

THE IMPORTANCE OF THERMAL NEUTRON
SCATTERING LAWS IN REACTOR CALCULATIONS

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ANTONY JOHN HUDSON GODDARD, B.Sc.

Nuclear Power Section
Department of Mechanical Engineering
Imperial College of Science and Technology
London, S.W.7.

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ABSTRACT

Multigroup transport theory methods have been used in a study of the importance of coolant and moderator thermal neutron scattering laws in determining neutron spectra, flux peaking and reactivity in cold, clean Steam Generating Heavy Water (SGHW) reactor lattices, at two different coolant densities. It is shown that the reactivity effects of scattering laws are just significant for design purposes.

Spectral density distributions of the normal modes of hydrogen motion in Santowax-R (terphenyl) have been fitted to the phased-chopper measurements of Brugger. Spectral density distributions for solid and liquid forms are compared. Calculated total cross sections and diffusion lengths are compared with available experimental data.

A study of the ability of various scattering models for hydrogen in light water to predict neutron diffusion and spectrum distortion in aqueous solutions of boron, cadmium and gadolinium salts has been carried out. It is shown that the Haywood model differs from experiment to the same extent as does the earlier Nelkin model, though in the opposite sense.

The fitted model for Santowax-R and the Haywood model have been combined in a transport theory analysis of the 'two-temperature' experiment of Tremmer. Such an experiment is relevant to the physics of an SGHW with hot coolant. Predicted spectrum transients agree with experiment within the random and systematic errors of the latter. Evidence for the separability in space and energy of the spectrum transient in Santowax-R has been examined without conclusive result.

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CHAPTER 1

INTRODUCTION

During the last ten years, the emphasis of reactor physics work on the 'point' or cell calculation of fluxes and reactivity has been much influenced by the availability of large, fast, computers. The computer has allowed the abandonment of methods based upon simple correlations of the results of many expensive experiments in favour of methods which use basic nuclear data for cell materials and an accurate representation of the cell geometry. Increasing computing speed has also permitted improved treatment of neutron behaviour in energy and space by the use of many energy groups together with a numerical solution of the Boltzmann transport equation. Methods based upon the transport equation will soon be used for routine reactor design calculations.

Now the nuclear data cannot be better than that currently available and the simplest approximations to the transport equation, consistent with acceptable accuracy, will be chosen in order to economise on computer running time and to accommodate the calculation within computer storage. Thus the role of experiment lies in allowing checks to be made of the adequacy of data, cell representation and numerical approximations. Such experiments usually include the main features of the reactor lattice in question. Because the physical processes are well understood, the theoretical methods may be applied to a variety of lattices of the same general type without the need for further experimental studies.

This work has been concerned with one particular data problem; the importance of a correct treatment of inelastic scattering of neutrons by certain moderator atoms with particular reference to the physics design of the Steam Generating Heavy Water Reactor (SGHW). Thermal events assume particular importance in this reactor because while the

light water coolant passes around the clustered fuel rods, the heavy water which has the major moderating function is at near to ambient temperature. While there will be some interaction with the overall flux shape in the cell, the thermal spectrum in the fuel will be influenced by two main factors. These are the absorption induced hardening of the spectrum due primarily (in a 'clean' unburnt core) to U-235 absorption and, secondly, the extent to which neutrons at a different 'temperature' from the moderator penetrate the fuel cluster, thereby influencing the reactivity. In part because no hot coolant experimental results were available when the work was begun and partly in order to isolate the processes involved, these two factors have been studied separately.

The effect of using realistic models for molecular binding of hydrogen in light water and deuterium in heavy water, upon reactivity and reaction rates in cold, clean, air cooled (to simulate steam) and water cooled lattices is discussed in Chapter 2. Experiments performed by other workers at Imperial College have investigated the rethermalisation process, which is the dominant process affecting the penetration of 'cold' neutrons into the hot fuel cluster containing coolant. This process is discussed in detail in Chapter 5. Chapters 3 and 4 are concerned with the models for molecular binding and their effect upon scattering, for the light atoms in the two liquids used in the Imperial College experiments. These liquids were hot Santowax-R (terphenyl) and cold light water. Obviously the discussion of light water properties is also relevant in itself to SGHW physics design.

The cold, clean SGHW lattice considered was one used in the Winfrith series of experiments; it had fuel:coolant and fuel:moderator volume ratios close to those of the prototype. It is not sufficient to consider the effect of various scattering models for hydrogen in water only. In water cooled cores, due to neutron in-leakage into

the fuel cluster, the treatment of deuterium in heavy water must be studied and in a 'superheat' steam cooled cell the heavy water is the only moderator. Fission and other reaction rates in the fuel are considered together with the cell flux peaking and reactivity.

Santowax-R is an eminently suitable material for rethermalisation experiments for reasons similar to those that led to it being considered for use in organic cooled reactors. However, thermal scattering data suitable for input to the multigroup calculations has been lacking. In contrast, benzene ('mono-phenyl') has been studied both experimentally and theoretically by other workers and detailed calculations of spectra in infinite, poisoned solutions have been carried out. Diphenyl has been studied to a lesser extent, mainly through experimental work using the pulsed neutron technique. There is, however, little reason to rely on data for benzene and diphenyl in calculating scattering by Santowax-R, which has markedly different physical properties. In Chapter 3 the origins of a new model for the molecular binding of hydrogen in Santowax-R and the testing of this model against available experimental data are described.

The author has been closely associated with the analysis of a series of precise measurements of neutron diffusion lengths in light water poisoned with neutron absorbing elements. Although thermal neutron scattering by water has been studied for many years, there remain significant discrepancies between experimental measurements of such quantities as total cross section and diffusion lengths and the predictions of the present best model derived from double differential scattering measurements. Four different scattering models, one the present best model and another the most realistic model used in the SGHW analysis in Chapter 2, have been compared with the diffusion length measurements.

To isolate the rethermalisation process and to avoid the geometrical complexities of an SGHW reactor cell, the most satisfactory solution is the two-temperature problem, where uniform media at different temperatures are separated by a temperature discontinuity, usually plane. This problem has been considered theoretically by numerous workers, but experimental work has been limited. Severe difficulties face the experimentalist, particularly if the moderators concerned contain eight elements. In Chapter 5, the results of Imperial College rethermalisation experiments are compared with the same methods of calculation as were used in the calculations for the SGHW cell, but incorporating the best available scattering models for light water and Santowax-R. Important features of the rethermalisation process are discussed.

CHAPTER 2

SCATTERING LAW EFFECTS
IN COLD CLEAN SGHW LATTICES

Particular problems arise in the calculation of the thermal neutron spectrum in an SGHW cell because of the presence of two different, both strongly moderating, liquids, heavy and light water. Although the thermal neutron spectrum to which the fuel is exposed will be determined mainly by scattering by H in H_2O (together with the effects of absorption), this spectrum will also be influenced by the in-leakage of neutrons having a different energy distribution from the D_2O moderator. Thus the effect of changing both the H and D scattering laws must be investigated.

Previous work in this field has been concerned with uniform lattices having a single moderator. Macdougall (1963) has compared reactivity, temperature coefficient and fission rate calculations using different graphite scattering models, for Calder Hall, AGR and homogeneous plutonium/graphite ('Zenith') lattices. Smit and Stamm'ler (1965) have compared calculated reaction rates with measurements in UO_2 lattices moderated by pure H_2O and by an H_2O/D_2O mixture. Recently, Volpe (1967) has published an extensive comparison between reaction rate measurements in low-enrichment uranium H_2O moderated lattices and calculations which employed three different H scattering laws.

This chapter describes work carried out during 1963-65 at AEE-Winfrith and Imperial College, using computer programmes at Winfrith. In Section 2.2 the multigroup method used for calculating reactivity, fission and reaction rates is described and errors in the representation of cross sections and fluxes are discussed. Three different scattering laws for D in D_2O and H in H_2O have been used and these models are described in Section 2.3. Calculated

reaction and fission rate ratios are compared with experiment in Section 2.5 and the possible interactions between fine structure and fuel spectrum in Section 2.6. The effect upon k -infinity of changing moderator scattering laws is discussed in 2.7.

The majority of the results given in this Chapter have appeared in a Winfrith report (Goddard, 1966).

2.1 The SG 13 Lattice

The SGHW lattice, which is the subject of this study, is highly inhomogeneous, the cell and fuel cluster dimensions being large compared with the transport mean-free paths of thermal neutrons. Figure 1 shows the arrangement of the cell; this arrangement was called SG 13 in the series of Winfrith experiments described by Campbell et al (1964). Seventy-one uranium oxide rods 0.4 inches in diameter, clad in aluminium form a fuel cluster enclosed by the aluminium pressure tube containing the coolant. Fuel enrichment was 1.35% and the volume ratios coolant/fuel and moderator/fuel were close to those for the SGHW prototype. The "coolant" between the fuel rods was either full density room temperature H_2O or air (simulating steam) in the two lattices considered here. The D_2O moderator was also at room temperature and an air gap separated the pressure tube from the aluminium calandria tube.

2.2 Cell Calculation - The Programme THULE

Calculations for the SGHW lattice geometry shown in Figure 1 have been carried out with the THULE programme. THULE uses basic cross-sections in a transport calculation with the Winfrith DSN programme (Askew and Brissenden (1963) and Francescon (1963)) and computes k -infinity and multi-group fluxes in the reactor cell. PIXSE, written by Macdougall (1963) computes inter-group transfer cross-sections from an arbitrary scattering law $S(\alpha, \beta)$; the

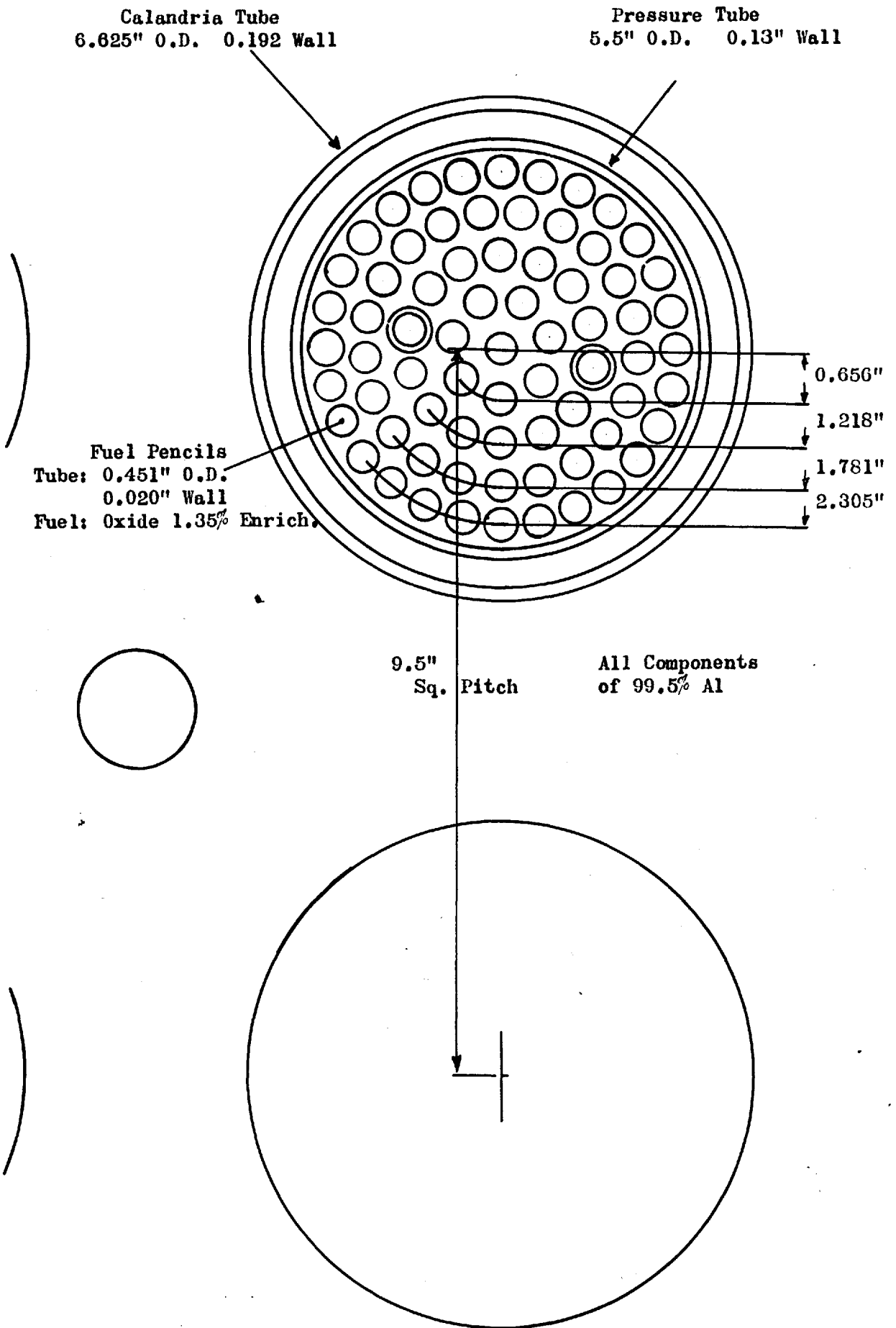


Figure 1 SG13 Lattice Cell

free gas model and the so-called effective width model are built into PIXSE. Descriptions of the physical basis and the detailed working of THULE have been published by Leslie and Terry (1963) and by Terry (1964). Two fast groups and one resonance group cover the energy ranges 10 MeV to 821 KeV; 821 KeV to 5.53 KeV and 5.53 KeV to 4 eV, while 42 interacting and non-overlapping groups cover the thermal range to 4 eV. Absorption cross-sections are taken from the Winfrith Data Library.

To convert the fuel clusters into cylindrical geometry THULE uses hyperfine structure factors for fuel, cladding and coolant that are calculated by one group collision probability methods in slab geometry for each ring of fuel separately, assuming this ring to be part of an infinite array. When subsequently calculating reaction rates in fuel rods, these factors are applied to the total thermal activity. This approach, which simplifies the actual calculation geometry, has been shown by Campbell et al to under-estimate the average hyperfine structure in an H₂O cooled cluster by 2 - 3%. In addition, thermal spectrum is calculated in a hyperfine weighted fuel/can/coolant smear and so spectrum hardening inside the (small) fuel rods is not taken into account.

Neutrons entering the thermal region from the resonance group are partitioned among the 42 thermal groups using cross-sections calculated by PIXSE, assuming a $\frac{1}{E}$ flux above 4 eV. The importance of a proper source distribution across the cell was shown in early studies by Leslie and Terry with THULE when it was found that the use of a correctly shaped thermal source instead of a flat source increased the flux in the calandria and pressure tubes of an SGHW cell relative to the minimum flux in the fuel by about 10%. A predicted spatial source distribution was therefore used for the present calculations.

The group structure of 42 groups was designed by Chudley (1963) for systems containing plutonium 239 and 240 resonances. The main reasons for the 4 eV boundary are that the plutonium 240 resonances is thereby included in the detailed thermal calculation and that measured spectra in homogeneous media show that molecular binding effects are significant up to energies of a few eV. Average absorption and fission cross-sections for these narrow groups are obtained by assuming a Maxwellian spectrum at the appropriate physical temperature for energies less than 4 kT and a $\frac{1}{E}$ spectrum for greater energies. To test this combination of group structure and cross-sections, average cross-sections below 0.625 eV calculated using the free gas model for hydrogen scattering with 42 groups and also with a more detailed 55 group set, were compared with exact free gas calculations using SOFOCATE (Amster and Saurez (1957)) programme calculations with the same Winfrith Data Library absorption cross-sections. The two group structures are compared in Figure 2 and Table I compares the reaction ratios Lutecium 176/Manganese 55 and the fission rate ratios Plutonium 239/Uranium 235 (each normalised to unity in a Maxwellian spectrum) for various mixtures of $\frac{1}{V}$ absorber with hydrogen.

Table I Effect of a More Detailed Group Structure
on Spectrum Indicators

$1/v$ barns/ H atom	Lu/Mn reaction rate ratio			239/235 fission rate ratio		
	SOFOCATE	42 group	55 group	SOFOCATE	42 group	55 group
4	1.291	1.278	1.294	1.358	1.357	1.357
8	1.480	1.454	1.481	1.653	1.650	1.652
12	1.605	1.570	1.605	1.900	1.895	1.898

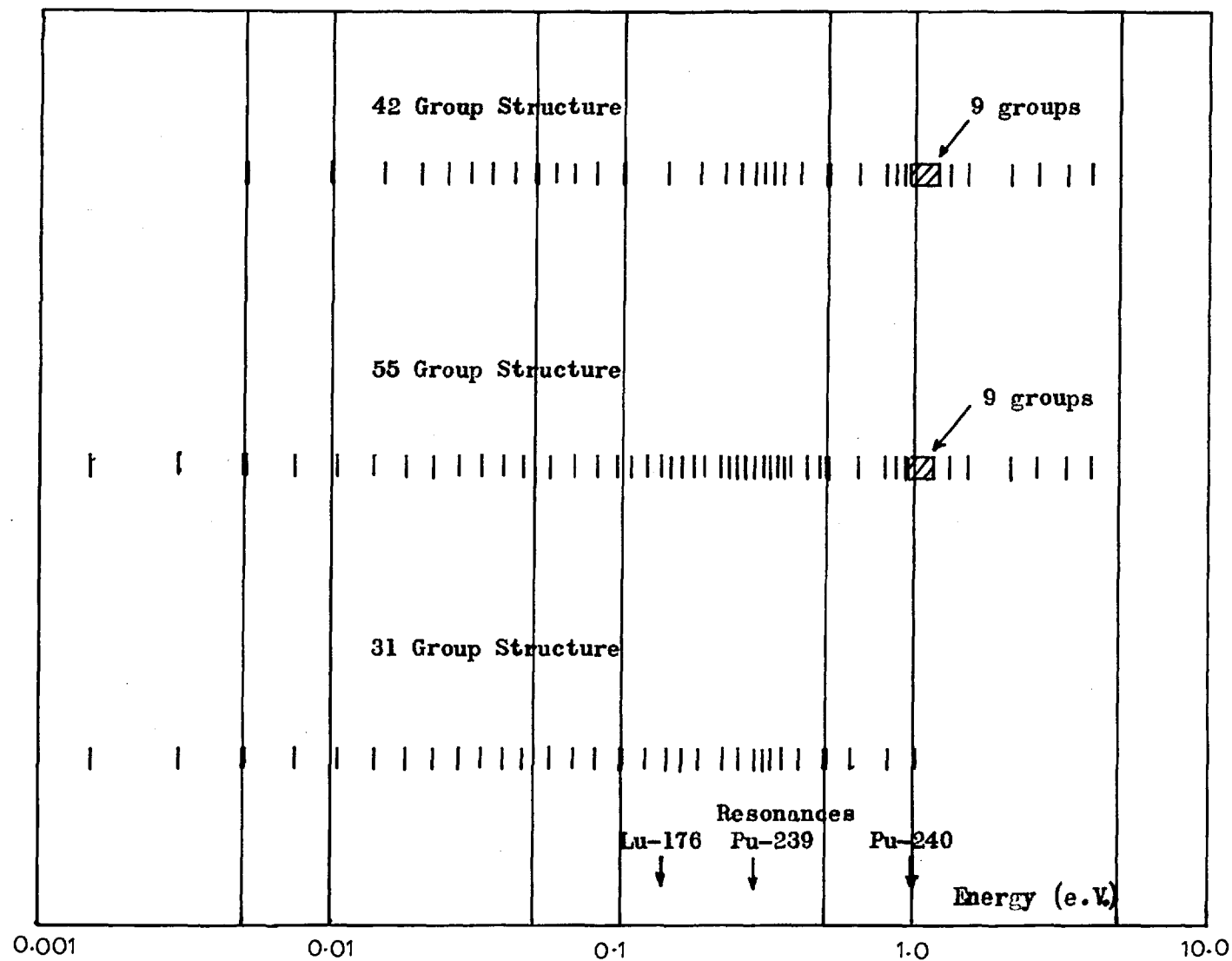


Figure 2 The Group Boundaries Used in the 45, 55 and 31 Group Calculations

Only for lutecium is the 42 group set inadequate; this is not unexpected as the group structure was not chosen with the lutecium resonance at 0.142 eV in mind. The SGHW spectra to be discussed are equivalent in "hardness" to 4 barns of $\frac{1}{V}$ absorber per H atom or less and so errors of up to 1% in calculated lutecium manganese ratios must be expected, and the necessary corrections have been made.

The version of the WDSN code used was restricted to isotropic scattering and in order to take some account of the anisotropy in the scattering by hydrogen and deuterium, the transport correction can be applied in PIXSE, whereby the total anisotropic scattering cross-section $\sigma_1(q)$ is subtracted from the diagonal (elastic) terms of the isotropic scattering matrix. This preserves the correct diffusion coefficient while leaving the spectrum as calculated in an infinite homogeneous medium unchanged. There is no direct evidence for the adequacy of the transport correction in SGHW geometry. The comparisons between measured and calculated flux peaking in section 2.6 suggest that the errors involved are small.

2.3 Scattering Models for H₂O and D₂O

The programme PIXSE requires the differential scattering cross-section as input and using the nomenclature of Egelstaff and Schofield (1961, 1962) this is given by

$$\sigma(E_0, E, \mu) = \frac{1}{kT} \frac{\sigma_b}{4\pi} \sqrt{\frac{E}{E_0}} e^{-\beta/2} S(\alpha, \beta) \quad (1)$$

where

$$\mu = \cos \Theta$$

$$\Theta = \text{the angle of scatter}$$

$$E_0, E \text{ are the initial and final energies}$$

$$\sigma_b = \text{the bound atom cross-section} = \frac{(A+1)^2}{A} \sigma_{\text{free}}$$

$$\alpha = \frac{E + E_0 - 2(EE_0)^{1/2}\mu}{AkT}$$

α is dimensionless, and is proportional to the square of the momentum transfer

$$\beta = \frac{E - E_0}{kT} \text{ is the dimensionless energy change}$$

$S(\alpha, \beta)$, the 'scattering law' can be thought of as the probability that if the neutron energy changes by β , then momentum α will be exchanged between neutron and the scattering atom.

Depending on the accuracy required in calculating spectra and neutron transport, theoretical models of molecular binding are commonly used instead of the more complex model based upon experiment. The three scattering models for H_2O and D_2O that have been used in this work are as follows; the first being the simplest, the last the most realistic:

(1) The free gas model

The free gas model neglects molecular binding and assumes the scattering atoms to be free; cross-sections for an ideal, monatomic gas were first calculated by Wigner and Wilkins (1944). In the scattering law representation, the spectral density distribution or density of the normal modes of vibration for a free gas is a δ function at $\beta = 0$ (Figure 3) and

$$S(\alpha, \beta) = \frac{1}{2\sqrt{\pi}\alpha} \exp\left(\frac{-(\alpha^2 + \beta^2)}{4\alpha^2}\right) \quad (2)$$

(ii) The width model

The width model is completely defined by a single parameter, the "width", which is adjusted so that some reactor physics constant, usually the neutron diffusion coefficient in the particular moderator, is predicted correctly. It was suggested by Egelstaff that the spectral density distribution $e(\beta)$ describing molecular diffusion might be a useful simple representation of the entire scattering law for H_2O and D_2O . This is not to imply that the scattering law is mostly diffusive in origin, but that a reasonable empirical fit to

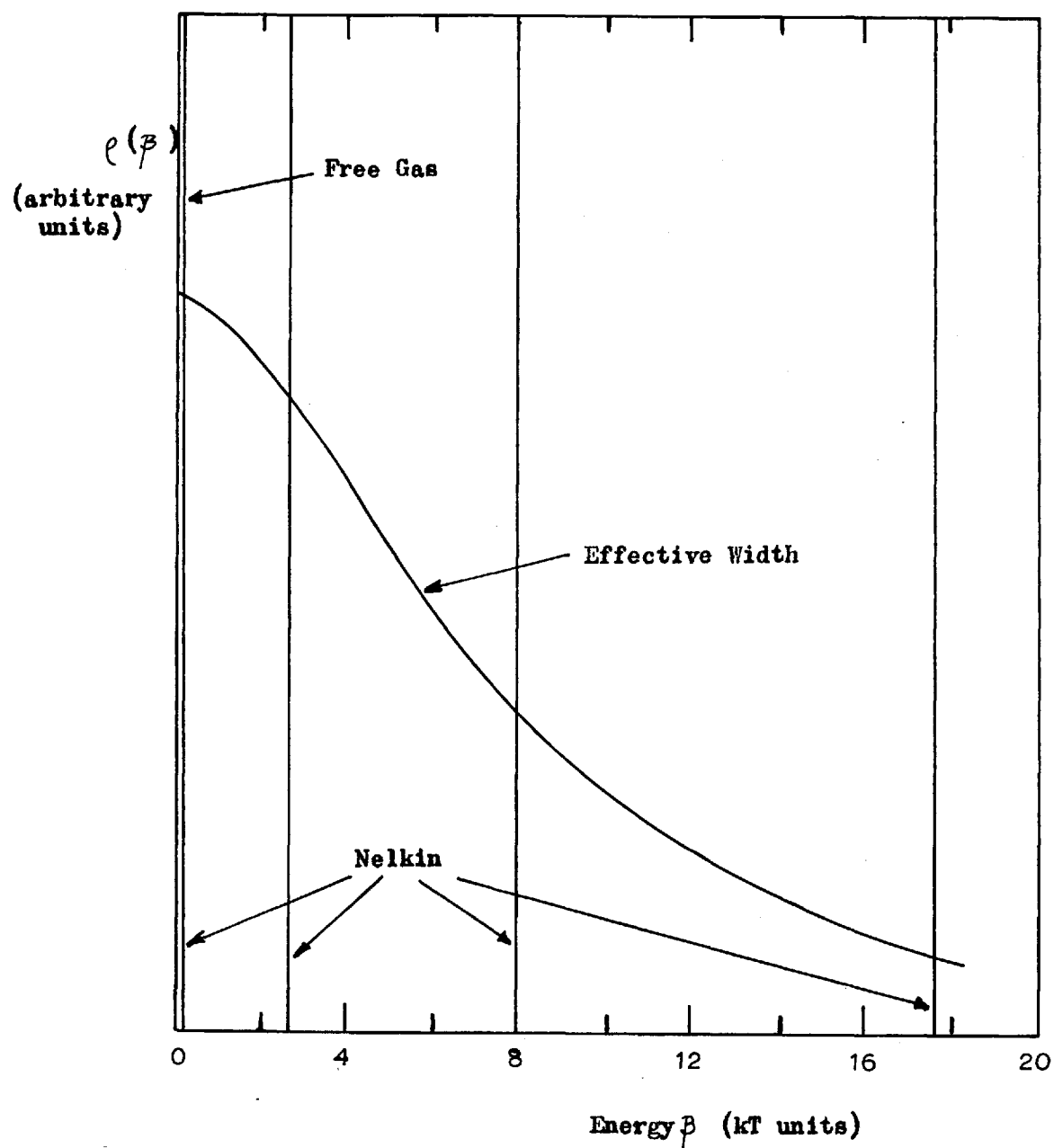


Figure 3 The Form of $\rho(\beta)$ for Free Gas, Effective Width and Nelkin Models of H-Atom Motion

the scattering behaviour might be obtained. Egelstaff and Schofield have suggested that $\varrho(\beta)$ for a single diffusion atom can be represented by

$$\varrho(\beta) = \frac{2 \sinh(\beta/2)(q^2 + 4)^{1/2}}{\pi q^2} K_1\left(\frac{(q^2 + 4)^{1/2}}{2q}\right) \quad (3)$$

where q is the width parameter, which is proportional to the width of the quasi-diffusive peak in $\varrho(\beta)$ (Figure 3). From this expression, $S(\alpha, \beta)$ can be obtained analytically. The width parameter can be fixed in a variety of ways; by fitting to scattering law data; to the mean kinetic energy of the atom $\bar{T}$, to the diffusion coefficient, or to the total cross-section. The first method is unlikely to be successful with such an empirical model. The other three are all interdependent and the diffusion coefficient was selected as the most convenient. At an early stage of this work, in 1963, q values of 4.3 and 2.7 for room temperature H_2O and D_2O were selected.

(iii) The Nelkin-type model for H_2O and D_2O

The Nelkin (1960) model for H_2O has been extensively used in spectrum calculations and in the calculation of diffusion coefficients. Translations of the molecule as a whole, hindered rotations and the vibrational modes are represented by δ -functions in $\varrho(\beta)$ as shown in Figure 3. For D_2O , Honeck (1962) has proposed an equivalent model.

Existing methods for producing cross-sections use approximations for certain of the modes, depending on the incident neutron energy, which results in discontinuities in the cross-section $\sigma(E_0 \rightarrow E)$ at the energy where the approximation changes. This is avoided in the method proposed in this report which treats the four modes exactly at all incident neutron energies up to 4 eV. It is based on the programme ADDELT written by McLatchie (1965) to convolve

the experimentally measured part of a scattering law with δ functions in $\rho(\beta)$ outside the range of measurement. The vibrations are assumed to be isotropic, and the Gaussian approximation on which the method depends implies that the vibration should also be harmonic. The theory of ADDELT is outlined in Appendix A.

In constructing the scattering model with the Nelkin parameters for H_2O , the input scattering law was that for a free gas model with mass 18 representing the translational mode at $\beta = 0$. The mass scale was increased by the factor of 18 by dividing the α scale of a scattering law derived from equation 1 by 18. ADDELT was then used to convolve successfully with this, the three δ functions in $\rho(\beta)$ corresponding to rotation and vibrations. The model for D_2O was formed in a similar way, starting from a free gas of mass 20. Honeck has shown that with this D_2O model, the effect of coherent scattering can be neglected without introducing serious error over the thermal range important in reactor physics.

2.4 Diffusion Coefficients Calculated by PIXSE

PIXSE will calculate the Maxwellian weighted transport cross-section and so permit the calculation of the diffusion coefficient for H_2O and D_2O for the various models.

This has been done in order to judge later the importance of any interaction between neutron diffusion and the spectrum in an SGHW cell, and the results are shown in Table II. The calculated diffusion coefficient for H_2O using the Nelkin model is, in fact, 2% lower than that given by the calculations of Clendenin (1964). However, the possible presence of small systematic errors in calculated diffusion coefficients should not invalidate the general conclusions. This discrepancy is tentatively ascribed to an inadequate angular and energy integration of equation 1 by PIXSE.

Table II Calculated H₂O and D₂O Diffusion Coefficients

H ₂ O diffusion coefficient		D ₂ O diffusion coefficient	
Scattering law model	PIXSE D (cm ⁻¹)	Scattering law model	PIXSE D (cm ⁻¹)
Free gas (isotropic matrix)	0.164	Free gas (isotropic matrix)	0.840
Width ($\frac{1}{q} = 4.3$)	0.147	Width ($\frac{1}{q} = 2.7$)	0.848
Nelkin	0.145	Nelkin-type model as proposed by Honeck	0.823
Experiment	0.143 $\pm$.001	Experiment	0.834 $\pm$.006

The experimental values quoted are unweighted means and mean deviations of two compilations by Springer (1964). Each of the three models for D₂O gives diffusion coefficients close to the experimental value, although agreement with the free gas model was only achieved by using a scattering matrix uncorrected for anisotropy. Making the correction for anisotropy to the free gas model cross-sections would result in diffusion coefficients of 0.26 cm⁻¹ and 0.97 cm⁻¹ for H₂O and D₂O respectively. The rather high value given by the width model for H₂O was the result of the width parameter being fitted at an early stage in the work when there were large uncertainties in the experimental value of the diffusion coefficient.

2.5 Dependence of Lattice Spectrum upon the H₂O and D₂O

Scattering Laws.

Two different integral spectrum measurements with foils have been made in the SGHW lattice cells used as examples in this chapter. These are the ratio of the activation of Lu-176 to that of Mn-55, and the ratio of the fission rate of Pu-239 to that of U-235, both ratios being normalized to unity in a Maxwellian spectrum. Foils

were placed in representative fuel rods in each fuel ring, and at the cell boundary in the D_2O . The experimental techniques have been described by Fox (1964).

THULE will calculate a chosen reaction rate or reaction rate ratio as a function of position in the cell. These are converted to fuel rod values by applying the hyper-fine weighting factors to the thermal activity and adding the resonance contributions. Small corrections have been applied to the Lu/Mn ratios to take account of the under-estimate of Lu activity due to the group structure (Section 2.2) and to correct for the fact that the Mn resonance at 337 eV lies above the main uranium resonances and is not therefore affected by the depression of the $\frac{1}{E}$ flux caused by these resonances.

(i) "Water-cooled" Lattice

Tables III and IV show calculated Lu/Mn and Pu/U ratios in the H_2O cooled lattice compared with experiment. Significant improvement is obtained by using the width model rather than the free gas model for H_2O . Changing the D_2O scattering model only affects the outer two rod rings as might be expected since thermalization in the interior of the cluster is dominated by the H_2O "coolant"; these outer rings, however, do have the highest flux weighting and contain the greatest number of rods. The use of the Nelkin-type models for H_2O and D_2O , rather than width models, does not produce a large change in spectrum. There remains a small discrepancy between theory and experiment, the experimental spectra being harder, although this difference is only greater than the experimental errors at the centre of the cluster. A similar difference has been observed in the analysis of the whole series of experimental SGHW lattices (Campbell et al (1964)) using the width model for H_2O . The discrepancy is consistent with the results of the analysis (Poole (1965)) of spectra in homogeneous

poisoned aqueous solutions, but the extensive analysis of regular lattices by Volpe (1967) is inconclusive on this point, the Nelkin model even being superior to more elaborate models for well-moderated lattices.

(ii) "Air-cooled" Lattice

The Lu/Mn and Pu/U ratios calculated for the "air cooled" lattice and given in Tables V and VI, show that the D_2O scattering model has in this case a large effect upon the fuel spectrum. The effect on the spectrum of changing from the width to the Nelkin-type model is small compared with that produced by using the width model instead of the free gas model. No Pu/U experimental results are available, but inside the fuel cluster the calculated Lu/Mn ratios, using the Nelkin-type model, agree with experiment within the experimental errors. The spatial variation of the spectrum within the fuel cluster also appears to be well represented. The excellent performance of the Nelkin-type model for D_2O , without correction for coherent scattering, for predicting spectral distortion, has been confirmed by Sinclair (1965) who has analysed boron poisoned D_2O solutions.

Table III "Water-cooled" Lattice. Lu/Mn Reaction Rate Ratios

Scattering models		Fuel rod rings					D_2O (Cell boundary)
H_2O	D_2O	1 (Centre rod)	2	3	4	5	
Free gas	Free gas	1.088	1.099	1.110	1.107	1.092	1.056
Width	Free gas	1.127	1.142	1.156	1.151	1.114	1.051
Width	Width	1.127	1.143	1.156	1.157	1.128	1.083
Nelkin	Nelkin	1.125	1.142	1.158	1.158	1.131	1.088
Experiment		1.152 ±.022	1.159 ±.022	1.169 ±.022	1.162 ±.022	1.142 ±.022	1.060 ±.020

Table IV "Water cooled" Lattice. Pu-239/U-235 Fission Rate Ratios

Scattering models		Fuel rod rings					D ₂ O (Cell boundary)
H ₂ O "coolant"	D ₂ O moderator	(1 centre rod)	2	3	4	5	
Free gas	Free gas	1.206	1.214	1.214	1.195	1.157	1.115
Width	Free gas	1.235	1.247	1.250	1.226	1.171	1.114
Width	Width	1.234	1.247	1.251	1.230	1.182	1.136
Nelkin	Nelkin	1.237	1.250	1.255	1.233	1.184	1.139
Experiment		1.299 ±.04	Not Measured	1.263 ±.037	1.233 ±.037	1.201 ±.036	1.160 ±.040

Table V "Air cooled" Lattice. Lu/Mn Reaction Rate Ratios

Scattering model D ₂ O	Fuel rod rings					D ₂ O (cell boundary)
	(1 centre rod)	2	3	4	5	
Free gas	1.268	1.265	1.254	1.230	1.181	1.075
Width	1.322	1.320	1.308	1.285	1.238	1.118
Nelkin	1.322	1.320	1.308	1.286	1.239	1.118
Experiment	1.318 ±.025	1.306 ±.025	1.296 ±.025	1.266 ±.024	1.240 ±.024	1.140 ±.022

Table VI "Air cooled" Lattice. Pu-239/U-235 Fission Rate Ratios

Scattering model D ₂ O	Fuel rod rings					D ₂ O (cell boundary)
	(1 centre rod)	2	3	4	5	
Free gas	1.428	1.423	1.403	1.367	1.308	1.189
Width	1.482	1.482	1.461	1.424	1.362	1.226
Nelkin	1.496	1.491	1.470	1.432	1.369	1.231

2.6 Neutron Transport and its Effect Upon Spectra, Flux Peaking and Reactivity

Changing the diffusion coefficients of the moderators in an SGHW cell will affect the flux peaking which may interact with the cell spectrum and both the spectrum and flux peaking changes will influence the reactivity.

(i) Adjustment of the Diagonal Elements of H and D

Scattering Models

An attempt has been made to vary diffusion coefficients while leaving the cell spectra substantially unchanged, by adjusting the diagonal (elastic) terms in the hydrogen and deuterium scattering matrices. Three alternative diffusion coefficients for H_2O and D_2O have been used, and were obtained by adjusting the scattering matrices for the width model between the extremes of an isotropic matrix and a matrix with the transport correction applied. Values of these diffusion coefficients listed in Table VII below can be compared with the values implied by the various scattering laws as given in Table II.

Table VII Effect of Transport Correction on the
Calculation of Diffusion Coefficients

	H_2O PIXSE D (cm^{-1}) (Width model $q = 3.7$)	D_2O PIXSE D (cm^{-1}) (Width model $q = 2.7$)
Isotropic matrix	0.111	0.737
Intermediate matrix	0.129	0.788
Anisotropic matrix	0.158	0.848

Different combinations of these alternative diffusion coefficients have been used in THULE calculations of the reaction rate of manganese as a function of position in the "water cooled" cell. The reaction

rate in a fuel rod is obtained by applying the hyperfine factor to the thermal activity and adding the resonance activity. Table VIII shows flux ratios in the cell, where ϕ represents manganese activity, and where 'PTCT' denotes the average value in the pressure and calandria tubes. Also shown are the variation of k-inf and the Pu-239/U-235 fission rate ratio in the outer ring of rods; changes of this ratio for the inner rod positions were negligible.

Table VIII "Water Cooled" Lattice. Varying D_{H_2O} and D_{D_2O}

$D_{H_2O}(\text{cm}^{-1})$	$D_{D_2O}(\text{cm}^{-1})$	$\frac{\bar{\phi}_{\text{PTCT}}}{\bar{\phi}_{\text{fuel}}}$	$\frac{\bar{\phi}_{D_2O}}{\bar{\phi}_{\text{fuel}}}$	$\frac{\text{Pu-239}}{\text{U-235}} \text{f.r.}$ outer ring	k-inf
0.158	0.848	1.707	2.024	1.1820	1.173
0.129	0.848	1.783	2.108	1.1787	1.170
0.111	0.848	1.849	2.178	1.1771	1.168
0.158	0.788	1.711	2.042	1.1815	1.173
0.158	0.737	1.706	2.049	1.1813	1.173

A decrease in D_{H_2O} of approximately 40% produces an increase in the fuel flux peaking of 4% and a 0.4% decrease in k-inf through increased absorption in the aluminium pressure and calandria tubes. Changing D_{D_2O} by 10% produced no significant changes in either flux peaking or reactivity. It is likely that in an air-cooled lattice, flux peaking and reactivity are similarly insensitive to the diffusion coefficient of the D_2O bulk moderator, although this has not been checked. A conclusion to be drawn from Tables VII and VIII is that, when only the elastic scattering is changed, the interaction between flux peaking and the form of the transient in the Pu-239/U-235 fission rate ratio in the fuel can be neglected. The fission ratio changes by only $\frac{1}{2}\%$ for a 40% change in D_{H_2O} .

(ii) The Effect of Different Scattering Models Upon Flux Peaking

In Tables IX and X, experimental results for flux peaking as indicated by manganese activation (x), in the water cooled and air cooled lattices are compared with THULE predictions using the various scattering models. The free gas models do not have the transport correction applied for the reason given in Section 2.4.

