INVESTIGATIONS ON THE MODERATING PARAMETERS OF "DOWTHERM A" AT DIFFERENT TEMPERATURES

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Moderating parameters of a medium consisting of 26,81% diphenyl and 73,19% diphenyl oxide have been tested at different temperatures. The temperature dependence of the diffusion length has been found to obey the empirical relation $L_D = 4{,}002 \exp(0{,}0014\ T)$. The slowing down length has been found to be $L_S = 11{,}24 \pm 0{,}06$ cm at 36° C. At higher temperatures the slowing-down length increased considerably, so that the testing tank could not be considered any longer as being of infinite size and therefore only the temperature dependence of the most probable slowing-down length for this tank of finite dimensions could be specifically given.

1. Introduction

A few years ago it was suggested to investigate whether certain organic compounds prove useful as moderating materials and coolants for reactors. As generally known [1], research aimed at developing nuclear reactors operating with organic moderators and coolants has been started on a large scale.

One group of cyclic organic compounds (diphenyl, isopropyl-diphenyl, terphenyl, etc.) shows especially favourable characteristics. Thus diphenyl e.g. proves to be very stable against decomposing effects of high-energy radiation and high temperature ; it has a high boiling point (255,6° C) and its vapour pressure retains its low value even at temperatures above the critical temperature of water (diphenyl vapour pressure amounts to only 1,4 atm at 418° C). It appears to be remarkable that fuel elements contacting diphenyl show almost no sign of corrosion at high temperatures, a favourable difference from fuel elements contacting water. As diphenyl does not contain oxygen, it does not become radioactive in the active zone. This circumstance contributes largely to simplify protection. Physical and chemical properties of diphenyl — defining the character of the diphenyl boundary layer forming on metallic surfaces — are very advantageous. Another favourable fact may be that a negative temperature coefficient has been found for the multiplication constant of a system containing diphenyl as a moderator. It may be expected that parameters of the diphenyl moderator will be superior to those of natural water, as the unit volume of diphenyl contains less hydrogen atoms than the unit volume of natural water.

Although diphenyl belongs to the group of organic compounds sufficiently resisting decomposing effects of both radiation and temperature it must be expected, however, that at high temperatures radiation effects will initiate chemical changes (polymerization, hydrogen formation, etc.) in diphenyl. These changes have been studied by several scientists [2]. The heat transfer coefficient of diphenyl seems to be not too favourable if compared with the same parameter of natural water, however, compared with the heat transfer coefficient of carbon dioxide used in gas-cooled reacher systems, diphenyl will prove to be definitely superior.

Pure diphenyl has a rather high melting point (70° C), while an eutectic mixture of diphenyl and diphenyl oxide shows a substantially lower melting point (12,3° C). This eutectic mixture of diphenyl and diphenyl oxide, known as Dowtherm A, is already familiar in the chemical industries as a heat transfer agent. Many data may be found in the literature on the general physical and chemical properties of Dowtherm A (also known as diphyl), but almost no data have been published on diphyl as a moderating material. The only exception is a recent study by BLACKMAN, LAKEY and PRESTON [3]. These authors have determined at low temperatures, probably at normal ambient temperature, the slowing-down length for the fission spectrum. Similar data are probably available at different research laboratories, where, as it is well known, great efforts are being made to develop reactors using organic moderators and coolants (especially diphyl).

Our measurements served the purpose of analysing moderator parameters and the temperature dependence of the latter to be used in design studies of diphyl-moderated nuclear reactors.

2. General physical and chemical properties of the Dowtherm A agent used in the experiments

Chemical analysis resulted in the following particulars concerning the composition of Dowtherm A as used in experiments by the authors :

C — 87,15%,

H — 5,97%,

O — 6,88%.

Conventional analytical methods were insufficient to prove the presence of any nitrogen, sulphur or haloid elements in the mixture (no ash remained after burning the test sample). The following Table has been prepared based on elemental analysis :

Denomination	molar %	weight %	weight % acc. to the literature [4]
diphenyl	28,33	26,81	26,5
diphenyl oxide ...	71,67	73,19	73,5

It can be seen that the composition of the mixture investigated does not differ significantly from that given in the sources quoted in the literature (corresponding to exactly the eutectic composition). Melting and boiling points were measured to have the following values :

Mixture	°C measured	°C literature [4]
melting point	13,0	12,3
boiling point (760 mm Hg)	256,0	258

Fig. 1 shows the temperature dependence of specific weight (density), viscosity, vapour pressure, specific heat and heat conductivity of diphyl as investigated by the authors. Measured values show a good agreement with the data available from the literature [5].

