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RESEARCH ESTABLISHMENT
LUCAS HEIGHTS

MEASUREMENT OF THE SLOWING DOWN TIME TO 0.3 eV IN BERYLLIUM OXIDE

by

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ABSTRACT

The reaction rates of plutonium-239 and boron (1/v) were measured as a function of space and time in a large block of beryllium oxide following the injection of a pulse of fast neutrons. The time-dependent reaction rates associated with the various Fourier spatial modes were derived from the experimental results. The experimental value of the peaking time of the plutonium-239 reaction rate associated with the fundamental spatial mode was found to be 19.85 ± 1.1 μsec. A theoretical value of 16.5 μsec was obtained using a time-dependent zero-dimensional diffusion code and a crystal scattering kernel for beryllium oxide. It is in marked disagreement with the experimental value.

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ABSTRACT (continued)

Measurements made of the time to peak in the plutonium-239 reaction rate as a function of distance from the source showed a total variation of 4 μsec in peaking time over a distance of 53 cm. Theoretical calculations of the peaking time as a function of distance showed a similar variation and the general trends were in good agreement with the experimental result. There was, however, a constant discrepancy between the theoretical and experimental results of the order of that noted for the fundamental mode.

NOTE: This work has been submitted to a journal. Further details may be obtained from the authors or the Director of the Research Establishment.

CONTENTS

1. INTRODUCTION

Many experiments have been reported in which the time-dependent reaction rate of a spectral indicator has been measured following the injection of a pulse of fast neutrons into a block of moderator. The common aim of these experiments has been to assess the various scattering kernels devised for the common moderators. Engelmann (1958), Moller and Sjostrand (1962, 1963, 1964), Profio and Eckard (1964) and Moller (1966a) have measured time-dependent reaction rates of various spectral indicators in water assemblies by prompt gamma ray detection techniques. Eichelberger (1962), Profio and Eckard (1964) and Moller (1966b) have worked with heavy water, and investigations with graphite assemblies have been reported by Kaneko and Sumita (1965), Maekawa and Yamamuro (1969) and Sumita and Takahashi (1967). Zhezherun et al. (1964) have measured time-dependent neutron transmission through several filters with low-energy resonances as well as the time-dependent response of filter-covered and bare plutonium-239 fission detectors in both beryllium metal and beryllium oxide. The authors (Maher et al. 1967) have also reported some preliminary measurements with beryllium oxide using a bare plutonium-239 fission detector.

In most of the experiments scant consideration has been given either to the effect of the finite duration of the fast neutron pulse or to the effect of the finite geometry of the moderator assembly. In addition, all the calculations reported to date have been space-independent (space dependence being either completely neglected or lost in a DB^2 term) but have still been compared with the results of measurements made at one particular location in a finite moderator assembly following the injection of a pulse of finite duration and of limited spatial extent.

It has been shown (Moller 1966a, 1966b; Maher et al. 1967) that for water, heavy water and beryllium oxide, the time-dependent reaction rate of a spectral indicator is a function of position in the moderator assembly. The results of Maekawa and Yamamuro (1969) also suggest the existence of this effect in graphite. Furthermore, calculations by Maher et al. (1967) show that the time-dependent reaction rate is a strong function of assembly size and the duration of the initial neutron pulse.

It will be shown later in this paper that there is a variation of some 4 μsec in the peaking time of the plutonium-239 reaction rate over a distance of 53 cm in a beryllium oxide assembly. It has already been predicted (Maher et al. 1967) that the variation of this same peaking time with buckling is ~ 8 per cent for a 0.01 cm^{-2} change in buckling and the variation with pulse length is ~ 2 per cent for a 1 μsec change in pulse length for pulse lengths in the range 0-20 μsec. These are comparable with the change of about 3 per cent (at 0.3 eV) in the

peaking time calculated when a sophisticated scattering kernel (e.g. one based on Sinclair's phonon frequency distribution for BeO) is used rather than a free gas kernel. It is obvious then that the effects of pulse length, finite size and spatial position cannot be neglected if this type of experiment is to produce consistent, unambiguous results. Furthermore, if this type of investigation is to serve its purpose, namely the assessment of current scattering kernels, then either

(i) the experiment must be performed in such a manner as to produce a space-independent time-dependent reaction rate distribution comparable with the effectively infinite medium calculations of the past, or

(ii) theoretical calculations must be improved to include spatial effects and be comparable with measurements made at one particular location in a finite assembly after a fast neutron pulse from a source of limited spatial extent.

This paper describes an experimental technique which was developed to satisfy the former of these alternatives. The technique was used to measure space-independent time-dependent reaction rates of the spectral indicator plutonium-239 in a large (60.96 x 60.96 x 58.42 cm^3) beryllium oxide assembly. The reaction rate of a 1/v (boron) detector was also measured to enable the extraction of the response due to the resonance (non-1/v) part of the plutonium-239 fission cross section from the total plutonium-239 response.

2. EXPERIMENTAL METHOD

2.1 Theoretical Basis

Let us confine our attention, for the sake of simplicity, to moderator assemblies in the form of rectangular parallelepipeds. Assuming that the neutron flux, $\Phi(\underline{x},E,t)$, is zero at some distance outside the surface of the assembly and that $\Phi(\underline{x},E,t)$ may be expanded in terms of the eigenfunctions $\psi_{ijk}(\underline{x})$ of the equation

$$\nabla^2\psi(\underline{x}) + B^2\psi(\underline{x}) = 0 \quad ,$$

we may write

$$\Phi(\underline{x},E,t) = \sum_i \sum_j \sum_k \phi(E,t,B^2_{ijk})\psi_{ijk}(\underline{x}) \quad , \qquad .....(1)$$

where

$$\psi_{ijk}(\underline{x}) = f(i,x,A) \quad f(j,y,C) \quad f(k,z,D) \quad ,$$

and

$$f(i,x,A) = \sin(i(x+A/2)\pi/A) \quad ,$$

if the origin is assumed to be at the centre of the parallelepiped and A, C and D are the extrapolated dimensions of the assembly.

The reaction rate of a spectral indicator as a function of space and time is

$$R(\underline{x},t) = \int_0^\infty \Phi(\underline{x},E,t)\Sigma_{SI}(E)\,dE \quad ,$$

where $\Sigma_{SI}(E)$ is the macroscopic cross section for the reaction of interest. Using expression (1) we obtain

$$R(\underline{x},t) = \sum_i \sum_j \sum_k R_{ijk}(t,B^2_{ijk})\psi_{ijk}(\underline{x}) \quad , \qquad .....(2)$$

with

$$R_{ijk}(t,B^2_{ijk}) = \int_0^\infty \phi(E,t,B^2_{ijk})\Sigma_{SI}(E)\,dE \quad . \qquad .....(3)$$

Thus, assuming that the neutron flux in a finite moderator assembly may be expanded in terms of Fourier spatial modes, the modal amplitudes are space-independent time-dependent reaction rates. These time-dependent modal amplitudes may be compared directly with the results of zero-dimensional diffusion calculations.

The measurement of space-dependent time-dependent reaction rates could be done in several ways to enable an evaluation of the $R_{ijk}(t)$. The following technique was used in the present investigations.

Consider the case of a moderator assembly presenting a square face to the external pulsed source and aligned with respect to the source as indicated in Figure 1. If it is assumed that a limited number of the terms in Equation 2 are sufficient to describe the measured spatial distribution, then Equation 2 can be simplified to

$$R(\underline{x},t) = \sum_{i=1}^{L} \sum_{j=1}^{2M-1} \sum_{k=1}^{2M-1} R_{ijk}(t,B^2_{ijk})f(i,x,A)f(j,y,C)f(k,z,C) \quad , \qquad .....(4)$$

where $i = 1, 2, 3, \ldots, L$ and $j,k = 1, 3, 5, \ldots, 2M-1$,

since y and z are interchangeable. In particular the measured spatial distribution of the reaction rate in the x-direction will have the form

$$F(x,t) = \sum_{i=1}^{L} A_i(y_o,z_o,t)f(i,x,A) \quad , \qquad .....(5)$$

so that a least squares fit of the experimental data at time t with the Fourier modes f(i,x,A) will yield estimates of the $A_i(y_o,z_o,t)$ and provide a set of L

equations of the form

$$A_i(y_o,z_o,t) = \sum_{j=1}^{2M-1} \sum_{k=1}^{2M-1} R_{ijk}(t,B^2_{ijk}) f(j,y_o,C) f(k,z_o,C)$$

The $R_{ijk}(t)$ constitute a set of $LM(M+1)/2$ unknown parameters. By undertaking the measurement of $M(M+1)/2$ independent scans in the x-direction and hence obtaining $LM(M+1)/2$ independent equations in the unknown parameters, the $R_{ijk}(t)$ may be determined.