Table IX "Water Cooled" Lattice

Variation of Flux Peaking with Scattering Model

Scattering models		$\frac{\bar{\rho}_{PTCT}}{\bar{\rho}_{fuel}}$	$\frac{\bar{\rho}_{D_2O}}{\bar{\rho}_{fuel}}$
H ₂ O "coolant"	D ₂ O "moderator"		
Free Gas	Free Gas	1.718	2.048
Width	Free Gas	1.761	2.093
Width	Width	1.729	2.049
Nelkin	Nelkin	1.741	2.069
Experiment		1.753 $\pm$.020	2.054 $\pm$.026

Table X "Air Cooled" Lattice

Variation of Flux Peaking with Scattering Model

Scattering model D ₂ O	$\frac{\bar{\rho}_{PTCT}}{\bar{\rho}_{fuel}}$	$\frac{\bar{\rho}_{D_2O}}{\bar{\rho}_{fuel}}$
Free Gas	1.502	2.084
Width	1.483	2.054
Nelkin	1.480	2.059
Experiment	1.484 $\pm$.017	2.014 $\pm$.026

The variations in flux peaking are small; this is to be expected in view of the results presented in Tables VII and VIII. When the Nelkin-type models are used for H_2O and D_2O the predictions for both lattices agree with experiment in the main within the experimental errors; it should be remembered, however, that the known $\sim 2\%$ underestimate in the calculated hyperfine structure implies an underestimate of this order in the calculation of $\bar{\nu}$ fuel, in the water cooled lattice. A particularly interesting feature is that while for the water cooled lattice changing the models for H_2O from free gas to width produces the expected increase in peaking, making the equivalent change for D_2O causes a small reduction in peaking. For the air cooled lattice a very similar reduction is observed. This is considered to arise through the smaller rate of exchange of energy for the bound model leading to the fuel cluster being exposed to a harder neutron spectrum and the peaking within the cluster thereby being smaller due to the $1/\nu$ fuel cross section. Thus in their effects on fuel flux peaking, neutron transport effects appear to be dominant within the fuel cluster, while D_2O scattering models are important mainly through their effect upon the spectrum.

2.7 The Reactivity Effect of Spectrum Changes

Even in lattices containing uranium-235 as the only fissile isotope, spectrum changes produce changes in reactivity, which although small, are significant for methods of analysis which aim to predict reactivity to better than 1%. These changes arise principally from the non- $1/\nu$ behaviour of the U-235 thermal cross section. From consideration of the reactivity effects of flux peaking given in Tables VIII and the flux peaking changes in Table IX resulting from the use of different models, it appears that $\sim 0.1\%$ of the reactivity change from gas to Nelkin can be attributed to flux peaking effects. This is the case for the water core; for the 'air cooled' core

peaking effects are likely to be small also, but will probably be positive rather than negative.

Table XI Changes in k-inf with Scattering Model

"Water cooled" Lattice			"Air cooled" lattice	
H ₂ O scattering model	D ₂ O scattering model	k-inf	D ₂ O scattering model	k-inf
Gas	Gas	1.177	Gas	1.217
Width	Gas	1.174	Width	1.211
Width	Width	1.172	Nelkin	1.210
Nelkin	Nelkin	1.171		

Table XI shows the overall reactivity effects of model changes for the two lattices. It appears that there is little incentive to improve on the Nelkin-type models on reactivity grounds, although this might be necessary if a natural uranium fuelled SGHW were contemplated as very accurate estimates of the initial reactivity would then be needed.

CHAPTER 3

SCATTERING LAW FOR SANTOWAX-R

Slow neutron scattering by the polyphenyls has been the subject of experimental and theoretical studies motivated by their possible use in organic cooled reactors. The more restricted aim of the work described in this chapter has been to provide a thermal neutron scattering model for liquid terphenyl, in the form of the Monsanto Chemical Co. product Santowax-R.

This model has been used in the analysis of rethermalization measurements. These measurements involved both Santowax and water (H_2O); the scattering by water is discussed in Chapter 4. Although a scattering model may be obtained by adjusting the results of calculation in an iterative manner to agree with differential neutron scattering measurements, such a model must not be accepted uncritically. Integral tests of the proposed model for Santowax-R have therefore been made by comparing predicted diffusion lengths and total cross sections with the available experimental values. Solid Santowax-R has also been investigated, though in less detail.

The iterative method of fitting a spectral density distribution (the density of normal modes of vibration as a function of the dimensionless energy variable β) to phased chopper velocity selector measurements of neutron scattering was proposed by Egelstaff and Schofield (1962) and was first applied by Haywood (1967) to H_2O . Beckurts and Gläser (1964) have analyzed their results using this method and show that the spectral densities of hydrogen atom motion for benzene and diphenyl differ significantly below 0.03 eV. In view of these differences, independent study of neutron scattering by terphenyl appears to be justified. Recently Ross et al (1967)

have studied scattering by benzene; their results confirm the general form of the results obtained by Beckerts and Gläser but show detailed differences. The hydrogen atom motion in benzene has been calculated by Crawford and Miller (1949) and applied to the calculation of neutron spectra by Boffi et al (1962) and by Young et al (1964). Sandroni and Zerbi (1965) have calculated the vibrational modes of diphenyl and have shown that although the low energy vibrations differ between benzene and diphenyl the energy of the C-H vibrations at 0.38 eV are essentially unchanged. Some reference must be made in this chapter to theory because the experimental results available for the fitting procedure only cover part of the energy range of the molecular vibrations.

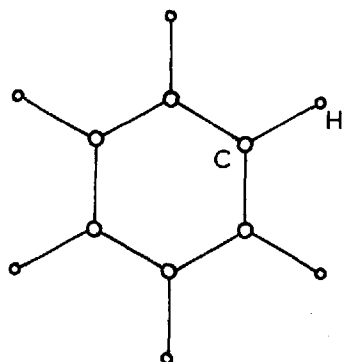
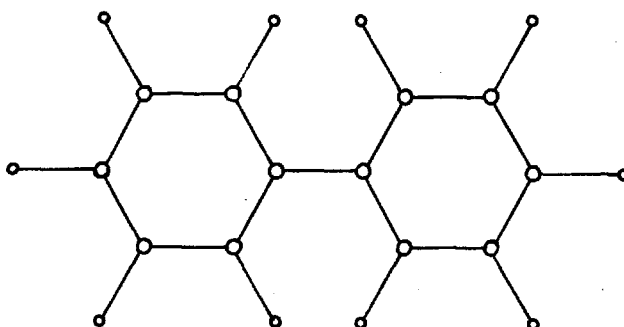
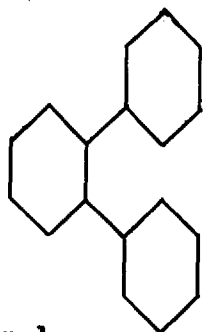
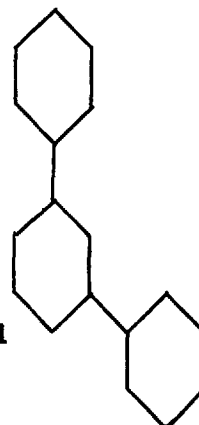
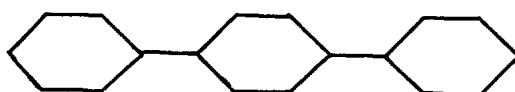
In Section 3.1 the main features and difficulties of the iterative method are outlined and the application in fitting spectral density distributions for Santowax-R to the differential measurements of Brugger (1962) at 23°C (solid) and 207°C and 267°C (liquid) is presented. Constraints on this distribution and the method of extending it outside the range of the available measurements are described in Section 3.2. The resulting spectral densities are compared in Section 3.3 with each other and with the results obtained by Beckerts and Gläser and by Ross et al for benzene and diphenyl. Existing experimental values for the thermal neutron diffusion length in liquid Santowax-R are discussed in Section 3.4 and are compared with the predictions of the proposed scattering model, together with the predictions of two alternative models. The second integral check of the scattering model, a comparison of calculated and experimental total cross section measurements, is described in Section 3.5. A description of the fitted model has been published (Goddard (1967)) and the paper is submitted with this thesis.

3.1 The Iterative Method Applied to Santowax-R

Brugger (1962) has derived the reduced partial differential scattering cross section, the scattering law $S(\alpha, \beta)$, where α is the square of the momentum change $\{E + E_0 - 2(EE_0)^{1/2} \cos \Theta\} / \Lambda kT$ (Λ is the atomic weight of the principal scattering atom; $\Lambda = 1$ has been used throughout this work) and β the energy change $(E - E_0) / kT$, by measuring the slow neutron differential cross section using a phased chopper velocity selector. $S(\alpha, \beta)$ is defined in Section 2.3. The Idaho MTR was the neutron source and the neutron energies incident on the samples were within the range 0.01 eV to 0.1 eV. Solid Santowax was studied at 23°C ($kT = 0.0255$ eV) and the liquid at 207°C and 267°C (0.0414 eV and 0.0467 eV). The velocity selector consisted of two slow neutron choppers 3.4m apart and two rotating collimators to limit fast neutron background. BF_3 detectors were placed at suitable angles 2m from the sample. The energy, time resolution and flux were determined by a beam monitor.

Santowax-R ($C_{18}H_{14}$) is a mixture of ortho, para and meta terphenyls in the approximate ratios 12:55:33. Reference to Figure 4, which illustrates the configuration of diphenyl and the terphenyls, shows that two rings are in a similar situation to the diphenyl rings, while the third ring has only four H atoms. It is probable therefore that the average hydrogen atom motion in the terphenyls resembles that in diphenyl more than diphenyl resembles benzene. In the analysis that follows the assumption is made that scattering by Santowax-R can be characterized by a single H atom spectral density. Brugger has studied scattering by the individual terphenyls without observing significant differences and the use of such an average is therefore justifiable.

Figure 5 shows a selection from the data of Brugger converted to $S(\alpha, \beta) / \alpha$ and plotted semi-logarithmically as a function of α .

Benzene**Diphenyl****Orthoterphenyl****Metaterphenyl****Paraterphenyl****Figure 4 The Polyphenyls**

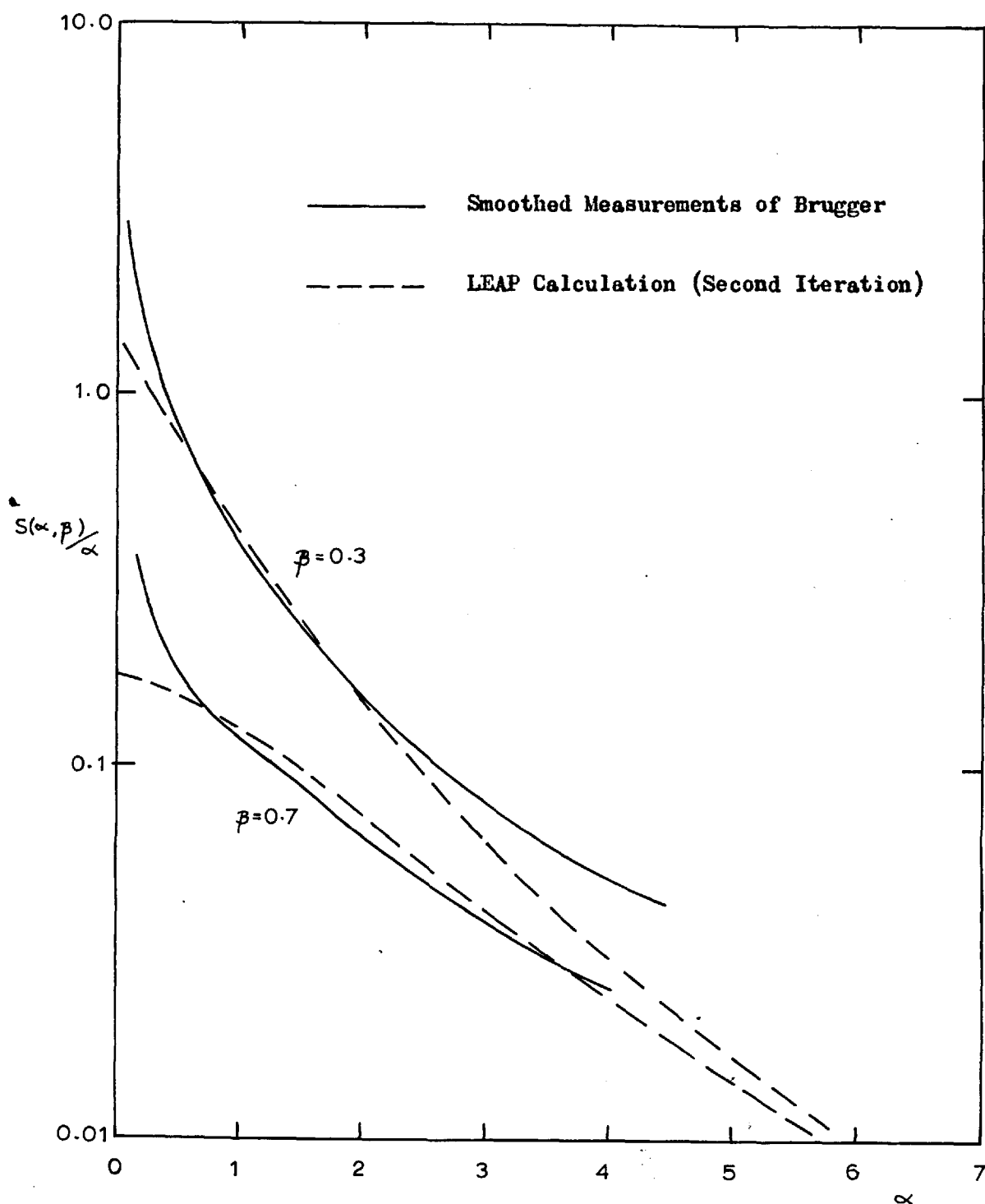


Figure 5 Typical Comparisons Between Experimental and Calculated $S(\alpha, \beta)$ vs. α for 267°C Santowax-R

Also shown are the extrapolations which were used to obtain the first estimates of $\rho(\beta)$, the spectral density, using the relation (derived in Appendix B).

$$\rho(\beta) = 2\beta \sinh \beta / 2 \lim_{\alpha \rightarrow 0} \frac{S(\alpha, \beta)}{\alpha} \quad (4)$$

This $\rho(\beta)$ was then fed as input to the programme LEAP - ADDELT, written by McLatchie (1965) and which was run on the University of London Atlas computer, to compute $S(\alpha, \beta)$. The theory of the LEAP part of the programme is outlined in Appendix C. Modes outside the range of measurement can be treated as δ - functions and are convolved with that part of $S(\alpha, \beta)$ generated from the fitted $\rho(\beta)$ using ADDELT (see Appendix A). $S(\alpha, \beta)$'s are calculated from the fitted part of $\rho(\beta)$ using three different methods, depending on the circumstances. At low α values the scattering law is expressed as a phonon series with the $(n + 1)$ th term giving the scattering law for an n -phonon process. Up to a maximum of 20 terms, the terms are calculated by direct numerical convolution. Beyond 20 terms an approximate form (Sjolander (1958)) is used. This series is an expansion in ascending powers of α and for larger α values an excessive number of terms would be required and so an alternative 'steepest descents' method (Egelstaff and Schofield (1961)) is used. This approach gives rise to an asymptotic expansion in inverse powers of α . On the basis of these calculations the new curve of $S(\alpha, \beta)/\alpha$ was adjusted up or down to give agreement over the middle α part of the measured range. Comparison of intercepts with the $\alpha = 0$ axis gives the change in $\rho(\beta)$ and this change may be large. These changes will mainly arise because multiphonon scattering from modes at lower β will give contributions at larger α values at higher β 's and thus prevent a single extrapolation from being successful. Thus equation 4 only holds at α values below the measured range or where multiphonon ~~points~~

effects are small. Five iterations were used in obtaining the final $\rho(\beta)$, but the major part of the change took place at the first iteration.

It will be seen from the $S(\frac{\alpha, \beta}{\alpha})$ plot in Figure 5 that experiment rises much more steeply than calculation at low α 's. This has been observed by workers studying scattering by other materials, and has been variously ascribed to multiple scattering in the sample or to anharmonic effects. No complete explanation has been proposed. Recently, however, Slaggie (1967) has calculated multiple scattering corrections for water samples and shown that these are large ($\sim 50\%$) at low α values. He considers that the technique used by Haywood and in this work of fitting to the middle α range eliminates the effect of these errors from $\rho(\beta)$.

Reference to Figures 6 to 8, where bound atom calculations are compared with the experimental data shows that there are very large discrepancies at $\beta = 0$. The reason for this is thought to be the crystalline model used which neglects diffusive motion of the molecule. Because of uncertainty concerning the mechanism by which diffusion occurs in Santowax-R, no attempt has been made to include this effect in the calculations. It should be possible to fit a free gas translation model to the $\beta = 0$ data by varying the mass of the free gas atom, but it will be shown in Section 3.4 that this procedure is not justified for reactor physics calculations.

Only hydrogen scattering was considered when fitting to $S(\alpha, \beta)$ for the whole molecule; assuming carbon to scatter according to the free gas model the contribution to $S(\alpha, \beta)$ per $C_{18}H_{14}$ molecule is only a few percent at $\beta = 0.1$ and is less than this for the higher values considered. In a reactor calculation carbon scattering would become significant in the few electron volt regions where 'free atom' cross sections prevail, but the use of the free gas model for carbon should then be adequate.

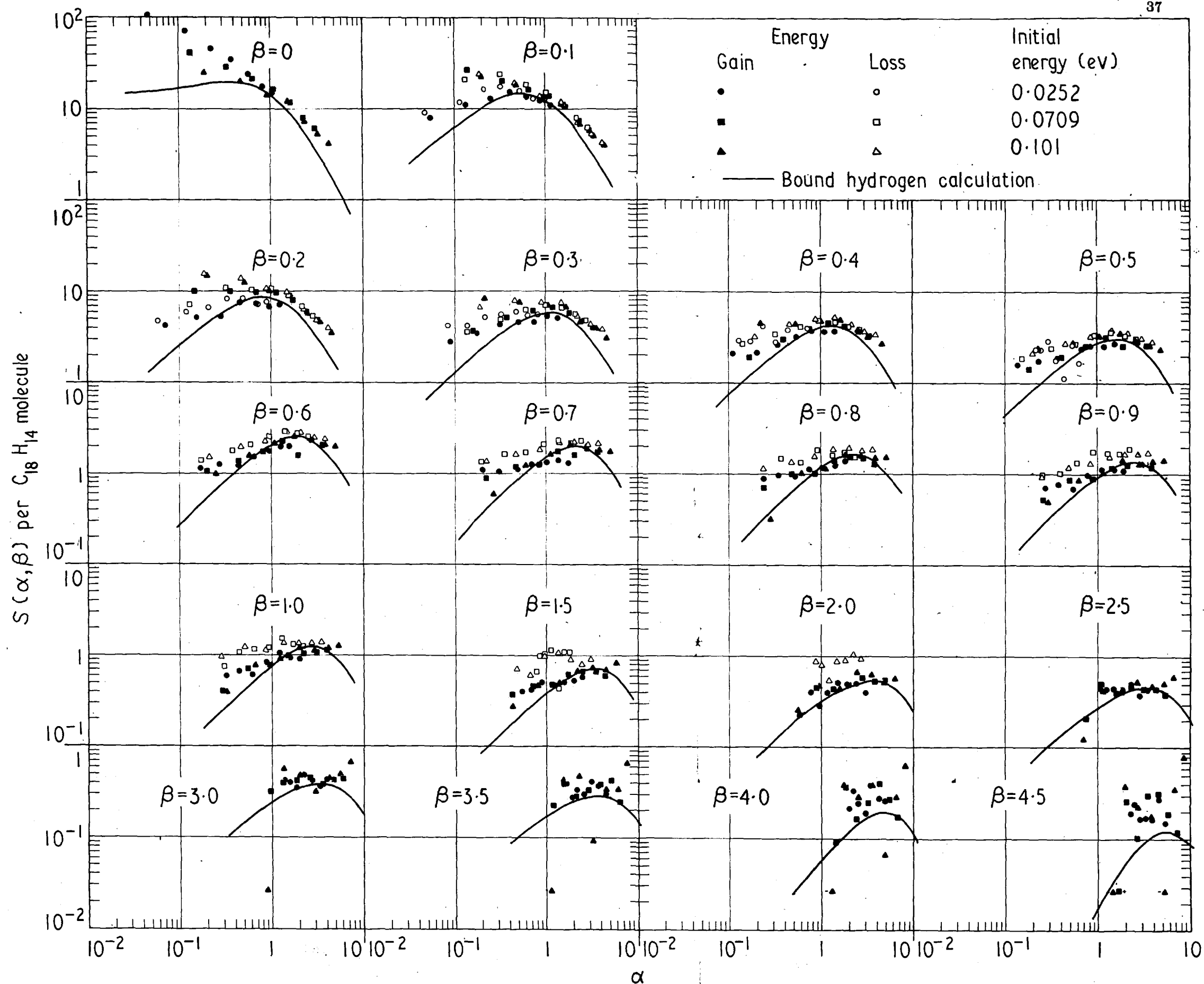


Figure 6 Scattering Law Calculations Based on the Fitted Model
for 207°C Santowax Compared with Measurements of Brugger

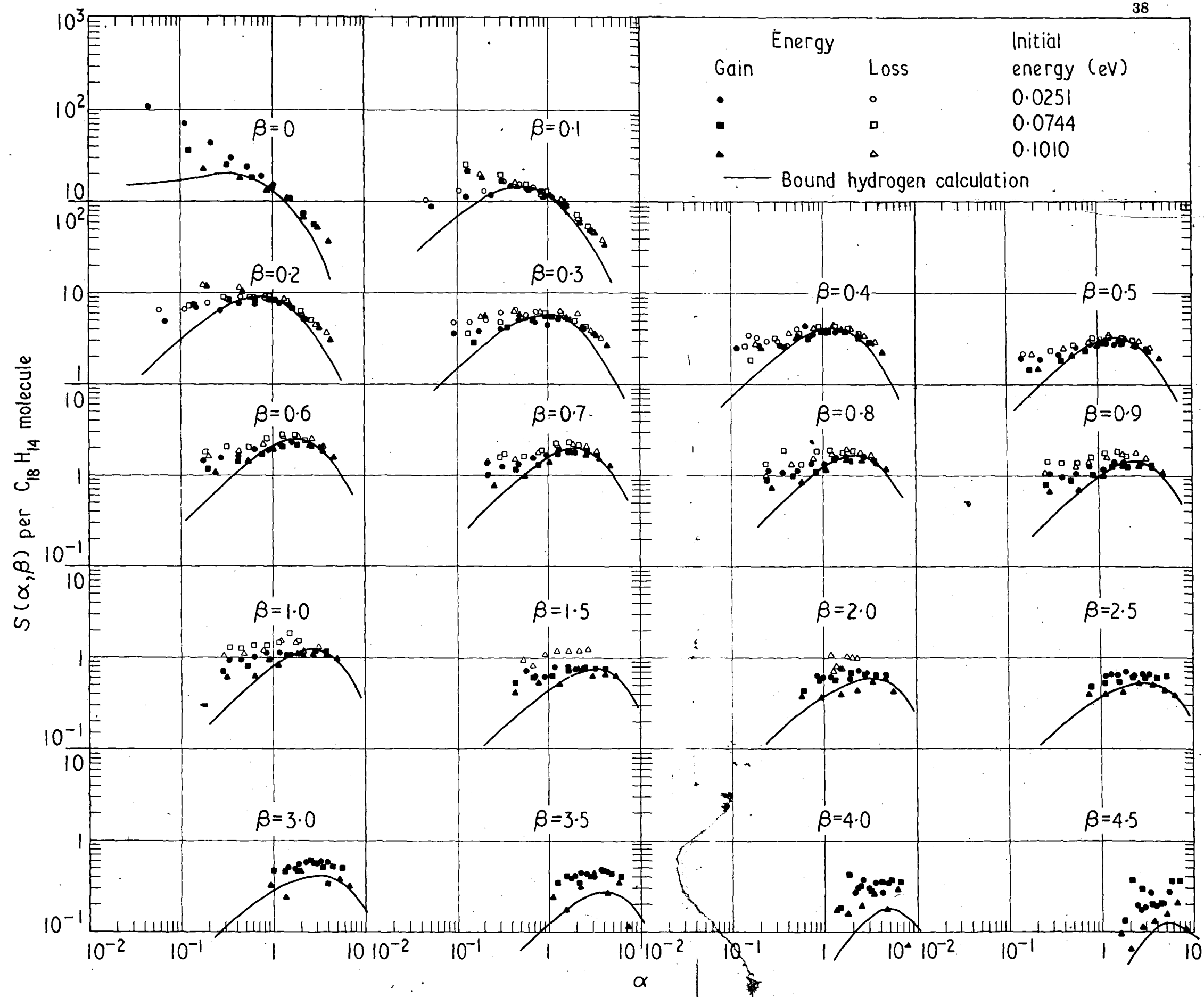


Figure 7 Scattering Law Calculations Based on the Fitted Model for 267°C Santowax Compared with Measurements of Brugger

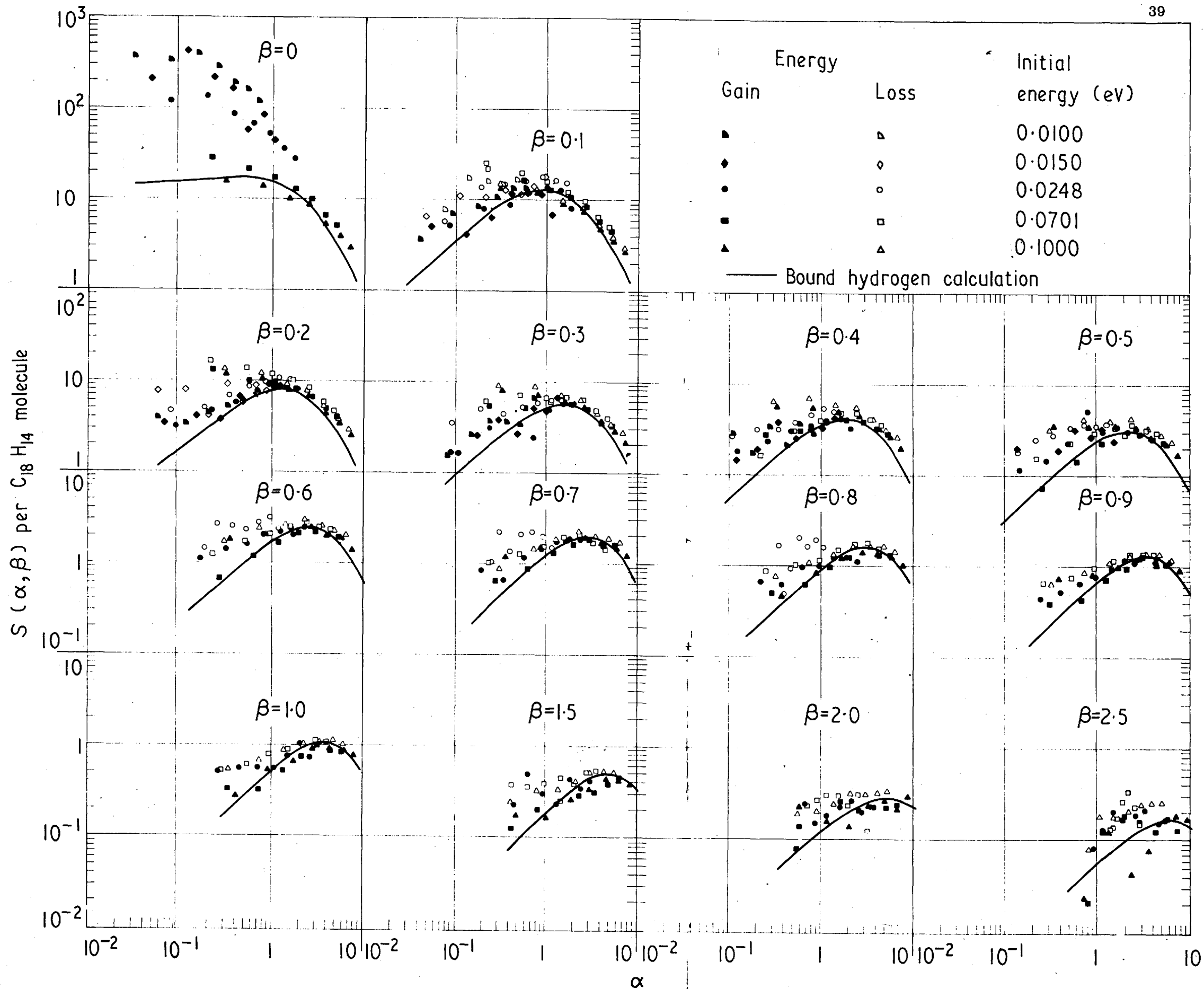


Figure 8 Scattering Law Calculations Based on the Fitted Model for 23°C Santowax Compared with Measurements of Brugger

3.2 Restrictions on the fitted

In Figures 6 to 8, where calculations are compared with experiment, there is the general disagreement, which has been noted, at low α values. Brugger considers that because of multiple scattering the most reliable experimental value of $S(\alpha, \beta)$ for $0.17 > \beta > 0.5$ is a line near the smaller values of $S(\alpha, \beta)$ and that at larger β values the energy loss points will fall above the neutron energy gain points. If this is taken into account the agreement is quite good for the solid data. The agreement is less good for the liquids, where there are discrepancies at large α values for low β values and all α values when $\beta > 2.5$. This latter discrepancy restricted the range of the $\ell(\beta)$ fit to $0.1 \leq \beta \leq 2.5$ for the liquid data.

In order to extend $\ell(\beta)$ beyond 2.5 for the liquids the available data for benzene and diphenyl was consulted. It is only necessary to construct an approximate model in this range, for if the modes are at high energies then their effect on thermal reactor calculations at these temperatures will be a second order one. For benzene the calculations of Crawford and Miller (1949) and Boffi, Molinari and Parks (1962) indicate that the C-H vibrations are concentrated at 0.38 eV and that there are few modes between this energy and the broad group of ring vibrations at ~ 0.14 eV. For diphenyl an extensive series of calculations and an analysis of experimental data on the fundamental frequencies has been made by Sandroni and Zerbi (1965). These show a similar absence of modes in the range 0.2 eV to 0.38 eV and a concentration of modes at 0.38 eV. We have therefore extrapolated the continuous, fitted, part of $\ell(\beta)$ to zero at 0.17 eV for the liquids and approximated the C-H vibrations by a δ function at 0.38 eV.

The second problem is the assignment of an area to the δ function so that the normalization

$$\int_0^{\infty} \ell(\beta) d\beta = 1 \quad (5)$$

is preserved. The relative areas in the δ function and under the rather ill-defined large peak in $\rho(\beta)$ were varied while the above normalization was maintained; it was found that the best overall agreement with the 'middle α ' data was obtained with a δ function of the form $0.35 \delta(\beta - \beta_1)$. A similar problem arose for the 23°C solid S(α, β) data. Because the temperature was lower, the modes observed were at lower energies and the data only extended to 0.07 eV. The fitted part of $\rho(\beta)$ extending up to ~ 0.07 eV was quite well defined, and the area was taken to be 0.12. It was assumed that the remainder of $\rho(\beta)$ had the same form as the liquid and that the large peak in the liquid data could be approximated by two δ functions to give a complete distribution of the form

$$\rho(\beta) = \rho(\beta)_{\text{fitted}} + 0.09 \delta(\beta - 3.7) + 0.44 \delta(\beta - 5.5) + 0.35 \delta(\beta - 15) \quad (6)$$

This representation appears justified by the satisfactory agreement obtained between calculation and experiment shown in Figure 8. Figure 9 shows the proposed spectral density distributions for Santowax-R at the three temperatures.

No attempt has been made to assign an error to the fitted $\rho(\beta)$ as this would require a knowledge of both random and systematic errors in the data besides an understanding of the importance of any non-Gaussian contribution to the scattering. Inspection of Figures 6 to 8 shows that the contribution of random errors in the data amounts to approximately 15% at low β values and approximately 30% at high β values.

3.3 Comparison Between Solid and Liquid Santowax, and Between Santowax, Diphenyl and Benzene.

As spectral density distributions have been derived both for solid and liquid Santowax, these may be compared in an attempt to detect differences arising from the change of state. The fitted spectral density distributions are plotted on a common energy scale in Figure 10. The two distributions are quite similar in the energy

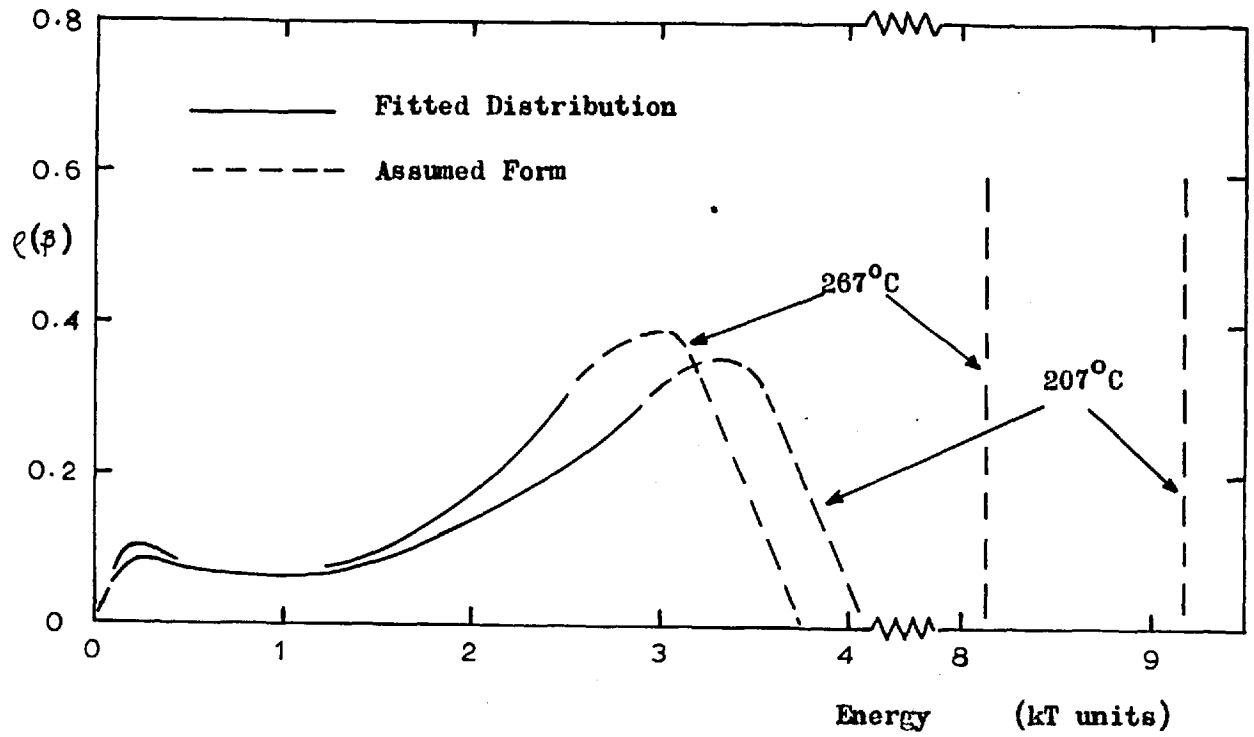


Figure 9(a) Fitted Spectral Density Distributions of H-atom Motion in Santowax-R at 207°C and 267°C

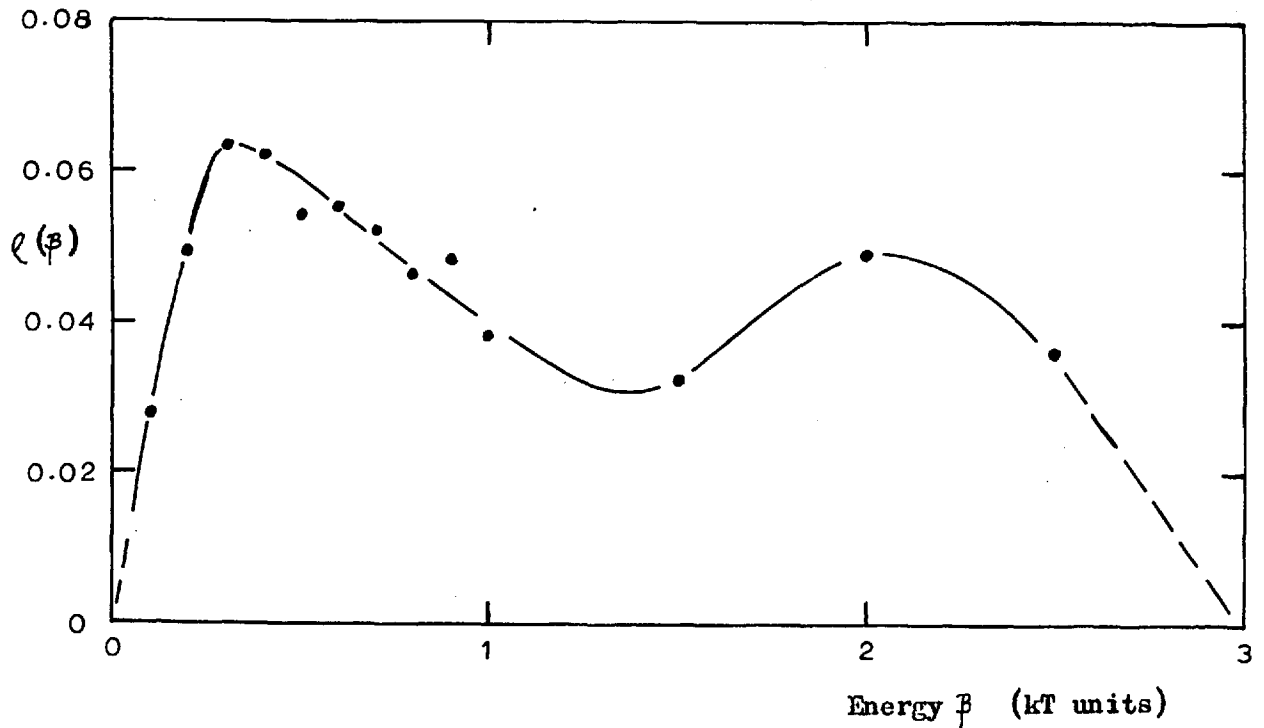


Figure 9(b) Fitted Spectral Density Distribution of H-atom Motion in Santowax-R at 23°C

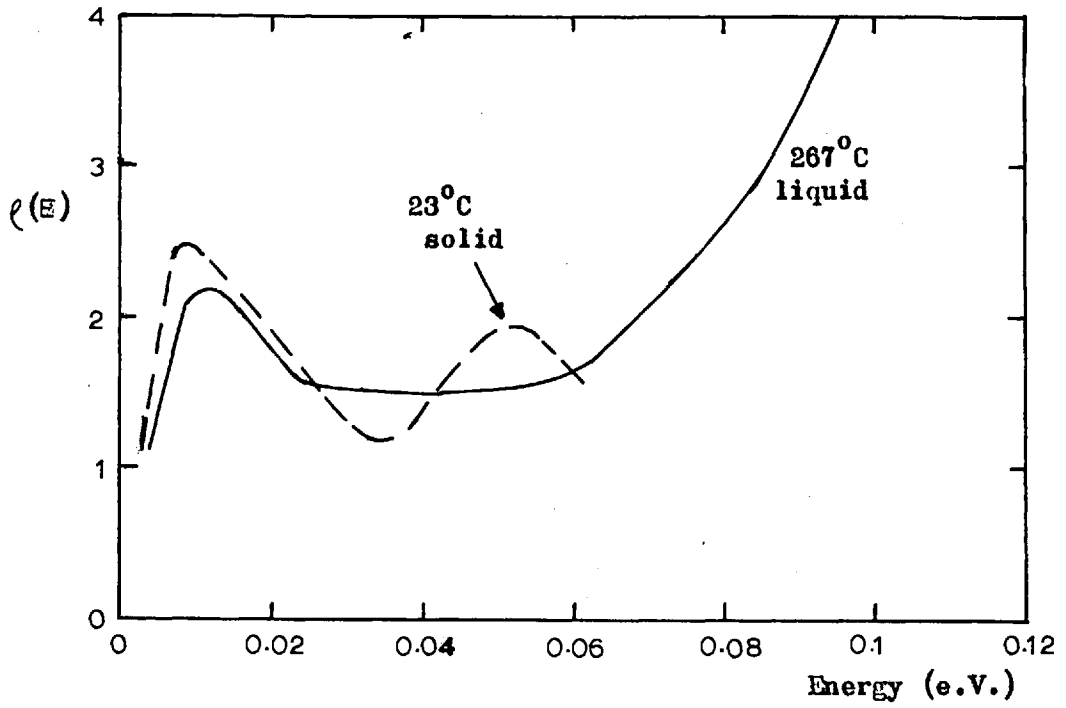


Figure 10(a) Comparison Between Fitted Spectral Densities Plotted on an Energy (e.V.) scale for Solid and Liquid Forms of Santowax-R

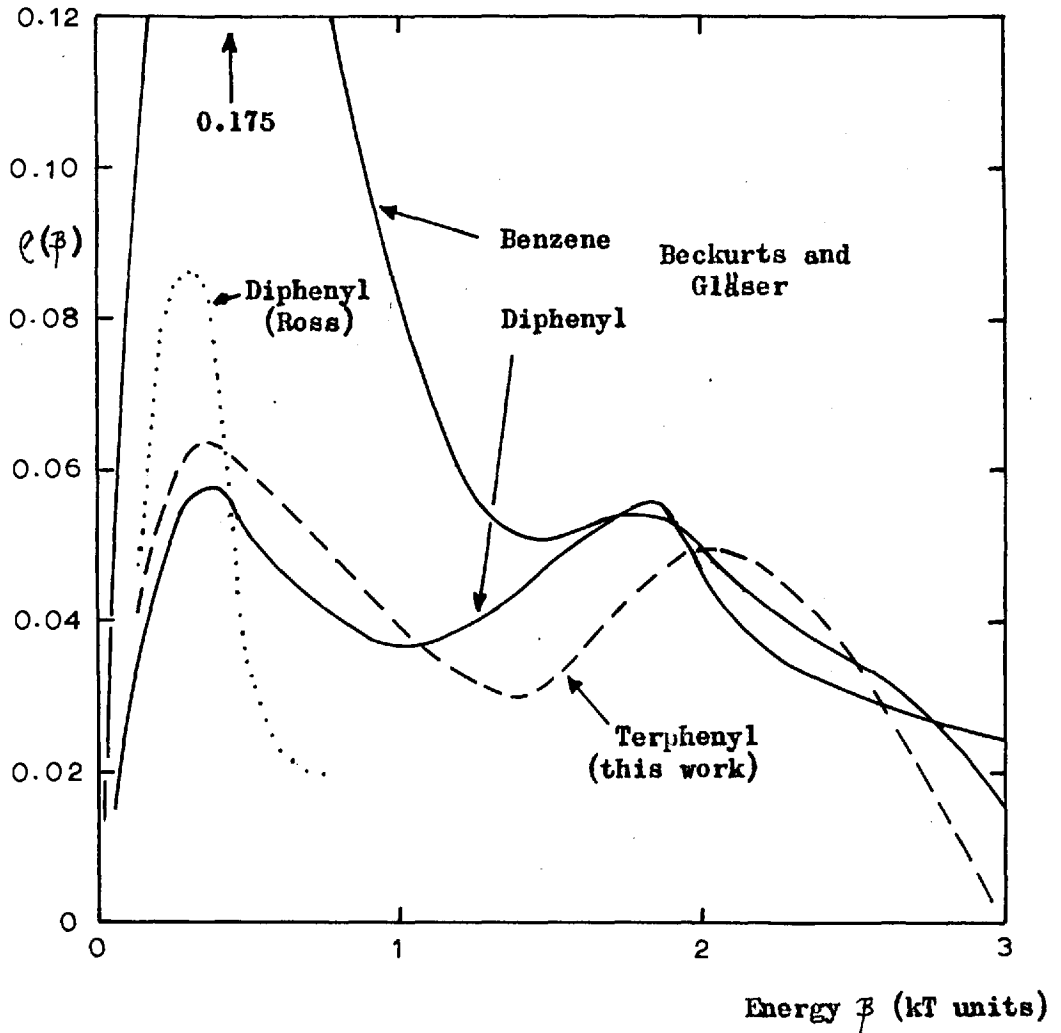


Figure 10(b) Comparison Between Fitted Spectral Densities at Room Temperature for Benzene, Diphenyl and Terphenyl

range 0.005 - 0.06 eV and in view of the possibly large errors in the fitted $\rho(\beta)$ the differences may not be significant. This absence of any effect of the phase change indicates that the modes observed are not due to motion of the whole molecule.