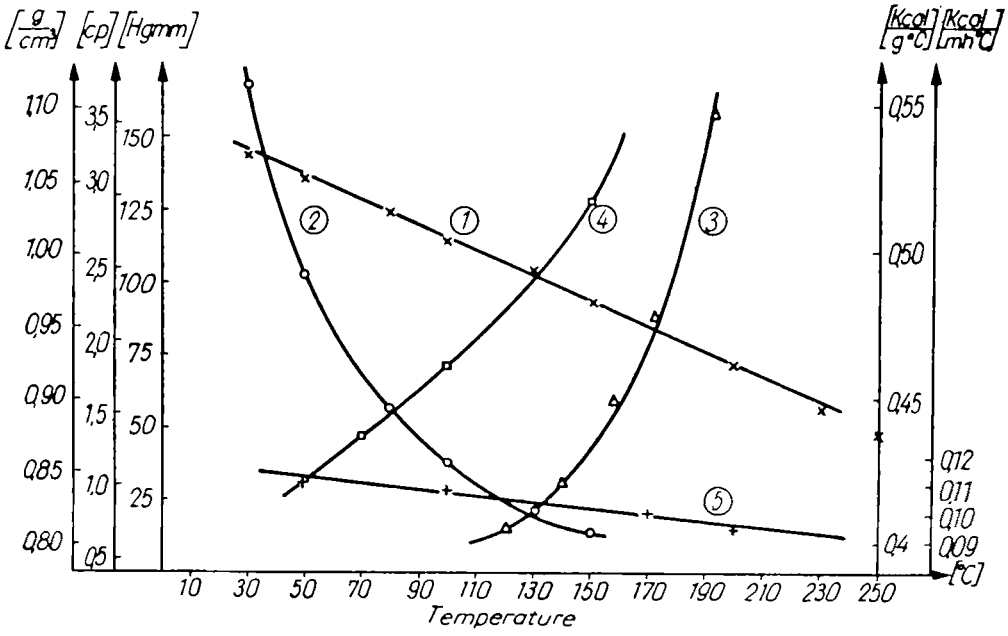

Fig. 1. Temperature dependence of the density ①, viscosity ②, vapour pressure ③, specific heat ④ and heat conductivity ⑤ of the diphyl (Dowtherm A) tested

3. Methods of measurement and description of the measuring equipment

Po-Be neutron sources have been used for our measurements. When starting the measurements, source activity amounted to $3{,}10^7$ n/sec. Later measurement data had to be corrected according to reduced source activity.

A silver foil with a diameter of 25 mm and a thickness of 98,16 mg/sec^2 has been used as neutron detector. The silver foil has been chosen with regard to the circumstance that investigations were to be continued up to temperatures exceeding the melting point of indium (i.e. 156,4° C). The silver foil has been used as a thermal neutron detector, i.e. the activity of the cadmium-coated silver foil has been always subtracted from that of the foil without cadmium cover. Thickness of the cadmium sheath fitted snugly to the foil amounted to 500/mg/sec^2.

As already known, Ag^{108} produced from Ag^{107} through neutron capture has a half-period of 2,3 minutes, while decay of the Ag^{110} as produced from Ag^{109} is characterized by a half-period of 24,2 seconds or 270 days resp. (for its isomer). Activation cross sections for thermal neutrons may be summarized in the following Table :

$$\sigma_a(Ag^{107} \rightarrow Ag^{108}) = 44 \pm 9b, \quad \sigma_a(Ag^{109} \rightarrow Ag^{110}) = 110 \pm 20b$$

and

$$\sigma_a^*(Ag^{109} \rightarrow Ag^{110*}) = 2{,}8 \pm 0{,}5b.$$

It can be seen that radioactive decay of isomeric Ag^{110*} having a much longer half-period plays an unsignificant role in the values of the measured activity. Relatively short half-periods make quick and reproducible processing of the foil absolutely necessary. As activation time 12 minutes was chosen while exactly 36 seconds after finishing the activation an 8-minute evaluating measurement was begun.

