REACTOR

MASTER

SAFETY

DIVISION

BROOKHAVEN NATIONAL LABORATORY

Proton Chemical Binding in Water and its Effect
on Neutron Absorption in the 1 eV Resonance of ^{240}Pu

Arthur J. Buslik

November 1974

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

**Proton Chemical Binding in Water and its Effect
on Neutron Absorption in the 1 eV Resonance of ^{240}Pu ***

**Arthur J. Buslik
Department of Applied Science
Brookhaven National Laboratory
Upton, New York 11973**

November 1974

NOTICE
This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Energy Research and Development Administration, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

*Research carried out at Brookhaven National Laboratory under contract with the U. S. Atomic Energy Commission.

I. Introduction

This report discusses proton chemical binding in water and its effect on neutron absorption in the 1 eV resonance of Pu-240. Various methods of including these effects are presented, and a method is chosen on the basis of a numerical comparison to a calculation using the Haywood scattering kernel for hydrogen; this calculation serves as a standard.

The numerical results to be presented will show that the use of the approximate hydrogen scattering kernel suggested by Cady, Kirouac and McInerney² is sufficiently accurate for the calculation of the resonance absorption in the Pu-240 1 eV resonance. We will call this kernel the CKM kernel.

The implementation of a method based on the CKM kernel in the HAMMER code⁶ requires changing certain parameters (the slowing down power $\xi\sigma_s$ and the Greuling-Goertzel parameter γ) in the hydrogen epithermal library, and making certain changes to the program - first of all, to ensure that these parameters are used everywhere they should be (HAMMER at present implicitly assumes the hydrogen values of ξ and γ are unity, in certain of its subroutines) and secondly, to modify the Dancoff factor used in the calculation of the resonance integral for the 1 eV resonance of Pu-240. The model employed corrects not only the resonance absorption in the Pu-240 1 eV resonance, but also gives a more accurate epithermal flux so that other quantities will also be calculated more accurately.

The effects of proton binding and thermal motion on the resonance absorption in the 1 eV resonance of Pu-240 might also possibly be included in reactor calculations by raising the thermal cutoff energy to around 2 eV. To use this procedure in HAMMER would likely require increasing the number of thermal groups. Moreover, even if the number of thermal groups were increased from the present 30 to, say, 35, the multigroup cross sections required might be composition-dependent. In fact, Poncelet,¹¹ in his burnup physics calculations, employed burnup-dependent thermal multigroup cross sections (see p. 77 of Ref. 11). These burnup dependent cross sections were obtained by averaging over the multigroup using a neutron flux spectrum derived from a free proton gas kernel; this may not be entirely adequate. Moreover, Liikala et al.¹² have found (see p. 65 of Ref. 12) that when the Nelkin kernel for water was used with a thermal cutoff energy of 2 eV, numerical difficulties were encountered (oscillating behaviour in the flux spectrum) and have also chosen to use a model based on the CKM kernel. They have employed this model in a code which uses scattering transfer matrices instead of a Greuling-Goertzel model of slowing-down, and have made no test on the accuracy of the method.

Epithermal proton chemical binding effects could also be taken into account to a certain extent by a method described by J. Hardy, Jr.¹⁵ In this method, values of σ_0 for hydrogen are obtained so as to reproduce the Corngold asymptotic expansion¹⁶ fluxes exactly in a multigroup calculation for plain

water, keeping the parameter γ equal to unity. This method does not, however, correct the resonance integral calculation for the Pu-240 1 eV resonance; moreover, allowing γ to vary (as is done when σ_0 and γ are derived from the CKM kernel) gives one the potential of improved epithermal flux shapes in strongly absorbing media.

The various approximate methods are compared to the Haywood scattering kernel for a homogeneous mixture of Pu-240 and hydrogen. In addition, for a heterogeneous medium, the resonance integral for the 1 eV resonance of Pu-240 was calculated using the intermediate resonance approximation, and the change in resonance absorption due to the chemical binding and thermal motion of the proton is calculated, using the approximate method derived in Ref. 1.

II. The Calculations with the Haywood Scattering Kernel

Calculations using the Haywood scattering kernel⁸ were performed by first using the FLANGE code³ to obtain the scattering kernel from the $S(\alpha, \beta)$ scattering law on the ENDF/B-III tape (ENDF/B tape label No. 320). Then a code was written to solve the homogeneous problem of a mixture of Pu-240, a background $\frac{1}{v}$ absorber, and water.

Eighty-five energy groups were used, with an upper energy cut of 2 eV. Forty energy groups were used in the energy range from .85 eV to 1.30 eV, so that the Pu-240 resonance absorption would be accurately calculated. The FLANGE kernel was calculated for a water temperature of 500°K. (This did not require interpolation between scattering laws at different temperatures). An effective temperature of 1437°K was used. This temperature is used to calculate $S(\alpha, \beta)$ for energy transfers larger than those tabulated in the ENDF file, and to compute the scattering kernel for these energy transfers. The value of the effective temperature used, 1437°K, corresponds to a Nelkin kernel¹⁴ and not a Haywood kernel. The effects on the results are thought to be quite small. (The Haywood kernel value of the effective temperature is about 1370°K).