In Figure 10 the $\rho(\beta)$ distributions for 23°C solid Santowax (this work) and for benzene and diphenyl derived by Beckurts and Gläser are compared. The distributions for diphenyl and Santowax-R do not differ significantly, but while both show a small peak at 0.01 eV, benzene shows a large peak near this energy. The results obtained by Ross et al for benzene are also shown, which differ from those of Beckurts and Gläser; it should be noted that Ross et al relied on a simple extrapolation and did not use the iterative method.

3.4 The Diffusion Length for Santowax-R

The diffusion length for Santowax-R has been measured by Hohki (1965) et al over the temperature range 200°C to 400°C. A loop pressurized by nitrogen was used and neutrons were fed into the pressure vessel by a Po-Ba source in a graphite plinth. Axial and radial distributions were measured using ceramic D₂O₃ discs. An iterative method was used to eliminate a small harmonic content.

Tremeer (1966) has published measured diffusion lengths over the more restricted range of 170°C - 230°C which resulted from rethermalisation studies in an H₂O/Santowax-R two-tank assembly. These studies are discussed in more detail in Chapter 5. The 10.3 cm thick cold water tank was next to the graphite thermal column. 'Axial' measurements in water and Santowax were made with fission chambers. Transverse measurements were made from the axis outwards in only one direction and an extrapolated width of 93 cm was obtained. Tremeer assumes that this is a genuine buckling and that the extrapolated width in the other transverse direction is determined by the tank physical size. However, the 61 cm square tank was not well matched to the 95.2 cm

square thermal column. Any harmonic content would not be attenuated by the water tank due to the small relaxation length differences arising from the small transverse buckling ($\frac{1}{\gamma_{11}} = 2.7 \text{ cm}$ $\frac{1}{\gamma_{33}} = 2.38 \text{ cm}$). In the Santowax tank, however, the difference between γ_{11} and γ_{33} (γ_{22} cannot be present in this symmetric system) are again small, but great enough to affect the deduced L . For example with a diffusion length of 6.1 cm, $\frac{1}{\gamma_{11}} = 5.6 \text{ cm}$ and $\frac{1}{\gamma_{33}} = 4.8 \text{ cm}$. In order to estimate possible corrections to Tremeer's data the relative proportions of (1,1) and (3,3) components in the flux entering the Santowax tank were obtained by a graphical method and were found to be 95% and 5%. Using these proportions, the percentage by which L would be overestimated (ϕ_{33} was negative on the axis) if harmonics were ignored and the transverse buckling were assumed to be fixed by the tank size, was estimated and found to be 2.2%. Now Tremeer took a relaxation length possibly too great by this percentage and combined it with a transverse buckling, possibly too small. The underestimate of L due to a possibly erroneous transverse buckling was 2.3%. Thus even if harmonics are postulated to explain the large transverse buckling, the value of L will not change. However, a possible systematic error of $\pm 2\%$ has been assigned to Tremeer's data.

In the calculation of diffusion length, besides the Santowax model fitted to Brugger's data, two other models of hydrogen scattering have been used. These are, the free gas model which has been described in Chapter 2, and a model which we have called the Ardente model. Ardente (1967) has proposed a simple Nelkin-type model based on the possible dynamics of the benzene molecule. The energies and effective masses of the modes are as follows:-

	Mass	Energy (eV)
Translation	78	0
Rotation	21.3	0.02
Vibration	(2 x Mv	0.12
	(2 x Mv	0.38

$$\text{Where } \frac{1}{M_T} + \frac{1}{M_R} + \frac{1}{M_V} = 1 \quad (7)$$

This normalisation is equivalent to the normalisation to unity of $\rho(\beta)$. The molecule is assumed to vibrate with two internal modes and these modes are positioned at the approximate energies of ring and C-H vibrations in benzene. Ardente suggests that this model may be adequate for the polyphenyls.

A limitation of the models fitted to Brugger's data is that they were, for the liquid, evaluated at 207°C and 267°C and a method of interpolation and extrapolation is obviously necessary. Now the spectral density distribution does not change greatly with temperature on an energy (eV) scale and the chosen procedure has been to convert the $\rho(\beta)$ to $\rho(E)$ and linearly interpolate and extrapolate. Using this procedure the $\rho(\beta)$ curves for 170°C, 200°C and 230°C have been obtained and are illustrated in Figure 11.

Diffusion lengths were calculated using the programme DIS written by P. W. Johnson (1966). Cross sections for this calculation are evaluated as follows; the angular integration of equation is accomplished by double Gaussian quadrature (order 15) while the energy integration to obtain total cross sections is of the Lagrange type with a cubic equation being fitted to successive sets of four out of 32 energy points and Simpson's rule being used to integrate across the central interval. The fundamental space eigenvalue (the diffusion length) of the source-free one dimensional transport equation is obtained by B_L analysis, using a method proposed by Honeck (1962). Differential cross sections for the free gas and Ardente models for hydrogen are obtained analytically while for the fitted models DIS will interpolate linearly in a matrix of $S(\alpha, \beta) e^{\beta/2}$ values. $S e^{\beta/2}$ varies more slowly with β than does S . In the Santowax calculation typically 35 β and 32 α values were used; these rather small matrices were used to economise on LEAP running times; the errors in the total cross section arising from this interpolation are thought to be 1 - 2%. In the absence of

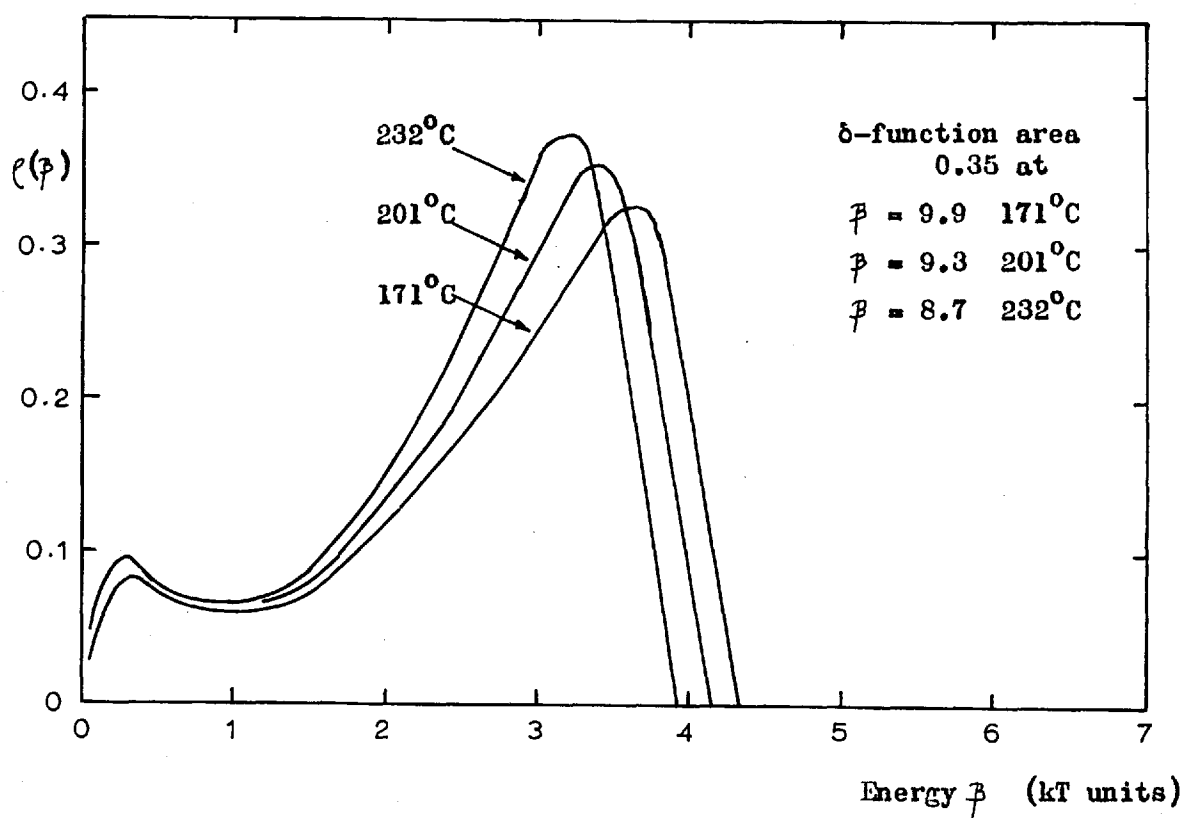


Figure 11. Spectral Density Distributions for Santowax-R at 171°C, 201°C and 232°C Obtained by Interpolation

any treatment of diffusive motion the quasi-elastic scattering is simply the first term in the phonon series

$$S(\alpha, \beta) = e^{-\alpha\lambda} \phi(\beta) \quad (8)$$

where λ is the Debye-Waller coefficient which is computed by LEAP:

$$\lambda = \int_0^{\infty} \frac{\coth \frac{1}{2}\beta}{\beta} \phi(\beta) d\beta \quad (9)$$

Carbon has been assumed to scatter according to the free gas model and the densities of Santowax at the various temperatures are those measured by Trilling (1957). Uncertainties in density may represent the major possible error in the calculations.

Calculated diffusion lengths are compared with experiment in Figure 12 and Table XII. The B_1 approximation has been used in the calculations, to economise on computing time; some comparative calculations with the B_2 approximation produced changes of 0.02%. Densities used are also given in the Table in order to facilitate comparison with future work. Calculations with the Ardente model were taken to higher temperatures in order to verify the trend with temperature of the measurements of Hohki et al.

Table XII Measured and Calculated Diffusion Lengths

Temperature (deg. C)	Measured (cm) (Tremeer)	Calculated (cm)			Assumed density (gm cm ⁻³)
		Goddard	Ardente	Free Gas	
171*	5.88 ± 0.12	5.46	5.57	6.93	0.991
201*	6.16 ± 0.12	5.80	5.87	7.21	0.967
207		5.85	5.97	7.27	0.962
232*	6.48 ± 0.13	6.01	6.18	7.51	0.943
267		6.34	6.55	7.85	0.916
300			6.92	8.22	0.887
400			8.01	9.31	0.812

*The Goddard model at these temperatures was obtained by interpolation or extrapolation.

Figure 12 shows that the measurements of Hohki et al are not of sufficient accuracy to distinguish between the models for H scattering

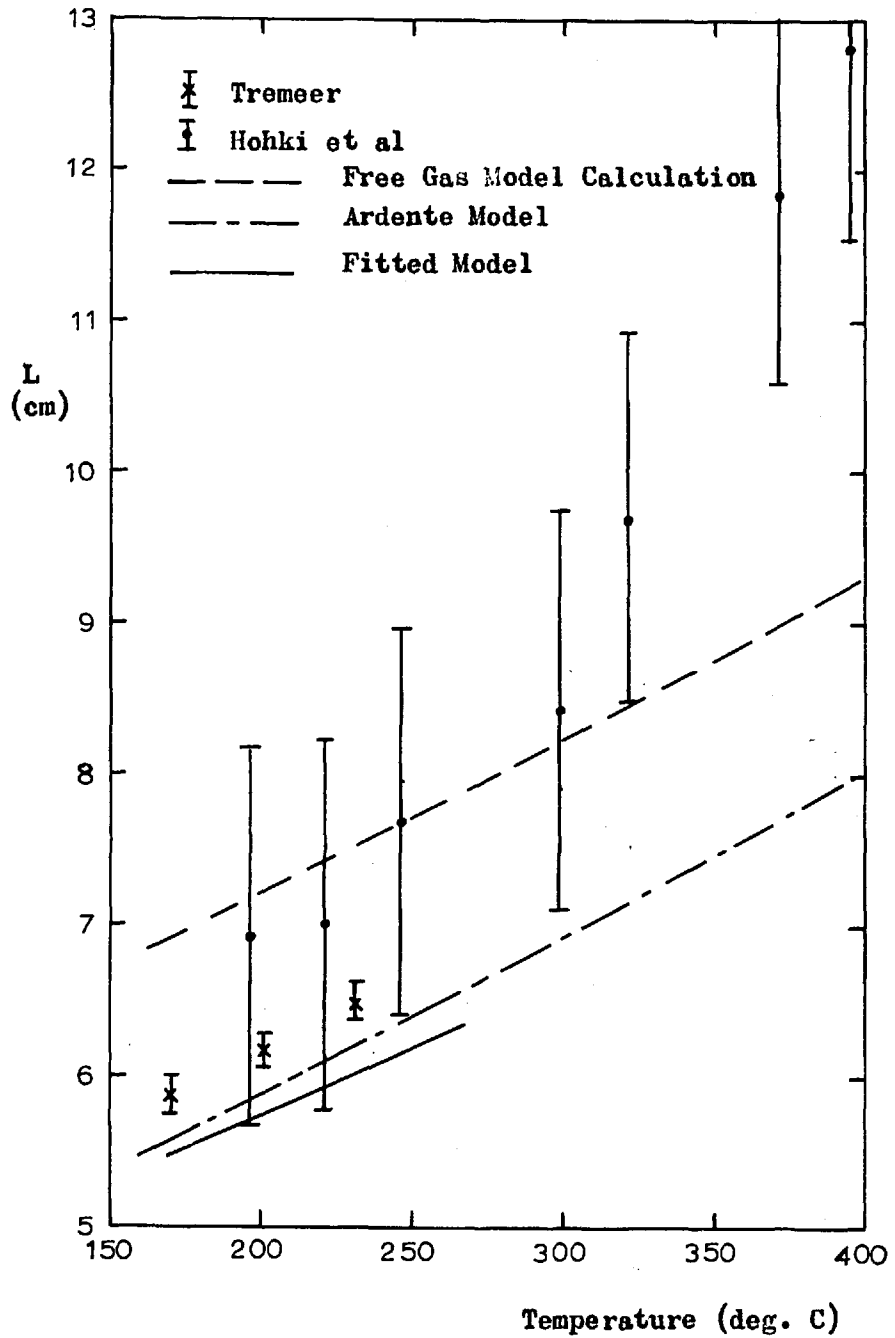


Figure 12 Thermal Neutron Diffusion Length in Santowax-R.
A Comparison Between Calculated Values Using Three Models
and Available Experimental Data

and in fact agree best with the free gas predictions. At 400°C the difference between these experiments and calculation is considerably greater than the errors assigned to the measurements. The predictions of the fitted model and the simple Ardente model do not differ significantly but lie below the experimental values of Tremear by 6%, that is by more than the 2% assigned systematic error of the measurements. However, if density and $S(\alpha, \beta)$ matrix interpolation errors are taken into account then this difference could lie just within the sum of experimental and theoretical errors. Total cross sections were examined as described in the next section in order to obtain more information about this discrepancy.

An unresolved question which arose from the fitting of the model to Brugger's data where these remained a large discrepancy at $\beta = 0$ was ascribed to the absence of diffusive motion in the theory. A programme BOND specified by the author and P. W. Johnson and written by P. W. Johnson has been used to convolve a mass 230 (total mass of the Santowax molecule) with the 207°C fitted model.

$$S(\alpha, \beta) = \int_0^\infty S_{\text{gas}}(\alpha, \beta'/230) S_{207}(\alpha, \beta - \beta') \alpha \beta' \quad (10)$$

Figure 13 shows that this model with free gas translations gives improved agreement with Brugger's elastic data, but the calculated diffusion length changes from 5.85 cm to 5.87 cm - not a significant change.

3.5 The Total Cross Section of Santowax-R

Brugger has measured the total cross section of liquid Santowax at 0.0253 and has obtained 810 ± 24 barns per molecule at 207°C and 791 ± 24 barns at 267°C. Ardente model calculations using the method outlined in Section 3.4 yield 925 barns and the Goddard model 895 barns in this temperature range. This comparison is presented graphically

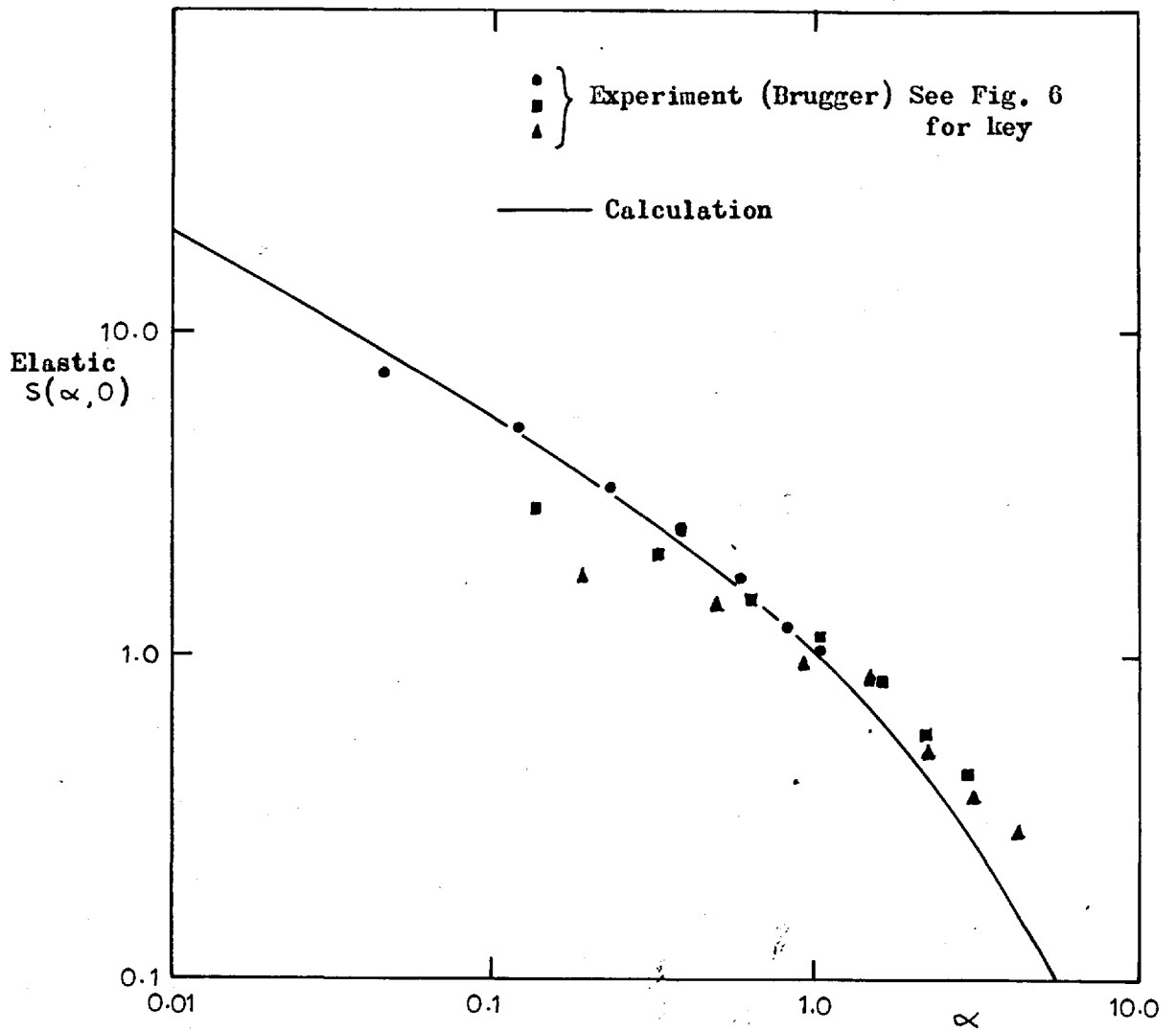


Figure 13 Quasi-Elastic Scattering for Santowax Measured by Brugger
 Compared with Calculation Using Free Gas Mass 230

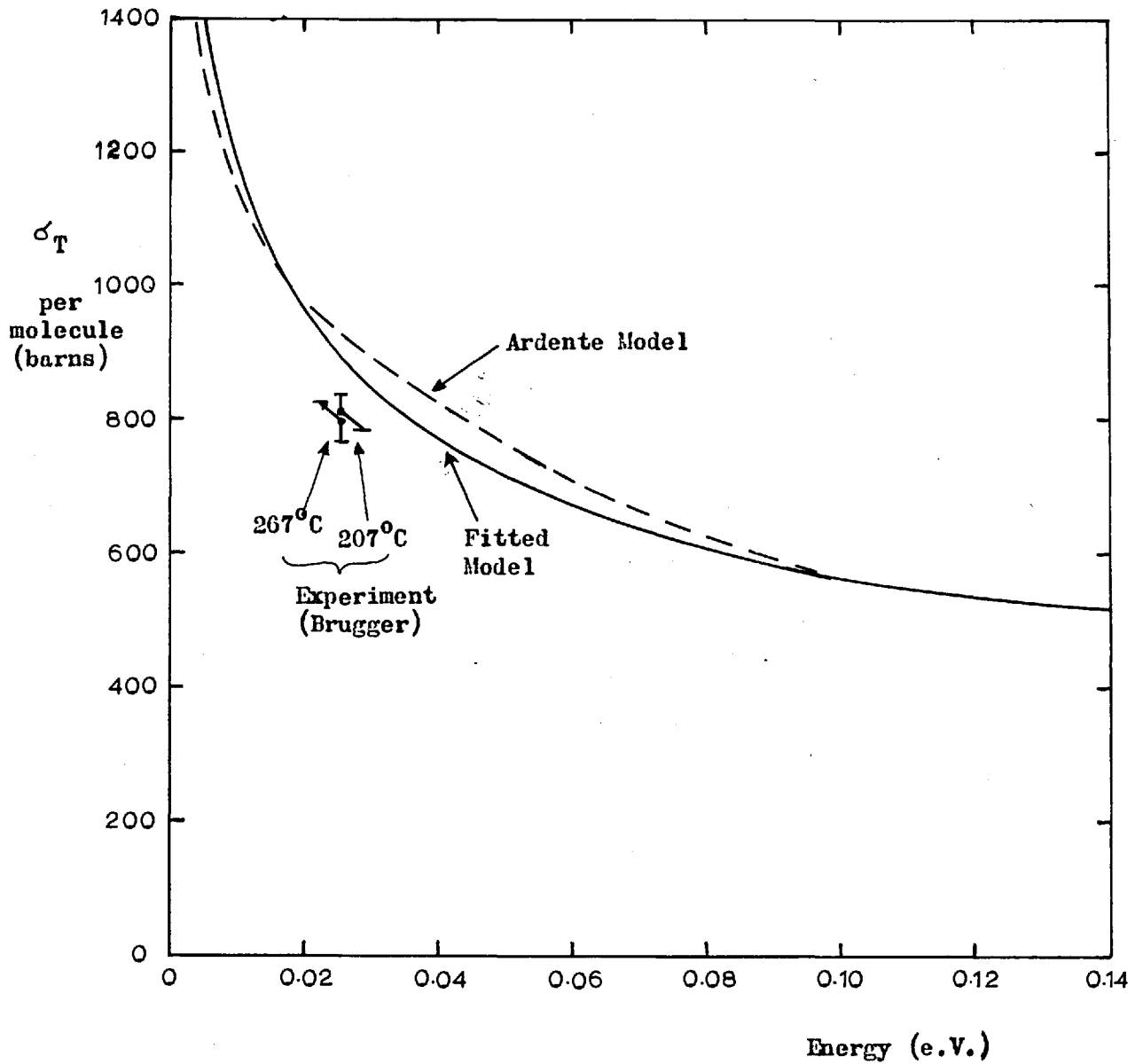


Figure 14 Total Cross Section of Santowax Calculated Using Two Models Compared with the Measurements of Brugger

in Figure 14. The discrepancy suggests an overestimate in cross sections derived from the fitted model which is in the same direction as that indicated by the discrepancy between experimental and calculated diffusion lengths. However, the experimental information is so restricted at the present time that it cannot be regarded as certain that the fitted model is deficient.

CHAPTER 4

NEUTRON SCATTERING BY LIGHT WATER

Neutron interaction with water has been studied more extensively than for any other moderator. One reason for this concentration of effort has been the need to predict thermal neutron transport and spectrum distortion in water reactors. The second reason has been the relative simplicity of the dynamics and environment of the molecule in liquid water and the fact that the scattering is predominantly by hydrogen and is essentially incoherent. Several scattering models for hydrogen have been proposed ranging from a representation of molecular modes by widely spaced discrete harmonic oscillators to a continuous mode density distribution derived from experiment. At the present time no one model is completely satisfactory. In this Chapter the predictions of four different scattering models are compared with the precise diffusion length measurements of Johnson (1966). The accuracy of the available experimental data compensates for the fact that the experiments will only reveal properties averaged over an energy range near to the Maxwellian peak. This is in contrast to direct spectrum measurements in poisoned solutions or total cross section measurements, which cover a wide energy range but with rather lower accuracy at the present time.

The first realistic model for bound proton scattering was due to Nelkin (1960) and assumes the dynamics of a proton to be translation of the entire molecule plus a number of harmonic oscillators describing rotation and vibration. Koppel and Young (1964) have proposed modifications of the Nelkin model to take account of the spatial anisotropy of the vibrations. A model with possible future potential has been proposed by McMurtry, Russell and Brugger (1966) who attempt to take

account of the varying environment of a molecule in water by using a model which also originates from the Nelkin concept. Several authors have interpreted double differential scattering experiments in terms of a continuous energy density of modes of vibration but only Haywood (1967) has so far proposed a complete model for use in reactor calculations. Important modifications to both the Nelkin and the Haywood models have been proposed recently by Koppel (1966). Slaggie (1967) has calculated multiple scattering corrections to the basic measurements and has commented on the errors arising from this source in the McMurry-Russell and Haywood models. Recently Beyster (1968) has reviewed experimental data and models for neutron scattering by water and has concluded that it is not possible, at the present time, to calculate both total cross section and differential scattering from any one model. Precise total cross section measurements (which agree with the earlier measurements of Melkonian (1949)) and an evaluation of the mean kinetic energy per bound proton have been performed by Russell, Neill and Brown (1966). Numerous authors (for example, Clendenin (1964)) have compared predictions, using individual scattering models, of the diffusion constant with experiment. However, no systematic studies have been made, using a range of scattering models, of the diffusion constants and diffusion heating or cooling coefficients for $\frac{1}{v}$ and non- $\frac{1}{v}$ absorbers. An interesting feature of the comparison presented here is the light thrown on the consistency of cross section sets for cadmium and gadolinium.

The probable structure of water and the motion of the molecule within this structure is outlined in Section 4.1. Four models for hydrogen scattering have been constructed in order to highlight particular parts of the spectral density distribution. Of these four, the Haywood model and related assumptions is described in Section 4.2 and the other three models in Section 4.4. In Section 4.5 the predictions

obtained with these four models are compared with the measured diffusion parameters of Johnson. This comparison uses more recent cross sections for cadmium and gadolinium than were used in the published work.

(Goddard, Grant, Johnson and Tremere (1967)). Calculated total cross sections and mean kinetic energies are compared with experiment in Section 4.6. The majority of the work described in this Chapter has been carried out in collaboration with P. W. Johnson and is to be published in "Nuclear Science and Engineering".

4.1 The Structure of Water

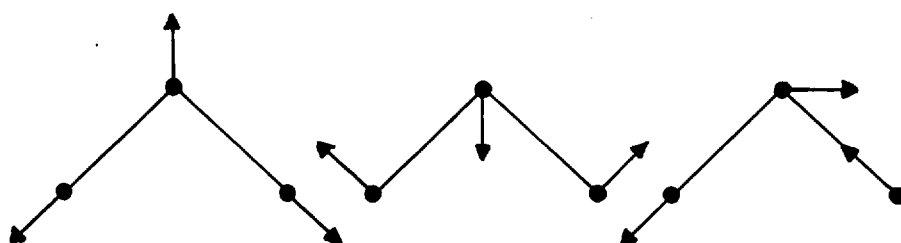
Present ideas of the structure of water assume the presence of crystal-like aggregates containing large numbers of molecules which are constantly breaking up and reforming. Hydrogen bonds maintain this structure. The first experimental evidence from neutron scattering of a structure for water came from the measurements of Brockhouse (1958) who used a crystal spectrometer technique to study the angular distribution of elastically scattered neutrons. It was found that the intensity varied with angle in the manner of a Debye-Waller factor $\exp(-\alpha\lambda)$, where α is the neutron momentum change, and this form normally arises from thermal motion within a crystalline lattice. In addition the energy spread of the elastically scattered neutrons could not be explained on the basis of simple diffusion and Brockhouse had recourse to a jump diffusion model which assumed that the molecule remains bound for a certain time before undergoing simple diffusion for a further, shorter, time. This residence time, which may be regarded as time spent in an aggregate of molecules, allows the development of a solid-like spectral density distribution of molecular motion.

The crystalline character of water, at least near to the freezing point, is further illustrated by the fact that the cold neutron scattering measurements of Larssen and Dahlberg (1962) made upon water

immediately above and below the freezing point indicate that neutron interactions occur with acoustic and optical vibrations and that the positions of these modes do not change significantly between phases. Similar modes are also observed at $\sim .01$ and $\sim .025$ eV in the Haywood (1967) spectral density distribution. There is little evidence at present as to how the size or binding of molecular aggregates change with temperature.

The three fundamental modes of vibration of the water molecule are illustrated in Figure 15 and comprise a bending vibration and symmetric and asymmetric stretching vibrations. These modes have been observed in infra-red absorption measurements (see, among several authors, Harzberg (1960)) and while these measurements indicate the energies of the vibrations they cannot be used to predict hydrogen atom amplitudes because infra-red absorption depends upon dipole changes of the vibrating molecule. Haywood has discussed the relevance of Raman and infra-red measurements to the study of neutron scattering processes. He points out that the vibrational potential is not quadratic and that the zero amplitude stretching vibration calculated by Herzberg, 0.474 eV (symmetric) and 0.488 eV (asymmetric), differ from the observed energies of 0.428 eV and 0.450 eV (liquid phase) and 0.453 eV and 0.466 eV (vapour phase). The vibrational modes are observed in infra-red spectra as part of a broad distribution but there is no evidence at the present time whether neutrons 'see' discrete or broadened vibrational modes. Haywood comments that the bending mode energy is only slightly changed by the liquid - vapour phase change, from 0.198 eV to 0.205 eV.

It remains to discuss the manner of molecular rotation. We have noted that the liquid is formed from transitory aggregates of molecules. In contrast to the situation in the vapour, where the molecule will be



Symmetrical
'Stretching'
Vibration

~ 0.48 e.V. H_2O

~ 0.35 e.V. D_2O

'Bending'
Vibration

~ 0.20 e.V. H_2O

~ 0.15 e.V. D_2O

Asymmetric
'Stretching'
Vibration

~ 0.48 e.V. H_2O

~ 0.35 e.V. D_2O

Figure 15 Normal Modes of Vibration of the
 H_2O and D_2O Molecules

free to rotate in a manner near to the classical, in a liquid the molecule will be restrained by the hydrogen bonds maintaining the aggregate and may perform small amplitude torsional oscillations. It is, however, unlikely that an oscillation at a single energy will exist, for several reasons. The moment of inertia of the molecule about the three principal axes of rotation will not be the same, the potential may not be quadratic but more shallow and lastly, because of the irregular environment, the bonding will differ depending on whether the molecule is within or at the edge of an aggregate or is free. A broad band of hydrogen atom frequencies due to rotation is to be expected and has been observed by Haywood, Larssen and Dahlberg and by Kottwitz, Leonard and Smith (1962) using different measuring techniques.

4.2 The Haywood Model and Assumptions Relating to it

A particular assumption made by Haywood in the analysis of his experimental results was that all hydrogen atoms scatter in an identical manner and that it is meaningful to consider a 'single average atom'. This approach, which greatly simplifies a situation where, in fact, molecules may be in completely different environments, either free or within clusters. McMurry, Russell and Brugger (1966) treat water as a mixture of 10% free molecules and of 45% each of two types of aggregate. The total masses of these aggregates are 150 and 75 and the energy and density of vibrational modes below 0.125 eV were adjusted to give agreement with differential scattering measurements. However, Slaggie (1967) has published calculated multiple scattering correction factors for these measurements which throws doubt on the present McMurry-Russell model. This model represents, however, a powerful concept and one which may be used in the future if, as seems likely, it does not prove possible to reconcile differential measurements with total cross section and integral measurements.

The Haywood analysis makes a second major assumption; that the harmonic oscillators (there is no evidence that the use of the harmonic oscillator model introduces significant errors) representing molecular vibrations are isotropic in space. As our aim is to comment on the Haywood model and to understand the importance of the shape of the spectral density distribution we have also used the isotropic approximation for the three comparative models. Koppel and Young (1964) have noted that the angle between the two O-H bonds is effectively constant and that the 'bending' vibration and two 'stretching' vibrations must be correlated in space. They assume an infinitely heavy oxygen atom and this leads to two stretching vibrations, one bending vibration and three completely hindered rotational modes. When applied to the calculation of total cross sections this anisotropic model produced a local increase of up to 4% in total cross section, compared with the Nelkin model (isotropic), between 0.02 and 0.08 eV (Young and Koppel (1965)). Beyster (1968) has quoted diffusion coefficients D_0 of $37906 \text{ cm}^2 \text{ sec}^{-1}$ for Nelkin (in good agreement with the calculated value in Table XIV) and $36352 \text{ cm}^2 \text{ sec}^{-1}$ for the Koppel-Young model. These changes are significant.

It is assumed conventionally that vibrational motion, internal or of the molecule as a whole, can be separated from the diffusive motion that is characteristic of a molecule in a liquid and which is responsible for line broadening in elastic scattering. Haywood neglects diffusive motion completely, quoting Schofield to the effect that the area of the spectral density distribution allocated to diffusive modes is less than 0.5%. With this assumption the elastic scattering is just the Debye-Waller factor

$$S(\alpha, \beta) = e^{-\alpha\lambda} \quad (10)$$

where λ is the Debye-Waller coefficient

$$\lambda = 2 \int_0^\infty \frac{1}{\beta} \coth \beta / 2 \quad \epsilon(\beta) d\beta \quad (11)$$

The importance of diffusive motion is discussed further in Section 4.4.

Haywood derived a spectral density distribution $\rho(\beta)$ for water using extrapolation techniques which have been described and applied to terphenyl in Chapter 3. His spectral density distribution is illustrated in Figure 16. The main features are two small peaks at $\beta = 0.5$ (0.0125 eV) and $\beta = 1$ (0.025 eV) which may be attributed to hindered translational motion of the whole molecule, a main peak at $\beta = 2.7$ (~ 0.07 eV) due to hindered rotation and two delta-functions representing the vibrational motions. The fitting procedure was only possible up to $\beta = 4$ and above this a constant $\rho(\beta)$ was assumed up to $\beta = 10$. The form of the constant part was such as to satisfy the normalisation of $\rho(\beta)$, to maintain 50% of the area of $\rho(\beta)$ under the continuous part and to force the mean kinetic energy $\bar{K}$ of the bound proton into agreement with experiment.

Scattering law values suitable for input to the programme for calculating diffusion lengths and total cross sections were obtained by using the programme LEAP-ADDELT, described in Chapter 3, to calculate a matrix of $S(\alpha, \beta) e^{\beta/2}$ values. Earlier calculations published by Johnson (1966) used a 31×35 matrix, whereas the present work uses 51×60 values. These values covered a range such that scattering cross sections in the range 0 - 1 eV could be calculated. Changing from the coarse to the fine $S(\alpha, \beta)$ matrix produced a change in diffusion constant of $\sim 1\%$. A proportion of this difference arises from the use of an improved energy mesh in the later calculations and it is likely that the remaining error arising from the use of the 51×60 matrix is appreciably less than 1%.

4.3 Models Used for Comparison with Haywood

The Nelkin (1960) model for hydrogen scattering has been used due to its extensive use in reactor and basic studies and its success in predicting integral, though not differential, scattering effects.