These considerations form the basis of the measurements to be described. Space-dependent time-dependent reaction rates were measured for boron and the spectral indicator plutonium-239. The experimental results were analyzed in the manner described to yield the time-dependent amplitudes of the three-dimensional Fourier spatial modes of the detector reaction rates.

2.2 Description of Apparatus

The neutrons were produced by the $^9Be\ (d,n)\ ^{10}B$ reaction, using the beam from a 3 MeV Van de Graaff accelerator which had been modified (Fraser et al. 1968) to produce square beam pulses. The pulse width throughout the experiment was 20 μsec with about 1 mA of beam in the peak of the pulse. A post-acceleration deflection unit (see Figure 2), used in conjunction with the main beam pulsing, ensured a very low ($< 10^{-5}$) off-current to on-current ratio.

The BeO assembly and its associated shielding have been described previously (Ritchie 1968, Rainbow and Ritchie 1968). For this experiment the BeO tiles were arranged in the form of a rectangular parallelepiped 58.42 x 60.96 x 60.96 cm^3 with five scan holes each of cross section 1.02 x 1.02 cm^2 passing through the assembly. Four of these had their lengths parallel to the x-direction and one had its length parallel to the y-direction. Figure 1 details the orientation of the BeO tiles and the location of the scan holes while Table 1 lists the co-ordinates of the scan holes.

The plutonium-239 fission detector was a cylindrical ionization chamber of 0.485 cm cathode diameter, 0.159 cm anode diameter and 2.54 cm active length with a 300 μg loading of plutonium-239 oxide ($^{239}PuO_2$) in the form of a thin film on the cathode. Its counting efficiency for thermal neutrons was approximately 5×10^{-4} counts per second per unit thermal flux and $d\Sigma_a$, the product of the absorption (including fission) cross section and the film thickness, was estimated to be 1.8×10^{-4} for thermal neutrons and 1.0×10^{-3} for neutrons in the region of the 0.3 eV resonance.

The low sensitivity of the detector and the small values of $d\Sigma_a$ imply that:

(i) flux depression in the region of the detector is very small, and

(ii) self-absorption is negligible and the detector count rate is proportional to

$$\int_0^\infty \sigma_f(E)\ \phi(\underline{x},E,t)\ dE \quad ,$$

where σ_f is the plutonium-239 fission cross section.

The 1/v detector was a silicon surface barrier detector with a thin boron film on the sensitive surface (Rose 1967). The active area was approximately 0.95 cm^2 and its active length 1.86 cm. The counting efficiency was low and comparable with that of the plutonium-239 fission detector.

Either of these detectors could be moved in the scan holes by means of a motorized screw which could position the detectors with a precision of ± 0.03 cm. The time distributions measured at each spatial position were normalized by accumulating data until a monitor detector, which was fixed in position, had reached a preset number of counts. The monitor was a Twentieth Century 5EB70 1/4 in. BF_3 detector and was located at the position shown in Figure 1.

A block diagram of the electronics is shown in Figure 2. The time-mark generator was the master timing unit but also provided synchronous time marks which could be used to check the timing of other parts of the system. Pulse generators 1, 2 and 3 determined the time of occurrence and the length of the initial neutron pulse. Pulse generator 4 controlled the monitor gate which was opened 2 msec after the end of the neutron pulse for a period of 3 msec. These two times were chosen to obtain good counting statistics from the monitor and also to ensure that the monitor count rate change with time was very close to exponential, hence allowing realistic dead time corrections to be made. The fifth pulse generator controlled the time after the neutron pulse at which the time analyzer started. The start pulse to the time analyzer was fed through a gate controlled by the monitor scaler. This gate opened when the monitor scaler was started at the beginning of a measurement and closed when the monitor scaler reached its preset count, thus normalizing each time distribution measurement to the same number of monitor counts.

The time analyzer used in this series of experiments was more sophisticated and capable of better time resolution than the one used in the experiments described previously (Maher et al. 1967). It consisted basically of two 16 MHz scalers interfaced to a PDP-7 on-line computer. The scalers were operated alternately; while one scaled, the contents of the other were transferred to a buffer store in the computer's memory. The minimum channel width possible was

3 μsec which was set by the cycle time of the computer and the number of instructions required to transfer the contents of a scaler to the buffer store. The important feature of the instrument was that there was a negligible (< 30 nsec) time interval between channels. The number of channels was variable over the range 16 to 512 by factors of 2, and the channel width from 3 μsec to 511 μsec in steps of 1 usec. The unit was operated with 256 channels, 5 μsec wide, during most of the present measurements.

The time delays associated with the delayed triggering pulses were generated by R-C circuits. Because of this the timing system was the major source of error in the experimental arrangement and contributed some 1.5 per cent to the error in the normalized detector counts.

2.3 Experimental Method

Preliminary measurements were carried out to measure the transverse spatial distribution of both the 1/v and plutonium-239 reaction rates. These measurements showed that the distribution was symmetric, as was to be expected from the source conditions, and that the amplitudes of the higher modes were quite small. For example the amplitude of the mode with i=3 was only two or three times its experimental error at times immediately after the pulse and about 2 per cent of the amplitude of the fundamental. Hence it was decided to limit the analysis to producing estimates of the $R_{i11}(t)$ and $R_{i13}(t)$, modes such as i33 and i15 being neglected. From Section 2.1 it follows that only two independent scans need to be done to derive values of the $R_{i11}(t)$ and $R_{i13}(t)$, but in fact the four scan holes shown in Figure 1 were used to check that the distribution was indeed cylindrically symmetric.

The time spectra from both the 1/v and plutonium-239 detectors were measured at thirteen points along each of the four scan holes. At each point the detector counts were normalized to 450,000 monitor counts. Immediately before and after each spatial scan, normalized measurements were made of the time distribution at a standard position in the transverse scan hole. This allowed a check on the consistency of operation of the equipment between and during the various spatial scans.

Immediately after measurement of the time-dependent spatial distributions the total scan time of the time analyzer was extended to 3.7 msec by increasing the analyzer channel widths from 5 to 15 μsec. Normalized measurements were made of the time-dependent reaction rates of the plutonium-239 and boron detectors at the centre position of the transverse scan hole. These measurements yielded the ratio of the plutonium-239 reaction rate to the boron reaction rate as a function of time and allowed an evaluation of the asymptotic $^{239}Pu/^{10}B$ ratio.

2.4 Measured Space-Dependent Time-Dependent Reaction Rates

Figure 3 presents the time distributions of the plutonium-239 and boron reaction rates measured in scan hole 1 at x = -13.2 cm and at x = +13.2 cm. The large peak in the plutonium-239 time distributions is due to the slowing down of neutrons through the energy region corresponding to the 0.3 eV plutonium-239 fission resonance. The smaller, less well defined peak in the time distributions of both the plutonium-239 and boron reaction rates obtained at x = +13.2 cm is the result of a wave of essentially thermalized neutrons diffusing from the source side of the assembly.

Figure 4 presents the spatial distributions of the plutonium-239 and boron reaction rates measured in scan hole 1 at various times after the end of the pulse. The spatial distributions exhibit marked asymmetry in the beam direction even as long as 1.2 msec after the neutron pulse.

