Determination of Diffusion Coefficients and Activation Energy of Selected Organic Liquids using Reversed-Flow Gas Chromatographic Technique
(Penentuan Pemalar Penyebaran dan Tenaga Pengaktifan bagi Cecair Organik Terpilih Menggunakan Teknik Kromatografi Gas Aliran-Berbalik)

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ABSTRACT

Evaporation of vaporize organic liquid has ecological consequences when the compounds are introduced into both freshwater and marine environments through industrial effluents, or introduced directly into the air from industrial unit processes such as bioreactors and cooling towers. In such cases, a rapid and simple method are needed to measure physicochemical properties of the organic liquids. The Reversed-Flow Gas Chromatography (RF-GC) sampling technique is an easy, fast and accurate procedure. It was used to measure the diffusion coefficients of vapors from liquid into a carrier gas and at the same time to determine the rate coefficients for the evaporation of the respective liquid. The mathematical expression describing the elution curves of the samples peaks was derived and used to calculate the respective parameters for the selected liquid pollutants selected such as methanol, ethanol, 1-propanol, 1-butanol, n-pentane, n-hexane, n-heptane and n-hexadecane, evaporating into the carrier gas of nitrogen. The values of diffusion coefficients found were compared with those calculated theoretically or reported in the literature. The values of evaporation rate were used to determine the activation energy of respective samples using Arrhenius equation. An interesting finding of this work is by using an alternative mathematical analysis based on equilibrium at the liquid-gas interphase, the comparison leads to profound agreement between theoretical values of diffusion coefficients and experimental evidence.

Keywords: Diffusion coefficient; organic liquid; reversed-flow gas chromatography

ABSTRAK

Penyejatan cecair organik yang meruap adalah suatu ekologi berbahaya yang membawa maut dengan bahan dialirkan ke kawasan aliran air tawar dan air masin melalui bahan buangan industri, atau dialirkan secara terus ke udara daripada pemprosesan unit industri seperti bioreaktor dan menara penyejuk. Dalam kes yang demikian, suatu kaedah yang cepat dan ringkas diperlukan untuk mengkaji sifat fiziko-kimia cecair-cecair organik. Teknik pengsampelan Kromatografi Gas Aliran-berbalik adalah kaedah yang mudah, cepat dan tepat. Ia telah digunakan untuk mengukur pemalar penyebaran wap daripada cecair ke dalam gas pembawa dan dalam masa yang sama menentukan pemalar kadar bagi proses penyejatan bagi cecair yang terlibat. Ungkapan matematik yang menjelaskan puncak lengkungan elusi sampel yang diterbitkan dan digunakan untuk menaakul parameter berkaitan bagi cecair pencemar yang terpilih seperti metanol, etanol, 1-propanol, 1-butanol, n-pentana, n-heksana, n-heptana dan n-heksadesana yang tersejat ke dalam gas pembawa nitrogen. Nilai pemalar penyebaran eksperimen kemudiannya dibandingkan dengan nilai-nilai teori yang didapati secara kaedah pengiraan atau nilai-nilai yang telah dilaporkan di dalam jurnal, dan seterusnya nilai-nilai pemalar penyejatan ini digunakan bagi menentukan tenaga pengaktifan bagi setiap sampel menggunakan persamaan Arrhenius. Suatu penemuan yang menarik dalam penyelidikan ini ialah dengan menggunakan analisis matematik alternatif berdasarkan keseimbangan di fasa perantara cecair-gas, nilai-nilai teori pemalar penyebaran adalah selari dengan nilai eksperimen.

Kata kunci: Cecair organik; kromatografi gas aliran berbalik; pemalar penyebaran

INTRODUCTION

The world is facing environment pollution for many years. Industrial wastes and even household leachates are dissipated without monitoring. The liquid waste is evaporated off to the environment and affects human life and ecosystem. Evaporation of liquid is a type of vaporization and happens only on the surface of a liquid. The other type of vaporization is boiling, which instead occurs on the entire mass of the liquid system. It is also part of the water cycle. Evaporation is also a type of phase transition; it is the process by which molecules in a liquid state spontaneously become gaseous. The air–liquid interface is a region of intense gradients where evaporation occurs and imposes interesting constraints on the lives and productivity of microorganisms. Thus, research in this area requires working at scales far smaller than those normally associated with the bulk processes on either side of the interface, and requires new experimental and theoretical approaches.

