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PHYSICS RESEARCH QUARTERLY REPORT

OCTOBER, NOVEMBER, DECEMBER, 1965

BY

THE STAFFS
of
REACTOR PHYSICS,
EXPERIMENTAL PHYSICS RESEARCH
and
CRITICAL MASS PHYSICS

JANUARY 14, 1966

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of
Reactor Physics,
Experimental Physics Research,
and
Critical Mass Physics

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LIGHT MODERATOR REACTOR PHYSICS

Integrated Burnup Equations for Uranium-Plutonium Systems - R. P. Matsen

The equations governing burnup analysis can be written in closed form for the case in which microscopic cross sections remain constant as a function of exposure. The simplicity brought about by the closed form of these equations would make worthwhile an attempt to fit them to experimental data by the method of least squares. The success of such a venture would point the way to predicting isotopic concentrations during burnup of power reactor systems, assuming, of course, that a suitable method of determining microscopic cross sections is available.

Neglecting neutron capture by U^{236} and Pu^{242} and the decay of Pu^{241}, the differential equations governing burnup for plutonium-uranium systems are the following:

1a. $\frac{dN^{25}}{d\tau} = -\sigma_a^{25} N^{25}$

1b. $\frac{dN^{25}}{d\tau} = -\sigma_a^{25} N^{25}$

2a. $\frac{dN^{26}}{d\tau} = \sigma_c^{25} N^{25}$

2b. $\frac{dN^{26}}{dN^{25}} = -\frac{\sigma_c^{25}}{\sigma_a^{25}}$

3a. $\frac{dN^{28}}{d\tau} = -\sigma_a^{28} N^{28}$

3b. $\frac{dN^{28}}{dN^{25}} = d\,\frac{N^{28}}{N^{25}}$

4a. $\frac{dN^{49}}{d\tau} = \sigma_c^{28} N^{28} - \sigma_a^{49} N^{49}$

4b. $\frac{dN^{49}}{dN^{25}} = -\delta d\,\frac{N^{28}}{N^{25}} + a\,\frac{N^{49}}{N^{25}}$

5a. $\frac{dN^{40}}{d\tau} = \sigma_c^{49} N^{49} - \sigma_a^{40} N^{40}$

5b. $\frac{dN^{40}}{dN^{25}} = -\alpha a\,\frac{N^{49}}{N^{25}} + b\,\frac{N^{40}}{N^{25}}$

6a. $\frac{dN^{41}}{d\tau} = \sigma_a^{40} N^{40} - \sigma_a^{41} N^{41}$

6b. $\frac{dN^{41}}{dN^{25}} = -b\,\frac{N^{40}}{N^{25}} + c\,\frac{N^{41}}{N^{25}}$

7a. $\frac{dN^{42}}{d\tau} = \sigma_c^{41} N^{41}$

7b. $\frac{dN^{42}}{dN^{25}} = -\gamma c\,\frac{N^{41}}{N^{25}}$

where: $d = \frac{\sigma_a^{28}}{\sigma_a^{25}}$, $\delta = \frac{\sigma_c^{28}}{\sigma_a^{28}}$, $a = \frac{\sigma_a^{49}}{\sigma_a^{25}}$, $\alpha = \frac{\sigma_c^{49}}{\sigma_a^{49}}$, $b = \frac{\sigma_a^{40}}{\sigma_a^{25}}$, $c = \frac{\sigma_a^{41}}{\sigma_a^{25}}$, $\gamma = \frac{\sigma_c^{41}}{\sigma_a^{41}}$

and τ is the exposure (flux time). The differential equations have been cast into two equivalent forms; the 'b' set having been obtained by dividing the 'a' set by Equation 1a.

