

1967

Permeation of organic liquid through fully swollen polyethylene employing a tracer technique

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PERMEATION OF ORGANIC LIQUID
THROUGH FULLY SWOLLEN POLYETHYLENE
EMPLOYING A TRACER TECHNIQUE

by
Frank August Pollak

A Thesis
Presented to the Graduate Faculty
of Lehigh University
in candidacy for the Degree of
Master of Science
in
Chemical Engineering

Lehigh University

1967

CERTIFICATE OF APPROVAL

This research report is accepted and approved in partial fulfillment of the requirements for the Degree of Master of Science in Chemical Engineering.

April 18 1967
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ACKNOWLEDGEMENT

The author wishes to express his gratitude for the privilege of being selected a National Defense Education Act Fellow and also to Dr. Robert W. Coughlin, Assistant Professor of Chemical Engineering, for suggesting the problem and for his helpful guidance during the work. This work was also materially assisted by use of the liquid scintillation spectrometer made available through the Office of Saline Water Grant No. 14-01-0001-753.

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ABSTRACT

The permeation of toluene through polyethylene film which was fully swollen by an organic liquid (toluene, chlorobenzene, mesitylene, cumene, ethylbenzene, cyclohexane, tetrahydronaphthalene, or decahydronaphthalene) was studied in the temperature range 25 to 40 °C. During all experiments, the 1.5 mil thick polyethylene film was in complete equilibrium with each swelling liquid used.

It was possible to study diffusion under these conditions by using a new and simplified technique. This involved employing a small quantity of tritiated toluene as the diffusing specie.

For the permeation process corresponding to each swelling liquid the data were fitted to an Arrhenius type plot to determine the energy of activation for permeation of toluene through the swollen polyethylene membrane.

This resulted in the following values:

Swelling Solvent	Activation Energy cal./g. mole	D_0 cm. ² /sec.
Toluene	15,772.35	4.89326×10^4
Chlorobenzene	16,218.53	9.12629×10^4
Mesitylene	14,253.56	3.40398×10^3
Cumene	16,675.40	1.29250×10^5
Ethylbenzene	11,414.84	3.65872×10^1
Cyclohexane	14,753.32	1.32904×10^4
Tetrahydronaphthalene	12,166.11	6.72825×10^1
Decahydronaphthalene	16,535.79	1.01184×10^5

INTRODUCTION

While many studies of diffusion of gases and liquids through polymeric materials have been reported in the literature, almost all of these studies employed a pressure differential across the polymer film itself. The diffusing gas or liquid was placed into a chamber on one side of the polymeric material and diffusion was allowed to occur through the polymer film to the low pressure side. This method always required a pressure difference across the membrane. The equipment for the pressure differential approach has become standard in diffusion work and is best described by Schumacher (25), Park (18), and Paul and DiBenedetto (21), each having their own modification. However, the present work does not incorporate any pressure differential across the film, thereby, limiting the need for extensive equipment. In this work the polymeric film is held in place between two chambers, each of which is filled with the organic liquid which causes the swelling. After equilibrium swelling is attained, a radioactive tracer is injected into one chamber and its rate of diffusion to the other chamber is measured by taking small samples at appropriate times and measuring their radioactivity concentration. Thereby an accurate measure of the rate of diffusion is possible. As a result, there is no chance that pressure gradients influence the data.

This concept of employing radioactive tracers has been employed before; but only on a very limited scope, and

then usually employing a thick layer method (32) for measuring the rate of diffusion. The tracer is the essential agent in the present work since without it, it would be impossible to measure permeation through the film, since this is the only way to determine accurately the amount of the diffusing specie which has diffused. Some interesting work using tracers has been done by Gromov (5) on the diffusion of the antioxidants, 2:6-di-t-butyl-4-methylphenol and phenothiazine in isotactic polypropene and polyformaldehyde and by deBrouckere (3) on Cl^{36} activated poly(chlorobutyl acrylate) dissolved in Me_2CO through cellophane films. Work on the permeation of polyethylene using trace quantities of tritiated water vapor has also been done (29). However, in all these cases using tracer elements no mention is made that the film was allowed to reach equilibrium before measurements were begun.

In the present work, the polyethylene film is allowed to attain complete equilibrium with the swelling liquids even before the tracer element is added to one chamber. In this swollen condition, which indicates the existence of a liquid solution of the swelling liquid and the diffusing specie and the polymer film, the increased thickness could lead to lower permeability values due to possible effect of thickness on concentration gradients (9) within the film.

The experimental work on the diffusion of organic substances through polymer films had until ten years ago

been very limited, the diffusion work being mainly concerned with the simple gases. Since that time extensive work has been completed. Barrer (1) in his book presents an extensive review of diffusion work primarily with the simple gases, while Paul and DiBenedetto (21) have recently performed new studies with the simple gases. Prager and Long (23), Michaels et. al. (16), and Sobolev et. al. (26) have done extensive work on hydrocarbons in polyisobutylene, xylene in polyethylene, and methyl bromide and isobutene in polyethylene, respectively. Raff and Allison (24) have compiled most of the significant data on polyethylene in their publication. Prager and Long (23) and Sobolev et. al. (26) in their work had a concentration influence in their diffusion constant while Michaels et. al. (16) were not troubled by a concentration influence having preswollen their polyethylene film in xylene.

The employment of a tritiated toluene tracer in the present work has reduced the need for extensive equipment to a minimum.

The results for each organic liquid-polyethylene system were studied in order to formulate and to verify an Arrhenius type relationship of the form $D_s = D_0 \exp(-\Delta E/RT)$.

THEORETICAL BACKGROUND

Concept of Diffusion

The permeation process of a liquid through a polymer film occurs in three steps (7):

1. solution of the permeating liquid molecules at the surface of the film
2. diffusion of these liquid molecules through the film
3. desolution of the diffused liquid molecules at the opposite film surface.

Of these three steps, the second, diffusion through the film, is by far the slowest and is, accordingly, the rate controlling step.

The concept of diffusion has been and still is a process poorly understood. Several theories as to the exact mechanism have been proposed and supported; but to date none is more widely accepted than Eyring's Hole Theory of Diffusion for the diffusion through solids.

In the Eyring Hole Theory of Diffusion an amorphous polymer is visualized as a random mass of polymer chains and holes. These segments of polymer chains and holes are thought of as being arranged in some quasi-crystalline lattice. Above the glass transition temperature of the polymer, thermal motion results in the continual disappearance and reformation of these holes in the polymer. It is this availability of holes that promotes diffusion. Molecules are supposed to diffuse by "jumping"

from hole to hole as a result of a concentration gradient being set-up in the polymer. Thus, the diffusing molecules work their way through the polymer, the speed of their diffusion depending on the availability of a hole. This hole must be at least the same size as the diffusing molecule; therefore, the larger the diffusing molecule, the less chance there is that it would find a suitably large hole and the slower it should diffuse.

There is also some energy attributable to hole formation. This "activation energy" for the diffusion process is associated with the energy required for the hole formation against the cohesive forces of the polymer. It is also related to the energy a diffusing molecule must acquire to enable it to "jump" from one hole to another. Therefore, the looser a polymer is; in other words, the less crystalline or the more amorphous it is, the faster the diffusion should be due to the lower activation energy required. Lack of symmetry in a polymer leads to a larger diffusion constant for it than for another polymer of similar cohesive energy but more symmetric. Similarly, for two symmetric polymers, the polymer which is more polar, thus exhibiting higher cohesive energy, would produce a lower diffusion rate than the non polar polymer.

Now that an explanation of the mechanism of diffusion has been presented, let us consider the calculation of the diffusion rate.

Consider a film x cm. thick with a cross-sectional area for diffusion A cm.². This film is exposed to a fluid containing solute (tracer) at concentration C_A^* on one side and at a lower concentration C_B^* on the other side as shown in Figure 1.

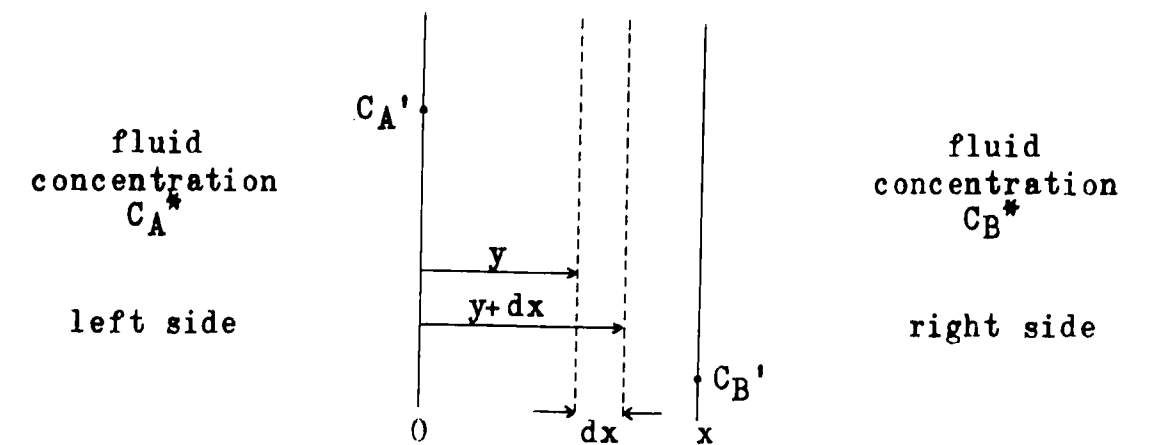


Figure 1

The boundary conditions resulting from this presentation are:

1. at $x=0$, $C=C_A'$
2. at $x=x$, $C=C_B'$

where the concentrations, C_A' and C_B' , are the concentrations in the first and last layer of the film, respectively, in the permeation direction.

On assuming Henry's law, these concentrations can also be expressed as follows:

$$C_A' = sC_A^* \quad [1]$$

$$C_B' = sC_B^* \quad [2]$$

where s is the solubility of the fluid in the polymer.

Let us now consider a differential element, dx , in the film, such that, at a distance y , the rate of permeation will be J^* cc./sec., and correspondingly, at a distance $(y+dx)$, the rate of permeation will be $J^* + (dJ^*/dx)dx$.

Therefore the amount of diffusant retained per differential volume will be $-(dJ^*/dx)/A$.

This is equal to the rate of change of concentration, C , in the film with respect to time.

$$-\frac{dJ^*}{dx} = \frac{dC}{dt} \quad [3]$$

For steady state diffusion, $dC/dt=0$ and dJ^*/dx is a constant, thus, J^* is also a constant (28).

Fick's first law of diffusion (1), which expresses the rate of permeation in terms of a concentration gradient across the film, is given by:

$$\frac{J^*}{A} = -D \frac{dC}{dx} \quad [4]$$

where D is the diffusion constant.

Substituting [4] into [3] and rearranging,

$$\frac{dC}{dt} = \frac{d}{dx} \frac{dC}{dx} D \quad [5]$$

Assuming D is independent of concentration:

$$\frac{dC}{dt} = D \frac{d^2C}{dx^2} \quad [6]$$

which is Fick's second law of diffusion (1).

Equations [4] and [6] are the basic diffusion equations. For steady state diffusion, which is approximated in the present work, the above equations reduce to a more convenient form. The approximation for steady state diffusion is justified in the present work since the left side concentration does not drastically change during the experiment. A calculation made for the effect of back diffusion from the right side to the left side showed the

result to be negligible when compared to the right side concentration. As has previously been stated, in steady state diffusion J^* is a constant, so that, equation [5] can be directly integrated to give a simple equation to solve for the diffusion constant.

$$\frac{J^*}{A} \int_{x=0}^{x=x} dx = -D \int_{C=C_A'}^{C=C_B'} dC$$

or,

$$\frac{J^*}{A} = \frac{D(C_A' - C_B')}{x}$$

$$D = J^* \frac{x}{A (C_A' - C_B')} \quad [7]$$

Equation [7] defines the diffusion constant in terms of the rate of permeation, J^* , and the concentration difference of the diffusing specie across the film of thickness x .

In the present work, this film concentration will be in reality, the concentration of the radioactively labeled specie in the film. Now substituting equations [1] and [2] into equation [7], the following is arrived at:

$$(Ds) = \frac{J^* x}{A (C_A^* - C_B^*)} \quad [8]$$

From equation [8] it is seen that the measurement of the rate of permeation yields the diffusion constant coupled with the solubility of the diffusing specie in the film.

Several expressions have been proposed to relate the diffusion constant to temperature as well as the permeability and the solubility to temperature; the most widely accepted form is that of an Arrhenius equation

which applies to activated processes such that:

$$D = D_1 \exp(-\Delta E_D/RT) \quad [9]$$

$$P = P_1 \exp(-\Delta E/RT) \quad [10]$$

$$S = S_1 \exp(-\Delta H/RT) \quad [11]$$

where ΔE_D is the activation energy for the diffusion process, ΔE is the energy for the permeation process, and ΔH is the heat of solution of the diffusing molecules in the polymer and D_1 , P_1 , and S_1 are constants.

The permeability is the combination of the diffusion constant and the solubility at a temperature, such that:

$$P = (DS) \quad [12]$$

then,

$$P = D_1 S_1 \exp\{-[\Delta E_D + \Delta H]/RT\} \quad [13]$$

or,

$$P_1 = D_1 S_1 \quad [14]$$

$$\Delta E = \Delta E_D + \Delta H \quad [15]$$

Therefore, measurements of the rate of permeation through a polymer film do not directly yield a value for the activation energy for the diffusion process alone. A simplified mathematical approach, for the determination of the diffusion constant alone has been devised (1, 28) and, was recently modified by Paul and DiBenedetto (21) to yield both the diffusion constant and the solubility with their corresponding energy terms from just the permeability data alone. Both methods employ a time-lag technique to calculate the diffusion constant. Figure 2 is a typical curve for the amount of fluid permeated versus time.