2.5 Fourier-Mode Reaction Rates and Space-Dependent Peaking Times

The first part of the data analysis consisted of checking that the measured spatial distribution did not change when y and z were interchanged. This was done by comparing data obtained along equivalent scan holes, that is, by comparing data obtained along scan hole 1 with that obtained along scan hole 2 and data obtained along scan hole 3 with that obtained along scan hole 4. The time-integrated data for the various positions and the time- and space-integrated data obtained along equivalent scan holes were found to agree within the experimental errors.

Data from equivalent scans were then summed to produce two independent data sets for each detector. These were fitted with the Fourier modes by a weighted least sum of squares fitting routine and, using the method indicated in Section 2.1, the $R_{ill}(t)$ and the $R_{il3}(t)$ were evaluated.

The value used for the extrapolation distance was 1.149 cm (Ritchie 1968). After correcting this value for the moderator density (2.877 g cm^{-3}) and for voidage (Ritchie 1968), the extrapolated dimensions of the moderator assembly were evaluated as 63.36 cm in the transverse direction and 60.87 cm in the beam direction. The corresponding value for the fundamental buckling was $B^2_{111} = 7.58 \times 10^{-3}$ cm^{-2}.

Figure 5 presents plots of some of the $R_{ill}(t)$ for plutonium-239 and boron. It was found that the spatial distributions in the beam direction for plutonium-239 could be adequately fitted with eight Fourier modes. Six beam modes was found to be sufficient for boron. The values of the $R_{il3}(t)$ are small, the $R_{113}(t)$ being only two or three times their experimental errors.

Figure 6 presents a plot of the $R_{ill}(t)$ for the response due to the non-1/v

(or resonance) part of the plutonium-239 fission cross section. These were evaluated directly from the $R_{111}(t)$ for plutonium-239 and boron. The $R_{111}(t)$ for boron was scaled by the asymptotic $^{239}Pu/^{10}B$ ratio (0.863 ± 0.020) and subtracted from the $R_{111}(t)$ for plutonium-239.

The peaking times of the plutonium-239 reaction rate were estimated from the various time distributions by using a fitting routine (POISFIT). This routine which employed the weighted least sum of squares criterion to obtain the best fit, and included a facility for adjusting the representative time values for each channel, fitted data in the region of a peak with the Poisson distribution

$$A[\lambda(t-B)]^n \exp[-\lambda(t-B)] \quad ,$$

where A, λ, B and n are the paramters of fit. The method proposed by Fletcher and Powell (1963-4) was used to locate the minimum of the weighted squares function, which was found to converge rapidly. The relation

$$T_p = \frac{n}{\lambda} + B$$

was used to evaluate the peaking time (T_p) from the fitted parameters. The use of the Poisson distribution for this purpose has little physical basis; its usefulness derives from the fact that it peaks asymmetrically, as does the plutonium-239 reaction rate, and gives a good fit to the experimental data. The errors on the peaking times were derived from the fitting routine in the normal way. These varied between about 0.5 μsec and 3 μsec depending on the spatial position at which the time distribution was measured. In comparison with these a systematic error of about 0.08 μsec due to ion drift time in the counter was neglected.

2.6 Theoretical Calculations

The zero-dimensional code TENDS (Maher et al. 1967, Ritchie et al. 1970) has been used to calculate the time-dependent reaction rates $r(t,B^2)$ of plutonium-239 and boron corresponding to the fundamental three-dimensional Fourier spatial mode. TENDS approximates leakage by a DB^2 term and uses a square pulse of variable duration as the source condition.

A 28-group cross section set condensed over a Westcott spectrum from the 120-group GYMEA cross section library (Pollard and Robinson 1966) was used in the calculations. The energies ranged from 1.026×10^{-3} eV to 10 MeV with one group below the Bragg cut-off and fourteen groups between 0.001 eV and 1 eV. The transport cross section for beryllium oxide was that calculated by Maher and Trimble (1968 - A.A.E.C. unpublished report) and corrected for extinction in the BeO. Table 2 presents the group boundaries used and the corresponding group-averaged BeO transport cross sections, both extinction corrected and uncorrected.

Time-dependent reaction rates of plutonium-239 and boron were calculated with and without absorption ($\lambda_a = 1.954 \times 10^2$ sec^{-1} at 2.96 g cm^{-3}, Ritchie 1968) for the fundamental buckling of the moderator assembly. They were normalized to the measured $R_{111}(t)$ at 1 msec after the pulse. Figure 7 presents a comparison of the measured and calculated plutonium-239 reaction rates.

Space-dependent time-dependent reaction rates of the plutonium-239 detector were constructed from the results of several TENDS calculations. The code was used to calculate the time-dependent reaction rate of the detector corresponding to various Fourier modes and these were summed to produce the space-dependent time-dependent reaction rates. The weights of the various modes were assigned by assuming that the initial spatial distribution could be described by a δ-function in the beam direction and the lowest Fourier modes in the transverse directions.

With only a few modes the peaking time as a function of position showed marked oscillations on the side of the stack away from the source. With more than eight modes the oscillations became vanishingly small. Figure 8 presents a comparison of the experimental plutonium-239 peaking time as a function of position, with a theoretical calculation in which nine Fourier modes were used.

3. DISCUSSION

From Figure 5 it can be seen that the decay of the Fourier modes appears exponential at times greater than about 50 μsec after the pulse. This indicates that there is little coupling between modes and that the various modes have an independent existence. The nature of the curves and further evidence to be presented below also suggest that there is little coupling at times immediately after the pulse. This means that the assumption that the time-dependent neutron flux in a finite moderator assembly may be expanded in terms of Fourier spatial modes is reasonable in the present application.

Figure 7 shows that there is a marked disagreement between theory and experiment in the peaking time of the plutonium-239 reaction rate. The theoretical distribution peaks at 16.5 μsec after the pulse while the measured distribution peaks at 19.85 ± 1.1 μsec. The difference of 3.35 μsec is significantly greater than the experimental error. In addition the leading edge of the theoretical curve is quite different from that of the measured curve and theory under-estimates the amplitude of the peak by about 16 per cent. At later times (> 100 μsec after the pulse) the curves are quite similar but the measured distribution decays slightly faster than the theoretical distribution. The theoretical time-dependent reaction rate of boron is quite similar to the measured distribution. However, once again the measured distribution decays slightly faster than the theoretical distribution. Only at very early times (< 30 μsec) after the pulse is there any marked disagreement. The marked disagreement between theory and experiment at

early times could be explained in terms of one or more of the following:

(i) inadequacies of the calculational technique,

(ii) errors in the detector cross sections,

(iii) inadequacies of the BeO scattering kernel.

The diffusion calculations described have predicted a rate of thermalization which is too fast. This effect has been noted by other investigators who have used diffusion theory. Recently Menzel (1968) conducted an investigation of time-dependent neutron spectra in water. He found that a space-dependent diffusion calculation using the Haywood-II kernel and a set of thirty bucklings predicted a rate of thermalization which was considerably faster than that observed experimentally. This evidence suggests that the use of diffusion theory as a basis for time-dependent thermalization calculations is unsatisfactory.

The discrepancy between theory and experiment in the amplitude of the plutonium-239 reaction rate peak is very large. It is possible that the data for the 0.3 eV plutonium-239 fission resonance in the GYMEA cross section library are in error. Another possibility is that the use of a finer group structure might reduce the discrepancy between theory and experiment.

The disagreement at long times is not severe ($\sim$ 2 per cent in the decay constant) and is not considered to be very important. There are two possible explanations for the measured distributions decaying faster than the theoretical distributions. Firstly, during analysis of the present results it was observed that failure to include sufficient modes in the Fourier analysis of the beam direction spatial distributions resulted in a systematic underestimate of the amplitude of the fundamental mode. It is possible that the neglect of higher transverse modes has also resulted in a systematic deviation of the $R_{111}(t)$ from their true values. The systematic difference between the true value of the $R_{111}(t)$ and the experimental value decreases with time as the higher transverse modes decay.