Foil activity has been measured in a plexiglass-lined lead column using end-window counter tubes of the Soviet SI-2V type. The foil was placed at a distance of 30 mm from the 40 mm dia. mica window of the counter tube, the window thickness being 2,5 mg/cm^2, Soviet PS-10 000 type pulse counters have been used for recording the pulses.

The squared migration length has been determined according to the usual method using the position function of the activity of the silver foil utilised for thermal neutron detection. Let us denote by $A_1(r)$ the activity of the silver foil (without cadmium cover) placed at a distance r from the point-like neutron source, subsequent to an activation period of given duration. Denote by $A_2(r)$ the activity of a silver foil placed into a cadmium sheath and irradiated under identical conditions. If the quantity $A(r) = A_1(r) - A_2(r)$ is known, relation

$$< r_{Cd}^2 > = \frac{\int_0^\infty r^4 A(r)\, dr}{\int_0^\infty r^2 A(r)\, dr} \tag{1}$$

permits to compute the squared migration length for neutrons slowed down below the energy level of cadmium cut-off, because, in a moderating medium of infinite dimensions, for the case with spherical symmetry we have $< r_{Cd}^2 > = = 6 L_M^2$. Integrals figuring in the expression $< r_{Cd}^2 >$ have been determined by planimetration of the experimental curves. At large distances from the neutron source, where measurement data are no longer available, activity values extrapolated from the last segment of the experimentally-defined curve and, as already known, satisfying the asymptotic relation $A(r) \sim r^{-2} \exp(-r/\lambda)$ have been used.

In a system which may be regarded as having infinite dimensions, migration, slowing-down and diffusion lengths are interconnected by the relation $L_M^2 = L_S^2 + L_D^2$. By determining — through use of a convenient method — the actual value of the diffusion length L_D the squared slowing-down length corresponding to the energy level of cadmium cut-off may be simply computed.

For measuring the diffusion length, FERMI's method has been chosen, because the relaxation length λ of the neutrons emitted by the Po-Be neutron source — as measured in diphyl — exceeds the diffusion length to be expected. It is known [6] that for this case the diffusion length cannot be directly determined from the distribution of thermal neutrons. Placing, however, a plane consisting of a cadmium plate of given thickness (this plane being considered as of infinite dimensions at a predetermined distance from the neutron source, the neutron density measured behind this plane is given, as well known, by the relation $n_2(r) = n_1(r) - \exp(-r/L_D)$, where $n_1(r)$ denotes the neutron density for the case without the cadmium sheath. Thus the diffusion length can be simply computed from the difference $n_1(r) - n_2(r)$, in so far as the medium can be regarded as having infinite dimensions.

A schematic diagram of the measuring apparatus appears in Fig. 2. This is essentially a tinplated cylindrical steel tank having a volume of 800 litres, a diameter of 940 mm and a height of 1000 mm (6). This tank is surrounded by flanges along its upper and lower rims, while three tubes of 90 mm dia. placed at 120° to each other pass through the flanges. The lower extensions of these tubes form the pedestal of the tank, this being shut-off through welded-in disks. This tube pedestal contains the mixer blades (7) fixed to the hollow shaft and driven by the mixer motors (2). Counter-current circulation provides a full exchange of the total fluid volume within five minutes. Circulation path follows from the bottom part of the tank through a tube stub (9) to the tube

