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Measurement of the Time Dependence of Neutron Slowing-Down and Thermalization in Heavy Water

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SUMMARY

The behaviour of neutrons during their slowing-down and thermalization in heavy water has been followed on the time scale by measurements of the time-dependent rate of reaction between the flux and the three spectrum indicators indium, cadmium and gadolinium. The space dependence of the reaction rate curves has also been studied. The time-dependent density at 1.46 eV is well reproduced by a function, given by von Dardel, and a time for the maximum density of 7.1 ± 0.3 μs has been obtained for this energy in deuterium gas in agreement with the theoretical value of 7.2 μs. The spatial variation of this time is in accord with the calculations by Claesson. The slowing-down time to 0.2 eV has been found to be 16.3 ±2.4 μs. The approach to the equlibrium spectrum takes place with a time constant of 33 ±4 μs, and the equilibrium has been established after about 200 μs. Comparison of the measured curves for cadmium and gadolinium with multigroup calculations of the time-dependent flux and reaction rate show the superiority of the scattering models for heavy water of Butler and of Brown and St. John over the mass 2 gas model. The experiment has been supplemented with Monte Carlo calculations of the slowing-down time.

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1. INTRODUCTION

The measurements of differential cross sections for the scattering of neutrons do not cover all energies and angles, and there is thus a need for complementary measurements of integral kind, which can be used for estimation of the validity of interpolated cross section data or theoretically derived data from model calculations. The experimental study of the time-dependence of the degradation of the energy of fast neutrons, injected in a moderator, is one of several methods to obtain such information. Time-dependent spectra and related quantities are obtained from such experiments. In practice, a point source has to be used in the measurements, and the analysis of the results must therefore take into account the spatial variation of the energy-time relations. Thus, such experiments are also of value for the comparison with theoretical investigations of the effects of diffusion during the moderation. Until recently, only a few measurements have been performed on heavy water, and before describing the present investigation, a summary will be given of the results obtained earlier.

2. EARLIER WORK ON TIME DEPENDENCE OF SLOWING-DOWN AND THERMALIZATION IN HEAVY WATER

2.1 Theory

Of the numerous papers giving expressions for the velocity - time distribution in an infinite medium during the slowing-down of high energy neutrons in a deuterium gas, the most fundamental are those of Marshak [1], Waller [2], von Dardel [3] and Svartholm [4]. More complete references and the latest developments are given by Koppel [5] and Boffi [6]. For the purpose of describing the time-dependent neutron density at energies in the eV region, the following "trial" function of von Dardel [3] is a good approximation.

$$n(v,t) = v^{5/4} (\Sigma_s t)^{9/4} e^{-\left(v \Sigma_s t + \frac{b}{v \Sigma_s t}\right)} \qquad (1)$$

The neutrons start at the time $t = 0$ with velocities which are very much higher than the velocity v. Σ_s is the macroscopic scattering cross section, assumed to be energy independent, and b is a constant. A value for

b of 0.903 is given by von Dardel by requiring an optimum fit of the moments of the trial distribution function to the exact moments, which are known. The maximum number of neutrons of a velocity v' are found at the time $t_{max.n}$, given by

$$t_{max.n} = \frac{1}{v' \Sigma_s} \left(\frac{9}{8} + \sqrt{\frac{81}{64} + b}\right) \tag{2}$$

Using Eq. (1), the time-dependent slowing-down density is found to be

$$q(v',t) = \frac{d}{dt} \int_o^{v'} n(v,t)dv =$$

$$= (v' \Sigma_s)^{9/4} t^{5/4} \cdot e^{-\left(v' \Sigma_s t + \frac{b}{v' \Sigma_s t}\right)} \tag{3}$$

the maximum of which is found at the time

$$t_{max.q} = \frac{1}{v' \Sigma_s} \left(\frac{5}{8} + \sqrt{\frac{25}{64} + b}\right) \tag{4}$$

The mean slowing-down time to the velocity v' can be calculated from the following equation:

$$\bar{t} = \frac{\int_o^\infty tq(v',t)dt}{\int_o^\infty q(v',t)dt} \tag{5}$$

In pure heavy water, Σ_s of the deuterium atoms present is $0.222\ cm^{-1}$. For 1.46 eV we find the maximum neutron density at the time 7.03 μs, and the maximum slowing-down density at 4.77 μs. The slowing-down time is found to be 7.28 μs by the use of $q(v',t)$ from Eq. (3) in Eq. (5). These figures should, however, be regarded as approximate, founded as they are on the Eq. (1), which is known to deviate slightly from the exact function [7]. The presence of oxygen in heavy water will modify the time distributions and shift them towards shorter times. Heavy water of reactor quality has a small content of light water, and this also influences the slowing-down time, although to a smaller extent.

The thermal motion of the moderator atoms has the opposite effect, increasing the slowing-down time by at most 2 per cent [8].

In heavy water, the diffusion of the neutrons during the slowing-down is appreciable, and the velocity-time distribution will be space dependent. In experiments, using a pulsed source, this effect is important. Attempts to solve this problem have been made by Dyad'kin and Batalina [9], who treated the space, velocity and time-dependent case for mixtures of elements, and by Claesson [10], who calculated $n(r, v, t)$ in a P1-approximation, an extension of an earlier work for the case of hydrogen [11].

The Monte Carlo method is well suited to the calculation of slowing-down time distributions. No such calculations seem to have been published for heavy water.

The time dependence ot the gradual approach to equilibrium has been treated by many authors. von Dardel [3] showed, that during the transition from the slowing-down distribution to the final spectrum the neutrons may be assumed to have a Maxwellian distribution with an associated neutron temperature, which falls exponentially with time to the temperature of the moderator with a decay constant, characteristic for each moderator. This description is rather good for hydrogen, and improves with increasing atomic mass. An expression was given for the thermalization time constant, which was found to be 58 μs for heavy water, assuming the molecule to be rigid. [12].

Purohit [13] and others have treated the problem by using a polynomial expansion of the energy, space and time-dependent flux and by solving the Boltzmann equation as an eigenvalue problem. This results in a description of the time-dependent flux as a series of energy eigenfunctions, each decaying exponentially with a decay constant, the corresponding eigenvalue, which is a function of the integral parameters of the scattering law. Some limitations are inherent with the method and have been extensively discussed in the literature during the last years, and also at the conferences dealing with neutron thermalization, at Brookhaven 1962 [14] and at Karlsruhe 1965 [15]. The establishment of the equlibrium is expressed by the higher eigenfunctions, and if the first eigenfunction dominates during the thermalization, the time constant is found from the following formula:

$$t_{th} = \frac{3\sqrt{\pi}}{2\, m_2\, v_o\, N}\, f_t \qquad (6)$$

where m_2 is a parameter, which expresses the thermalizing power of the moderator, v_o is the most probable velocity of thermal neutrons and f_t is a correction factor for higher order polynomials in the eigenfunction expansion. N is the number of molecules per cubic centimeter. An m_2 value of 10.7 barns per molecule, calculated from a scattering kernel for the realistic D_2O model of Butler [16] has been given by Purohit and Sjöstrand [17]. By the use of an f_t value of 1.24, calculated for the deuterium gas model by Shapiro [18], the time constant is found to be 42 μs.

A new attempt to connect the regions of slowing-down and thermalization in an analytical treatment has been made by Koppel [19], who used the time moment method for the heavy gas model. A more practical method for such connection is to use the multigroup formalism for the calculation of $\Phi(E,t)$ for the whole time and energy region. The slowing-down distribution, acting as a time-dependent source feeding neutrons into energy groups below 1 eV, may be obtained by using many groups in the high energy region or by using an analytical expression for $n(v,t)$. The former method has been used by Ghatak and Krieger [20] for light water calculations, and the latter by Purohit [21] for light and heavy water.

2.2 Experiment

The first measurements of the time scale of the moderation process in heavy water were made by von Dardel [3]. Transmission measurements with a cadmium filter on the leakage flux from a 6 litre volume with a time resolution of 2 μs were found to be in satisfactory agreement with predictions from Eq. (1) during the first 30 μs after the neutron burst. The curves showing the leakage flux as a function of time indicate a time for the establishment of the equilibrium flux of the order of 200 μs. The approach to equlibrium takes place in an exponential manner as derived theoretically. The experimental limitations, mainly low source intensities and small available water quantities, did not allow a quantitative determination of the thermalization time constant. The pulsed neu-

tron work on heavy water has later been concentrated on the determination of diffusion parameters with methods, also outlined by von Dardel. The next study of the slowing-down process in heavy water was made by Eichelberger [22], who used the method, applied by Engelmann [23] for light water, to detect and perform a time analysis on gamma rays from the capture of 1.46 eV neutrons in an indium foil, introduced in the medium. The time distribution of the counts was expected to display a peak at 6 μs after the injection, representing the time for the maximum of Eq. (1), modified for the leakage out of the 11 litre volume during slowing-down. The measurement gave a slight increase in the counting rate around 7 μs, and the result was interpreted as an indication of a chemical binding effect in the eV region. Profio and Eckard [24] used the same method for measurements in 21 litre water. A NaI scintillation counter was placed in the center of the container, and indium was introduced as foils and in solution. The presence of 1.46 eV neutrons in the time region 3 - 8 μs was shown. The experimental conditions make it difficult to draw further conclusions from the results, influenced as they are by the small geometry, time shift effects due to a rather high indium concentration, and the perturbation, caused by the detector. The calculated time of the maximum in the infinite medium, 7 μs, should be decreased by the first two effects and increased by the last.