The Reversed-Flow Gas Chromatography (RF-GC) (Gavril 2010) technique has been used for various physicochemical measurements, such as of rate coefficients and diffusion coefficients for the evaporation of pure liquids (Khalisanni 2011), mass transfer coefficients regarding the evaporation of liquids (Rashid et al. 2004), activity coefficients and diffusion coefficients in multicomponent liquid mixtures (Gavril et al. 2006) as well as for the estimation of the solubility and interaction parameters in binary liquid mixtures (Gavril et al. 2004).

RF-GC system comprises of modified commercial gas chromatography, detector, six port valves and a simple cell placed in chromatographic oven. The carrier gas such as nitrogen is used as mobile phase while the sample performed as stationary phase. The type of detector depends to the type of sample.

The technique is effortless and not limited to environmental research. It can be applied in the medical and catalysis sectors as well as for research and development centres.

In the present work, the reversed-flow gas chromatography was used to measure simultaneously the rate coefficient for the evaporation of pure liquids and the diffusion coefficient of these vapors into a carrier gas passing over the surface of the liquids at a remote point (Figure 1). The liquids used were methanol, ethanol, 1-propanol, 1-butanol, n-pentane, n-hexane, n-heptane and n-hexadecane while the carrier gas used was nitrogen.

THEORY

The common chromatographic sampling equation, describing the elution curves which follow the carrier gas flow reversals is:

$$c = c_1(l', t_0 + t' + \tau)\, u(\tau) + c_2(l', t_0 + t' - \tau)[1 - u(\tau - t')] \times [u(\tau) - u(\tau - t_M')] + c_3(l', t_0 - t' + \tau)\, u\,(t_0 + \tau - t')\{u(t - t') [1 - u(\tau - t_M')] - u(\tau - t')[u(\tau) - u(\tau - t_M')]\},$$
(1)

where c is the concentration of vapor at the detector, c_1 $(l', \ldots)$, $c_2(l', \ldots)$, and $c_3(l', \ldots)$ are concentrations at the point $x = l'$ (Figure 1) for the times shown (t_0 is the total time from placing the liquid in column L to the last backward reversal of gas flow, t' = time interval of backward flow, τ is the $t - t_M$, t being the time from the last restoration of the carrier gas flow, and t_M the gas hold-up time of column section l; finally the various u's are unit step functions for the arguments shown in parentheses and t_M' is the gas hold-up time in the section l'.

For t' smaller than both t_M and t_M', each sample peak produced by two successive reversals is symmetrical and its maximum height h from the ending baseline is given by:

$$h \cong 2c(l', t_o),$$
(2)

where $c(l', t_0)$ is the vapor concentration at $x = l'$ and time t_0. The concentration of the liquid can be found from the diffusion equation in the column L (Figure 1):

$$\partial c_z / \partial t_0 = D \partial^2 c_z / \partial z^2,$$
(3)

where D is the diffusion coefficient of the vapor into the carrier gas. The solution of (3) is sought under the initial condition:
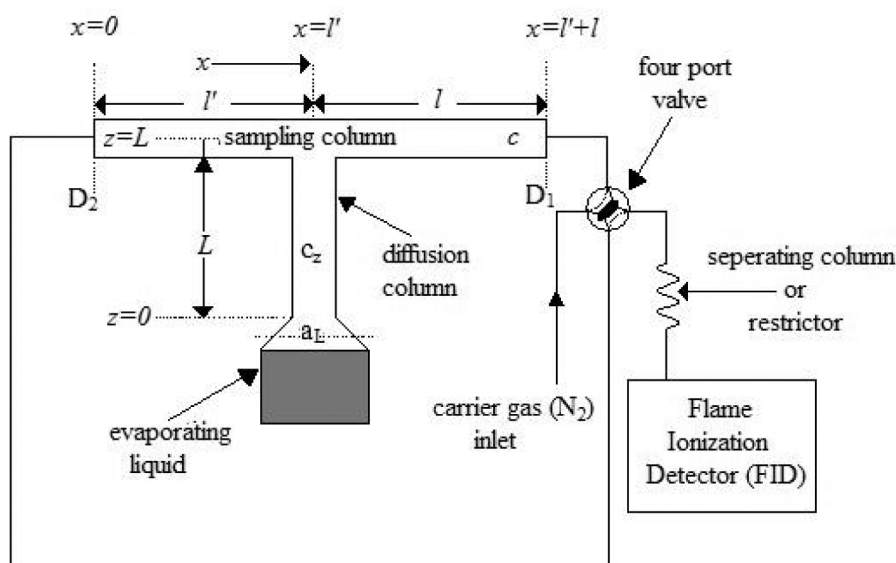
$$c_z(z, 0) = 0,$$
(4)