Assuming that the microscopic cross sections remain constant as a function of exposure, the differential equations can be solved analytically using standard techniques. In particular, proper choices of integrating factors facilitate solving set 'b'. With the zero subscript indicating initial values, the solutions are as follows:

1. $N^{25} = N_o^{25} e^{-\sigma_a^{25}\tau}$

2. $\frac{\sigma_c^{25}}{\sigma_a^{25}} \left(N^{25} - N_o^{25}\right) + N^{26} - N_o^{26} = 0$

3. $N^{28} - K_o\left(N^{25}\right)^d = 0$

where: $K_o = N_o^{28}\left(N_o^{25}\right)^{-d}$

4. $N^{49} + \frac{\delta d}{d - a} N^{28} - K_1\left(N^{25}\right)^a = 0$

where: $K_1 = \left(N_o^{49} + \frac{\delta d}{d - a} N_o^{28}\right)\left(N_o^{25}\right)^{-a}$

5. $N^{40} + \frac{\alpha a \delta d}{(d - b)(a - b)} N^{28} + \frac{\alpha a}{a - b} N^{49} - K_2\left(N^{25}\right)^b = 0$

where: $K_2 = \left[N_o^{40} + \frac{\alpha a \delta d}{(d - b)(a - b)} N_o^{28} + \frac{\alpha a}{a - b} N_o^{49}\right]\left(N_o^{25}\right)^{-b}$

$$6.\quad N^{41} + \frac{\alpha ab\delta d}{(d-c)(a-c)(b-c)} N^{28} + \frac{\alpha ab}{(a-c)(b-c)} N^{49} + \frac{b}{b-c} N^{40} - K_3 \left(N^{25}\right)^c = 0$$

$$\text{where: } K_3 = \left[N_o^{41} + \frac{\alpha ab\delta d}{(d-c)(a-c)(b-c)} N_o^{28} + \frac{\alpha ab}{(a-c)(b-c)} N_o^{49} + \frac{b}{b-c} N_o^{40}\right]\left(N_o^{25}\right)^{-c}$$

$$7.\quad \frac{1}{\gamma}\left(N^{42} - N_o^{42}\right) + \alpha\left[\delta\left(N^{28} - N_o^{28}\right) + N^{49} - N_o^{49}\right] + N^{40} - N_o^{40} - N^{41} - N_o^{41} = 0 \quad .$$

Equation 7 is most easily obtained by multiplying '3b' by $\alpha\delta$, '4b' by α, '7b' by $\frac{1}{\gamma}$, and then adding these modified equations to '5b' and '6b' and integrating. Also, since d is a small number, Equation 3 may be written

$$\frac{N_o^{28} - N^{28}}{N_o^{28}} = d\,\ell n \frac{N_o^{25}}{N^{25}}$$

which is accurate to first order in d. To the extent that microscopic cross sections are constant, most probable values of the cross section ratios– α, a, b, etc.– may be obtained by performing a simultaneous least squares fit of Equations 2 through 7 to available experimental burnup data.

The least squares fit required here is considerably more complex than is ordinarily encountered. First of all one considers only variables, as opposed to considering a dependent variable and one or more independent variables. In our case the variables are the isotopic concentrations (the N's in Equations 2 through 7). The errors associated with each data point must be described by a matrix as opposed to the simpler situation, where only the error in the dependent variable is required. Lastly Equations 2 through 7 must be fitted to the data simultaneously. A least squares fitting routine with the required sophistication to solve this problem is under development by B. H. Duane in this laboratory.

The analogous equations for a plutonium system are easier to deal with. Applying the same procedure as above, one has:

$$\frac{dN^{49}}{d\tau} = -\sigma_a^{49} N^{49} \qquad \frac{dN^{49}}{d\tau} = -\sigma_a^{49} N^{49}$$

$$\frac{dN^{40}}{d\tau} = \sigma_c^{49} N^{49} - \sigma_a^{40} N^{40} \qquad \frac{dN^{40}}{dN^{49}} = -\alpha + b\frac{N^{40}}{N^{49}}$$

$$\frac{dN^{41}}{d\tau} = \sigma_a^{40} N^{40} - \sigma_a^{41} N^{41} \qquad \frac{dN^{41}}{dN^{49}} = -b\frac{N^{40}}{N^{49}} + c\frac{N^{41}}{N^{49}}$$

$$\frac{dN^{42}}{d\tau} = \sigma_c^{41} N^{41} \qquad \frac{dN^{42}}{dN^{49}} = -\gamma c\frac{N^{41}}{N^{49}}$$

