ENERGY TRANSPORT BY GAMMA-RAY EXCITATION IN FLUORESCENT LIQUID SOLUTIONS

By

A. KAWSKI and T. WAŚNIEWSKI

Luminescence Research Group, Institute of Physics, University of Gdańsk (Poland)
Institute of Physics, Technical High School, Białystok (Poland)

Experimental results on the radioluminescence efficiency η_s of a number of scintillators (solutes) in two different solvents under gamma-ray excitation are presented. These results are compared with the Kallmann—Furst theory and the three Kallmann parameters P, Q, R are determined. Linear dependence of η_s on the ratio of P/R is found for different solutes in a given solvent. The physical significance of the Kallmann parameters is studied.

Introduction

A very large number of organic compounds have been tested hitherto as solutes for scintillator solutions [1, 2]. FURST and KALLMANN [3] have measured the light intensites of the solutions as a function of the solute concentrations. The light emission of the solutions in the case of gamma-ray excitation was referred to the emission of an anthracene crystal of the same mass and in the same geometrical setting. Such relative fluorescence intensity values at the optimum concentration were described by KALLMANN and FURST as the relative physical efficiencies. Since the gamma-ray results for all the investigated solutions are referred to the same mass, which means approximately the same amount of absorbed gamma-ray energy, the number of electrons per gramme (of the solvent excited in unit time) is essentially the same for all the organic solutions. The experimental results of the relative yield are in good agreement with the theory given by KALLMANN and FURST [3, 4]. In the theoretical expression three parameters appear. These can be determined from the experimental data (see below).

In this paper we present further experimental data about the physical efficiences for some compounds in two different solvents, and in particular, the physical significance of the Kallmann parameters is studied.

Experimental

The light intensities of all investigated solutions were measured as a function of the solute concentrations, using an EMI 9514S photomultiplier. With this photomultiplier, the spectrum from 350 nm to 550 nm can be adequately covered. The relative radioluminescence efficiency was determined with an arrangement USB—2

A. KAWSKI AND T. WAŚNIEWSKI

described previously [5, 6]. The solutions were bombarded with gamma-radiation (^{60}Co). The gamma-ray results for all the solutions are referred to the same mass and to the emission yield of an anthracene crystal, similarly as in the investigations of FURST and KALLMANN [3].

Results and discussion

The results for 2-phenylindole in toluene and xylene are reproduced in Fig. 1, where the points give the experimental values and the full lines are curves calculated to fit the experimental points closely by means of the three Kallmann parameters occurring in Kallmann and Furst's theory. The theoretical expression found by
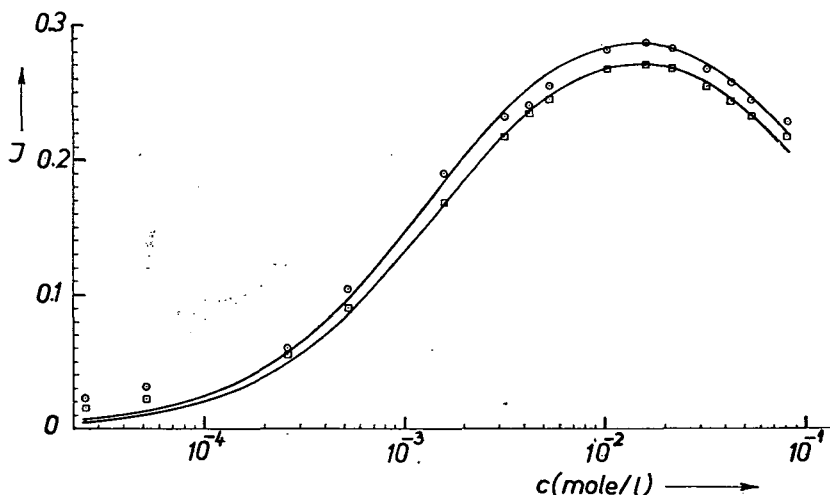

Fig. 1. Variation of emitted light intensity for various concentrations of 2-phenylindole in toluene (circles) and xylene (squares) for gamma-ray excitation. Full curves, theoretical curves calculated by means of equation (1)

KALLMANN and FURST [4, 7] for the variation of intensity I with concentration c is

$$I = \frac{P \cdot c}{(Q+c)(R+c)},\qquad (1)$$

where P, Q and R are the Kallmann parameters.