Measurements of time-dependent neutron spectra in the energy region below 1 eV have been performed by Poole and Wydler [25], who studied the time region 44 - 1000 μs with a moderator volume of 280 litres. Kryter et al. [26] measured in the time interval 20 - 2000 μs and had a 125 litres water cube. The neutron bursts were produced by linear accelerators and the spectra were measured as a function of time by means of a chopper. The first group obtained good agreement between the experiments and spectra, calculated by the use of the realistic scattering kernel, whereas the second found this kernel not to be perfect, but better than a free gas model with the fictitious mass 3.6. Both groups found, that the spectra did not change in shape at times later than 200 μs.

3. THE PRESENT INVESTIGATION

The review of the earlier experimental work in the field shows that the existing measurements have been done under conditions, which made

it difficult to confirm in detail the theoretical prediction for the time dependence of the slowing-down process in heavy water. The approach to thermal equilibrium should be explored under better conditions. The effects of diffusion during moderation would also be interesting to study. It was therefore decided to make measurements on heavy water by the use of a method, developed earlier for measurements in light water.

The principles of the time-dependent reaction rate method have been described in earlier papers by Möller and Sjöstrand [27] and by Möller [28]. After the production of a burst of neutrons in a moderator at the time $t = 0$, the flux $\Phi(E, r, t)$ at the position r is studied by measuring its (space-dependent) rate of reaction $R(r, t)$ with a spectrum indicator, having an energy-dependent macroscopic cross section $\Sigma_a(E)$. The measurement is performed by the detection of gamma rays from neutron capture in the indicator. The reaction rate is given by the following formula:

$$R(r, t) = \int \Phi(E, r, t)\, \Sigma_a(E)\, dE \qquad (7)$$

Small quantities of the indicator are distributed homogenously in order to keep the perturbation of the flux low. Time analysis is performed on the pulses from the gamma ray detector. The analysis of the experimental results depends on the indicator used. If it has only one resonance, situated in the eV region, the result will be the $\Phi(E, r, t)$ (or $n(v, r, t)$) curve for the energy of the resonance. For indicators with resonance in the near thermal region, a great energy interval will contribute to the momentary rate, and the analysis has to be done by the use of integral concepts, or by calculation of $\Phi(E, r, t)$ for specific models and integration of these curves over the indicator cross section, before comparing with experimental data. The use of an indicator with a $1/v$ cross section will yield $n(r, t)$.

In practice, an indicator container of a finite volume must be used, and the measured reaction rate will refer to the integral over this volume. In this integration a spatial weighting factor should also be included, representing the efficiency of the gamma detector for each point.

The aim of the experiment was to obtain information that could be related to the case of a point source in an infinite medium. Pre-

liminary experiments with a cylindrical vessel with a diameter of 40 cm and a volume of 50 litres with the source outside the water, showed a great spatial dependence of the reaction rate curves for cadmium. Close to the boundary near the source, the maximum rate was found at 17 μs, whereas the measurement at the centre gave a time for the maximum of 23 μs. A quantitative treatment of these leakage effects, which depend on the source spectrum and the geometrical conditions, would be difficult. In order to obtain a stationary spectrum for the infinite medium in heavy water, several cubic meters must be used. Time-dependent measurements in such volumes would be impractical. However, since we are only interested in following the neutrons until they are completely thermalized, a smaller tank can be accepted. By using Spiegel and Richardson's data [29] for the neutron age in heavy water in the formula of the age theory for the spatial distribution around a point source, we find that of the neutrons starting with an energy of 2.6 MeV, 98 per cent will be found within a sphere having a radius of 50 cm at the time, when they have become thermalized. 89 per cent are found within 40 cm. For a cubic tank with 1 m side, the figures will be lower because of the absence of the inscattering from the outer region, but the percentage of just thermalized neutrons within the tank will be reduced only slightly. The sum of measurements along a radius in the tank, weighted by r^2, will therefore, with reservation for the missing contribution of the escaped neutrons, represent a measurement in an infinite medium during the time to complete thermalization. If the source energy is decreased, the escaped fraction of neutrons will be still lower.

Three indicators were used; indium, which yields the time-dependent density at 1.46 eV, cadmium with a resonance at 0.178 eV, close to the thermal region, and gadolinium, the cross section of which rises in the lower thermal region.

4. THE PERFORMANCE OF THE EXPERIMENT

The measurements were made with the same equipment as was used in the measurements in light water [27, 28]. The neutrons were produced by means of the $Li^7(p,n)Be^7$ reaction from a thick target. The proton energy was 3.7 MeV, and the resulting fast neutron spectrum had a maximum energy in the forward direction of 2 MeV. The neutron pro-

duction was monitored by means of a small boron counter, placed far away from the indicator container. The water tank, made of stainless steel, had the dimensions 97 x 97 x 107 cm, and the water, 875 litres, had the composition 99.5 % D_2O and 0.5 % H_2O. The water height above the target was 39 cm. Boron-loaded plastic sheets were used as neutron shields around the tank and also inside the tank in the upper half in order to obtain a reduction of the background of gamma rays from neutron capture in the steel walls. The target was situated in the centre of the tank in an aluminum tube, 2.5 cm in diameter and with a wall thickness of 0.2 cm. The container for the indicator solutions, made of aluminum, 0.15 cm thick, was cylindrical with a diameter of 25.0 cm and a height of 10.5 cm, thus having a volume of about 5 litres. It was suspended under the scintillation counter with its bottom facing the target. The diameter of the plastic scintillator was 5 cm and its length 20 cm. The distance between the lower end of the scintillator and the container boundary was 6 cm. The assembly of scintillator and container could be moved along a line extending horizontally from the target tube in the forward direction relative to the proton beam with the axis of the container on the line. The time analysis of the detector pulses was performed by means of a time-to-pulse-height converter and a multi-channel analyzer. The time analyzer was started before the neutron burst. A discriminator rejected pulses corresponding to gamma ray energies lower than 2.2 MeV.

The indium measurements were made with an indium sulphate solution with a macroscopic cross section at 1.46 eV of 0.237 cm^{-1}, corresponding to a resonance escape probability of about 0.95 and a thermal lifetime of 2790 μs. A test run with a solution with one fifth of this concentration gave a too low signal to background ratio. A shift of the time-dependent neutron density curves, measured with a strong resonance, towards shorter times will occur as shown in [28]. At 1.46 eV, the macroscopic scattering cross section of heavy water, including oxygen, is 0.348 cm^{-1}. The mean time a neutron will spend at this energy will then be reduced from $(v \Sigma_s)^{-1} = 1.72$ μs to $(v \Sigma_s + v \Sigma_a)^{-1} =$ $= 1.02$ μs, that is by 0.72 μs. The exchange of neutrons with the surroundings will reduce the shift correction. As the figure 0.72 μs refers only to the peak of the cross section, the real shift will be much smaller.

For cadmium, measurements were made with two solutions of water free sulphate, one with $\Sigma_a = 0.00573$ for thermal neutrons, which means a lifetime of 600 μs, and one with one third of this concentration, i.e. a lifetime of 1800 μs. For gadolinium, measurements were made with a concentration for a lifetime of 600 μs. The gadolinium solution was prepared by dissolving the oxide in a slight excess of concentrated sulphuric acid and adding heavy water. The absorption of the aluminum of the container corresponds to a macroscopic cross section of 0.00074 cm^{-1}, if the material is distributed over the container. The light water content of the water results in a macroscopic absorption cross section of 0.00011 cm^{-1}.

Measurements were made at four places, the distance between the target and the container being 0, 10, 20, and 30 cm. At each position, a background measurement was made with an identical container without dissolved indicator. The measured curves were corrected for the dead time of the time analyzer. The background curves were smoothed before being subtracted from the indicator measurements. In the cadmium measurement, a minor contribution from a deposit of indium in the container, left from the indium measurement, was also eliminated. The overlapping background from preceding bursts was subtracted. At the time of injection, this background was very low for the indium measurement, about 1 % of the peak reaction rate, and varying with the distance between 2 and 10 % for the cadmium and gadolinium measurements. For the latter, decay constants of the overlapping background were found from measurements with different time ranges.