where: $\alpha = \frac{\sigma_c^{49}}{\sigma_a^{49}}$, $b = \frac{\sigma_a^{40}}{\sigma_a^{49}}$, $c = \frac{\sigma_a^{41}}{\sigma_a^{49}}$, $\gamma = \frac{\sigma_c^{41}}{\sigma_a^{41}}$

$$N^{49} = N_o^{49} e^{-\sigma_a^{49}\tau}$$

$$N^{40} + \frac{\alpha}{1-b} N^{49} - K_o (N^{49})^b = 0$$

where: $K_o = \left(N_o^{40} + \frac{\alpha}{1-b} N_o^{49}\right)\left(N_o^{49}\right)^{-b}$

$$N^{41} + \frac{\alpha b}{(1-c)(b-c)} N^{49} + \frac{b}{b-c} N^{40} - K_1 (N^{49})^c = 0$$

where: $K_1 = \left[N_o^{41} + \frac{\alpha b}{(1-c)(b-c)} N_o^{49} + \frac{b}{b-c} N_o^{40}\right]\left(N_o^{49}\right)^{-c}$

$$\frac{1}{\gamma}\left(N^{42} - N_o^{42}\right) + \alpha\left(N^{49} - N_o^{49}\right) + N^{40} - N_o^{40} + N_{41} - N_o^{41} = 0$$

It must be pointed out that the original assumption of constant microscopic cross sections is at best a weak one. Clearly, the microscopic cross section of any isotope which exhibits a strong and hence self-shielding

resonance (Pu^{240} is a good example) will vary as the isotope changes concentration. None the less, the integrated equations given above could still be worthwhile for some applications. Meantime, the simplicity implicit in the closed form of these equations provides sufficient motivation for further investigation of this point.

CRITICAL MASS PHYSICS

Criticality Safety of PuO_2-SS Driver Fuel in the FFTF - C. L. Brown

When the FFTF design was reviewed for nuclear safety in July 1965,[1] the review was based on FFTF driver fuel pins of 20.0 vol% PuO_2-SS and 0.20 in. diameter. Also at that time, several conservative assumptions were made concerning the calculation of the critical parameters for driver fuel in light water; in particular, the pins were assumed to contain PuO_2 at theoretical density; the minimum Pu^{240} content was assumed to be 5%; and the pins were assumed to have no cladding.

Since July 1965 the maximum proposed enrichment for the driver fuel has been increased slightly to 21.6 vol% PuO_2 and the pin diameter to 0.21 in. To evaluate the effect of these changes on the conclusions made in the original study, the critical parameters for the FFTF driver fuel pins in light water were recalculated. In the later calculations, the fuel pins were defined as accurately as possible; and no conservative allowances were made in density, Pu^{240} content, or cladding thickness. The results are presented in Tables I and II.

TABLE I

CHANGES MADE IN FFTF DRIVER FUEL DESCRIPTION

Assumption	Original Study	Present Study
Fuel Pin Diameter, in.	0.200	0.210
Fuel Pin Length, in.	36	33.46
Cladding, in.	None	0.008 (SS)
PuO_2 Content, vol%	20.0	21.6
Pu^{240}, %	5	10
PuO_2 Density, g/cc	11.46 (theo.)	10.31 (90% theo.)
Plutonium Density in Fuel, g Pu/cc	2.021	1.964

(1) BNWL-193, Physics Research Quarterly Report, July, August, September, 1965.

Age Theory and Time Dependent One-Group Diffusion

Thermal pulsed neutron sources are not common, however; and a modification of the model is needed to give results suitable for comparison with experimental data obtained using a typical high energy neutron source. Since the migration of high energy neutrons is usually significant, some consideration must be given to this aspect of the problem. In a first approximation this can be done using Fermi Age Theory to describe the slowing down process; the assumption of time dependent Fermi Age Theory combined with time dependent diffusion theory has been successfully made by Kylstra and Uhrig[9] in deriving spatially dependent transfer functions. In our analysis, time independent age theory and time dependent diffusion theory were used since in water, the medium of prime concern here, slowing down times are very short. Time-dependent age theory could be incorporated with very little increase in complexity.