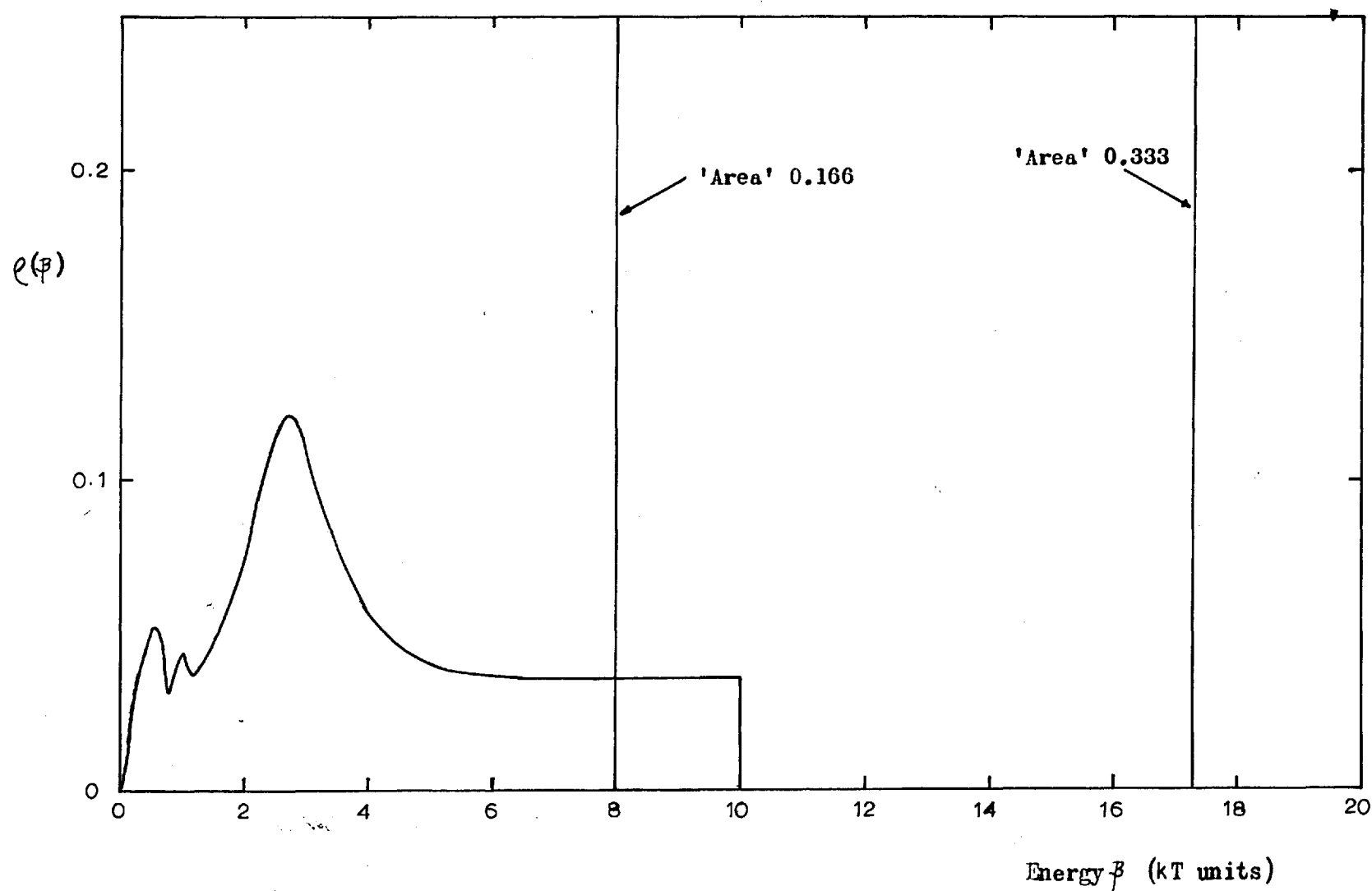


Figure 16 The Haywood Model for $\rho(\beta)$,
The Spectral Density of Normal Modes of H-Atom Vibration

Molecular hindered rotation is approximated by a single harmonic oscillator at 0.06 eV and vibrational motion by one oscillator at 0.205 eV and two degenerate oscillators at 0.48 eV. Motion of the molecule as a whole is represented by a free gas of mass 18. The method of Nelkin was used as modified by Frederici and Goldman (1962) and programmed by Johnson in the code DIS. Energy transfers are classified into the ranges 0 - 0.205 eV, 0.205 - 0.48 eV and 0.48 eV - ∞ . In each range one mode (the .06 eV mode in the first region) is treated exactly. All lower modes are treated in the free gas model limit and all higher in the elastic limit. It is of interest to compare predictions obtained using this approach with those obtained with the 'exact' method proposed by the author in Section 2.3 (iii). An $S(\alpha, \beta)$ matrix for comparison with the 'exact' data was generated using the DIS code in an identical $35\beta \times 30\alpha$ form. For these two matrices calculated diffusion lengths in water differed by only 0.3% and the total cross section by, at the most, 0.4% over the energy range 0.02 eV - 0.08 eV.

A modified Nelkin (hereafter denoted MN) model has been constructed in which the delta-function representing hindered rotations in $\ell(\beta)$ is broadened in an arbitrary symmetric manner to form a distributed peak of the same weighting (Figure 17). This modification is similar to one proposed independently by Koppel (1966) who has used a Gaussian form for the broadened line. We have seen that broadening of this hindered rotation line is the main feature of the experimentally based Haywood model. As the modified $\ell(\beta)$ now has a continuous part, together with the two δ -functions representing vibrations, LEAP-ADDELT was used to calculate $S(\alpha, \beta)$. LEAP-ADDELT will only calculate the 'bound' part of the scattering law $S_b(\alpha, \beta)$, and in order to include the free gas mass 18 translations in the modified model a programme BOND was specified by the author and P. W. Johnson and programmed by

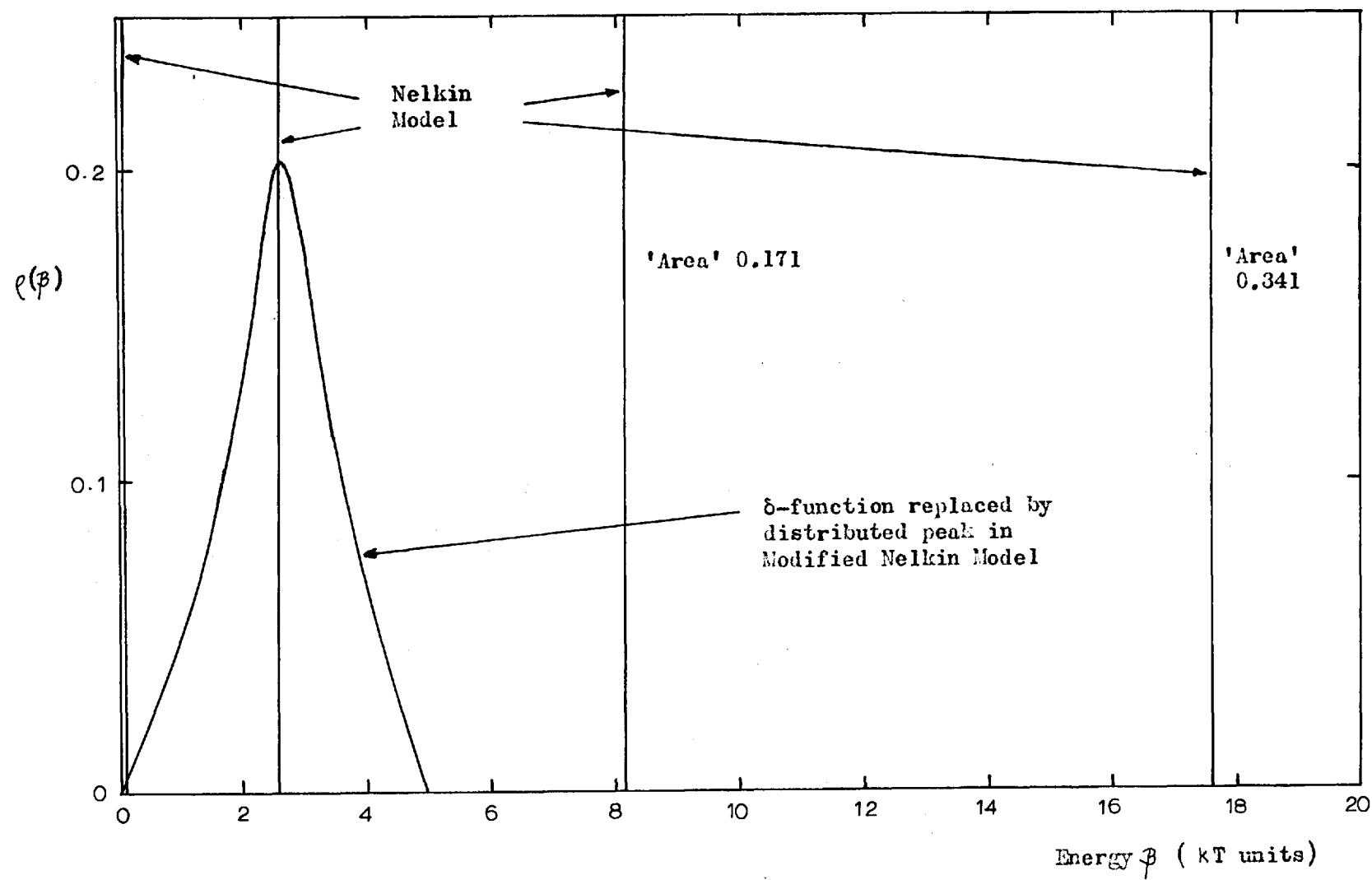


Figure 17 Spectral Density $\rho(\beta)$ for Nelkin and Modified Nelkin Models

the latter to perform the convolution

$$S(\alpha, \beta) = e^{-\alpha\lambda} S_g(\alpha/18, \beta) + \int_0^\infty S_g(\alpha/18, \beta') S_b(\alpha, \beta - \beta') d\beta' \quad (12)$$

by numerical integration. $S_g(\alpha/18, \beta)$ is the free gas model scattering law (with the α scale reduced by a factor of 18 to give the correct mass) and $e^{-\alpha\lambda}$ the Debye-Waller factor.

A third comparative model, 'modified Haywood' (hereafter denoted MH) was chosen to have some features of the Haywood model, but to have mass 18 translations added, so that the only difference (apart from a small difference of the upper vibrational mode) between MN and MH models is in the shape of the continuous part of $e(\beta)$. Again BOND was used to convolve a free gas $S(\alpha, \beta)$ with the Haywood $S(\alpha, \beta)$. A gas of mass 17 was used initially and the combined $e(\beta)$ then had an area of 1.055. Now the normalisation to unity of $e(\beta)$ is equivalent to a normalisation of inverse mode masses. As the dimensionless momentum transfer, α , is inversely proportional to the particle mass (equation (1)), the normalisation to unity was achieved by increasing the α scale of the $S(\alpha, \beta)$ matrix by 5.5%. The translational mass thereby became $17 \times 1.055 = 18$.

4.4 Self-Diffusive Motion

In Sections 4.2 and 4.3 the quasi-elastic scattering has been treated in two ways, by the Debye-Waller factor only and also according to the free gas mass 18. A partial study has been carried out to investigate a model which represents the process of self-diffusion more realistically. Several models have been proposed, ranging from the simple Fick diffusion model (Vineyard, 1958) to the 'jump diffusion' model (see, for example, Singwi and Sjölander (1960)) where a molecule oscillates within an aggregate for a certain mean time and then diffuses for a second specified mean time. We have used, however, a model due

to Egelstaff and Schofield (1962) which, while not representing the physical processes in such detail, can be fitted to experimental data and has been expressed in terms of the dimensionless variables α and β .

Egelstaff and Schofield propose that the function $W(t)$, proportional to the mean square displacement of the molecules at time t , be given for diffusion by:-

$$W_d(t) = \frac{2d}{\pi} \left\{ (t^2 + c^2 + \frac{1}{4})^2 - c \right\} \quad (13)$$

c is a measure of the time delay during which the molecules oscillate around their initial position before diffusing away. The $\rho(\beta)$ derived from (13) is

$$\rho(\beta) = \frac{4d}{\pi} (c^2 + \frac{1}{4})^{\frac{1}{2}} \sinh \beta/2 K_1 \left\{ (c^2 + \frac{1}{4})^{\frac{1}{2}} \beta \right\} \quad (14)$$

The 'area' under this $\rho(\beta)$ is $\frac{d}{c}$ and the inverse of this ratio, the 'mass', gives the size of the cluster of atoms that affects the short time motion of an individual molecule. The scattering law may be obtained from (14) analytically.

We have assumed a value of the self-diffusion coefficient for water of $2.13 \text{ cm}^2 \text{ sec}$ which was obtained by Simpson and Carr (1958) and is close to the value of $2.17 \text{ cm}^2 \text{ sec}$ obtained by Pope and Nation (1965). These values are within the spread of a number of other measurements. A plot of the half width at half maximum $((\Delta\beta)_{\frac{1}{2}})$ of the quasi-elastic peak as a function of α is linear at low α values and

$$(\Delta\beta)_{\frac{1}{2}} = 2d\alpha \quad (15)$$

as is the case for Fick diffusion for all α (d is a dimensionless form of D). At larger α values experimental half widths fall below this line due to the retention of molecules for a period with clusters. We have fitted the Egelstaff-Schofield model to this non-linear part by adjusting the free parameter θ . Using for D $2.13 \text{ cm}^2 \text{ sec}$, the equivalent dimensionless parameter d in (15) is 0.034 and in order

to reproduce the non-linear part of the measurements of Pope and Nation a value of c of 30 was chosen. Thus the ratio $c/d \approx 1,000$ indicating a cluster of ~ 50 molecules; these figures must be treated with great reserve on account of the simple model and fitting procedure. However, it is clear that the effective mass is large compared to the free molecular mass.

It has been noted that Haywood quotes Schofield to the effect that the 'area' of the diffusive part of $e(\beta)$ must be less than 0.5% (equivalent to a mass of 200) and Haywood does not take account of diffusive motion. This is legitimate as his work does not involve energy transfers less than $\beta = 0.1$ and differences between quasi-elastic models are not important above this energy. However, elastic scattering is important in the calculation of integral quantities such as the diffusion length.

An attempt has been made to convolve the self-diffusion model with the Haywood $S(\alpha, \beta)$ using BOND and to use the resulting $S(\alpha, \beta)$ to calculate diffusion lengths. The convolution was successful, but the diffusion length calculation was not, a partial reason being that the angular integration to give the Legendre moments of the scattering kernel (see the next Section) involves an α -integration from $\alpha = 0$ to $4E/kT$. Figure 18 compares the elastic $S(\alpha, 0)$ for Haywood with three different quasi-elastic scattering models and the reason for the difficulty becomes clear. For the self-diffusion model $S(\alpha, 0)$ varies approximately as $\frac{1}{\alpha}$, compared with $\frac{1}{\sqrt{\alpha}}$ for the free gas model. A second equally acute problem is that the half-width of the elastic peak as a function of β is $2d\alpha$ for the diffusion model. While the peak is wide for large α , it resembles a δ -function for small α and the energy integration (effectively an integration in β) becomes difficult to perform. The numerical methods used here do not appear to be capable of handling realistic models of quasi-elastic scattering.

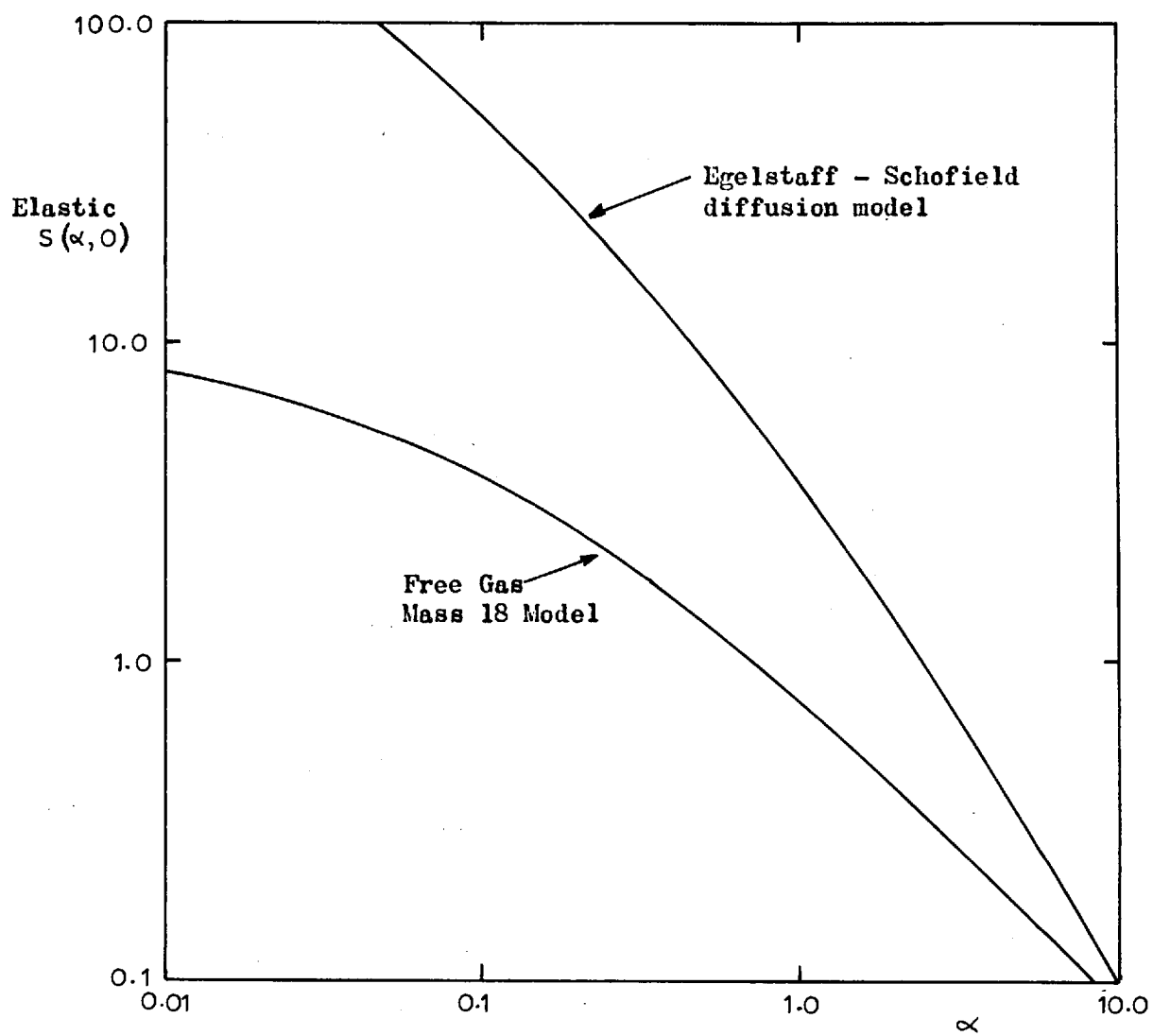


Figure 18 Quasi-Elastic Scattering $S(\alpha, 0)$ for Haywood Model Combined with Two Different Water Molecule Translation Models

4.5 Calculation of Diffusion Lengths

The method used to obtain diffusion lengths has been outlined in Section 3.4 and described in detail by Johnson (1966). It involves the calculation of the Legendre moments of the scattering kernel

$$\sigma_n(E_0, E) = 2\pi \int_{-1}^1 \sigma(E_0, E, \mu) P_n(\mu) d\mu \quad (16)$$

This integral is carried out by double Gaussian quadrature;

$\sigma(E_0, E, \mu)$ is obtained from equation (1), the required values of $S(\alpha, \beta)$ being obtained by a linear interpolation of $S(\alpha, \beta) e^{\beta/2}$ in α and β . For the basic Haywood model the elastic scattering is the Debye-Waller factor and there is an additional contribution to the n th moment of the scattering kernel.

$$c_n(E_0) \delta(E_0 - E) = \frac{\sigma_b}{2} \int_{-1}^1 e^{-\alpha\lambda} P_n(\mu) d\mu \delta(E - E_0) \quad (17)$$

In the case of the basic Nelkin model $\sigma(E_0, E, \mu)$ is calculated directly using the analytical method outlined in Section 4.3. Throughout this work scattering by oxygen is represented by a free gas of mass 16.

The diffusion length, the fundamental space eigenvalue of the one-dimensional neutron transport equation

$$\left\{ \mu \frac{\partial}{\partial z} + \Sigma_T(E) \right\} \phi(z, E, \mu) = \int_{-1}^1 d\mu' \int_0^\infty dE' \Sigma_s(E', E, \mu_0) \phi(z, E', \mu') \quad (18)$$

where μ_0 is the cosine of the angle between μ and μ' , is evaluated using the ' B_L ' method proposed by Honeck (1962). Fluxes were computed at 32 energy points covering the energy range 0 - 0.625 eV. Ten calculations using the $L = 2$ approximation were performed for each of the three absorbing solutions containing boron, cadmium or gadolinium. These ten calculations were equally spaced in absorber concentration between the pure water case and the Σ_T (min) limit (Corngold (1964)) above which no fundamental space eigenvalue exists. The hydrogen

2200 m sec⁻¹ cross section was taken to be 0.332 b after Stehn et al (1964); this cross section was increased to 0.337 b for the cadmium cases to allow for the small, known, boron contamination. An earlier analysis of this type has been published by Goddard et al. Since this work was published the calculations for boron have been extended and those for cadmium and gadolinium repeated using more recent cross section data.

Considering first absorption by the non-¹/v absorbers, both calculated and experimental (Johnson) values of κ^2 ($\kappa \equiv 1/L$) were fitted to a series expansion

$$\kappa^2 = \beta_0 + \beta_1 N - \beta_2 N^2 \quad (19)$$

where $N(\text{cm}^{-3})$ is the absorber number density and the coefficient β_0 represents absorption by hydrogen (and small quantities of boron in the cadmium solutions) and is κ^2 for pure water. The coefficient β_2 represents the effect of spectrum distortion. To demonstrate the effect of diffusion cooling and heating, Johnson has suggested a rearrangement of the series expansion in the form

$$\frac{\kappa^2 - \beta_0}{\Sigma_{00}} = \beta_1 / \sigma_{00} - \frac{\beta_2}{\sigma_{00}^2} \Sigma_{00} + O(\Sigma_{00})^2 \quad (20)$$

where Σ_{00} is the product of the non-¹/v absorber density and its microscopic absorption cross section averaged over a Maxwellian spectrum σ_{00} . Plotting $(\kappa^2 - \beta_0)/\Sigma_{00}$ against Σ_{00} yields a graph whose slope demonstrates the effect of diffusion heating or cooling.

A puzzling feature of the comparison already published was that when plotted according to (20), experimental cadmium absorption data was in good agreement with the predictions of the MN model while for gadolinium and boron experiment lay between predictions of the MH and MN models. Additional evidence comes from the so-called Laguerre polynomial analytic approach in which the flux is represented by a

Maxwellian modified by a series of Laguerre polynomials. Johnson has extended this method to non- $1/v$ absorbers and, in particular, has shown that $\beta 1/\sigma_{00}$, the intercept of the plot of equation (20), is given by $\frac{1}{D_{00}}$ (where D_{00} is the Maxwellian averaged diffusion constant for water) minus a small extra term. This term is $\sim 2.8\%$ for gadolinium and $\sim 0.5\%$ for cadmium and when equation (20) is plotted the gadolinium intercept should lie below the cadmium. This was true for the calculated curves, but for experiment the cadmium intercept was 3.2% below the gadolinium, indicating an inconsistency between true and assumed cross sections of these isotopes of $\sim 5\%$.

For the above study both gadolinium and cadmium absorption cross section were taken from BNL-325 (1st ed.). Wydler (1967) has suggested that an arbitrary normalisation of $\sim 7\%$ applied to the cadmium data of Rainwater (1947) in making this compilation, was not justified. Recently Widder and Brunner (1968) have performed a very detailed series of total cross section measurements which confirm the data of Rainwater, and the Widder data, together with that of Hohne (1961) at very low energies, has been used in the calculations shown in Table XIII and Figure 19. For this data σ_{00} is 2841 b compared with 2986b for the old BNL-325 data.

A new correlation of the gadolinium absorption cross section has been prepared by Goldberg et al (1966) and this has been used in the present calculations for gadolinium poisoned water. The new σ_{00} is 35680 b compared with 37520 for the old data. Fitted series constants for both calculation and Johnson's experiments at 22.3°C are given in Table XIII and illustrated in Figure 20.

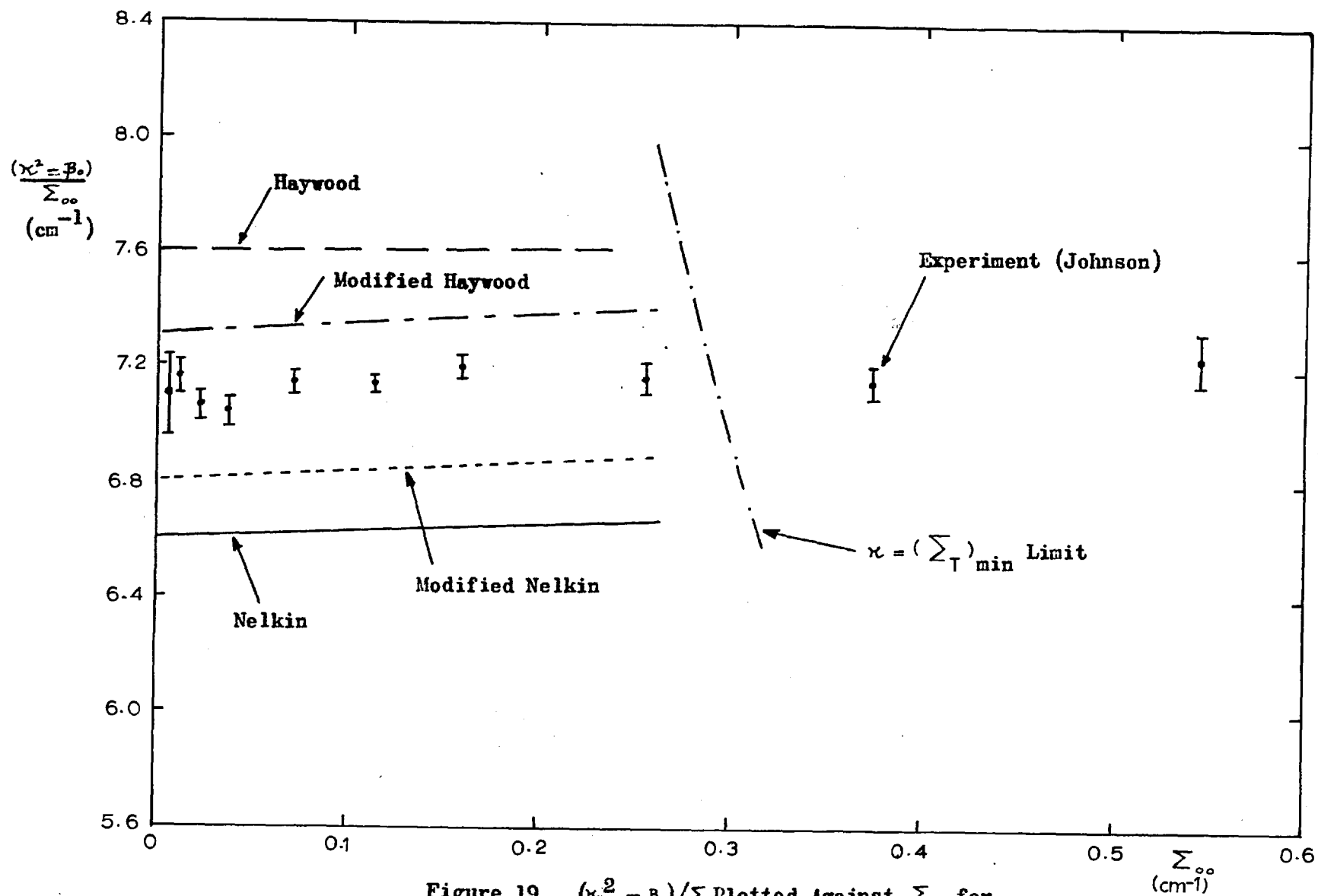


Figure 19 $(\kappa^2 - \beta_0) / \Sigma_{\infty}$ Plotted Against Σ_{∞} for Cadmium Sulphate Solutions at 22.3°C

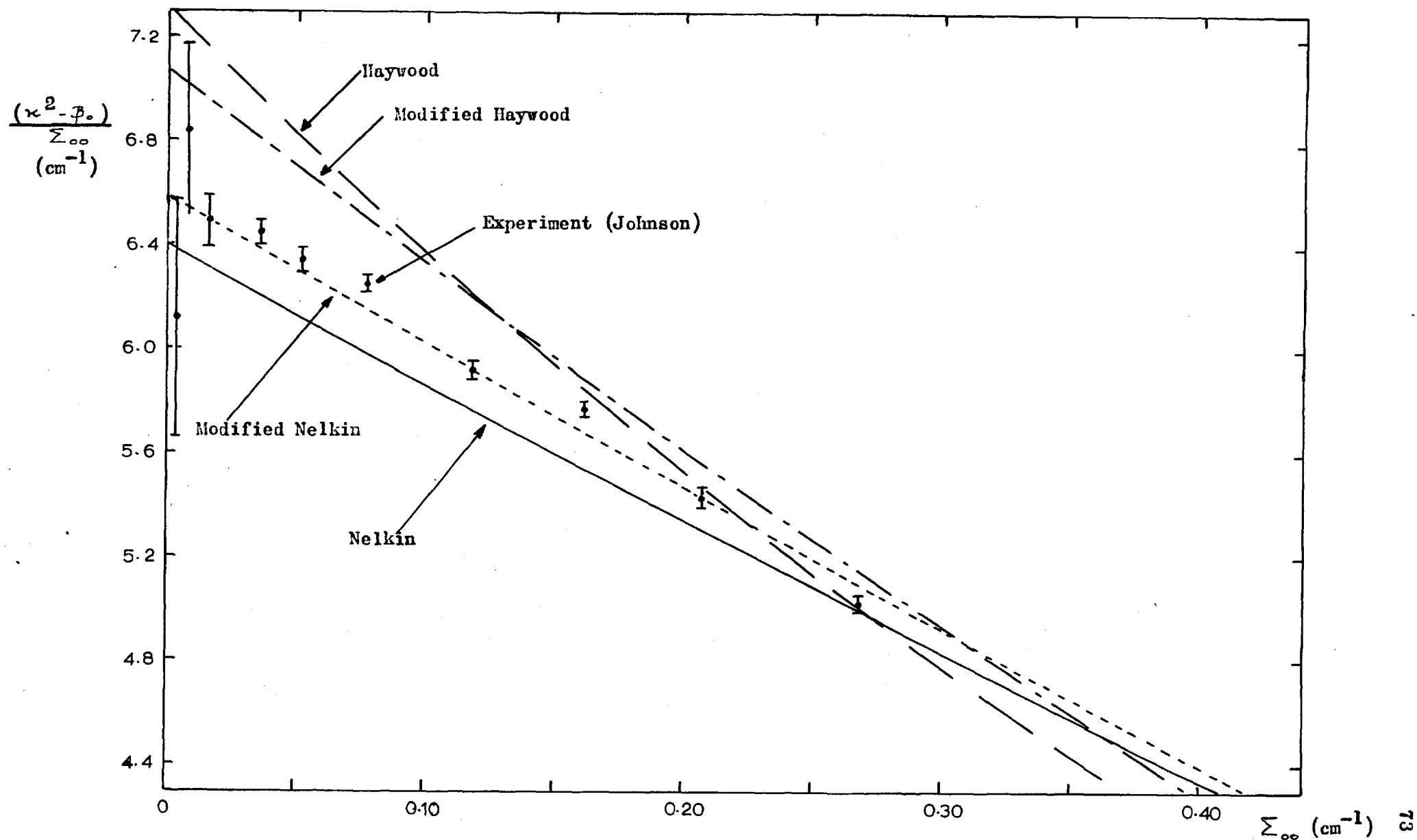


Figure 20 $(\kappa^2 - p_0) / \Sigma_{\infty}$ Plotted Against Σ_{∞} for Gadolinium Nitrate Solutions at 22.3°C

Table XIII Non- $1/v$ Coefficients For Water at 22.3°C

Expt./Model	Element	σ (cm ⁻²)	1×10^{19} (cm)	2×10^{38} (cm ⁴)
Experiment	Cd	.1356 $\pm$.0005	.202 $\pm$.002	-.0002 $\pm$.0001
Haywood	Cd	0.1490	0.2160	-0.00003
Mod. Haywood	Cd	0.1429	0.2075	-0.00033
Mod. Nelkin	Cd	0.1331	0.1928	-0.00037
Nelkin	Cd	0.1293	0.1874	-0.00026
Experiment	Gd	.1315 $\pm$.0002	2.49 $\pm$.02	.86 $\pm$.03
Haywood	Gd	0.1466	2.747	1.39
Mod. Haywood	Gd	0.1407	2.662	1.10
Mod. Nelkin	Gd	0.1309	2.473	0.81
Nelkin	Gd	0.1273	2.402	0.77

Turning near to $1/v$ absorption, the series expansion is

$$\kappa^2 = \alpha_1 \sum_{a0} - \alpha_2 \sum_{a0}^2 + \alpha_3 \sum_{a0}^3 \quad (21)$$

where $\sum_{a0}$ is the sum of the 2200 m sec⁻¹ macroscopic absorption cross sections for boron and hydrogen and the α 's are coefficients to be fitted. The magnitude and sign of the coefficient α_2 indicates the effect of diffusion cooling or heating. Fitted coefficients at 22.3°C are given in Table XIV where the more familiar pulsed parameters have been obtained through the expressions quoted by Honeck (1962).

Table XIV $1/v$ Coefficients For Water at 22.3°C

Experiment	T(°C)	D_0 (cm ² sec ⁻¹)	C (cm ⁴ sec ⁻¹)
Johnson	22.3	35620 $\pm$ 230	2800 $\pm$ 260
Starr and Koppel	21	35850 $\pm$ 100	2900 $\pm$ 350
Küchle	22	35400 $\pm$ 700	4200 $\pm$ 800
Calculation			
Haywood	22.3	32660	3750
Mod. Haywood	22.3	34203	2960
Mod. Nelkin	22.3	36820	2920
Nelkin	22.3	37850	3230

The rearrangement used for graphical presentation is

$$\kappa^2 / \sum_{a0} = \alpha_1 - \alpha_2 \sum_{a0} + \alpha_3 \sum_{a0}^2 \quad (22)$$

and if $\kappa^2 / \sum_{a0}$ is plotted vs. $\sum_{a0}$, then the gradient demonstrates the effect of diffusion heating. This is done for the calculated value and the experimental values of Johnson in Figure 21.

From being below that of gadolinium, the intercept β_1 / α_{00} for cadmium is near 7% greater, compared with the expected $\sim 2.3\%$. Comparing the diffusion constants D_0 for boron and β_1 for cadmium and gadolinium with the experimental values, the latter lie between the predictions of the MH and MN models. There is an indication that as the amount of spectrum distortion increases, from cadmium through boron to gadolinium, then experiment agrees better with the MN model. The effect of spectrum distortion is contained in the constants C and β_2 . β_2 for cadmium is not sufficiently well-defined to permit a comparison with theory, but it is shown without doubt to have a negative value, indicating diffusion cooling. The experimental values of C for boron of Johnson, which he has shown to be in good agreement with the typical static value of Starr and Koppel (1962) but lower, though only $1\frac{1}{2}$ standard deviations less, than the typical pulsed value of Kluchle (1960), agree within the experimental errors with the predictions of the MN and MH models, but is significantly less than that of the Haywood model. For gadolinium β_2 lies between the predictions of the MH and MN models.

4.6 Total Cross Section and Mean Kinetic Energy

Valuable confirmation of the adequacy of scattering models can be obtained by studying the related quantities total cross section and mean kinetic energy per bound proton $\bar{K}$. Russell, Neill and Brown (1966) have recently published total cross section measurements for H_2O which, while agreeing with the earlier measurements of

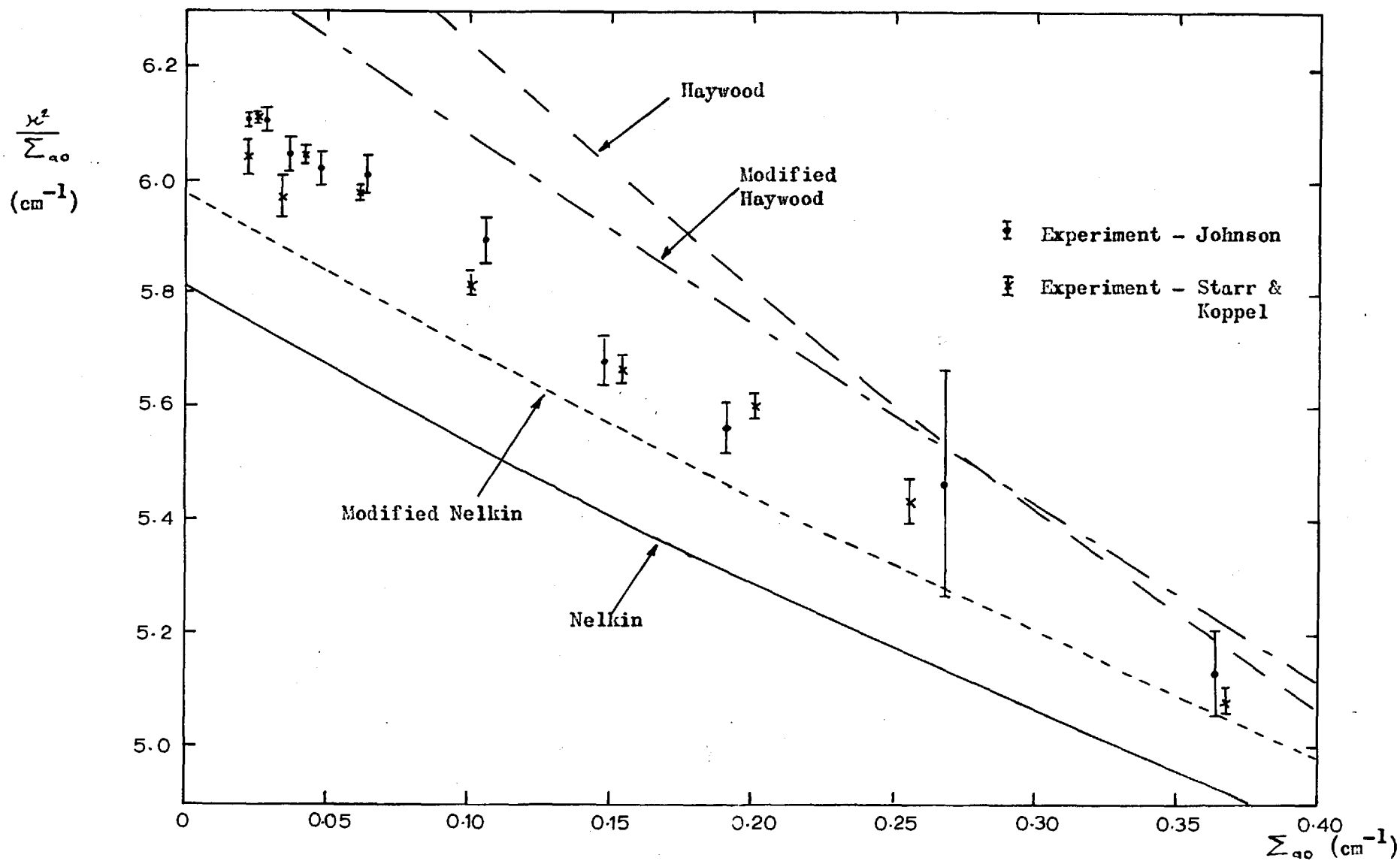


Figure 21 $\frac{n^2}{\sum_{a_0}}$ Plotted Against $\sum_{a_0}$ for Boric Acid Solutions at 22.3°C

Melkonian (1949) within the errors of the latter, are more precise and more closely spaced in energy. They have also obtained a new value for $\bar{K}$.

In re-calculating the total cross section for the four models for hydrogen scattering, a particular effort has been made to reduce numerical errors. Most important, the number of energy points has been increased to 64 from 32. This change had little effect on the total cross section below 0.06 eV, but produced detailed improvements in the form, compared with the published curves of Goddard et al, above this energy. Figure 22 compares calculation with the measurements of Russell et al. Around 0.025 eV the experimental values lie between the MH and the Nelkin-type models. The Nelkin model drops below experiment by $\sim 5\%$ above 0.04 eV and then rises above experiment due to energy loss scattering through the discrete mode at 0.06 eV. Broadening this line in the MN model gives good agreement. Haywood and MH predictions are both high above ~ 0.02 eV.

By fitting the total cross section in the high energy 'joining' region to the Placzek asymptotic expansion, Russell et al have determined a new value for the mean kinetic energy per bound proton $\bar{K}$. In practice we work with $\bar{T} = 2/3 \times \bar{K}$ which is equal to kT for a free proton gas. In terms of kT ,

$$\bar{T} = \frac{1}{2} \int_0^{\infty} \beta^2 \coth(\beta/2) \rho(\beta) d\beta \quad (23)$$

In Table XV the values of $\bar{T}$ for the various models are compared with the value of Russell et al.