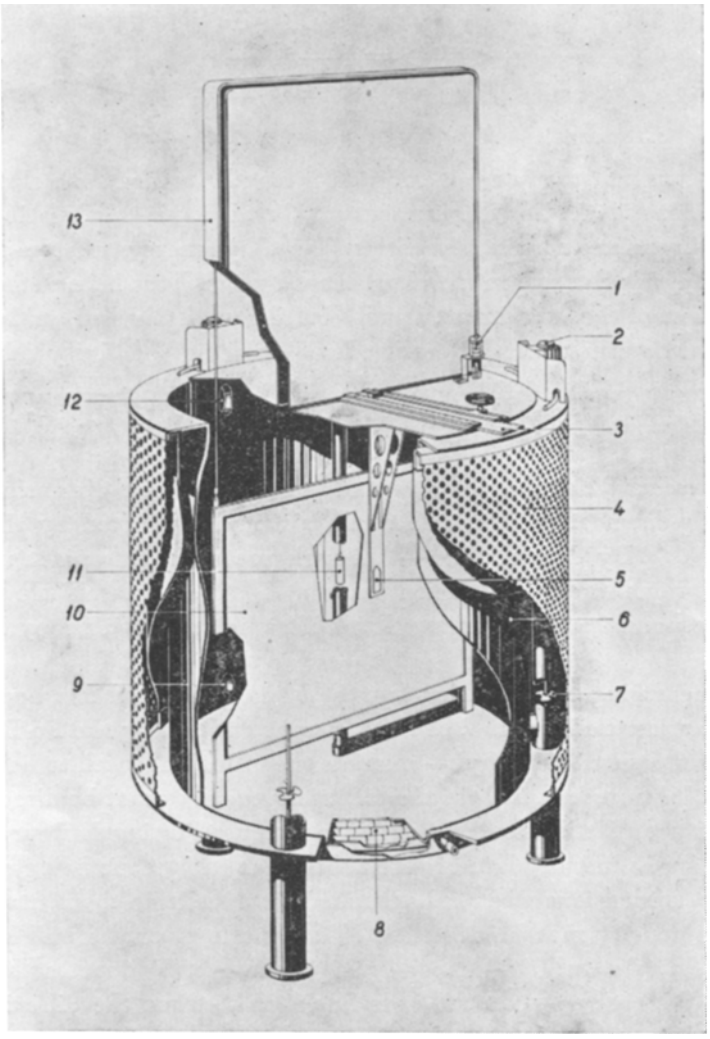

Fig. 2. Schematic diagram of testing equipment. (See pages 177, 178 and 179)

pedestal, while the fluid is re-introduced to the tank through the upper channel (12). Thermosyphon type circulation is operating to produce an opposite effect. As a net result efficient mixing and maximum temperature stability is being obtained.

The electrical heater (8) is a three-phase wye-delta connected unit, ensuring a maximum power output of some 25 kW. Heating elements made from flat "Kanthal" wire and insulated by ceramic beads have been mounted into three sector-shaped steel plate holders and fixed to the tank bottom.

The heater is surrounded by a second, heat-insulating case mounted with a gap of some 5 cm, just in the way as the outer case (4) surrounds the tank walls.

Within the tank the cadmium separator plate (10) can be moved along channel-shaped guide ways and can be lifted — being balanced by a counterpoise — to the upper chamber of the cooling tower (13). Behind the cadmium plate an aluminium-alloy frame (11) holding the neutron source has been mounted ; this can be moved to any arbitrary position, while its position data may be read off a scale.

The foil to be activated is to be inserted into an externally-accessible groove of the holder (5) made of thin, high-strength aluminium sheet. This holder (5) has been mounted on to a slide structure made of heavier-gauge aluminium-plate, its lateral surface being provided with rack teeth, so that it can be brought nearer to or removed from the neutron source by means of a gearwheel. Distance can be read off a measuring mechanism graduated in millimetres.* The constant temperature value of the medium to be introduced into the tank is being ensured by a contact thermometer (1) giving a precision of $\pm$ 0,2° C. Parts of the contact thermometer protruding from the tank, as well as mixer motors have been surrounded by a water-cooled jacket, which enables the apparatus to be operated at higher temperatures (around 200 to 250 centigrades).

The controlling contact thermometer has been placed upstream of one of the inflow openings, which brought forth a considerable increase in the sensitivity of temperature control, as the warmed-up fluid drawn from the heater surface made direct contact with the contact thermometer. For the sake of greater reliability a second contact thermometer has also been installed, the latter operating at a temperature exceeding by a few centigrades that of the first thermometer. In case of a failure of the first thermometer a safety switch disconnects the whole arrangement and simultaneously operates an alarm signal.

The tank is being hermetically closed by the sealed, bolted-down flange of column (13).

In the design of the equipment attention has been paid to enable all necessary control and regulation operations, insertion and removal of neutron source and measuring foil to be performed without disassembling the closed column.

In case of high-temperature experiments the fluid condensating along the large, cold internal surfaces of the cooling column will flow back to the tank.

* The foil is inserted and removed by means of a small servomotor placed into the cylindrical tube (not to be seen in the figure) mounted on the slot above the foil-holder which is opened and closed by a magnetic switch. The slat one-sidedly toothed carrying the foil and moving on a guided track is driven by this motor. The magnetic switch and the servomotor are operated by the time switch determining the activation time.