5. RESULTS OF THE MEASUREMENTS

Fig. 1 a shows the results from the indium measurements. The channel width and the time resolution were 0.317 μs. The time separation of the neutron bursts was 1460 μs and each had a duration of 0.2 μs. The curves have been normalized relative to each other by means of the monitor. The number of counts per channel at the peak ranges from 3000 to 240 in the not normalized records. The counts at the first four channels have been omitted since they display statistical fluctuations without significant meaning. With increasing distance from the source, the time for the maximum rate is found to be 5.2, 5.9, 6.8 and 7.4 μs.

In Fig. 1 b the curves from Fig. 1 a have been summed after being weighted by r^2. The effective values of r have been taken as 7, 15, 25, and 35 cm. The peak occurs at the time 6.3 μs. The contribution to these curves from the reaction with the higher resonances of indium at 3.86 and 9.10 eV is quite small as can be seen in the earlier calculated corresponding curves for light water [28]. The maxima of these distributions would appear at the times 4.3 and 2.8 μs according to Eq. (2).

Close to the source, a measurement was also made with a 1 litre indicator volume in order to get an improved spatial resolution. The peak appeared at 4.9 μs in this measurement.

These measurements were repeated with a proton energy of 4.7 MeV in order to check the possible energy dependence of the time of the peak. Except for the intensities, the results did not differ noticeable from those shown in Fig. 1.

In Fig. 1 b a full line curve has also been included, showing the time-dependent neutron density at 1.46 eV according to Eq. (1) for the case of free deuterium atoms at rest.

The results of the measurements with cadmium are shown in Figs. 2 and 3 for the lifetime 600 μs with the time resolutions 1.90 and 4.65 μs. The curves for the lifetime 1800 μs are not shown since they are, except for the statistical accuracy, almost identical with the Figs. 2 and 3. The gadolinium curves are shown in Figs. 4 and 5. The experimental points have been replaced by smooth curves, and these curves have been normalized to the same monitor counts and weighted by r^2. The pulse separation was 2500 μs for the cases with 1.90 μs resolution, and 10000 μs for those with 4.65 μs resolution. The number of counts per channel was 6800 at the peak of the cadmium curves closest to the source in Figs. 2 and 3, and 3750 at the largest distance. The gadolinium curves had about the same accuracy, whereas the cadmium runs with the weaker solutions were less accurate at the larger distances. At the top of each of the Figs. 2 - 5 is shown the sum of the four normalized and weighted curves. The sum curves have been kept unsmoothed in order to illustrate the accuracy of the results.

Precautions were taken to keep the equipment stable during the measurements, and the detector efficiency was regularly checked by

means of a standard radioactive gamma ray source. Comparisons between the measurements under different conditions, normalized by means of the monitor counts and corrected for different concentrations and channel widths, showed differences less than 3 % for both the sum curves and the separate runs.

In order to investigate the influence on the measured curves of the presence of the gamma ray detector, a cadmium measurement was made at the distance 10 cm with a plexiglass cylinder of the same size as the detector, hanging by its side. No difference could be seen between this run and the earlier one. It was concluded that the perturbation caused by the detector could be disregarded.

6. MONTE CARLO CALCULATIONS

It is important to know how much the presence of oxygen and hydrogen in the heavy water, used in the experiment, will change the distribution given by Eq. (1) and the time values derived therefrom. Monte Carlo calculations of the time-dependent slowing-down density were obtained by the use of the existing program of Högberg, Ljunggren and Persson [30], which, originally intended for calculations of space-dependent distributions, was changed by its originators to generate time distributions. Three cases were run, each with about 25000 neutron histories. The heavy water was given the composition 99.5 % D_2O and 0.5% H_2O. First only the deuterium was taken into account, then the corresponding amount of oxygen was added, and last the light water was also introduced. The source energy was 1 MeV and isotropic scattering was assumed. Distributions were taken for the energies 5.00 and 1.46 eV. The deuterium result for 1.46 eV is shown in Fig. 6, where the results for the two other compositions are also included as smooth broken line curves. The peak in the distribution occurs at the time 5.0 μs for the deuterium case, shifts only slightly when oxygen is added and comes at 4.6 μs when the light water is introduced. The mean slowing-down time was calculated according to Eq. (5), the results being shown in Table 1. The slowing-down time to 1.46 eV decreases from 6.8 to 6.3 μs when the oxygen and hydrogen are added to the deuterium.

A least squares fit of Eq. (3) with b kept as a parameter was made for the distribution at 1.46 eV in the deuterium case. A very good fit was

obtained and a value of $b = 0.96 \pm 0.02$ was found, somewhat higher than von Dardel's value 0.903. The fitted curve is included in Fig. 6 as a full line curve.

7. DISCUSSION OF THE RESULTS

7.1 The time distributions during the slowing-down

The space integral of the indium measurements shown in Fig. 1 b agrees in shape quite well with the calculated time-dependent density curve according to von Dardel, included in the figure. The peak appears at 6.3 ± 0.2 μs instead of 7.0 μs for the case of slowing-down against deuterium atoms at rest. If we want to compare them, corrections have to be applied for the various effects mentioned earlier. The shift towards shorter time caused by the capture in indium is lower than 0.7 μs, and assuming that the observed reduction of the correction from 0.07 to 0.02 in the case of light water [28] can be extended to our case, we find a correction of 0.2 μs. The higher indium resonances also shift the measured curves to the left, and the correction for this effect is estimated to be 0.2 μs by using the correction found for light water [28] and multiplying by the time scale ratio. The presence of oxygen and light water shifts the peak of the Monte Carlo calculated slowing-down density curve by 0.4 μs, and this correction should also be applicable to the case of the number density. The total correction of 0.8 μs with an estimated uncertainty of 0.2 μs to the measured time for the peak thus yields a time of the maximum density in deuterium of 7.1 ± 0.3 μs. The theoretical value of 7.03 μs should be increased by 2 % by the effect of thermal motion [8] to 7.2 μs. We find, therefore, good agreement between the experiment and von Dardel's expression for the time-dependent density. The Monte Carlo calculation results are also well reproduced by the equation for the time-dependent slowing-down density derived from von Dardel's expression.

The space dependence of the measured curves has been compared with the predictions from the P1 calculations by Claesson [10], where an expression is given for $n(r, v, t)$. The formula has been evaluated with the source energies 0.1 and 1.0 MeV for the distance 0, 5, 10, ...

50 cm from the source at the velocity corresponding to 1.46 eV, and the time for the maximum, $t_{max.}$, determined. The difference in the $t_{max.}$ values is very small for the two different source energies. The $t_{max.}$ observed in the measurements at different positions was corrected by 0.8 μs according to the preceding discussion in order to represent slowing-down in deuterium gas. Fig. 7 shows the results of the experiments and the calculated relation between $t_{max.}$ and the distance from the source (full line).

The experimental results are in good agreement with the theoretical curve, especially if it is observed, that the calculated values should be increased by 2 % for the thermal motion, and that the theoretical approach used implies a rather high uncertainty near the source. The experimental points close to the source may also be influenced by the leakage of neutrons through the target tube and by absorption in the target.

In Koppel's paper [5], an approximate method was shown to give a value of b (Eq. (1)) of 1.5. Since this is quite high compared to von Dardel's value and the result of the Monte Carlo calculation, Claesson's calculations were checked on this point by summing $n(r, v, t)\, r^2$ in order to get an infinite medium curve. When Eq. (1) was fitted to the result in the time region 4 - 12 μs, a value for b of 1.1 was obtained. This shows that the calculations have not introduced too much simplification and distortion of the infinite medium curve.

The spatial variation of $t_{max.}$ was also calculated according to the equation given by Dyad'kin and Batalina [9]. For the distances 0 - 50 cm from the source, $t_{max.}$ was found to vary from 11.6 to 27.8 μs, which is far beyond reasonable values. This result indicates a need for further theoretical treatment of the time-dependent slowing-down and its space dependence in mixtures of elements.

