

Study the Effect of Imposing Surfactants toward the Evaporation of Low Molecular Weight Alcohol

Mohammad Hafiz Hamzah, Sharifuddin Mohd Zain, Rashid Atta Khan, Khalisanni Khalid

Abstract—In this paper, Reversed-Flow Gas Chromatography (RF-GC) is utilized to investigate the evaporation of low molecular weight alcohol. Evaporation rates as well as the diffusion rates of methanol are determined with a surfactant monolayer on the surface of the liquid; while nitrogen acts as carrier gas, at 313 K. The precision (>99.9%) and accuracy of this investigation demonstrates the potential of current methodologies for environmental impact studies; this is further verified when the results are compared with the available literature. The varying evaporation rates of methanol in the presence of varying amounts of Triton X-100 reflects that application of surfactants do damper the evaporation rates of liquid pollutants; without interference with the former's diffusion coefficients. High amounts of Triton X-100 are required for retardation of evaporation rates, suggesting the formation of a densely packed surface monolayer or the formation of an insoluble monolayer.

Index Terms—Evaporation, diffusion coefficients, rate coefficients, surfactants.

I. INTRODUCTION

The ecological fate and evaporation of pollutant liquids are co-related to each other, especially when the compounds are introduced into both freshwater and marine environments through industrial effluents, or introduced directly into the air from industrial unit process units such as bioreactors and cooling towers [1]. The process is highly dependent on the concentration of the pollutant liquid, it will travel according to the concentration gradient, i.e. from high concentration to low concentration of the respective liquid. There are barriers that contribute to the evaporation rate [3] which includes the “stagnant” gaseous and liquid layer close to the surface, across which the vapor must diffuse. One area of application is industrial accidents, for example such as when the MV Bunga Alpinia 3 exploded while being loaded with methanol on 26 July 2012 at the PETRONAS jetty in Labuan [2]. As a result, methanol – one of the liquids under study, spilled into the sea near Labuan. The Reversed-Flow Gas Chromatography (RF-GC) method can be used as an indicator to ‘measure’ the evaporation rates of liquid

pollutants and can quickly guide actions to be taken by the respective authorities, such as applying a suitable surfactant to curb the methanol from being evaporated into the air – this effort will help prevent the people nearby from inhaling the hazardous vapors of methanol. This may be possible if the application of surfactant on the liquid pollutant results a drastic retardation of the evaporation rate, mainly because of the presence of adsorbed monolayers [3]. Because RF-GC sampling is fast and accurate, it is a reliable method to investigate how much liquid pollutant has been evaporated into the air. Besides that, the methodologies can also embrace the area of food chemistry, chemical kinetics, catalyst and catalysis [4]. Surfactants which form monolayers at the air-liquid interface have been found to cause a significant reduction in the evaporation rate of water. Long chain alcohols decrease the evaporation rate of the water on an actual lake by a factor of 46-50% [16-22]. This paper demonstrates the utilization of RF-GC in the study of evaporation of a low molecular alcohol weight (methanol), with various amounts of the nonionic surfactant Triton X-100.

II. METHODOLOGY

A. Preparation of the TritonX-100-methanol solution

The Gibbs adsorption equation (solutions of nonionic surfactants) as stated below [5]

$$\Gamma_1 = -\frac{1}{2.303RT} \left(\frac{\partial \gamma}{\partial \log C} \right) \quad (1)$$

is used to prepare the various Triton X-100 – methanol solutions in order to determine the amount of Triton X-100 per unit area.

- γ is the surface tension in N.m^{-1} initially and then converted to J.m^{-2} .
- C_1 is the molar concentration of the surfactant at the experimental absolute temperature $T = 298.15 \text{ K}$
- $R = 8.314 \text{ J.mol}^{-1}.\text{K}^{-1}$ is the ideal gas constant
- Γ_1 is the surface excess constant of the surfactant in mol.m^{-2} .

