THERMAL NEUTRON CROSS SECTIONS AND DIFFUSION PARAMETERS OF DIPHENYL AND DOWTHERM-A USING A SYNTHETIC SCATTERING FUNCTION

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We have used a synthetic scattering function to evaluate thermal neutron cross sections and diffusion parameters for Diphenyl and Dowtherm-A, over a range of temperatures of practical interest. The present formalism is based on our benzene model, which has been already validated in a previous application. Our calculated results for Diphenyl are in excellent agreement with several measured quantities, whereas those for Dowtherm-A are at variance with the scarce available information.

1. Introduction

The availability of neutron scattering cross section data is a basic requirement to perform reactor physics calculations. In particular, the cross sections of moderator materials play the most important role in the neutron thermalization process. First-principles theories were developed in the past to describe the slow neutron - molecule interaction, most of them based on the essentially exact Zemach-Glauber formalism [1]. However, those theoretical approaches usually require a detailed information about the system under study, and the resulting computer codes - while producing unnecesarily rich descriptions of the scattering process for reactor calculation purposes - are rather expensive in terms of computing time. For those reasons, standard Nuclear Data Libraries contain scattering cross sections only for the most common moderators, such as water or graphite, but not for other hydrogeneous molecules such as Dodecane, Diphenyl, Butyl-phosphate, etc., which are also of great interest from a practical point of view. A procedure frequently adopted to handle these hydrogeneous systems is to substitute the (non existent) actual Hydrogen microscopic data by those of Hydrogen bounded in water, while the rest of atomic species are treated as a free gas. This leads to the evaluation of 'estimators' for the real values, but the effect of such a procedure can hardly be properly established.

In the last few years there has been considerable interest in the concept of High Conversion Light Water Reactors (HCLWR), motivated by its potential advantages concerning fuel utilization. Much work has been already done in this field, but unacceptable

discrepancies have been observed in the main reactor physics parameters, especially in the void coefficient [2-4]. Recently, a series of experiments were carried out at the PROTEUS-LWHCR facility [5-8] aimed to obtain some integral magnitudes in a typical HCLWR lattice, using Dowtherm-A to simulate an intermediate voidage state.

In this work, we have applied a synthetic scattering function (SSF) [9] to represent the scattering law of Diphenyl and Dowtherm-A, taking advantage of its flexibility to describe any hydrogeneous molecule and the analytic character of the resultant expressions for scattering kernels and total cross sections [10]. We present here our evaluated results for several integral magnitudes and thermalization constants, and their comparison with experimental values and other calculations for those molecules. A closely related system, benzene, is considered as starting point for our Diphenyl and Dowtherm-A models, bearing in mind their structural similarity and the fact that the SSF for benzene has been already validated [11].

In a forthcoming contribution we will present reactor calculations for different types of HCLWRs, using our evaluated Dowtherm data and simulated 'Dowtherm' data to show and quantify the influence of the materials replacement procedure mentioned above.

2. The model for Diphenyl and Dowtherm-A

The basic hypothesis and approximations involved in the SSF have been described previously [9,10], as well as some of its applications to common neutron moderator materials [11-13], so

here we will only summarize its main features.

In terms of the synthetic model, the double-differential scattering cross section of a molecular unit is written as

$$\frac{\partial^2\sigma}{\partial\omega\,\partial\Omega} = \frac{k'}{k} \sum_{\nu} N_{\nu} \frac{\sigma_{\nu}}{4\pi} T_{\nu}(Q,\omega;E) \quad , \tag{1}$$

where k and k' denote the modulus of incident and scattered neutron wavevectors, respectively, ν runs over all the nuclear species of the molecule and N_{ν} is the number of nuclides dynamically equivalent, each of them with a bound scattering cross section σ_{ν}. Finally, $T_{\nu}(Q,\omega;E)$ represents the basic expression of the SSF:

$$T(Q,\omega;E) = S_{\mu\tau\Gamma}(Q,\omega) - \sum_{\lambda} \frac{P_{\lambda}}{\hbar\omega_{\lambda}\, M_{\lambda}} \left[n_{\lambda} \frac{\partial}{\partial\Gamma} S_{\mu\tau\Gamma}(Q_{\lambda}^{+},\omega_{\lambda}^{+}) + (1 + n_{\lambda}) \frac{\partial}{\partial\Gamma} S_{\mu\tau\Gamma}(Q_{\lambda}^{-},\omega_{\lambda}^{-}) \right] , \tag{2}$$

where $S_{\mu\tau\Gamma}(Q,\omega)$ is the scattering law for the neutron interaction with a quasi-rigid molecule, in which a momentum $\hbar Q$ and energy $\hbar\omega$ have been exchanged. The quantities μ, τ and Γ represent the effective mass, temperature and vibrational factor, respectively, that the scattering nucleus would present in the interaction, and they depend explicitely on the incident neutron energy E.

The second term on the right-hand side of Eq.(2) is a corrective one which stands for processes where the neutron exchanges energy with the λ internal modes of the molecule, by creating or annihilating one phonon. The summation over inelastic

processes is performed under the assumption that each internal mode λ is represented by an Einstein oscillator, with eigenfrequency ω_λ and effective mass M_λ. These quantities are obtained from a realistic frequency spectrum and, in particular, the effective masses associated to each of those motions are taken from the areas of the corresponding part of it, and constrained to satisfy a proper normalization condition [9].

The reduced number of input parameters required for the SSF and the analytic character of the derived expressions, make this formalism a powerful and practical tool for describing the slow neutron - molecule interaction. In particular, it provides analytic expressions for the l-th scattering kernels

$$\sigma_l(E,E') = 2\pi \int_0^\pi \sin(\theta)\, \cos^l(\theta)\, \frac{\partial^2\sigma}{\partial\omega\, \partial\Omega}\, d\theta \quad , \qquad (3)$$

the total scattering cross section

$$\sigma_s(E) = \sigma_0(E) = \int_0^\infty \sigma_0(E,E')\, dE' \quad , \qquad (4)$$

and the average cosine of the scattering angle

$$< \cos(\theta) >_{(E)} = \frac{\sigma_1(E)}{\sigma_0(E)} \quad , \qquad (5)$$

The molecules studied in this work were Diphenyl [C_5H_6-C_6H_5], and Dowtherm-A [(C_5H_6-O-C_6H_5) * 0.735 + (C_5H_6-C_6H_5) * 0.365] and, as stated before, we also considered benzene as a reference. Figure 1 shows the geometrical conformation of the molecular unit in each of those systems.

In the equilibrium configuration, the benzene molecule presents an hexagonal planar geometry, while its frequency spectrum shows two vibrational peaks around 0.12 eV and 0.38 eV [14], and a rotational band at very low energies which is completely excited at room temperature.

Diphenyl is also a planar molecule, formed by two benzene rings joined at two carbons. Its vibrational spectrum is similar to that of benzene [15,16], the most significant difference arising between 0.012 eV and 0.05 eV, as the equivalent rotational mode for this molecule appears at higher energies.

Dowtherm-A is an eutectic containing 26.5 wt% of Diphenyl and the rest of Diphenyl oxide. The structure of the latter is similar to the Diphenyl molecule, except that the link between the two benzene rings is provided by an oxygen. Based on this fact, we have considered the frequency spectrum of Diphenyl oxide equal to that corresponding to Diphenyl [17], as far as the hydrogen contributions are concerned.