Using a delta function source of fast monoenergetic neutrons

$$\delta_f(r,t) = A\,\delta(r)\,\delta(t), \tag{3}$$

and ignoring capture during slowing down, the predicted thermal neutron density in an infinite medium with this model is

$$n_{th}^{ii}(r,t) = \frac{A \exp(-\alpha t) \exp\left[-r^2/4(Dvt + \tau_{th})\right]}{\left[4\pi(Dvt + \tau_{th})\right]^{3/2}} \tag{4}$$

where $\alpha = v\Sigma_A[1 - k^\infty(1 - \beta)]$.

Since slowing down time has been ignored here, the time variable t refers to the time elapsed since the neutrons were slowed down.

Figure 2 shows the transient neutron density as predicted by Equation (4) for various values of the separation distance in light water; the energy of the source neutrons was assumed to be 14 MeV. The transient densities predicted by Equation (4) are significantly different from those obtained using diffusion theory alone. The transient curves are much straighter and each could be well approximated by a single exponential term.

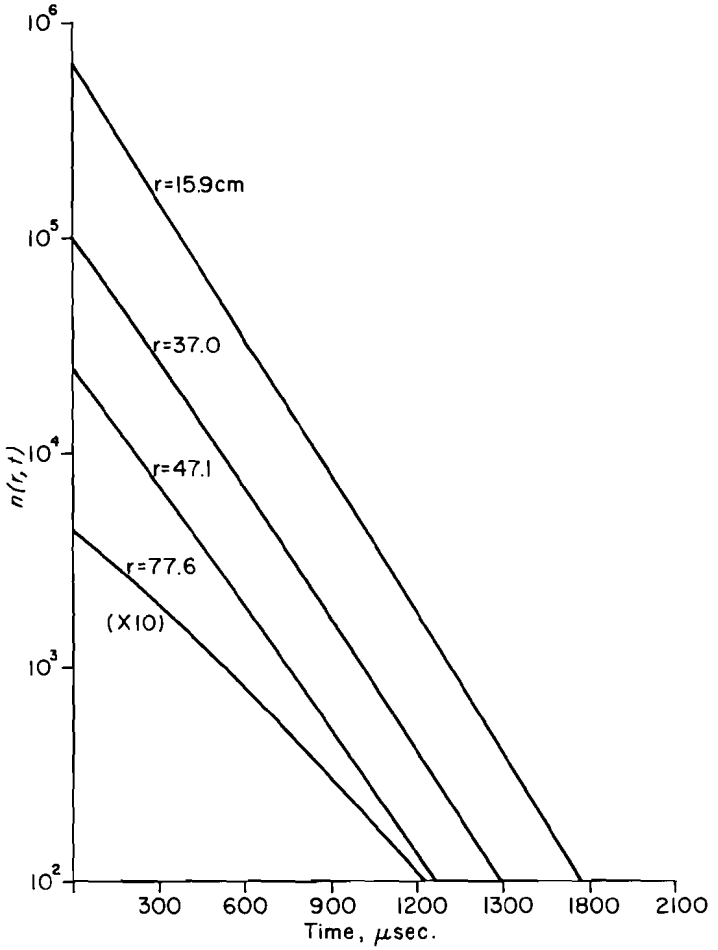

FIGURE 2. Transient Neutron Density in Water as Predicted by Age Theory Plus Time-Dependent One-Group Diffusion

In addition, the spatial dependence is far less than before, due to the widespread dispersion of neutrons from their point of origin as they slow down.

For a nonmultiplying medium, the combined age and thermal diffusion model is exactly equivalent to a two-group diffusion model, where source neutrons are permitted to disperse as neutrons in the high energy group for a specific time before entering the thermal group.

First Flight, Age, and Diffusion

Since the dispersion of fast neutrons strongly influences the thermal neutron density, an important effect, especially in aqueous media, is likely to be first flight correction. Such a correction was suggested by a

comparison of the results of Equations (2) and (4); it was further indicated by the poor agreement between experiment and Equation (4), and it has also been suggested by Kylstra and Uhrig.[9] To add this correction, the fast neutron source term, $S_f(r,t)$, is not expressed as a delta function, but as a first flight collision density.

$$S_f(r,t) = \frac{A\Sigma_o e^{-\Sigma_o r}}{4\pi r^2} \tag{5}$$

where

Σ_o = macroscopic scattering cross section.