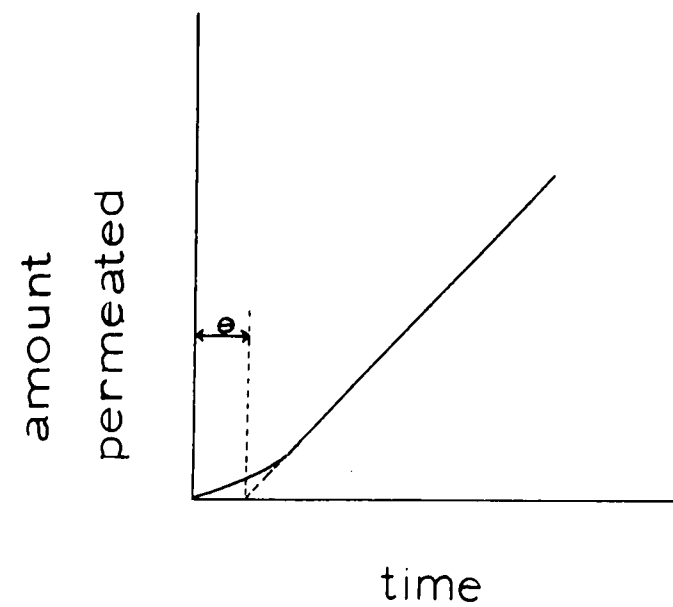


Figure 2

This time-lag method employs the extrapolation of the steady state portion of the curve back to zero amount permeated. This value of time, θ , is called the time-lag for steady state attainment and the diffusion constant can be calculated directly from this value by (1,28):

$$D = x^2/6\theta \quad [16]$$

where x is the film thickness.

Thus, the solubility is readily obtainable from equation [12]. The method of Paul and DiBenedetto (21) which is more rigorous and exact, is concerned primarily with gas permeation but can be modified for a liquid permeation process.

Tracer Technique

Tritated toluene, the tracer used in the present work, emits negatively charged beta particles. The initial concentration of the tracer is 0.141 millicuries per cubic centimeter. This means that $(3.7 \times 10^7)(0.141)$ beta particles are emitted within one milliliter of tritated toluene per second. Through the use of a liquid scintillation spectrometer, it is possible to count beta particle emission with a high degree of efficiency and discrimination. If this one milliliter of tritated toluene is diluted to one hundred milliliters, the total emission of the one hundred milliliters is that for the original one milliliter; but now the emission per milliliter has been reduced ninety-nine fold. This type of dilution is that which occurs in the present work. In order to calculate the radioactive concentration in any given sample, the number of beta emissions per sample is counted for a preset time. This emission rate divided by the efficiency of the liquid scintillation spectrometer yields the number of tritium molecules which decayed. This amount of tritium molecules which have decayed is the activity of the sample. On dividing the activity of the sample by the sample volume and using appropriate conversion factors, the radioactive concentration of the sample is calculated.

Factors Effecting Permeation

The solubility of a fluid in a polymer depends on their mutual compatibility and, in general, the principle of "like dissolves like" is applicable. Likewise, the permeability also increases with similarity in chemical structure between the polymer and the fluid penetrant. This borne out by the fact that the permeation rate through polyethylene is lowest with strongly polar penetrants and greatest with hydrocarbons (15,22). These effects were confirmed by the data from the present work.

The permeation rate decreases as the symmetry and the cohesive energy density of the polymer increases. (31)

The effect of polymer cross-linking on permeation is significant. The permeation rate through polyethylene (27) is seen to decrease as the degree of cross-linking in the polymer is increased. It has been proposed that the decrease in the permeation rate is due to a decrease in the entropy of activation for the diffusion process. This entropy is related to the probability of the polymer chains moving away from some central point, thereby, creating a hole for diffusion to occur. This probability will decrease if the polymer chain segments are tied together at intervals by cross-linking (28).

The crystallinity of a polymer also effects the permeation rate. As the degree of crystallinity is increased the permeation rate decreases. As a result of their work, Michaels and Parker (17), it has been proposed that a

polymer should be considered a "porous medium," the "particles" of which are the crystallites and the "pores" of which are the amorphous phase. Impedance to diffusion is shown to depend primarily on the geometry of the crystalline (impermeable) phase, its volume concentration and to be independent of the crystallite size. These crystallites reduce polymer chain segment mobility in the amorphous phase, thus increasing the energy barrier for diffusion and decreasing the diffusion rate.

The value of the diffusion constant depends very highly on the concentration of solvents in the polymer film. Numerous expressions have been proposed to relate the diffusion constant to the solubility of the solvent in the film but none is more widely accepted than the following:

$$D = D_0 \exp(ac^*) \quad [17]$$

where D is the diffusion constant, c^* is the concentration of the solvent in the polymer, and D_0 and a are constants.

Figure 3 represents a typical concentration gradient in a polymer film.

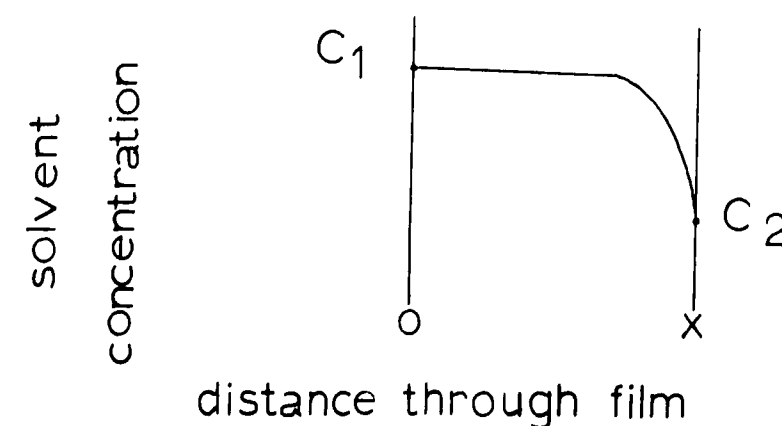


Figure 3

As can be seen in Figure 3, there is a very steep solvent concentration gradient near the downstream side of the polymer film. This indicates the fact that essentially all resistance to permeation is near the downstream side of the film. (7)

The size and shape of the penetrant molecule also have a marked effect on the permeation rate. Park (19,20,28) has concluded that the probability of a diffusing molecule in a polymer moving from one position to another is proportional both to the probability of finding a gap between the polymer chains wide enough to pass the penetrant and to the probability of finding a hole in the polymer matrix large enough to accommodate it. Thus the larger the penetrant molecule, the further must the polymer segments move to allow the penetrant to pass and thus causing an increase in the activation energy and, correspondingly, a decrease in the diffusion constant. However, other factors such as chain flexibility and the segmental chain length involved per unit diffusion step must also effect the ease of diffusion so that the stated dependence on hole size and volume alone can only be part of the actual conditions governing diffusion. Except for small and simple molecules, however, the effect of penetrant solubility usually overshadows the influence of penetrant molecular diameter (7).

Permeation through swollen and unswollen polymers does not yield the same results. As a "dry" film begins

the uptake of a liquid solvent it undergoes a change in thickness which can result in a changing value for the diffusion constant. However, more important is the fact that the film undergoes a constant increase in the solvent concentration until the equilibrium concentration is reached. During this time, which is usually several days, the diffusion constant is not a constant value but continually changes due to change in solvent concentration, as shown by equation [17]. Therefore, calculations made for the diffusion constant during the equilibration period must always take account of the fact that the diffusion constant is not uniform throughout the film when the concentration is not uniform throughout the film. In the work reported here, these difficulties were avoided by measuring the diffusion of a small quantity of radioactively labeled component through a film swollen to equilibrium.

DESCRIPTION OF APPARATUS AND MATERIALS

Experimental Apparatus

Several types of diffusion cells have been developed to measure permeation through polymer films, but none is simpler or as trouble-free as the apparatus required for the present work. Previous researchers have required extensive equipment and usually the need for a complicated high vacuum system to aid in their investigations, but this is not the case in this work.

The basic piece of equipment is a twin-chamber brass diffusion cell, which was constructed from three-sixteenth inch thick brass plate. This rectangular cell, as depicted in Figure 4, has overall dimensions of $4\frac{9}{16}$ in. by $2\frac{3}{8}$ in. by $2\frac{7}{16}$ in. Each chamber has inner dimensions of 2 in. by 2 in. by $2\frac{1}{4}$ in. The cell is silver soldered and thereby made leak tight. The chambers are separated by a brass partition which has dimensions of 2 in. by $\frac{3}{16}$ in. by $2\frac{1}{4}$ in. and itself lead soldered into place. A $1\frac{1}{8}$ inch diameter circular hole was drilled into the center partition with its center at a distance of 1 inch from the bottom and each side.

Brass flanges, one for each side of the partition, were then fashioned to fit into the center hole. The flanges were beveled as shown in Figure 5. Each flange was dimensioned so that when the unit was assembled, the beveled portions made contact with each other. Each flange was drilled and tapped to accommodate four brass

DIFFUSION CELL



Figure 4

FLANGE SCHEMATIC

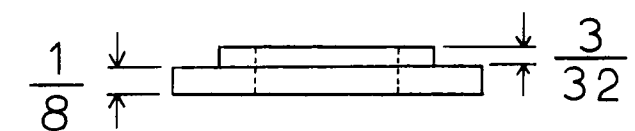
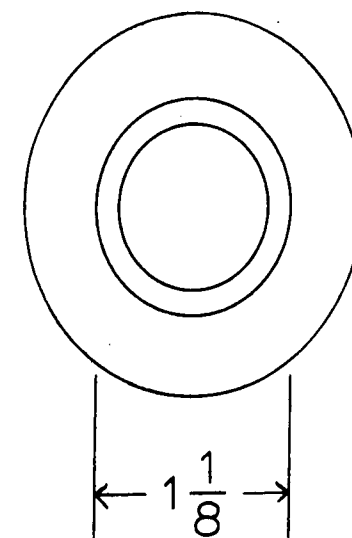
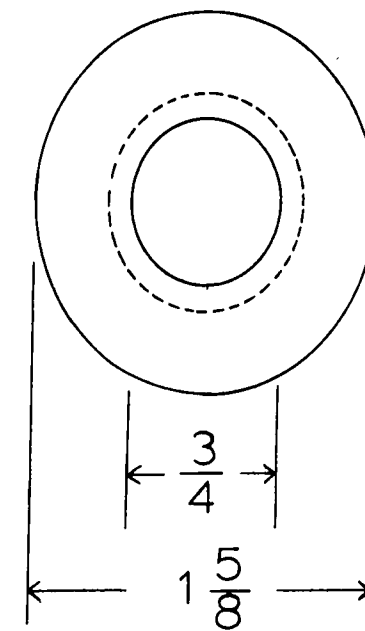


Figure 5

screws in order to secure it to the center partition.

Two thiokol rubber gaskets were fashioned to fit the flanges and give a tighter fit between the center partition and the flange.

Two additional thiokol rubber gaskets were fashioned to fit the beveled portion of the flanges exactly. It was between these two gaskets that the polymer film to be studied was placed.

A brass plate was made to fit the top of the diffusion cell. Two 3/4 inch diameter holes were drilled in the plate so that when the plate was placed on the diffusion cell, the center of each hole would correspond to the center of each chamber. These holes enabled a glass stirring rod, 16 mm. diameter propeller, to be admitted to each chamber.

A constant temperature water bath controlled to ± 0.02 C was employed in order to assure a constant liquid temperature in the diffusion cell. The diffusion cell was placed into this water bath and rested upon a tripod. The liquid level in the water bath was maintained at a distance of 1/8 inch from the top of the diffusion cell.

One stirrer was needed for mixing the contents of each diffusion chamber. Each stirrer was attached to a powerstat. This enabled the same degree of mixing in each chamber to be achieved without splashing. Mixing was always maintained sufficiently to insure that the only resistance to transport was diffusion through the film.

The entire apparatus is shown in Figure 6.

Since the novel feature of the present work is the employment of the tracer technique, an instrument was needed to measure the concentration of the tracer. Shown in Figure 7 is the Tri Carb Liquid Scintillation Spectrometer Model 2101 which was used for all tracer measurements. The spectrometer counts the number of beta particles emitted by a radioactive sample for any preset time. Then, from this emission rate, the actual amount of tracer element present in the sample can be calculated. The operation (33) of the spectrometer will not be dealt with here in detail except to mention that each sample in order to be counted by the spectrometer had to be placed in 15 milliliters of a specially prepared scintillation cocktail. This scintillation cocktail converted the beta particle energy emitted by the tracer to light quanta which were detected by the spectrometer and recorded.