FIGURE 1. Schematic arrangement showing the instrumentation of RF-GC which the diffusion column L connected to the chromatographic column $l + l'$ and carrier gas flows from D_2 to D_1 or vice versa

the boundary conditions at $z = L$:

$$c_z(L,t_0)=c(l,t_0), \qquad (5)$$

$$-D(\partial c_z/\partial z)_{z=L} = vc(l',t_0), \qquad (6)$$

where v is the linear velocity of carrier gas, and the boundary condition at $z= 0$:

$$-D(\partial c_z/\partial z)_{z=0} = k_c(c_0-c_z(0)), \qquad (7)$$

where $c_z(0)$ is the actual concentration at the liquid interface at time t_0, c_0 the concentration of the vapor which would be in equilibrium with the bulk liquid phase, and k_c a rate coefficient for the evaporation process. Equation (7) expresses the equality of the diffusion flux for removal of vapors from the liquid surface and the evaporation flux due to departure of c_z at the surface from the equilibrium value c_0.

When the Laplace transform of (3) is taken with respect to t_0, a linear second-order differential equation results. It can be solved by using z Laplace transformation yielding:

$$C_z = C_z(0) \cosh qz + \frac{C_z{}'(0)}{q} \sinh qz, \qquad (8)$$

where

$$q = (p_0/D0)^{1/2}, \qquad (9)$$

and $C_z(0)$ and $C_z{}'(0)$ are the t_0 Laplace transform of $c_z(0)$ and $(\partial c_z/\partial z)_{z=0}$ respectively. If one combines (8) with the t_0 transforms of the boundary conditions (5), (6) and (7), the Laplace transform of $c(l',t_0)$, denoted as $C(l',p_0)$, is founds by:

$$C(l',p_0) = \frac{k_c c_0}{p_0} \frac{1}{(Dq+vk_c/Dq)\sinh qL+(v+kc)\cosh qL}. \qquad (10)$$

The application of inverse Laplace transformation of this equation to find $c(l',t_0)$ is difficult. Thus, it can be achieved by using certain approximations which are different for small or for long times. In the first case qL is large, allowing both $\sinh qL$ and $\cosh qL$ to be approximated by $\exp(qL)/2$. Then (10) becomes:

$$C(l',p_0) = \frac{k_c c_0}{p_0 Dq} \frac{2\exp(-qL)}{(1+k_c/Dq)(1+v/Dq)}. \qquad (11)$$

For high enough flow rates, the equation further reduces to:

$$C(l',p_0) = \frac{2k_c c_0}{vD} \frac{\exp(-qL)}{q(q+k_c/D)}. \qquad (12)$$

Taking now the inverse Laplace transform of this equation, one finds:

$$c(l',t_0) = \frac{2k_c c_0}{v} \exp\left[\frac{k_c L}{D} + \frac{k_c 2t_0}{D}\right] erfc\left[\frac{L}{2(Dt_0)^{1/2}} + (t_0)^{1/2}\right]. \qquad (13)$$

Finally, if one uses the relation erfc $x \cong \exp(-x^2)/(\tau\pi^{1/2})$, which is a good approximation for large values of x, equation (13) becomes:

$$c(l',t_0) = \frac{2k_c c_0}{v} \left[\frac{(D)}{\pi}\right]^{1/2} \exp\left[\frac{-L^2}{4Dt_0}\right]\left[\frac{L}{2t_0^{1/2}} + k_c t_0^{1/2}\right]^{-1}. \qquad (14)$$

