Measurement of Neutron Slowing Down Time in Graphite

Yoshihiko KANEKO*, Ryosuke KUROKAWA*, Fujiyoshi AKINO* and Kenji SUMITA**

Received September 8, 1966

A series of pulsed neutron experiments was performed to investigate the chemical binding effect on the neutron slowing down time. Bursts of D-T neutrons of 1 μsec width were generated in a hexagonal prism with 240 cm high with 120 cm flanks, made of reactor grade graphite with a density of 1.54. The slowing down neutrons were detected by bare as well as energy-selective filter-covered BF3 counters, and analyzed with a 256 channel time analyzer. Slowing down times in graphite were determined by interpreting the increment of the difference of events between the two counters to be due to the contribution made by the fraction of the slowing down neutrons at the time of measurement that was below the cut-off energy of the filter. The results of measurements showed good agreement with calculation based on the crystal model.

I. INTRODUCTION

One of the most important problems in nuclear reactor physics is the determination of neutron slowing down properties of reactor materials. The energy decrease of fast neutrons in their collision with moderator nuclei can be described by classical mechanics. However, when these neutrons are slowed down into the energy range of the chemical binding of the moderator, the simple classical model which simulates the collision of neutrons to that of billiard balls no longer remains valid; more elaborate models have been proposed, incorporating such concepts as phonon exchange between the neutrons and moderator lattice. Several studies have been devoted to proving the validity of these models. In the present work, the effect of chemical binding on the neutron slowing down time has been investigated by pulsed neutron experiments.

II. PRECEDING WORK

A number of different definitions have been in current use for the slowing down time with

* Japan Atomic Energy Research Institute, Tokai-mura, Ibaraki-ken.
** Now at Faculty of Engineering, Osaka University, Miyakojima-ku, Osaka.
regard to the different measured quantities (neutron density, neutron flux, slowing down density). In this work, the following two definitions will be adopted.

One is the average time elapsed before the neutrons are slowed down below a definite energy $E_c$, i.e., the first moment of the slowing down density at energy $E_c$, or $q(E_c,t)$:

Slowing down time to below $E_c$

\[ \langle t \rangle = \frac{\int_0^\infty q(E_c,t)dt}{\int_0^\infty q(E_c,t)dt} \quad (1) \]

The other is the instantaneous time when the neutron density of energy $E_r$ reaches its maximum:

Slowing down time to $E_r$

The principles for measuring the slowing down times defined above will be given here.

After a burst of 14 MeV D-T fast neutrons, a bare BF$_3$ counter and a BF$_3$ counter covered with filter, located in the resulting field of neutron density $n(E,t)$, will show the following responses.

- The counting rate of the bare BF$_3$ counter
  \[ (CR)_{\text{bare}} = \int_0^n S(E)n(E,t)dE \quad (2) \]
  where $T(E)$ is the transmission of the filter, and $S(E)$ the counting efficiency of the BF$_3$ counter—or $a/v$, $a$ being a proportional factor.

- The counting rate of the BF$_3$ counter covered with filter
  \[ (CR)_{\text{filter}} = \int_0^n T(E)S(E)n(E,t)dE, \quad (3) \]
  can be related to the net slowing down density below $E_c$, $q(E_c,t)$, through the equation

\[ q(E_c,t) = \frac{\langle t \rangle}{\int_0^n n(E,t)dE} \]

The above arguments lead to the conclusion that the slowing down time below $E_c$ is

\[ \int_0^{E_c} (CR)_{\text{bare}} - (CR)_{\text{filter}} dt \]

\[ \int_0^{E_c} \frac{d}{dt} [(CR)_{\text{bare}} - (CR)_{\text{filter}}] dt. \]

1. Slowing Down Time below $E_c$

If the filter is ideally of sharp-cut off type as illustrated in Fig. 1(a),

\[ T(E) = \begin{cases} 1, & E > E_c \\ 0, & E < E_c \end{cases} \quad (4a) \]

\[ (CR)_{\text{bare}} = a \int_0^n n(E,t)dE \quad (5) \]

\[ (CR)_{\text{filter}} = a \int_0^n n(E,t)dE. \quad (6) \]