The flux in the homogeneous mixture of water, $\frac{1}{v}$ absorber, and Pu-240 was calculated by a computer code written especially for this task (the UPS code). Only scattering from hydrogen was considered (i.e., energy transfer by the oxygen and Pu-240 was neglected). The Pu-240 resonance absorption cross section

was calculated with Doppler broadening neglected. It seems quite reasonable to assume that any approximate means of taking into account the effects of proton chemical binding on Pu-240 resonance absorption which is accurate for this model problem will also be accurate for the heterogeneous problem with Doppler broadening and non-hydrogenous moderation included.

The equation solved by the UPS code is a numerical approximation to

$$[\Sigma_s(E) + \Sigma_a(E)]\phi(E) = \int_0^{E_c} \Sigma_s(E \leftarrow E')\phi(E')dE' + S(E)$$

Here, $\phi(E)$ is the flux, $\Sigma_s(E)$ is the macroscopic scattering cross section of hydrogen, $\Sigma_s(E \leftarrow E')$ is the hydrogen scattering kernel, and $S(E)$ is the source to energy E due to neutrons whose last scattering collision occurred above the upper energy cutpoint E_c . The energy shape of $S(E)$ is assumed to be that associated with a free gas kernel at an effective temperature T^* , so that

$$S(E) = \text{const} \times \text{erf} \sqrt{\frac{E}{kT^*}}.$$

The constant is chosen such that $\int_0^{E_c} S(E)dE = 1$. The absorption cross section consists of a sum of a $\frac{1}{v}$ part and the resonance absorption cross section of the 1.056 eV resonance of the Pu-240,

$$\Sigma_a(E) = \frac{k_v}{\sqrt{E}} + \frac{N \sigma_0 \frac{E}{E_0}}{1 + \left(\frac{E - E_0}{\Gamma/2}\right)^2} \quad (2)$$

where N is the number density of Pu-240, k_v is an input constant giving the magnitude of the $\frac{1}{\sqrt{E}}$ absorption, and the other symbols have their conventional meanings.

When an energy mesh $\{E_i, i=1, 2, \dots, N\}$ is chosen and the integral on the right-hand side of Eq. (1) approximated by a quadrature rule, one obtains

$$[\Sigma_s(E_i) + \Sigma_a(E_i)] \phi(E_i) = \sum_{j=1}^N \Sigma_s(E_i \leftarrow E_j) W_j \phi(E_j) + S(E_i) \quad (3)$$

$i = 1, 2, \dots, N$

where the W_j are the integration weights. An integration rule was chosen such that the points E_i were midpoints of energy intervals of length W_i .

An option in running the FLANGE problem was chosen such that the diagonal elements of the kernel were adjusted so that

$$\Sigma_s(E_i) = \sum_{j=1}^N \Sigma_s(E_j \leftarrow E_i) W_j \quad (4)$$

holds exactly.

Equation (3) was solved by the following iterative method:

$$\varphi^{(n)}(E_i) = \frac{1}{D_i} \sum_{j,i} \sum_s (E_i \leftarrow E_j) W_j \varphi^{(n-1)}(E_j) + \frac{1}{D_i} S(E_i) \quad (5)$$

where

$$D_i = \sum_s (E_i) + \sum_a (E_i) - \sum_s (E_i \leftarrow E_i) W_i \quad (6)$$

and $\varphi^{(n)}$, $\varphi^{(n-1)}$ are the n^{th} and $(n-1)^{\text{th}}$ iterates. This is a conventional point Jacobi iterative method.⁴ The iterations were terminated when

$$|\varphi^{(n)}(E_i) - \varphi^{(n-1)}(E_i)| < \epsilon$$

for all i . Here, ϵ is an input parameter. Convergence was checked by seeing how closely the integral of the source equalled the absorption rate.

In these problems, a value of $\epsilon = .001$ was used, and neutron balance held within a few tenths of a percent. (In a very slowly converging problem, such as one with a very small ratio of absorption to scattering, it would be necessary to use a tighter convergence criterion.)

The results of the calculations with the Haywood kernel were compared to the results with an effective temperature kernel, and with the CKM kernel. These results will be given after a brief description of these two kernels is given. In addition, the possibility of a Greuling-Goertzel type kernel with moments inferred from the Haywood kernel will be considered.