Coming now to the other extreme, i.e. long time approximations, qL is small and the functions $\sinh qL$ and $\cosh qL$ of equation (10) can be expanded in Mc Laurin series, retaining only the first three terms in each of them. Then, from equation (10) one obtains:

$$C(l',p_0) = \frac{k_c c_0}{p_0} \frac{1}{(Dq+vk_c/Dq)qL+(v+kc)(1+q^2L^2/2)}, \qquad (15)$$

and by using equation (9) and rearranging this becomes:

$$C(l',p_0) = \frac{k_c c_0}{Lp_0} \frac{1}{p_0[1+(v+k_c)L/2D]+vk_c/D+(v+k_c)/L}. \qquad (16)$$

For high enough flow rates k_c can be neglected compared to v and 1 can be neglected in comparison with $vL/2D$. For instance, in a usual experimental situation it was calculated that $vL/2D = 420$. Adopting these approximations, equation (16) reduces, after some rearrangement, to:

$$C(l',p_0) = \frac{2k_c Dc_0}{vL^2 p_0} \frac{1}{p_0 + 2(k_c L+D)/L^2}. \qquad (17)$$

Finally, inverse Laplace transformation of this relation yields:

$$C(l',t_0) = \frac{k_c Dc_0}{v(k_c L+D)}\left\{1-\exp\left[-2(k_c L+D)t_0/L^2\right]\right\}, \qquad (18)$$

by considering maximum height h of the sample peaks in equation (2) and substituting in it the right hand side of Eq 18 for $c(l',t_0)$, one obtains h as an explicit function of time t_0. In order to linearize the resulting relation, an infinity value h_∞ for the peak height is required:

$$h_\infty = 2k_c Dc_0/[v(k_c L+D)]. \qquad (19)$$

Using this expression, we obtain:

$$\ln(h_\infty-h)=\ln h_\infty - [2(k_c L+D)/L^2]t_0. \qquad (20)$$

Thus, the long enough times, for which (18) was derived, a plot of $\ln(h_\infty - h)$ vs. t_0 is expected to be linear, and from the slope $-2(k_c L+D)/L^2$ a first approximate value of k_c can be calculated from the known value of L and a literature or theoretically calculated value of D. This value of k_c can now be used to plot small time data according to (14) which is substituted now for $c(l',t_0)$ in (2). After rearrangement logarithms are taken, and there results:

$$\ln[h(L/2t_0^{1/2} + k_c t_0^{1/2})] = \ln[4k_c c_0/v(D/\pi)^{1/2}] - (L^2/4D)(\,1/t_0).$$

(21)

Now, a plot of the left hand side of this relation vs $1/t_0$ will yield a first approximation of experimental value for D from the slope $-L^2/4D$ of this new linear plot.

MATERIALS AND METHODS

CHEMICALS

Methanol, ethanol, 1-propanol, 1-butanol, n-pentane, n-hexane, n-heptane and n-hexadecane from Merck-AR graded were used as evaporating liquids (stationary phase). Nitrogen of 99.99% purity from MOX (Malaysia) was used as the carrier gas (mobile phase).

APPARATUS

The experimental setup for the application of reversed-flow gas chromatography is shown in Figure 1. A conventional gas chromatograph with a flame ionization detector (FID) was modified as to include a six-port gas sampling valve, by means of which the carrier gas either enters at D_2 and meet at D_1 (valve position indicated by the solid lines) or vice versa (valve switched to the dotted-line position). All column sections of the cell were located inside the oven of chromatograph. They were empty stainless steel of 6.35 mm, chromatographic tube with a 4 mm i.d. and lengths $L = 28.5$ cm and $l = l'=57$ cm. They were connected at the junction $x = l'$ by a 6.35 mm. Swagelok tee union. Another simple 6.35 mm union was used to connect a short tube (2 cm) containing 0.5 cm^3 of liquid at the end of diffusion column L.

