and Gd/Sm is negligible. Figure 12 shows a comparison between the calculated time-dependent reaction ratio for Sm/Cd corrected for the burst width and the weighted average of the measured values at four positions in the pile. From this figure, it is seen that the calculated time-dependent reaction ratio for Sm/Cd based on the Page-Haywood phonon model is in agreement with the experimental values in the time range up to ~200 μsec after fast neutron burst.

V. CONCLUSION

The conclusions obtained from the studies reported here can be summarized as follows.

1) The reaction ratio technique based on
measurement of the time-dependent reaction rates of materials with different energy responses in the thermal energy region is a powerful tool for studying time-dependent thermalization, and for evaluating the different phonon models for crystalline moderator.

(2) Space-dependence of the time-dependent thermal neutron energy spectra in graphite has been demonstrated theoretically and experimentally: during thermalization, the time-dependent thermal neutron spectrum is harder at positions removed from the fast neutron source placed at the center of the pile.

(3) The space-dependence of the peaking times of the reaction ratio between the resonance material and the material of 1/ν energy dependence can be explained from the space-dependence of the time-dependent slowing-down and thermal neutron spectra in the moderator.

(4) The Page-Haywood phonon and the Yoshimori-Kitano phonon models for graphite fit the results of the measured time-dependent reaction ratios better than the Young-Koppel model. This result indicates that time-dependent thermalization in graphite is significantly influenced by the high-energy cutoff of the phonon spectrum.

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References