Following those ideas, our model for Diphenyl contains the same vibrational energies and effective masses as that for benzene [11]; another low energy (rotational) mode was included at 0.017 eV, with a Sachs-Teller mass value taken from Refs. [15,16] and a molecular mass of 77.0 (neutron mass units).

To apply the SSF to Diphenyl oxide, we assumed that the hydrogen and carbon atoms behave in the same manner as in the Diphenyl molecule, whereas the oxygen contribution has been simply represented by that of the carbons but normalized to the proper cross section. This latter approximation is well justified on the basis of their similar masses and the smallness of their contributions to the total scattering as compared to that of the

hydrogen nuclei.

3. Calculations and results

In Table I we present the complete set of input parameters required to evaluate the neutron cross sections of benzene, Diphenyl, and Dowtherm-A according to the synthetic model.

Figure 2 shows the total cross section of benzene over the thermal neutron energy range, where our evaluated curve is compared with the experimental points of Sprevak *et al.* [18]. The SSF results are in excellent agreement with the data, except in the vicinity of 0.1 eV where it produces a structure not observed in the measurement. This behaviour is associated with the crude approximations borned into the model by the use of Einstein oscillators to represent an actually continuous spectrum; however, those shortcomings of the SSF do not spoil its overall performance in terms of simplicity, speed, and accuracy [19]. Comparison of this model results with other neutron scattering and thermalization properties of benzene, also shows a very satisfactory agreement [11].

Our evaluated scattering kernel, Eq.(3), for Diphenyl at room temperature is shown in Fig.3(a), whereas Fig.3(b) shows the ratio {Bencene/Diphenyl} of that magnitude per hydrogen atom. Over a large portion of the (E_i, E_f) plane both kernels are almost equal, as the vibrational modes adopted for both molecules are essentially the same except for a small difference in the width of the lower energy mode (see Table I). This causes the features observed in the low-energy corner of Fig.3(b), which display the difference in the (up/down) scattering power of those systems in

that energy region.

The total cross section (per Hydrogen atom) of Diphenyl at room temperature is shown if Fig.4, where our model calculations are compared with the experimental results of Antonini and Paoletti [20] and the calculations performed by Sprevak *et al.*[15]. Once again, the agreement between the SSF results and the experimental data is highly satisfactory.

We have also calculated the average cosine of the scattering angle and the diffusion constant as a function of energy:

$$D_0(E) = \left[3\, N\, \sigma_s(E)\, [1 - \cos(\theta)] + \sigma_a \right]^{-1} , \qquad (6)$$

where

$$\sigma_s(E) = \sum_j N_j \sigma_j \qquad \text{and}$$

$$N = N_A \frac{\rho}{M_{Mol}}$$

The magnitudes defined by Eqs.(5) and (6) are shown in Figs.5 and 6, respectively, for Diphenyl at 21^0C and 140^0C, together with the corresponding experimental values [21]. The integral diffusion constant

$$D_0 = (K_B T)^{-2} \int_0^\infty D_0(E) \exp(-E/K_B T)\, dE , \qquad (7)$$

is presented in Fig.7 for Diphenyl over a wide range of temperatures. The SSF evaluation is compared with experimental data from Refs.[21-23], showing a very good agreement. The density values used in this calculation are given in Table II, while our

results for the main diffusion parameters of Diphenyl at 21°C are summarized in Table III. It must be emphasized that little changes in the (lower energy mode) parameters of the model, constrained to preserve its agreement with the observed total cross section [20], produce changes of the order of a few percents in the calculated diffusion parameters.

In the case of Dowtherm-A, there is less information available. The diffusion constant for this material is shown in Fig.8, where the measured and calculated values of Sefidvash and Grant [16] and the experimental points of Küchle [26] have been plotted. Also, the evaluated curve obtained from the SSF for Dowtherm-A is presented in this figure, showing consistently higher values. For the sake of completeness, our predicted diffusion parameters for this material at 21°C are also given in Table III, whereas the calculated total cross section at the same temperature is given in Table IV.

Bearing in mind that the hydrogen cross section is much higher than those of carbon and oxygen, and considering the structural similarity of the three molecular units studied in this work, we decided to reduce all calculated and experimental values of the diffusion constant to a density removed magnitude, taking Diphenyl as the reference:

$$D_0^* = \rho \, D_0 \, (N_H / M_{Mol}) \cdot (M_{Mol}^{Dip} / 10) \quad , \qquad (8)$$

where ρ is the density of the substance, and N_H is the number of hydrogen atoms in the molecule with molecular weight M_{mol}. The result of applying this procedure to the available information on Bencene, Diphenyl, and Dowtherm-A is shown in Fig.9 in the form of

D_0° *versus* temperature. Clearly, there is a very good agreement between all those data points measured on benzene and Diphenyl and the SSF evaluation. However, a systematic discrepancy there exists between the above data sets and the measured and calculated values of Refs. [16] and [26] performed on Dowtherm-A. This conflicting situation is well demonstrated by the fact that the Sefidvash-Grant model, optimized for Dowtherm, gives values for the diffusion constant of Diphenyl [16] which are 10% lower than the experimental data (see Fig.7); on the other hand, our SSF evaluation which proved to be in very good agreement with different measured magnitudes for both benzene and Diphenyl, produces results for the diffusion constant of Dowtherm-A which are consistently higher than the measurements of Küchle and Sefidvash-Grant.

4. Conclusions

Based on its successful applications to several common moderators, benzene in particular, we have presented in this work evaluated thermal neutron cross sections and diffusion parameters for Diphenyl and Dowtherm-A produced by a Synthetic Scattering Function. The reduced number of physically meaningful parameters which are required to model any molecule in the frame of the this formalism, makes the SSF a very useful tool to tackle these kinds of problems.

In the specific case of Dowtherm-A, our evaluated results - based on input parameters well validated by its applications to

benzene and Diphenyl - are at variance with the scarce available information. Additional experimental results are required to settle this discrepancy.

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Figure Captions

Fig. 1: Structural conformation of the molecular units considered in this work. Filled squares represent carbon atoms, circles represent hydrogens, and the (filled) hexagon in c) represents the oxygen atom.

Fig. 2: Total cross section of benzene over the thermal neutron energy range. The experimental points are from Ref.[18]. See text for details.

Fig. 3: a) The energy-transfer scattering kernel for Diphenyl at room temperature, according to the Synthetic model. The group structure displayed corresponds to that of WIMS for the thermal range.

b) Ratio of the scattering kernels for Benzene and Diphenyl, per hydrogen atom and over the same energy mesh as in a).

Fig. 4: The total cross section (per hydrogen atom) of Diphenyl at room temperature over the thermal neutron energy region. The experimental data are from Ref. [20], whereas the dashed line represents a GASKET-FLANGE calculation [15].

Fig. 5: The energy dependent diffusion constant (Eq.6) of Diphenyl at two temperatures. The experimental points are from Ref. [21].

Fig. 6: Average cosine of the scattering angle for Diphenyl at two temperatures. The experimental points are from Ref. [21].