EXPERIMENTAL APPARATUS SETUP

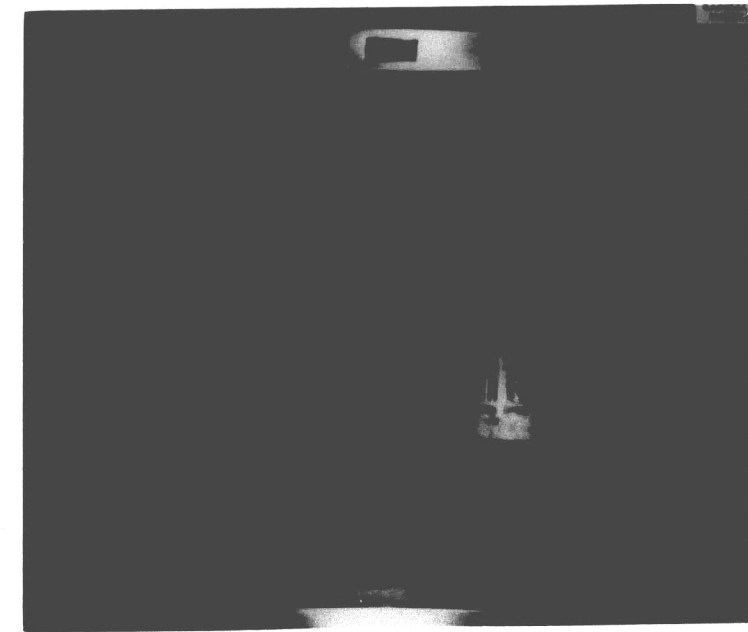


Figure 6

TRI CARB LIQUID SCINTILLATION SPECTROMETER



Figure 7

Reagents and Materials

The polyethylene film used in the present work was 1.5 mils thick with a density of 0.920 to 0.930 grams per cubic centimeter and a number average molecular weight of 40-50,000. This film was supplied by the Celanese Plastics Company.

The tracer used in this work was tritiated toluene having an initial activity of 0.141 millicuries per cubic-centimeter. The tracer was obtained from Isotopes Inc.

The scintillation cocktail was prepared by adding 0.3 grams 1,4-bis-(2-4(methyl-5-phenyloxazolyl)) benzene and 5.0 grams 2,5 diphenyloxazole together in a one liter volumetric flask which was then brought up to volume with toluene (6). The 1,4-bis-(2-4(methyl-5-phenyloxazolyl)) benzene and the 2,5 diphenyloxazole were obtained from the Packard Instrument Co. Inc.

The organic liquids employed as the swelling agents were all Baker Analyzed Reagent Grade with the exception of cyclohexane and tetrahydronaphthalene. The tetrahydronaphthalene was the Baker Practical Grade, and the cyclohexane was an Eastman Chemical Company product which was of a higher quality than the Baker Analyzed Reagent Grade.

The gasket material was a thiokol rubber obtained from the Reliable Rubber Company. This gasket material was found the most suitable for the organic liquids used since it did not swell as much as other rubber gasket materials tested. The thiokol rubber also did not release

its high molecular weight components into the organic liquids causing a color change as the other rubber products did when tested. The other rubber gasket materials tested were neoprene and Buna-N. Teflon was also tested but did not yield a leak tight seal.

EXPERIMENTAL TECHNIQUE

Eight organic liquids, each used at four different temperatures in the temperature range 25 to 40 °C in intervals of about 5°C, were employed as the swelling agents in the present work. These eight organic liquids were toluene, chlorobenzene, mesitylene, cumene, ethylbenzene, cyclohexane, tetrahydronaphthalene, and decahydronaphthalene.

In the present work the diffusion chamber into which the initial tracer was added will be referred to as the "hot" chamber; and, similarly, the chamber into which no tracer was initially added will be referred to as the "cold" chamber.

For each run, the second set of thiokol gaskets, those which were to be placed on the beveled portions of the brass flanges, were swollen to equilibrium in the organic liquid to be used as the swelling agent prior to insertion in the cell. The gaskets were allowed to remain in the swelling agent about twelve hours. This insured no further swelling of the gaskets while in the diffusion cell, thus keeping the cross-sectional area of the polymer film for diffusion constant.

A one inch diameter section of polyethylene film served as the medium for diffusion. This section was cut from a sheet of the film. One of the two brass flanges was then secured, though not completely in the diffusion cell along with its accompanying gasket. Then one of the

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swollen gaskets was placed in the circular portion of the center partition on the beveled portion of the flange. The polymer film was now placed on this swollen gasket and immediately the second swollen gasket was placed upon the polyethylene film. Finally, the second flange and its gasket were secured to the center partition and both flanges tightened fast.

Now the organic liquid to be used as the swelling agent was measured and placed in the diffusion cell. One hundred and nineteen milliliters of the liquid were placed in the "hot" chamber and one hundred and twenty milliliters were placed in the "cold" chamber.

The diffusion cell, as now prepared, was placed on the tripod in the constant temperature water bath. The cover was placed over the diffusion cell and the stirring rods placed in the chambers.

The diffusion cell was allowed to remain in the constant temperature bath for forty-two hours before diffusion measurements were initiated by the addition of the tracer. This time was chosen to insure equilibrium swelling of the polyethylene film by the organic liquid.

While waiting for the attainment of equilibrium, a determination of the liquid evaporation rate from the cell under the same conditions to be employed for diffusion measurements was made. The stirring rate was maintained as constant as possible for each chamber by the use of a powerstat. The speed of the stirrer was such to avoid

splashing of the contents of the cell. The calculation of the evaporation rate would lend itself to calculation of an average volume for each chamber. The effect of evaporation itself makes no difference in the determination of the "hot" or "cold" chamber concentration, since on evaporation from either chamber the liquid evaporated has the same concentration of tracer as that liquid in the chamber, thereby, resulting in no net concentration change. This average volume value was needed in order to calculate a permeation constant. The chambers were maintained at their original volume during this period of equilibration, except for the short time when the evaporation rate determination was being carried out, by addition of liquid at the same temperature as that in the cell.

After the equilibration period, the chambers were brought up to their respective volumes for the final time prior to the addition of the tracer.

As soon as this final volume attainment was made, one milliliter tritiated toluene having a concentration of 0.141 millicuries per milliliter was added by means of a pipette to the "hot" chamber and time zero for diffusion measurements was realized. Thus, at the start of the tracer diffusion, both chambers were at equal volumes.

For the next four hours, at thirty minute intervals after the addition of the tritiated toluene, one-half milliliter samples were withdrawn from each chamber by means of a pipette. In order to take a sample, the stirrers were

turned off, and pipettes, one for each respective chamber, were inserted through the holes in the cover into the "hot" or "cold" chamber, respectively, and a sample was withdrawn. After samples were withdrawn from both sides, the stirrers were again turned on. This procedure was followed throughout the four hour period.

Prior to the addition of the tritiated toluene to the cell, fifteen milliliters of the specially prepared scintillation cocktail were pipetted into twenty-two milliliter glass sample bottles equipped with a polyseal insert in the plastic cap for a sure seal.

After the one-half milliliter sample was withdrawn from the cell, it was emptied into the previously prepared sample bottle. The fifteen milliliters of scintillation cocktail in each sample bottle were held constant for all samples to insure the same value of counting efficiency for any given system. Each sample bottle's cap was secured tightly to avoid evaporation of the contents. The sample bottles with their radioactive contents were then placed into a dark room, in which the scintillation spectrometer was kept, for a period of at least twelve hours. This so called "cooling-off" period allowed the contents of the sample bottles to reach room temperature. But mainly, since the scintillation cocktail is sensitive to the ultraviolet rays of the sun and to the fluorescent lights in the laboratory, this period allowed decay of ultraviolet activated molecules to the ground state. A small red light

was the only light in the room.

After the "cooling-off" period each sample bottle was placed in the scintillation spectrometer and the number of beta particles emitted for a preset time was counted. Half-way through the counting of the samples from any one run, a Tri Carb standard was placed in the spectrometer and counted. This standard served to determine the efficiency of the spectrometer.

To calculate the effect of background radiation a blank cocktail was counted. This blank cocktail consisted only of the scintillation cocktail and a non-radioactive one-half milliliter sample of the respective organic liquids. For the Tri Carb standard, which was used to determine the efficiency of the scintillation spectrometer, a Tri Carb blank standard was used to determine its background. After the blank was counted, subtraction of its value from that for the sample would yield the sample's true reading.

This procedure was followed throughout the present work for all the organic liquids with the exception of cyclohexane, in which the actual sample time was cut to two hours and sampling was carried out every fifteen minutes. The purpose of this deviation was caused by the higher diffusion rate for cyclohexane and the desire of the researcher to keep the amount of tracer transferred fairly consistent with that observed during the other trials.

DISCUSSION OF RESULTS

The data from the present work may be found in tabular form in Tables 1 to 16 with the corresponding final permeation equations in Table 20.

The present work yielded only the values for the permeation constant and the energy for the permeation process. The diffusion constant could not be directly calculated as indicated by equation [16] due to the inaccessability of the value for the time-lag. As can be seen from a typical plot for the "cold" chamber tracer concentration versus time, Figure 8, the steady state tracer permeation through the polyethylene film was achieved in a very short time, in most cases less than two minutes after addition of the tracer. Thus with such a small time increment and the drawing of the "best" straight line through the data, the time-lag was seen to be indeterminate. With reference to the plots which are extrapolated to give a negative time-lag, this was due to the retention of some radioactive tracer in the gaskets from a prior run, since the gaskets were interchanged after each run. Correction was made for this initial radioactive tracer but still no certain time-lag was obtained.

The use of the method of Paul and DiBenedetto (21) to obtain the solubility of the liquid in the polymer could not be carried out due to the uncertain time-lag values.

The permeation constant values at the different temperatures conform to an Arrhenius type relation as shown by the graph in Figure 9. However there is some inconsistency shown near 30°C where the logarithm of the permeability does not fall on the straight line indicated by the other data. It is supposed that this fact is due to some inherent physical or chemical property of the polyethylene film itself and not to the data or the method since the effect can be observed for most of the liquids.

The permeability does not seem to follow any set pattern with regard to molecular size, as is seen by Table 21, although the factor of solubility could be highly important.

The solubilities of toluene and chlorobenzene in polyethylene (8) are approximately the same, while the solubility of cyclohexane in polyethylene (8) is much greater. From the values calculated for the activation energy of the permeation process a pattern can possibly be recognized since the energy values for toluene and chlorobenzene are very close while that for the cyclohexane is very low, possibly indicating the effect of solubility on the permeation energy. The increased solubility could somehow lend itself to decrease the resistance to permeation. The use of ethylene dibromide and nitrobenzene, both having low solubilities in polyethylene (8), were considered; however, due to the high cost and the high toxicity, respectively, of these substances, no attempt was made to

employ them.

An evaluation was made to determine whether the heat of solution term, as shown in equation [15], could be considered negligible in comparison to the activation energy for the permeation process, thereby, arriving at a satisfactory value for the activation energy for the diffusion process. However, Michaels et. al. (16) arrived at a value of 4.1 Kcal./g. mole for the heat of solution of liquid p-xylene in polyethylene film while Sobolev et. al. (26) computed a value of 5.7 Kcal./g. mole for the heat of solution of methyl bromide in polyethylene film. Klute and Franklin (10) found a value of 5.5 Kcal./g. mole for the heat of solution of water vapor in polyethylene film. These findings lend support to the fact that the heat of solution value is significant and separate work must be done to determine it and, correspondingly, to determine the true value for the activation energy for the diffusion process.

Any attempt to compare the values for the activation energy for the permeation process of the present work to those found by other researchers is meaningless. Since, as has been pointed out, there are many factors, such as degree of crystallinity and cross-linking, low density or high density polyethylene, which cause the polyethylene used by one observer to differ from that used by another observer. Also each processor of the polyethylene film does not prepare the film in exactly the same fashion

again adding to the differences in the polyethylene films.

Figure 10 which depicts a plot of $\ln(D_0)$ versus ΔE , shows a straight line relationship with some scatter. This is a modified form of the "linear free energy relationship" which is said to exist for a process in similar systems whose rate constants show the temperature dependence of the Arrhenius type (2,11,12,21). Very good agreement is thus shown, considering the work of other researchers who have realized more scatter than found in the present work (21).

Tables 17 and 18 present the pertinent values leading to the calculation of the permeation constant while Table 19 lists the values obtained for the energy for the permeation process for each system. Presented in Table 20 are the final empirical equations for the permeation of the toluene specie through the swollen polyethylene film in the temperature range 25 to 40 °C.

However the most important feature of the present work is the highly successful operation of the novel technique employed for the measurement of the liquid permeation through the polymer film. The conformity of the data to the Arrhenius equation and the "linear free energy relationship" is satisfactory.

CONCLUSIONS AND RECOMMENDATIONS

The effect of the substitution of the isopropyl side chain on the benzene ring in place of the ethyl side chain on ethylbenzene is seen to produce a marked effect on the permeation energy yielding 16,675.40 cal./g. mole and 11,414.84 cal./g. mole for cumene and ethylbenzene, respectively. Consideration should be given to the use of straight-chain and branched-chain hydrocarbons for liquid permeation measurements.

The effect of solubility of the liquid in the polyethylene film is the most important factor in this work. Work must be done with liquids of varying solubilities in the polyethylene film in order to test the hypothesis of the increased permeability with the increased solubility of the liquid in the polyethylene film.

To check the accuracy of the Paul and DiBenedetto method when applied to the present work, it is proposed that numerous determinations of the solubilities of various liquids in the polyethylene film be made through the construction of a separate solubility apparatus and the experimental results compared to their theoretical results.

To employ equation [16] for the direct determination of the diffusion constant it is suggested that thicker films be utilized, which will give a larger and more certain time-lag value.

The effect often observed of temperature on the

permeation constant was verified to be a linear relationship, and this fact was further verified by the agreement with the "linear free energy relationship" aspect.

The use of new gaskets for each run is strongly suggested to offset any radioactive tracer buildup in the old gaskets which might lead to an erroneous time-lag value if not corrected.

Along with processing of new data with regard to organic liquid permeation through polyethylene film, the successful operation of a new and simplified technique for measuring the liquid permeation through a polymer film is the prime contribution this work has presented.