4. Measurement results

Determining the migration length characteristic for a quasi-infinite medium may seem to be a simple task when $< r_{Cd}^2 >$ is known. However — with size characteristics of the medium as investigated by the authors — the relation $< r_{Cd} > = 6L_M^2$ cannot be regarded as being satisfied for every case.

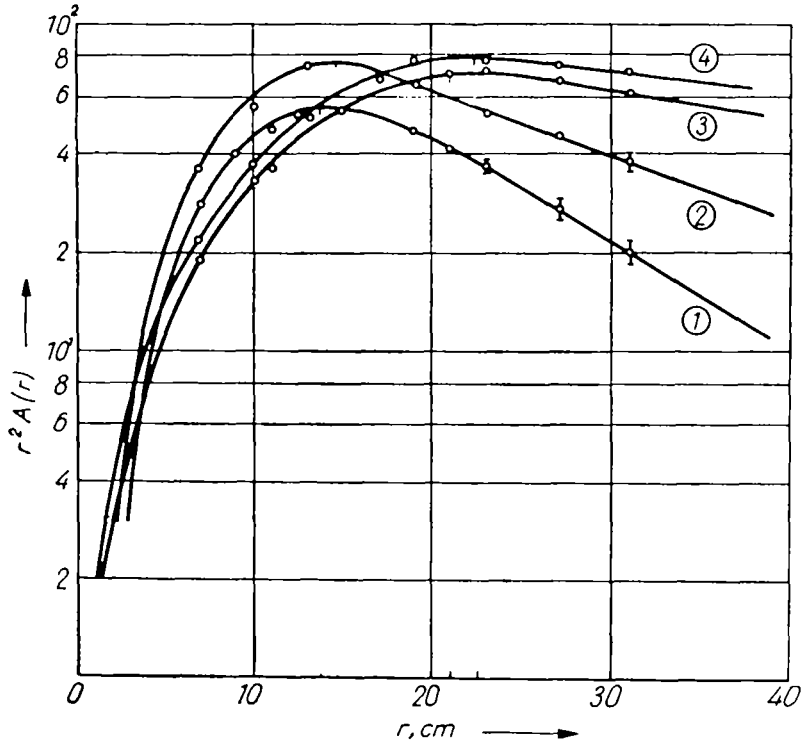

Fig. 3. $r^2 A(r)$ versus distance for different temperatures: ① for 36,0° C, ② for 92,0° C, ③ for 146,0° C and ④ for 192,0° C

Fig. 3. shows the variations of $r^2 A(r)$ for 36,0, 92,0, 146,0 and 192,0 centigrades. In case of diphyl the slowing-down length corresponding to neutrons emitted by the Po-Be neutron source already attains considerable values for ambient temperature, as proved by the maximum place of the $r^2 A(r)$ curve. Any increase in temperature shifts this maximum towards larger values of r. The curves show that computing the mean value has only sense for 36° C as for a higher temperature the medium cannot be regarded as being of infinite dimensions. Consequently, increments of the integrals figuring in the expression of the mean $< r_{Cd}^2 >$ obtained by extrapolation will make a substantial part of the integral value. For measurements performed at 36° C

extrapolation increments of the integral $\int_0^\infty r^2 A(r)\,dr$ amount to 8,2 per cent of the integral value, while attaining 65,6 per cent for the integral $\int_0^\infty r^4 A(r)\,dr$. Extrapolation increments will be anyway much lower for the medium of infinite dimensions than increments determined by measurements for the same space. This is due to the fact that because of leakage neutron density is reduced in the proximity of the boundary surface. Of course there will be the more leakage neutrons from a medium of given size the higher the actual value of slowing-