Table XV Mean Kinetic Energy Per Bound Proton
For Models Compared with Experiment

Model/Experiment	T (eV)
Experiment Russell et al	0.1173 ± 0.0068
Haywood	0.1172
Mod. Haywood	0.1111
Mod. Nelkin	0.1102
Nelkin	0.1168

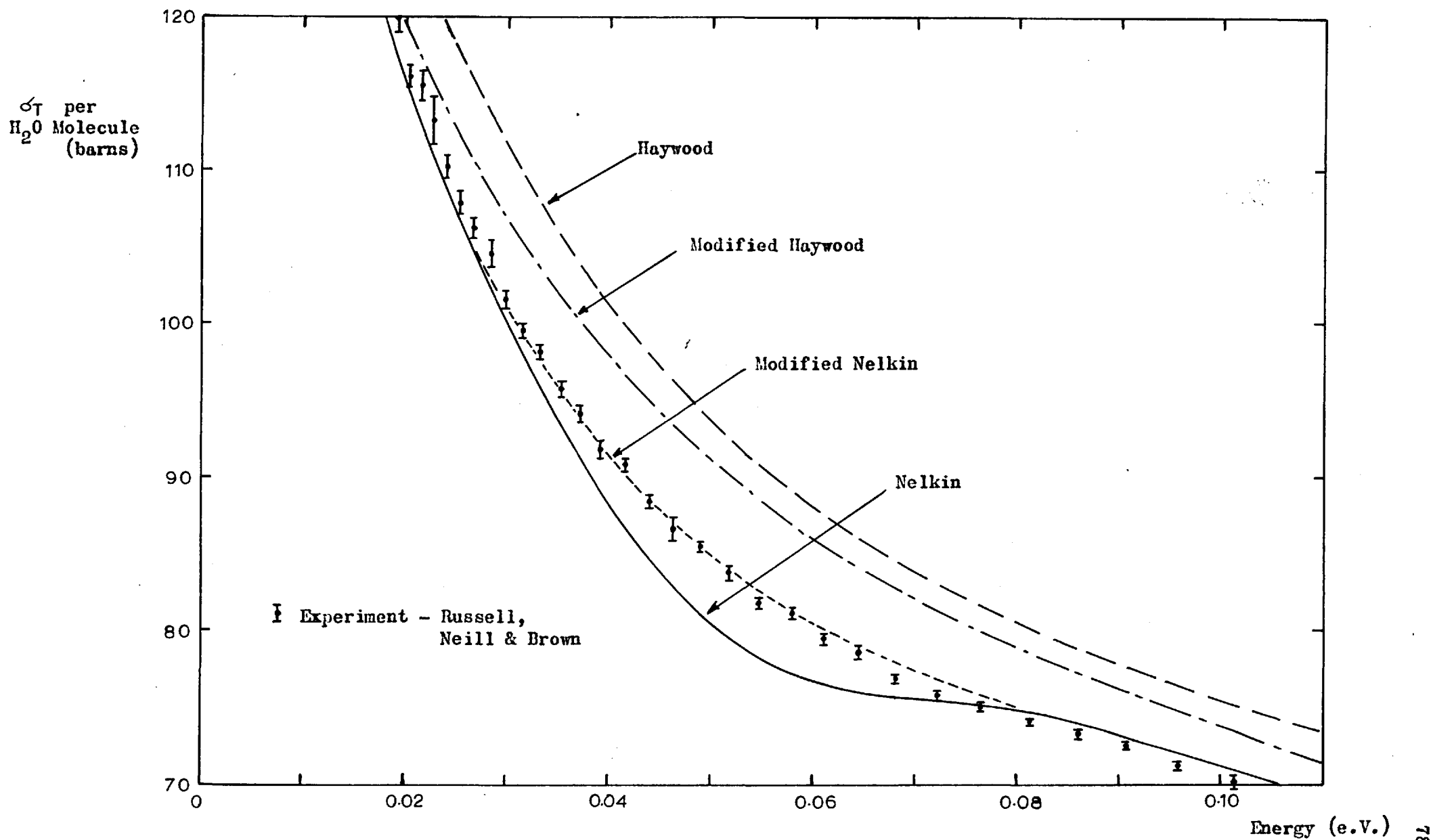


Figure 22 H_2O Total Cross Section; Comparison of Calculation With Measurements of Russell, Neill and Brown

The Haywood model was adjusted to give agreement with a measured $\bar{K}$ value and this is seen to have been successful. Modifying Haywood to include mass 18 translations reduces $\bar{K}$ below measurement. Both Nelkin and MN models agree with experiment within the assigned error. While achieving the correct $\bar{K}$ is a very important constraint, $\bar{K}$ is not a sensitive parameter for low thermal effects, mainly because $\bar{K}$ depends on the high β parts of $\rho(\beta)$. Compared with the MN model, the area displaced in $\rho(\beta)$ from the rotational peak appears around $\beta = 7$ in the Haywood model and so will not greatly affect $\bar{K}$ ($\bar{K} = 6.9$ in kT units for Haywood).

CHAPTER 5

THE RETHERMALISATION PROCESS

In this chapter features of chapters 2, 3 and 4 are brought together in the discussion of the rethermalisation problem. It must be accepted that some important points have not been resolved completely, but these points are indicated and suggestions are made for future work. In chapter 2 the sensitivities of fission and reaction rate ratios in a cold, clean, SGHW lattice to changes in the hydrogen and deuterium scattering models were studied. An operating SGHW power reactor will, among other differences, have hot coolant at $\sim 270^{\circ}\text{C}$ and the fuel cluster spectrum will now depend more strongly upon the rate of exchange of energy between the coolant and the 'softer' neutron spectrum entering from the cold moderator. This rate of exchange will influence the manner in which the spectrum approaches the asymptotic spectrum for the cluster. In order to study this 'rethermalisation' process we have taken scattering laws studied in detail in chapters 3 and 4, namely the Haywood model for light water and the model proposed by the present author for Santowax-R, and applied them in an analysis of the two-temperature experiment of Tremeer (1966). This experiment combined cold water and hot Santowax-R in slab geometry. Essentially the same numerical method of calculation has been used in the analysis presented here as was used for the cell calculations described in chapter 2. By using pure moderators and simple geometry the rethermalisation aspect should be isolated in both experiment and calculation. A second part of this work has involved the re-analysis of the experimental results of Tremeer in an attempt to verify the existence or otherwise of a single discrete 'rethermalisation length' for Santowax-R.

The analysis of the temperature discontinuity problem was initiated

by Kottwitz (1960) who used the heavy gas scattering model and proposed a solution in terms of an effective neutron temperature. Selengut (1958) introduced the two overlapping thermal group model and Leslie (1962) has discussed the calculation of rethermalisation cross sections between such overlapping groups. The work of Kottwitz has been extended by De Ladonchamps and Grossman (1962) and Kuscer^{VV} (1962). Pearce and Kennedy (1964) have performed 40-group diffusion theory calculations and obtained good agreement with the model of Selengut. Using the heavy gas model, Williams (1965) has solved the exact neutron transport equation to give the mean neutrons energy as a function of position near a plane temperature discontinuity. He shows that a transport transient may be expected near to the discontinuity in addition to the rethermalisation transient. In a second paper relevant to this work Williams (1966) has studied the existence of space eigenvalues of the transport equation using the free gas model. For a free proton gas he concludes that no space eigenvalues other than the fundamental exist (i.e. there is no discrete rethermalisation length) and that the first higher eigenvalue only exists for $A \gtrsim 2$. Saastamoinen (1967) has also used an analytical approach and has assumed the flux to be the sum of two overlapping components. However, these fluxes were assumed to be angle-dependent and for light water the transport equation was solved by the Pn method with $n = 3$ and the different Legendre components of the rethermalisation flux were compared. These differed markedly in their transient behaviour and Saastamoinen links this with the fact that all violate the $(\sum_{\tau})_{\min}$ limit. A numerical approach has been used by Day (1967) who has analysed, with the WDSN Code, his time of flight measurements of the spectrum in differentially heated graphite blocks.

Although this chapter is concerned with the analysis, rather than the conduct of experiments, it is relevant to consider other experimental

work in addition to that of Tremeer, which will be discussed in the next section. Bennett (1963) has reported measurements in an annular arrangement of graphite and water placed at the centre of a reactor core. This experiment appears to be subject to uncertainties in the overall slowing down neutron source. Day in his work already referred to, has reported measurements which represent the culmination of a series of experiments using differentially heated graphite. These used a well-defined thermal source originating at the LIDO reactor and entering the experiment perpendicular to the interface. In the series of experiments performed at General Atomic (see, for example, Dardes, Neill and Houghton (1964)) neutron spectra near to poison and temperature discontinuities in graphite and beryllium were measured by the time of flight method. A pulsed electron beam falling on a uranium target provided a fast neutron source external to the experiment. In the initial work of Saastamoinen (1966) the provision of time-of-flight beam extraction resulted in very poor definition of spectrum changes near to the discontinuity between water tanks. Recently Saastamoinen (1967) has reported time-of-flight measurements in water close to an interface with hot zirconium hydride. The reported measurements were restricted to a verification of the space dependence of the transient flux angular distribution referred to above.

In Section 5.1 the experimental arrangement used by Tremeer in measuring the fission rates of Plutonium-239 and Uranium-235 near a plane temperature discontinuity is discussed. A detailed re-analysis of the form of the spectrum transient in Santowax-R is presented in Section 5.2; evidence for the existence of a discrete rethermalisation length in this medium is discussed. The methods used in performing transport theory calculations of fission rates, together with the scattering models used and the representation of the experimental conditions are described in Section 5.3. Some comparison between

calculated and experimental fission rate distributions are also presented in this Section. A preliminary analysis of the experiments of Tremeer have been published (Tremeer, Goddard and Grant (1966)) and recently Goddard, Grant, Johnson and Tremeer (1967) have carried the analysis further.

5.1 Rethermalisation Experiments at Imperial College

Let us first consider the design requirements for an experimental study of the rethermalisation problem. These are that the rethermalisation transient be readily isolated and measured with sufficient accuracy for meaningful comparison with one-dimensional transport theory. All design features arise from these requirements.

The source-free case of no flux gradient far from the interface and no absorption, originally studied theoretically by Kottwitz, cannot be realised in practice and the need for an external neutron source brings with it the problem of overall flux gradients, which will be particularly severe with hydrogenous moderators. Attempts by Saastamoinen (1966, 1967) to reduce the overall gradient thermal column are not regarded with favour by the present author as the overall flux shapes in the water are not easily measurable and no measurements are reported. The most satisfactory arrangement is that used by Day and by Tremeer; the overall neutron current flow is perpendicular to the temperature discontinuity and can therefore be taken account of in a one-dimensional theory. A reactor has been used as a neutron source by both Day (LIDO) and Tremeer (CONSORT). Now the transverse dimensions of both the neutron source and the experimental tanks are linked by the need for a fundamental transverse flux mode in the tanks which means that transverse bucklings of both source and tanks must be matched. The transverse dimensions are fixed by the requirement that the transverse leakage correction be small and for this correction to be made

easily the overall flux should be harmonic-free. Finally, the source neutron spectrum should be close to the equilibrium spectrum in the tank nearest to the source.

The experiment of Tremeer was installed on a face of the London University 100 Kw reactor CONSORT II, in a high flux cave separated by 82 cm of graphite from the reactor tank. The fast flux entering the experiment as a percentage of the total flux was 16% and a negative source technique was used to simulate a near Maxwellian source. An aluminium reinforced cadmium shutter could be lowered to close a square aperture, matching the size of the experimental tanks, in a cadmium screen. Collisions in the shutter gave the negative source a small fast component.

Two tanks fabricated from 0.16 cm aluminium contained cold water and hot Santowax. A 0.32 cm gap between the tanks was maintained by 0.48 cm diameter insulating discs fixed to the cold tank face. The cold tank was 10.3 cm long and the hot tank 25.4 cm long, both were 61 cm wide and were filled to a depth of 61 cm with the liquids. The mismatch of the transverse bucklings of the tank and the main 95.2 cm square graphite neutron feed meant that harmonic corrections had to be considered; this has been discussed in Chapter 3. Demineralised water in the cold tank was kept close to room temperature by circulation through a heat exchanger while the Santowax temperature was automatically controlled using hot plates with a total output of 5.1 Kw beneath the tank. The sides and rear face of the tanks were covered with cadmium sheet and thermal insulation.

A miniature fission chamber which could be moved horizontally by a remotely operated screw drive was used and a second vertical screw drive together with a wiper seal to remove liquid enabled the chamber to be transferred from one tank to the other. A horizontal positional reproducibility of 0.013 cm was achieved in the rethermalisation experiment.

Two pairs of U-235 and Pu-239 chambers were available, mounted in 0.32 cm and 0.3 cm outside diameter gas-filled Nimonic tubes of such a length that the centre of the active regions were at the tank centre line. The fissile coatings, which were of 0.5 cm and 0.25 cm diameters had thicknesses of 1 mgm/cm^2 for U-235 and 0.3 mgm/cm^2 for Pu-239.

Fission chamber gain changes with temperature between cold water and hot Santowax tanks were determined by the method of graphical superposition of attenuation bias curves. The greatest correction applied was 1.5%. A constant systematic error of approximately 2 mm in the position of the 0.3 cm detectors in the hot tank has been shown to be due to temperature induced internal gas pressure changes straightening out small bends in the rather flexible counter stems. A possible error in the negative source technique arose through radiative capture γ -rays from cadmium giving photo-fission counts in the measuring fission chambers. From the change in counting rate produced by lowering the cadmium screen in front of an additional boron screen it was shown that the photo-fission contribution must be less than 4.9% of the fast flux or typically 1% of the difference counting rate observed in the rethermalisation experiment.

Unpublished experimental work by Petr (1967) has used the same experimental tanks but with a number of important differences. Diphenyl was substituted for light water in the cold tank, a greater thickness of graphite between the neutron core and the tanks reduced the fast flux and a special cadmium insert in the thermal column gave a source with the correct transverse flux shape. The amount of aluminium in the shutter was reduced to cut down any fast component of the difference flux. To avoid the systematic position errors associated with the fission chambers and to obtain greater spectral sensitivity, lutecium and manganese foils were used, mounted in foil holders

positioned rigidly against the temperature interface. This work has not yet been analysed completely.

5.2 Analysis of Spectrum Transients in Santowax-R

The re-analysis of Tremeer's experimental results has been made in the light of the theoretical work of Williams and Saastamoinen already mentioned. Their work has thrown doubt upon the existence of a discrete rethermalisation length when the scattering is by protons and its existence is not certain for light water, in spite of the increase in H atom effective mass due to molecular binding. We have already noted Williams' conclusion that as the mass of a free gas increases then more discrete space eigenvalues appear. Now Tremeer has fitted the two overlapping group model to his measured fission rate distributions and has obtained typical values for κ_1 the inverse transient (rethermalisation) relaxation length of $\sim 2.5 \pm 0.2 \text{ cm}^{-1}$ (at 20°C) for H_2O and $\sim 1.33 \pm 0.04 \text{ cm}^{-1}$ (at 200°C) for Santowax-R. These values should be compared with the corresponding values of $(\sum_\tau)_{\min}$ which are 1.48 cm^{-1} for H_2O and 0.948 cm^{-1} for Santowax-R; both limits are violated. For Santowax $\sum_\tau$ falls below the value of the above fitted κ_1 , at $\sim 0.14 \text{ eV}$ and above this energy the transient relaxation length should deviate from the apparent discrete value and be influenced by the $\sum_\tau$ variation down to the limiting $(\sum_\tau)_{\min}$ value. Thus it may be expected that the transient flux will not be separable in space and energy and the aim of the analysis has been to detect this.

The approach to the analysis has been influenced partly by known systematic errors in Tremeer's measurements and partly by the relevant theoretical work. The principal systematic error was the uncertainty in the fission chamber positioning although it must be emphasised that reproducibility and the distance between counter positions in

each tank were known to a high degree of accuracy. This temperature induced bending of the $1/8$ " detector stems meant that the cold tank and the hot tank measurements were not properly related to each other, and analysis of the hot tank points alone was a solution to this difficulty. Rethermalisation effects in the H_2O tank occurred over too short a range to be analysed accurately.

A result of theory which must influence the analysis is the conclusion of Williams that there will be a 'transport transient', near to the interface, in addition to the main rethermalisation transient. It may be appreciated readily that, at a short distance inside the hot medium, the transient flux perpendicular to the interface will have a higher mean energy than that travelling parallel to it. Williams shows, for a heavy gas of mass 20, that the transport transient has a shorter range than the main 'diffusion theory' transient. For lighter scattering atoms, where a neutron may reach equilibrium at a single collision, then the transport transient may extend over the whole range of the rethermalisation process. Such a situation has been studied theoretically by Saastamoinen for light water; he finds that the Legendre components of the transient flux have very different relaxation lengths, indicating that the angular distribution is changing continuously. A complete experimental approach to the problem would thus involve a study of the separability of the transient flux with respect to the space, energy and angular variables. In the case of Tremere's results we are restricted to a comparison between measurements with the two fissile isotopes Pu-239 and U-235 evidence of space/energy separability and to a study of goodness of fit to an exponential transient for rather indirect evidence for space/angle separability. If a second eigenvalue exists then beyond a certain distance from the interface, close to which transport and higher spectrum transients might be significant, a fission rate distribution in the hot tank

might be represented by

$$F(r) = A_1 e^{-\gamma_1 r} + A_2 e^{-\gamma_0 r} + A_3 e^{\gamma_0 r} \quad (24)$$

where γ_0 and γ_1 are the inverse relaxation lengths of the fundamental and transient components.

In the method of non-linear fitting used the function was linearised by expanding it in a Taylor series in the unknown parameters about initial guessed values. Application of the least squares criterion led to a set of normal equations involving the corrections to the initial values of the parameters. This process was repeated until accuracy criteria were satisfied.

Convergence was not obtained when fitting expression (24) to the experimental data owing to the small influence at the measuring positions of leakage from the back face of the hot tank. The data were then fitted to

$$F(r) = A_1 e^{-\gamma_1 r} + A_2 \left(e^{-\gamma_0 r} - e^{-2\gamma_0 H} \cdot e^{\gamma_0 r} \right) \quad (25)$$

where H is the extrapolated thickness of the hot tank. By fitting expression (25) to the experimental data using different fixed values of H it was shown that the value of H expected from the physical dimensions of the hot tank was close to the value indicated by the goodness of fit.

Table XVI shows the values of the fitting parameter χ^2 for various values of $\exp(-2\gamma_0 H)$, denoted z , corresponding to different values of H . In five cases the minimum χ^2 occurs near Z^* and Z was therefore fixed at these values for all subsequent fitting.

Table XVI Effect Upon χ^2 of Varying Extrapolated Thickness

Expt. No.	(Z) χ^2	(Z*) χ^2	(Z) χ^2	(Z) χ^2
	(.000134)	(.000056)	(.00003)	(.000014)
G170	53.0	46.4	46.4	47.1
H170	35.5	25.4	23.5	22.8
J170	40.3	32.8	30.9	30.0
K170	-	11.3	11.1	11.3
	(.000182)	(.0000825)	(.000045)	(.000022)
G200	20.5	11.4	10.5	10.7
H200	24.0	24.0	26.2	28.1
J200	24.7	23.2	23.7	24.3
K200	56.0	29.3	29.9	30.6
	(.000348)	(.000125)	(.00008)	(.000035)
G230	53.8	20.4	17.3	15.7
H230	23.9	12.8	13.9	16.3
J230	28.4	35.3	-	41.7
K230	33.7	27.7	29.9	33.4

Table XVII shows the results of fitting expression (25) to the experimental data. For the first points included in the fit the detector centres were 0.4 cm and 0.5 cm from the interface for 0.62 cm and 0.3 cm detectors respectively. For the same Santowax-R temperature there are no systematic differences in the fitted γ_0 , either with fissile isotope or detector size. The values of chi-squared when compared with the theoretical χ_0^2 corresponding to the number of degrees of freedom and a 5% level of significance show that the differences between experiment and expression (25) were not significantly greater than the experimental errors.

Comparing the fitted values of γ_1 there are significant differences between Pu-239 and U-235 experimental results obtained using 0.62 cm detectors. These results must however be treated with some caution because of the large detector diameter.

Table XVII Experimental Fundamental and
Transient Relaxation Lengths

Expt. No.	Detector dia. (cm) /Isotope	Hot Tank Temp. (°C)	γ_0 (cm ⁻¹)	γ_1 (cm ⁻¹)	χ^2	χ_0^2
G170	0.62 U	171.7	.1827 $\pm$.0004	1.35 $\pm$.05	22	26
H170	0.62 Pu	170.5	.1806 $\pm$.0005	1.17 $\pm$.07	25	29
J170	0.3 U	170.5	.1811 $\pm$.0007	1.16 $\pm$.07	27	25
K170	0.3 Pu	171.0	.1810 $\pm$.0005	1.03 $\pm$.08	10	25
G200	0.62 U	201.7	.1740 $\pm$.0003	1.25 $\pm$.03	12	29
G200A	0.62 U	202.4	.1754 $\pm$.0006	1.32 $\pm$.05	33	26
G200B	0.62 U	201.9	.1732 $\pm$.0004	1.25 $\pm$.04	22	29
H200	0.62 Pu	200.1	.1727 $\pm$.0005	1.12 $\pm$.08	24	29
J200	0.3 U	198.8	.1738 $\pm$.0006	1.13 $\pm$.05	16	25
K200	0.3 Pu	200.5	.1722 $\pm$.0006	1.00 $\pm$.10	17	25
G230	0.62 U	232.9	.1666 $\pm$.0004	1.28 $\pm$.04	21	29
H230	0.62 Pu	231.7	.1639 $\pm$.0005	1.19 $\pm$.05	5	20
J230	0.3 U	232.4	.1653 $\pm$.0006	1.14 $\pm$.05	20	25
K230	0.3 Pu	231.9	.1659 $\pm$.0005	1.24 $\pm$.14	21	25

With the 0.3 cm detectors the resulting γ_1 values are less than those obtained with the larger detectors and the differences between Pu-239 and U-235 detector results are not greater than the experimental errors. The errors were greater because of inferior counting statistics.

To detect any deviation from an exponential form of the transient which might not be revealed by the goodness-of-fit test, up to 8 points were dropped next to the interface and the fit repeated. No systematic changes in either γ_0 or γ_1 were observed. Typical results are shown in Figure 23.

In an effort to explain the apparent differences between values of γ_1 for 0.62 cm and 0.3 cm detectors, numerical integrations of the fission rate around the counter circumference were performed, assuming

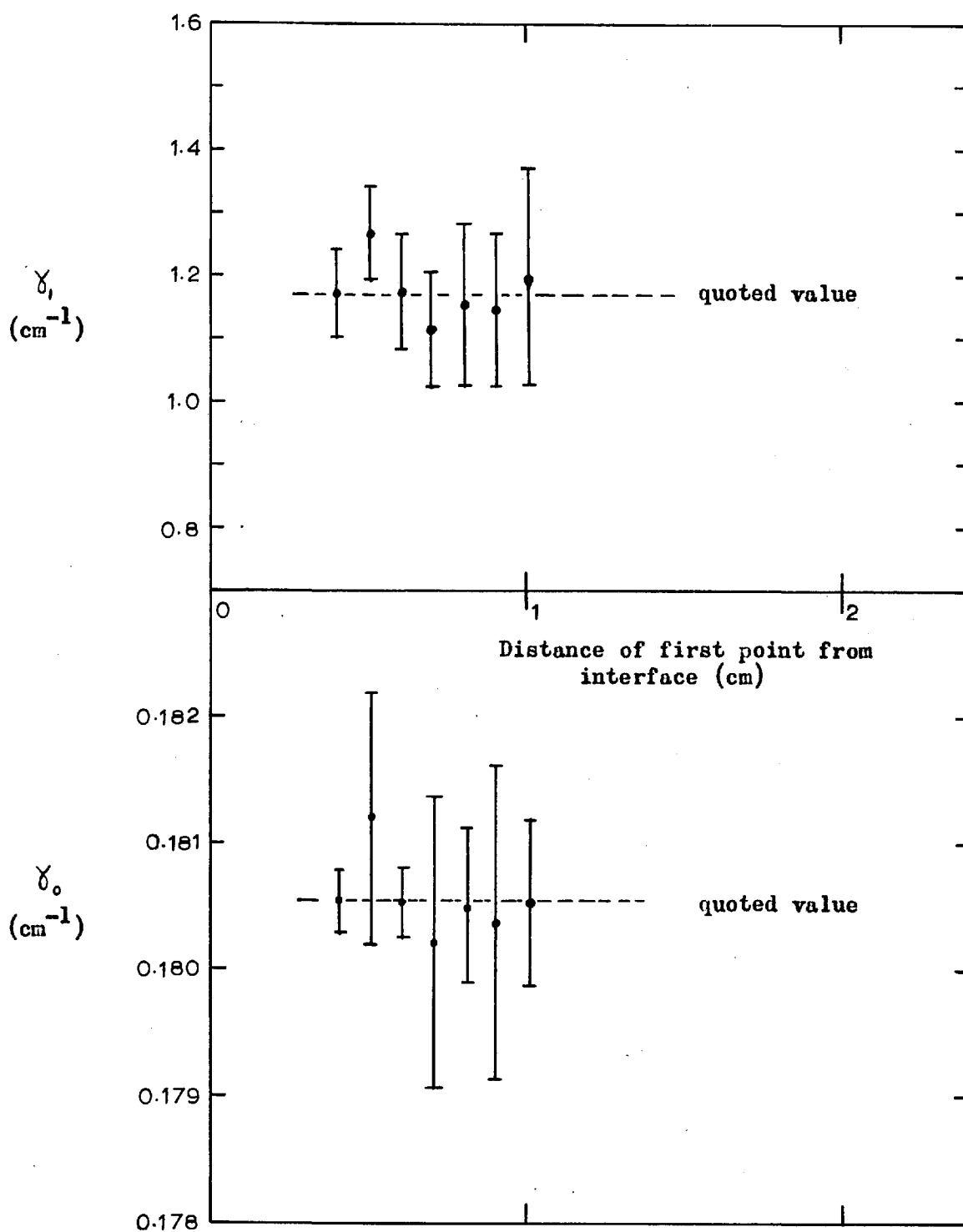


Figure 23 Typical Effect Upon γ_0 and γ_1 for Santowax-R of Dropping Interface Points from the Fit.
H-170 Data, 0.62 cm Pu-239 Chamber.

the overall flux distribution to be the initial fitted one. Corrected fission rates were then fitted to equation (25) but without any significant change in the γ_1 values. The differences observed are most likely to result from moderator displacement and the resulting neutron streaming, rather than from the size effect alone.

Two further aspects of the measurements need to be considered; these have implications for the reliability of the transient relaxation lengths quoted in Table XVII and for the design of experiments of this type. In Section 3.4 it was noted that a 5% harmonic content in the experimental neutron source was a strong possibility, although Tremear did not consider this. In order to check the likely effect of harmonics upon transient relaxation lengths the data were fitted to a modified form of equation (25)

$$F(r) = A_1 e^{-\gamma_1 r} + A_2 (e^{-\gamma_0 r} - e^{-2\gamma_0 H} \cdot e^{\gamma_0 r}) - 0.05 A_2 e^{-r \cdot \text{sqrt}(\gamma_0^2 + B)} \quad (26)$$

The constant B is $16 \left(\frac{\pi}{a} \right)^2$ where a is the tank extrapolated width and $B = 0.0408 \text{ cm}^{-2}$.

Table XVIII Effect Upon γ_1 of Harmonic Contamination

Expt. No.	$\gamma_0 (\text{cm}^{-1})$	$\gamma_1 (\text{cm}^{-1})$	% change in γ_1
G170	.1851	1.30	-3.7
H170	.1830	1.10	-5.8
J170	.1832	1.11	-4.4
K170	.1828	0.95	-8.2
G200	.1761	1.20	-3.8
G200A	.1776	1.26	-4.2
G200B	.1753	1.21	-3.0
H200	.1748	1.03	-7.9
J200	.1759	1.08	-4.0
K200	.1743	0.92	-8.2
G230	.1686	1.23	-3.8
H230	.1659	1.14	-3.6
J230	.1673	1.10	-3.5
K230	.1678	1.12	-10.3

While the small difference between plutonium and uranium transients and between the 0.62 cm and 0.31 cm transients are, in general, maintained the typical change in γ_1 is of the same order as these differences. This undesirable state of affairs has been rectified in the recent experiments of Petr where a harmonic-free flux distribution was obtained.

It has been noted that collisions in the shutter gave the negative source a small fast component. Purely on the basis of the total scattering cross sections of the cadmium and its aluminium backing, 10% of fast neutrons were scattered in the shutter. However, the difference flux will only contain those neutrons thereby removed from the experiment, rather than those scattered into the experiment or back into the graphite. It has been assumed that one-third of the scattered neutrons are removed and the effect of this upon the experimental flux distributions has been simulated by reducing the measured shutter-down flux distributions by 3% and repeating the fitting procedures. Table XIX shows that, if the assumption of one-third of scattered neutrons lost is reasonable, which it may be because of the presence of an air gap between the graphite thermal column and the experiment tank, then this may be a significant systematic error in Tremere's measurements. In the subsequent measurements of Petr, more graphite was used in the thermal column and the major part of the aluminium was removed from the shutter.

Table XIX Effect Upon γ_1 of Additional Fast Neutrons in the Source

Expt. No.	$\gamma_0 (\text{cm}^{-1})$	$\gamma_1 (\text{cm}^{-1})$	% change in γ_1
G170	.1860	1.43	+ 6.0
H170	.1841	1.29	+ 11.5
J170	.1845	1.25	+ 8.1
K170	.1844	1.18	+ 11.4
G200	.1772	1.32	+ 5.5
G200A	.1788	1.41	+ 7.0
G200B	.1765	1.31	+ 5.1
H200	.1762	1.26	+ 12.6
J200	.1769	1.19	+ 5.3
K200	.1754	1.13	+ 13.0
G230	.1697	1.34	+ 4.7
H230	.1659	1.22	+ 2.5
J230	.1682	1.19	+ 4.6
K230	.1690	1.42	+ 14.5

5.3 Calculation of Fission Rates

With the object of calculating fission rates in as rigorous a manner as possible, a numerical solution of the transport equation has been used in contrast to the analytical methods used by Williams and Saastamoinen. These numerical methods are the same as those used in the cell calculation described in Chapter 2, and the aim has been to test these methods in plane geometry, with homogeneous regions, against Tremeer's measurements.

To deal first with the scattering models for the materials in Tremeer's experiment, the free gas model with isotropic scattering has been used for oxygen and carbon in the two moderators and for aluminium in the two interface walls. The scattering law representation of equation (1) in Chapter 2 has been used. Calculations have been performed for the three experimental temperature combinations, 171°C/20°C, 201°C/20°C and 232°C/20°C. An attempt has been made to use both the free gas model for hydrogen scattering and bound models. However, the free gas model transport theory calculations were not successful for reasons to be discussed later. The Haywood model as described and studied in Chapter 4 was used as the bound model for hydrogen in cold water. The spectral density distribution is shown in Figure 16. As before, the LEAP-ADDELT programme was used to generate matrices of $S(\alpha, \beta)$ values on cards. The method of generating bound models for hydrogen in Santowax-R at 171, 201 and 232°C has been described in Chapter 3. Spectral density distributions were obtained by interpolating and extrapolating linearly, on an energy (e.V.) scale, the models fitted by the present author to the measurements of Brugger. Linear interpolation and extrapolation was used because on an e.V. scale the curves of $\rho(\beta)$ fitted to Brugger's measurements differed by less than the likely systematic errors. Figure 11 shows the three spectral density distributions plotted against dimensionless energy transfer β .

A special group structure was devised for these calculations, based upon the 55 group structure studied in Section 2.2. By eliminating the dense cluster of groups around 1 e.V. and ending the group structure at 1 e.V. a 31 group set was obtained which is illustrated in Figure 2. As the problem is entirely thermal a slowing down source need not be considered and the number of neutrons above 1 e.V. will be negligible. Point fission cross sections for uranium-235 and plutonium-239 were taken from the Winfrith Data File. Multigroup fission cross sections were obtained by numerical integration by computer; the effect of changing the effective temperature of the Maxwellian weighting spectrum was investigated and found to be negligible. The chosen weighting spectrum had a temperature which was the mean of cold and hot tank temperatures.

Again following the procedures outlined in Chapter 2, the programme PIXSE was modified and run on the Imperial College 7090 computer. It was divided into two parts; the first, PIXSCAT, was used to write microscopic 31 group scattering matrices for all the elements concerned onto magnetic tape. The second part of the programme, PIXED, was used to edit this tape to give macroscopic region cross sections suitable for input to the transport theory programme WDSN. Now the version of this programme which was available was restricted to isotropic scattering and in order to improve the representation of overall flux gradients the transport correction was applied. This has been referred to in Chapter 2. In this correction the total anisotropic scattering cross section is subtracted from the diagonal (elastic) scattering cross section.

$$\sigma_{g \rightarrow g}^1 = \sigma_{g \rightarrow g} - \sum_{g'} \sigma_{1g \rightarrow g'}$$

The scattering is assumed to be linearly anisotropic. As we have noted, this correction gives the correct diffusion coefficient and leaves the spectrum as calculated in an infinite homogeneous medium

unchanged. It appears to the author that this approximation may give errors when the coolant of an SGHW is hot or in the temperature discontinuity experiment. The anisotropy in the energy change cross sections is ignored and therefore it is possible that the spatial rate of energy change (or rethermalisation) will be too great.

In order to reduce the long running times of the WDSN code for this problem, the thicknesses of the water and Santowax tanks were reduced to 6.6 cm and 22 cm. The S_4 approximation was used. Fifty space sub-regions were used in the calculation as illustrated in Figure 24. A neutron source having a 20°C Maxwellian energy distribution was assumed in the innermost three sub-regions in water, distributed spatially in an approximate first collision distribution. The effect of transverse leakage was not included as a DB^2 absorption term because this was a negligible correction to the short transient relaxation lengths. In retrospect, however, its inclusion would probably have improved the rate of problem convergence. For the calculation using bound models for hydrogen scattering in water and Santowax, the convergence of the transport theory calculation was subject to two tests. First a comparison of fluxes after 40 minutes and 60 minutes running times on the computer; this, however, is not necessarily a reliable test. The test which was regarded as conclusive was that the asymptotic flux distributions away from the interface should have an exponential form and should have relaxation lengths which agreed with the results of eigenvalue calculations.

Fission rates of U-235 and Pu-239 were obtained by editing an output tape from WDSN with the aid of the Winfrith programme WED. A typical calculated U-235 fission distribution is shown in Figure 24. Both transient and asymptotic distributions can be seen together with the effect of leakage from the outer Santowax boundary. Calculated fission rate ratios

$$\frac{(\text{Pu-239/U-235})_{\text{experiment}}}{(\text{Pu-239/U-235})_{20^\circ\text{C Maxwellian}}}$$

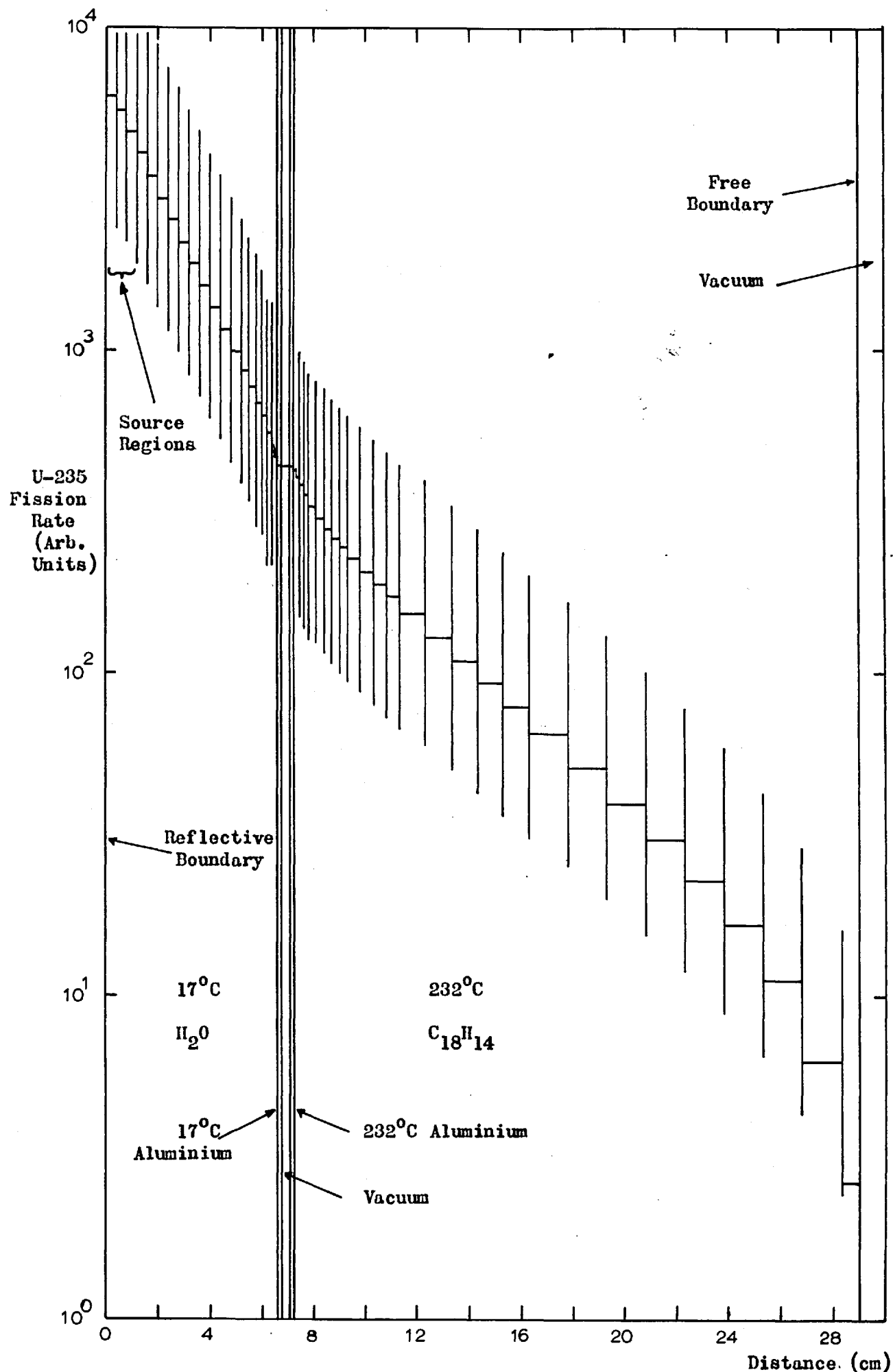


Figure 24 Space Mesh Intervals Used in Calculation and Typical Calculated U-235 Fission Rate Distribution

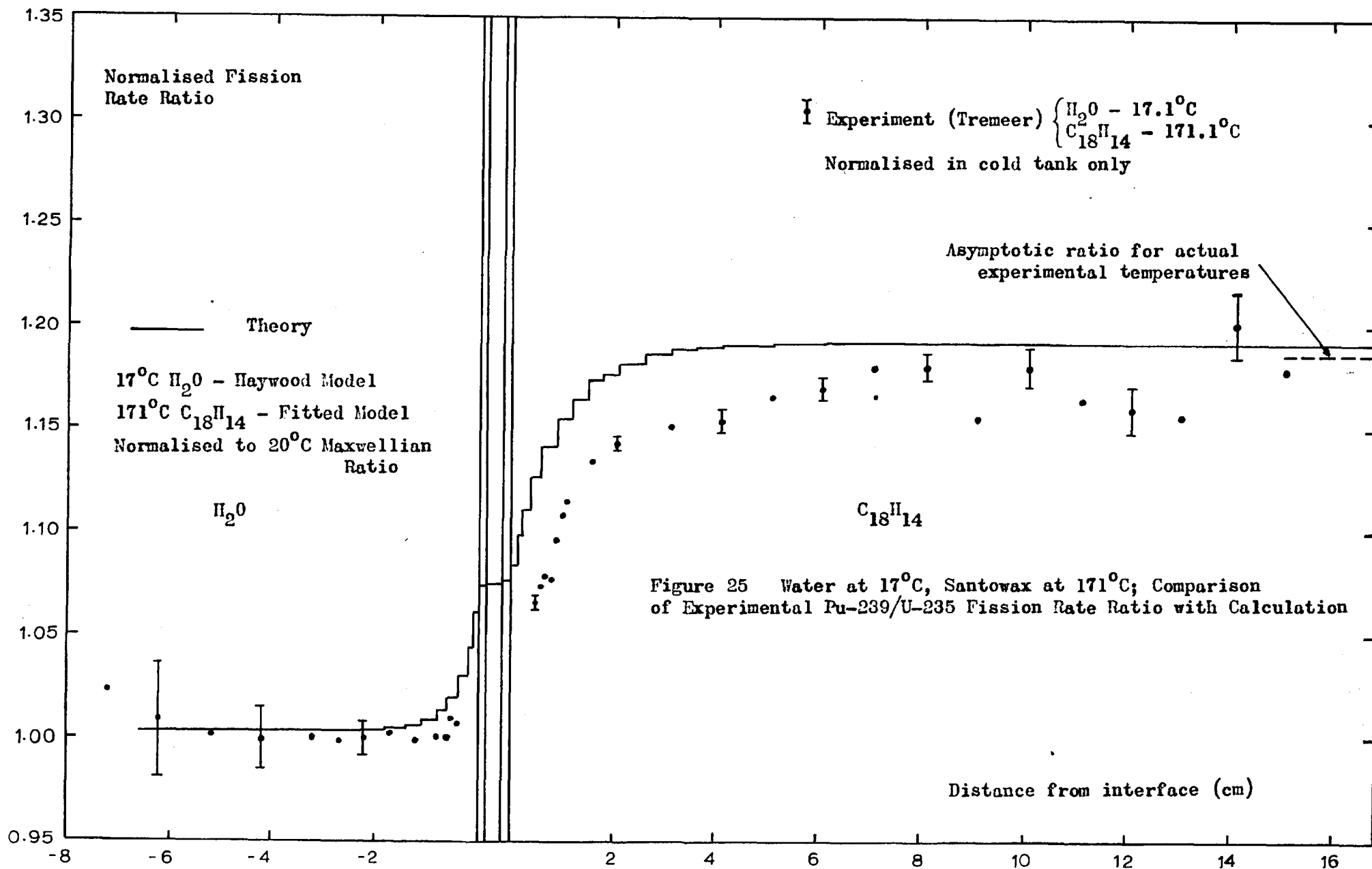
are compared with Tremereer's measurements using the 0.62 cm detector in Figures 25 to 27. A comparison with the 0.3 cm measurements was not possible because of the position uncertainties already referred to. The form of the transients are well represented although the experimental points in Santowax-R tend to lie below theory close to the interface. There is a strong possibility that this feature is due to neutron streaming across the large (0.62 cm) fission chambers.