APPENDIX

SAMPLE CALCULATION

The following is an example of the method followed in determining the permeability of the diffusing specie through the polyethylene film.

The following notation is used:

- C_a^+ number of beta particles counted per minute for the liquid sample in the "hot" chamber including background excluding efficiency (cpm)
- C_a^- number of beta particles counted per minute for the liquid sample in the "hot" chamber less background excluding efficiency (cpm)
- C_b^+ number of beta particles counted per ten minutes for the liquid sample in the "cold" chamber including background excluding efficiency (counts/10 min.)
- C_b^- number of beta particles counted per ten minutes for the liquid in the "cold" chamber less background excluding efficiency (counts/10 min.)
- C_A "hot" chamber tracer concentration less background excluding efficiency (mc./cc.)
- C_B "cold" chamber tracer concentration less background excluding efficiency (mc./cc.)
- C_A^* "hot" chamber tracer concentration less background including efficiency (mc./cc.)
- C_B^* "cold" chamber tracer concentration less background including efficiency (mc./cc.)
- V_{avg} average volume of chamber (cc.)
- x thickness of film (cm.)
- A cross sectional area of film for diffusion (cm.²)
- E spectrometer efficiency
- J permeation rate excluding efficiency (mc./sec.)
- J^* permeation rate including efficiency (mc./sec.)
- (D_s) permeability (cm.²/sec.)

- T temperature ($^{\circ}$ K)
- ΔE activation energy for the permeation process
(cal./g. mole)
- D_0 constant (cm.²/sec.)
- R gas constant (1.987 cal./g. mole $^{\circ}$ K)
- B background (counts/time)

For the calculation of the "hot" chamber and the "cold" chamber liquid sample counting rate less background the equations are seen to be:

$$C_a^- = C_a^+ - B \quad [18]$$

$$C_b^- = C_b^+ - B \quad [19]$$

Taking into account the efficiency of the liquid scintillation spectrometer, E, the following equations are found:

$$C_A^* = C_A/E \quad [20]$$

$$C_B^* = C_B/E \quad [21]$$

$$J^* = J/E \quad [22]$$

A millicurie (mc.) is defined (4) as 3.7×10^7 disintegrations per second. Therefore the "hot" chamber tracer concentration at any time, t, is found:

$$C_A = \frac{C_a^-(\text{cpm})}{1/2 \text{ cc.}} \frac{\text{min.}}{60 \text{ sec.}} \frac{\text{mc.}}{3.7 \times 10^7 \text{ dis./sec.}}$$

$$C_A(\text{mc./cc.}) = 9.00901 (10^{-10}) C_a^- \quad [23]$$

Correspondingly, the "cold" chamber tracer concentration at any time can be written as:

$$C_B = \frac{C_b^-(\text{counts}/10 \text{ min.})}{(1/2 \text{ cc.})(10 \text{ min.})} \frac{\text{min.}}{60 \text{ sec.}} \frac{\text{mc.}}{3.7 \times 10^7 \text{ dis./sec.}}$$

$$C_B(\text{mc./cc.}) = 9.00901 (10^{-11}) C_b^- \quad [24]$$

To determine the rate of permeation, J, a graph

of the "cold" chamber tracer concentration, C_B , versus time was plotted, the slope of which gave the rate of permeation per "cold" chamber volume. Since the chamber volume was decreased with time, an average value was computed using the volumes calculated at each of the extremes on the C_B versus time plot for any particular run. An analysis of the error introduced by this choice of an average volume will be presented in the following section on error analysis. The product of this average volume times the slope of the C_B versus time curve gives directly the rate of permeation, J .

Through the use of stirrers in the cell, an assumption of perfect mixing in each chamber is justified.

The value for the concentration of the tracer in the "hot" chamber is very large compared to that for the tracer in the "cold" chamber, so that, the value for the "cold" chamber can be considered negligible in comparison to the "hot" chamber tracer concentration. This further reduces equation [8] to:

$$(Ds) = J^* \frac{x}{A} \frac{1}{C_A^*} \quad [25]$$

In a separate experiment the difference between the thickness of a polyethylene film in the unswollen state with that for the film fully swollen in each respective swelling agent could not be detected using a micrometer with a sensitivity of ± 0.01 mil. This then justified the use of the unswollen film's thickness.

On substituting equations [20] and [22] into

equation [25], the final working equation for the determination of the permeability is found to be:

$$(Ds) = J \frac{x}{A} \frac{1}{C_A} \quad [26]$$

For the purpose of clarity, the calculation of the permeation of the tracer through the polyethylene film swollen in toluene at 25°C will be made in detail.

From the data in Table 1 a graph of the concentration of the tracer in the "cold" chamber versus time was plotted as shown by Figure 8.

On taking the slope of this plot, it is found:

$$\begin{aligned} \text{slope} &= \frac{(5.3093-0.5832)(10^{-6}) \text{ mc./cc.}}{242-32 \text{ min.}} \\ &= 2.2505(10^{-8}) \text{ mc./cc.}(min.) \end{aligned}$$

The chamber volume was therefore found by taking the initial chamber volume, 120.0 ml., and subtracting the volume evaporated and/or volume withdrawn by sampling at the time the sample was taken.

At the time of sample withdrawal	V ml.	V _B ml.	Reason
0	-	120.00	-
1	0.15	119.85	evaporation
2	0.65	119.20	evaporation & sample
3	0.65	118.55	evaporation & sample
4	0.65	117.90	evaporation & sample
5	0.65	117.25	evaporation & sample
6	0.65	116.60	evaporation & sample
7	0.65	115.95	evaporation & sample
8	0.65	115.30	evaporation & sample

The average volume is the average of the volumes corresponding to the extremes of the steady state portion of the C_B versus time plot, Figure 8.

$$V_{\text{avg}} = \frac{119.85 + 115.30}{2} \text{ cc.}$$

$$= 117.575 \text{ cc.}$$

The rate of permeation then is found to be:

$$J = \frac{2.2505(10^{-8}) \text{ mc. min.}}{\text{cc. min.}} \frac{117.575 \text{ cc.}}{60 \text{ sec.}}$$

$$= 4.4101(10^{-8}) \text{ mc./sec.}$$

The concentration of the tracer in the "hot" chamber is found by using equation [23] to be:

$$C_A = 9.00901(10^{-10}) C_a$$

$$= 9.00901(10^{-10}) (488295)$$

$$= 4.3994(10^{-4}) \text{ mc./cc.}$$

Therefore, solving for the permeability, it is found that:

$$(Ds) = \frac{4.4101(10^{-8}) \text{ mc.}}{\text{sec.}} \frac{3.81(10^{-3}) \text{ cm.}}{2.85 \text{ cm.}^2} \frac{\text{cc.}}{4.3994(10^{-4}) \text{ mc.}}$$

$$= 1.3401(10^{-7}) \text{ cm.}^2/\text{sec.}$$

where $x=3.81(10^{-3})$ cm. and $A=2.85 \text{ cm.}^2$, the respective thickness and cross sectional area of the film.

The determination of the activation energy for the permeation process can be seen to be from equations [10] and [12] the slope of a plot of $\ln(Ds)$ versus $(1/RT)$ where the constant P_1 is the intercept at $(1/RT)=0$.

For the system of the toluene tracer permeating through the polyethylene film swollen in toluene this calculation will be shown.

From the plot of $\log_{10}(Ds)$ versus $(1/T)$, Figure 9, it is found:

$$\text{slope} = \frac{2.3025(\log_{10}4.7989(10^{-7}) - \log_{10}1.3401(10^{-7}))}{0.0031932 - 0.0033539}$$

$$= \frac{2.3025((-6.318859)-(-6.872867))}{-0.0001607}$$

$$= -7937.78^\circ \text{K}$$

since,

$$\text{slope} = - \frac{\Delta E}{R}$$

$$- \frac{\Delta E}{R} = -7937.78 \text{ K}$$

therefore,

$$\Delta E = 15,772.35 \text{ cal./g. mole}$$

Now to determine the constant, D_0 , the following method is employed.

Since the permeation equation is:

$$(D_s) = D_0 \exp(-\Delta E/RT)$$

therefore,

$$\ln(D_s) = - \frac{\Delta E}{RT} + \ln(D_0)$$

using $T=313.16 \text{ K}$ $(D_s)=4.7989(10^{-7}) \text{ cm.}^2/\text{sec.}$

substituting,

$$2.3025(-6.318859) = - \frac{7937.78}{313.16} + \ln(D_0)$$

$$\ln(D_0) = 10.7982$$

$$D_0 = 4.89326(10^4)$$

Therefore the final permeation equation for the toluene-polyethylene system in the temperature range 25 to 40 °C is:

$$(D_s) = 4.89326(10^4) \exp(-15,772.35/RT)$$

TABLE 1

TOLUENE-POLYETHYLENE SYSTEM

T=25.0°C

B=16.7 cpm

Time min.	Ca ⁺ counts/ 1 min.	Ca ⁻ counts/ 1 min.
31	488312	488295
60	479004	478987
90	485217	485200
121	487374	487357
151	494338	494321
181	486194	486177
211	490737	490770
241	489608	489586

Time min.	Cb ⁺ counts/10 min.	Cb ⁻ counts/10 min.	C _B millicuries/cc.
32	6641	6474	5.8324x10 ⁻⁷
61	13694	13527	12.1365x10 ⁻⁷
91	21251	21084	14.9946x10 ⁻⁷
122	24499	23322	25.5243x10 ⁻⁷
152	35700	35532	32.0117x10 ⁻⁷
182	42670	42508	38.2910x10 ⁻⁷
212	51492	51325	46.2337x10 ⁻⁷
242	59199	58923	53.0923x10 ⁻⁷

T=30.0°C

B=19.7 cpm

Time min.	Ca ⁺ counts/ 1 min.	Ca ⁻ counts/ 1 min.
30.5	484911	483991
60.0	493545	493525
90.5	490904	490334
120.0	479648	479623
150.0	482135	482115
180.0	473155	473135
210.0	479069	479049
240.0	480755	480735

Time min.	Cb ⁺ counts/10 min.	Cb ⁻ counts/10 min.	C _B millicuries/cc.
31.5	12645	12448	1.1214x10 ⁻⁶
61.0	25951	25754	2.3202x10 ⁻⁶
91.5	39046	38849	3.4999x10 ⁻⁶
121.0	52743	52546	4.7339x10 ⁻⁶
152.5	65143	64951	5.8514x10 ⁻⁶
181.0	78313	78116	7.0375x10 ⁻⁶
211.0	92151	91954	8.2841x10 ⁻⁶
241.0	105165	104968	9.4566x10 ⁻⁶

TABLE 2

TOLUENE-POLYETHYLENE SYSTEM

T=35.0°C

B=19.7 cpm

Time min.	Ca ⁺ counts/ 1 min.	Ca ⁻ counts/ 1 min.
30	492902	492332
60	492384	492364
90	496401	496381
126	491976	491956
150	491732	491712
180	491791	491681
210	491597	491577
241	490997	490977

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
31	17120	16922	1.5255x10 ⁻⁶
61	34949	34752	3.1303x10 ⁻⁶
91	52443	52246	4.7063x10 ⁻⁶
127	74595	74398	6.7025x10 ⁻⁶
151	8207	8110	7.3378x10 ⁻⁶
181	107415	107218	9.6593x10 ⁻⁶
211	127114	126917	11.4340x10 ⁻⁶
242	145553	145371	13.0965x10 ⁻⁶

T=40.0°C

B=19.3 cpm

Time min.	Ca ⁺ counts/ 1 min.	Ca ⁻ counts/ 1 min.
30.5	491362	491342
60.5	500604	498932
90.5	436233	434663
120.5	441167	432549
150.5	477702	476106
180.5	437943	436313
210.5	490919	489312
240.5	474306	472721

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
31.5	25983	25791	2.3235x10 ⁻⁶
61.0	51406	51209	4.6132x10 ⁻⁶
91.5	30452	30259	7.2305x10 ⁻⁶
121.5	106053	105360	9.5369x10 ⁻⁶
151.5	130111	129918	11.7043x10 ⁻⁶
182.5	160588	160395	14.4500x10 ⁻⁶
211.5	188316	188623	16.9931x10 ⁻⁶
241.5	215778	215585	19.4221x10 ⁻⁶

TABLE 3

CHLOROBENZENE-POLYETHYLENE SYSTEM

T=25.3°C

B=19.3 cpm

Time min.	Ca ⁺ counts/ 2 min.	Ca ⁻ counts/ 2 min.
30.0	489303	489264
60.0	495112	495079
90.0	492353	492319
120.0	491826	491787
150.0	483935	483946
180.5	487040	487001
210.0	490553	490514
240.0	485403	485369

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
31.0	3256	3163	2.3496x10 ⁻⁷
61.0	6639	6446	5.8072x10 ⁻⁷
91.0	10113	9925	8.9414x10 ⁻⁷
121.0	13516	13322	12.0013x10 ⁻⁷
151.5	17039	16346	15.1766x10 ⁻⁷
181.5	20497	20614	18.5712x10 ⁻⁷
211.0	23515	23322	21.0108x10 ⁻⁷
241.0	27043	26355	24.1937x10 ⁻⁷

T=30.0°C

B=19.3 cpm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
30	12473559	12473366
60	12433240	12433047
90	12383399	12383206
120	12276382	12276639
150	12495144	12494951
180	12365367	12365174
212	12291731	12291533
240	12216639	12216446