Secondly, the 28-group cross section set used in the present calculations covered the range 1.026×10^{-3} eV to 10 MeV with only one group below the Bragg cut-off energy. Sub-Bragg neutrons play an important role in the decay of thermalized neutron populations in polycrystalline moderators. It is possible that insufficient attention has been given to them in the present calculations. If this is the case we could expect the decay rate to increase with the inclusion of lower energy groups.

As can be seen from Table 2 the times at which the $R_{111}(t)$ peak show a tendency to decrease as the order of the mode increases, although the errors are such that the existence of the trend cannot be completely justified. The trend

does, however, agree with that predicted by the zero-dimensional code TENDS and is further justification for the use of the Fourier mode concept in the present application. Indeed the various Fourier spatial modes appear to have quite separate identities, the variation in peaking times of the $R_{ill}(t)$ for plutonium-239 indicating that they are associated with different energy distributions.

Figure 8, which compares the measured and calculated peaking times of the plutonium-239 reaction rate versus position, has several interesting features. Both curves rise quite quickly ($\sim$ 0.15 μsec cm^{-1}) as x increases at negative x. In the region of positive x there is an apparent levelling-off of the experimental curve which is not matched by the theoretical curve. At negative x the discrepancies between equivalent theoretical and experimental peaking times are comparable with the space-independent value (3.35 μsec) whereas at positive x the discrepancy between theory and experiment is reduced.

The most important feature of the peaking time versus position measurements is the total variation of approximately 4 μsec in peaking time across the assembly. This implies that the neutron energy spectrum is quite different at different positions in a moderator assembly at times soon after the injection of a fast neutron pulse and that adequate consideration must be given to spatial effects in pulsed thermalization experiments.

Figure 6, which is a plot of the time-dependent response of the non-1/v part of the plutonium-239 fission cross section over a 260 μsec time interval from immediately after the pulse, reveals that there is no possibility of measuring a decay constant associated with a higher discrete energy mode for beryllium oxide. This response is due almost entirely to the 0.3 eV fission resonance and for times greater than about 35 μsec represents the decay of the 0.3 eV neutron population.

From Figure 6 it is obvious that the decay of the 0.3 eV neutron population is markedly non-exponential over the time range shown in the diagram, and exhibits no evidence of the existence of a higher discrete energy mode. This is consistent with the current theoretical prediction for polycrystalline moderators - that there exists at most only one discrete energy mode. It also demonstrates that it is not possible to parameterize (via a decay constant associated with a higher discrete energy mode) the time-dependent reaction rate of plutonium-239 in beryllium oxide. This suggests that any information concerning the thermalization properties of the moderator must be inferred from a comparison of measured and calculated spectral indicator reaction rates.

4. CONCLUSIONS

The experimental technique which has been proposed for the measurement of space-independent time-dependent detector reaction rates has been applied

successfully in measurements with a boron (1/v) detector and the spectral indicator plutonium-239. The experimental results indicate that the assumption that the neutron flux in a finite moderator assembly may be expanded in terms of Fourier spatial modes is good, even at times immediately after a fast neutron pulse. The various time-dependent Fourier spatial modes appear to exist as quite separate identities, each having a different energy spectrum.

Zero-dimensional multigroup diffusion calculations of the time-dependent reaction rates of plutonium-239 and boron, corresponding to the fundamental buckling of the assembly, do not match the measured distributions, the discrepancy between theory and experiment being particularly marked in the region of the peak in the time distribution of the plutonium-239 reaction rate. There are several possible explanations for the inadequacy of the theoretical calculations. However, it does appear that the use of diffusion theory as the basis for time-dependent thermalization calculations is unsatisfactory and could well be the major source of the discrepancy between theory and experiment.

The marked variation of the peaking time of the plutonium-239 reaction rate as a function of position in the moderator assembly implies that the neutron energy spectrum varies with position within a finite moderator assembly at times immediately after the injection of a pulse of fast neutrons. This demonstrates that adequate consideration must be given to spatial effects in thermalization experiments which involve the use of a pulsed source technique, and justifies the considerable attention given to spatial effects in the present measurements. Allowing for the discrepancy between the results of the zero-dimensional calculation and experiment, the agreement of the calculated plutonium-239 peaking times as a function of position with the experimental results is quite good. This indicates that, once the discrepancy between the zero-dimensional calculations and experiment is remedied, spatial effects might be handled quite adequately by a technique similar to that used in the space-dependent calculations described here.

There is an obvious need for the development of time-dependent multigroup codes which are based upon better approximations (such as the P_1 approximation) to transport theory than diffusion theory. The use of these codes in the analysis of time-dependent thermalization experiments would help to resolve the problem of whether the disagreement between theory and experiment is due to the inadequacy of the theory or the data. This would enable measurements of the time-dependent reaction rates of spectral indicators to serve as a means of assessing the adequacy of scattering kernels.

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TABLE 1

COORDINATES OF SCAN HOLES USED TO

MEASURE SPATIAL DISTRIBUTIONS

Scan Hole	y-Coordinate	z-Coordinate
1	+ 10.61 cm	+ 0.51 cm
2	+ 0.51 cm	+ 10.16 cm
3	- 20.32 cm	- 0.51 cm
4	+ 0.51 cm	- 20.32 cm
	x-Coordinate	z-Coordinate
Transverse	- 5.08 cm	- 0.51 cm

TABLE 2

GROUP BOUNDARIES USED IN THE CALCULATIONS WITH

CORRESPONDING GROUP-AVERAGED BeO TRANSPORT CROSS SECTIONS

Group	Energy Boundaries (eV)	σ_{tr} (barns)	
		Corrected*	Uncorrected
28	1.026×10^{-3} - 2.789×10^{-3}	0.59	0.586
27	2.789×10^{-3} - 1.250×10^{-2}	6.15	8.549
26	1.250×10^{-2} - 2.061×10^{-2}	11.22	13.522
25	2.061×10^{-2} - 3.389×10^{-2}	8.67	9.846
24	3.389×10^{-2} - 5.602×10^{-2}	10.17	10.625
23	5.602×10^{-2} - 8.358×10^{-2}	10.26	10.263
22	8.358×10^{-2} - 1.020×10^{-1}	9.727	9.727
21	1.020×10^{-1} - 1.246×10^{-1}	9.538	9.538
20	1.246×10^{-1} - 1.523×10^{-1}	9.402	9.402
19	1.523×10^{-1} - 2.055×10^{-1}	9.452	9.452
18	2.055×10^{-1} - 2.511×10^{-1}	9.485	9.485
17	2.511×10^{-1} - 4.139×10^{-1}	9.376	9.376
16	4.139×10^{-1} - 6.825×10^{-1}	9.278	9.278
15	6.825×10^{-1} - 1.125	9.248	9.248
14	1.125 - 1.855	9.216	9.216
13	1.855 - 3.059	9.195	9.195
12	3.059 - 5.043	9.191	9.191
11	5.043 - 8.315	9.187	9.187
10	8.315 - 2.260×10^{1}	9.183	9.183
9	2.260×10^{1} - 7.889×10^{1}	9.179	9.179
8	7.889×10^{1} - 2.753×10^{2}	9.177	9.177
7	2.753×10^{2} - 9.611×10^{2}	9.172	9.172
6	9.611×10^{2} - 3.354×10^{3}	9.180	9.180
5	3.354×10^{3} - 1.170×10^{4}	9.103	9.103
4	1.170×10^{4} - 4.086×10^{4}	8.789	8.789
3	4.086×10^{4} - 1.426×10^{5}	8.429	8.429
2	1.426×10^{5} - 4.987×10^{5}	8.289	8.289
1	4.987×10^{5} - 1.000×10^{7}	4.222	4.222

* Corrected for extinction effects

TABLE 3

MEASURED AND CALCULATED PEAKING TIMES

OF THE ^{239}Pu REACTION RATE

Mode	Buckling (cm^{-2})	Peaking Time (μsec)	
		Theoretical	Calculated
111	7.58×10^{-3}	16.5	19.85 ± 1.1
211	1.56×10^{-2}	15.5	19.1 ± 1.2
311	2.89×10^{-2}	14.2	17.9 ± 1.8