The Kallmann—Furst theory assumes that there is an energy transfer from the excited solvent molecules to the solute, where the energy is trapped. The parameter Q is associated with the energy transfer. It is shown in [8] that $Q\eta_{sv} = 1/\alpha \cdot \tau_{nt}$, where α is a measure of the cross section for transfer from the directly excited solvent to the solute, τ_{nt} the lifetime for nontransferring processes in the solvent alone and

η_{sv} is the solvent efficiency. A high value of Q indicates poor transfer. The parameters P and R are

$$P = \frac{n_e \delta}{\beta \tau_e} \tag{2}$$

and

$$R = \frac{1}{\beta}\left(\frac{1}{\tau_e} + \frac{1}{\tau_i}\right), \tag{3}$$

respectively, where n_e denotes the number of electrons of the solvent excited per unit time, capable of transferring their energy to the solute, δ is a constant which transforms the number of emited quanta into meter units, β a parameter (probability for unit time) connected with the process of self-quenching at higher concentrations, $\frac{1}{\tau_e}$ the probability (for unit time) of light emission of the solute, and $\frac{1}{\tau_i}$ the probability (for unit time) of internal quenching in the solute.

From (2) and (3) we get

$$\frac{P}{R} = \frac{n_e \delta}{1 + \dfrac{\tau_e}{\tau_i}}. \tag{4}$$

According to (4), the value P/R depends only on the number of excited solvent electrons n_e and the ratio of $\dfrac{\tau_e}{\tau_i}$.

Table I

Solution parameters. Solvent: toluene

No	Solute	η_s	c_{opt} (mole/l)	P	Q	R	P/R	$\frac{\tau_e}{\tau_i}$
2	Acenaphthene	0.044	0.039	0.0465	0.00143	0.980	0.0474	15.6
3	Eugenole	0.054	0.00653	0.0053	0.000431	0.0855	0.0620	11.68
4	Methyleugenole	0.052	0.00438	0.00567	0.000275	0.0983	0.0577	12.65
5	Isoeugenole	0.061	0.0164	0.0183	0.000626	0.2720	0.0671	10.75
6	Methylisoeugenole	0.075	0.0356	0.05245	0.00202	0.626	0.0838	8.37
7	N-phenyl-2-naphtylamine	0.093	0.0137	0.0185	0.00100	0.172	0.1076	6.32
8	2-phenylindole	0.288	0.0155	0.0510	0.00130	0.148	0.3446	1.28

Table I and II give the Kallmann parameters, the values of P/R and the physical efficiencies η_s for different solutes in toluene and xylene. We observed that the values of η_s corresponding to the optimum concentrations, of the different solutes are proportional to P/R for a given solvent (Figs. 2 and 3). It follows from Figs. 2 and 3 that
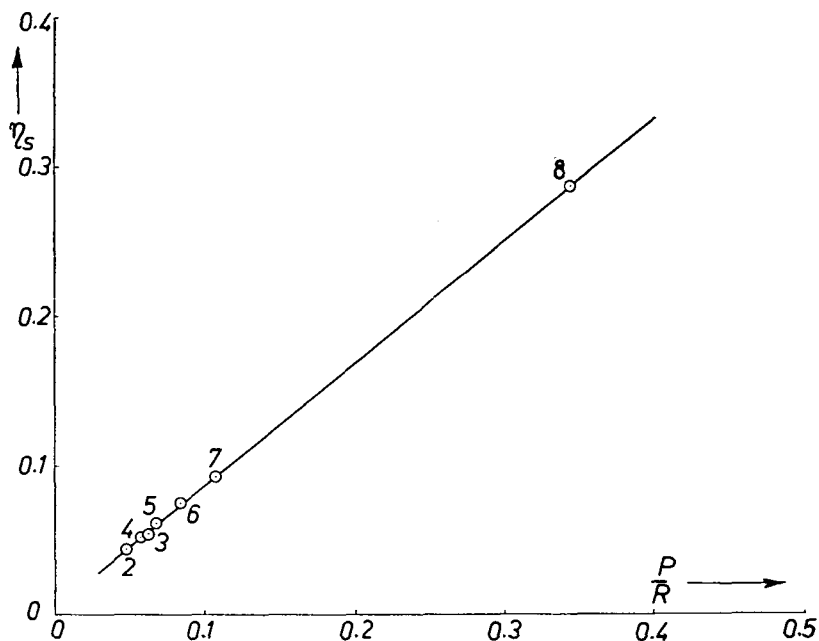
$$\eta_s = \alpha \frac{P}{R}, \tag{5}$$

Fig. 2. Relation between η_s and P/R for different solutes
(for the solutes see Table I) in toluene

where $a=1.27$ for toluene and 1.23 for xylene. From the definition of quantum yield and equations (5) and (4), we have $\alpha \approx \dfrac{1}{n_e \delta}$. Therefore, for a given mass of solvent the number of effective excited electrons available for energy transfer is constant ($a=$const.) and information concerning the dependence of $\dfrac{\tau_e}{\tau_i}$ on the solvent can be obtained by comparing the P/R values of a given solute in different solvents. For toluene and xylene, $n_e\delta=0.787$ and $n_e\delta=0.813$, respectively.