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
31	30679	30486	2.7465x10 ⁻⁶
61	59644	59451	5.3559x10 ⁻⁶
91	91343	91155	8.2122x10 ⁻⁶
121	119780	119587	10.7736x10 ⁻⁶
151	150842	150649	13.5720x10 ⁻⁶
181	179036	178843	16.1120x10 ⁻⁶
213	209169	208976	18.8267x10 ⁻⁶
241	239477	239284	21.5571x10 ⁻⁶

TABLE 4

CHLOROBENZENE-POLYETHYLENE SYSTEM

T=35.0°C

B=19.3 cpm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
30	2394978	2394785
60	2366571	2366378
90	2159861	2159668
120	2425175	2424932
150	2383969	2383776
180	2343260	2343067
210	2301562	2301369
240	2266423	2266236

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
31	15305	15612	1.4065x10 ⁻⁶
61	2342	23235	2.0932x10 ⁻⁶
91	31216	31028	2.7133x10 ⁻⁶
121	39157	38964	3.5103x10 ⁻⁶
151	47576	47383	4.2637x10 ⁻⁶
181	55316	55123	4.9660x10 ⁻⁶
211	61320	61637	5.5574x10 ⁻⁶
241	70740	70547	6.3556x10 ⁻⁶

T=39.3°C

B=20.0 cpm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
30.5	2533400	2533200
60.0	2500559	2500359
90.0	2503526	2503326
120.0	2445641	2445441
150.0	2424452	2424252
180.0	2436034	2435834
210.5	2409336	2409136
240.0	2332145	2331945

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
31.5	21724	21524	1.9391x10 ⁻⁶
61.0	33302	33102	2.9822x10 ⁻⁶
91.0	45107	44907	4.0457x10 ⁻⁶
121.0	57932	57732	5.2056x10 ⁻⁶
151.0	70710	70510	6.3522x10 ⁻⁶
181.0	82439	82239	7.4089x10 ⁻⁶
211.0	93661	93461	8.4199x10 ⁻⁶
241.0	103218	103018	9.2809x10 ⁻⁶

TABLE 5

MESITYLENE-POLYETHYLENE SYSTEM

T=25.4°C

B=17.4 cpm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
29	4718964	4718790
60	4713652	4713478
90	4635341	4635167
120	4667112	4666932
150	4647682	4647508
180	4641099	4640925
211	4576384	4576710
240	4571343	4571169

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _b millicuries/cc.
30	9501	9327	0.4403x10 ⁻⁶
61	16461	16327	1.4673x10 ⁻⁶
91	23143	22969	2.0693x10 ⁻⁶
121	30041	29367	2.6907x10 ⁻⁶
151	36951	36777	3.3132x10 ⁻⁶
181	43310	43136	3.3361x10 ⁻⁶
212	50260	50095	4.5131x10 ⁻⁶
241	61294	61120	5.5063x10 ⁻⁶

T=30.0°C

B=22.9 cpm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
31	4703013	4702739
60	4693460	4693231
90	4705431	4705202
120	4611597	4611368
150	4621443	4621219
180	4590216	4589987
210	4543726	4543497
240	4541026	4540797

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
32	9430	9201	0.8289x10 ⁻⁶
61	13213	17984	1.6202x10 ⁻⁶
91	27321	27092	2.4407x10 ⁻⁶
121	37313	37084	3.3409x10 ⁻⁶
151	46292	46063	4.1498x10 ⁻⁶
181	55219	54990	4.9541x10 ⁻⁶
211	65648	65419	5.8936x10 ⁻⁶
241	75156	74927	6.7502x10 ⁻⁶

TABLE 6

MESITYLENE-POLYETHYLENE SYSTEM

T=35.0°C

B=15.1 cm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
30	4820136	4819985
60	4817777	4817626
90	4763392	4763741
121	4769100	4763949
152	4691167	4691016
180	4699699	4699548
210	4689012	4688361
240	4574046	4573895

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
31	14393	14747	1.3236x10 ⁻⁶
61	29722	29571	2.6641x10 ⁻⁶
91	44579	44428	4.0025x10 ⁻⁶
122	59823	59677	5.3762x10 ⁻⁶
153	75922	75771	5.8262x10 ⁻⁶
181	88838	88689	7.9423x10 ⁻⁶
211	102116	102965	9.2761x10 ⁻⁶
241	115391	115740	10.4270x10 ⁻⁶

T=40.0°C

B=15.1 cm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
30	4500730	4300629
61	4404227	4304076
90	4717569	4717413
120	4708611	4703460
150	4599736	4599635
180	4524271	4524120
210	4503620	4503469
240	4438484	4438333

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
31	24474	24323	2.1913x10 ⁻⁶
62	45357	45706	4.1176x10 ⁻⁶
91	67143	66997	6.0358x10 ⁻⁶
121	36027	35876	7.7366x10 ⁻⁶
151	106947	106796	9.6213x10 ⁻⁶
181	130010	129859	11.6990x10 ⁻⁶
211	151979	151828	13.6782x10 ⁻⁶
241	172557	172406	15.5321x10 ⁻⁶

TABLE 7

CUMENE-POLYETHYLENE SYSTEM

T=25.4°C

B=15.1 cpm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
30	4795047	4794396
60	4801084	4800933
92	4753241	4753090
121	4640238	4640137
151	4722722	4722571
130	4704302	4704651
210	4677321	4677179
240	4633046	4632395

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
31	4112	3963	0.3569x10 ⁻⁶
61	2491	3289	0.7459x10 ⁻⁶
92	13529	13373	1.2052x10 ⁻⁶
122	13595	16444	1.4314x10 ⁻⁶
152	21372	21221	1.9118x10 ⁻⁶
131	25936	25335	2.3275x10 ⁻⁶
211	30457	30306	2.7303x10 ⁻⁶
241	34339	34633	3.1251x10 ⁻⁶

T=30.0°C

B=17.9 cpm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
30	4741569	4741390
60	4733030	4737901
90	4760912	4753333
120	4673036	4677877
150	4715677	4715498
180	4590327	4590643
210	4581623	4581444
240	4569913	4569739

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
31	6937	6753	0.6088x10 ⁻⁶
61	13203	13024	1.1733x10 ⁻⁶
91	19649	19470	1.7541x10 ⁻⁶
121	26233	26109	2.3522x10 ⁻⁶
151	32967	32788	2.9539x10 ⁻⁶
181	39383	39704	3.5769x10 ⁻⁶
211	46052	45901	4.1352x10 ⁻⁶
241	52770	52619	4.7405x10 ⁻⁶

TABLE 8

CUMENE-POLYETHYLENE SYSTEM

T=35.4°C

B=15.1 cpm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
30	4794753	4794602
60	4749593	4749442
90	4731477	4731326
120	4700238	4700137
150	4703866	4703715
180	4667054	4666903
210	4591764	4591613
240	4562301	4562150

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
31	13609	13544	1.2205x10 ⁻⁶
61	24952	24801	2.2343x10 ⁻⁶
91	35466	35315	3.1215x10 ⁻⁶
121	46334	46133	4.1606x10 ⁻⁶
151	57055	56904	5.1265x10 ⁻⁶
181	67937	67333	6.1114x10 ⁻⁶
211	79004	78353	7.1039x10 ⁻⁶
241	90059	89908	8.0993x10 ⁻⁶

T=39.3°C

B=17.9 cpm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
30	4344355	4344506
61	4733303	4733624
90	4759030	4758351
120	4705555	4705376
150	4638956	4633777
180	4655003	4654324
210	4629763	4629584
240	4601003	4600324

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
31	19492	19313	1.7399x10 ⁻⁶
62	36827	36648	3.3016x10 ⁻⁶
91	53555	53376	4.8086x10 ⁻⁶
121	68511	68332	6.1560x10 ⁻⁶
151	85709	85530	7.7054x10 ⁻⁶
181	100939	100760	9.0775x10 ⁻⁶
211	117573	117394	10.5760x10 ⁻⁶
241	133735	133556	12.0320x10 ⁻⁶

TABLE 9

ETHYLBENZENE-POLYETHYLENE SYSTEM

T=25.0°C

B=17.9 cpm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
30	4617662	4617433
60	4593713	4593539
90	4619301	4619712
120	4602093	4601914
150	4563415	4563236
180	4572166	4571937
210	4530976	4530797
240	4512210	4513031

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
31	7219	7040	0.6842x10 ⁻⁶
61	15043	14364	1.3391x10 ⁻⁶
91	22241	23162	2.0367x10 ⁻⁶
121	31047	30363	2.7309x10 ⁻⁶
151	39057	38373	3.5025x10 ⁻⁶
181	43733	43554	4.3742x10 ⁻⁶
211	56746	56567	5.0961x10 ⁻⁶
241	65243	65164	5.3707x10 ⁻⁶

T=30.0°C

B=17.7 cpm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
30	4671455	4671270
62	4603556	4603379
92	4655327	4655150
120	4628955	4626773
150	4537644	4537467
182	4542605	4542423
210	4532987	4532310
240	4519362	4519635

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
31	11971	11794	1.0625x10 ⁻⁶
63	24618	24441	2.2019x10 ⁻⁶
93	36518	36341	3.2740x10 ⁻⁶
121	47884	47707	4.2979x10 ⁻⁶
151	59815	59638	5.3728x10 ⁻⁶
183	72357	72180	6.5027x10 ⁻⁶
211	83608	83431	7.5163x10 ⁻⁶
241	95305	95128	8.5701x10 ⁻⁶

TABLE 10

ETHYLBENZENE-POLYETHYLENE SYSTEM

T=35.4°C

B=17.9 cpm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
30	4693476	4693297
60	4656370	4656691
90	4613188	4613009
120	4606023	4605844
150	4513992	4513812
189	4466315	4466636
210	4443102	4442923
240	4409367	4409633

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
31	14556	14377	1.2952x10 ⁻⁶
61	29760	29531	2.6650x10 ⁻⁶
91	44606	44427	4.0024x10 ⁻⁶
121	61332	61203	5.5138x10 ⁻⁶
151	76250	76071	6.8532x10 ⁻⁶
190	96433	96304	8.6760x10 ⁻⁶
211	112645	112466	10.1321x10 ⁻⁶
241	123003	127329	11.5162x10 ⁻⁶

T=40.0°C

B=17.9 cpm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
30	4733649	4773470
60	4696955	4696776
90	4375763	4675535
120	4601622	4601443
150	4573431	4573302
180	4561605	4561426
210	4552910	4552731
240	4530110	4529931

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
31	29826	29647	2.6709x10 ⁻⁶
61	51652	51473	4.6372x10 ⁻⁶
91	72780	73601	6.6370x10 ⁻⁶
121	94679	94480	8.5117x10 ⁻⁶
151	117016	116837	10.5258x10 ⁻⁶
181	138735	138556	12.4825x10 ⁻⁶
211	160495	160316	14.4429x10 ⁻⁶
241	182220	182041	16.4001x10 ⁻⁶

TABLE 11

CYCLOHEXANE-POLYETHYLENE SYSTEM

T=25.0°C

B=17.7 cpm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
14	4646531	4646354
29	4622754	4622577
44	4639655	4639578
59	4639642	4639465
74	4584687	4584510
89	4562181	4562004
104	4549299	4549122
119	4533317	4533640

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
15	4477	4809	0.3374x10 ⁻⁶
30	10074	9897	0.3916x10 ⁻⁶
45	15733	15556	1.4015x10 ⁻⁶
60	20186	20009	1.8026x10 ⁻⁶
75	26730	26603	2.3967x10 ⁻⁶
90	32646	32469	2.9251x10 ⁻⁶
105	38102	37925	3.4167x10 ⁻⁶
120	43590	43413	3.9111x10 ⁻⁶

T=29.6°C

B=17.7 cpm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
15	4745063	4744836
30	4636152	4635975
45	4695745	4695563
60	4703546	4703369
76	4709194	4709017
90	4705095	4704918
105	4634224	4684047
120	4679775	4679593

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
16	9412	9235	0.3320x10 ⁻⁶
31	16945	16768	1.5106x10 ⁻⁶
46	26230	26053	2.3471x10 ⁻⁶
61	34045	33868	3.0512x10 ⁻⁶
77	42918	42741	3.8505x10 ⁻⁶
91	50959	50782	4.5750x10 ⁻⁶
106	58903	58726	5.2906x10 ⁻⁶
121	67358	67181	6.0523x10 ⁻⁶

TABLE 12
CYCLOHEXANE-POLYETHYLENE SYSTEM

T=39.3°C

B=18.0 cpm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
15	4846195	4846015
30	4849176	4848996
46	4840776	4840596
60	4831976	4831796
76	4800791	4800611
90	4797931	4797751
105	4779331	4779651
120	4760921	4760741

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
16	13393	13213	1.6413x10 ⁻⁶
31	35564	35324	3.1377x10 ⁻⁶
47	57143	56963	5.1322x10 ⁻⁶
61	74395	74715	6.7311x10 ⁻⁶
77	95221	95041	8.5622x10 ⁻⁶
91	114094	113914	10.2625x10 ⁻⁶
106	113106	112926	11.9753x10 ⁻⁶
121	150753	150573	13.5656x10 ⁻⁶

T=35.0°C

B=17.7 cpm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
23	4771022	4770345
44	4753763	4753536
59	4321997	4821820
74	4743543	4743366
89	4754941	4754764
104	4741399	4741222
119	4739736	4739609
134	4725760	4725583

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
30	24927	24750	2.2297x10 ⁻⁶
45	37778	37601	3.3875x10 ⁻⁶
60	50639	50462	4.5461x10 ⁻⁶
75	64411	64234	5.7869x10 ⁻⁶
90	76735	76558	6.8971x10 ⁻⁶
105	89551	89374	8.0517x10 ⁻⁶
120	104530	104353	9.4012x10 ⁻⁶
135	115857	115680	10.4217x10 ⁻⁶