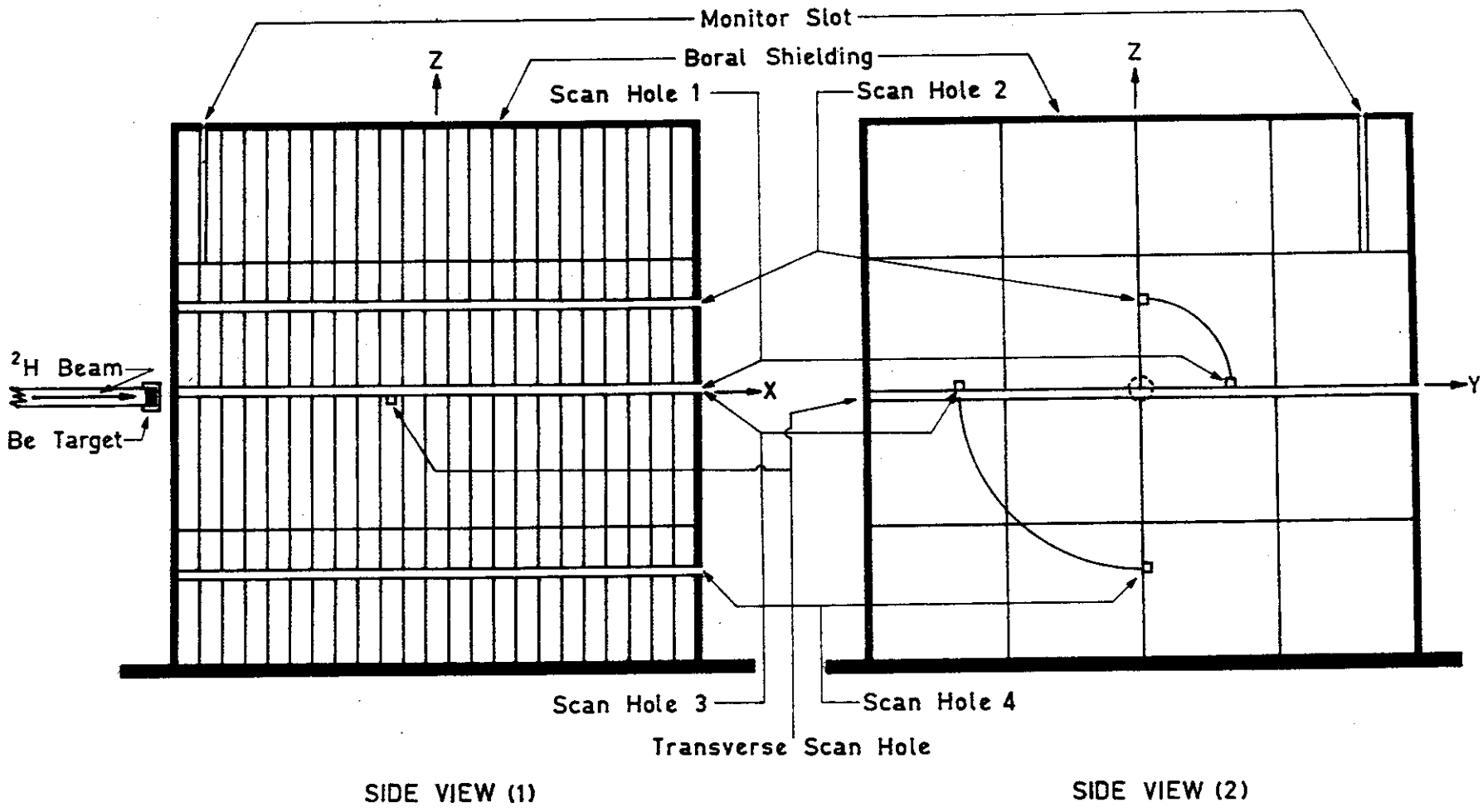

FIGURE 1. SCHEMATIC DIAGRAM OF THE MODERATOR ASSEMBLY

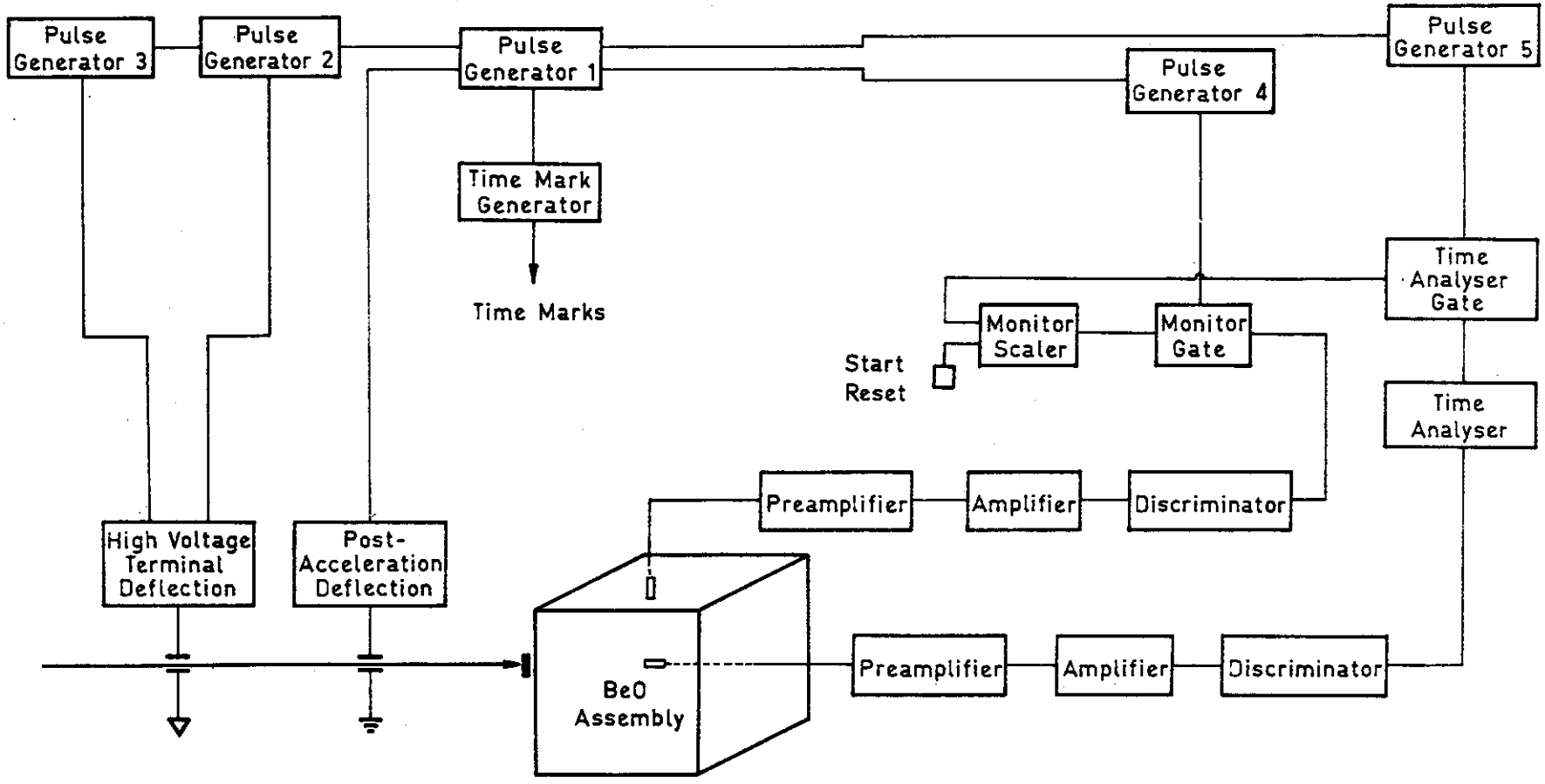

FIGURE 2. SCHEMATIC DIAGRAM OF THE EXPERIMENTAL ARRANGEMENT

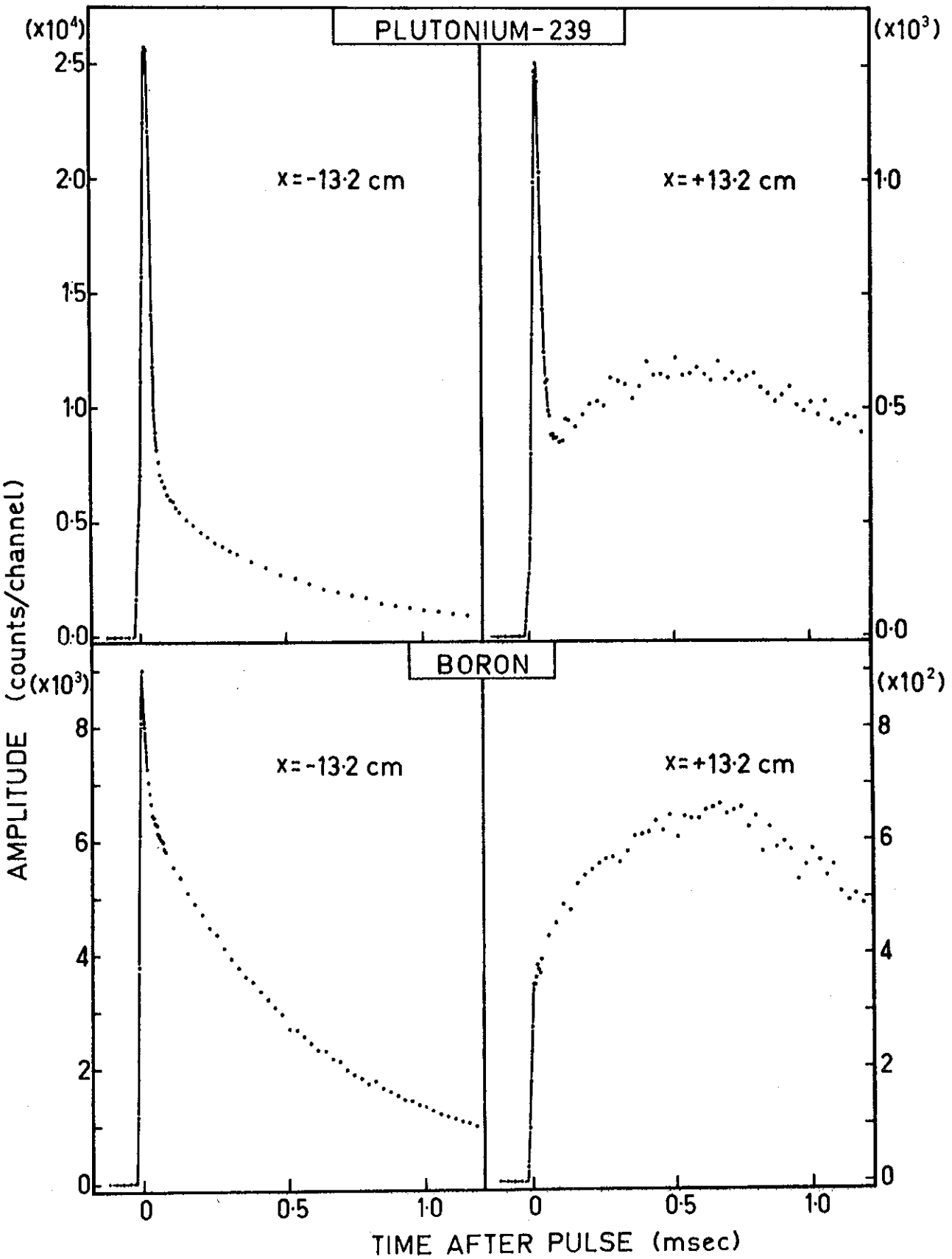