A conventional two-stage reducing valve and pressure regulator was followed by a needle valve, a 4 Å molecular sieve dryer, and a gas flow controller used to minimize variations in the gas flow rate. A restrictor placed before the detector served to prevent the flame from being extinguished, when the valve was switched from one position to the other.

PROCEDURE

After placing the liquid under study in its position (Figure 1) and waiting for a certain time, during which no signal is noted, we recorded a monotonously rising concentration-time curve for the vapor of the liquid. When this rising continous signal is high enough, the chromatographic sampling procedure is started by reversing the direction of the carrier gas flow for a time period *shorter* than the gas hold-up time in both column sections $l + l'$ (6 s). Then the gas flow is restored to its original direction. These flow reversals are done by switching the six port valve from one position (solid lines) to the other position (dotted-lines) and vice versa.

After certain dead time, an extra signal is recorded having the form of fairly symmetrical sample peak (Figure 2). This double reversal of the flow is repeated several times with always the same duration (6 s) of backward flow. It gives rise to a series of sample peak corresponding to various time t_0 from the beginning.

The pressure drop along column $l' + l$ was negligible, and the pressure inside the whole cell was set at 1atm. Temperature variations in the oven were less than ± 0.1K. The carrier gas flow rate was set at 1 cm^{3}s^{-1}.
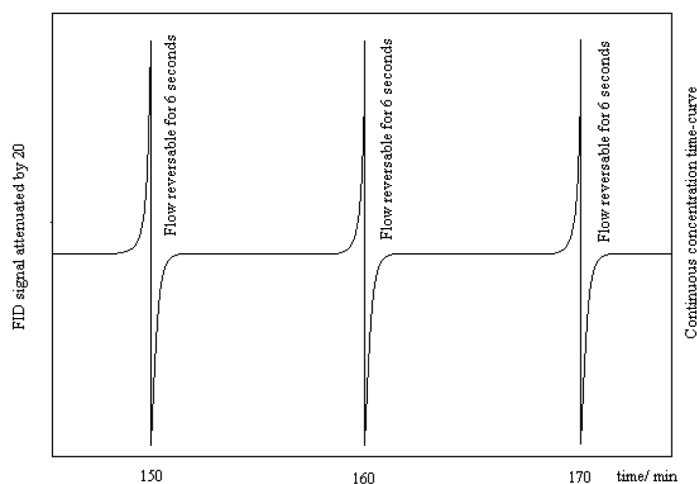

FIGURE 2. Reversed flow gas chromatogram showing two sample peaks for the adsorption of liquid pollutant at 312.15 K and 1atm (v=1.0 cm^3 s^{-1})

RESULTS AND DISCUSSION

Figure 3 shows the rise of the sample peak height with time for the diffusion of liquid vapor. In the analysis, the height, h of the sample peaks is used as a function of time t_0, when the flow reversal was made, which plotted on semilogarithmic scale. It shows the steep rise and the leveling off with time of the sample peak height.

From the plot of Figure 3, the highest peak value is taken for plotting $\ln(h_\infty-h)$ vs t_0 (cf. Figure 4). The height deviation which is taken as $\ln(h_\infty-h)$ of the sample peaks is used as a function of time t_0. The results from Figure 4 are iterated, which leaves 3 to 4 points, which corresponded to small times, and the rest of the experimental points are plotted according to (20), as shown in Figure 5.

From the slope of this plot, which is equal to $-2(k_cL+D)/L2$, according to (20), using the theoretically calculated value for D from Fuller-Schettler-Giddings equation[10], and the actual value of L, a value of k_c is calculated. This approximate value is now used to be plotted according to (21) as shown in Figure 5.

Table 1 summarizes the results obtained with all liquid studied. In the same table the diffusion coefficients determined here are compared with those calculated theoretically and/ or those found in the literature.