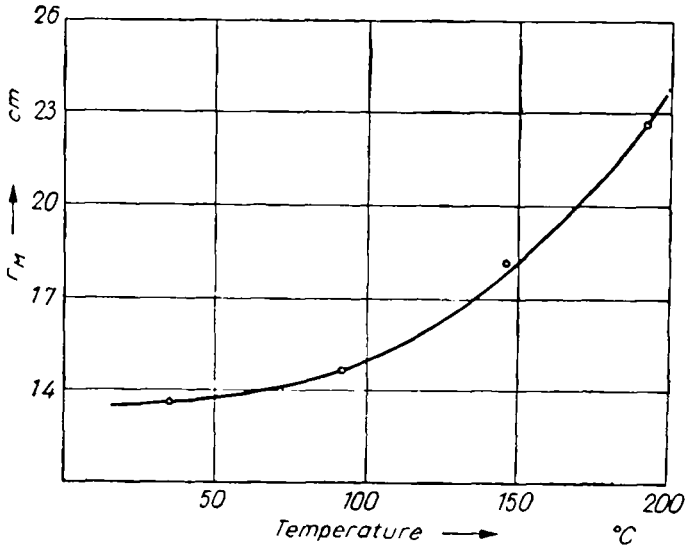

Fig. 4. Temperature dependence of the most probable slowing-down length

down length. Using the estimations of BLACKMAN, LAKEY and PRESTON [3] the error due to finite system size amounts to no more than 1,5% at 36° C. For a temperature of 36° C the following values are being obtained :*

$\lambda = 15{,}8 \pm 0{,}05$ cm, $< r_{Cd}^2 > = 864 \pm 12{,}04$ cm², $L_M^2 = 144 \pm 1{,}34$ cm².

These values exceed the values measured by BLACKMAN, LAKEY and PRESTON [3], i. e.

$\lambda = 7{,}4 \pm 0{,}3$ cm $< r_t^2 > = 396 \pm 19$ cm², $L_M^2 = 66 \pm 3{,}5$ cm²

because the mean energy of the neutrons emitted by the Po-Be neutron source also exceeds the mean energy of the fission neutron sources.

From the series of curves $r^2 A(r)$ measured at higher temperatures only the temperature dependence of the curve peaks is being shown by Fig. 4.

* The value of λ has been determined by the method of least squares, and its normal deviation through use of the inverse matrix method. The relatively low deviation value of L_M^2 may be explained by the fact that it has been determined from data obtained through six independent measurements.

Geometrical conditions have been much more favourable in so far as the measurements of the diffusion length are concerned. For relatively low values of the diffusion length our medium may be regarded as being of infinite dimensions. The table below shows the value of diffusion lengths as determined by the method of least squares for several temperature values, specifying

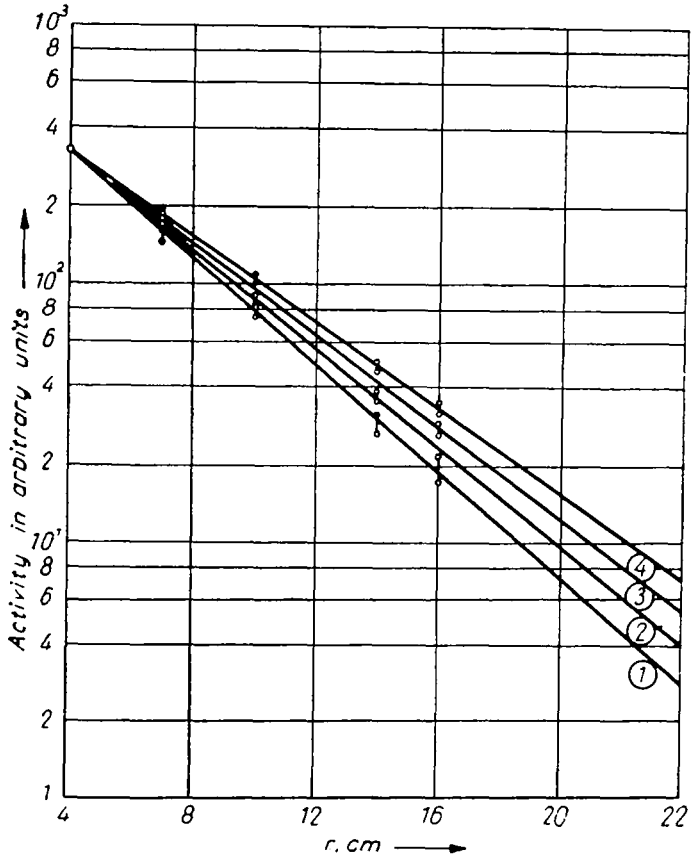

Fig. 5. Determination of the diffusion length on the basis of the position dependence of activity proportional to the neutron density as given by $n_1(r) - n_2(r) = \exp(-r/L_D)$. ① for 36,0° C, ② for 92,0° C, ③ for 146,0° C and ④ for 192,0 °C

T	36,0	92,0	14,60	192,0
L_D	4,214 ± 0,006	4,541 ± 0,002	4,934 ± 0,005	5,245 ± 0,004

simultaneously the deviation as computed by the inverse matrix method while Fig. 5 is a diagram of the position function of activity values proportional to the neutron density $n_1(r) - n_2(r) = \exp(-r/L_D)$.