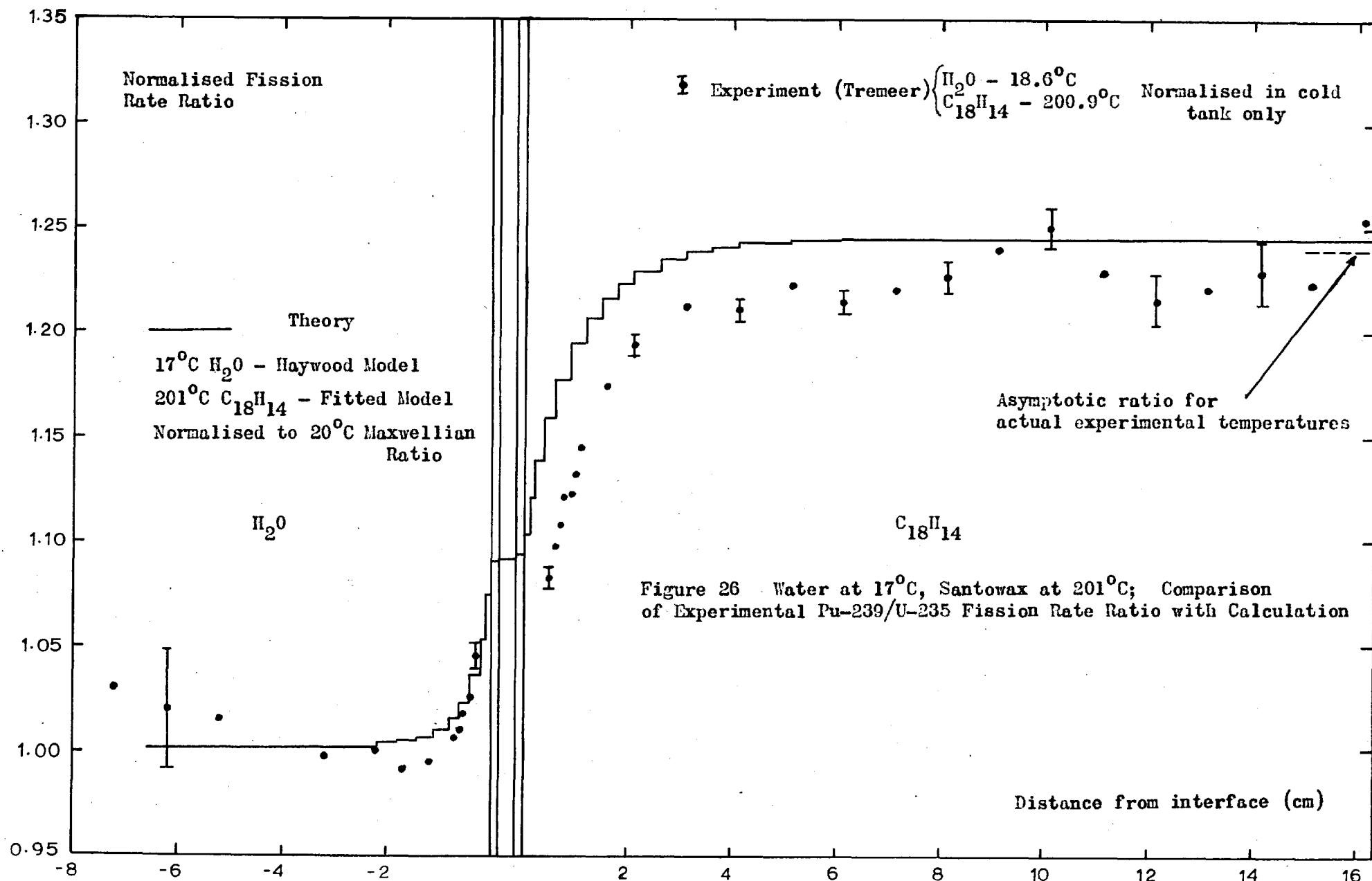
As with the experimental points, an attempt was made to fit a sum of three exponentials given by equation (25) to the calculated fission rates in the Santowax. A study was again made of the effect of dropping points next to the interface upon the fit. Contrary to the result of this study with the experimental data, a slow increase in the transient relaxation length was observed with point dropping. The inverse transient and fundamental relaxation lengths given in Table XX below correspond to the first point in the fit being at the same distance from the interface as was the first experimental point.

Table XX Calculated Fundamental and Transient
Relaxation Lengths

Santowax Temperature (°C)	Fissile isotope	γ_0 (cm ⁻¹)	γ_1 (cm ⁻¹)
171	U	0.184	1.13
	Pu		1.04
201	U	0.171	1.13
	Pu		1.03
232	U	0.159	1.12
	Pu		1.03

Fitting sums of exponential functions is notoriously difficult and the variation in the transient relaxation length might have been due to deviations of the asymptotic part from the hyperbolic sine form either due to incomplete convergence or to, more probably, the use of too wide finite difference sub-regions in the outer parts of the





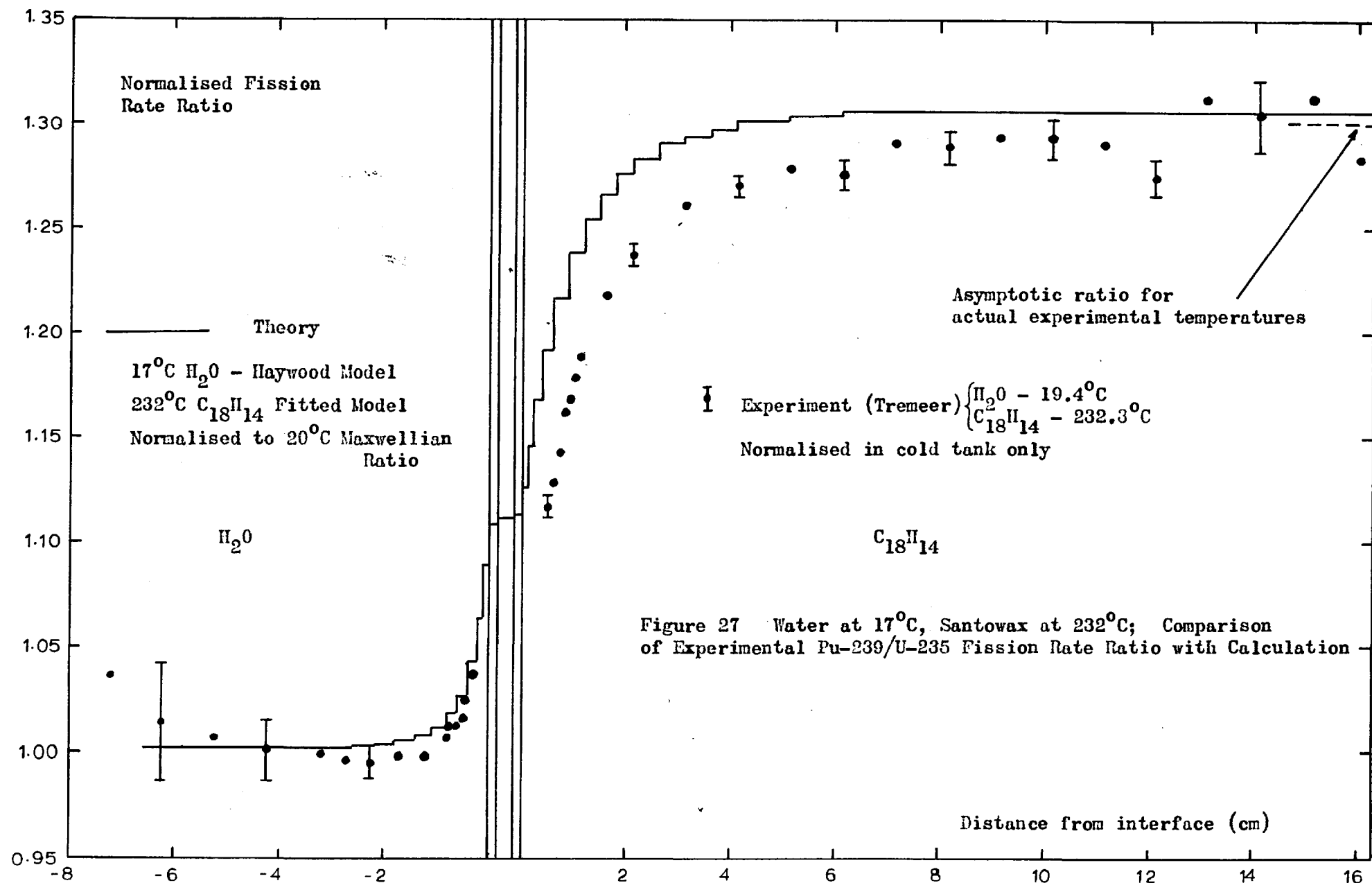


Figure 27 Water at 17°C , Santowax at 232°C ; Comparison of Experimental Pu-239/U-235 Fission Rate Ratio with Calculation

Santowax tank. There is an obvious conflict between the WDSN assumption of a linear flux variation across sub-regions and the overall exponential flux distribution. Very narrow intervals could not be used as this would result in very long running times. The difference between fitted transient relaxation lengths is of the same order as that observed (with rather doubtful statistical significance) for the experimental measurements.

No comparisons between bound and free gas hydrogen scattering model calculations was possible because WDSN calculations with free gas models failed to converge even after running times of ~ 1 hour. In a recent publication Engle and Mynatt (1968) comment that the flux will converge in a few iterations in or near source regions, but, depending on the proportion of neutrons lost by absorption or leakage at each iteration, several hundred iterations may be required to converge at a point many mean free paths from the source. They propose an iteration scheme which is claimed to reduce the running times for penetration problems by an order of magnitude and which, if applied to WDSN, would probably permit free gas model calculations.

CHAPTER 6

CONCLUSIONS

Although the work which has been described is broadly linked together, it does fall naturally into four distinct parts and the results are therefore considered in this form.

6.1 Scattering Law Effects in Cold, Clean SGHW Lattices

Free gas models, effective width models fitted to diffusion coefficients and Nelkin-type models for hydrogen and deuterium have been used in this study of scattering law importance. A new method has been proposed and used to generate cross sections for the Nelkin-type models.

Reaction rate ratios in the fuel, predicted using the effective width models, are very close to those obtained using Nelkin-type models. However, this result may not hold for higher coolant temperatures because it is not obvious how the width model should be generalised to cover all temperatures. There is some evidence that, for 'water-cooled' fuel clusters, the calculated spectra are too 'soft' when the Nelkin model is used. This conclusion must be tentative because of simplifying assumptions made, particularly the neglecting of energy-dependent hyper-fine structure.

From the rather limited evidence available, the width model for deuterium in heavy water or the Nelkin-type model with no corrections for coherent scattering, appear to yield reaction and fission rate ratios in agreement with experiment for cells containing only heavy water moderator, that is 'superheat' cells.

Interaction between flux peaking and fuel fission rate ratios was examined and found to be negligible. The prediction of cell flux peaking is only marginally affected by the choice of scattering model;

changing from a free gas to a width or Nelkin model typically alters the cell flux peaking by $1\frac{1}{2}$ - 2% in the SGHW lattice examined. The effect of these changes in flux peaking on reactivity is small compared with those arising directly from spectrum changes.

The use of the width or Nelkin model gives a harder spectrum than a free gas model and hence a lower predicted reactivity due to the non- $1/v$ behaviour of the U-235 thermal cross section. Typically, the value of K-infinity is reduced by 0.5%. While such changes in reactivity are probably just significant for an enriched SGHW design, they are likely to be very important in a natural-uranium fuelled SGHW where the excess reactivity is small and burn-up must be predicted accurately.

6.2 Scattering Law for Santowax-R

By the use of an iterative fitting method, spectral density distributions of the normal modes of hydrogen atom motion have been derived for solid Santowax-R at 23°C and liquid Santowax-R at 207°C and 267°C. Reduced partial differential cross sections generated from these distributions agree well with the measurements of Brugger (from which they were largely derived), particularly when likely systematic errors in these measurements are taken into account.

Spectral densities for the liquid and solid phases show no significant differences where the fitted energy ranges overlap. By comparing the spectral density for the liquid form with the equivalent results obtained by other workers for diphenyl and benzene, it has been shown that Santowax-R is closer to diphenyl than it is to benzene in its scattering behaviour.

Total cross sections at 0.025 eV calculated for the fitted model are $\sim 10\%$ greater than experiment. There is also a consistent discrepancy of 6% between calculated and the measured diffusion lengths in Santowax-R of Tremeer. A study has been made of the temperature dependence of the diffusion length. Both free gas model and the

Nelkin-type Ardente model have the same trend with temperature.

Over the restricted range $171^{\circ}\text{C} - 267^{\circ}\text{C}$ this trend is confirmed by Tremere's measurements and the calculations with the fitted model.

The trend is markedly different from that exhibited by the measurements of Hohki et al.

6.3 Neutron Scattering by Light Water

Diffusion lengths in cold light water poisoned with boron, cadmium and gadolinium measured by Johnson have been compared with the results of calculation using four different scattering models for hydrogen.

For both $1/v$ and non- $1/v$ absorbers the best agreement with the fitted experimental diffusion constants D_0 and β_1 and the spectrum distortion constants C and β_2 is not obtained with either Nelkin or Haywood models. Rather, experiment tends to lie close to the predictions of the modified Nelkin and modified Haywood models.

The reason why a model such as Haywood, which was fitted to differential measurements, does not predict integral experiments satisfactorily may lie both in the treatment of quasi-elastic scattering and in the form of the generalized frequency distribution. Koppel (1966) has suggested that, at least in the prediction of total cross sections, the difference between the Haywood model calculations and experiment in the energy range 0.02 to 0.10 eV arises from excessive weighting of the region in the frequency spectrum around 0.2 eV. The modified Haywood and modified Nelkin models differ principally at this point in the spectrum as both models have essentially the same treatment of free translational motion and of vibrational motion by two discrete oscillators. This work has been restricted to isotropic models and Koppel and Young have shown that the effect of spatial anisotropy of the hydrogen vibrations acts in such a way as to improve the agreement of the Nelkin model predictions of D_0 and total cross section with experiment. If it does not

prove possible to reconcile models based on differential experiments with integral measurements then it may be necessary to abandon the 'single typical atom' concept and adopt the approach of McMurry et al (1966) which takes account of the existence in water of free molecules and aggregates of differing sizes.

6.4 The Rethermalisation Process

The work presented has served to indicate the direction that future work on the rethermalisation problem should take. With regard to the space/energy separability of the spectrum transient in Santowax-R, apparent differences were observed between transient relaxation lengths for the two fissile isotopes in the case of the large diameter (0.62 cm) detectors only. In order to confirm these differences future experimental work should ensure that the fast neutron content of the flux and the harmonic content is reduced by at least an order of magnitude compared with Tremeer's experiments. Counting statistical accuracy should also be improved by at least a factor of two.

Bound hydrogen multigroup transport theory calculations of the normalised fission rate ratio plutonium-239/uranium-235 were in general agreement with the experimental results obtained by Tremeer with the 0.62 cm detector. There were, however, differences close to the interface which were most likely due to moderator displacement by the detectors. Transient relaxation lengths fitted to the individual calculated fission rate distributions were in better general agreement with the less accurate experimental results obtained with the 0.3 cm detectors. Within the accuracy of these comparisons it appears that the rethermalisation process in an SGHW reactor with hot coolant will be calculated correctly with the methods used. Future theoretical work should be concerned with multigroup methods with a proper treatment of anisotropic scattering.

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APPENDIX A

THEORY USED IN THE ADDELT PROGRAMME

These Appendices draw heavily on the publications of Egelstaff and Schofield and information communicated by McLatchie, who wrote the programmes ADDELT and LEAP.

We will consider only the 'self' or incoherent part of the scattering and neglect effects due to interference between parts of the neutron wave scattered from neighbouring atoms. The physical meanings of the various functions are not easy to envisage, but a qualitative description of each will be attempted. $S(\alpha, \beta)$ the "Scattering Law" measures the probability that, if the atomic system is given a "push" of momentum $\hbar \alpha$ then it will absorb or give up energy $\hbar \beta$. An atom is given this "push" and its energy initially either increases or decreases. Now the atom is coupled to the molecule, and as time passes the momentum concentrated on the initial atom will decrease through the various dynamic connections between the atom and the system, e.g. vibrational or rotational motions. The function which measures the fraction of the initial momentum, which still resides on the initial atom after a time t , is called the intermediate scattering function $I(\alpha, t)$, and due to their physical connection I and S are the Fourier transforms of one another. $I(\alpha, t)$ is defined by

$$S(\alpha, \beta) = \frac{1}{2\pi} \int_{-\infty}^{\infty} I(\alpha, t) \exp(i\beta t) dt \quad (1)$$

The behaviour of $I(\alpha, t)$ is discussed by Schofield who has shown that for small times and low values of α , and for large times, it has the form

$$I(\alpha, t) = \exp(-\alpha w(t)) \quad (2)$$

$w(t)$ (secs) is a decay constant in the usual sense, but in this case its variation is the question of interest. In the so-called Gaussian approximation, equation (2) is held to be true for all values of t . This corresponds to the assumption that the "self correlation function" $G(r, t)$, of which $I(\alpha, t)$ is the Fourier transform, is a Gaussian function of position. $G(r, t)$ may be thought of as describing the probability of the atom being at position r at time t , having started from $r = 0$ at $t = 0$, and with $w(t)$ being proportional to the mean square displacement of the atom at time t .

From equations (1) and (2) we can write for the Gaussian part of the scattering,

$$S(\alpha, \beta) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \exp.(-\alpha w(t)) + i\beta t) dt \quad (3)$$

the non-Gaussian part of the scattering being of the order α^2 . It may be shown that by integrating (3) by parts, and considering the limit of small α , where non-Gaussian terms vanish and the expression becomes exact,

$$\lim_{\alpha \rightarrow 0} \frac{1}{\alpha} S(\alpha, \beta) = \frac{1}{2\pi} \cdot \frac{1}{\beta^2} \int_{-\infty}^{\infty} \ddot{w}(t) \exp(i\beta t) dt \quad (4)$$

A function $p(\beta)$ is now defined, such that

$$p(\beta) = \beta^2 \lim_{\alpha \rightarrow 0} \frac{1}{\alpha} S(\alpha, \beta) \quad (5)$$

In the case of a solid, $p(\beta)$ as defined above is related to the spectral density distribution $\ell(\beta)$ by

$$\frac{2}{\beta} p(\beta) = \frac{\ell(\beta)}{\sinh \beta/2} \quad (6)$$

and this serves to define $\ell(\beta)$ for liquids also.

Let us take equation (4), invert the transform and substitute for $\ell(\beta)$,

$$\ddot{w}(t) = \int_{-\infty}^{\infty} p(\beta) \exp. (-i\beta t) d\beta \quad (7)$$

$$= 2 \int_0^{\infty} p(\beta) \cos \beta t d\beta \quad \text{since } w(t) \text{ is real.} \quad (8)$$

Integrating,

$$\dot{w}(t) = 2 \int_0^{\infty} \frac{p(\beta) \sinh \beta t}{\beta} d\beta \quad (9)$$

Integrating again,

$$w(t) = 2 \int_0^{\infty} \frac{p(\beta) [\cosh \beta/2 - \cos \beta t]}{\beta^2} d\beta \quad (10)$$

$$= \int_0^{\infty} \frac{\rho(\beta) [\cosh \beta/2 - \cos \beta t]}{\beta \sinh \beta/2} d\beta \quad (11)$$

If we consider a δ function in $\rho(\beta)$, of "area" A_1 (that is $\rho(\beta) = A_1 \delta(\beta - \beta_1)$) then for this δ function

$$w(t) = \frac{A_1 [\cosh \beta_1/2 - \cos \beta_1 t]}{\beta_1 \sinh \beta_1/2}$$

and if the subscript e denotes the experimentally determined part of $\rho(\beta)$, we have for the complete $S(\alpha, \beta)$, including the δ function contribution, from equation (3)

$$S(\alpha, \beta) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \exp. \left\{ -\alpha \left\{ w_e(t) + \frac{A_1}{\beta_1 \sinh \beta_1/2} (\cosh \beta_1/2 - \cos \beta_1 t) \right\} \right\} e^{i\beta t} dt$$

$$= \frac{e^{-\alpha \lambda_1}}{2} \exp. \left\{ -\alpha w_e(t) \right\} \exp. \left\{ \frac{\alpha A_1}{\beta_1 \sinh \beta_1/2} \cos \beta_1 t \right\} e^{i\beta t} dt \quad (12)$$

$$\text{where } \lambda_1 = \frac{A_1 \coth \beta_1/2}{\beta_1}$$

Now

$$\exp \left\{ \frac{\alpha A_1}{\beta_1 \sinh \beta_1/2} \cdot \frac{1}{2} (e^{-i\beta_1 t} + \frac{1}{e^{-i\beta_1 t}}) \right\} = \sum_{n=-\infty}^{\infty} e^{-in\beta_1 t} \ln \left(\frac{A_1}{\beta_1 \sinh \beta_1/2} \right) \quad (13)$$

where $\ln$ is the modified Bessel function

$$\therefore S(\alpha, \beta) = \frac{e^{-\alpha \lambda_1}}{2} \sum_{n=-\infty}^{\infty} \left\{ \frac{1}{2\pi} \int_{-\infty}^{\infty} \exp(-\alpha w_e(t)) e^{i(\beta - n\beta_1)t} dt \right\} \ln \left(\frac{\alpha A_1}{\beta_1 \sinh \beta_1/2} \right)$$

$$\text{But } S_e(\alpha, \beta - n\beta_1) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \exp(-\alpha w_e(t)) e^{i(\beta - n\beta_1)t} dt \quad (14)$$

and therefore

$$S(\alpha, \beta) = e^{-\alpha \lambda_1} \sum_{n=-\infty}^{\infty} S_e(\alpha, \beta - n\beta_1) \ln \left(\frac{\alpha A_1}{\beta_1 \sinh \beta_1 / 2} \right) \quad (15)$$

The scattering law with the δ function included in $\ell(\beta)$ may therefore be expressed as a series involving the scattering law based upon the experimentally measured part of $\ell(\beta)$ alone. ADDELT will, in fact, add on successively any number of δ functions up to a maximum of 50. This facility was used in the construction of Nelkin-type models for H_2O and D_2O , by adding to an $S(\alpha, \beta)$ representing the translational motion three δ functions corresponding to the rotational and vibrational motions.

APPENDIX B

THEORY OF EXTRAPOLATION PROCEDURE FOR $\ell(\beta)$

The derivation of the extrapolation procedure has been largely carried out in Appendix A. From equations (5) and (6) in Appendix A we have

$$\ell(\beta) = 2\beta \sinh \beta/2 \lim_{\alpha \rightarrow 0} S(\alpha, \beta) \quad (16)$$

and thus $\ell(\beta)$, the spectral density, may be obtained by examining the limiting behaviour of $S(\alpha, \beta)$ at small α . In practice, as has been noted, additional components appear in the experimental $S(\alpha, \beta)$ at low α , probably due to multiple scattering effects. The procedure of extrapolating the experimental data for each β to zero α can only yield a first estimate for $\ell(\beta)$. For the first iteration this is fed into LEAP - ADDELTA to generate calculated $S(\alpha, \beta)$'s. $\ell(\beta)$ is corrected on the basis of discrepancies in the 'middle α ' range, away from the effect of multiple scattering.

APPENDIX C

THEORY USED BY THE LEAP PROGRAMME

The LEAP part of the combined LEAP - ADDELT programme calculates $S(\alpha, \beta)$ from the continuous part of the spectral density. For low values of α the scattering law is expressed as a series involving $n = 0, 1, 2, \dots$ phonon interactions, where $n = 0$ represents elastic scattering.

$$S(\alpha, \beta) = e^{-\alpha\lambda} \sum_{n=0}^{\infty} \frac{1}{n!} (B\alpha)^n T_n(\beta) \quad (17)$$

$$\text{where } B = \int_{-\infty}^{\infty} p(\beta)/\beta^2 d\beta$$

$$T_0(\beta) = \phi(\beta)$$

$$T_1(\beta) = p(\beta)/B\beta^2$$

$$T_n(\beta) = \int_{-\infty}^{\infty} T_{n-1}(\beta') T_1(\beta - \beta') d\beta'$$

These numerical convolutions are time consuming and beyond 20 terms thus calculated LEAP uses an Edgeworth series approximation (Sjolander, (1958)) which assumes $\phi(\beta)$ to have a Gaussian form.

$$T_n = \frac{1}{(2\pi n E_2)^{1/2}} \left\{ 1 + \frac{1}{24n} \left(\frac{E_4}{E_2^2} - 3 \right) \left[\left(\frac{\beta^2}{nE_2} - 3 \right)^2 - 6 \right] \right\} e^{-\beta^2/2nE_2} \quad (18)$$

$$\text{where } E_i = \frac{1}{B} \int_{-\infty}^{\infty} \left[p(\beta)/\beta^2 \right] \beta^i \cosh \beta/2 d\beta \quad (i = 2, 4)$$

For large α an excessive number of terms is required and an alternative 'short collision time' approach is used. This gives an asymptotic expansion in inverse powers of

$$S(\alpha, \beta) = \frac{1}{\sqrt{2\pi\alpha f_2(\tau)}} \exp \left[-\alpha(\tau f_1(\tau) - f_0(\tau)) \right] \cdot \left\{ 1 + \frac{1}{\alpha f_2(\tau)} c_1(\tau) + \frac{1}{(\alpha f_2(\tau))^2} c_2(\tau) + \dots \right\} \quad (19)$$

$$\text{where } f_0(\tau) = 2 \int_0^{\infty} \frac{p(\beta)}{\beta^2} (\cosh \beta\tau - \cosh \beta/2) d\beta$$

and where

$$f_{2n+1}(\tau) = 2 \int_0^\infty p(\beta) \beta^{2n-1} \sinh \beta \tau d\beta$$

$$f_{2n+2}(\tau) = 2 \int_0^\infty p(\beta) \beta^{2n} \cosh \beta \tau d\beta$$

τ is given by $f_1(\tau) = \beta/\alpha$ and the C_m are simple functions of f_n/f_2 ($n \geq 3$)

Switching parameters are arranged in the programme to ensure a smooth transition between regions where SCT and the series approximations are used. At large α values the SCT approximation is used and at small α values the series. For intermediate α values the approximation may change at a point in the β range.

Delta-functions in the spectral density outside the measured range are automatically convolved with the continuous part with the ADDEL T section of the combined programme.

A.J.H. Goddard
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THERMAL NEUTRON DIFFUSION AND RETHERMALIZATION IN HYDROGENOUS MATERIALS

A. J. H. GODDARD, P. J. GRANT,
P. W. JOHNSON* AND G. E. B. TREMEER†
IMPERIAL COLLEGE OF SCIENCE AND TECHNOLOGY,
LONDON, UNITED KINGDOM

Abstract

THERMAL NEUTRON DIFFUSION AND RETHERMALIZATION IN HYDROGENOUS MATERIALS.

Using the thermal columns of a reactor as neutron sources two experiments have been performed, the aim of the first being to test the ability of models for the scattering of neutrons by hydrogen bound in water to predict thermal neutron diffusion lengths in solutions of $1/v$ and non- $1/v$ absorbers. In the second experiment rethermalization in Santowax-R (terphenyl) near a temperature discontinuity has been studied.

Diffusion length measurements in both pure water and boric acid solutions have been carried out at 22.3 and 65°C, and in solutions of cadmium and gadolinium at the lower temperature only. In each case measurements were made over a wide range of concentrations. All measured inverse diffusion lengths lie within the $(\Sigma_T)_{\min}$ limit of Comgold with the exception of those from the two most strongly absorbing cadmium solutions. Representation of each set of results as a power series in absorber concentration has enabled the coefficients up to the spectral distortion term to be determined and it is shown that considerable diffusion heating occurs in gadolinium solutions in contrast to the slight diffusion cooling observed in cadmium solutions. In a theoretical analysis four scattering models for bound hydrogen were used. Diffusion lengths were calculated by solving the transport equation for the lowest space eigenvalue in the B_L approximation with $L = 2$, using the method described by Honeck.

The differentially heated experiment combined cold water and hot Santowax-R in slab geometry. Fission rates of ^{235}U and ^{239}Pu were measured with fission chamber detectors as a function of position across the tanks containing the liquids. Two diameters of chamber, 0.6 cm and 0.3 cm, were used to identify finite detector size effects. The general features of the fission rate distributions in the water and Santowax are discussed. In view of current interest concerning the existence of a rethermalization length when the principal scattering is by bound protons, the form of the transient flux in the hot tank is examined in detail. For comparison with the experimental results the fission rates of ^{235}U and ^{239}Pu have been calculated using the Winfrith DSN transport theory code. Scattering by hydrogen in water has been calculated from the Haywood model and in Santowax from a model fitted to the phased chopper velocity selector measurements of Brugger.

1. Introduction

Thermal neutron diffusion length measurements in room-temperature water containing dissolved $1/v$ absorber have been carried out by a number of investigators, in particular Starr and Koppel [1], Bretscher and Meyer [2] and Baumann and Pellarin [3]. Aqueous solutions of non- $1/v$ absorber have been studied by Miller [4] who reports measurements using cadmium in high concentration, and by Russell et al. [5] who used two concentrations each of cadmium and gadolinium. Measurements of this type provide a useful test of the ability of proposed models for the scattering of neutrons by bound

* Present address: Central Electricity Generating Board, Berkeley Nuclear Laboratories, Berkeley, Gloucestershire, UK

† Present address: South African Atomic Energy Board, Pretoria, South Africa.

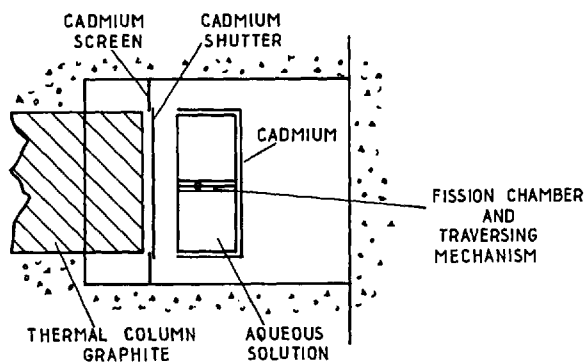


FIG. 1. Experimental arrangement for diffusion length measurements.

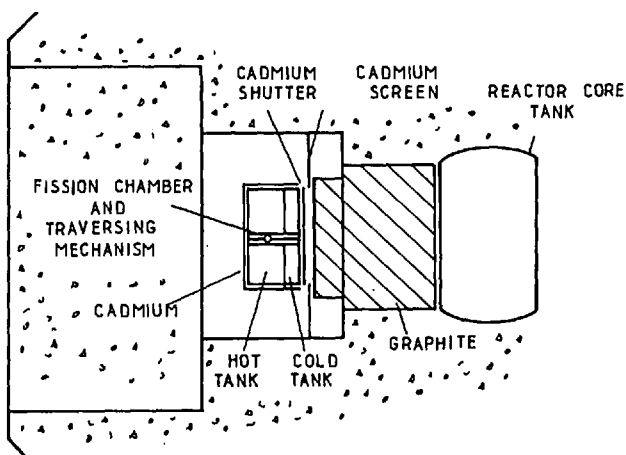


FIG. 2. Experimental arrangement for rethermalization measurements.

hydrogen to predict neutron diffusion and diffusion heating. In the present work neutron diffusion lengths have been measured in water containing dissolved boron at 22.3°C and 65°C and dissolved cadmium and gadolinium at 22.3°C only. A wide range of concentrations has been used and the results are compared with calculation using four different hydrogen scattering models.

In a second experiment rethermalisation across a temperature discontinuity between two liquid moderators, cold water and hot Santowax-R (terphenyl), has been studied in slab geometry using miniature fission chamber detectors. Relatively few measurements of this type have been performed in liquid moderators. Bennett [6] has studied cylindrical graphite-water systems using foil detectors and Saastamoinen [7] the spectrum changes near a temperature discontinuity in water using time-of-flight methods. In a theoretical study of the space eigenvalues of the Boltzmann equation Williams [8] has suggested that if the scattering is by hydrogen and

is according to the free-gas model then the rethermalisation length does not exist. In the analysis of experimental results presented here, the evidence for the separability in space and energy of the well-defined spectrum transient in the Santowax-R, where the principal scattering is by bound hydrogen, is examined in detail.

2. Experimental Techniques

The two experiments were installed on opposite faces of the London University 100-kW reactor CONSORT II, the diffusion experiment (Fig. 1) against the 95.2-cm square thermal column and the rethermalisation experiment (Fig. 2) in a high flux cave separated by 82 cm of graphite from the reactor tank. Fast fluxes entering the experiments as a percentage of the total flux were approximately 5% in the first and 16% in the second and in both cases a negative source technique was used to simulate a near Maxwellian source. An aluminium reinforced cadmium shutter could be lowered to close a square aperture, matching the size of the experimental tanks, in a cadmium screen. Collisions in the shutter gave the negative source a small fast component which was negligible in the case of the diffusion measurements.

An aluminium tank 96.5 cm wide, 38 cm long contained the aqueous absorbing solutions to a depth of 95.2 cm. The sides, rear and base of the tank were covered with heating blankets, insulation and cadmium sheets. A cooling coil and a stirrer were mounted at the rear of the tank where their effect upon the measured flux distribution was negligible. Solution temperatures could be controlled to better than $\pm 1^\circ\text{C}$ over the range $20 - 70^\circ\text{C}$. In addition to pure water, ten different concentrations of boric acid were used at 65°C , nine at 22.3°C and ten each of cadmium sulphate and gadolinium nitrate at 22.3°C . A small quantity of boron (8.7 ± 0.9 p.p.m.B) was detected spectrographically in the cadmium solutions and is thought to have been adsorbed onto the tank surface during the boron measurements.

In the rethermalisation measurements two tanks fabricated from 0.16-cm aluminium contained cold water and hot Santowax. A 0.32-cm gap between the tanks was maintained by 0.48-cm diameter insulating discs fixed to the cold tank face. The cold tank was 10.3 cm long and the hot tank 25.4 cm long; both were 61 cm wide and were filled to a depth of 61 cm with the liquids. The mismatch of the transverse bucklings of the tank and the main 95.2-cm square graphite neutron feed meant that harmonic corrections had to be considered. Demineralised water in the cold tank was kept close to room temperature by circulation through a heat exchanger while the Santowax temperature was automatically controlled using hot plates with a total output of $5\frac{1}{2}$ kW beneath the tank. The sides and rear face of the tanks were covered with cadmium sheet and thermal insulation.

A miniature fission chamber which could be moved horizontally by a remotely operated screw drive was used in both experiments, and in the rethermalisation experiment a second vertical screw drive together with a wiper seal to remove

liquid enabled the chamber to be transferred from one tank to the other. Horizontal positional reproducibilities of 0.002 cm and 0.013 cm were achieved in the diffusion and rethermalisation experiments respectively. Two pairs of ^{235}U and ^{239}Pu chambers manufactured by 20th Century Electronics Ltd were available, mounted in 0.62 cm and 0.3 cm outside diameter gas-filled Nimonic tubes of such a length that the centre of the active regions were at the tank centre line. The fissile coatings, which were of 0.5-cm and 0.25-cm diameters had thicknesses of 1 mg/cm² for ^{235}U and 0.3 mg/cm² for ^{239}Pu . In the diffusion experiment a 0.3-cm ^{235}U chamber only was used.

Fission chamber gain changes with temperature between cold water and hot Santowax tanks were determined by the method of graphical superposition of attenuation bias curves. The greatest correction applied was 1.5%. A constant systematic error of approximately 2 mm in the position of the 0.3-cm detectors in the hot tank has been shown to be due to temperature-induced internal gas pressure changes straightening out small bends in the rather flexible counter stems. A possible error in the negative source technique arose through radiative capture γ -rays from cadmium giving photo-fission counts in the measuring fission chambers. From the change in counting rate produced by lowering the cadmium screen in front of an additional boron screen it was shown that the photofission contribution must be less than 4.9% of the fast flux or typically 1% of the difference counting rate observed in the rethermalisation experiment.

3. Diffusion Length Results

After making corrections for dead-time effects and reactor power fluctuations the difference $\Phi(z)$ between the corrected shutter open and closed counting rates as a function of distance z from the front face of the tank was fitted by a least-squares technique to the expression

$$\Phi(z) = Ae^{-\gamma z} \quad (1)$$

by optimising A and γ . No corrections for the tank being finite in the z direction were necessary. No systematic deviations from the exponential form were found, either near to the front face, where the first point was 0.6 cm from the tank wall, or for points farthest from the wall, where errors arising from the negative source technique might have been apparent. The fundamental space eigenvalue was obtained by applying a correction for lateral leakage

$$\kappa^2 = \gamma^2 - 2 \left(\frac{\pi}{a} \right)^2 \quad (2)$$

where a is the tank extrapolated dimension in the transverse directions. The maximum contribution (in the case of pure water) of the leakage term was only 1.7% in κ^2 and the uncertainty in this term was estimated to produce an error of only 0.05% in κ^2 for pure water. The results for absorbing solutions are presented in Tables I and II. The two most

TABLE I. Boron absorption at 22.3°C and 65°C ^a

Solution No.	22.3°C		65°C	
	L(cm)	g/litre H ₃ BO ₃	L(cm)	g/litre H ₃ BO ₃
1	2.383 ± .004	.996 ± .005	2.563 ± .004	1.000 ± .005
2	2.139 ± .003	1.99 ± .01	2.305 ± .003	2.00 ± .01
3	1.866 ± .002	3.56 ± .02	2.014 ± .002	3.59 ± .02
4	1.621 ± .002	5.68 ± .03	1.754 ± .003	5.69 ± .03
5	1.269 ± .002	11.40 ± .06	1.351 ± .002	11.50 ± .06
6	1.094 ± .002	17.10 ± .08	1.170 ± .003	17.20 ± .08
7	.9738 ± .0015	22.9 ± .1	1.0325 ± .0013	22.85 ± .10
8	.8278 ± .0019	33.5 ± 1.0	.8793 ± .0023	33.5 ± 1.0
9	.7348 ± .0017	46.4 ± .5	.7755 ± .0020	46.4 ± .5
10			.6581 ± .0021	69.3 ± 1.4

^a The concentrations are given in g/litre as determined at 23°C

TABLE II. Cadmium and gadolinium absorption at 22.3°C

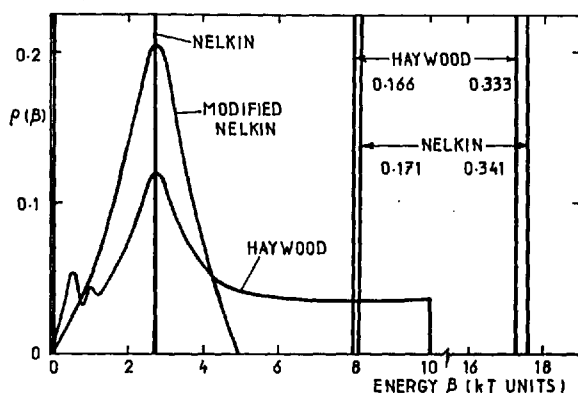
Solution No.	Cadmium		Gadolinium	
	L(cm)	g/litre 3CdSO ₄ ·8H ₂ O	L(cm)	g/litre Gd(NO ₃) ₃ ·6H ₂ O
1	2.408 ± .003	.774 ± .001	2.487 ± .004	.0992 ± .005
2	2.138 ± .003	1.732 ± .004	2.301 ± .006	.169 ± .003
3	1.859 ± .003	3.251 ± .005	2.044 ± .004	.335 ± .001
4	1.566 ± .002	5.772 ± .010	1.658 ± .002	.725 ± .004
5	1.234 ± .002	10.90 ± .02	1.458 ± .002	1.062 ± .002
6	1.0251 ± .0014	17.07 ± .03	1.279 ± .002	1.546 ± .003
7	.8843 ± .0014	23.72 ± .04	1.0995 ± .0014	2.370 ± .005
8	.7165 ± .0019	37.82 ± .08	.9775 ± .0018	3.251 ± .006
9	.5983 ± .0019	55.52 ± .10	.8937 ± .0018	4.155 ± .008
10	.4925 ± .0029	82.15 ± .13	.8222 ± .0015	5.405 ± .010

heavily absorbing cadmium solutions lay outside the $\Sigma_T(\min)$ limit.