Table II.

Solution parameters. Solvent: xylene

| No	Solute	η_s	c_{opt} (mole/1)	P	Q	R	P/R	$\dfrac{\tau_e}{\tau_i}$
1	2,6-Dimethylnaphtalene	0.044	0.013	0.0081	0.00095	0.158	0.0513	14.85
2	Acenaphthene	0.048	0.039	0.0530	0.00156	1.020	0.0520	14.65
3	Eugenole	0.052	0.00653	0.0051	0.000584	0.0835	0.0611	12.30
4	Methyleugenole	0.054	0.00438	0.00556	0.000265	0.0928	0.060	12.50
5	Isoeugenole	0.060	0.0164	0.0189	0.000593	0.287	0.0658	11.33
6	Methylisoeugenole	0.069	0.0356	0.04438	0.00225	0.569	0.0780	9.42
7	N-phenyl-2-naphthylamine	0.097	0.0137	0.0180	0.00100	0.1585	0.1136	6.15
8	2-phenylindole	0.274	0.0155	0.0460	0.00148	0.1380	0.3333	1.44
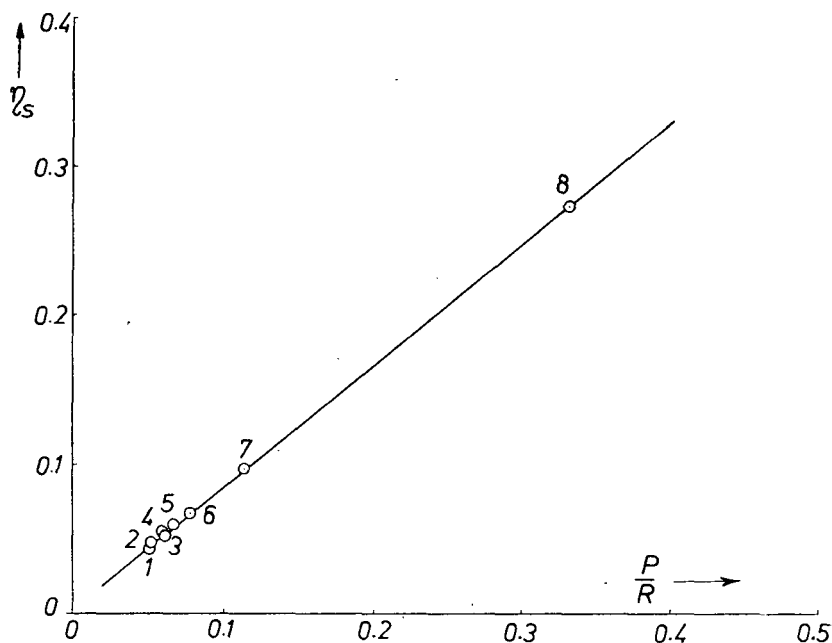
Fig. 3. Relation between η_s and P/R for different solutes
(for the solutes see Table II) in xylene

In Table I and II, also the values of $\dfrac{\tau_e}{\tau_i}$ for different solutes in toluene and
xylene are given.

If the decay time τ is known $\left(\dfrac{1}{\tau} = \dfrac{1}{\tau_e} + \dfrac{1}{\tau_i}\right)$, the time constants τ_e and τ_i can be individually evaluated from the following equations

$$\tau_e = \tau\,\frac{n_e\delta}{P/R} \tag{6}$$

and

$$\tau_i = \tau\,\frac{n_e\delta}{n_e\delta - P/R}. \tag{7}$$

For example, the measured value of the decay time τ for acenaphthene in xylene is $\tau = 10.6$ nsec, and from this $\tau_e = 166$ nsec and $\tau_i = 11.3$ nsec. It can be seen that if the solution is not of the highest efficiency (for acenaphthene in xylene $\eta_s = 0.048$), the lifetime τ is only approximately given by τ_i. However, for 2-phenylindole, the time constants τ_e and τ_i are comparable (see Tables I and II).

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ПЕРЕНОС ЭНЕРГИИ В ФЛУОРЕСЦИРУЮЩИХ ЖИДКИХ РАСТВОРАХ, ВОЗБУЖДЁННЫХ ГАММА-ЛУЧАМИ

А. Кавски и Т. Васьневски

Приведены экспериментальные результаты эффективности радиолюминесценции для ряда сцинтилляторов (растворенных молекул) в двух различных растворителях при возбуждении гамма-лучами. Эти результаты сравнили с теорией Калмана — Фурста и определили три параметра по Калману P, Q и R. Авторы нашли линейную зависимость эффективности от отношения P/R для различных сцинтилляторов в том же самом растворителе. Проведено обсуспсдение физического смысла параметров по Калману.