The evaporation rates of liquid pollutants over the temperature range 298.2–333.2 K were measured in order to obtain the activation energy of the respective liquid evaporation. The evaporation rate in units of cm s^{-1} was
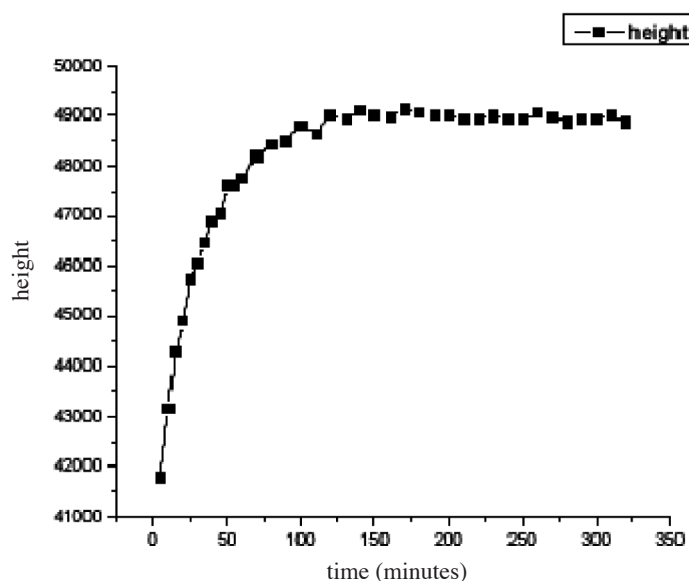
FIGURE 3. The rise of the sample peak height with time for the diffusion of liquid vapor (ethanol) into nitrogen (v=cm^3 s^{-1}), 313.15 K and 1 atm
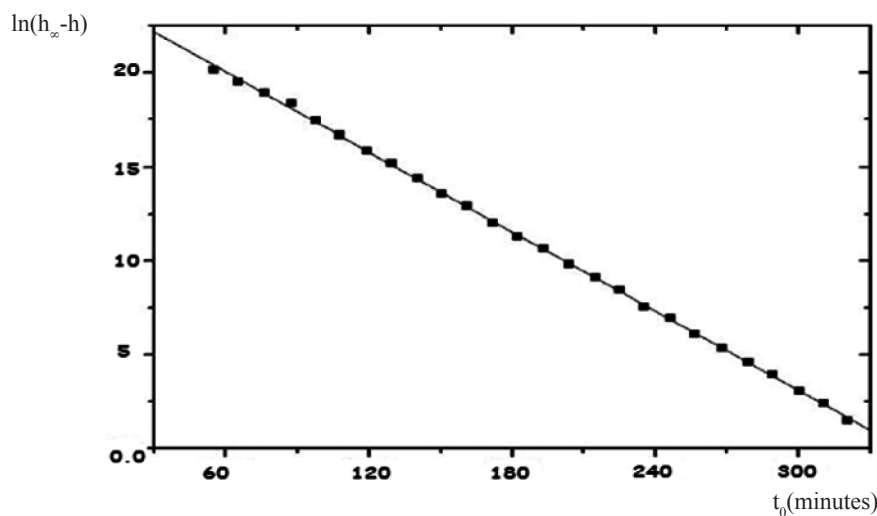
FIGURE 4. Example of plotting (20) for the diffusion of liquid vapor (ethanol) into carrier gas at 313.15K and 1atm (v=1.0cm^3 s^{-1})
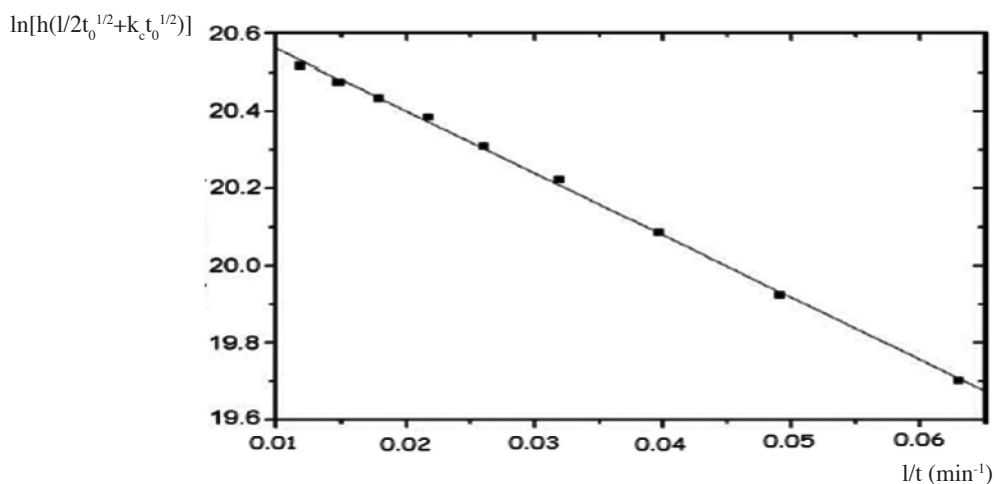
FIGURE 5. Data from evaporation of liquid vapor (ethanol) into carrier gas at 313.15 K and 1atm
(v=1.0 cm³s⁻¹), plotted according to (21)