Thus, the counting rate of the bare BF$_3$ counter $(CR)_{\text{bare}}$ indicates the number of neutrons of all energies, which is approximately constant in time, because the leakage and absorption of neutrons during moderation are very small. On the other hand, $(CR)_{\text{filter}}$, the counting rate of the BF$_3$ counter covered with filter gives the number of neutrons hav-
where $\Delta E < E_r$.  

\[ T(E) = \begin{cases} 
1, & |E - E_r| > \Delta E \\
0, & |E - E_r| < \Delta E, 
\end{cases} \quad (10) \]

Thereby, the slowing down time to $E_r$ can be determined in terms of the time when $(CR)_{filter}$ reaches its minimum.

3. Effective Cut Off and Effective Resonance Energies of the Filter

The transmissions of the In-, Cd- and Gd-filters used in the experiment are shown in Fig. 2.

![Fig. 2 Transmission of Filters used in the Experiments](image)

While these transmissions are obviously not ideal step functions such as shown in Fig. 1, the principle of the measurement can be considered well founded provided suitable effective cut off and effective resonance energies, which should be calculated by the following criteria:

1. The effective cut off energy $E_{c,eff}$ should be determined in such manner that, for the probable slowing down spectrum, the right-hand side of Eq.(9) measured by the actual filter would be the same as that measured by the ideal filter.

2. The effective resonance energy $E_{r,eff}$ should be determined in such manner that, for the probable slowing down spectrum, the time when $(CR)_{filter}$ measured by the actual filter reaches its minimum would be the same as the corresponding time with the ideal filter.

In this work, the probable slowing down spectrum is assumed to be, for calculating effective cut off energy, the continuous slowing down spectrum by age theory, and for calculating the effective resonance energy the Gaussian spectrum.

Small errors due to the deviation of these assumed spectra from the true spectra to be investigated are within experimental error.  

The calculated effective cut off energy $E_{c,eff}$, and the effective resonance energy $E_{r,eff}$ are listed in Table 1.  

IV. EXPERIMENTAL ARRANGEMENT

In this experiment, pulsed neutrons were generated near the center of the graphite pile.  

The slowing down neutrons were detected first by bare BF$_3$ counter and then by the same counter covered with filter.  

The neutron induced pulses were analyzed with a 256 channel time analyzer.  

A block diagram for the measuring arrangement is shown in Fig. 3.

1. Pulsed Neutron Source

A 200 kV Cockcroft-type compact pulsed neutron source was used in this experiment.  

The slowing down experiment was undertaken with the use of a thin tritium target, bursts of 14 MeV neutrons being supplied with a pulse width of 1 $\mu$sec.

2. Experimental Assembly

Approximately 2800 hollow graphite tubes and rods, each 120 cm long and 6.5 cm in outer diameter are stacked to form a hexagonal prism such as shown Fig. 4.  

Insertion of graphite rods of 5.5 cm outer diameter into these hollow tubes produced a graphite pile of nuclear grade with a density of 1.54/cm$^3$; and then after completion of experiments on this pure graphite pile, the tubes in a given portion of the lattice had the graphite rods removed to be replaced by fuel rods, to obtain a graphite pile with an annular fuel zone of 63 cm outer and 50 cm inner diameter.  

Each fuel rod is encased in a graphite sleeve containing fuel pellets made from a mixture of
20% enriched uranium oxide and graphite grains. The atomic ratio of carbon to $^{235}$U is 5.378 in the core region.

In both piles, several of the hollow graphite tubes have also been used for providing void channels into which were inserted neutron detectors and an extension tube of the accelerator. The distance from the wall of the graphite channel to the center axis of the detector was 2.0 cm.

A thin tritium target was placed along with neutron detectors on the mid plane of the pile in the manner shown in Fig. 4. The detector position designated 8C6 coincides approximately with the center of the fuel region of the annular core.