Fig. 7: The (density removed) diffusion constant of Diphenyl over a wide temperature range. The dashed line represents the Synthetic model results, while the experimental points are from Refs.[21-23].

Fig. 8: The diffusion constant of Dowtherm-A as a function of temperature. The dashed line is the prediction of the

Synthetic model for that magnitude. The experimental data are from Refs.[16] and [26]. See text for details.

Fig. 9: Diffusion constant for the three polyphenyls considered here, reduced according to Eq.8 and taking Diphenyl as a reference. The upper set of experimental points is from Refs. [21-23], whereas the lower set is from Refs. [16] and [24]. See text for details.

TABLE I. Values of the parameters for the synthetic model used in the calculations. Energies are given in eV, masses in neutron mass units and cross sections in barns.

	Benzene		Diphenyl		Dowtherm-A	
	H	C	H	C	H	C
$\hbar w_1$	0.120		0.017		0.017	
$\hbar \sigma_1$	0.030		0.0098		0.0098	
M_1	1.531	17.67	21.3	386	21.0	285
$\hbar w_2$	0.380		0.120		0.120	
$\hbar \sigma_2$	0.018		0.040		0.040	
M_2	3.345	392.3	1.55	15.55	1.55	15.55
$\hbar w_3$			0.380		0.380	
$\hbar \sigma_3$			0.018		0.018	
M_3			3.387	289.8	3.387	289.8
#at.	6	6	10	12	10	12
σ_{bound}	81.66	5.55	81.66	5.55	81.66	5.81
M_{mol}	20.94	42.20	77.0		82.88	
$\sigma_{abs.}$	0.33	---	0.33	---	0.33	---

TABLE II. Density values used for diphenyl in these calculations.

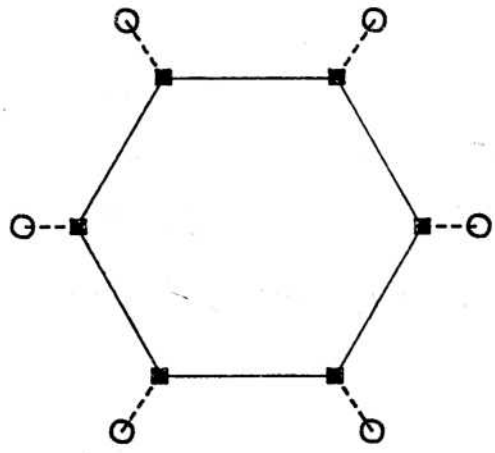
T (°C)	ρ (gr/cm^3)
21	1.0620
80	0.9862
110	0.9619
150	0.9295
207	0.8840
243	0.8850

TABLE III. Measured and calculated values of diffusion parameters near room temperature.

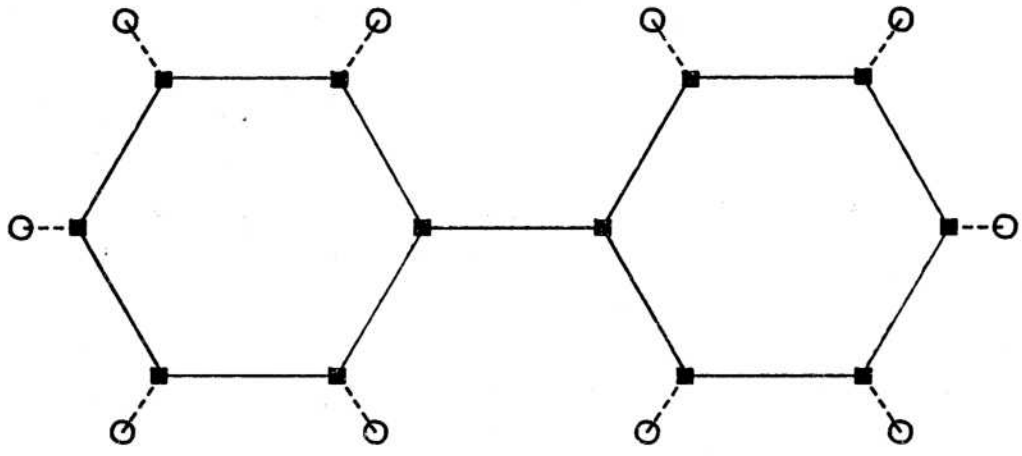
Reference	$\bar{v}\bar{\Sigma}_a$ (sec^{-1})	ρD_o (10^3 $\frac{gr}{cm\ sec}$)	$C(10^4\ \frac{cm^4}{sec})$	T(°C)
DIPHENYL				
21	2835	50.8 ± 0.93		21
23	3114 ± 26	51.6 ± 0.7	2.19 ± .29	24
24	3497 ± 281	45.6 ± 2.0	1.33 ± .29	21
25	3704 ± 150	44.3 ± 1.7	0.77 ± .18	20
this work	3038	51.9	1.123	21
DOWTHERM				
this work	2859	55.3	1.361	20

TABLE IV. Total cross section in barns for hydrogen in Dowtherm-A at 21•C calculated with the present model.

Energy (eV)	σ_{tot} for H (barn)
1.00 E-4	2.00 E+2
1.78 E-4	1.59 E+2
3.16 E-4	1.30 E+2
5.62 E-4	1.11 E+2
1.00 E-3	9.78 E+1
1.47 E-3	9.16 E+1
2.15 E-3	8.65 E+1
3.16 E-3	8.20 E+1
3.83 E-3	7.97 E+1
4.64 E-3	7.74 E+1
5.62 E-3	7.50 E+1
6.81 E-3	7.24 E+1
8.25 E-3	7.02 E+1
1.00 E-2	6.80 E+1
1.47 E-2	6.35 E+1
2.15 E-2	5.85 E+1
2.61 E-2	5.57 E+1
3.16 E-2	5.26 E+1
3.83 E-2	4.90 E+1
4.64 E-2	4.53 E+1
5.62 E-2	4.14 E+1
6.81 E-2	3.73 E+1
8.25 E-2	3.47 E+1
1.00 E-1	3.47 E+1
1.21 E-1	3.27 E+1
1.47 E-1	2.97 E+1
1.78 E-1	2.75 E+1
2.15 E-1	2.63 E+1
3.16 E-1	2.43 E+1
4.64 E-1	2.26 E+1
5.62 E-1	2.22 E+1
6.82 E-1	2.19 E+1
8.25 E-1	2.17 E+1
1.00 E+0	2.14 E+1
1.78 E+0	2.10 E+1