For source neutrons, the manipulations are involved for obtaining the thermal neutron density. The simplest method is to solve first the one-dimensional problem for a plane source and then convert to the point source case via a plane-to-point kernel conversion,[10] viz,

$$\rho_{pt}(r,t) = -\frac{1}{2\pi r}\frac{\partial}{\partial r}\rho_{p\ell}(r,t). \tag{6}$$

This method was used for all models, and the results are summarized in Table I. Equations (2) and (4) in Table I have already been presented, while Equation (7) is new and accounts for the distance traveled in first flight in addition to age and thermal group diffusion. The τ' appearing in Equation (7) is different from τ appearing in Equation (4). The actual value of τ used in calculations was obtained from experimental values of the mean squared slowing down distance, $\overline{r^2}$.[11] The value of τ' was modified to account for the distance traveled during the first flight. All values of the slowing down and diffusion parameters of water used in the predicted curves of this paper are given in Table II.

TABLE I

DISTRIBUTION OF THERMAL NEUTRONS, n(x,t) AND n(r,t) FOLLOWING A PULSE OF NEUTRONS AT THE ORIGIN

	Plane Source, $A\delta(x)\,\delta(t)$	Point Source, $A\,\delta(r)\,\delta(t)$	Equation No.
One-group time dependent diffusion	$n^{i}(x,t) = \dfrac{Ae^{-x^2/4Dvt}e^{-\alpha t}}{(4\pi Dvt)^{1/2}}$	$n^{i}(r,t) = \dfrac{Ae^{-r^2/4Dvt}e^{-\alpha t}}{(4\pi Dvt)^{3/2}}$	(2)
Fermi age slowing down plus thermal time-dependent diffusion	$n^{ii}(x,t) = \dfrac{Ae^{-x^2/4(Dvt+\tau)}e^{-\alpha t}}{\left[4\pi(Dvt+\tau)\right]^{1/2}}$	$n^{ii}(r,t) = \dfrac{Ae^{-r^2/4(Dvt+\tau)}e^{-\alpha t}}{\left[4\pi(Dvt+\tau)\right]^{3/2}}$	(4)
First flight collision density plus age plus thermal time-dependent diffusion	$n^{iii}(x,t) = \left\{\dfrac{A\Sigma_o e^{\Sigma_o^2(Dvt+\tau')}e^{-\alpha t}}{4}\right\}$ $\left\{e^{\Sigma_o x}\operatorname{erfc}\dfrac{2\Sigma(Dvt+\tau')+x}{2(Dvt+\tau')^{1/2}}\right.$ $\left. + e^{-\Sigma_o x}\operatorname{erfc}\dfrac{2\Sigma_o(Dvt+\tau')-x}{2(Dvt+\tau')^{1/2}}\right\}$	$n^{iii}(r,t) = \dfrac{A\Sigma_o^2 e^{\Sigma_o^2(Dvt+\tau')}e^{-\alpha t}}{8\pi r}$ $\left\{e^{-\Sigma_o r}\operatorname{erfc}\dfrac{2\Sigma(Dvt+\tau')-r}{2(Dvt+\tau')^{1/2}}\right.$ $\left. - e^{\Sigma_o r}\operatorname{erfc}\dfrac{2\Sigma_o(Dvt+\tau')+r}{2(Dvt+\tau')^{1/2}}\right\}$	(7)

D, v = thermal group diffusion coefficient and speed

t = time variable

$\alpha = v\Sigma_a\left[1 - k^{\infty}(1-\beta)\right]$

$\tau = \frac{1}{6}r^2$; $\tau' = \frac{1}{6}\left(r^2 - \frac{2}{\Sigma_o^2}\right)$