7.2 The cadmium measurements

Whereas the indium results are dominated by the reaction from neutrons with energies within a relatively narrow resonance, the measurements with cadmium and gadolinium show the reaction of a flux extending over the whole thermal region and varying strongly with space and time. The curves obtained do not give easily interpreted informa-

tion for well defined energy intervals. In Fig. 2, we see that at r = 7 cm the neutrons with energies around 0.18 eV have their maximum reaction at 23 μs. Later the reaction rate decreases rapidly with time and changes into an exponential decay. During this period, both energy loss and leakage from the source region cause the reduction of the reaction rate. For the distances 15 and 25 cm, the peak comes later and the decay becomes slower as the distance is increased. This is explained by the diffusion during the moderation. For these distances, the leakage out of the container region is more compensated by the leakage into it than in the case of r = 7 cm. For the distance 35 cm, the raction rate shows first how the neutrons pass over the resonance peak and then how the neutron density stays constant up to about 225 μs, after which time it increases. This is explained by the wave of neutrons from the centre which in the time region 225 - 500 μs overcompensates the leakage out from the region around 35 cm. In Fig. 3, the extended time allows us to follow the change to fundamental mode decay also for this distance. The asymptotic decay, typical for a pulsed experiment for diffusion parameter determination, has apparently not yet been reached after 800 μs.

Although we cannot separate energy, time and density for the individual curves, the sum curve can give us information about the moderation on the energy - time scale, since it is connected to the infinite medium case, for which the analysis is less complicated.

If the measurements had been performed under the ideal condition of no absorption, the sum curve would show how the neutron flux passes the resonance region 0.1 - 0.25 eV, giving the maximum reaction rate around 26 μs, and how the thermalization proceeds with a continuous change of the spectrum into the equilibrium Maxwellian distribution. During the last stage, the reaction rate for cadmium would decay exponenetial to a constant value. As shown in [27], the assumption of a neutron temperature, which approaches the moderator exponentially, leads to the interpretation of the decay constant for the reaction rate as the inverse of the thermalization time constant. For our finite geometry, we would expect a break in the constant asymptotic reaction rate when the neutrons begin to leave the tank. Fig. 3 shows this break at the time 450 μs. However, we also see, that we have an exponential decay of the sum curve during the earlier time. This is due to the ab-

sorption and leakage in the indicator volume and the absorption of the light water present. For the measurements shown in Figs. 2 and 3, the indicator concentration corresponds to an infinite medium lifetime of 600 μs for thermal neutrons within the container. The slope of the sum curve corresponds to a lifetime of 1150 μs. For the measurements with the three times weaker solution, the slope corresponds to a somewhat longer lifetime. The differences in the calculated and observed values have their explanation mainly in the diffusion of the neutrons at the container boundaries, and, since the container occupies only a small part of the moderator, the decay of the neutrons in the tank as a whole actually takes place with an effective decay constant which is much smaller than is shown by the sum curve.

The dependence of these effects on position, time and geometry make a precise calculation of corrections to convert the sum curve into a curve for the zero absorption case very difficult. A reasonable approximation, however, is to consider the deacy observed in the region 200 - 400 μs to represent an effective absorption and to extrapolate it towards earlier times. For the time 0 - 200 μs, this represents a virtual loss of 14 % of the injected neutrons. As far as the absorption of the indicator is concerned, this approximation should be right since the two concentrations gave only small differences in the measured curves.

In order to obtain the time constant for the approach to equilibrium in the same way as for the measurements on light water [27], a least squares fit of the following function was made to the sum curves for the two concentrations.

$$R(t) = A_o e^{-\lambda_o t} + A_1 e^{-\lambda_1 t} = (A_o + A_1 e^{-(\lambda_1 - \lambda_o)t}) e^{-\lambda_o t} \qquad (8)$$

Data up to 350 μs were used, starting at the time 76 μs after the injection, at which time all neutrons have slowed down below 0.2 eV. The results are found in Table 2. The thermalization time constant t_{th} should be calculated from the following equation:

$$\frac{1}{t_{th}} = \lambda_1 - \lambda_o \qquad (9)$$

The weighted mean value for the two cadmium concentrations of $\lambda_1 - \lambda_o$ equals $(3.16 \pm 0.36)\ 10^4\ s^{-1}$.

A more detailed interpretation of the individual cadmium curves would require a theoretical treatment of the thermalization in energy, space and time, including the energy spectrum of the source. Since this is a formidable task, we have only made a comparison between the space dependence of the time for the maximum rate and the predictions from the slowing-down calculations of Claesson. As is shown in Fig. 7, the calculations and the experiment are still in reasonable agreement for the energy of 0.18 eV. However, the calculated curve should be corrected for thermal motion, and the experimental points do not refer to a well defined energy, so the value of this agreement is limited.

7.3 The gadolinium measurements

After the preceding discussion, the interpretation of the gadolinium curves in Figs. 4 and 5 is rather obvious. The gradual increase of the gadolinium cross section at lower energies is most clearly reflected by the sum curve, which mainly shows how neutrons are fed down into the lower thermal energy region. The individual curves show the composite effect of slowing-down, thermalization and diffusion, and we also note here the increase with time of the number of neutrons at the greatest distance before the leakage out becomes dominating.

The sum curve shows the same behaviour for longer times as the cadmium curves. Fitting Eq. (8) to the data, we should also obtain the decay constants for thermalization and for virtual absorption. The result, included in Table 2, shows rather good agreement with the cadmium results with respect to the values, but the accuracy of λ_1 is not so good. A_1 has a negative sign since the temperature coefficient of the effective absorption cross section is negative. Since this coefficient is about one third of that for cadmium, A_1/A_o is lower by this factor. This smaller sensitivity of gadolinium results in the large error in the λ_1 value. The fact that there are two capturing isotopes in gadolinium, implying that the flux reacts differently with the different cross sections, may also result in a slightly time-dependent detection efficiency if the gamma ray spectra differ for the two cases. For these reasons the parameters for gadolinium will be disregarded in the discussion of the thermalization time constant.

7.4 Reaction rate curves for infinite medium and no absorption

The time-dependent flux in a moderator may be calculated by numerical solution of the Boltzmann equation in the multigroup formalism, using scattering cross section from some model. Purohit [21] has performed such calculations for heavy water, using a Ferranti Mercury computer program. The program makes use of von Dardel's trial function (Eq. (1)) for feeding down neutrons into the region below 1 eV, which is divided into 22 groups. Scattering kernels for the gas model with mass 2, the Butler model, and the model of Brown and St. John [31] were used. The flux is integrated over suitable indicators, and the result is shown in Fig. 8. The models of Butler and of Brown and St. John give almost the same curves, for which reason the latter has been omitted. The mass 2 gas model results in a much faster thermalization and a lower peak in the reaction rate curve. The result of the experiment is also shown in the figure. The measured curve has been multiplied by $e^{\lambda_o t}$ in order to eliminate the virtual absorption, the λ_o value being taken from Table 2. The agreement between the experiment and the calculated curve for the Butler model is seen to be rather good, although the experimental peak comes at a later time. The difference indicates a smaller cross section for large energy transfers than predicted from the model. Our conclusion is therefore, that although the main features of the measurements are reproduced by these models, perfect agreement requires slight modifications of the models.

7.5 The thermalization time constant

From Eq. (9) we obtain a value for the thermalization time constant t_{th} of 32 ± 4 μs for our heavy water. In order to refer the time constant to pure heavy water, a correction should be applied for the light water present. Assuming that the decay constant is a sum of the contributions from the two components, we find a correction of 1 μs to the time constant. Our result is thus

$$t_{th} = 33 \pm 4 \text{ μs for } D_2O.$$

This value is considerably smaller than the earlier mentioned value 58 μs for the rigid molecule, and also smaller than the value 42 μs calculated from Eq. (6) with parameters based on the Butler model. A calculation of the time constant from the diffusion cooling coefficient and the temperature dependence of the diffusion constant, using parameters from [17], yields a value of 41 μs with a 66 % error. A fit of a sum of a constant and an exponential function to the later part of Purohit's calculated curve in Fig. 8 has resulted in an average time constant of 33 $\pm$ 2 μs for starting times between 79 and 96 μs, but for the time region 115 - 410 μs, a value of 42 $\pm$ 2 μs was obtained.

It must be realized, when considering these figures, that attempts to fit exponential functions to results from laboratory experiments, or to results from a multigroup calculation, always introduce errors if a "true" asymptotic thermalization time constant is aimed at. In order to obtain a fit, representing the establishment of the asymptotic equilibrium spectrum, one should start the procedure as late as possible. The statistical fluctuations in the data will reduce the accuracy of the parameter value more, the later the starting time. The time constant, we obtain, represents a parametric description of the thermalization in a certain time region. The value evaluated from the scattering kernel by the use of a Maxwellian flux of moderator temperature may well be somewhat different. Our time constant value is in good agreement with Purohit's result for the corresponding time region. The larger value obtained by Purohit for later starting times is close to that expected from the eigenvalue treatment with the Butler model scattering kernel. The shorter time constant at an earlier time should, in the eigenvalue formalism, imply the presence of higher energy modes at earlier times. It is, however, not possible to resolve higher modes from our data, and the theoretical ground for such a procedure is not very firm since the λ-values will approach the limit $(v\,\Sigma_s)_{min}$ of the moderator, where the discrete eigenfunction representation becomes inadequate. For heavy water $(v\,\Sigma_s)^{-1}_{min}$ is about 30 μs, which is close to the values mentioned above.