a) Benzene



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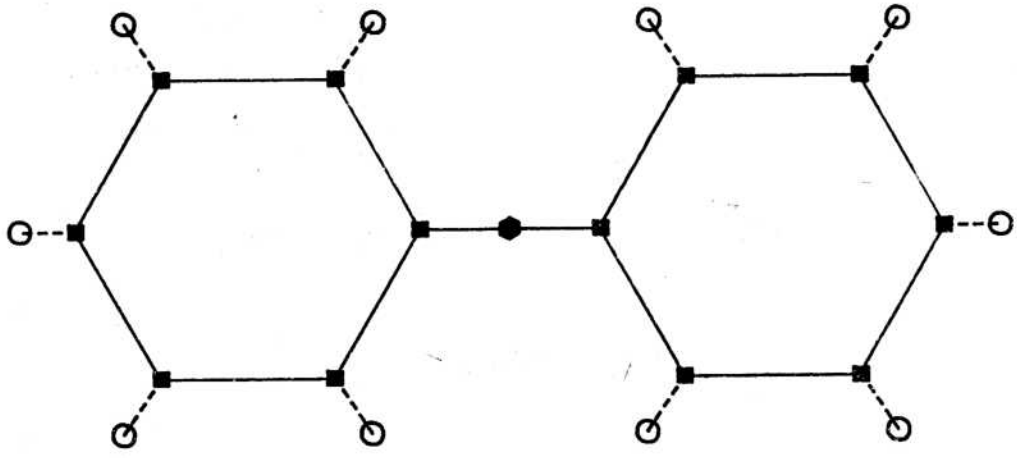
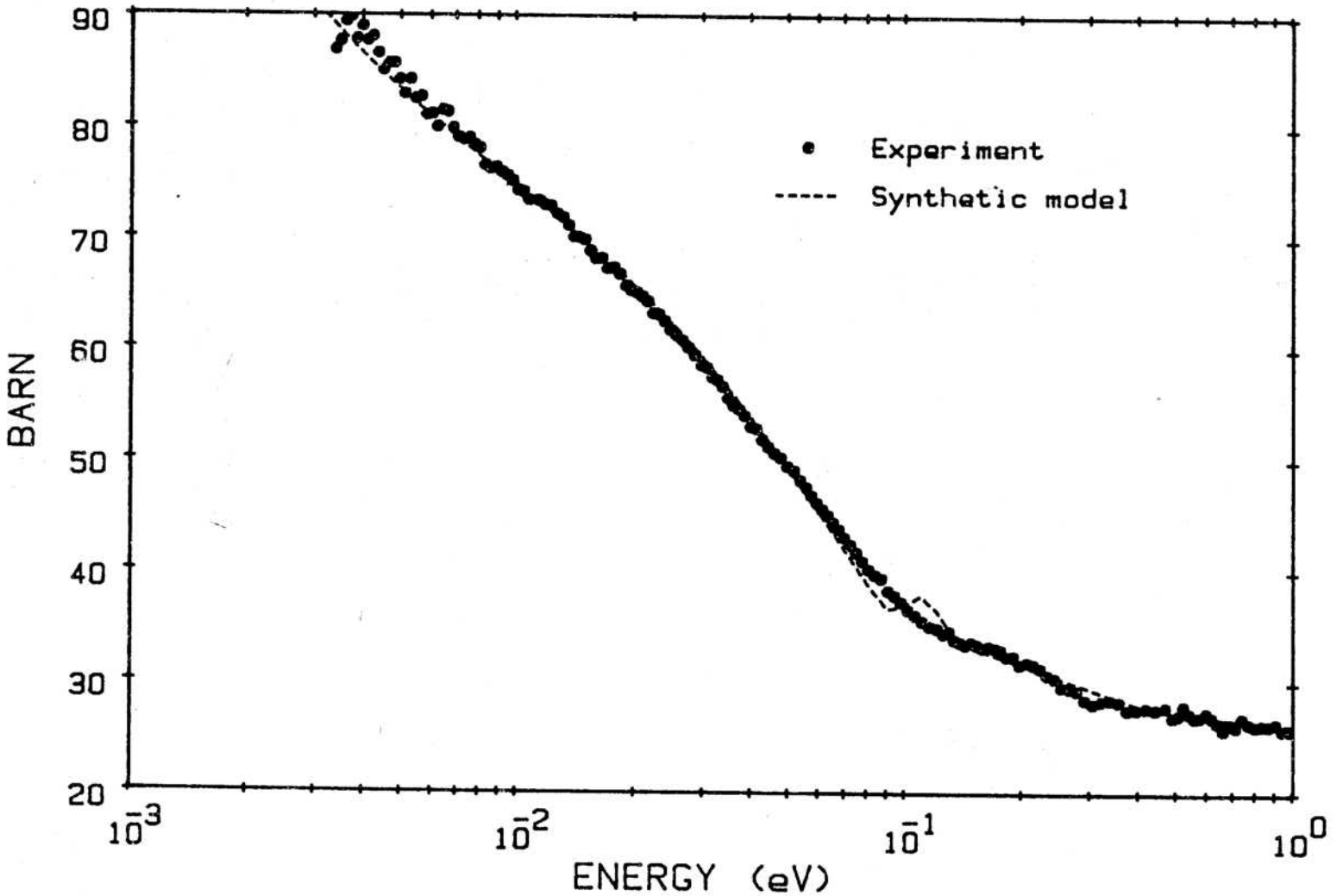