Σ_o = Macroscopic scattering cross section for source neutrons

TABLE II

SLOWING DOWN AND DIFFUSION PARAMETERS OF WATER USED IN CALCULATIONS

Item	Value
A	10^{10} neutrons
D	0.16 cm
v	2.47×10^5 cm/sec
Σ_A	0.0191 cm^{-1}
Σ_o	0.1 cm^{-1}
τ	150 cm^2
τ'	117 cm^2

Predicted values from Equation (7) for a point source of 14 MeV neutrons are plotted in Figure 3. On comparing these curves with those of Figure 2, it is apparent that the first flight correction is significant. The changes result in less curvature of the semilog plots as well as less spatial variation in the time decay of neutron density. The curves of Figure 3 approach very nearly the behavior one would expect for the asymptotic, space-independent transient, i.e., an exponential decay proportional to $e^{-\alpha t}$. To illustrate the closeness of approach, the partial time derivative of Equation (7) is plotted as a function of time in Figure 4 and may be compared with $-\alpha$.

EXPERIMENTAL RESULTS

Experiments have been conducted at the Critical Mass Laboratory where neutron pulses, generated by a Kaman, sealed tube neutron generator, were introduced into various media and the subsequent neutron density transient measured with a 3 in. liquid scintillator tube. The first experiments were conducted in an underground waste storage crib containing some fissionable material. The crib itself was very large, but the experiment was not expecially precise since the pulsed neutron generator tube and the detector could not be lowered into the medium but were suspended above it. The

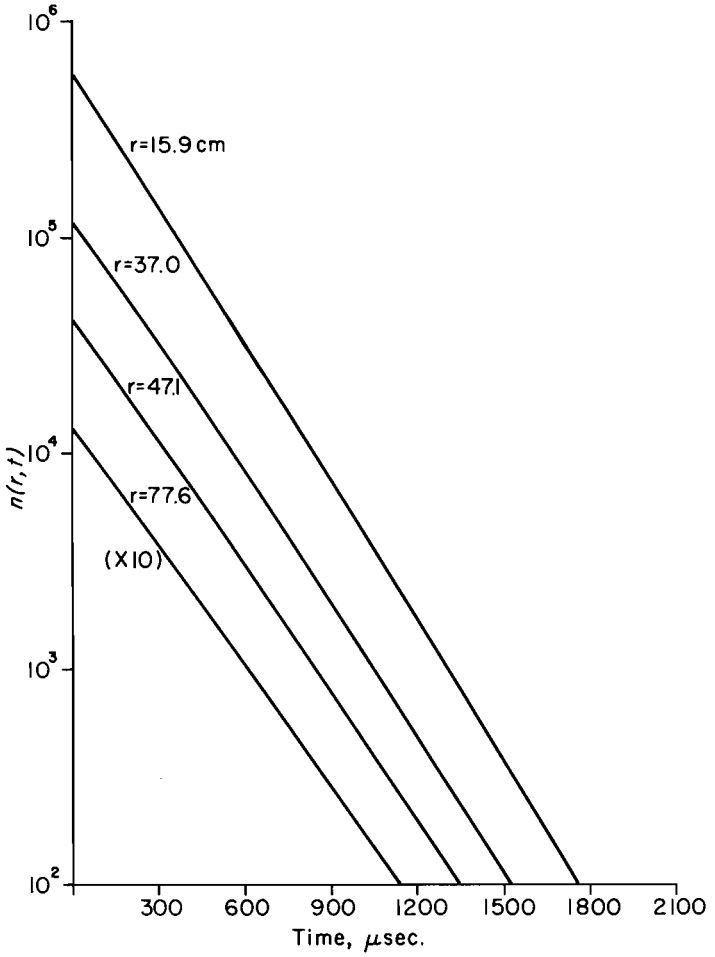

FIGURE 3. Transient Neutron Density in Water as Predicted by a Combination of First Flight, Age, and Time-Dependent Diffusion