7.6 The time for complete thermalization

The sum curves in Figs. 2 - 5 show, that the transient, representing the thermalization, has died out about 200 μs after the fast neutron burst. This is apparently the time required for complete thermalization. The time value is in accord with the observations of Poole and Wydler [25], Kryter et al. [26] and Daughtry and Waltner [32].

7.7 The slowing-down time to 0.2 eV

The number of neutrons below a selected energy may be obtained as a function of time by the time-dependent reaction rate method by the use of an indicator with a cross section of the 1/v type, but with zero cross section at energies higher than the specified limit. It was shown in [27], that a mixture of cadmium and gadolinium can approximate such a cross section. By summing curves, measured individually for the two indicators and normalized to the same asymptotic value, in the proportion 1:4.64, the desired curve may be obtained. This procedure was undertaken with the experimental curves in Fig. 8, and the slowing-down time evaluated. The result obtained is

$$t = 16.3 \pm 2.4 \ \mu s \text{ for } 0.2 \text{ eV},$$

the error being estimated to have the same magnitude as for the light water case [27]. From Eq. (5) a value of 20.0 μs is obtained. The difference is due to the presence of hydrogen and oxygen, to the effects of chemical binding, and to thermal motion.

8. CONCLUSIONS

The application of the time-dependent reaction rate method to the study of the slowing-down and thermalization of neutrons in heavy water has been shown to yield useful information about these processes and their dependence on time, energy and position in the moderator. The comparison of the results for the slowing-down to the indium resonance at 1.46 eV with the theories for slowing-down in deuterium has demonstrated the applicability of the trial function for the infinite medium given by von Dardel, and has also illustrated the validity of the calculations of Claesson for the space and time-dependent slowing-down. The process

of thermalization, followed in the measurements with cadmium and gadolinium, is well described by a gradually cooling spectrum with a temperature, approaching that of the moderator with a time constant of 33 ± 4 μs. The slowing-down time to 0.2 eV has been found to be 16.3 ± 2.4 μs, and the thermalization has been completed after about 200 μs. The comparison of the experimental curves with the calculations by Purohit has shown, that the models of Butler and of Brown and St. John describe the scattering process quite well, although not perfectly. This supports the conclusion reached from measurements at other laboratories of time-dependent neutron spectra in heavy water. The detailed interpretation of the measurements at different positions in the medium demands a method for the connection of the processes of moderation and diffusion in energy, time and space. Our curves should be useful for checking different theoretical approaches to solve this problem. The results from the indium measurements indicate a need for further theoretical work on the slowing-down in mixtures.

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Table 1. Slowing-down times in heavy water, obtained from Monte Carlo calculations

Moderator composition	Slowing-down time, μs	
	5.00 eV	1.46 eV
99.5 % D_2	3.96	6.84
99.5 % D_2O	3.64	6.49
99.5 % D_2O + 0.5 % H_2O	3.51	6.27

Table 2. Parameters from experimental curve fittings

Measurement	A_o, 10^4	λ_o, 10^3 s^{-1}	A_1, 10^3	λ_1, 10^4 s^{-1}
Cd, 600 μs	1.37 ± 0.02	0.93 ± 0.05	3.72 ± 0.20	3.35 ± 0.39
Cd, 1800 μs	0.45 ± 0.03	0.75 ± 0.25	1.33 ± 0.23	2.43 ± 0.89
Cd, 600 μs	1.13 ± 0.01	1.15 ± 0.05	- 0.70 ± 0.16	4.55 ± 2.34

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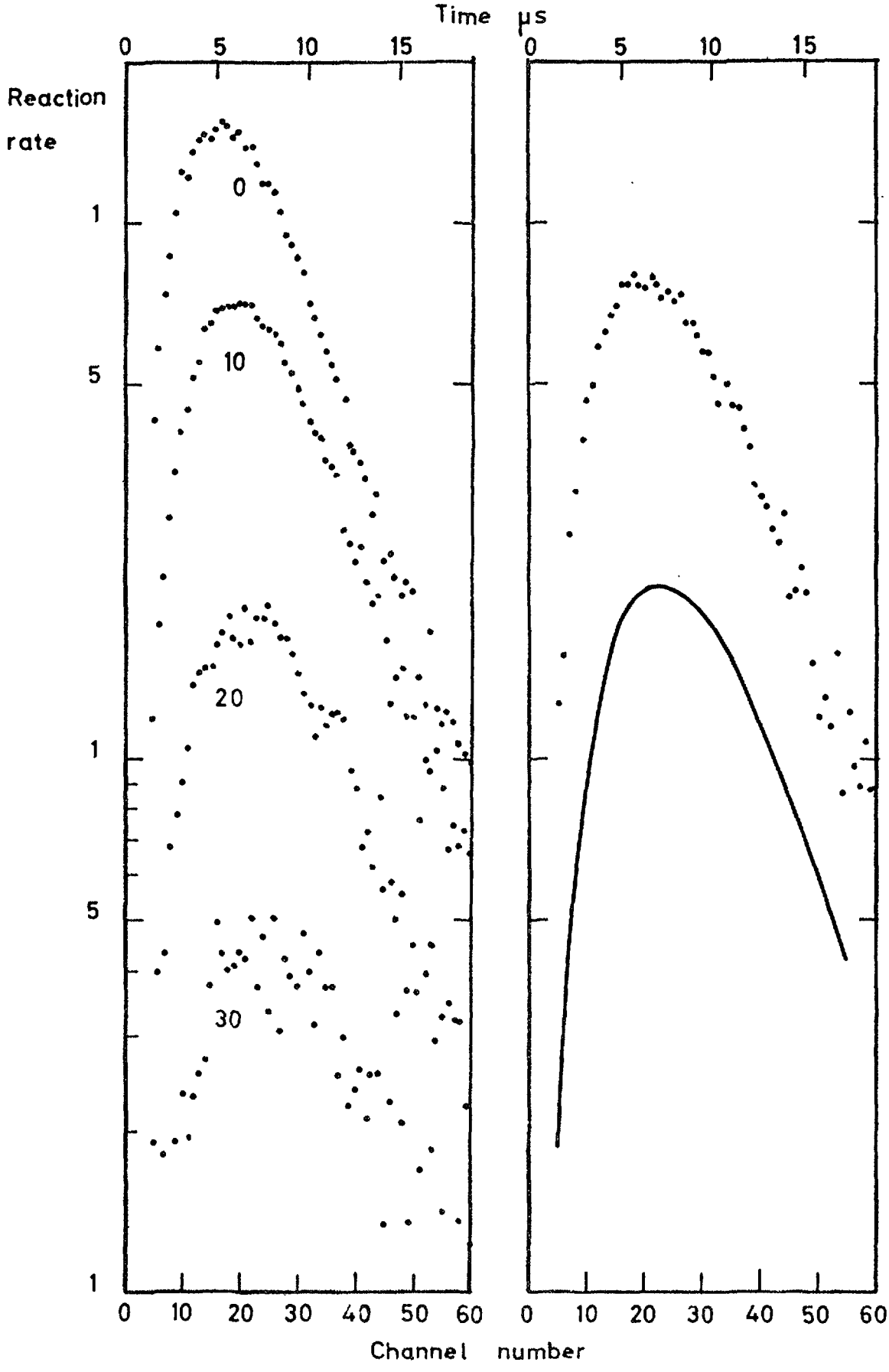

Fig. 1 a. Measurement with an indium solution at the distances 0 - 30 cm between the source and the container. The time resolution is 0.317 μs.

Fig. 1 b. The weighted sum of the curves of Fig. 1 a, representing the infinite medium case. The full line curve shows the time-dependent density in deuterium at 1.46 eV according to von Dardel.