FIGURE 3. TIME DISTRIBUTIONS OF THE PLUTONIUM-239 AND BORON REACTION RATES

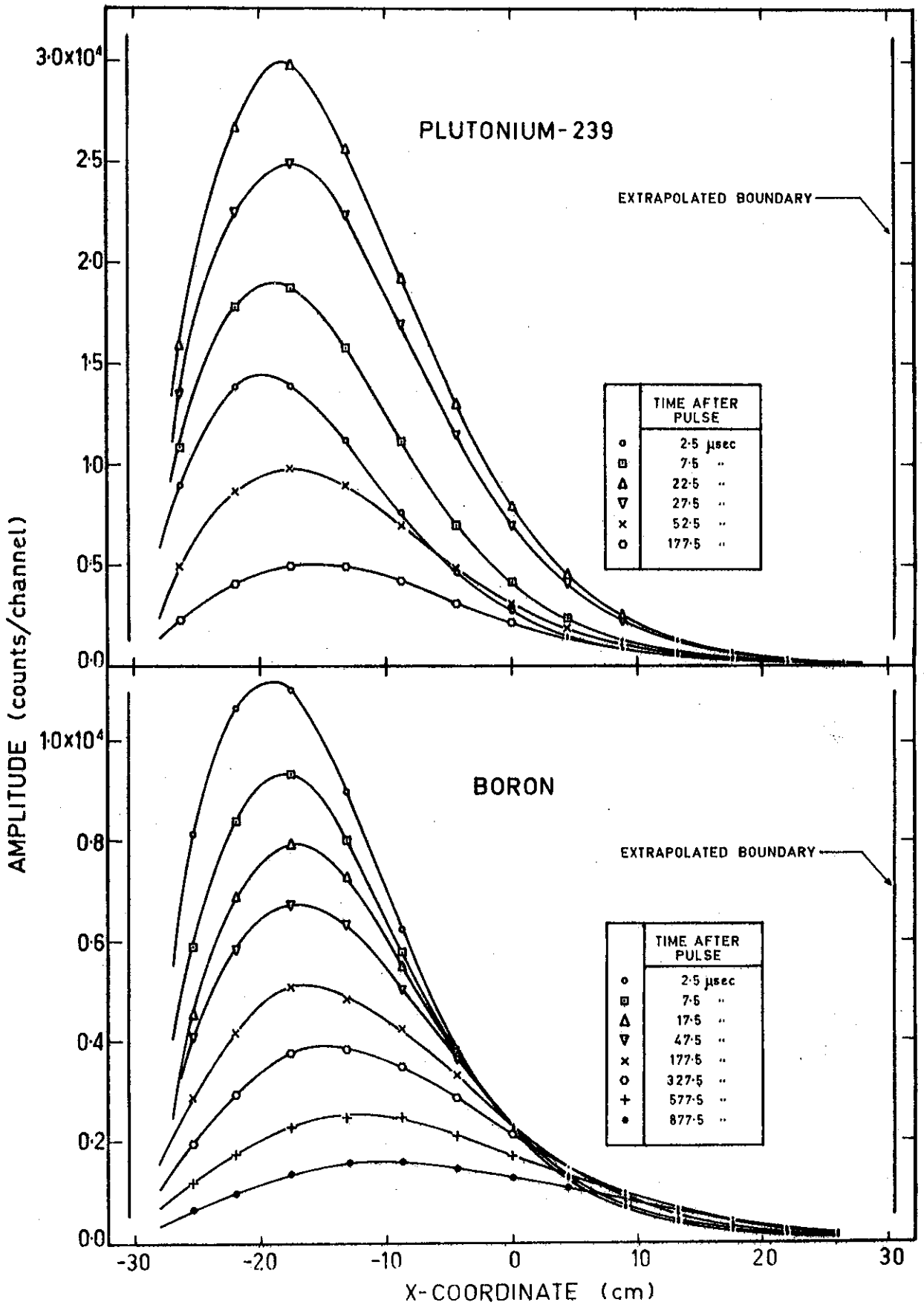

FIGURE 4. SPATIAL DISTRIBUTIONS OF THE PLUTONIUM-239 AND BORON REACTION RATES

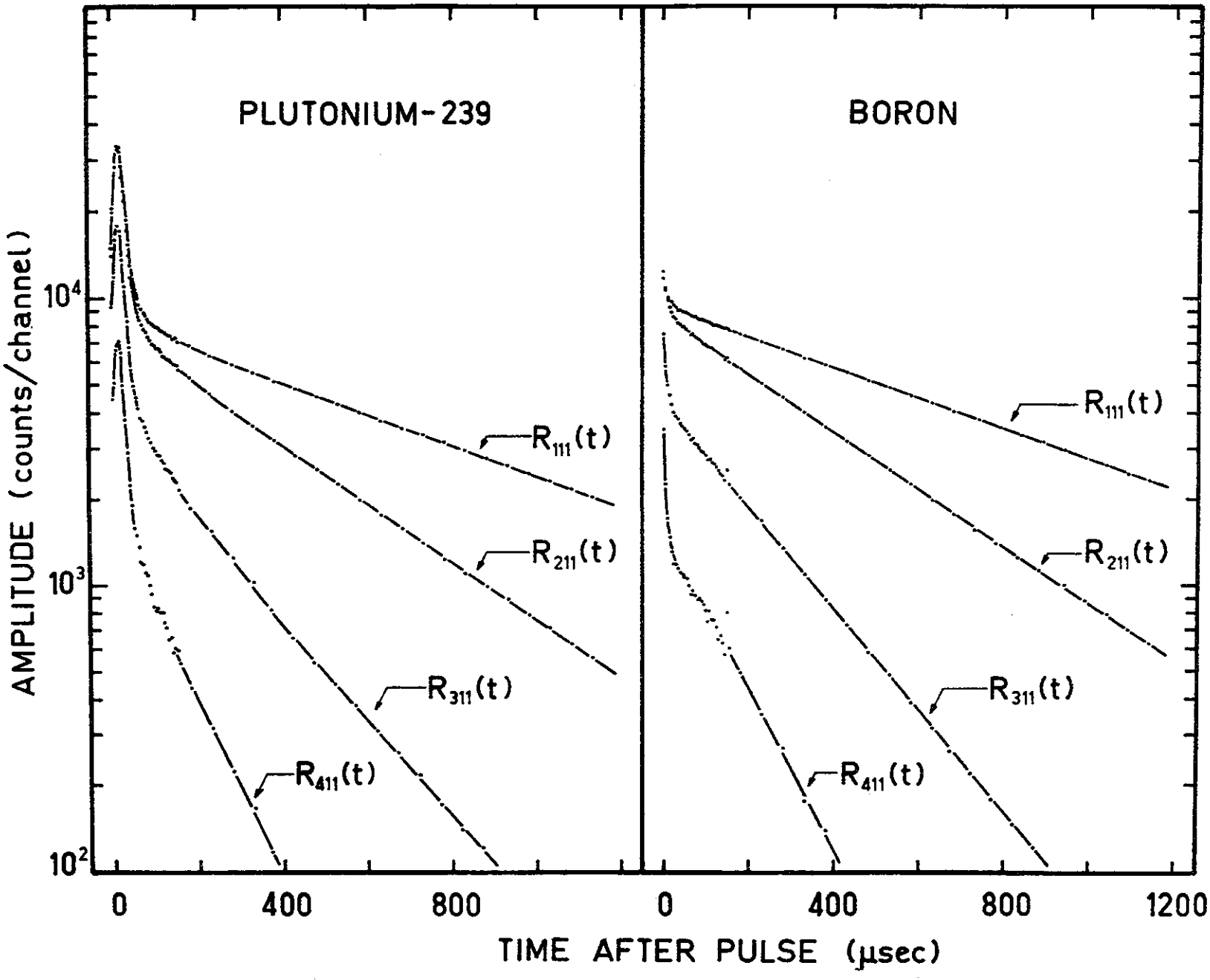

FIGURE 5. TIME-DEPENDENT REACTION RATES OF PLUTONIUM-239 AND