Two independent measurements made of the diffusion length in pure water at 22.3°C yielded the results $2.767 \pm .004$ cm and $2.755 \pm .004$ cm and the mean value $2.761 \pm .006$ cm was adopted. This is in excellent agreement with the average value $2.767 \pm .008$ cm of several stationary measurements quoted by Beckurts [9]. At 65°C the result obtained was $2.971 \pm .005$ cm.

4. Scattering by Water and Calculation of the Eigenvalue

In considering the hydrogen atom motion in water some simplifying assumptions have been made. The isotropic

FIG. 3. Frequency spectrum models for hydrogen in H_2O .

harmonic approximation has been used throughout and molecular self-diffusion has not been included in the calculations although for one model results are presented for the two limiting situations of no translational motion and translation as a free gas of mass 18.

In addition to the basic Nelkin model [13] a modified Nelkin model has been used in which the δ -function representing hindered rotations in the generalised frequency spectrum is broadened in an arbitrary symmetric manner to form a distributed peak of the same mass (Fig. 3). This modification to the Nelkin model is similar to one employed by Koppel [10] and has been constructed purely for the purpose of comparison with other distributed frequency spectrum models. As a second basic model the Haywood model [11] shown in Fig. 3 has been used.

Scattering Law $S(\alpha, \beta)$ data, where α and β are the dimensionless momentum and energy transfers, for both the modified Nelkin and the Haywood model have been generated using the programme LEAP-ADDELT of McLatchie [12]. $S(\alpha, \beta)$ matrices were generated with 51 α and 60 β values covering a range such that scattering cross-sections in the range 0 - 1 eV could be calculated.

LEAP-ADDELT will only calculate the inelastic part of the Scattering Law $S_1(\alpha, \beta)$ and in order to include the free gas mass 18 in the modified Nelkin model, the convolution

$$S(\alpha, \beta) = e^{-\alpha^2 \lambda} S_G(\alpha/18, \beta) + \int_0^\infty S_G(\alpha/18, \beta') S_1(\alpha, \beta - \beta') d\beta' \quad (3)$$

was carried out by numerical integration. $S_G(\alpha, \beta)$ is the free-gas model scattering law and $e^{-\alpha^2 \lambda}$ is the Debye-Waller factor. Using the same procedure a fourth model was constructed by combining the Haywood model with mass 18 free translations; in order to preserve the normalisation of the frequency spectrum the Haywood spectrum was reduced in amplitude overall by 5.5%. This will be referred to as the modified Haywood model.

The differential scattering cross-section is related to the scattering law by

$$\sigma(E_0, E, \mu) = \frac{1}{T} \frac{\sigma_b}{4\pi} \sqrt{\frac{E}{E_0}} e^{-\beta/2} S(\alpha, \beta) \quad (4)$$

where E_0, E are the initial and final energies, μ is the cosine of the scattering angle and σ_b the bound atom cross-section.

The first stage in the computation of the diffusion length is the calculation of the Legendre moments of the scattering kernel, defined by

$$\sigma_n(E_0, E) = 2\pi \int_{-1}^1 \sigma(E_0, E, \mu) P_n(\mu) d\mu \quad (5)$$

where $P_n(\mu)$ is the Legendre polynomial of order n . The integration in equation (5) is carried out by a double Gaussian quadrature scheme; $\sigma(E_0, E, \mu)$ is calculated from equation (4), the required values of $S(\alpha, \beta)$ being obtained by linear interpolation of $S(\alpha, \beta)e^{\beta/2}$ in α and β . For the basic Haywood model where the elastic scattering is simply the Debye-Waller factor, there is an additional component to the n th moment of the scattering kernel

$$C_n(E_0) \delta(E_0 - E) = \frac{\sigma_b}{2} \int_{-1}^1 e^{-\alpha\lambda} P_n(\mu) d\mu \delta(E_0 - E) \quad (6)$$

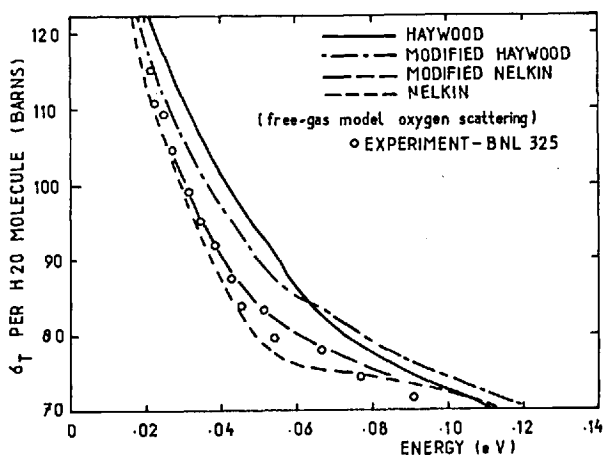
In the case of the basic Nelkin model $\sigma(E_0, E, \mu)$ is calculated directly using the analytic expressions which are obtained when the well-known approximations of Nelkin [13] are made. In all cases scattering from the oxygen in water is represented by adding a contribution from a free-gas model of mass 16.

The second stage is the solution of the stationary neutron transport equation for the source-free half space $z > 0$ bounded by a uniform plane source at the origin. This may be written

$$\begin{aligned} & \left[\mu \frac{\partial}{\partial z} + \Sigma_T(E) \right] \varphi(z, E, \mu) \\ & = \int_{-1}^1 d\mu' \int_0^\infty dE' \Sigma_S(E' \rightarrow E, \mu_0) \varphi(z, E', \mu') \end{aligned} \quad (7)$$

where μ_0 is the cosine of the angle between μ and μ' , and $\Sigma_T(E)$ is the total macroscopic cross-section at energy E . The method adopted for the solution of equation (7) to obtain the fundamental space eigenvalue (the ' B_L ' method) has been described by Honeck [14].

Ten calculations with 32 energy points covering the energy range 0 - 0.625 eV were carried out for each absorber using the $L = 2$ approximation with the exception of the 65°C case where $L = 1$ was used. These ten calculations were

FIG.4. H_2O total cross-section; comparison of calculation with experiment.TABLE III. $1/v$ coefficients for water at $22.3^\circ C$ and $65^\circ C$

Experiment	T($^\circ C$)	D_o ($cm^2 sec^{-1}$)	C ($cm^4 sec^{-1}$)	σ_{aH} (mb)
Present	22.3	35620 ± 230	2800 ± 260	321.9 ± 2.1
Starr and Koppel	21	35850 ± 100	2900 ± 350	326.9 ± 1.6
Küchle	22	35400 ± 700	4200 ± 800	326 ± 6
Present	65	41340 ± 390	2470 ± 400	326.5 ± 3.8
Calculation				
Haywood	22.3	32660	3750	-
mod. Nelkin	22.3	36820	2920	-
Nelkin	22.3	37850	3230	-
Nelkin	65	43350	3340	-

equally spaced in absorber concentration between the pure water case and the $\Sigma_T(\min)$ limit (Corngold [15]) above which no fundamental space eigenvalue exists. Cadmium and gadolinium absorption cross-sections were read from BNL-325; the hydrogen 2200-m/s absorption cross-section was taken as 0.332 b after Stehn, et al. [16] and that for boron 759 barns. The hydrogen cross-section was increased to 0.337 b for the cadmium runs to allow for the boron contamination mentioned in Section 2. Only Nelkin model calculations were performed for boron absorption at $65^\circ C$ and calculations with the modified Haywood model were not carried out for boron absorption at $22.3^\circ C$.

5. Comparison of Experiment and Theory

5.1. 1/v absorption at 22.3°C and 65°C

Corrections were applied to the experimental eigenvalues to allow for the reduction in the moderator number density on addition of solute. Both corrected experimental and calculated eigenvalues have been fitted by the method of least-squares to a series expansion of the form

$$\kappa^2 = \alpha_1 \Sigma_{ao} - \alpha_2 \Sigma_{ao}^2 + \alpha_3 \Sigma_{ao}^3 \quad (8)$$

where Σ_{ao} is the sum of the 2200 m sec⁻¹ macroscopic absorption cross-sections for boron and hydrogen and the α 's are coefficients to be fitted. The magnitude and sign of the coefficient α_2 indicates the effect of diffusion cooling or heating. Fitted coefficients obtained at 22.3°C and 65°C are given in Table III where the more familiar pulsed parameters have been obtained through the expressions quoted by Honeck [14]. An experimental value of the hydrogen absorption cross-section was determined by including a constant term in (8), in the nature of a correction to the assumed value.

The experimental values, at the two temperatures, of the parameter F were close to 370 $\pm$ 500 cm⁶ sec⁻¹ and it is clear that coefficients of higher order than C are elusive. The 22.3°C experimental value for D₀ is in excellent agreement with results obtained by other experimenters using pulsed and stationary methods and the value of C at this temperature agrees well with stationary measurements as typified by the results of Starr and Koppel [1] but is somewhat less than values obtained from pulsed experiments of which the results of KÜchle [17] are given in Table III.

Differences between calculation and experiment are best illustrated graphically by re-arranging the series expansion in the form

$$\kappa^2 / \Sigma_{ao} = \alpha_1 - \alpha_2 \Sigma_{ao} + \alpha_3 \Sigma_{ao}^2 \quad (9)$$

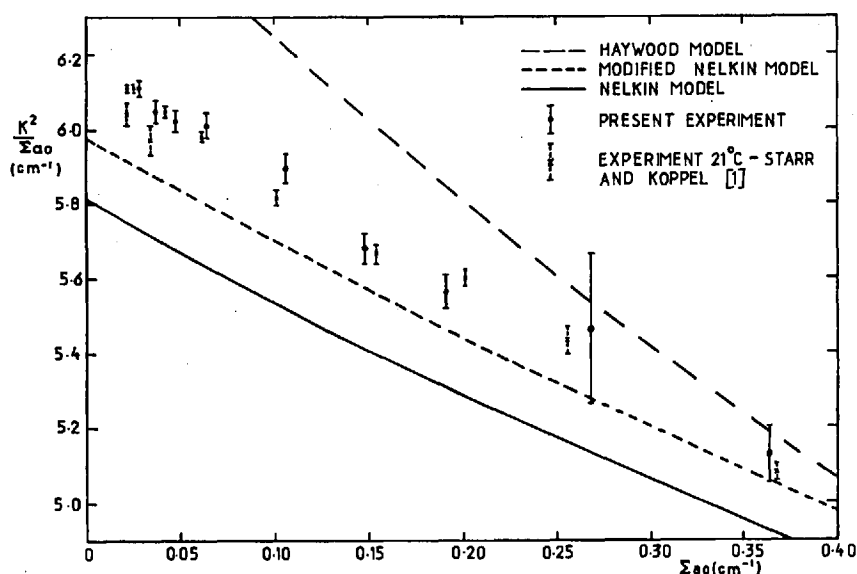
and plotting κ^2 / Σ_{ao} vs. Σ_{ao} to give a graph which by its gradient demonstrates the effect of diffusion heating. This is done for 22.3°C in Fig. 5.

5.2. Non-1/v absorption at 22.3°C

Moderator number density reductions caused by the solute were only significant in the cadmium solutions and the correction applied was 0.18% in the most concentrated case. For non-1/v absorbers the experimental and calculated eigenvalues are fitted to the expansion

$$\kappa^2 = \beta_0 + \beta_1 N - \beta_2 N^2 \quad (10)$$

where $N(\text{cm}^{-3})$ is the absorber number density and the coefficient β_0 representing the effect of absorption by hydrogen

FIG. 5. κ^2/Σ_{ao} plotted against Σ_{ao} for boric acid solutions at 22.3°C.TABLE IV. Non- $1/v$ coefficients for water at 22.3°C

Expt/Model	Element	β_0 (cm ⁻²)	$\beta_1 \times 10^{19}$ (cm)	$\beta_2 \times 10^{38}$ (cm ⁴)
Experiment	Cd	.1356 $\pm$.0005	.202 $\pm$.002	-.0002 $\pm$.0001
Haywood	Cd	.1488	.2254	-.00003
mod. Haywood	Cd	.1428	.2164	-.0004
mod. Nelkin	Cd	.1328	.2014	-.0003
Nelkin	Cd	.1291	.1956	-.0002
Experiment	Gd	.1315 $\pm$.0002	2.49 $\pm$.02	.86 $\pm$.03
Haywood	Gd	.1466	2.60	1.02
mod. Haywood	Gd	.1407	2.53	.83
mod. Nelkin	Gd	.1310	2.36	.63
Nelkin	Gd	.1273	2.29	.60

(and trace quantities of boron in the case of the cadmium solutions) is just κ^2 for water alone. The coefficient β_2 represents the effect of spectrum distortion. Fitted coefficients are shown in Table IV. The small negative value of β_2 obtained by experiment and calculation for cadmium shows that diffusion cooling is taking place whereas in gadolinium solutions diffusion heating is observed.

If the series expansion is re-arranged in the form

$$\frac{(\kappa^2 - \beta_0)}{\Sigma_{oo}} = \frac{\beta_1}{\sigma_{oo}} - \frac{\beta_2}{\sigma_{oo}^2} \Sigma_{oo} + O.(\Sigma_{oo}^2) \quad (11)$$

where Σ_{oo} is the product of the non-1/v absorber density and its microscopic absorption cross-section averaged over a Maxwellian spectrum σ_{oo} , then plotting $(\kappa^2 - \beta_0)/\Sigma_{oo}$ vs. Σ_{oo} yields a graph whose slope demonstrates the effect of diffusion cooling or heating. Experiment and calculation are compared in this form in Figs. 6 and 7.

5.3. Discussion

In the eigenvalue calculations for the three absorbers at 22.3°C as shown in Figs. 5, 6 and 7 a basic trend exists in that the Haywood model consistently gives the greatest, the Nelkin model the least, and modified Haywood and modified Nelkin intermediate values.

In the case of cadmium absorption shown in Fig. 6 the modified Nelkin model gives the best overall agreement. The magnitude of the diffusion cooling is very small and it is not possible to distinguish between the predictions of β_2 for the various models. Spectral distortion effects are small because in the diffusion approximation these depend upon the difference between the variation of absorption cross-section and diffusion constant with energy and this difference is small because of the presence of the cadmium resonance. Fig. 4 shows total cross-sections calculated for the four models compared with experimental values from BNL-325. It appears that stationary diffusion experiments in cadmium solutions give information of the same nature as do total scattering cross-section measurements. The Haywood-type models give rather better agreement with the gadolinium measurements. This suggests that the Haywood model may be better able to predict spectrum distortion effects.

It has been shown that for the three absorbers experimental points in general lie between the predictions of the Nelkin-type model with a distributed rotational peak and the Haywood model modified to include free-gas mass 18 translational motion. The differing predictions given by these two models must arise primarily from the different form of the distributed parts of the frequency spectrum as both models have essentially the same treatment of free translational motion and of vibrational motion by two discrete oscillators. Koppel [10] has suggested that, at least in the prediction of total cross-sections, the difference between the Haywood model calculations and experiment in the energy range 0.02 - 0.1 eV arises from excessive weighting of the region in the frequency spectrum around 0.2 eV.

6. Fission Rate Distribution across the Temperature Discontinuity

Thermal fission rates as a function of position in cold and hot tanks were derived by subtracting shutter up and

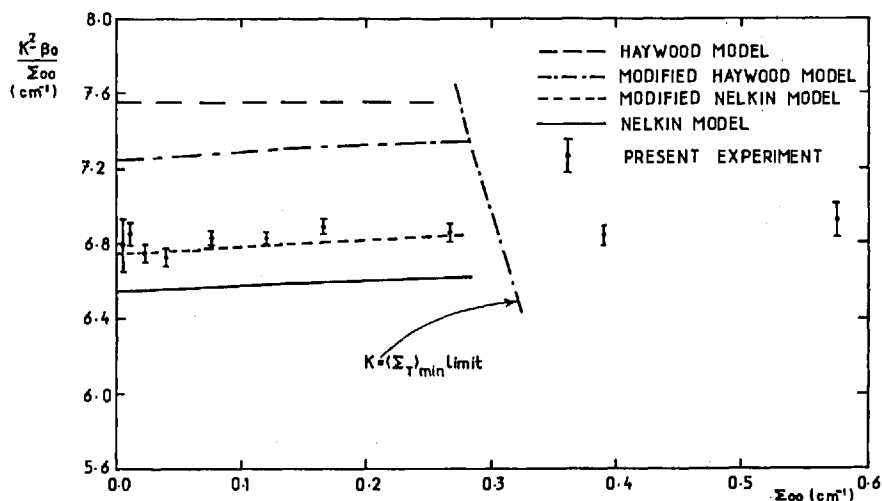


FIG. 6. $(\kappa^2 - \beta_0)/\Sigma_{00}$ plotted against Σ_{00} for cadmium sulphate solutions at 22.3°C.

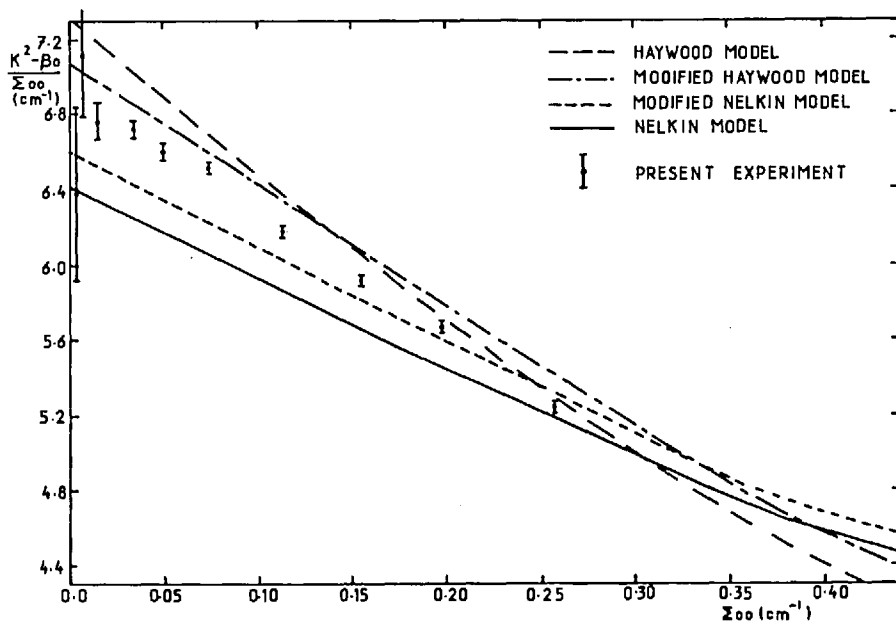


FIG. 7. $(\kappa^2 - \beta_0)/\Sigma_{00}$ plotted against Σ_{00} for gadolinium nitrate solutions at 22.3°C.

shutter down counting rates and applying dead time and neutron power fluctuation corrections. Three Santowax-R temperatures, 171°C, 201°C and 232°C were used, the nominal water temperature being 17°C. Fig. 8 shows fission rate distributions, measured with a Santowax temperature of 201°C, expressed as deviations from the unperturbed exponential form. The transients are clearly defined, particularly in

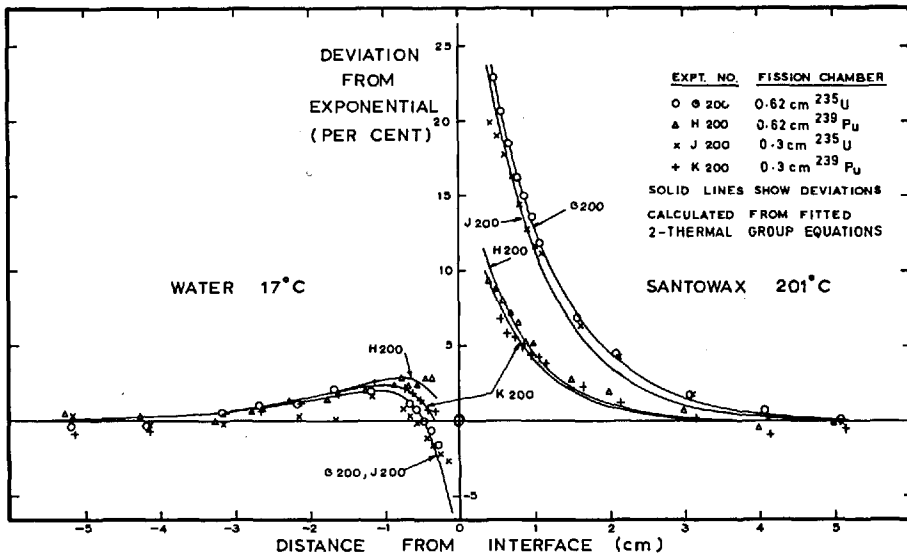


FIG. 8. Measured transients in the fission rates of ^{239}Pu and ^{235}U near a plane discontinuity between cold H_2O and hot Santowax-R.

the Santowax where because of the lower hydrogen density the rethermalisation process extends over a greater distance than in water. The initial rise in the deviation from exponential in the water tank as the interface is approached is due to the second part of the fundamental distribution. Also shown are the predictions of a two overlapping thermal group model fitted to individual fission rates by the method of least-squares. The analysis of the measurements in Section 8 is related to the validity of this model.

7. Calculation of Fission Rates

7.1. Scattering laws

With the object of providing realistic models for scattering by hydrogen bound in Santowax-R, generalised frequency distributions fitted [18] to the measurements of Brugger [19] at 207°C and 267°C have been extended to yield models for 171°C, 201°C and 232°C. Linear interpolation and extrapolation at corresponding energies (in eV) yielded the distributions shown in Fig. 9. The LEAP-ADDELT programme has been used to generate Scattering Law, $S(\alpha, \beta)$ data. No account has been taken of translational motion of the molecule. As a test of the proposed models Fig. 10 shows the result of performing the necessary angular and energy integration of equation (4) to derive the total cross-section. The differences between calculation and the measurements of Brugger are not large but are greater than the experimental error.

The Haywood model [11] has been used for the scattering by hydrogen in water and the free-gas model for oxygen, carbon and the aluminium in the interface walls.

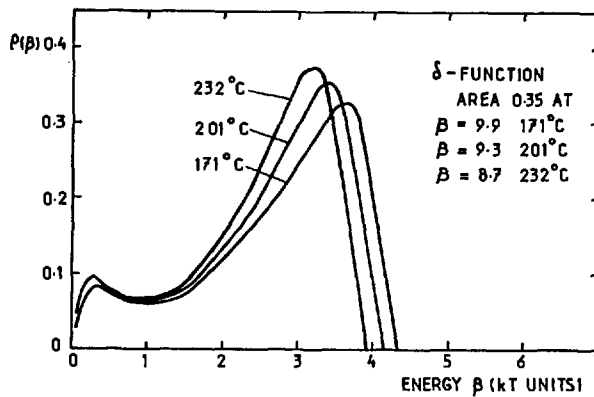


FIG. 9. Frequency spectrum models for hydrogen in Santowax-R.

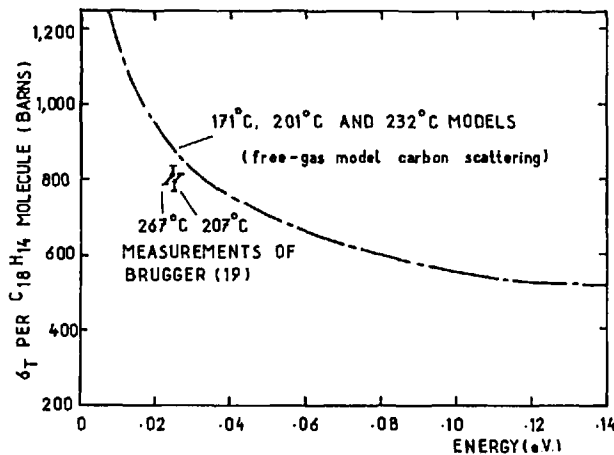


FIG. 10. Santowax-R total cross-section; comparison of calculation with experiment.

7.2. Spatial calculation

Intergroup transfer cross-sections for a slab version of the transport theory programme WDSN [20] were prepared with the PIXSE [21] programme. 31 energy groups were used covering the range 0 - 1 eV. The version of the WDSN programme used was restricted to isotropic scattering and for hydrogen scattering in the two moderators the first approximation to the correction for anisotropic scattering

$$\sigma_o(g \rightarrow g)_{\text{modified}} = \sigma_o(g \rightarrow g) - \sum_{g'} \sigma_1(g \rightarrow g') \quad (12)$$

was made by modifying the diagonal element in the scattering matrix.

TABLE V. Experimental fundamental and transient relaxation lengths

Expt. No.	Detector dia.(cm) /Isotope	Hot Tank Temp. (°C)	γ_0 (cm ⁻¹)	γ_1 (cm ⁻¹)	χ^2	χ_0^2
G170	0.62 U	171.7	.1827 $\pm$.0004	1.35 $\pm$.05	22	26
H170	0.62 Pu	170.5	.1806 $\pm$.0005	1.17 $\pm$.07	25	29
J170	0.3 U	170.5	.1811 $\pm$.0007	1.16 $\pm$.07	27	25
K170	0.3 Pu	171.0	.1810 $\pm$.0005	1.03 $\pm$.08	10	25
G200	0.62 U	201.7	.1740 $\pm$.0003	1.25 $\pm$.03	12	29
G200A	0.62 U	202.4	.1754 $\pm$.0006	1.32 $\pm$.05	33	26
G200B	0.62 U	201.9	.1732 $\pm$.0004	1.25 $\pm$.04	22	29
H200	0.62 Pu	200.1	.1727 $\pm$.0005	1.12 $\pm$.08	24	29
J200	0.3 U	198.8	.1738 $\pm$.0006	1.13 $\pm$.05	16	25
K200	0.3 Pu	200.5	.1722 $\pm$.0006	1.00 $\pm$.10	17	25
G230	0.62 U	232.9	.1666 $\pm$.0004	1.28 $\pm$.04	21	29
H230	0.62 Pu	231.7	.1639 $\pm$.0005	1.19 $\pm$.05	5	20
J230	0.3 U	232.4	.1653 $\pm$.0006	1.14 $\pm$.05	20	25
K230	0.3 Pu	231.9	.1659 $\pm$.0005	1.24 $\pm$.14	21	25

TABLE VI. Calculated fundamental and transient relaxation lengths

Santowax Temperature (°C)	Fissile isotope	γ_0 (cm ⁻¹)	γ_1 (cm ⁻¹)
171	U Pu	0.184	1.13 1.04
201	U Pu	0.171	1.13 1.03
232	U Pu	0.159	1.12 1.03

Fixed source calculations in the S_4 approximation using 50 space points were carried out for three Santowax temperatures of 171, 201 and 232 and with a water temperature of 17°C. In order to avoid using unduly wide mesh intervals the cold and hot tanks were taken to be 6.6 cm and 22 cm wide respectively. The source had a Maxwellian energy distribution in the first three mesh intervals. Group-averaged fission cross-sections for ^{239}Pu and ^{235}U for use in the

calculation of fission rates were derived using cross-sections from the Winfrith Nuclear Data File.

8. Analysis of Fission Rate Distributions in Santowax-R

The measured and calculated fission rate distributions have been examined for the possible existence of a discrete eigenvalue other than the fundamental. If a second eigenvalue exists then beyond a certain distance from the interface, close to which transport and higher spectrum transients might be significant, a fission rate distribution in the hot tank might be represented by

$$F(r) = A_1 e^{-\gamma_1 r} + A_2 e^{-\gamma_0 r} + A_3 e^{\gamma_0 r} \quad (13)$$

where γ_0 and γ_1 are the inverse relaxation lengths of the fundamental and transient components.

If the transient flux is not separable in space and energy then different fitted values of γ_1 for ^{239}Pu and ^{235}U distributions might be obtained and also the assumption of an exponential form for the transient might not be valid.

In the method of non-linear fitting used [22], the function was linearised by expanding it in a Taylor series in the unknown parameters about initial guessed values. Application of the least-squares criterion led to a set of normal equations involving the corrections to the initial values of the parameters. This process was repeated until accuracy criteria were satisfied.

Convergence was not obtained when fitting expression(13) to the experimental data owing to the small influence at the measuring positions of leakage from the back face of the hot tank. The data were then fitted to

$$F(r) = A_1 e^{-\gamma_1 r} + A_2 (e^{-\gamma_0 r} - e^{-2\gamma_0 H} \times e^{\gamma_0 r}) \quad (14)$$

where H is the extrapolated thickness of the hot tank. By fitting expression(14) to the experimental data using different fixed values of H it was shown that the value of H expected from the physical dimensions of the hot tank was close to the value indicated by the goodness of fit.

Table V shows the results of fitting expression(14) to the experimental data. For the first points included in the fit the detector centres were 0.4 cm and 0.5 cm from the interface for 0.62 cm and 0.3 cm detectors, respectively. For the same Santowax-R temperature there are no systematic differences in the fitted γ_0 , either with fissile isotope or detector size. The values of chi-squared when compared with the theoretical χ_0^2 corresponding to the number of degrees of freedom and a 5% level of significance show that the differences between experiment and expression(14) were not significantly greater than the experimental errors.

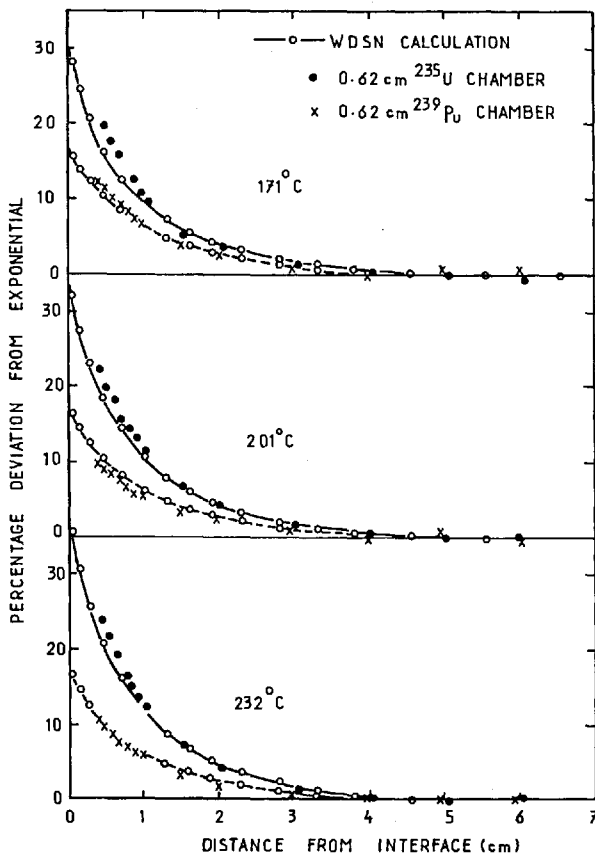


FIG. 11. Calculated fission-rate transients in Santowax-R compared with 0.62-cm chamber measurements.

Comparing the fitted values of γ_1 there are significant differences between ^{239}Pu and ^{235}U experimental results obtained using 0.62 cm detectors. These results must however be treated with some caution because of the large detector diameter which resulted in displacement of moderator. With the 0.3-cm detectors the resulting γ_1 values are less than those obtained with the larger detectors and the differences between ^{239}Pu and ^{235}U detector results are not greater than the experimental errors. The errors were greater because of inferior counting statistics. Dropping up to 8 experimental points near to the interface and repeating the fit produced no systematic change in γ_1 for either diameter of detector. The differences between values of γ_1 fitted to ^{239}Pu and ^{235}U measurements were not affected by either the inclusion of a harmonic term in expression 14 or the correction of the data for fast neutron collisions in the neutron shutter.

Expression (14) was fitted to fission rate distributions in the hot tank calculated using WDSN and the resulting

values of γ_1 and γ_0 shown in Table VI are those obtained with the first point in the fit 0.5 cm from the interface and with the relative weighting of points similar to that in the experiments. Corrections for lateral leakage have not been applied to the calculated γ_0 and γ_1 ; this correction would be negligible in the case of γ_1 . A 10% difference between γ_1 fitted to ^{239}Pu and ^{235}U distributions is indicated. As calculated points closer than 0.3 cm from the interface were included in the fitting process the goodness of fit became markedly worse, although the difference between γ_1 from ^{239}Pu and ^{235}U was essentially unchanged. The influence of the temperature dependence of the effective cross-section of ^{235}U upon the amplitude of the observed transient is shown in Fig. 11, where calculated and measured fission rate distributions are compared.

The results of experiment and calculation suggest that the transient flux is not separable in space and energy although the evidence is not conclusive. There is a need for the experiment to be repeated with improved statistical accuracy or with detectors more sensitive to thermal neutron spectrum changes and for the calculations to be repeated with a more rigorous treatment of anisotropic scattering.

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Neutron scattering by liquid terphenyl

A. J. H. GODDARD

Nuclear Power Group, Mechanical Engineering Department, Imperial College of Science and Technology, London

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Abstract. Generalized frequency distributions for the normal modes of hydrogen motion in liquid Santowax-R (terphenyl) at 207°C and 267°C have been fitted to the phased-chopper velocity selector measurements of Brugger. The fitting procedure is based upon the method of Egelstaff and Schofield. It is shown that the hydrogen frequency distribution is similar to the equivalent distribution for the solid over the energy range 0.005–0.06 eV. There is a discrepancy of approximately 12% between the total cross section at 0.025 eV calculated from the fitted model and the direct measurements of Brugger.

1. Introduction

Liquid Santowax-R ($C_{18}H_{14}$), which is a mixture of ortho-, meta-, para- and quarterphenyls (in the approximate proportions 9 : 52 : 30 : 8), is the most commonly used coolant in organic cooled nuclear reactors. The high boiling point of approximately 380°C, which makes pressurization largely unnecessary, also makes Santowax-R an attractive material for basic neutron rethermalization experiments. The work described here was undertaken to provide scattering cross sections for a transport theory analysis of a 'two-temperature' experiment, with Santowax-R as the hot moderator, that has recently been completed using the University of London CONSORT reactor as a neutron source.

Partial differential scattering data for solid Santowax-R at 23°C and for the liquid at 207°C and 267°C obtained by Brugger (1962) using a phased-chopper velocity selector have been analysed using the iterative method based on the theory of Egelstaff and Schofield (1962) to obtain a fitted generalized frequency distribution for the hydrogen atom motion. This method of analysing neutron scattering data has been used by B. C. Haywood (1965, private communication) for water and by Beckurts and Gläser (1964) for benzene and diphenyl. From a fitted frequency distribution, scattering cross sections can be generated covering the whole range of neutron momentum and energy transfers required for reactor physics calculations in the thermal energy range. The total scattering cross section for Santowax-R has been calculated as a function of energy and compared with the measurements of Brugger at 0.025 eV.

2. Method of analysis

Brugger has derived the reduced partial differential scattering cross section, the scattering law $S(\alpha, \beta)$, where α is the momentum change $\{E + E' - 2(EE')^{1/2} \cos \theta\}/AkT$ (A is the atomic weight of the principal scattering atom; $A = 1$ has been used throughout this work) and β the energy change $(E' - E)/kT$, by measuring the slow neutron differential scattering cross section using a phased-chopper velocity selector. $S(\alpha, \beta)$ is defined by

$$\frac{\partial^2 \sigma}{\partial \Omega \partial E} = \frac{\langle |a|^2 \rangle}{kT} \left(\frac{E'}{E} \right)^{1/2} e^{-\beta/2} S(\alpha, \beta) \quad (1)$$

where $\langle |a|^2 \rangle$ is the mean square of the modulus of the bound atom scattering lengths.

The iterative method described by Egelstaff, Haywood and Thorson (1963) for water has been used to fit a generalized frequency distribution $\rho(\beta)$ of the normal modes of the

hydrogen atom motion to the data; in the incoherent Gaussian approximation to the self-correlation function $G(\mathbf{r}, t)$ it has been shown by Egelstaff and Schofield (1962) that

$$\rho(\beta) = 2\beta \sinh \frac{1}{2}\beta \lim_{\alpha \rightarrow 0} \frac{S(\alpha, \beta)}{\alpha} \quad (2)$$

and this is used to obtain the first estimate from the data. Succeeding iterations attempt to obtain agreement between calculation and experiment over the whole energy range. The Gaussian part of $S(\alpha, \beta)$ is generated from $\rho(\beta)$ for each iteration by using the programme LEAP-ADDELT (R.C.F. McLatchie 1965, private communication) for the Atlas computer. LEAP is based upon the method of Egelstaff and Schofield which, for low momentum transfer, uses a phonon series expansion with up to 20 terms calculated by direct numerical convolution, while for high momentum transfer the method of steepest descents is used. High-energy vibrational modes outside the measured range are included in $\rho(\beta)$ with the programme ADDELT which uses a method due to Zemach and Glauber (1956) to convolve successively δ functions in $\rho(\beta)$ with $S(\alpha, \beta)$ computed from the continuous part of $\rho(\beta)$.

3. Comparison with experimental data

Bound atom calculations for the solid and liquid are compared with the experimental data in figures 1-3. There is a general disagreement at low α values; similar discrepancies have been observed in analysing scattering by water (Egelstaff *et al.* 1963). Brugger considers that because of multiple scattering effects the most reliable experimental value of $S(\alpha, \beta)$ for $0.1 < \beta < 0.5$ is a line near the smaller value of $S(\alpha, \beta)$ and that at larger β values the energy loss points will fall above the neutron energy gain points. If this is taken into account, then the agreement is quite satisfactory for the solid data. The agreement is less good for the liquids, where there are discrepancies at large α values for low β values and at all α values when $\beta > 2.5$. This latter discrepancy restricted the range of the $\rho(\beta)$ fit to $0.1 < \beta < 2.5$ for the liquid data. The calculated curves are for hydrogen only; assuming carbon to scatter according to the free-gas model then the contribution to $S(\alpha, \beta)$ per $\text{C}_{18}\text{H}_{14}$ molecule is only a few per cent at $\beta = 0.1$ and is negligible for higher β values. The large differences between experiment and the bound atom calculation at $\beta = 0$ are thought to be due to molecular diffusive motion. Because of uncertainty concerning the mechanism by which diffusion occurs in Santowax-R, no attempt has been made to include this effect in the calculations.

The proposed $\rho(\beta)$ distributions for liquid Santowax-R at 207°C and 267°C are compared in figure 4. $\rho(\beta)$ can be divided approximately into three parts: a low-energy region where a peak is clearly indicated at 0.012 eV, an intermediate energy region where a large peak at 0.14 eV is indicated, though not completely defined by the data, and a high-energy region outside the measured range where the C-H vibrations have been approximated by a single δ function at 0.38 eV, which is the energy of the C-H vibrations in benzene as calculated by Crawford and Miller (1949). While preserving the normalization of $\rho(\beta)$

$$\int_0^\infty \rho(\beta) d\beta = 1 \quad (3)$$

the areas in the δ function and under the large $\rho(\beta)$ peak were varied; it was found that the best overall agreement with the experimental $S(\alpha, \beta)$ was obtained with a δ function of the form $0.35 \delta(\beta - \beta_i)$. In the case of the 23°C solid results the area under the fitted part of $\rho(\beta)$ was better defined and was taken to be 0.12. It was assumed that the remainder of $\rho(\beta)$ had the same form as for the liquid and the large peak in the liquid $\rho(\beta)$ was approximated by two δ functions to give a complete distribution of the form

$$\rho(\beta) = \underbrace{\rho(\beta)}_{\text{fitted}} + 0.09 \delta(\beta - 3.7) + 0.44 \delta(\beta - 5.5) + 0.35 \delta(\beta - 15).$$

This representation of $\rho(\beta)$ appears justified by the satisfactory agreement obtained between calculation and experiment.