TABLE 1. Binary gaseous diffusion coefficients, D_{AB} (cm²s⁻¹) (A: trace solute, B: carrier gas), measured
by the reversed flow-gas chromatographic techniques (v = 1cm³s⁻¹)

| Binary system A-B | T, K | $10^2 k_c$, cm s⁻¹ | D, cm² s⁻¹ | | | | |
			This work D^{exp}	Theory D^{theory}	Reference D^{lit}	Precision[b] (%)	Accuracy[c] (%)
CH_3OH-N_2	313.15	156.99±0.13[a]	0.1843±0.0007[a]	0.1843	-	0	-
	323.15	148.59±0.17[a]	0.1947±0.0003[a]	0.1947	-	0	-
	333.15	140.87±0.11[a]	0.2054±0.0006[a]	0.2054	0.2448[e]	0	19
$C_2H_5OH-N_2$	313.15	206.08±0.04[a]	0.1404±0.0009[a]	0.1403	0.1300[f]	0.06	7[d]
	323.15	195.11±0.05[a]	0.1483±0.0006[a]	0.1483	-	0.03	-
	333.15	185.02±0.09[a]	0.1563±0.0003[a]	0.1564	-	0	-
	343.15	175.58±0.07[a]	0.1648±0.0009[a]	0.1647	-	0.07	-
$C_3H_7OH-N_2$	313.15	247.45±0.14[a]	0.1169±0.0002[a]	0.1169		0	
	323.15	234.31±0.15[a]	0.1234±0.0001[a]	0.1234	-	0	-
	333.15	222.04±0.11[a]	0.1303±0.0001[a]	0.1303	-	0	-
	343.15	210.84±0.13[a]	0.1372±0.0001[a]	0.1372	-	0	-
	353.15	200.50±0.14[a]	0.1443±0.0001[a]	0.1443	-	0	-
	363.15	190.94±0.15[a]	0.1515±0.0001[a]	0.1515	0.1530[g]	0	1[d]
$C_4H_9OH-N_2$	313.15	283.57±0.02[a]	0.1020±0.0005[a]	0.1020	-	0.01	-
	323.15	268.08±0.07[a]	0.1079±0.0015[a]	0.1078	-	0.13	-
	333.15	254.39±0.10[a]	0.1137±0.0004[a]	0.1137	-	0.04	-
	343.15	241.65±0.05[a]	0.1197±0.0002[a]	0.1197	-	0	-
	353.15	229.80±0.12[a]	0.1259±0.0001[a]	0.1259	-	0	-
	363.15	218.75±0.04[a]	0.1322±0.0008[a]	0.1322	-	0.04	-
	373.15	208.68±0.08[a]	0.1386±0.0002[a]	0.1387	-	0.07	-

(continue)

Continued (TABLE 1)