III. The Effective Temperature Kernel

In the short collision time approximation⁵ the scattering kernel for hydrogen is, to a first approximation, a free proton scattering kernel at an effective temperature T^* . The short collision time approximation is valid for large energy transfer (i.e., energy transfers somewhat larger than the largest vibrational frequency of the water molecule, which occurs at about .48 eV). Since, for large incident neutron energy most neutrons scatter with large energy transfer, it would appear reasonable to use an effective temperature kernel at incident neutron energies on the order of 1 eV. However, an effective temperature kernel obeys detailed balance at the effective temperature T^* instead of at the true moderator temperature T . This means that if the downscattering part of the effective temperature kernel were correct, then too many neutrons would be upscattered. (This is a consequence of the fact that the effective temperature is much larger than the true temperature.) One might therefore infer that with an effective temperature kernel too many neutrons would be scattered from below the Pu-240 1 eV resonance into the resonance, and consequently that the resonance absorption would be overestimated. As will be seen, the numerical results obtained show that this is indeed the case.

1V. The CKM Kernel

The CKM kernel modifies the effective temperature kernel by assuming that all those neutrons which would upscatter in an effective temperature model scatter without energy change. In this way the excessive upscattering in the effective temperature model is corrected.

Figure 1 gives curves of $\sigma_{SH}(E_i \leftarrow E_j)$ vs. E_i for $E_j = 1.0581$ eV and $E_j = .6064$ eV, for both the Haywood kernel and the effective temperature kernel. (Here, $\sigma_{SH}(E_i \leftarrow E_j)$ is the hydrogen scattering kernel for scattering from E_j to E_i .) One sees that, indeed, there is too much upscattering in the effective temperature model, and that the approximation of Cady, Kirouac and McInerney² of assuming that the kernel is given by the downscattering part of the effective temperature kernel plus a term proportional to $\delta(E-E')$ (corresponding to scattering without energy change) appears reasonable.

As will be seen, the CKM kernel gave good agreement with the Haywood scattering kernel; it gave a slight underestimate of the resonance absorption in the Pu-240 1 eV resonance, as might be expected from its not having any upscattering of neutrons into the resonance from energies below the resonance.

The CKM kernel enables proton chemical binding effects to be taken into account very simply in the HAMMER code.⁶ If scattering from hydrogen is calculated as if the proton were free and at rest, then the slowing down density due to hydrogen q_H is related to the flux by

$$\frac{\partial q_H}{\partial u} + f_H(u) = \Sigma_s \phi(u) \quad (7)$$

But, as will be shown below, if the CKM kernel is used for scattering by hydrogen, then q_H is related to ϕ by

$$\gamma_H \frac{\partial q_H}{\partial u} + f_H = \xi_H \Sigma_{SH} \phi \quad (8)$$

where the parameters γ_H and $\xi_H \Sigma_{SH}$ will be determined below.

Equation (8) is of the form of the Greuling-Goertzel relation, and consequently the use of the CKM kernel may be implemented in HAMMER by the modification of the parameter γ_H and the slowing-down power $\xi_H \Sigma_{SH}$ for hydrogen.

The parameters γ_H and $\xi_H \Sigma_{SH}$ are determined as follows. In the CKM approximation the downscattering part of the kernel is given by the downscattering part of the effective temperature kernel, so that

$$\Sigma_s(E \leftarrow E') = \Sigma_s \exp\left(-\sqrt{\frac{E}{2T}}\right) \frac{1}{E'}, \quad E' > E \quad (9)$$

where T is the asymptotic value of the hydrogen scattering cross-section at energies above the region where chemical binding and thermalization effects are important. The up-scattering part of the effective temperature kernel is replaced by scattering without energy change in the CKM model, and this scattering, like that of the scattering by an infinitely heavy scatterer, does not contribute to the slowing-

down density. Accordingly, the slowing-down density is given by

$$\begin{aligned} q(E) &= \int_0^E dE'' \int_E^\infty \Sigma_s(E'' \leftarrow E') \phi(E') dE' \\ &= \int_0^E dE'' \int_E^\infty \exp\left(-\sqrt{\frac{E''}{kT^2}}\right) \frac{\Sigma_s}{E'} \phi(E') dE', \end{aligned} \quad (10)$$

where Eq. (9) has been used. We obtain from Eq. (10),

$$q(E) = \int_E^\infty \left\{ \int_0^E \exp\left(-\sqrt{\frac{E''}{kT^2}}\right) dE'' \right\} \frac{\Sigma_s}{E'} \phi(E') dE' \quad (11)$$

But

$$\int_0^E \exp\left(-\sqrt{\frac{E''}{kT^2}}\right) dE'' = (E - \frac{1}{2}kT^2) \exp\left(-\sqrt{\frac{E}{kT^2}}\right) + \frac{1}{2\sqrt{kT^2}} \sqrt{\frac{E}{kT^2}} e^{-E/kT^2} \quad (12)$$

and for energies in the epithermal range

$$\int_0^E \exp\left(-\sqrt{\frac{E''}{kT^2}}\right) dE'' = E - \frac{1}{2}kT^2 \quad (13)$$

is a very good approximation. Therefore, from Eqs. (11) and (13),

$$q(E) = (E - \frac{1}{2}kT^2) \int_E^\infty \frac{\Sigma_s}{E'} \phi(E') dE' \quad (14)$$