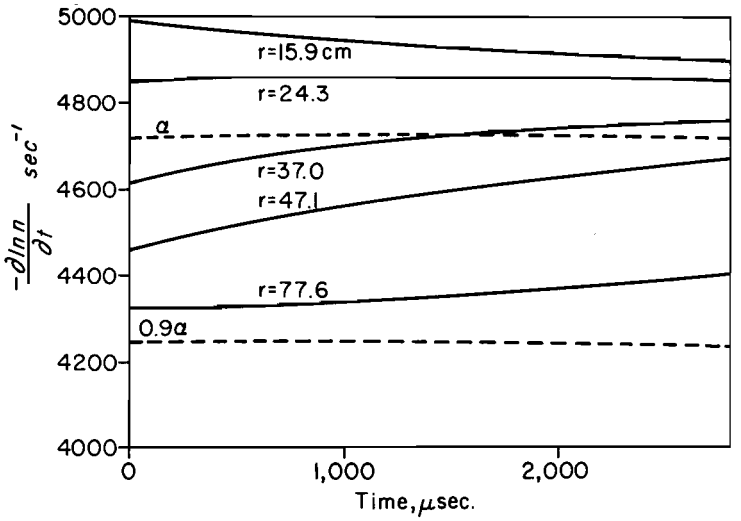

FIGURE 4. Comparison of the Asymptotic Decay Constant, α, with the Apparent Decay Constant Predicted by Equation (7)

spacing between pulser and the detector could not be varied over a significant range, not could the spacing be carefully controlled. Although the results[12] were not conclusive, they did indicate that the neutron die-away was quite similar to a simple exponential decay, which was unexpected in light of the predictions obtained from one-group time dependent diffusion theory.

Experiments were conducted in a large (15 ft diameter, 20 ft high) cylindrical tank filled with light water. The results of these experiments confirmed those previously obtained on, and provided experimental data on, a medium having well-established nuclear properties for comparison with theory. A typical set of experimental neutron density and theoretical predictions is shown in Figure 5. For comparison between theory and experiment, all transients have been normalized to the experimental value at 600 μsec. This procedure was purely arbitrary and was selected mainly on the basis of illustrating the difference in curvature and slope between experiment and predictions. Values obtained using time-dependent thermal diffusion theory have been corrected for a nominal slowing down time in water of 10 μsec.

DISCUSSION

The experimental results and a comparison of them with the predictions of the three simple models outlined above are very revealing. First, it is clear that the use of one-group (thermal) diffusion theory to describe or interpret normal pulsed neutron measurements in large aqueous media is apt to be in gross error, not only quantitatively, but qualitatively. The dispersion of fast source neutrons is very important and must be included. If the fast dispersion is accounted for by a Fermi Age Model, the resulting prediction is improved significantly. If, in addition to Fermi Age, a first flight correction to the initial source distribution is made, the prediction is still further improved and the agreement with experimental results is quite satisfactory.

Second, it seems worth noting that the decay of neutrons in a large, homogeneous, aqueous medium appears very much like an exponential decay,