BORON ASSOCIATED WITH VARIOUS FOURIER SPATIAL MODES

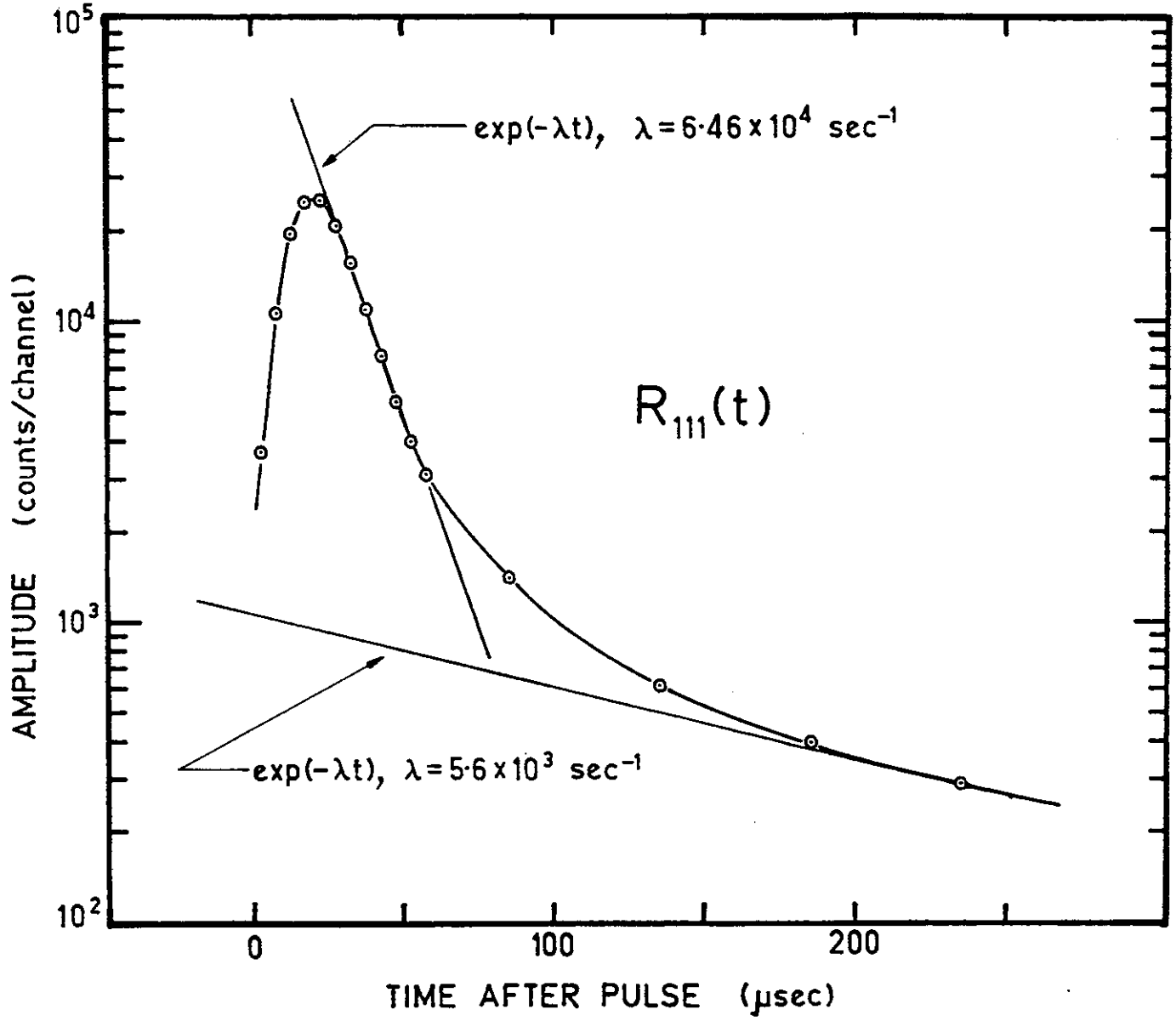

FIGURE 6. THE TIME-DEPENDENT REACTION RESPONSE DUE TO THE NON-1/v PART OF THE PLUTONIUM-239 FISSION CROSS SECTION

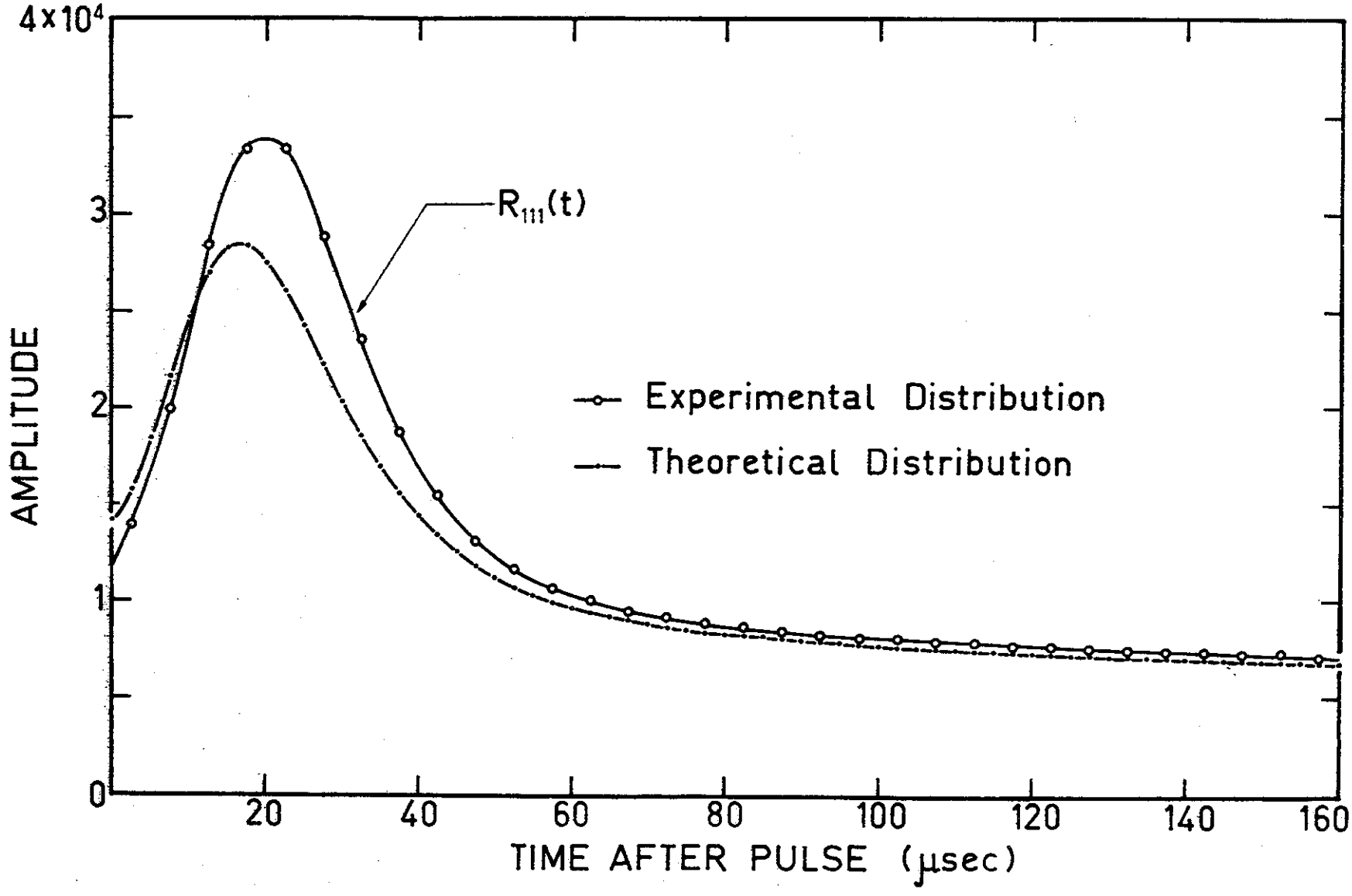