Therefore,

$$\frac{d\phi}{dE} = \int_E^{\infty} \frac{\Sigma}{E'} \phi(E') dE' - (E - \frac{1}{2} kT^*) \frac{\Sigma}{E} \phi(E) \quad (15)$$

Solving Eq. (14) for the integral on the right-hand side, and substituting into Eq. (15) one obtains

$$\frac{d\phi}{dE} = \frac{\phi(E)}{E - \frac{1}{2} kT^*} - (E - \frac{1}{2} kT^*) \frac{\Sigma}{E} \phi(E) \quad (16)$$

or

$$-E \left(1 - \frac{1}{2} \frac{kT^*}{E}\right) \frac{d\phi}{dE} + \phi(E) = E \left(1 - \frac{1}{2} \frac{kT^*}{E}\right)^2 \Sigma \phi(E) \quad (17)$$

Converting from the independent variable E to the lethargy u ,

$$\left(1 - \frac{1}{2} \frac{kT^*}{E}\right) \frac{d\phi}{du} + \phi(u) = \left(1 - \frac{1}{2} \frac{kT^*}{E}\right)^2 \Sigma \phi(u) \quad (18)$$

Comparing Eq. (18) to Eq. (8) we see that

$$\gamma_H = 1 - \frac{1}{2} \frac{kT^*}{E}, \quad \Sigma_{sH} = \left(1 - \frac{1}{2} \frac{kT^*}{E}\right)^2 \Sigma \quad (19)$$

The value of Σ_{sH} obtained agrees with that obtained by Cadz et al.⁴; however, the value of γ_H given in reference 2 is simply unity.

Another way of obtaining the values of $\epsilon_H \Sigma_{SH}$ and γ_H which correspond to the CKM kernel is to make use of the solution to the equation for the neutron flux in an infinite medium consisting of an absorber and hydrogen, when the CKM kernel is used. This solution is given in the paper by Cady et al.,² and may be written in the form

$$\phi(E) = \frac{q(E)}{E \left\{ \left(1 - \frac{1}{2} \frac{A_T^0}{E}\right)^2 \Sigma + \left(1 - \frac{1}{2} \frac{A_T^0}{E}\right) \Sigma_a \right\}} \quad (20a)$$

$$q(E) = \exp \left(- \int_E^\infty \frac{dE'}{E'} \frac{\Sigma_a(E')}{\left(1 - \frac{1}{2} \frac{A_T^0}{E'}\right) \Sigma + \left(1 - \frac{1}{2} \frac{A_T^0}{E'}\right) \Sigma_a} \right) \quad (20b)$$

Differentiating Eq. (20b) we find that

$$E \frac{dq}{dE} = q(E) \frac{\Sigma_a(E)}{\left(1 - \frac{1}{2} \frac{A_T^0}{E}\right)^2 \Sigma + \left(1 - \frac{1}{2} \frac{A_T^0}{E}\right) \Sigma_a} \quad (20c)$$

$$= q(E) \frac{1}{\left(1 - \frac{1}{2} \frac{A_T^0}{E}\right)} \left[1 - \frac{\left(1 - \frac{1}{2} \frac{A_T^0}{E}\right)^2 \Sigma}{\left(1 - \frac{1}{2} \frac{A_T^0}{E}\right)^2 \Sigma + \left(1 - \frac{1}{2} \frac{A_T^0}{E}\right) \Sigma_a} \right]$$

Using Eq. (20a) in Eq. (20c),

$$E \frac{dq}{dE} = q(E) \frac{1}{1 - \frac{1}{2} \frac{A_T^0}{E}} - \left(1 - \frac{1}{2} \frac{A_T^0}{E}\right) \Sigma_a \phi(E) \quad (20d)$$

Eq. (20d) may be written as

$$- \left(1 - \frac{1}{2} \frac{A_T^0}{E}\right) \Sigma_a \phi(E) + q(E) = \left(1 - \frac{1}{2} \frac{A_T^0}{E}\right)^2 \Sigma \phi(E) \quad (20e)$$

which is identical to Eq. (17). It then follows that after converting to the lethargy u as the independent variable an equation of the form of Eq. (8) is obtained, with γ_H and $\bar{\sigma}_H^T$ given by Eq. (19).