The surface tension, γ is plotted against the log of the concentration, C_1 to determine the surface area per molecule, α_1^s as shown in Fig. 1

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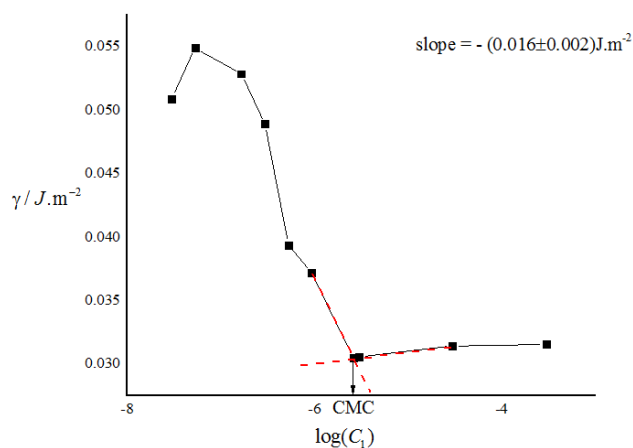


Fig. 1. Plot of the surface tension ($\gamma/\text{J.m}^{-2}$) against the log of C_1 , where C_1 is the bulk phase concentration of the various aqueous solutions of Triton X-100 surfactant, at 298.15 K.
 **CMC = Critical micelle concentration Surface tension measurements were performed by using a Kruss Tensiometer K100.

The surface excess concentration of Triton X-100 was calculated: $\Gamma_1 = (2.78 \pm 0.04) 10^{-6} \text{ mol.m}^{-2}$ by using the slope of Fig. 1. From the value Γ_1 obtained, a specific surface area per molecule at the interface was calculated as follows [6]:

$$\alpha_1^s = \frac{10^{20}}{N_A \Gamma_1} \quad (2)$$

where N_A is Avogadro's number and Γ_1 is in mol.m^{-2} . The value of α_1^s was found as $(5.975 \pm 0.080) \cdot 10^9 \text{ m}^2$. The values of Γ_1 and α_1^s were found with the same magnitude of the similar homologous head group of nonionic surfactants given on the page 78 of Ref 5. The CMC** for the aqueous solutions of Triton X-100 was found to be $C_1 = 2.57 \cdot 10^{-4} \text{ M}$ or 0.0002% vol. The theoretical coverage of the Triton X-100 monolayer, ranging from 1 to 4 monolayers was determined [3], since the surface area of the bottle containing the evaporating liquid under study, a_L as well as the specific surface area per molecule at the interface, α_1^s were known.

B. Materials

The solutes used as evaporating liquids (stationary phase) were Merck "SupraSolv[®]" (methanol) and TritonX-100 (iso-Octylphenoxypolyethoxyethanol, $d = 1.06 \text{ g.ml}^{-1}$ and $M = 646.37 \text{ g.mol}^{-1}$). Purified nitrogen (99.9% purity) supplied by Malaysian Oxygen Berhad (MOX) was used as the carrier gas (mobile phase). Hydrogen and compressed air used to fuel the FID were supplied by MOX as well.

C. Techniques

The apparatus used and the experimental procedure followed have been described elsewhere [8],[9]. A conventional gas chromatograph (Shimadzu, Series GC-14B) with a flame ionization detector (FID) and Shimadzu Solution as the software program was used for this experiment. The diffusion column (28.5 cm x 4.0 mm i.d.) was placed perpendicular to the center of the sampling

column l' and l of [(57 + 57) cm x 4.0 mm i.d.], as shown in Figure 2. Both columns were connected by a 6.35 mm Swagelok tee union. The columns used in this chromatography were 6.35 mm diameter stainless-steel, unfilled with any solid material. All column sections were located inside the oven. The carrier gas was adjusted to a $1 \text{ cm}^3 \text{ min}^{-1}$ continuously flows through the sampling column, while it is stagnant inside the diffusion column, L . A glass bottle (2 cm^3) filled with methanol (4 cm^3) was placed at the lower close end of the diffusion column, with a 6.35mm Swagelok stainless nut.