c) Diphenyl oxide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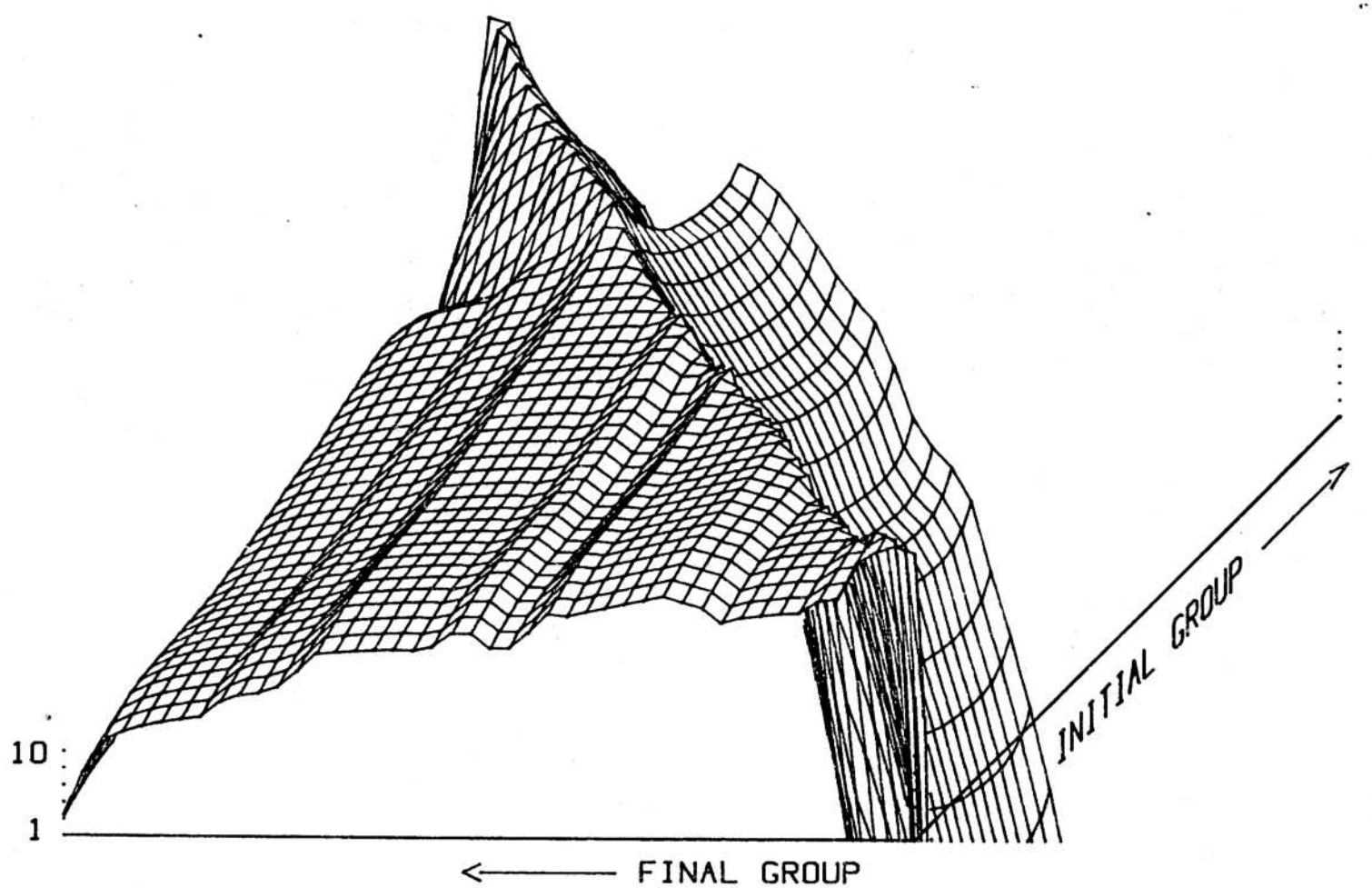
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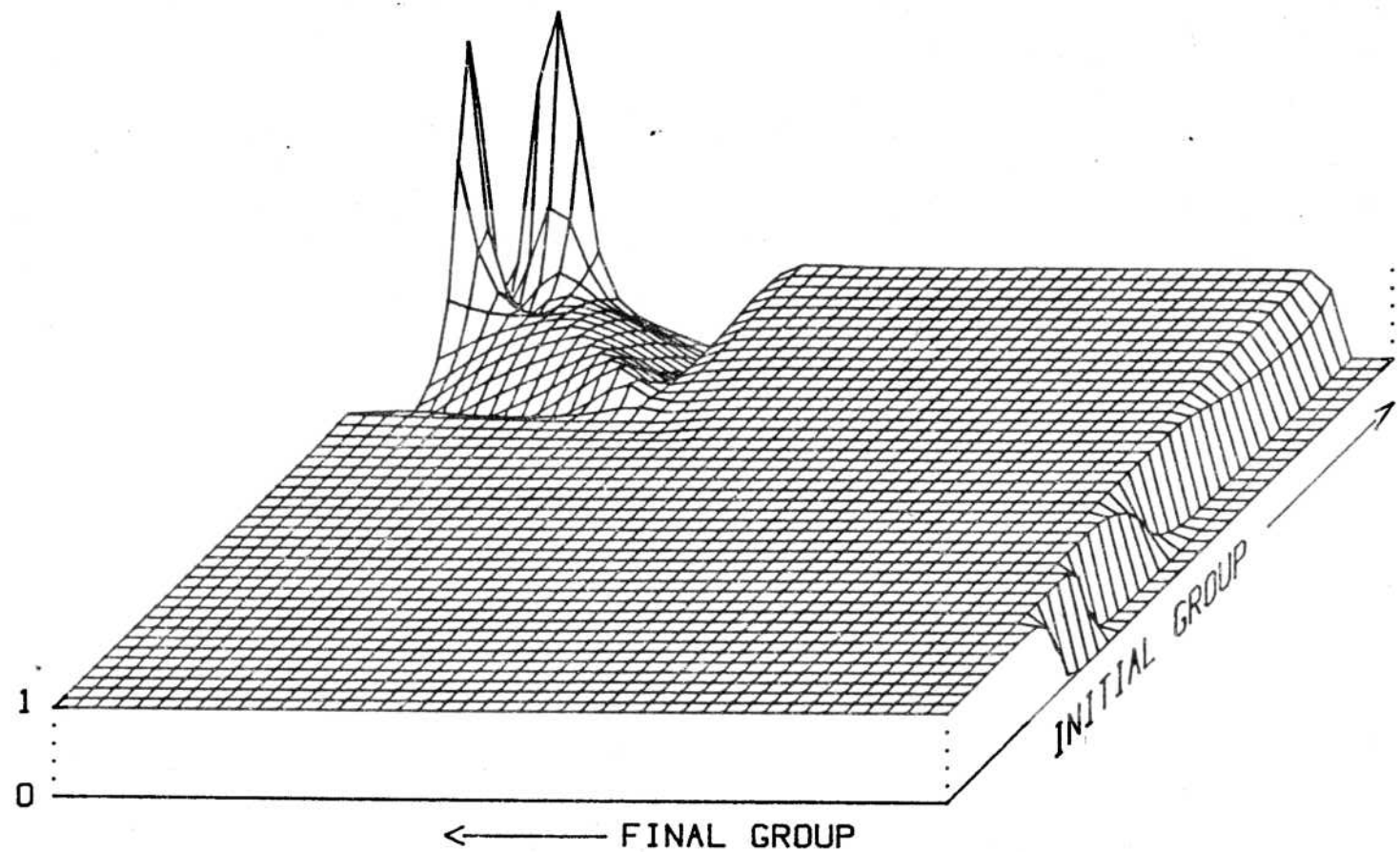
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