In addition to the changes in $\bar{\sigma}_s$ and γ for hydrogen, it is necessary to make changes to the resonance integral treatment, if the effects of the proton chemical binding and thermal motion on the Pu-240 1 eV resonance absorption are to be taken into account properly. These changes stem from the fact that the chemical binding and thermal motion of the protons makes it less likely that a neutron travelling at resonance energy will be scattered out of the resonance energy range on a collision with a proton. Consequently, the Dancoff factor is modified. The theory of this correction is described in reference 1, and leads to

$$\frac{1}{D_{eff}} = \frac{1}{D} + \left(\frac{\gamma}{\bar{\sigma}} - 1 \right) \frac{1}{\bar{\sigma}_s(E_0) \bar{L}_m}, \quad (21)$$

where the γ , $\bar{\sigma}$ are evaluated at the resonance energy E_0 , $\bar{\sigma}_s(E_0)$ is the moderator hydrogen scattering cross-section at energy E_0 , D is the uncorrected Dancoff factor, D_{eff} is the effective Dancoff factor with proton chemical binding effects included, and $\bar{L}_m$ is the mean chord length $\frac{4V}{S}$ for the moderator. Now HAMMER sometimes treats the lattice escape probabilities not by a Dancoff factor, but by the method of cosine currents. The treatment of the resonance integral may be extended to this case by noting that in the rational approximation,

$$\frac{1}{D} = 1 + \frac{1}{\bar{\sigma}_s(E_0) \bar{L}_m} \quad (22)$$

Then, from Eqs. (21) and (22),

$$\frac{1}{\Sigma_{eff}} = 1 + \frac{1}{(\xi/H) \Sigma_s(E_0) \bar{L}_m} \quad (23)$$

Thus, the effect of proton chemical binding and thermal motion on the lattice escape probabilities may be included by using $(\xi/\gamma) \Sigma_s(E_0)$ in place of $\Sigma_s(E_0)$ in the calculation of the lattice escape probabilities, when these are calculated by the cosine current method. For simplicity, we have chosen to use the same technique even when HAMMER determines the lattice escape probabilities using a Dancoff factor instead of the cosine current method. That is to say, the Dancoff factor is calculated with $\frac{\xi}{\gamma} \Sigma_s(E_0)$ replacing $\Sigma_s(E_0)$ in the calculation of the Dancoff factor. Moreover, the Pu-240 1 eV resonance at typical Pu-240 concentrations is essentially wide with respect to scattering by oxygen, or isotopes heavier than oxygen. (More precisely, in a typical case, the intermediate resonance parameter for the oxygen (in the fuel pin) in the intermediate resonance approximation of Goldstein and Cohen¹⁰ is about 1/4, so that only $\frac{1}{4}$ of the oxygen scattering contributes to removal from the resonance energy range.) Consequently, in calculating the Dancoff factor in the multigroup (group 53) in which the 1 eV resonance of Pu-240 occurs, only the hydrogen scattering is included in the removal cross section on which the Dancoff factor is based.

It turns out that the HAMMER program does not use the ξ/γ and γ parameters for hydrogen which are on the library

tape in all of its subroutines; in some portions of the code (in particular, subroutine *SORE*) ϵ and γ are assumed equal to unity regardless of their values on the library tape. This must be modified if the HAMMER program is to calculate resonance absorption properly in those neutron energy groups where the effects of proton chemical binding and thermal motion are important. If these changes are not made, the group resonance escape probability will not be calculated properly, even if the resonance integrals are calculated properly.

A good general reference describing the HAMMER code and the computational model it uses is a paper by Jabbawy, Karni, Rothenstein and Velner.¹³

V. Greuling-Goertzel Parameters from the Haywood Kernel

Instead of using the values of $\xi_H \sigma_{SH}$ and γ_H derived from the Q kernel, one might try to define them such that ξ_H is the mean lethargy charge per collision, and $2\gamma_H \xi_H$ the mean squared lethargy charge per collision, in the same way that these parameters are defined for a Greuling-Goertzel synthetic kernel approximation⁷ to heavy element slowing down when the target nucleus is assumed free and at rest. Then

$$\xi_H \sigma_{SH}(E) = \int_0^\infty d\eta \frac{E}{E'} \sigma_{SH}(E' \leftarrow E) dE' \quad (24)$$

$$2 \xi_H \gamma_H(E) \sigma_{SH}(E) = \int_0^\infty \left(d\eta \frac{E}{E'} \right)^2 \sigma_{SH}(E' \leftarrow E) dE' \quad (25)$$

Consider an infinite medium consisting of hydrogen and an absorber. Let us assume that a Greuling-Goertzel type relationship between q_H and ϕ holds, as given by Eq. (8). We also have

$$\frac{dq}{du} = - \Sigma_a \phi \quad (26)$$

Substituting Eq. (26) into Eq. (8) and solving for ϕ , we obtain

$$\phi(u) = \frac{q(u)}{\gamma_H \Sigma_a(u) + \xi_H \Sigma_{SH}(u)}$$

or

$$\phi(E) = \frac{1}{E} \frac{q(E)}{\frac{1}{4} \Sigma_a + \frac{5}{4} \Sigma_{sh}} \quad (27)$$