3. Neutron Detectors and Time Analyzer

The slowing down neutrons resulting from the D-T 14 MeV neutron injections were detected first by bare BF$_3$ counter (2.5 cm diam. BF$_3$ 36 cm Hg) and were then detected by the same BF$_3$ counter covered with filter. Four of the filters used in the experiment were of cut off type, three of cadmium and one of gadolinium, while one In-filter of the resonance type was also used. The Cd- and In-filters were made of metal foil, while Gd$_2$O$_3$ dispersed in resin layer on an aluminum cylinder was used for the Gd-filter.

It was feared that possible small inhomogeneities in the Gd$_2$O$_3$ and the presence of hydrogen atoms in the resin layer might affect the experimental results. But no appreciable change in the results obtained was observed from measurements undertaken for verification with the use of metallic gadolinium. The physical properties of these filters are listed in Table 1. The effective cut off and resonance energies appearing on this table have been determined by the criteria discussed earlier for isotropic neutron flux.

The neutron induced pulses from the de-
tector were amplified by preamplifier and non-
overloading amplifier with 0.1 msec rise time,
and the pulse height selector outputs were
fed to a 256 channel time analyzer, whose
analysis channel length could be selected
among the values of 0.25, 0.5 and 1.0 μsec.
The time origin of the measurement was
taken to be the instant at which the counting
rate of the bare BF₃ counter amounted to
half of the step increment caused by the in-
jection of the pulsed neutrons.

The delay time between the triggering of
the time analyzer and pulsed neutron injec-
tion was chosen to be 10 μsec. A monitor
channel was also provided to measure the
total neutron yields in a run.

V. RESULTS OF THE EXPERIMENT

The counting rates of bare, In-filter covered
and 1.36 mm Cd-filter covered BF₃ counters
are represented as functions of time in Figs.
5~7 respectively.

The dip in response seen in the case of
In-filter is accounted for by the resonance
captures at 1.46 eV. The time of attainment
of minimum response was determined to be
24.5 ± 1 μsec from visual observation. The in-
crement of the difference of events between

![Fig. 5 Time Response of Bare BF₃ Counter in Graphite Pile](image-url)
Fig. 6 Time Response of In-filter covered BF$_3$ Counter in the Graphite Pile

Fig. 7 Time Response of Cd-filter covered BF$_3$ Counter in Graphite Pile

the bare and 1.36 mm Cd-filter covered BF$_3$ counters can be considered to result from the contribution of the fraction of the neutrons
slowing down at the instant of the measurement below the cut off energy of the filter, which is 0.61 eV. The same interpretation is possible with the other Cd-filters and the Gd-filter. The difference increments with the 1.36 mm Cd-filter and with the other filters are shown in Figs 8~11.

Slowing down time to 1.25 eV and corresponding times to energies below 0.61, 0.50, 0.40 and 0.18 eV were determined through the formulas (8) and (11) with the use of the data in Figs. 3, 8~11 respectively. Here, to correct small changes in the neutron density, the reciprocals of the counting rate of the bare BF3 counter, \((CR)_{\text{bare}}^{-1}\), were multiplied by the difference between \((CR)_{\text{bare}}\) and \((CR)_{\text{filter}}\). The results are listed in Table 1 together with the theoretically predicted values. Experimental errors have been estimated to be within 5%. The following corrections were considered:

1. Leakage of neutrons from the finite graphite pile before slowing down below the cut off energy
2. Time of flight effect from the wall of the graphite channel to the BF3 counter
3. Background neutrons caused by the post-accelerated beam
4. Slowing down by hydrogen contained in the graphite.

The effect on the slowing down time caused by the small void space between the stacked graphite rods was taken into account by homogenizing the void with the graphite rods. The experimental values of the slowing down time did not show appreciable dependence on the detector positions and this may be due to the fact that the space-dependence of the slowing down spectrum is not as strong as the observed dependence of the neutron density itself.

These experimental results were compared with the theoretical values. No theoretical work has so far been performed on the time moments of slowing down density \(q(E,t)\) in pure moderator, and these values have therefore been approximately evaluated with the use of the results on the first time moments of neutron flux \(\Phi(E,t)\) obtained by Marshak and Williams.