TABLE 13

TETRAHYDRONAPHTHALENE-POLYETHYLENE SYSTEM

T=25.2°C

B=13.0 cpm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
16	3768434	3768254
30	3711302	3711122
60	3713776	3713596
86	3656005	3655825
120	3721747	3721567
150	3633323	3633143
130	3654233	3654103
210	3727145	3726965

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
17	1912	1732	1.5605x10 ⁻⁷
31	3093	2913	2.6283x10 ⁻⁷
61	5936	5756	5.1356x10 ⁻⁷
87	3007	7327	7.0514x10 ⁻⁷
121	11026	10346	9.7712x10 ⁻⁷
151	13305	13625	12.2748x10 ⁻⁷
131	16639	16159	14.5576x10 ⁻⁷
211	19039	13859	16.9901x10 ⁻⁷

T=30.0°C

B=13.0 cpm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
30	4663337	4663657
60	4648755	4648575
90	4597286	4597106
120	4521734	4521554
150	4537307	4537127
130	4465923	4465743
210	4437603	4437423
240	4400343	4400163

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
31	7450	7270	6.5496x10 ⁻⁷
61	11765	11585	10.4369x10 ⁻⁷
91	16264	16084	14.4901x10 ⁻⁷
121	21499	21319	19.2063x10 ⁻⁷
151	26460	26280	23.6757x10 ⁻⁷
181	31592	31412	28.2991x10 ⁻⁷
211	36661	36481	32.8654x10 ⁻⁷
241	42031	41851	37.2241x10 ⁻⁷

TABLE 14

TETRAHYDRONAPHTHALENE-POLYETHYLENE SYSTEM

T=34.8°C

B=18.0 cm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
30	4555459	4555279
60	4506901	4506721
89	4478569	4473381
123	4424377	4424697
151	4311427	4311247
180	4309867	4309687
210	4270942	4200762
240	4259431	4259251

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
31	9193	9013	0.3120x10 ⁻⁶
61	16079	15399	1.4323x10 ⁻⁶
90	21629	21449	1.9323x10 ⁻⁶
124	29577	29397	2.6434x10 ⁻⁶
152	25731	35601	3.2073x10 ⁻⁶
181	42231	42201	3.3019x10 ⁻⁶
211	43743	43563	4.3755x10 ⁻⁶
241	55416	55236	4.9762x10 ⁻⁶

T=39.3°C

B=18.0 cm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
30	4392107	4391927
60	4327012	4326832
90	4319386	4319706
120	4296343	4296663
150	4195205	4195025
180	4154219	4154039
210	4110937	4110817
240	4099937	4099817

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
31	11396	11216	1.0104x10 ⁻⁶
61	19332	19152	1.7254x10 ⁻⁶
91	28634	28454	2.5634x10 ⁻⁶
121	37433	37303	3.3606x10 ⁻⁶
151	46318	46638	4.2016x10 ⁻⁶
181	55916	55736	5.0213x10 ⁻⁶
211	65123	64943	5.8507x10 ⁻⁶
241	74338	74158	6.6809x10 ⁻⁶

TABLE 15

DECALYDRONAPHTHALENE-POLYETHYLENE SYSTEM

T=25.2°C

B=17.7 cpm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
30	4619966	4619789
57	4558019	4557342
90	4432197	4432020
120	4473915	4473733
150	4399337	4399710
180	4323701	4323524
210	4298703	4298526
240	4253701	4253524

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
31	3328	3751	3.3793x10 ⁻⁷
53	7410	7233	6.5162x10 ⁻⁷
91	11674	11497	10.3577x10 ⁻⁷
121	16062	15335	14.3108x10 ⁻⁷
151	20365	20138	18.1374x10 ⁻⁷
181	24496	24319	21.9090x10 ⁻⁷
211	28617	28440	25.6217x10 ⁻⁷
241	32701	32524	29.3007x10 ⁻⁷

T=30.0°C

B=17.7 cpm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
31	4632410	4632233
62	4569601	4569424
90	4551074	4550827
125	4467364	4467187
165	4438545	4438368
180	4429660	4429483
210	4403720	4403543
240	4336792	4336615

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
33	3037	7260	0.7081x10 ⁻⁶
63	13546	13369	1.2044x10 ⁻⁶
91	19636	19509	1.7576x10 ⁻⁶
126	26919	26742	2.4092x10 ⁻⁶
166	35732	35555	3.2032x10 ⁻⁶
181	39029	38852	3.5002x10 ⁻⁶
211	45487	45310	4.0820x10 ⁻⁶
241	52071	51894	4.6751x10 ⁻⁶

TABLE 16

DECAHYDRONAPHTHALENE-POLYETHYLENE SYSTEM

T=35.0°C

B=17.6 cpm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
30	4556315	4556139
60	4522526	4522410
89	4474344	4474163
124	4372236	4372060
150	4255617	4255441
180	4280623	4280507
210	4239763	4239537
240	4220942	4220766

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
31	11335	11679	1.0522x10 ⁻⁶
61	21202	21026	1.8942x10 ⁻⁶
90	30442	30263	2.7267x10 ⁻⁶
125	41934	41303	3.7665x10 ⁻⁶
151	50570	50394	4.5409x10 ⁻⁶
181	60614	60433	5.4449x10 ⁻⁶
211	70473	70297	6.3331x10 ⁻⁶
241	80339	80213	7.2264x10 ⁻⁶

T=39.7°C

B=17.6 cpm

Time min.	Ca ⁺ counts/10 min.	Ca ⁻ counts/10 min.
30	4034134	4034003
60	4045692	4045516
90	4004637	4004511
120	3950331	3950205
148	3294537	3294361
180	3236901	3236723
202	3722655	3722479
240	3750177	3750001

Time min.	C _b ⁺ counts/10 min.	C _b ⁻ counts/10 min.	C _B millicuries/cc.
31	15631	15455	1.3923x10 ⁻⁶
61	28717	28541	2.5713x10 ⁻⁶
91	41916	41740	3.7604x10 ⁻⁶
121	57403	57232	5.1560x10 ⁻⁶
149	67141	66965	6.0329x10 ⁻⁶
181	81629	81453	7.3381x10 ⁻⁶
203	91219	91043	8.2021x10 ⁻⁶
241	108597	108421	9.7677x10 ⁻⁶

TABLE 17

SYSTEM	TEMPERATURE °C	SPECTROMETER EFFICIENCY %	EVAPORATION RATE cc./hr.
Toluene- Polyethylene	25.0	39.57	0.302
	30.0	39.43	0.415
	35.0	39.54	0.563
	40.0	39.17	0.746
Chlorobenzene- Polyethylene	25.3	39.66	0.200
	30.0	39.47	0.323
	35.0	39.13	0.507
	39.3	39.14	0.691
Mesitylene- Polyethylene	25.4	39.13	0.150
	30.0	39.32	0.200
	35.0	39.30	0.246
	40.0	39.12	0.497
Cumene- Polyethylene	25.4	39.25	0.200
	30.0	39.13	0.312
	35.4	39.20	0.492
	39.3	39.13	0.700
Ethylbenzene- Polyethylene	25.0	38.65	0.375
	30.0	38.33	0.500
	35.4	38.60	0.637
	40.0	38.73	0.759
Cyclohexane- Polyethylene	25.0	39.09	1.300
	29.6	38.95	1.800
	35.0	39.23	2.440
	39.3	38.96	3.340
Tetrahydronaphthalene- Polyethylene	25.2	39.24	0.071
	30.0	38.31	0.038
	34.3	38.53	0.110
	39.3	38.33	0.132
Decahydronaphthalene- Polyethylene	25.2	39.10	0.138
	30.0	38.93	0.250
	35.0	38.66	0.294
	39.7	39.07	0.332

TABLE 18

SYSTEM	TEMPERATURE	AVERAGE	J x10 ⁸	Ds x10 ⁷
	°C	VOLUME cc.	mc./sec.	cm. ² /sec.
Toluene- Polyethylene	25.0	117.575	4.4101	1.3401
	30.0	117.260	7.7755	2.3839
	35.0	116.945	10.6900	3.2184
	40.0	116.930	15.3900	4.7989
Chlorobenzene- Polyethylene	25.3	117.300	1.9955	1.2104
	30.0	117.521	17.5450	2.0864
	35.0	117.107	4.5993	2.3502
	39.3	117.116	7.0474	4.1231
Mesitylene- Polyethylene	25.4	118.200	3.9755	1.2501
	30.0	117.300	5.5624	1.7551
	35.0	118.008	3.6340	2.6735
	40.0	117.130	12.4016	3.3334
Cumene- Polyethylene	25.4	117.300	2.5330	0.3009
	30.0	117.205	3.3712	1.2116
	35.4	117.143	6.3957	1.9794
	39.3	116.375	9.5304	2.9192
Ethylbenzene- Polyethylene	25.0	117.404	4.3794	1.5631
	30.0	117.125	6.9733	2.2170
	35.4	116.314	9.4753	2.9960
	40.0	116.540	13.6934	3.9475
Cyclohexane- Polyethylene	25.0	116.765	6.5309	2.0353
	29.6	116.270	9.6343	3.0130
	35.0	114.395	14.9400	4.6463
	39.3	113.930	21.5641	6.6031
Tetrahydronaphthalene- Polyethylene	25.2	113.033	1.5653	0.5035
	30.0	117.730	2.9213	0.3913
	34.3	113.002	3.9000	1.1919
	39.8	117.670	5.3992	1.3242
Decahydronaphthalene- Polyethylene	25.2	117.327	2.4241	0.7786
	30.0	117.375	3.3143	1.2387
	35.0	117.265	5.7396	1.3356
	39.7	117.503	7.3106	2.3379

TABLE 19

SYSTEM	ΔE cal./g. mole	D_0 cm. ² /sec.
Toluene- Polyethylene	15,772.35	4.39226×10^4
Chlorobenzene- Polyethylene	16,213.53	9.12629×10^4
Mesitylene- Polyethylene	14,253.56	3.40393×10^3
Cumene- Polyethylene	16,675.40	1.22250×10^5
Ethylbenzene- Polyethylene	11,414.84	3.65372×10^1
Cyclohexane- Polyethylene	14,753.32	1.32904×10^4
Tetrahydronaphthalene- Polyethylene	12,166.11	6.72325×10^1
Decahydronaphthalene- Polyethylene	16,525.79	1.01134×10^5

TABLE 20

SYSTEM

Toluene- Polyethylene	$D_s = 4.89326 \times 10^4 \exp(-15,772.35/RT)$
Chlorobenzene- Polyethylene	$D_s = 9.12629 \times 10^4 \exp(-16,218.53/RT)$
Mesitylene- Polyethylene	$D_s = 3.40393 \times 10^3 \exp(-14,253.56/RT)$
Cumene- Polyethylene	$D_s = 1.29250 \times 10^5 \exp(-16,675.40/RT)$
Ethylbenzene- Polyethylene	$D_s = 3.65379 \times 10^1 \exp(-11,414.34/RT)$
Cyclohexane- Polyethylene	$D_s = 1.32904 \times 10^4 \exp(-14,753.32/RT)$
Tetrahydronaphthalene- Polyethylene	$D_s = 6.72825 \times 10^1 \exp(-12,166.11/RT)$
Decahydronaphthalene- Polyethylene	$D_s = 1.01134 \times 10^5 \exp(-16,535.79/RT)$

TABLE 21

SWELLING SOLVENT	MOLECULAR* VOLUME cc./g. mole	ΔE cal./g. mole	D_0 cm. ² /sec.
Ethylbenzene	149.4	11,414.84	3.65872×10^1
Tetrahydronaphthalene	162.4	12,166.11	6.72325×10^1
Mesitylene	162.6	14,253.56	3.40393×10^3
Cyclohexane	118.2	14,753.32	1.32904×10^4
Toluene	118.2	15,772.35	4.39326×10^4
Chlorobenzene	92.3	16,218.53	9.12629×10^4
Decahydronaphthalene	134.6	16,535.79	1.01134×10^5
Cumene	163.3	16,675.40	1.29250×10^5

* calculated by Lebas Equation (13)