FIGURE 7. A COMPARISON OF THE MEASURED AND CALCULATED PLUTONIUM-239 REACTION RATE ASSOCIATED WITH THE FUNDAMENTAL FOURIER SPATIAL MODE

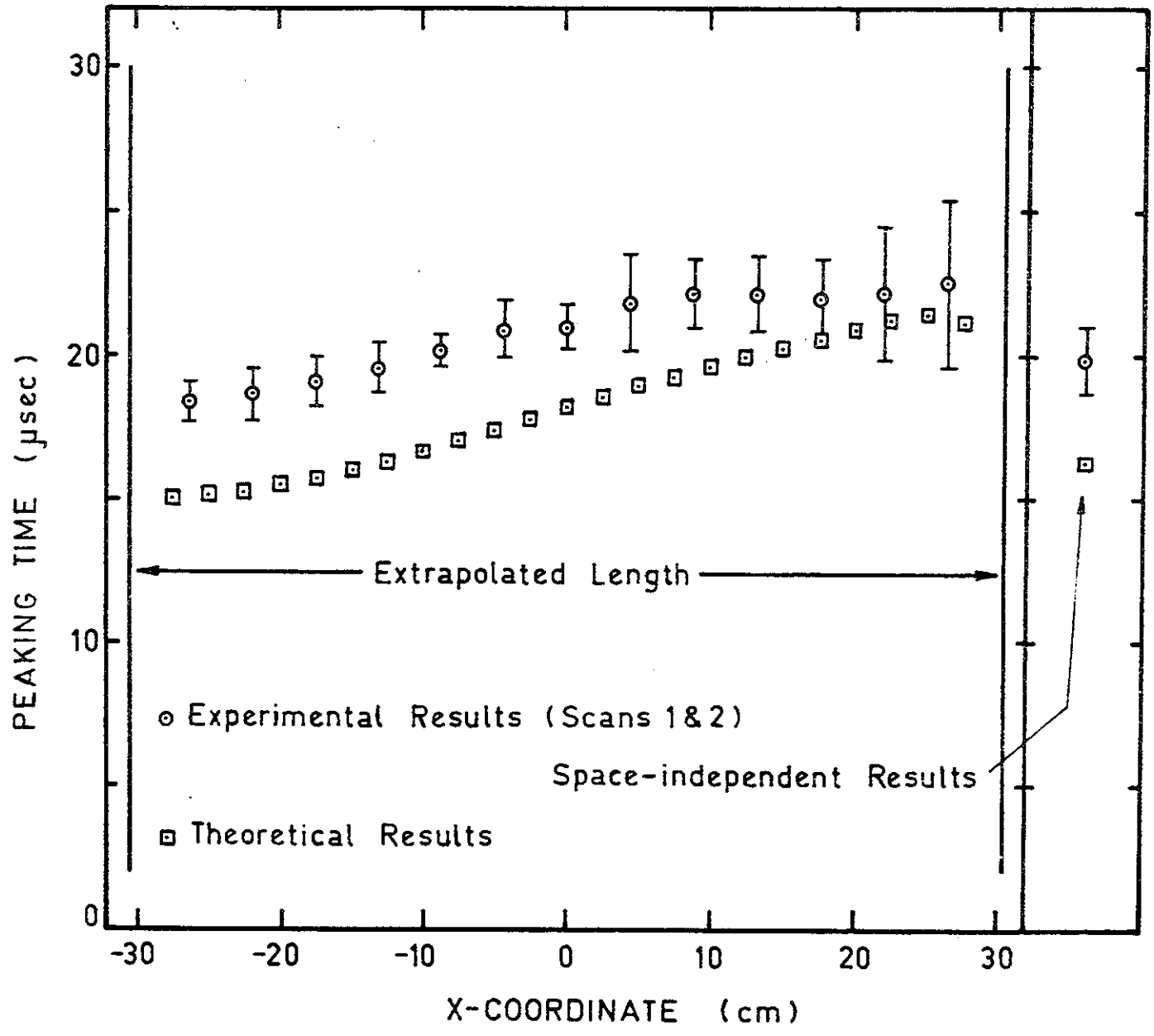

FIGURE 8. A COMPARISON OF THE MEASURED AND CALCULATED PEAKING TIME OF THE PLUTONIUM-239 REACTION RATE VERSUS POSITION