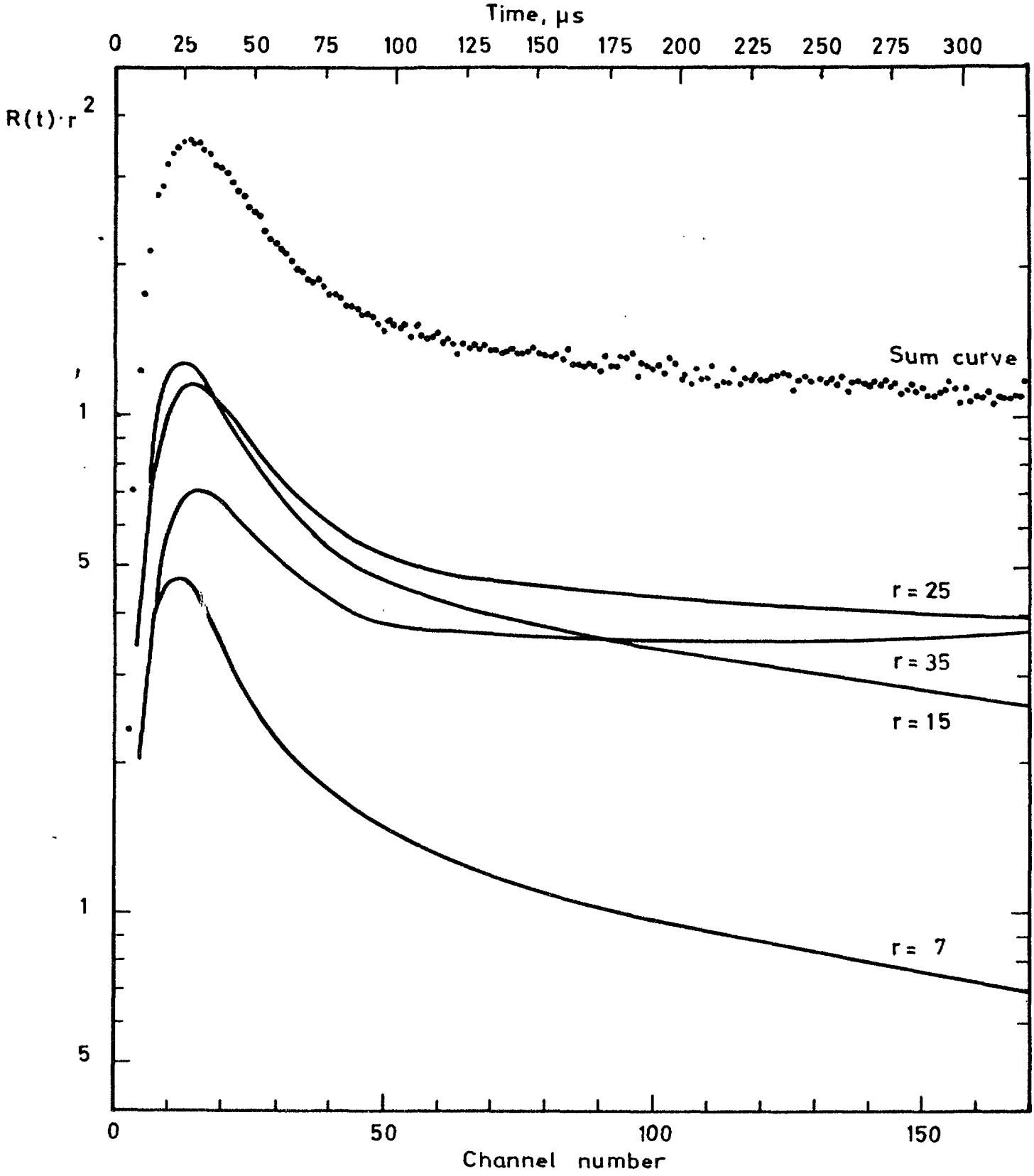

Fig. 2. Reaction rate curves for cadmium at various positions (full lines), normalized and weighted by the square of the effective radius. The upper curve is the sum of the lower curves. Time resolution 1.90 μs.

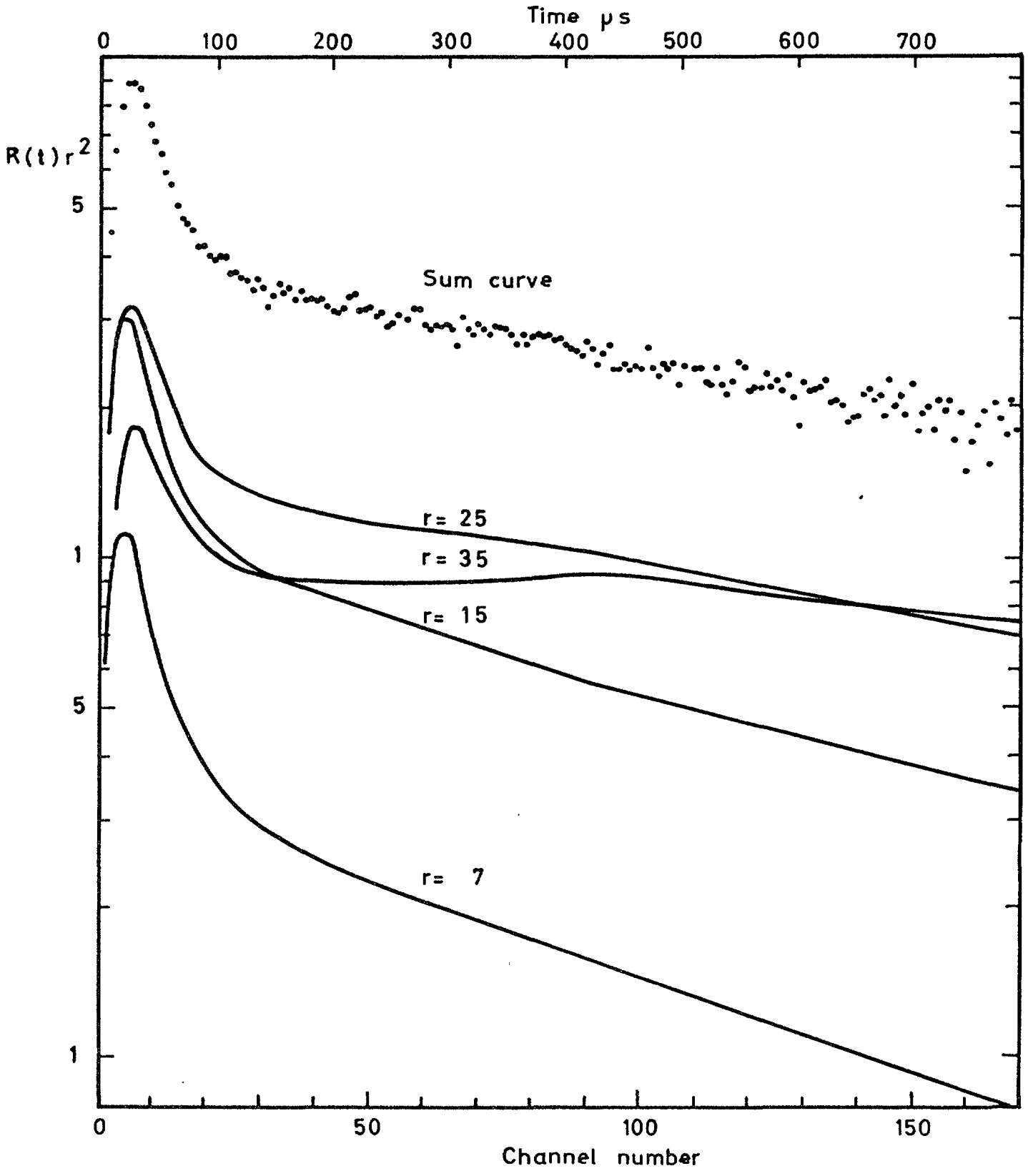

Fig. 3. Reaction rate curves for cadmium, measured with a time resolution of 4.65 μs.