FIGURE 8

C_B VERSUS TIME
FOR THE
TOLUENE - POLYETHYLENE SYSTEM

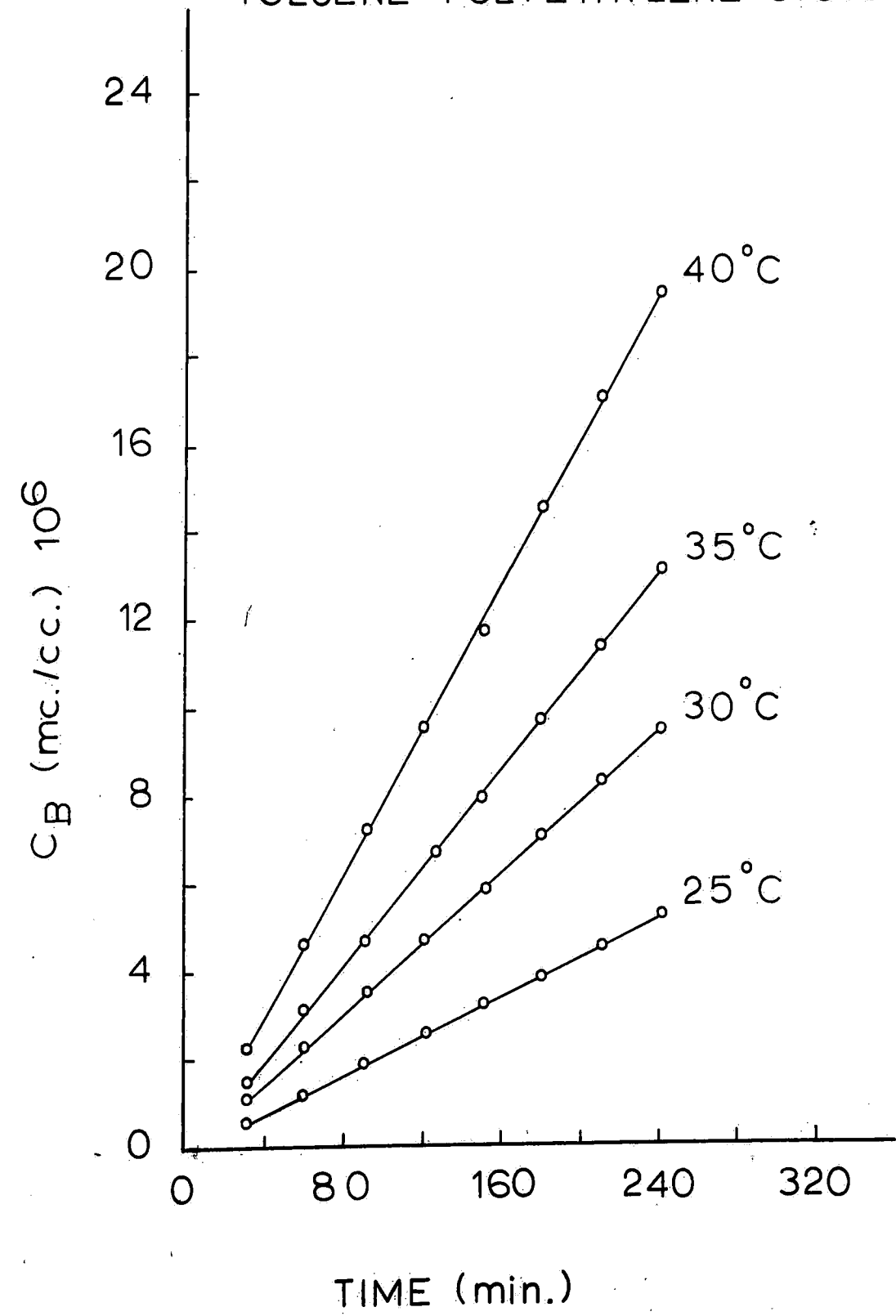


FIGURE 9

LOG₁₀(Ds) VERSUS 1/T

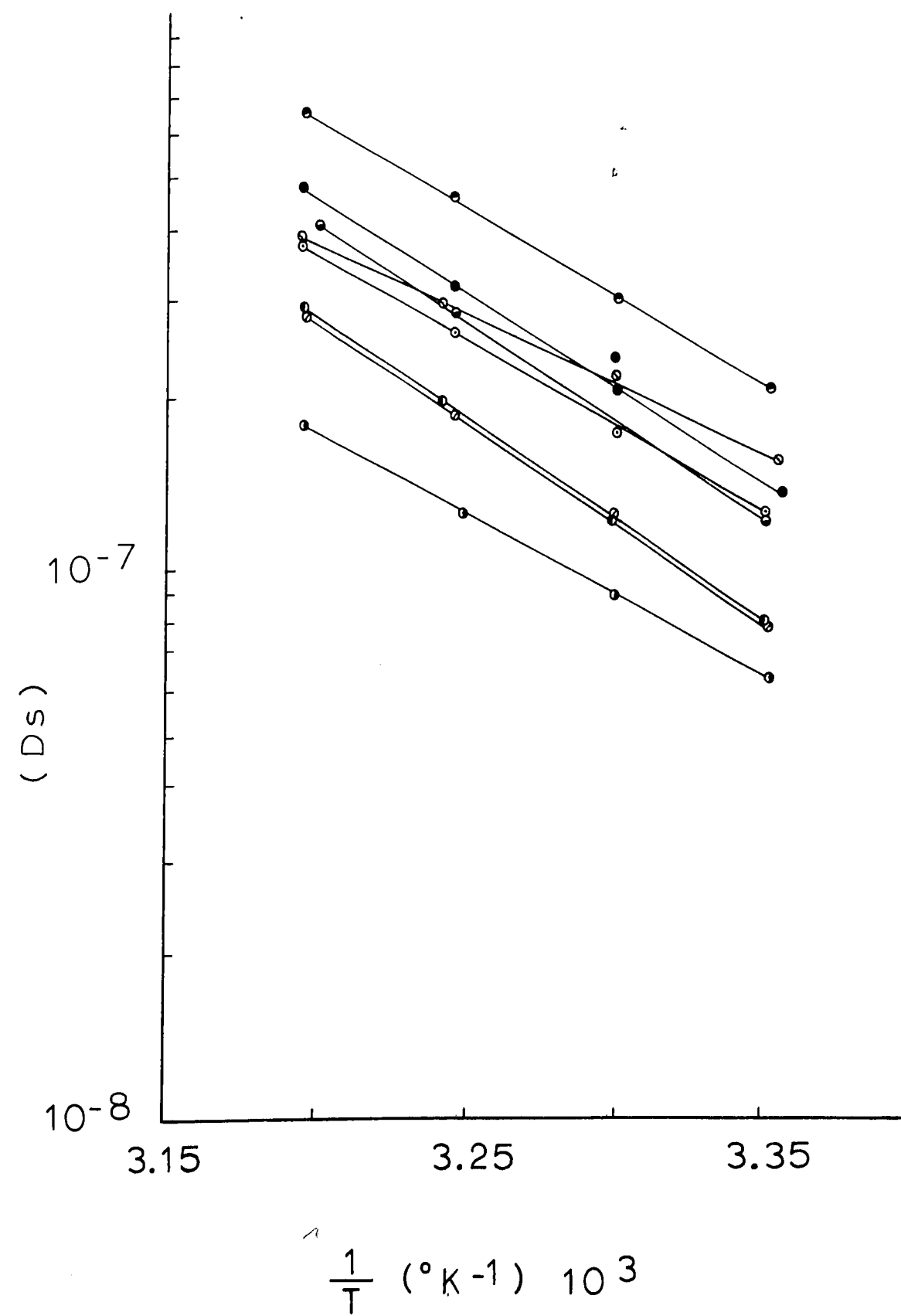
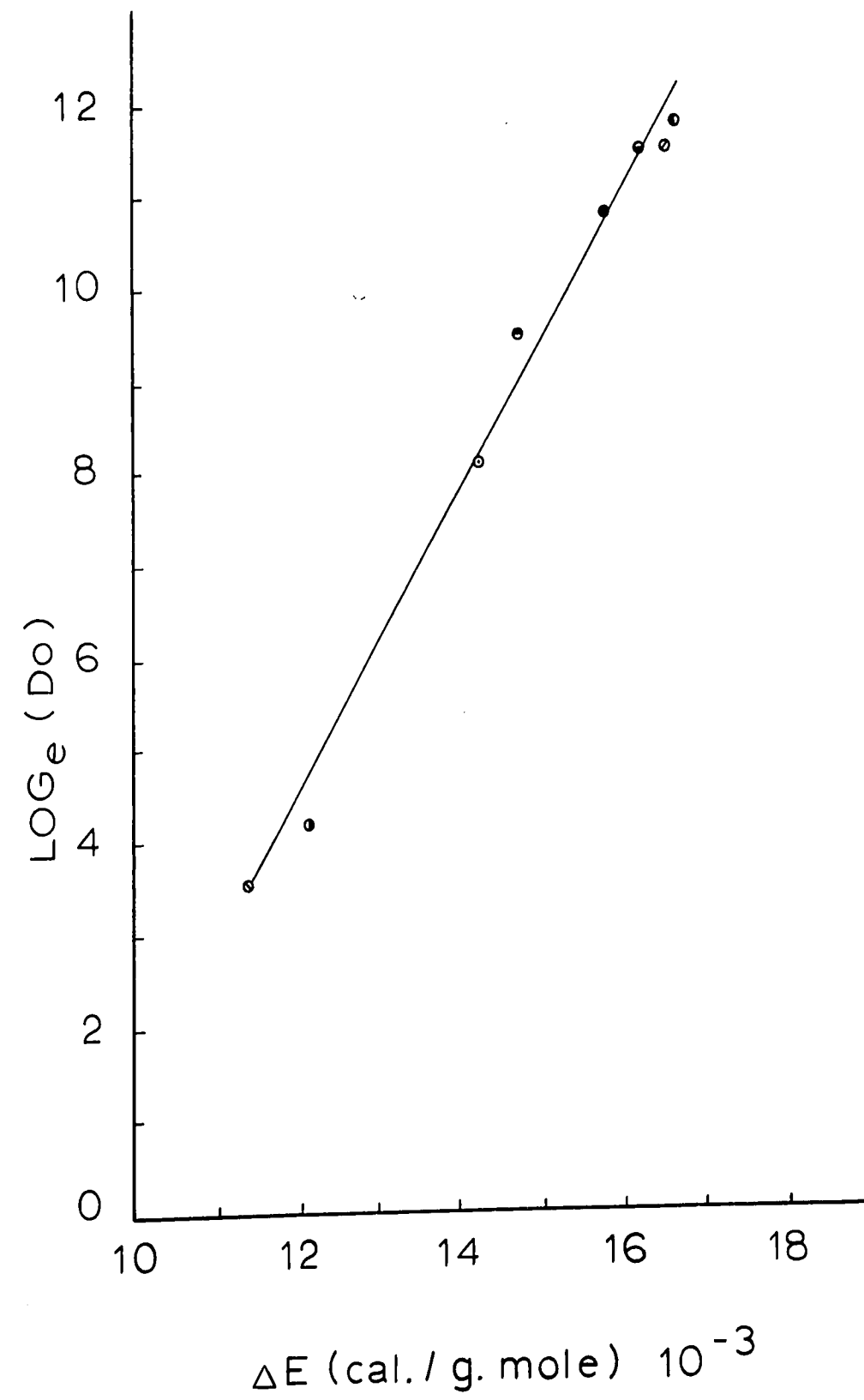


FIGURE 10

$\text{LOG}_e(D_0)$ VERSUS ΔE



SYMBOL NOTATION

Symbol	System
•	Toluene-Polyethylene
◦	Chlorobenzene-Polyethylene
◦	Cyclohexane-Polyethylene
◦	Tetrahydronaphthalene-Polyethylene
◦	Cumene-Polyethylene
◦	Decahydronaphthalene-Polyethylene
◦	Ethylbenzene-Polyethylene
◦	Mesitylene-Polyethylene

ERROR ANALYSIS

This section will present an analysis of the accuracy of the values calculated for the permeability and for the activation energy for the permeation process.

The errors encountered can be divided into the following:

1. counting error
2. sampling error
3. error resulting from use of average volume
4. curve fitting error

The error encountered in the counting of any sample is expressed as:

$$\% \text{ Error} = \frac{1}{(t)^{0.5}} \frac{(\text{count rate})^{0.5}}{\text{count rate}} (100)$$

The error is greatest for the least count rate. The maximum per cent error in the present work for the "cold" chamber tracer concentration is 0.76 for tetrahydronaphthalene and 0.32 for mesitylene; this being for the initial sample with the per cent error decreasing sharply as further samples were taken having a higher count rate. The error encountered for the "hot" chamber tracer concentration is about 0.05% for all the systems.

The sampling error is that for the tolerance of the measuring pipette. The one-half milliliter pipettes used had a maximum tolerance of ± 0.003 milliliters, this yielding a per cent error of ± 0.6 .

The use of the average volume over the extremes of the "cold" chamber tracer concentration versus time plot

compared to that using the "true" cold chamber volume at each sampling time in order to calculate the rate of permeation is felt by the researcher to be justified. The chamber volumes are not actually measured at each sampling time, such that, the "true" chamber volume is not really a known quantity. It is calculated from the results of a previous evaporation rate measurement, so that, any change in stirring rate can effect the "true" volume. Also added to this is the error of the sampling pipettes which again can cause a difference in the volume computed to the "true" volume in each chamber.

An example of the error estimation for the toluene-polyethylene system at 25.0°C follows. A least squares analysis is made on the plot of C_B versus time for both the case of an average volume assumption and also for the case employing the "true" volume at each sampling time to determine the permeation rate.

An outline of the least squares analysis for the plot of "cold" chamber tracer concentration in millicuries per cubic centimeters versus time in minutes employing the use of an average volume to calculate the rate of permeation follows.