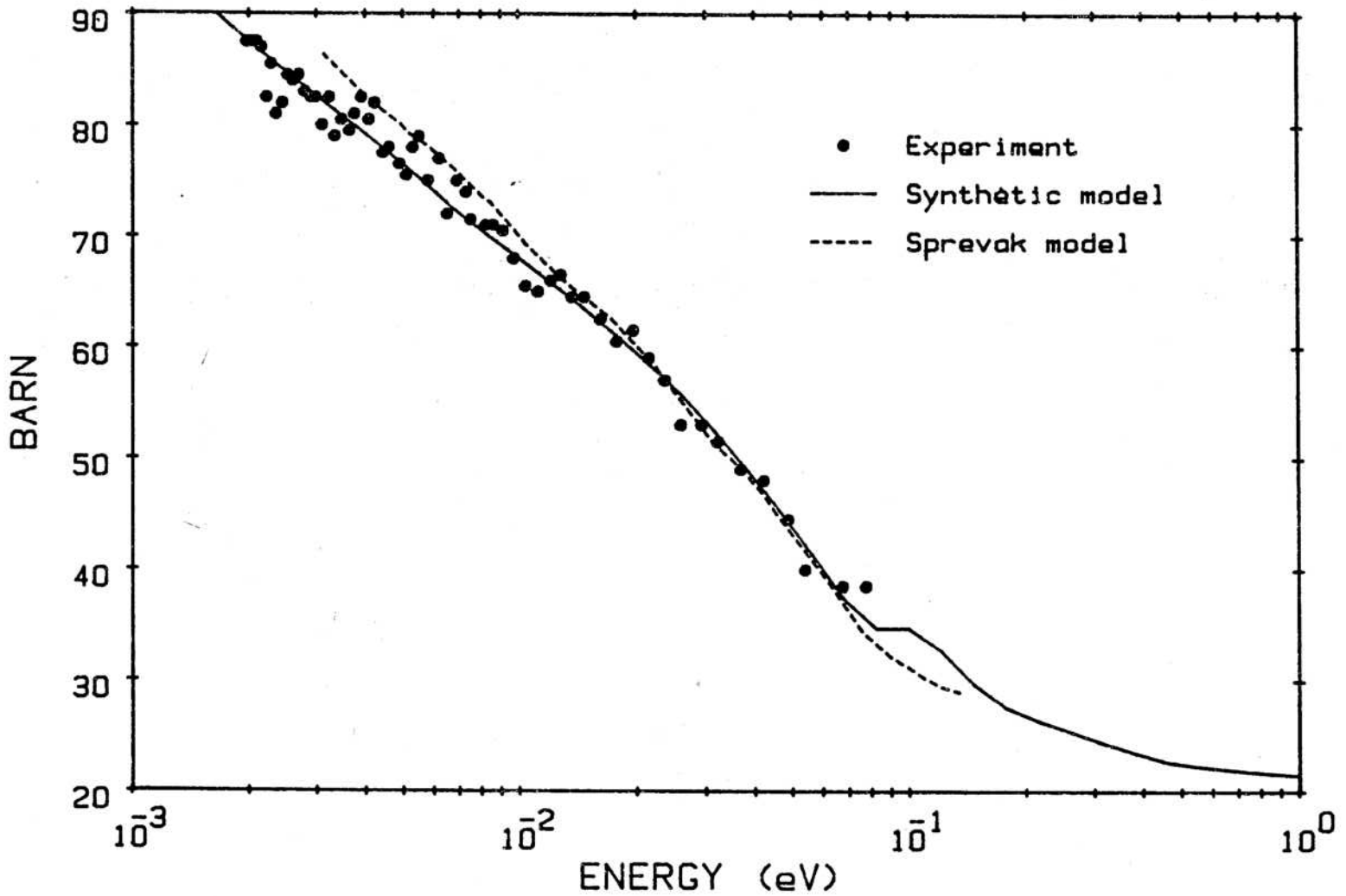
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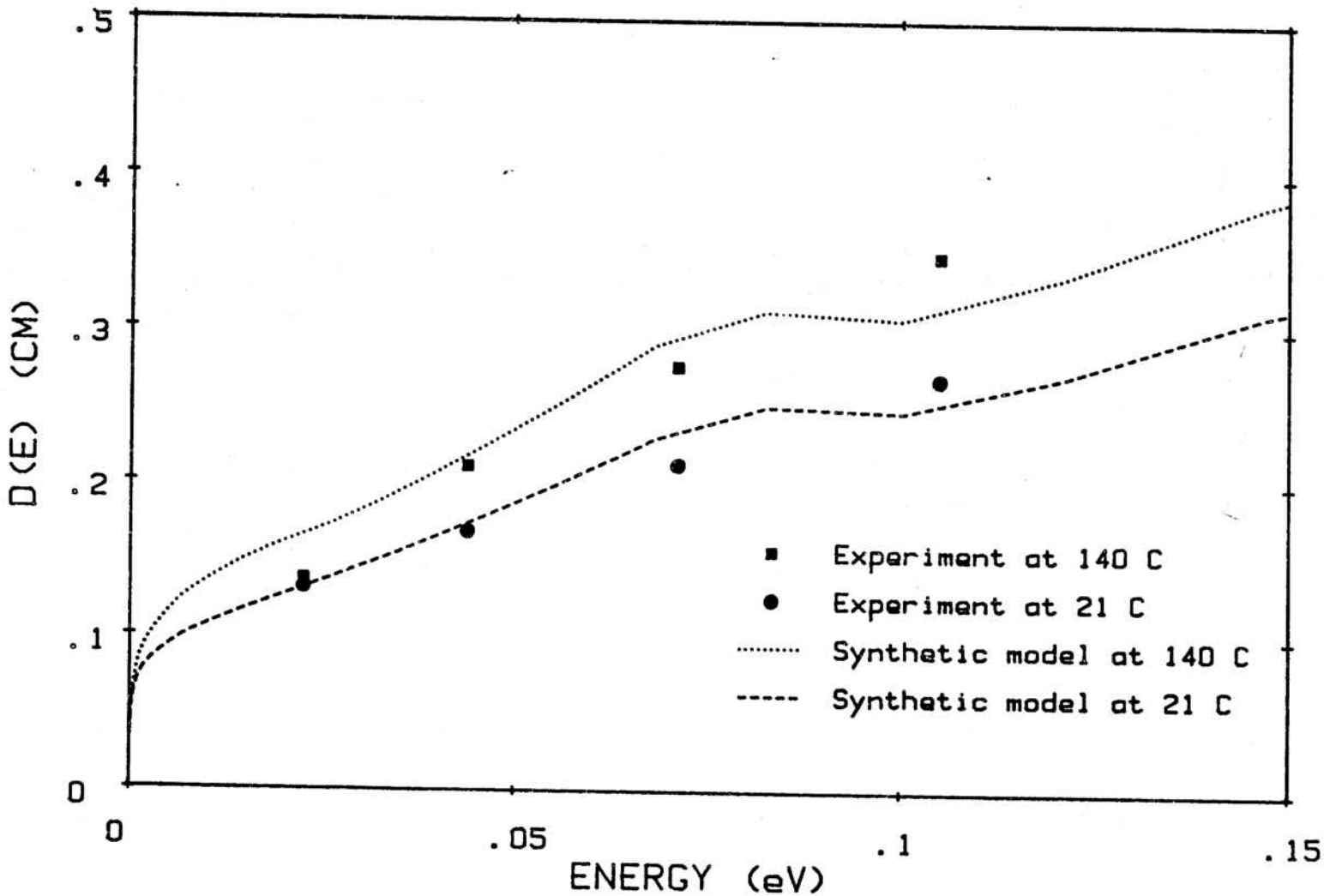
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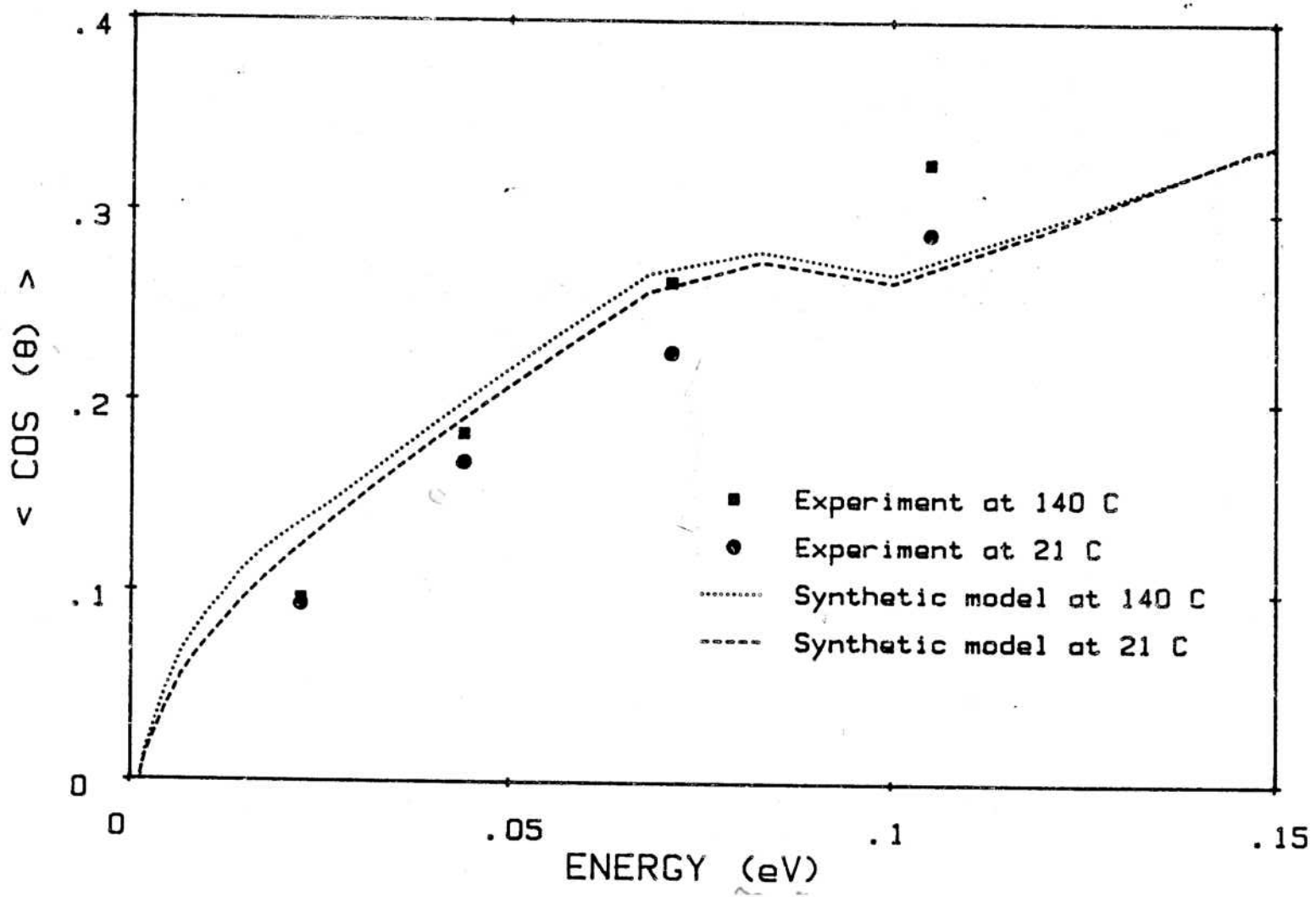