Suppose now that we obtain $q(E)$ from a numerical calculation using the Haywood kernel. Suppose further that we compute Σ_{HSH} and γ_H from Eqs. (24) and (25) and then compute $\phi(E)$ from Eq. (27). We can compare this $\phi(E)$ to the $\phi(E)$ obtained from the numerical calculation. For the homogeneous problem discussed later this was done for $E = 1.2944$ eV. For this problem, at this energy

$$\begin{aligned} q(E) &= .968194 \text{ neutrons/cm}^2 \\ \left. \begin{aligned} \Sigma_{HSH} &= 16.951 \text{ barns} \\ \gamma_H &= .82234 \end{aligned} \right\} && \text{From Eqs. (24) and (25).} \\ \Sigma_a(E) &= .096196 \text{ cm}^{-1} \\ N^H &= .03 / \text{barn-cm} \quad (\text{number density of hydrogen}) \end{aligned} \quad (28)$$

Using the data of Eqs. (28) in Eq. (27) we find that

$$\phi(E) = 1.2729 \text{ neutrons/cm}^2 \cdot \text{eV} \quad (29)$$

However, the numerical calculation using the UPS code gave

$$\phi(E) = 1.1703 \text{ neutrons/cm}^2 \cdot \text{eV} \quad (\text{UPS code}) \quad (30)$$

The error in the flux at this energy is therefore 8.8%, when γ_H and Σ_{HSH} are defined in terms of the moments of the kernel,

as given by Eqs. (24) and (25).

The values of γ_H and $\epsilon_H \Sigma_{SH}$ based on the CKM kernel (see Eqs. (19)) give a much better value of $\phi(E)$. Using Eqs. (19) with $kT^* = .1238$ eV (corresponding to a material temperature of 500°K with Nelkin kernel parameters), we find that at $E = 1.2944$ eV

$$\begin{aligned} (\int \epsilon_{SH})_{\text{CKM}} &= 18.496 \text{ eV} \\ \gamma_{\text{CKM}} &= .952 \end{aligned} \quad (31)$$

A computation of $\phi(E)$ from Eq. (27) yields

$$\phi(E) = 1.1570 \text{ neutrons/cm}^2\text{-sec} \quad (32)$$

which differs from the value calculated by UPS (Eq. (30)) by only -1.1%. It seems clear, therefore, that the calculation of ϵ_{SH} and γ by Eqs. (19) is preferable to that of calculating them in terms of the moments of the Haywood kernel, as given by Eqs. (24) and (25).

VI. Numerical Results for the Homogeneous Problem

The homogeneous test problem used is described (in general outline) under the description of the UPS code given in section II. The atom density of Pu-240 used was

$$N^{240} = 8.5 \times 10^{-5} \text{ atoms/bn-cm}$$

while that of hydrogen was

$$N^H = .03 \text{ atoms/bn-cm}$$

The $\frac{1}{v}$ background absorption cross section (see Eq. (2)) was

$$\Sigma_a(E) = \frac{h_r}{\sqrt{E}}, \quad h_r = .035 \frac{\text{b-cm}}{\sqrt{\text{eV}}}$$

The moderator temperature was 500°K.

Table I gives a comparison of the absorption rates in various energy ranges using the Haywood scattering kernel, the effective temperature kernel, the CKM kernel, and a kernel which neglects all chemical binding and thermal motion effects (free proton at rest kernel). It is seen that the CKM kernel gives the best agreement with the Haywood scattering kernel results.

One should point out that although the Haywood scattering kernel results are used as a standard here, there are errors here, not only in inaccuracies in the phonon spectrum but also in the numerical procedures used in GASKET⁹ to obtain the scattering law which is on the ENDF/B tape, and in the numerical procedures used in the FLANGE code to obtain

Table I

<u>Energy Range</u> (eV)	<u>Haywood Kernel</u>	<u>Absorption Rates</u>		<u>CKM Kernel</u>
		<u>Free H</u> <u>at rest Kernel</u>	<u>Effective Temp-</u> <u>erature Kernel</u>	
(.84 , .905)	.01182	.01056 (-10.6%)	.01490 (26.0%)	.01144 (-3.2%)
(.905,1.130)	.1312	.1210 (- 7.7%)	.1494 (13.8%)	.1281 (-2.3%)
(1.130,1.317)	.0378	.03450 (- 8.6%)	.03850 (1.9%)	.03695 (-2.2%)
(1.317,2.0)	.0318	.02900 (- 8.8%)	.03205 (.8%)	.03107 (-2.3%)

the scattering kernel from the scattering law function $S(\alpha, \beta)$. It is unlikely that these errors would affect the resonance absorption by more than about 1%.