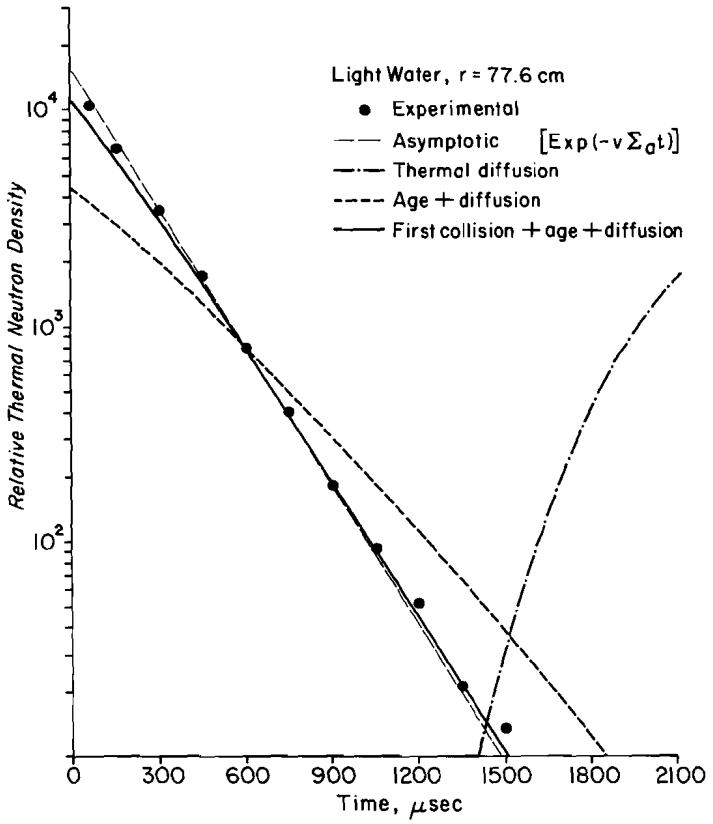

FIGURE 5. Experimental and Theoretical Neutron Density in Water; 77.6 cm Separation Between Source and Detector

from which a decay constant may be estimated. This "apparent" decay constant is a good estimator of the asymptotic value and in these experiments was always within 10%, and usually within 5%, of the asymptotic.

Third, these results suggest that pulsed neutron experiments in very large multiplying systems may also give useful results. In multiplying systems it would be essential to account for high energy neutrons through the use of time-dependent age or few-group theory, but in principle the treatment would be identical. In aqueous media the apparent decay constant is usually less than the true asymptotic constant, and only for a very small separation distances is it greater. Therefore, this indicates that an estimate of k^∞ based on a pulsed neutron experiment would nearly always be conservative in that the apparent k^∞ would be

higher than the true value. To make any estimate of k_∞ of course, one must know β/ℓ, but engineering estimates can often be made with fair precision.

Finally, it should be noted that these results apply primarily to aqueous media. In a system such as graphite, where the thermal diffusion length is long and where there is not such a disparity between fast and thermal neutron mean free paths, the qualitative nature of the transient neutron density will be different. Figure 6 gives an indication of the difference and shows that here again the effects of fast neutron dispersion are significant, but in this case the apparent decay constant is significantly greater than the asymptotic value, and that the time scale for establishing an exponential decay is significantly greater than in an aqueous medium.

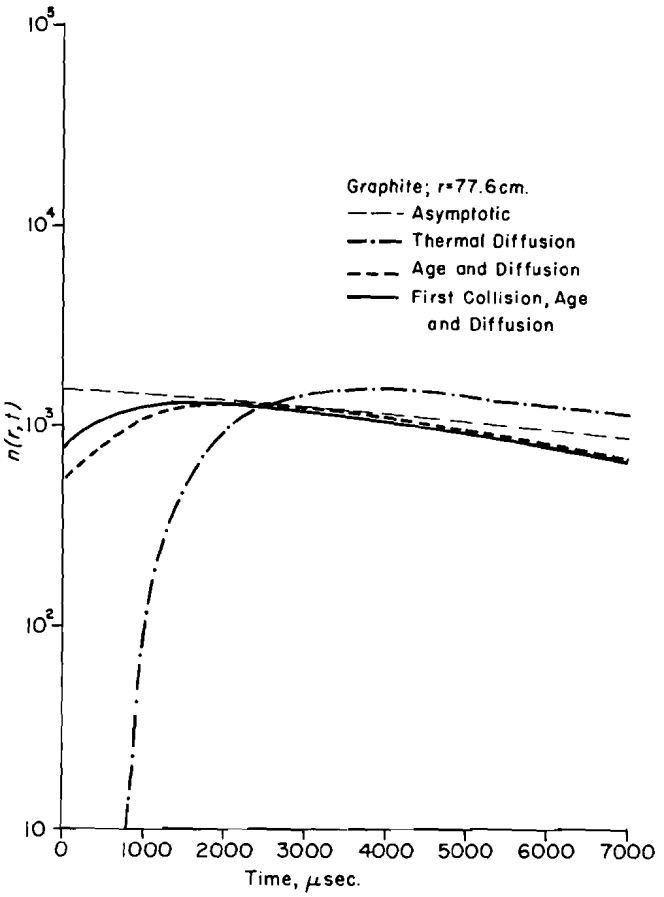

FIGURE 6. Predicted Transient Neutron Density in Graphite

ACKNOWLEDGEMENT

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