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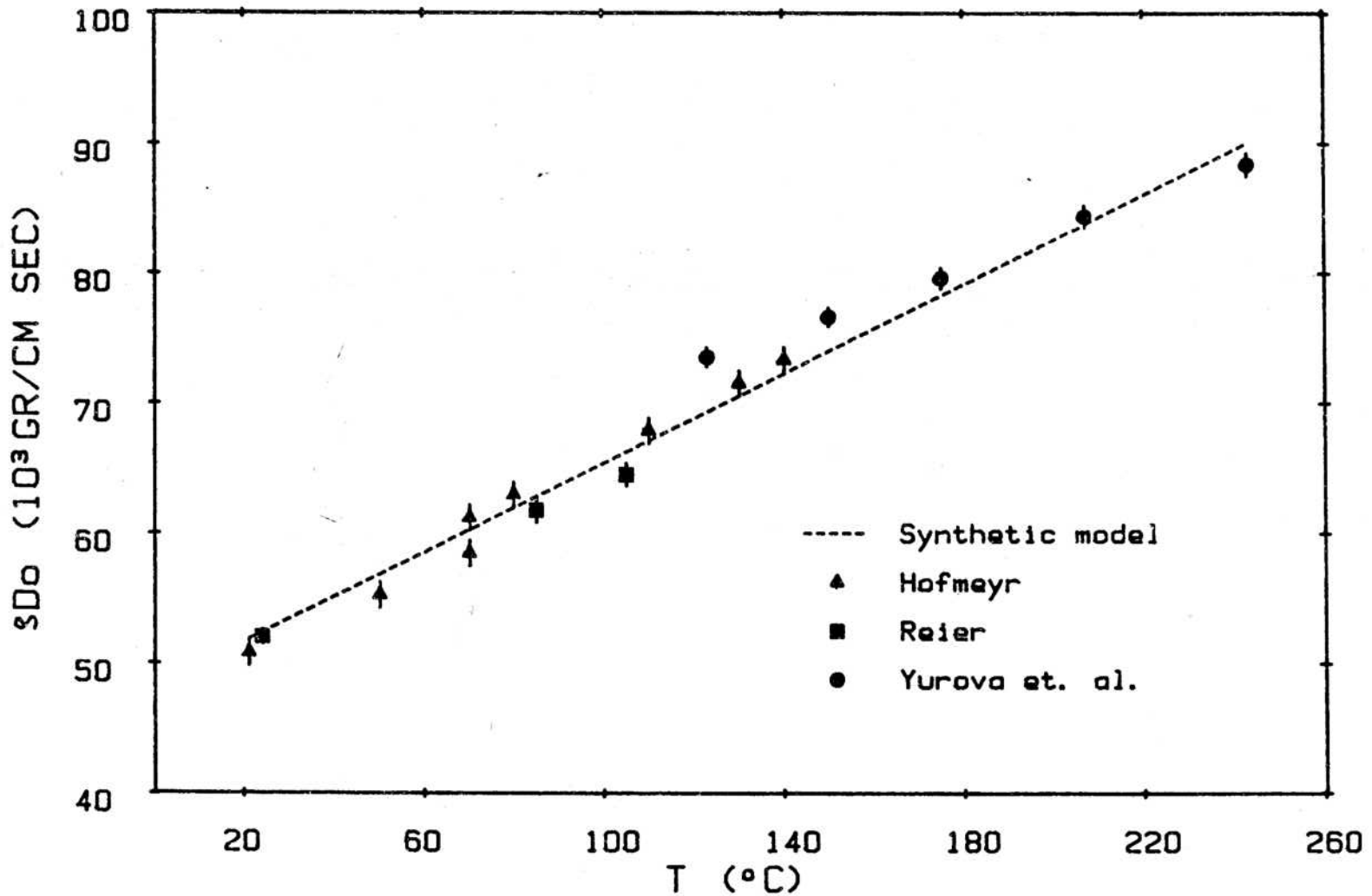
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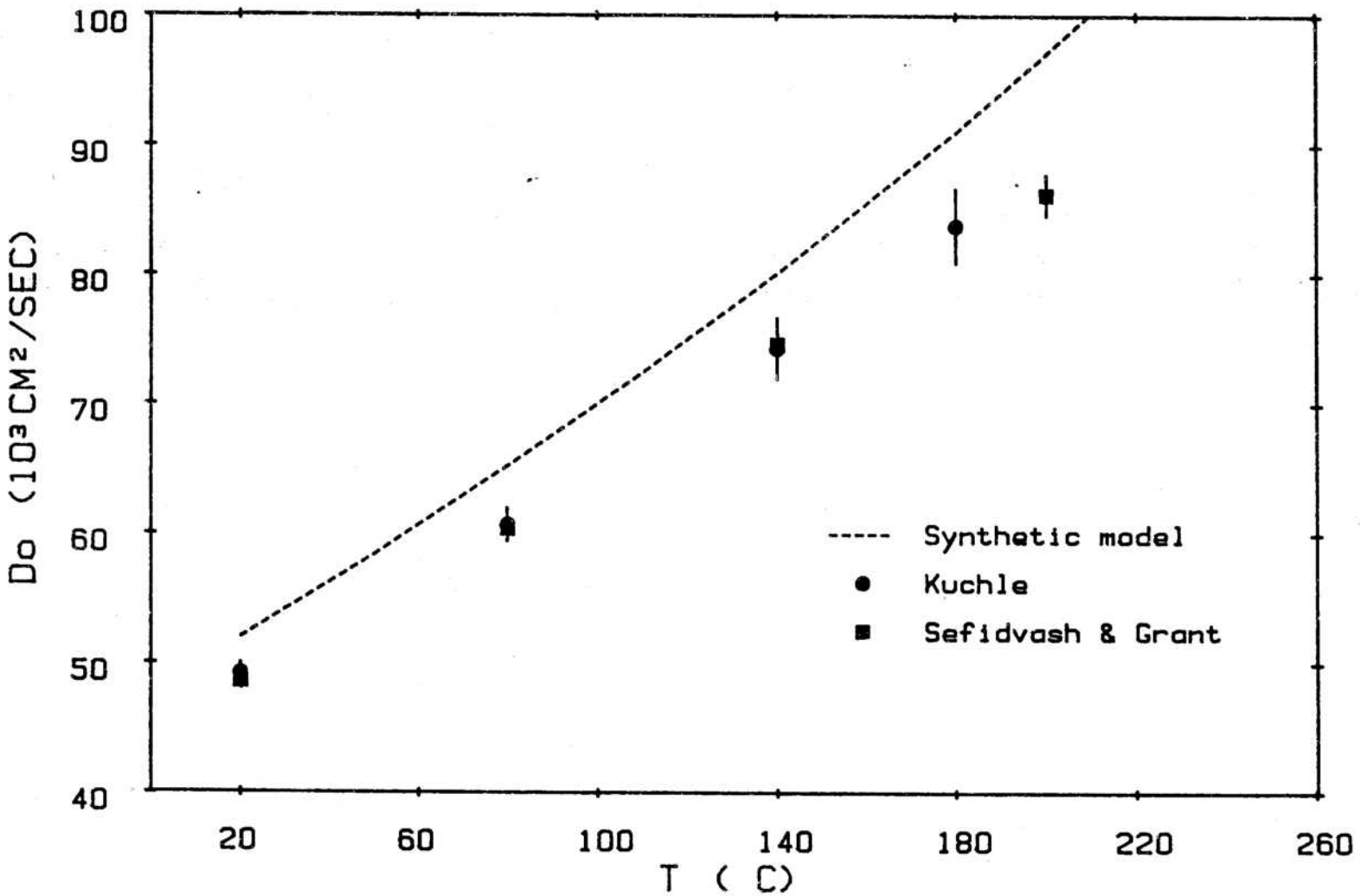
Yurova et. al.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