The effective temperature kernel results were obtained in the same way as the Haywood scattering kernel results were obtained, by using the UPS code with the effective temperature scattering kernel calculated with analytical formulas.

The equation for the flux which is solved when the CKM kernel is used may be derived as follows. The complete kernel, including the $\delta(E-E')$ contribution, is given by

$$\Sigma_s(E \leftarrow E') = \begin{cases} \Sigma \text{inf} \sqrt{E/RT^*} \frac{1}{E'} + \Sigma \frac{RT^*}{E} \delta(E-E'), & E' \geq E \\ 0, & E' < E \end{cases} \quad (33)$$

The source term $S(E)$ of Eq. (1) is due to the scattering of neutrons from above E_c to energies below E_c , and is given in this model by

$$S(E) = \text{const} \times \text{inf} \sqrt{\frac{E}{RT^*}}$$

The normalization condition

$$\int_0^{E_c} S(E) dE = 1$$

leads to

$$S(E) = \frac{\text{inf} \sqrt{E/RT^*}}{E_c - \frac{1}{2} RT^*} \quad (34)$$

where use has been made of Eq. (13). Moreover, in the short collision time approximation the scattering cross section is

given to sufficient accuracy by

$$\Sigma_s(E) = \Sigma \left(1 + \frac{1}{2} \frac{kT^*}{E} \right) \quad (35)$$

Substituting Eqs. (33), (34) and (35) into Eq. (1), one obtains

$$\left[\Sigma_s(E) + \Sigma \left(1 + \frac{1}{2} \frac{kT^*}{E} \right) \right] \phi(E) = \Sigma \frac{kT^*}{E} \phi(E) + \Sigma \operatorname{erf} \left(\sqrt{\frac{E}{kT^*}} \right) \int_E^{\infty} \frac{\phi(E')}{E'} dE' + \frac{\operatorname{erf} \sqrt{E/kT^*}}{E_0 - \frac{1}{2} kT^*} \quad (36)$$

Since at epithermal energies $\operatorname{erf} \sqrt{E/kT^*} = 1$ is a good approximation, we may write

$$\left[\Sigma_s(E) + \Sigma \left(1 + \frac{1}{2} \frac{kT^*}{E} \right) \right] \phi(E) = \Sigma \frac{kT^*}{E} \phi(E) + \Sigma \int_E^{\infty} \frac{\phi(E')}{E'} dE' + \frac{1}{E_0 - \frac{1}{2} kT^*} \quad (37)$$

The integral on the right-hand side of Eq. (37) is approximated by a quadrature rule (the same one which was used for the Haywood scattering kernel). Because there is only downscattering, the fluxes $\phi(E_i)$ may be solved for successively without iteration.

The energy mesh was the same for all the approximation methods so that any error due to finite mesh intervals would be minimized.

VII. The Heterogeneous Problem

As mentioned in section IV, the effects of the chemical binding and thermal motion of the protons on the absorption in the Pu-240 1 eV resonance may be taken into account by modifying the calculation of the Dancoff factor, in the way given by Eq. (21), and also by using the values of $\Sigma_H \Sigma_{SH}$ and γ_H consistent with the CKM kernel in the epithermal flux calculation. The resonance absorption in the Pu-240 1 eV resonance is given by

$$f \left(1 - e^{-N I V_0 / \Sigma_H \Sigma_{SH} V_1} \right) \approx \frac{N I V_0}{\Sigma_H \Sigma_{SH} V_1} \bar{\phi} \quad (38)$$

where $\bar{\phi}$ is the off-resonance flux, I is the resonance integral, $\Sigma_H \Sigma_{SH}$ is the slowing down power of the hydrogen in the moderator, N is the number density of Pu-240 in the rod, and V_0 and V_1 are the fuel rod and moderator volumes, respectively.

For a test heterogeneous case, the resonance integral was calculated with a Dancoff factor uncorrected for proton chemical binding, and also with the corrected Dancoff factor. In addition, the effect on the resonance absorption due to the deviation of the value of $\Sigma_H \Sigma_{SH}$ from the asymptotic value was computed. The test case consisted of a fuel pin containing a Pu-240 atom-density of .00023 atoms/bn-cm. The pellet radius was .47 cm, and the fuel pin temperature was taken as 1000°K. The scattering due to oxygen in the fuel was treated in the IR approximation, as was the uranium and the plutonium.