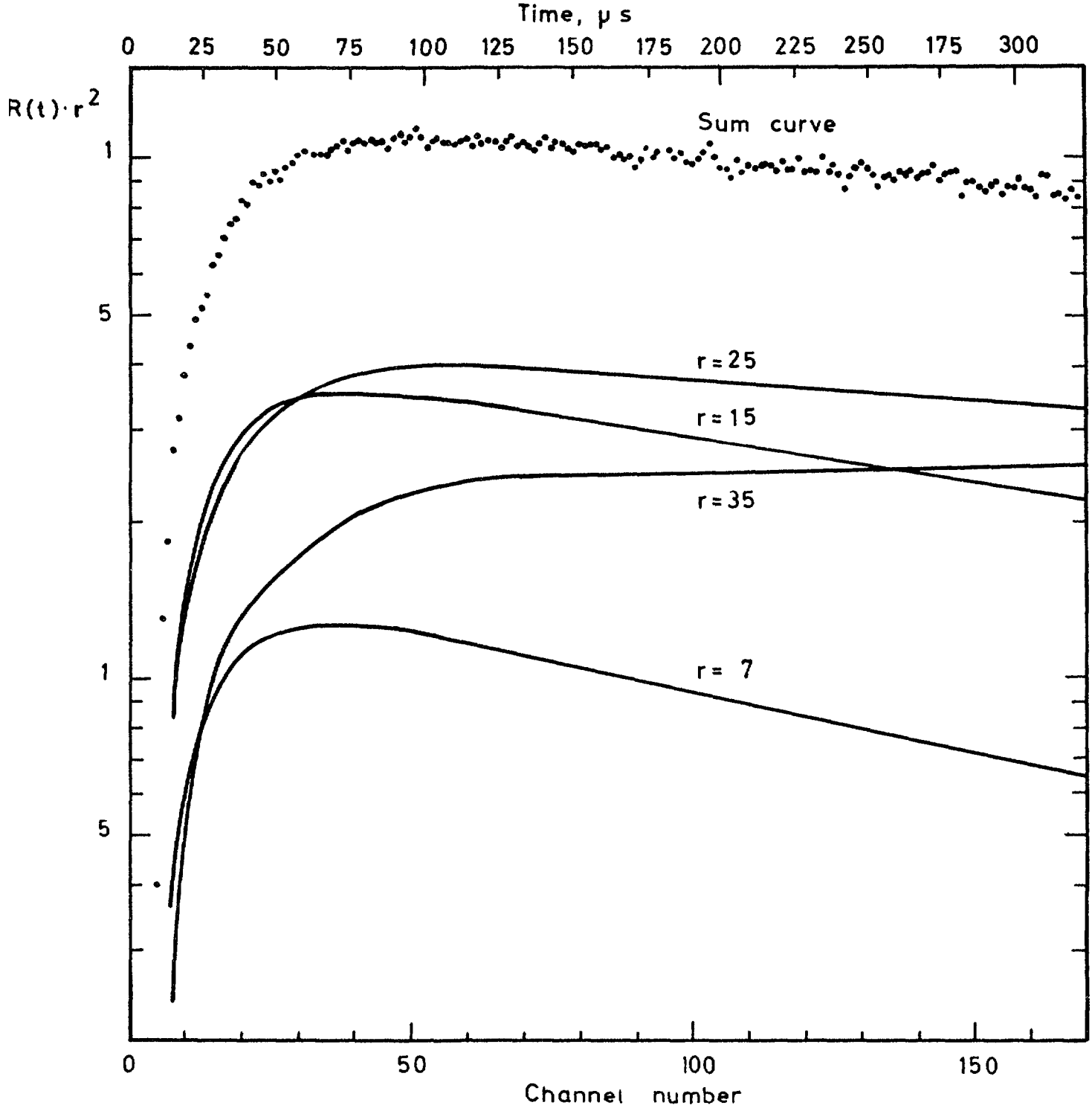

Fig. 4. Reaction rate curves for gadolinium at various positions (full lines) and their sum (upper curve). Time resolution 1.90 μs.

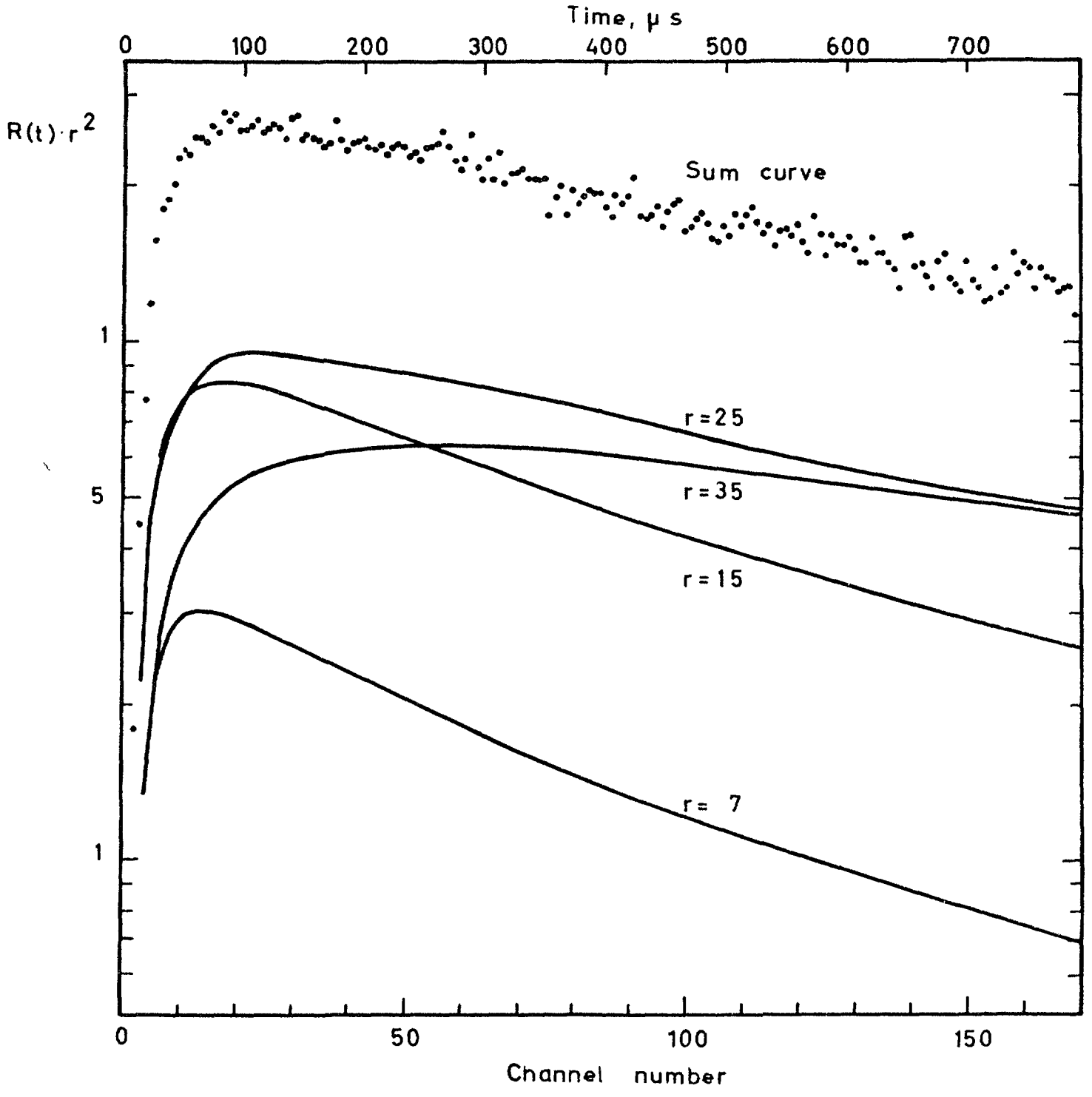

Fig. 5. Reaction rate curves for gadolinium, measured with a time resolution of 4.65 μs.