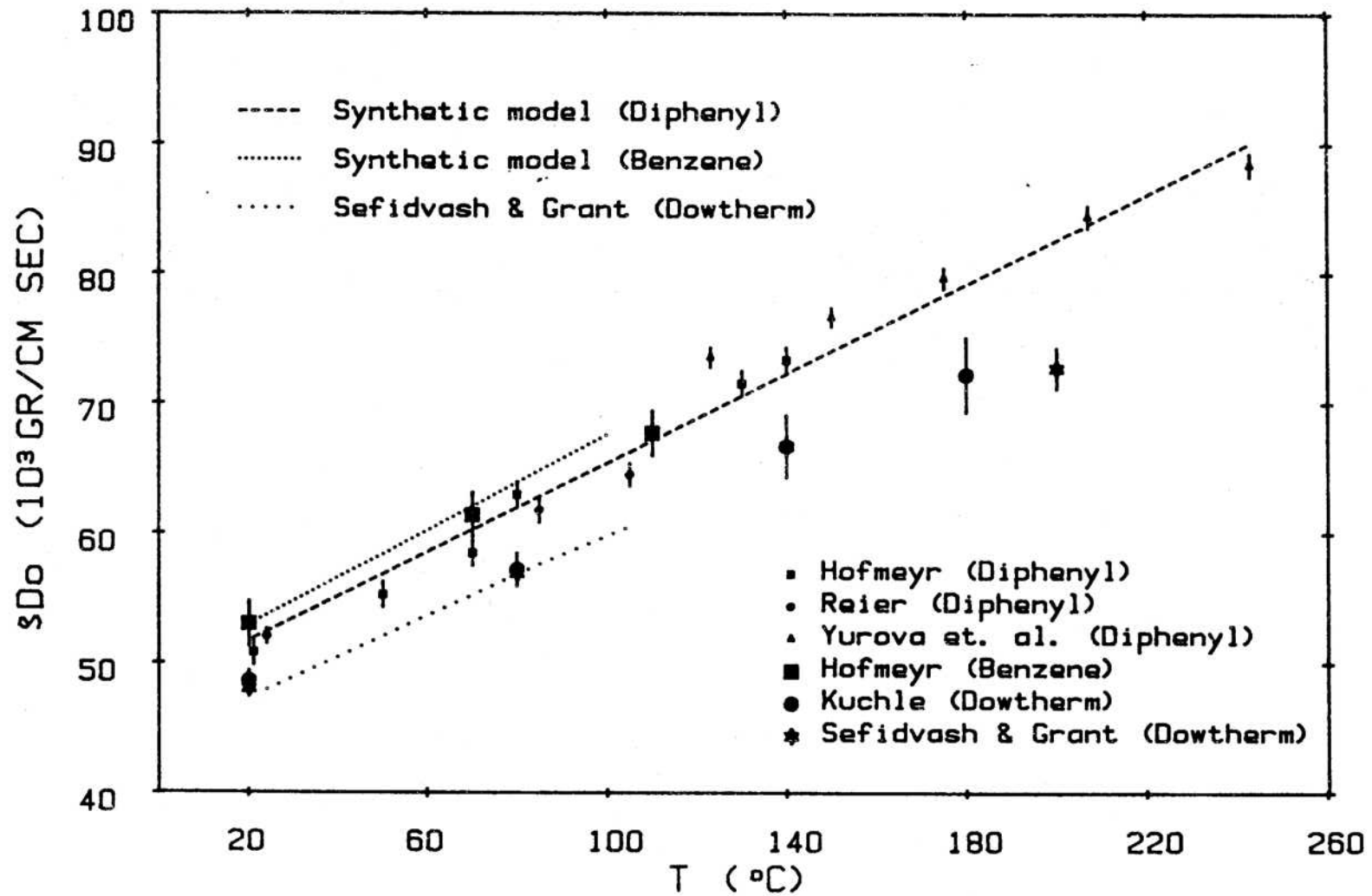
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Sefidvash & Grant (Dowtherm)
Hofmeyr (Diphenyl)
Reier (Diphenyl)
Yurova et. al. (Diphenyl)
Hofmeyr (Benzene)
Kuchle (Dowtherm)
Sefidvash & Grant (Dowtherm)