Interference scattering was neglected. The uncorrected Dancoff factor employed was $D = 1 - C = .682$. Equation (21) was used to derive the Dancoff factor corrected for the proton chemical binding and thermal motion. The corrected value obtained was $D_{\text{eff}} = 0.6477$. The resonance integral for the 1 eV resonance of Pu-240 was 1267 barns using the uncorrected Dancoff factor and 1230 barns using the corrected Dancoff factor, a decrease of 3%. The resonance absorption is proportional, according to Eq. (38), to the resonance integral I and inversely proportional to $\xi_H \Sigma_{\text{SH}}$. If the value of $\xi_H \Sigma_{\text{SH}}$ uncorrected for chemical binding effects is taken as the asymptotic value of $\Sigma_{\text{SH}} = N^H \times 20.4 \text{ cm}^{-1}$ (where N^H = number density of hydrogen in atoms/bn-cm), and if the corrected value is obtained from Eq. (19), then it turns out that

$$\xi_H \Sigma_{\text{SH}} = \left(1 - \frac{4I^v}{E_0}\right)^2 \Sigma = .886 \Sigma$$

Thus, the ratio of the resonance absorption uncorrected for chemical binding effects to the resonance absorption corrected for chemical binding effects is

$$\frac{\left(\frac{I}{\xi \Sigma_0}\right)_{\text{uncorrected}}}{\left(\frac{I}{\xi \Sigma_0}\right)_{\text{corrected}}} = .886 \frac{I_{\text{uncorrected}}}{I_{\text{corrected}}}$$

so that the net error in resonance absorption due to the neglect of chemical binding is approximately 9%.

VIII. Summary and Conclusions

Through numerical comparisons of the CKM kernel of reference 2 to the Haywood scattering kernel, when these are used to compute absorption rates in a mixture of Pu-240 and hydrogen, it has been shown that the CKM kernel can be used as the basis for including the effects of proton binding and thermal motion in the epithermal flux range and in particular the effects on the resonance absorption in the 1 eV resonance of Pu-240. The method of implementing the use of this kernel in the HAMMER program was discussed. For a heterogeneous medium estimates of the magnitude of proton binding and thermal motion effects on the resonance absorption in the 1 eV resonance of Pu-240 were made; the neglect of the proton binding and thermal motion produced an underestimate of about 9% in the resonance absorption, in this case.

References

1. A. Buslik, "Calculational Model for the Resonance Absorption in the Pu^{240} 1 eV Resonance, BNL 19046/ RP-1014, June 1974.
2. K. B. Cady, G. J. Kirouac, and J. J. McInerney, Nucl. Sci. Eng. 29, 299 (1967).
3. H. C. Honeck and D. R. Finch, "FLANGE-II, A Code to Process Thermal Neutron Data from an ENDF/B Tape", DP-1278, Savannah River Laboratory, E. I. duPont de Nemours & Co., Aiken, So. Carolina (1971).
4. R. S. Varga, Matrix Iterative Methods, Prentice-Hall, New Jersey, 1962, pp. 56 ff.
5. M. M. R. Williams, The Slowing Down and Thermalization of Neutrons, John Wiley & Sons, Inc., New York (1966), p. 58 ff.
6. J. E. Suich and H. C. Honeck, "The HAMMER System - Heterogeneous Analysis by Multigroup Methods of Exponentials and Reactors", DP-1064, Savannah River Laboratory (January, 1967).
7. G. Goertzel and E. Greuling, Nucl. Sci. Eng. 7, 60 (1960).
8. J. U. Koppel, "Neutron Scattering by Hydrogenous Moderators", in Reactor Physics in the Resonance and Thermal Regions, edited by A. J. Goodjohn and J. C. Pomraning, MIT Press, Mass., Vol. 1, pp. 27 ff. See also GA-7055 with the same author and title.

9. J. U. Koppel, J. R. Triplett and Y. D. Haliboff, "GASKET - A Unified Code for Thermal Neutron Scattering", GA-7417 (Rev.), March 1967.
10. B. R. Sehgal and R. Goldstein, Nucl. Sci. Eng. 25, 174 (1966); also, Goldstein and Cohen, Nucl. Sci. Eng. 11, 132 (1962).
11. C. G. Poncelet, "Burnup Physics of Heterogeneous Reactor Lattices", WCAP-6069 (June 1965).
12. R. C. Liikala, V. O. Uotinen, and U. P. Jenquin, "Uncertainties in the Analysis of Plutonium-Fueled Light Water Moderated Assemblies", BNWL-1656 (May 1973).
13. S. Jabbawy, J. Karni, W. Rothenstein and S. Veiner, "Water-Moderated Reactor Analysis with ENDF/B Data", in Nuclear Data in Science and Technology, Vol. II, Proceedings of a Symposium, Paris, March 12 to 16, 1973, International Atomic Energy Agency (1973), paper IAEA-SM-170/B.
14. M. Nelkin, Phys. Rev. 119, 741 (1960).
15. J. Hardy, Jr., Nucl. Sci. Eng. 27, 135 (1962).
16. N. Corngold, Ann. Phys. 11, 338 (1960).

A COMPARISON OF THE HAYWOOD KERNEL TO THE EFFECTIVE TEMP. KERNEL

FIGURE 1.