Binary system A-B	T, K	$10^2 k_c$, cm s^{-1}	D, cm^2 s^{-1}				
			This work D^{exp}	Theory D^{theory}	Ref. D^{lit}	Precision[b] (%)	Accuracy[c] (%)
C$_5$H$_{12}$-N$_2$	298.15	338.27±0.20	0.0854±0.0004	0.0855	-	0.04	-
C$_6$H$_{14}$- N$_2$	313.15	332.17±0.13	0.0871±0.0005	0.0886	-	0.26	-
	323.15	315.19±0.15	0.0918±0.0007	0.0918	-	0	-
	333.15	298.84±0.08	0.0968±0.0010	0.0968	-	0	-
C$_7$H$_{16}$-N$_2$	313.15	362.22±0.12	0.0798±0.0009	0.0799	-	0.04	-
	323.15	342.01±0.18	0.0846±0.0004	0.0844	-	0.20	-
	333.15	324.94±0.17	0.0890±0.0004	0.0890	-	0	-
	343.15	308.57±0.12	0.0937±0.0007	0.0937	-	0	-
	353.15	293.39±0.17	0.0986±0.0003	0.0986	-	0	-
	363.15	279.40±0.16	0.1035±0.0016	0.1035	-	0	-
C$_{16}$H$_{14}$-N$_2$	313.15	563.54±0.04	0.0513±0.0009	0.0513	-	0	-
	323.15	533.37±0.07	0.0542±0.0007	0.0542	-	0	-
	333.15	505.40±0.15	0.0574±0.0008	0.0572	-	0.05	-
	343.15	480.11±0.04	0.0603±0.0012	0.0602	-	0.01	-
	353.15	456.63±0.09	0.0633±0.0007	0.0633	-	0	-
	363.15	434.87±0.07	0.0665±0.0008	0.0665	-	0	-
	373.15	414.66±0.05	0.0697±0.0014	0.0697	-	0	-

[a] Uncertainty based on standard error value

[b] Precision has been defined as (|D^{exp} - D^{theory}| / D^{exp}) × 100 [11]

[c] Accuracy has been defined as (|D^{exp} - D^{lit}| / D^{exp}) × 100 [11]

[d] Corrected to the experimental temperature

[f] Karaiskakis et al. 1986

[g] Agathonos & Karaiskakis et al. 1989

[h] Koliadima et al. 1988

calculated from equation (20). The activation energy of liquid evaporation from the surface can be calculated from the temperature dependence of the evaporation rate employing the Arrhenius equation,

$$\ln k = \ln A - \frac{E_a}{RT}, \qquad (22)$$

where k is the evaporation rate per unit area (cm s^{-1}), Ea is the activation energy (J mol^{-1}), T is absolute temperature (Kelvin), R is the gas constant taken as 8.3145 J mol^{-1} K^{-1}, and A is an arbitrary constant. Thus, Ea can be obtained by analyzing the evaporation rates according to equation (22). This definition applies regardless of whether the Arrhenius plot is linear or not; if it is not, the activation energy changes with temperature.

The activation energy calculated for respective samples at various temperatures were given about the same amount of values (Table 2). It is proven by the definition of activation energy where the minimum amount of energy is required to initiate the reaction process. In this experiment, the reaction can be defined as evaporation process, where the liquid pollutants were transformed into gaseous vapors. Therefore, the above mentioned activation energy is the minimum amount of energy required for liquid molecules to transit to the gaseous phase.

CONCLUSION

In conclusion, the hypothesis of equilibrium at the liquid-gas interphase leads to profound agreement with the experimental evidence. This precision is better than 1% in all cases. An evaluation of k_c values determined in this work with other literature is not possible, because such values could not be found. While the comparison of

TABLE 2. The activation energy values of respective samples calculated using Arrhenius equation and measured by the reversed flow-gas chromatographic techniques ($v = 1\text{cm}^3\text{s}^{-1}$)

Sample	Activation Energy (J mol^{-1})
$CH_3OH\text{-}N_2$	4697.925
$C_2H_5OH\text{-}N_2$	4766.189
$C_3H_7OH\text{-}N_2$	4904.346
$C_4H_9OH\text{-}N_2$	4958.178
$C_5H_{12}\text{-}N_2$	4432.657
$C_6H_{14}\text{-}N_2$	4583.339
$C_7H_{16}\text{-}N_2$	4889.463
$C_{16}H_{14}\text{-}N_2$	4964.504

diffusion coefficients of respective liquids pollutant and previous work is unattainable, because limited literatures for the work at respective temperatures are published. The basic advantage of the method is its precision and experimental simplicity. Absolute evaporation rates, which are independent of carrier gas flow-rate and, furthermore, related to the physical properties of the evaporating liquids, are determined simultaneously with the respective diffusion coefficients of the evaporating vapors into the carrier gas.

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