It can be easily shown that for the temperature dependence of the diffusion length in Dowtherm A an empirical relation of the form

$$L_D = L_D^0 \exp(\vartheta T)$$

$$(L_D^0 = 4{,}002 \text{ cm}, \qquad \vartheta = 0{,}0014 \text{ grad}^{-1})$$

may be deduced. The primary reason for the temperature dependence of the diffusion length is the change in diphyl density. However, the temperature coefficient of the inverse density is somewhat less than the temperature coefficient of the diffusion length. PETRIE, STORM and ZWEIFEL [7] have carried through a series of computations to define the temperature dependence of the diffusion length in diphenyl.* According to their computations the temperature coefficient of the diffusion length in diphenyl is again higher than that for the corresponding inverse density. Fig. 6 shows the temperature dependence of the diffusion length for the Dowtherm A sample tested by the authors of this paper.

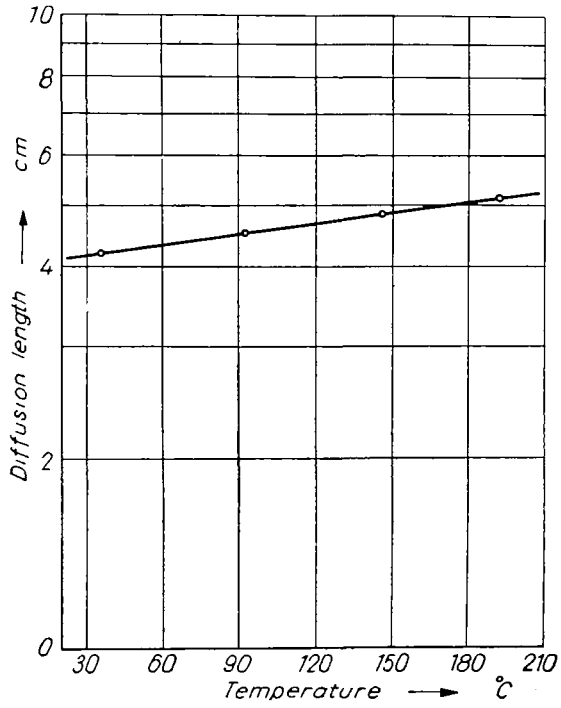

Fig. 6. Diffusion length in Dowtherm A versus temperature ($L_D = 4.002 \exp(0{,}0014\ T)$)

* Temperature dependence of the diffusion length in diphenyl may be described by a formula similar to that given above with the only difference that $L_D^0 = 4{,}143$ cm and $\vartheta = 0{,}0021$ grad^{-1}.

Subtracting the value of the diffusion length measured at 36° C from the migration length equally measured at 36° C gives the squared slowing-down length for the energy level of cadmium cut-off. It has been found that $L_s^2 = 126{,}3 \pm 1{,}34$ cm². This is again higher than the squared slowing-down length as defined by BLACKMAN, LAKEY and PRESTON.

It should be noted that perturbations due to finite thickness of the foil to be activated as well as of the cadmium sheath have been taken into account by introducing appropriate corrections.

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ИССЛЕДОВАНИЯ ЗАМЕДЛЯЮЩИХ СВОЙСТВ ОРГАНИЧЕСКОГО ВЕЩЕСТВА ДИФИЛЯ ПРИ РАЗНЫХ ТЕМПЕРАТУРАХ

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Резюме

Параметры замедления нейтронов в органическом веществе дифиля (состав: 26,81% дифенил и 73,19% окись дифениля) были определены при разных температурах. Для температурной зависимости диффузионной длины было найдено следующее эмпирическое соотношение

$$L_D(T) = 4{,}002\,[1 + 1{,}4\,10^{-3}\,T + 0{,}98\,10^{-6}\,T^2]\,.$$

Для длины замедления при температуре 36 °C найдено значение $L_S = 11{,}24 \pm 0{,}96$ cm для нейтронов Po-Be источника.