$C_B (10^6)$	(mc./cc.)	0.5832	1.2186	1.8995	2.5524
time	(minutes)	32	61	91	122
$C_B (10^6)$	(mc./cc.)	3.2011	3.8291	4.6239	5.3093
time	(minutes)	152	182	212	242

Let the equation of the line be:

$$C_B = a + bt$$

The form of the residual equation is then:

$$v_n = a + bt_n - C_{Bn}$$

The residuals equations are then:

$$v_1 = a + 32b - 0.5832(10^{-6})$$

$$v_2 = a + 61b - 1.2186(10^{-6})$$

$$v_3 = a + 91b - 1.8995(10^{-6})$$

$$v_4 = a + 122b - 2.5524(10^{-6})$$

$$v_5 = a + 152b - 3.2011(10^{-6})$$

$$v_6 = a + 182b - 3.8291(10^{-6})$$

$$v_7 = a + 212b - 4.6239(10^{-6})$$

$$v_8 = a + 242b - 5.3093(10^{-6})$$

Multiplying the right-hand members of each residual equation by the coefficient of the first unknown in that member, adding the products obtained, and equating their sum to zero, it is found:

$$8a + 1094b - 23.2171(10^{-6}) = 0$$

Multiplying the right-hand members of each residual equation by the coefficient of the second unknown in that member, adding the products obtained, and equating their sum to zero, it is found:

$$1094a + 187646b - 4025.8251(10^{-6}) = 0$$

The normal equations are then:

$$8a + 1094b = 23.2171(10^{-6})$$

$$1094a + 187646b = 4025.8251(10^{-6})$$

Solving by determinates it is found that:

$$a = \frac{\begin{vmatrix} 23.2171(10^{-6}) & 1094 \\ 4025.8251(10^{-6}) & 187646 \end{vmatrix}}{\begin{vmatrix} 8 & 1094 \\ 1094 & 187646 \end{vmatrix}}$$

$$= -1.565944(10^{-7})$$

$$b = \frac{\begin{vmatrix} 8 & 23.2171(10^{-6}) \\ 1094 & 4025.8251(10^{-6}) \end{vmatrix}}{\begin{vmatrix} 8 & 1094 \\ 1094 & 187646 \end{vmatrix}}$$

$$= 2.236732(10^{-8})$$

The equation is then:

$$C_B = -1.565944(10^{-7}) + 2.236732(10^{-8})t$$

The slope of the C_B versus t plot would be:

$$\text{slope} = 2.2367(10^{-8}) \text{ mc./cc.}(min.)$$

and the rate of permeation is:

$$J = 2.6298(10^{-6}) \text{ mc./min.}$$

The following is an outline of the least squares analysis on the plot of "cold" chamber tracer activity in millicuries versus time in minutes employing the "true" volume at each sampling in order to calculate the rate of permeation.

C_B' (10^4) (mc.)	0.6989652	1.4525712	2.25185725	3.0092796
time (min.)	32	61	91	122
C_B' (10^4) (mc.)	3.75328975	4.4647306	5.36141205	6.1216229
time (min.)	152	182	212	242

Let the equation of the line be:

$$C_B' = a + bt$$

The form of the residual equation is then:

$$v_n = a + bt_n - C_B'_n$$

The residuals equations are then:

$$v_1 = a + 32b - 0.6989652(10^{-4})$$

$$v_2 = a + 61b - 1.4525712(10^{-4})$$

$$v_3 = a + 91b - 2.25185725(10^{-4})$$

$$v_4 = a + 122b - 3.0092796(10^{-4})$$

$$v_5 = a + 152b - 3.75328975(10^{-4})$$

$$v_6 = a + 182b - 4.4647306(10^{-4})$$

$$v_7 = a + 212b - 5.36141205(10^{-4})$$

$$v_8 = a + 242b - 6.1216229(10^{-4})$$

Applying the same conditions as before, the normal equations are found to be:

$$8a + 1094b = 27.1137(10^{-4})$$

$$1094a + 187646b = 4684.1579(10^{-4})$$

Solving the normal equations by determinates, it is found that:

$$a = \frac{\begin{vmatrix} 27.1137(10^{-4}) & 1094 \\ 4684.1579(10^{-4}) & 187646 \end{vmatrix}}{\begin{vmatrix} 8 & 1094 \\ 1094 & 187646 \end{vmatrix}}$$

$$= -1.205637(10^{-5})$$

$$b = \frac{\begin{vmatrix} 8 & 27.1137(10^{-4}) \\ 1094 & 4684.1579(10^{-4}) \end{vmatrix}}{\begin{vmatrix} 8 & 1094 \\ 1094 & 187646 \end{vmatrix}}$$

$$= 2.566563(10^{-6})$$

The equation is then:

$$C_{P'} = -1.2056(10^{-5}) + 2.5666(10^{-6})t$$

The slope of the $C_{P'}$ versus t plot would be:

$$\text{slope} = 2.5666(10^{-6}) \text{ mc./min.}$$

and the rate of permeation is:

$$J = 2.5666(10^{-6}) \text{ mc./min.}$$

A least squares analysis on the plot of the logarithm of the permeability versus reciprocal temperature for the toluene-polyethylene system follows. In this analysis the value of the permeability of the tracer at 30.0°C is not used since, as had been discussed before, due to some inherent property of the polymer the data near 30°C were inconsistent with the other data points. Employment of the value at 30.0°C would yield a value for D_0 about half that found by disregarding that point.

(D_s)	(10^7)	$(\text{cm.}^2/\text{sec.})$	1.3401	3.2184	4.7989
$1/T$	(10^3)	(K^{-1})	3.3539	3.2451	3.1932

Let the equation of the line be:

$$\log_{10}(D_s) = \log_{10}(D_0) - \log_{10}e (1/RT)(\Delta E)$$

The form of the residual equation is then:

$$v_n = \log_{10}(D_0) - \log_{10}e (1/RT_n)(\Delta E) - \log_{10}((D_s)_n)$$

The residuals equations are then:

$$v_1 = \log_{10}(D_0) - 0.7330549(10^{-3})(\Delta E) + 6.872867$$

$$v_2 = \log_{10}(D_0) - 0.7092747(10^{-3})(\Delta E) + 6.492358$$

$$v_3 = \log_{10}(D_0) - 0.6979310(10^{-3})(\Delta E) + 6.318859$$

Applying the same procedure as before, the normal equations are found to be:

$$3 \log_{10}(D_0) - 2.140261(10^{-3})(\Delta E) = -19.68408$$

$$-2.140261(10^{-3}) \log_{10}(D_0) + 1.527548(10^{-6})(\Delta E) = 14.053182(10^{-3})$$

Solving the normal equations by determinates it is found that:

$$\log_{10}(D_0) = \frac{\begin{vmatrix} -19.68408 & -2.140261(10^{-3}) \\ 14.05318(10^{-3}) & 1.527548(10^{-6}) \end{vmatrix}}{\begin{vmatrix} 3 & -2.140261(10^{-3}) \\ -2.140261(10^{-3}) & 1.527548(10^{-6}) \end{vmatrix}}$$

$$= 4.72061$$

$$(D_0) = 5.25544(10^4)$$

$$\Delta E = \frac{\begin{vmatrix} 3 & -19.68408 \\ -2.140261(10^{-3}) & 14.05318(10^{-3}) \end{vmatrix}}{\begin{vmatrix} 3 & -2.140261(10^{-3}) \\ -2.140261(10^{-3}) & 1.527548(10^{-6}) \end{vmatrix}}$$

$$= 15,816.61$$

The equation is then:

$$(D_s) = 5.25544(10^4) \exp(-15,816.61/RT)$$

The energy of activation for the permeation process

is:

$$\Delta E = 15,816.61 \text{ cal./g. mole}$$

and the value of the constant is:

$$(D_0) = 5.25544(10^4) \text{ cm.}^2/\text{sec.}$$

The following is a determination of the confidence interval for the least squares equation for the plot of the "cold" chamber activity in millicuries versus time in minutes.

The least squares equation was found to be:

$$C_B' = -1.2056(10^{-5}) + 2.5666(10^{-6})t$$

Let y_i represent the value of C_B' obtained from the data and \bar{y}_i represent the value of C_B' calculated from the above least squares equation at the appropriate times.

Also let x_i represent the values for the time of sampling in minutes.

y_i	(10^4)	0.6989652	1.4525712	2.25185725	3.0092796
\bar{y}_i	(10^4)	0.7007365	1.4450397	2.2150086	3.0106432
$(y_i - \bar{y}_i)$	(10^6)	-0.17713	0.75315	3.68486	-0.13636
$(y_i - \bar{y}_i)^2$	(10^{12})	0.0313750	0.5672349	13.5781932	0.0185940
y_i	(10^4)	3.75328975	4.4647306	5.36141205	6.1216229
\bar{y}_i	(10^4)	3.7806121	4.5505810	5.3205499	6.0905188
$(y_i - \bar{y}_i)$	(10^6)	-2.73223	-8.58504	4.08621	3.11041
$(y_i - \bar{y}_i)^2$	(10^{12})	7.4650808	73.7029118	16.6971122	9.6746504

$$\sum_{i=1}^n (y_i - \bar{y}_i)^2 = 121.7351523(10^{-12})$$

$$S_{y/x} = \sqrt{\frac{\sum_{i=1}^n (y_i - \bar{y}_i)^2}{n-2}}$$

$$= \sqrt{\frac{121.7351523(10^{-12})}{6}}$$

$$= 4.5043525(10^{-6})$$

$$\bar{x} = \frac{\sum_{i=1}^n x_i}{n} = \frac{1094}{8} = 136.75$$

x_i	32	61	91	122	
$(x_i - \bar{x})$	-104.75	-75.75	-45.75	-14.75	
$(x_i - \bar{x})^2$	(10^{-4})	1.0972562	0.5738062	0.2093062	0.0217562
x_i	152	182	212	242	
$(x_i - \bar{x})$	15.25	45.25	75.25	105.25	
$(x_i - \bar{x})^2$	(10^{-4})	0.0232562	0.2047562	0.5662562	1.1077562

$$\sum_{i=1}^n (x_i - \bar{x})^2 = 3.8041496(10^4)$$

Using confidence coefficient $(1-\alpha)$ of 0.95, so that the value of α is 0.05.

The degrees of freedom are $(n-2)$ or 6.

Therefore, from Student's t -distribution it is found that the value of $t_{\alpha/2; n-2}$ is 1.943.

The confidence interval for $b = 2.5666(10^{-6})$ is:

$$b \pm t_{\alpha/2; n-2} \frac{S_y/x}{\sqrt{\sum_{i=1}^n (x_i - \bar{x})^2}}$$

$$b \pm 1.943 (4.5043525(10^{-6})) (3.8041496(10^4))^{-0.5}$$

$$b \pm 0.0448721(10^{-6})$$

Therefore, the value of the slope, b , with a confidence of 95% is:

$$2.5666 \pm 0.0449 (10^{-6}) \text{ mc./min.}$$

or,

$$2.5666(10^{-6}) \pm 1.75\% \text{ mc./min.}$$

Correspondingly, the diffusion constant with a confidence of 95% is:

$$1.3401(10^{-7}) \pm 1.75\% \text{ cm.}^2/\text{sec.}$$

The following is a determination of the confidence interval for the least squares equation for the plot of the logarithm of the permeability versus reciprocal temperature for the toluene-polyethylene system.

The least squares equation was found to be:

$$\log_{10}(D_s) = 4.72061 - (1/RT) \log_{10}e (15,816.61)$$

Let y_i represent the value of $\log_{10}(D_s)$ obtained from the data and \bar{y}_i represent the value of $\log_{10}(D_s)$ calculated from the above least squares equation at the appropriate temperatures. Also let x_i represent the value of $\log_{10}e (1/RT)$ at the respective temperatures.

y_i	-6.87287	-6.49236	-6.31886
\bar{y}_i	-6.87382	-6.49770	-6.31828
$(y_i - \bar{y}_i) (10^4)$	9.5	53.4	-5.8
$(y_i - \bar{y}_i)^2 (10^8)$	90.25	2851.56	33.64

$$\sum_{i=1}^n (y_i - \bar{y}_i)^2 = 2975.45(10^{-8})$$

$$S_{y/x} = \sqrt{\frac{\sum_{i=1}^n (y_i - \bar{y}_i)^2}{n-2}}$$

$$= \sqrt{2975.45(10^{-8})}$$

$$= 5.4548(10^{-3})$$

$$\bar{x} = \frac{\sum_{i=1}^n x_i}{n} = \frac{2.140261(10^{-3})}{3} = 0.713420(10^{-3})$$

x_i	0.733055	0.709275	0.697931
$(x_i - \bar{x}) (10^6)$	19.635	-4.145	-15.489
$(x_i - \bar{x})^2 (10^{12})$	385.5332	17.1810	239.9091

$$\sum_{i=1}^n (x_i - \bar{x})^2 = 6.426233(10^{-10})$$

Using confidence coefficient $(1-\alpha)$ of 0.90, so that the value of α is 0.10.

The degrees of freedom are $(n-2)$ or 1.

Therefore, from Student's t-distribution it is found that the value of $t_{\alpha/2; n-2}$ is 3.078.

The confidence interval for $b = 15,816.61$ is:

$$b \pm t_{\alpha/2; n-2} \frac{S_{y/x}}{\sqrt{\sum_{i=1}^n (x_i - \bar{x})^2}}$$

$$b \pm 3.078 (5.4548(10^{-3})) (6.426233(10^{-10}))^{-0.5}$$

$$b \pm 662.32$$

Therefore, the value of the slope, ΔE , with a confidence of 90% is:

$$15,816.61 \pm 662.32 \text{ cal./g. mole}$$

or,

$$15,816.61 \pm 4.19\% \text{ cal./g. mole}$$

The confidence interval for $a = 4.72061$ is:

$$a \pm t_{\alpha/2; n-2} (S_{y/x}) \sqrt{\frac{1}{n} + \frac{\bar{x}^2}{\sum_{i=1}^n (x_i - \bar{x})^2}}$$

$$a \pm 3.078 (5.4548(10^{-3})) \sqrt{\frac{1}{3} + \frac{(0.713420(10^{-3}))^2}{6.426233(10^{-10})}}$$

$$a \pm 0.048234$$

Therefore, the value of $\log_{10}(D_0)$ with a confidence of 90% is:

$$4.72061 \pm 0.04823$$

or,

$$4.72061 \pm 1.0\%$$

Finally a value of D_0 with a confidence of 90% is found to be:

$$5.25544(10^4) \pm 0.61727(10^4)$$

$$-0.55234(10^4)$$

or,

$$5.25544(10^4) \pm 11.7\%$$

$$-10.5\%$$

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