From their analysis, the first time moment of neutron flux on the 0 K free gas model \(<t>\Phi^{0K}\) is given by

\[
<t>\Phi^{0K} = \frac{1}{1 + 2 + 1 + \frac{r}{1 + r}} \frac{I}{v},
\]

where \(r = \frac{M-1}{M+1}\), \(M\): Atomic mass of nuclei

\(I\): Mean free path, \(v\): Neutron velocity.

The first time moment of neutron flux on the 290 K free gas model, \(<t>\Phi^{290K}\), and that on the crystal mode, i.e., Krumhansl-Brooks model, \(<t>\Phi^{\text{crystal}}\), are given respectively by

\[
<t>\Phi^{290K} = <t>\Phi^{0K}(1 + 1.0753 T/E_c) \tag{13}
\]

\[
<t>\Phi^{\text{crystal}} = <t>\Phi^{0K}(1 + 2.5402 T/E_c) \tag{14}
\]

where \(T\) is the moderator temperature.

Generally speaking, the slowing down density \(q(E,t)\) can be related to the neutron flux by

\[
q(E,t) = \int_{E_c}^{E} \sum_{E'} \sum_{E'_c} (E' \rightarrow E) \phi(E') dE dE'
\]

where \(\sum_{E'} \sum_{E'_c}\) : Scattering cross-section from \(dE'\) to \(dE\).

If, \(\Phi(E,t)\) is approximated by

\[
\Phi(E,t) = \Phi_{E=E_c}(E-E_c)
\]

on the 0 K free gas model, the right-hand side of Eq.(15):

\[
= \text{const.} \int_{E_c}^{E} \phi(E,t) + \frac{\partial \phi}{\partial E} \bigg|_{E=E_c}
\]

--- 25 ---
Therefore, by substituting Eq.(17) into Eq.(1), the slowing down time below $E_c$ on the $0^\circ K$ free gas model is given by,

\[
\frac{1-r^2 E/E_c}{(1-r^2) E/E_c} \frac{dE}{E} \approx \text{const.} \left[ \phi(E_c,t) + \frac{\partial \phi}{\partial E} \right]_{E=E_c} \frac{\ln r^2 - r^2/2 + 1/2 \tau^2}{\ln r^2 - (1-r^2)},
\]

where

\[
\phi(E_c+E_c, t) = \phi(E_c, t) \frac{\ln r^2 - r^2/2 + 1/2 \tau^2}{\ln r^2 - (1-r^2)}. \tag{18}
\]

which can be evaluated with the help of Eq.(12) by

\[
\frac{1}{1-2/3 (1+r^2)} \frac{1}{(1+r)} \int_0^\infty \phi(E_c, t) dt.
\]

The slowing down time below $E_c$ on the $300^\circ K$ free gas model and on the crystal model i.e., Krumhansl-Brooks model, are determined through a similar procedure using the relations of Eqs.(13) and (14).

On the other hand, the slowing down time to the energy $E$, can be also approximated by

\[
\int_0^\infty \phi(E_c, t) dt \int_0^\infty \phi(E_c, t) dt,
\]

because the neutron density can be expressed with good accuracy by the Gaussian function centered about the right-hand side of Eq.(20).

For the fuel containing moderators, a rigorous theoretical treatment has not been performed, and no comparison between experiment and theory was undertaken. However it can be inferred that the content of the fuel in this experiment would probably be too small to have significant effect.

VI. CONCLUSION

Slowing down times 1.25 and below 0.61, 0.50, 0.40 and 0.18 eV in graphite pile as well as in fuel containing graphite pile were measured by the transmission method. The measured results show no considerable difference between the pure graphite pile and the fuel containing graphite pile.

The pure graphite results showed fairly good agreement with the theoretical values obtained with account taken of chemical binding in the graphite lattice. It may thus be concluded that the effect of the chemical binding force in the graphite lengthens the neutron slowing down time.

The experimental data obtained include some information on the higher time moments of the slowing down density, so that a direct comparison between the measured counting ratio and that calculated from the various slowing down models is now possible, and planned to be undertaken in the future.

ACKNOWLEDGMENT

The authors wish to thank Messrs. A. Takahashi of the Osaka University and H. Wakabayashi of the University of Tokyo for their cooperation in the experiments.

REFERENCES