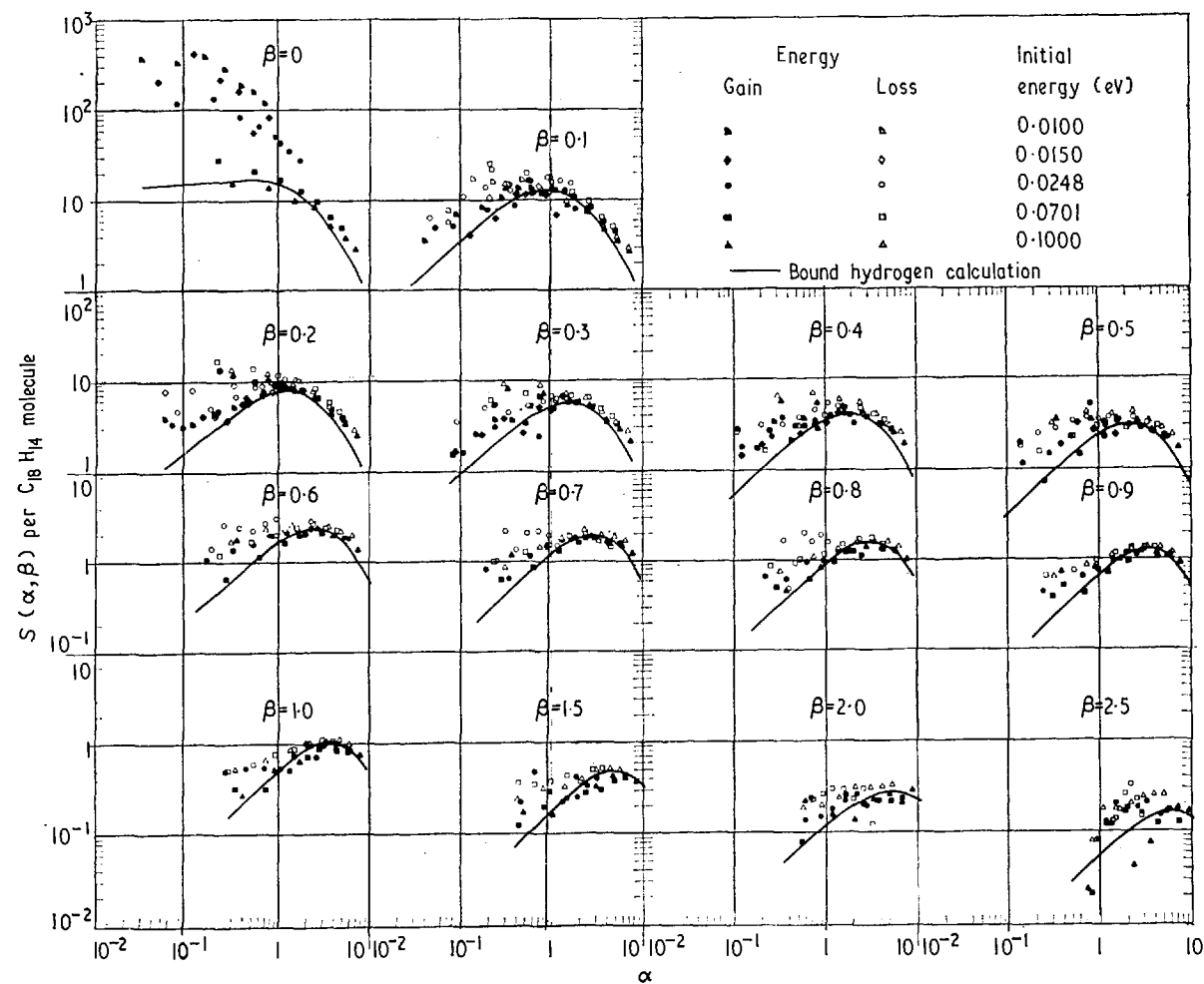


Figure 1. Calculated scattering law for solid Santowax-R at 23°C compared with the experimental data of Brugger (1962).

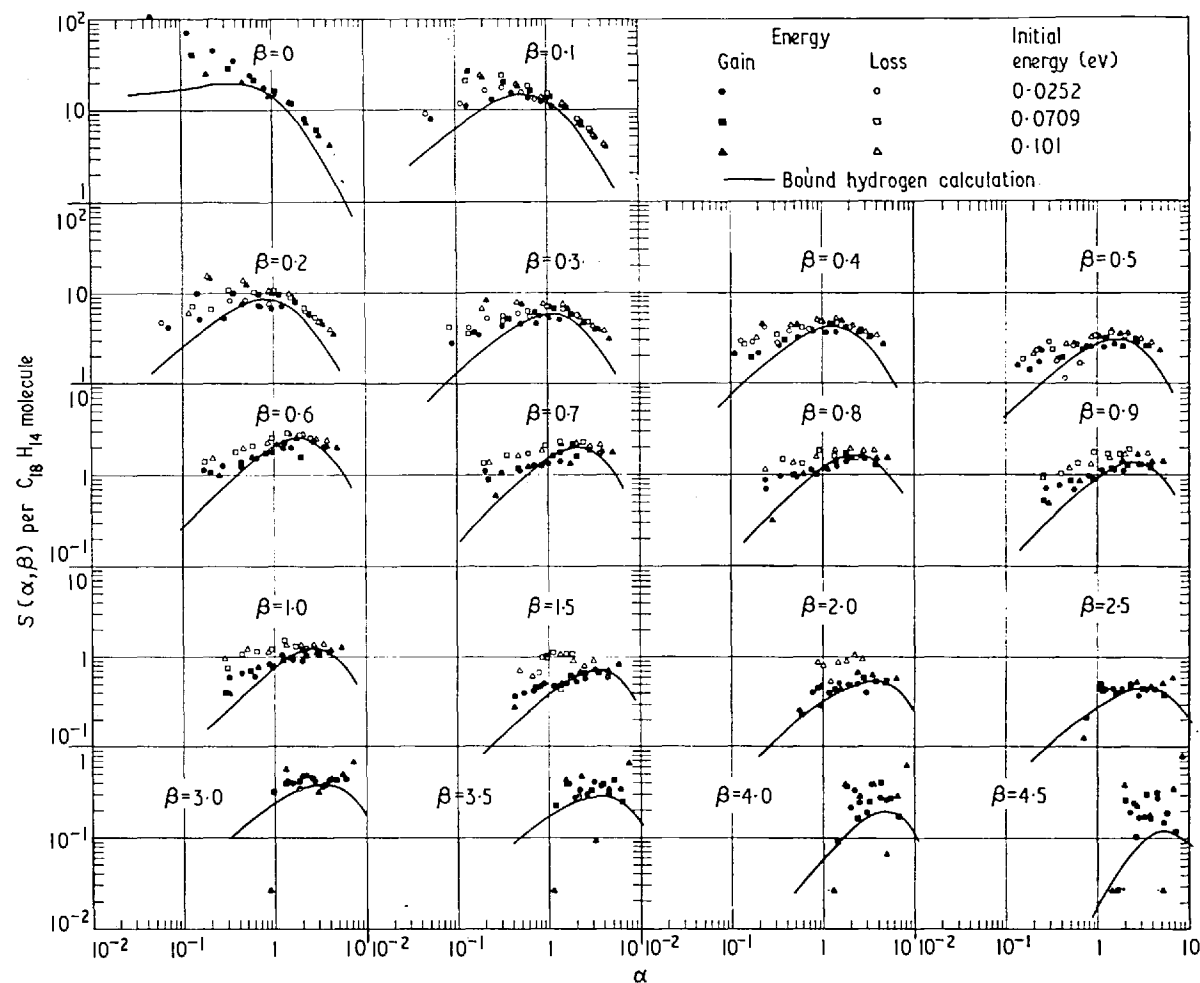


Figure 2. Calculated scattering law for liquid Santowax-R at 207°C compared with the experimental data of Brugger (1962).

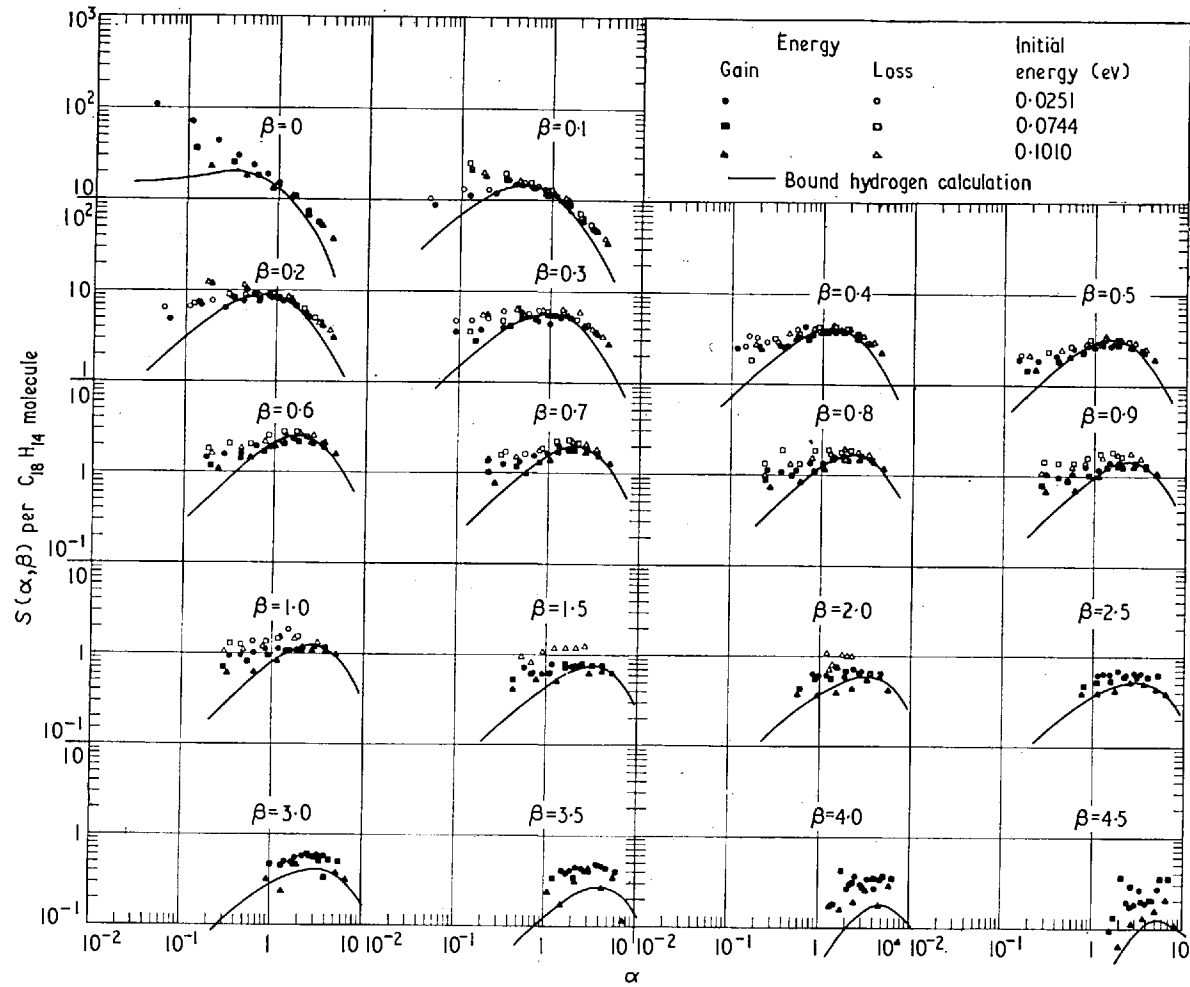


Figure 3. Calculated scattering law for liquid Santowax-R at 267°C compared with the experimental data of Brugger (1962).

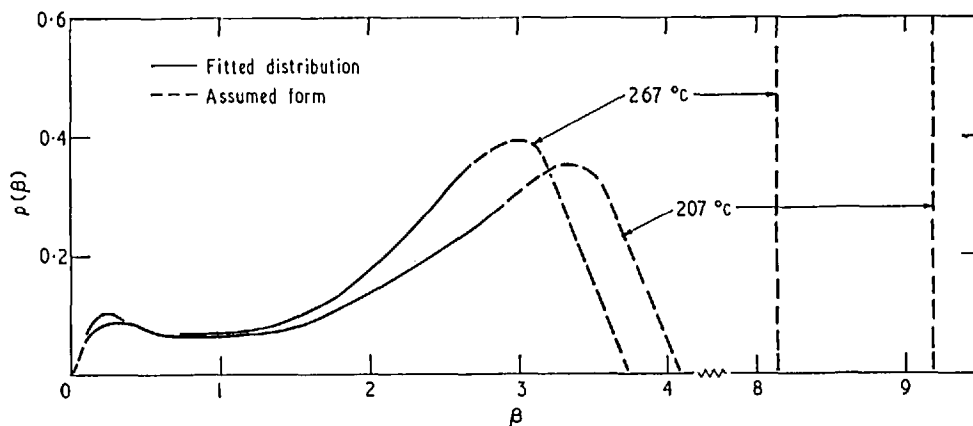


Figure 4. Generalized frequency distributions for 207°C and 267°C, used in the calculations in figures 2 and 3, plotted against dimensionless energy transfer $\beta = E/kT$.

No attempt has been made to assign an error to the fitted $\rho(\beta)$ as this would require knowledge of random and systematic errors in the experimental data besides an understanding of the importance of any non-Gaussian contribution to the scattering. Inspection of figures 1–3 shows that the contribution of random errors in the data amounts to approximately 15% at low β values and approximately 30% at high β values.

4. Comparison between solid and liquid

By plotting on a common scale, the fitted frequency distributions for the liquid and solid can be compared. This is done in figure 5. The two distributions are quite similar in the energy range 0.005–0.06 eV. In view of the possibly large errors in the fitted $\rho(\beta)$ the differences may not be significant, indicating that the frequency distribution observed for the liquid over this energy range is due to molecular vibrational motion.

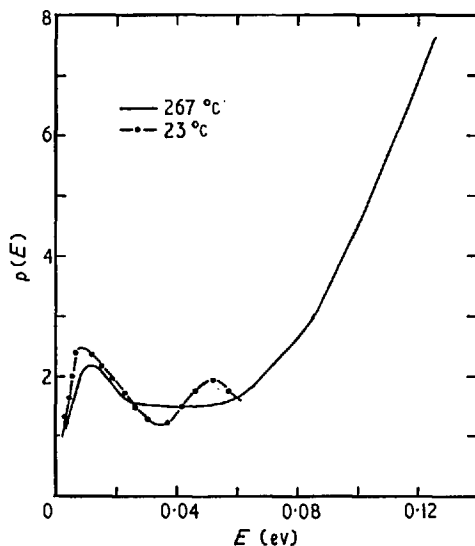


Figure 5. The fitted parts of the frequency distributions for solid (23°C) and liquid (267°C) Santowax-R plotted against a common energy scale.

5. Total scattering cross sections

The total scattering cross sections at 0.025 eV have been calculated from the proposed $\rho(\beta)$ distributions using the programme DIS (P. W. Johnson 1966, private communication). DIS will perform the necessary angular and energy integrations of equation (1) to give the total cross section from an input matrix of $S(\alpha, \beta)$ values. The angular integration employs double Gaussian quadrature while the energy integration uses a basic set of 32 energy points within which the integral between any two points was obtained by integrating the cubic interpolating polynomial through these points and an adjacent point on either side. In the absence of any treatment of diffusive motion the quasi-elastic scattering is simply the first term in the phonon series

$$S(\alpha, \beta) = e^{-\alpha\lambda} \delta(\beta) \quad (4)$$

where λ is the Debye-Waller coefficient which is computed by LEAP and is given by

$$\lambda = \int_0^{\infty} \frac{\coth \frac{1}{2}\beta}{\beta} \rho(\beta) d\beta.$$

Total cross sections per molecule as a function of energy have been calculated for the liquid at 207°C and 267°C and are shown in figure 6. Carbon has been assumed to scatter

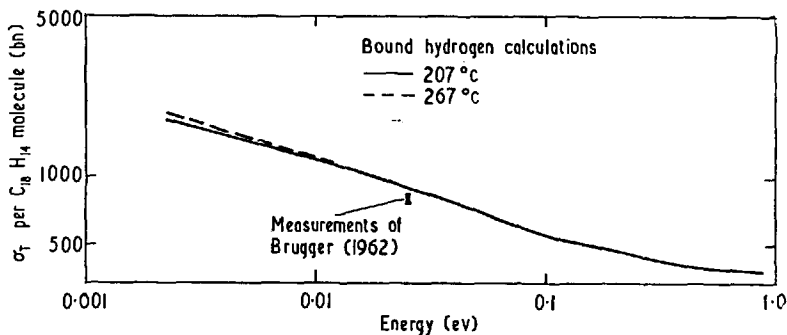


Figure 6. Calculated total cross section as a function of energy for liquid Santowax-R at 207°C and 267°C.

according to the free-gas model. Brugger (1962) has measured the total cross section at 0.025 eV and his results are compared with the calculated values in the table.

Total cross section per $C_{18}H_{14}$ molecule at 0.025 eV

Temperature (°C)	σ_T (bn) (Brugger 1962)	σ_T (bn) (calculated)	Debye-Waller coeff. λ
207	810 ± 24	901	2.91
267	791 ± 21	912	3.26

The differences between the measured and calculated total cross sections are four times greater than the experimental error and the trends with temperature appear to be in opposite directions. Among possible reasons for these discrepancies now under investigation are the omission of diffusive motion from the calculations and systematic errors in the fitting procedure.

6. Conclusions

It has been shown that reasonable agreement with scattering law data for Santowax-R can be obtained with methods based upon the Gaussian approximation for the motion of

the scattering atom. Although some features of the experimental data limited the range of the fitting process, the results suggest that the differences between the hydrogen atom motion in solid and liquid are small, and that only vibrational modes were observed. Total cross sections computed from the fitted models are approximately 12% greater than experimental values at 0.025 eV.

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A.J.H. Goddard
Ph.D. Material.

Neutron rethermalization studies in liquids using miniature fission chamber detectors

G. E. B. TREMEER†, A. J. H. GODDARD and P. J. GRANT

Nuclear Power Group, Department of Mechanical Engineering, Imperial College of Science and Technology, London, England

Abstract. Miniature ^{235}U and ^{239}Pu fission chambers have been used to separate the space and energy variations in neutron density in the region of a plane interface between a tank containing room-temperature water and a tank containing hot organic liquid, Santowax-R. A cadmium-shutter technique has been used to simulate a purely thermal-neutron source and an attempt was made to detect γ -ray-induced photofissions in the fission chambers. Methods used to monitor temperature-induced gain changes and to record reactor power fluctuations are discussed. Examples are given to illustrate the two types of analysis of the results that are currently in use, namely the two overlapping thermal-group and multi-group transport theory methods.

1. Introduction

The transport of thermal neutrons across a temperature discontinuity between two liquid moderators has been studied using the London University CONSORT reactor as a neutron source. This problem, besides being of fundamental interest in reactor physics, is of practical importance for reactor systems such as the steam generating heavy-water-moderated reactor, where the thermal-neutron flow in the cell is from the cold moderator to the hot fuel/coolant region.

Few experimental studies near temperature discontinuities have been reported and none has used only liquid moderators with well-defined source geometry. Bennett (1963) has studied cylindrical graphite-water systems in the core of the Hanford physical constants testing reactor. At the United Kingdom Atomic Energy Authority, Harwell, Day and Partridge (1964) have used both foil and time-of-flight techniques to study neutron spectrum changes within two adjacent graphite blocks maintained at different temperatures.

2. Mechanical design of equipment

The experiment was carried out on the 'bare face' of CONSORT, separated by a total of 32 in. of graphite from the reactor tank. Thermal- and fast-neutron fluxes at 100 kw reactor power were 5×10^9 and 10^9 neutrons/sec cm² respectively over an area of 9 ft². Figure 1 shows the general arrangement of the apparatus which consisted of two rectangular tanks of $\frac{1}{8}$ in. aluminium held together in a frame to form a unit, but thermally insulated from each other by a $\frac{1}{8}$ in. air gap, this gap being maintained by small insulating disks. The supporting frame was designed to accommodate differential thermal expansion between the two tanks. Some local stiffening of the interface wall with T-section aluminium strips was necessary, but this was 3 in. from the central measuring axis of the tanks. Each of the two tanks had a depth of 26 in. and a width of 24 in. and was filled to a depth of 24 in. with liquid. The 'cold' tank had a length of 4 in. and was normally filled with demineralized water; the 'hot' tank was 10 in. long and could be filled with water or Santowax-R (terphenyl).

The rear wall of the hot tank and the side walls of both tanks were covered with 0.015 in. cadmium sheet to provide a black boundary for thermal neutrons and these areas were also covered with 1 in. thick asbestos fibre thermal insulation. Above the tanks the thermal insulation was arranged on either side of the fission chamber traversing mechanism.

† On leave from the South African Atomic Energy Board.

The hot tank was heated from below by two 8 in. $\times$ 12 in. double-element hot plates having a maximum heating power of $5\frac{1}{2}$ kw but variable down to zero. A simple arrangement of a thermocouple operating a heavy duty contactor via an electronic potentiometer was used to control the temperatures. Demineralized water in the cold tank was kept within a few degrees of ambient temperature by circulating it through a heat exchanger outside the reactor shield. Cold water entered at the base of the tank and the displaced water flowed under gravity from an outlet pipe set 24 in. above the base of the tank to a surge tank and finally through a water-cooled heat exchanger before being returned to the cold tank. A total of ten chromel–alumel thermocouples were installed. Three were in the cold tank, at the water inlet, outlet and in a central position; the remainder were distributed through the hot tank in a pattern most likely to locate any temperature gradients.

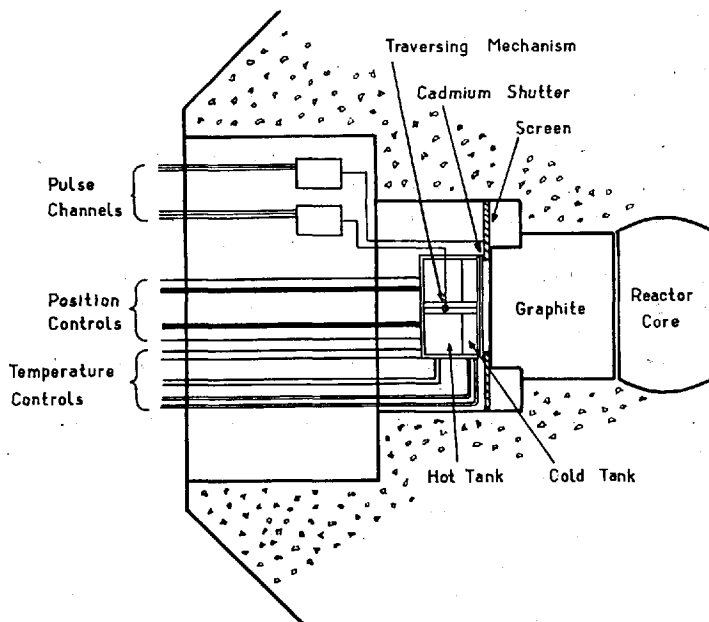


Figure 1. Arrangement of the experiment on the CONSORT reactor.

Because the neutron flux from the reactor contains a substantial proportion of fast neutrons and because the most straightforward analysis of the experiment required a pure thermal source to enter the cold tank, a negative source technique was adopted. A cadmium shutter was built in front of the apparatus and the difference was taken between neutrons detected with the shutter raised and lowered. Although such a technique can be very useful, care must be taken to ensure that moving the cadmium shutter produced no undesired effects. In order to prevent the shutter from affecting thermal neutrons which could enter the assembly from the side, a screen was constructed across the whole width of the cave having an aperture matching the dimensions of the tanks, across which the shutter moved. Both the screen and the shutter were of 0.015 in. cadmium sandwiched between aluminium sheets. The shutter ran between vertical guide rails and was raised and lowered by an electric motor.

3. Neutron detection

The two most convenient reaction-rate ratios that can be measured in a study of neutron temperatures using integral techniques are the $^{239}\text{Pu}/^{235}\text{U}$ fission-rate ratio and the $^{176}\text{Lu}/^{55}\text{Mn}$ reaction-rate ratio. The former involves the use of either foils or fission chambers while the latter is restricted to foil techniques only. Although the Lu/Mn ratio

is more sensitive to neutron temperature changes, the choice was made to use fission chambers because of the ease with which good counting statistics and accurate positioning can be achieved and any doubtful measurements repeated. In addition the necessity of removing a large quantity of shielding material to obtain access to the experiment would make a series of foil irradiations very time consuming.

The fission chambers (manufactured to order by 20th Century Electronics Ltd.) were suitable for operation up to 350°C. In order to check upon any systematic errors in the measurements due to the finite size of detectors, two pairs of chambers were used, $\frac{1}{4}$ in. and $\frac{1}{8}$ in. in diameter, with active regions 1.0 cm and 3.8 cm long respectively. Each chamber was constructed with a 36 in. long gas-filled Nimonic alloy extension.

The measuring fission chamber was moved by a traversing mechanism fixed above the tanks and connected by two flexible drives to a control box outside the reactor shield. Duplicate 1 mm pitch screws were provided on the control box and the traversing mechanism, so that manual rotation of the control box screw moved a slide against a scale in step with the movement of the fission chamber. The flexible drives were in two sections: a robust 16 ft length between the control box and a connection point at the base of the dual tank assembly, and a light 4 ft length for final connection to the mechanism. The former was sufficiently stiff to reduce backlash while the latter by its low weight minimized bending loads on the mechanism. Provision was made for moving the fission chamber both horizontally, to examine flux distributions normal to the tank interface, and vertically either to make flux measurements or to transfer the chamber from one tank to the other during an experiment. Cams at the end of the horizontal and vertical screws on the mechanism actuated microswitches once every revolution so that the movement of the fission chamber could be signalled in steps of 1 mm. The slides carrying the fission chambers incorporated polytetrafluoroethylene bearing material and ran on hard anodized aluminium bearing surfaces. A necessary feature of the vertical part of the mechanism was a high-temperature silicone rubber seal whose function was to wipe water or hot wax from the counter during transfer between the tanks.

An additional fission chamber was used as a reactor power monitor and was mounted in a tube between the cadmium screen and the end of the graphite extension from the reactor. This position was a compromise between the conflicting requirements of obtaining a suitable counting rate and avoiding effects due to the movement of the cadmium shutter. Both counting channels used conventional valve electronic equipment comprising Dynatron type 1430 head/main amplifiers and Ekco scaler/timer units.

4. Experimental procedure

A typical sequence of operations during a flux traverse with one counter was as follows. After running the heaters at maximum power to melt the wax and achieve the desired hot tank temperature, the reactor power was adjusted to give a counting rate on the experimental channel of approximately 10 000 counts per second at positions close to the tank interface. This counting rate gave good statistics with small dead-time corrections over the region of greatest interest. Before the start of the measurements, while the fission chamber was in the cold tank, a bias curve was recorded using the amplifier attenuator and this was repeated at the conclusion of the measurements with the chamber in the hot tank. The results of these gain checks are discussed in the next section.

A flux traverse was started in the cold tank with the shutter in the raised position. Proceeding from a nominal zero position counts were recorded on experimental and monitor channels at positions up to the interface, the points being spaced close together near the interface. This traverse was then repeated with the shutter down, after which the chamber was transferred to the hot tank and the count rates recorded at selected positions, again with the cadmium shutter in the raised and lowered positions.

Both before and after a traverse the fission chamber was positioned at the nominal zero of the mechanism, and the distance from the chamber to the interface was measured with a micrometer.

The different combinations of liquids and operating temperatures are shown in table 1 below.

Table 1. Liquids in the cold and hot tanks

Experiment	Cold tank liquid	Cold tank temperature (°C)	Hot tank liquid	Hot tank temperature (°C)
1	H ₂ O	17	H ₂ O	90
2	H ₂ O	17	Santowax-R	171
3	H ₂ O	17	Santowax-R	201
4	H ₂ O	17	Santowax-R	232

The range of Santowax temperatures was determined by the melting point of 145°C and the flame-point temperature of about 230°C.

5. Reduction of raw data

All essential corrections were applied to the data. There were no significant short-term variations of reactor power but at high powers the heating-up of the reactor cooling water produced slow changes in the monitor counting rate. The experimental channel dead time was measured by varying the reactor power over a wide range using each of the different experimental fission chambers and over different count-rate ranges. At very high counting rates the dead time was found to be count-rate dependent, increasing significantly above rates of 40 000 counts/sec. Measured dead times were used to correct the data; because of the steep thermal-neutron flux gradient the increased values only applied to a few experimental points near to the reactor face of the cold tank.

The difference between counts taken at each position with the shutter up and down gave the desired thermal-neutron reaction rate. Hot tank reaction rates were normalized to those in the cold tank on the basis of channel gain measurements made in both the cold and hot tanks. These measurements were undertaken because it was thought that heating of the gas column in the fission chambers might affect performance. In assessing the typical gain changes shown in table 2 the simple method of graphical superposition of attenuation bias curves has been used; the accuracy of this method is approximately 0.1 dB or 0.2% in counting rate.

Table 2. Typical measured gain changes produced by a temperature change 17–201°C

Fission chamber	e.h.t. (v)	Gain changes (dB)	Correction factor (%)
$\frac{1}{8}$ in. ²³⁵ U	500	-0.6	+0.8
$\frac{1}{8}$ in. ²³⁹ Pu	500	+0.5	-0.8
$\frac{1}{4}$ in. ²³⁵ U	250	-0.2	+0.4
$\frac{1}{4}$ in. ²³⁵ U	500	-1.0	+1.5
$\frac{1}{4}$ in. ²³⁹ Pu	500	0.0	0.0

Although some gain changes were significant, no systematic trend was observed. After an initial experiment an e.h.t. voltage of 500 v was used for $\frac{1}{4}$ in. chambers; at 250 v there was a small change in bias curve shape with temperature although the corresponding change in gain was still very small.

6. Systematic errors in the measurements

Two possible systematic errors in the measurements arose from the negative source technique. Although primarily attenuating thermal neutrons, the shutter of cadmium sandwiched between aluminium sheets had a finite cross section in the epithermal and fast energy ranges, thus giving the negative source a small fast component. The calculated proportion of the fast component making a collision in the shutter is 6.8%, resulting in a

typical contribution to the total difference counting rate of 1.2%. A second error could arise from additional capture gamma radiation from cadmium with the shutter in the 'down' position giving photofission counts in the measuring chamber. Thresholds for photoneutron production are 5.5 MeV in ^{239}Pu and 5.75 MeV in ^{235}U , and a significant proportion of cadmium radiative capture gamma rays exceed these energies. An experimental test was made using a ^{239}Pu chamber as the experimental counter by placing a screen, comprising a $\frac{3}{8}$ in. wide boric-acid-filled gap between hardboard sheets, immediately in front of the cold tank. From the change in counting rate produced by lowering the cadmium shutter in front of the boron screen it was shown that the photofission contribution must be less than 4.9% of the fast flux or typically less than 1% of the difference counting rate.

The greatest difference between results from $\frac{1}{4}$ in. and $\frac{1}{8}$ in. detectors arose from systematic positioning errors of the $\frac{1}{8}$ in. detectors in the hot tank; these errors have been shown to be due to temperature-induced internal gas pressure changes straightening out small bends in the rather flexible counter stems. When allowance was made for these effects, no systematic differences were observed between reaction-rate distributions obtained with $\frac{1}{4}$ in. and $\frac{1}{8}$ in. detectors.

7. Interpretation and analysis of results

A typical fission-rate distribution obtained with a ^{235}U chamber is shown in figure 2. Away from the interface the distribution is determined by the thermal diffusion lengths in the two liquids. Near the interface transients are produced by the change in the effective cross section of ^{235}U . The effective cross section of ^{239}Pu is less sensitive to neutron temperature, resulting in smaller transients at the interface.

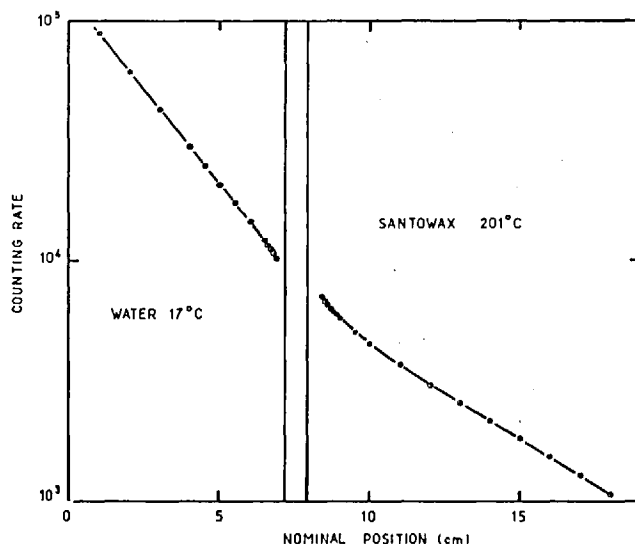


Figure 2. ^{235}U fission-rate distribution measured with a $\frac{1}{4}$ in. detector.

Figures 3 and 4 show fission-rate distributions, expressed as deviations from the unperturbed exponential form, for hot tank temperatures of 171 and 232°C and for each of the four detectors used. In the hot tank the $\frac{1}{8}$ in. and $\frac{1}{4}$ in. measurements are separated because corrections for the position error, discussed in the previous section, have not been applied. It will be seen that transients of the order of 3% amplitude and extending over about 2 cm are clearly defined. One method of analysis of the experimental results consists of fitting to individual fission-rate distributions, by the method of least squares, two overlapping thermal group equations (Bennett 1963) and figures 3 and 4 show that this is

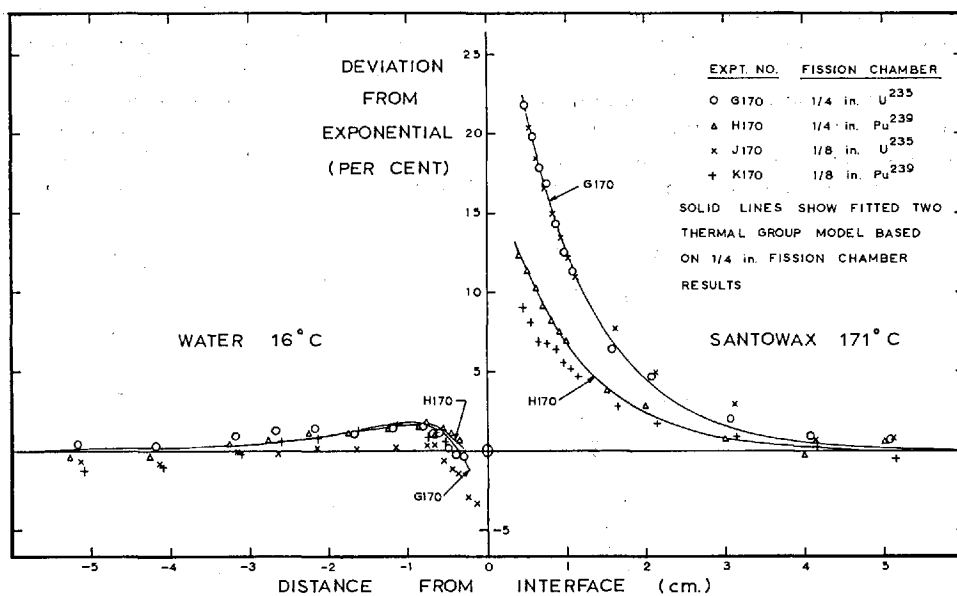


Figure 3. Transients across a temperature step between water at 16°C and Santowax at 171°C.

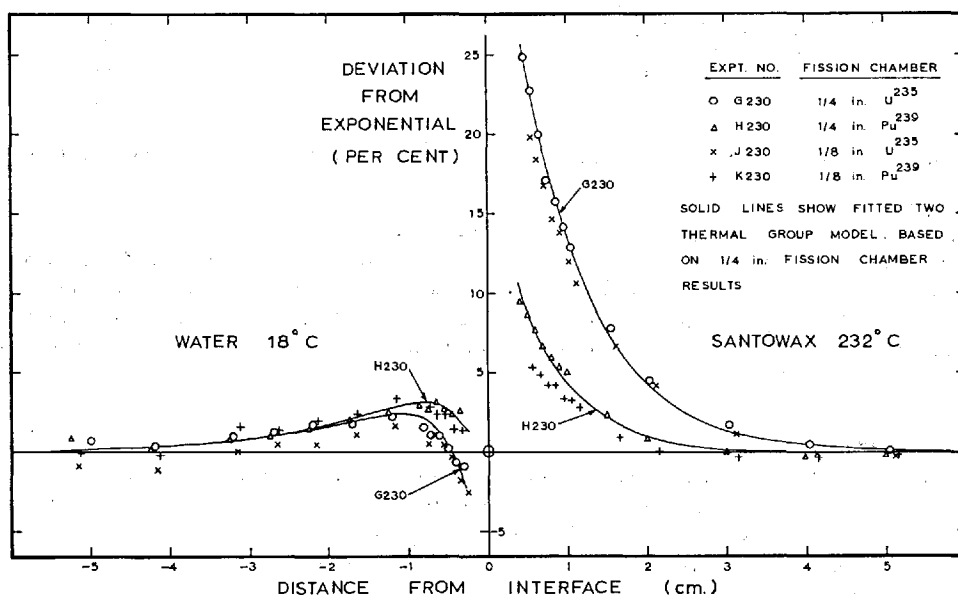


Figure 4. Transients across a temperature step between water at 18°C and Santowax at 232°C.

possible to a high degree of accuracy. This procedure yields rethermalization lengths and cross sections which can be used in two thermal group reactor cell calculations or compared with predicted values.

Figure 5 shows the fission-rate ratio $^{239}\text{Pu}/^{235}\text{U}$ measured with $\frac{1}{4}$ in. detectors with the hot tank at 201°C, normalized to unity in the cold tank. Although a conventional 'thermal calibration' of the pairs of fission chambers in a known Maxwellian spectrum was not carried out, the measurements in the cold tank away from the interface are closely equivalent to such a calibration, for the fission ratio per atom in water calculated using multi-group methods (see below) differed by less than 0.5% from the calculated ratio in a Maxwellian spectrum.

Evidence that a purely thermal distribution was established is provided by the fact that, for the case shown in figure 5, the change in fission-rate ratio between hot and cold tanks away from the interface agrees within the experimental errors with the calculated change, 23.8%. A second essential requirement was that the distribution in the cold tank should yield a thermal-neutron diffusion length for water in agreement with the best accepted value; this was the case. As an alternative analytical method the Winfrith DSN (transport theory code) (Askew and Brissenden 1963) is being used with 31 energy groups to predict

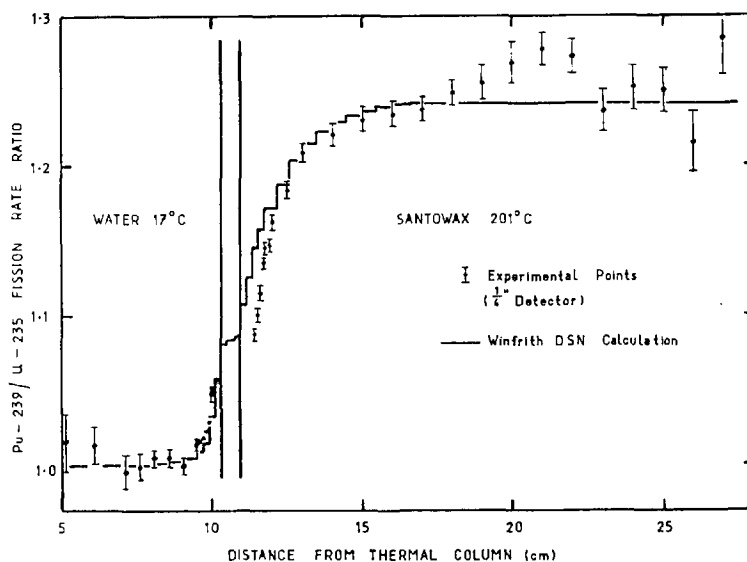


Figure 5. Measured $^{239}\text{Pu}/^{235}\text{U}$ fission-rate ratio compared with the Winfrith DSN calculation.

fission-rate distributions and figure 5 shows the fission-rate ratio distribution calculated in the DS4 approximation and with hydrogen in both water and Santowax scattering according to the free gas model. The differences between this calculation and experiment are not large, but are significant near to the interface. The calculations are to be repeated with proper treatment of the effect of hydrogen molecular binding upon the scattering cross sections.

8. Conclusions

It has been shown that a negative source technique can be used to stimulate a thermal-neutron source in the presence of a mixed spectrum and that miniature fission chamber detectors can be used successfully to study thermal spectrum transients in liquid moderators. The results obtained are of practical interest for reactor physics design besides yielding basic information on neutron rethermalization.

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