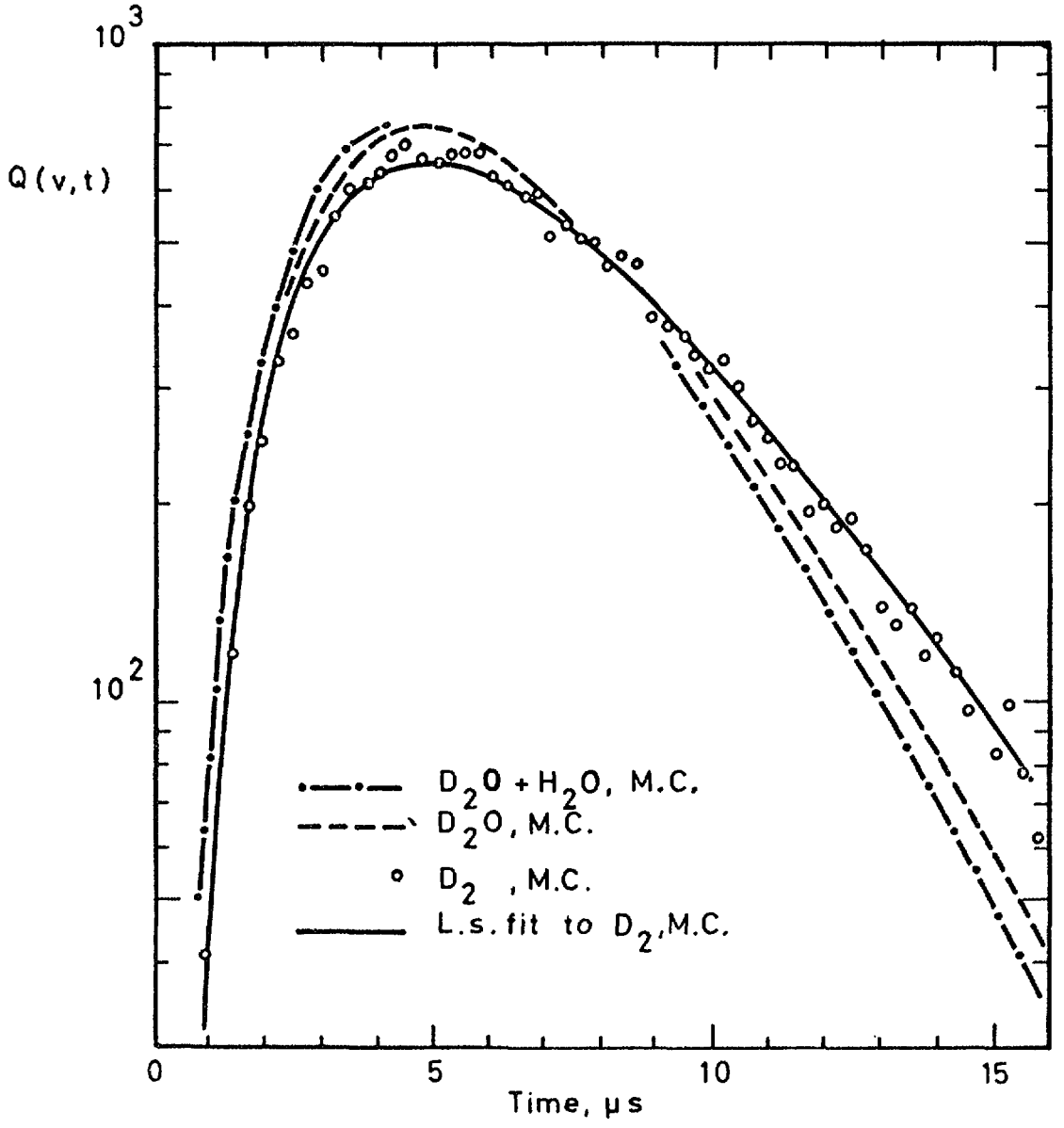

Fig. 6. The time-dependent slowing-down density, calculated by the Monte Carlo method for pure deuterium (circles) and for heavy water with and without 0.5 % light water. The full line curve is the fitted theoretical function derived from von Dardel's density function.

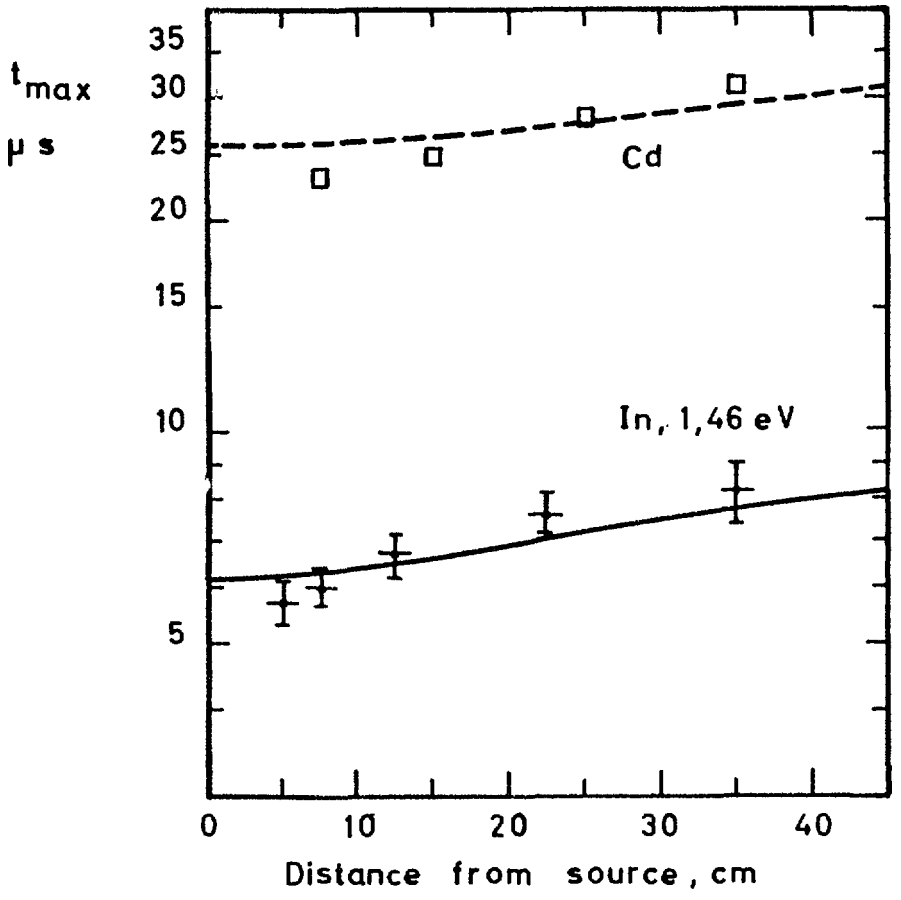

Fig. 7. Experimental values for the maximum of the time-dependent density at various distances and the relation for this dependence given by Claesson.

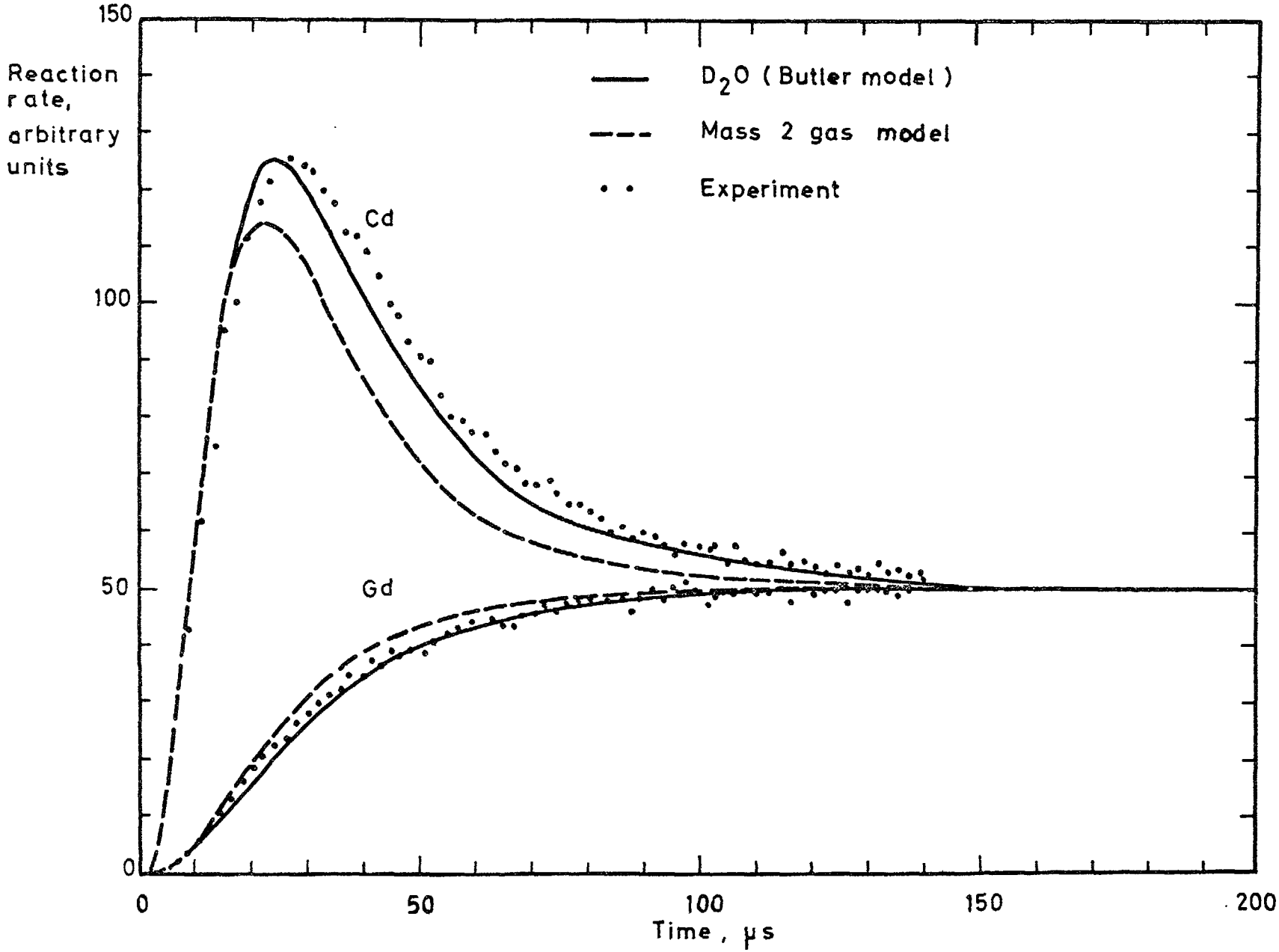

Fig. 8. Calculated reaction rate curves in heavy water for cadmium and gadolinium according to Purohit with the experimental results included.

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