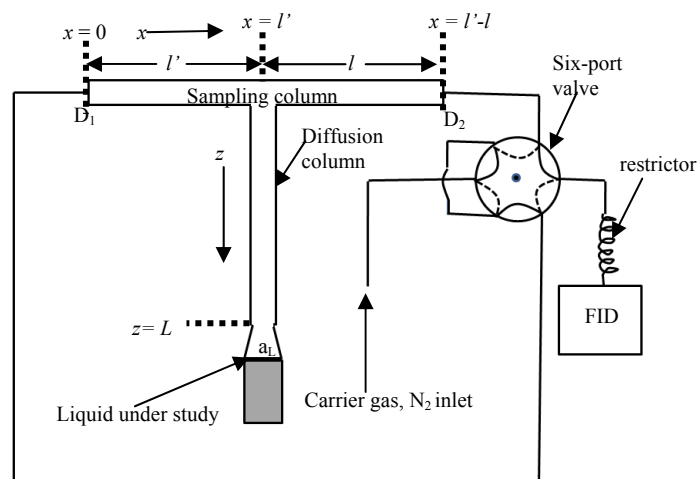


Fig. 2. The apparatus of the reversed-flow gas chromatography technique, for measuring rate coefficients and diffusion coefficients of liquids.

A six-port valve as shown in fig 2 was used to permit the carrier gas to enter either from D_2 and out at D_1 then immediately to the FID (valve position indicated by the solid line) or vice versa (the valve is switched to the dotted-line position). The valve reversed the direction of the carrier gas for 6 s, which a shorter time period than the gas's hold-up time in both column section l' and l , then the gas was returned to its original direction, with resulting extra chromatographic peaks (sample peaks) superimposed on the continuous elution curve as depicted in fig 3.

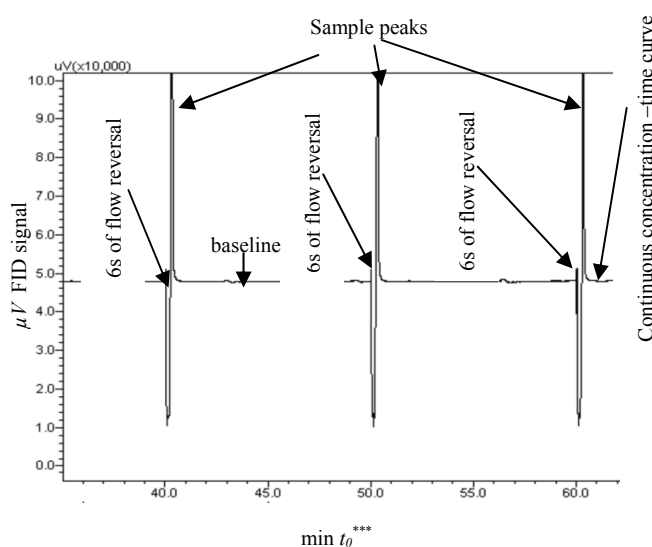


Fig. 3. A reversed flow chromatogram showing two sample peaks for the diffusion of liquid vapors into carrier gas nitrogen at 323.15 K and 101325 Pa (volumetric flow rate = $1 \text{ cm}^3 \text{ min}^{-1}$)

*** t_0 = Time from the beginning to the last reversal of gas flow

The process was repeated during the experiment, lasting for 5 hours and 40 minutes. The pressure drop along section l' and l was negligible as the carrier gas flow rate was kept constant. ($1.0 \text{ cm}^3 \cdot \text{s}^{-1}$)

D. Theoretical basis

The height, h of the sample peaks from the continuous signal, taken from baseline to the maximum, was plotted as $\ln h$ versus time, giving a diffusion bands as shown in Fig. 4.

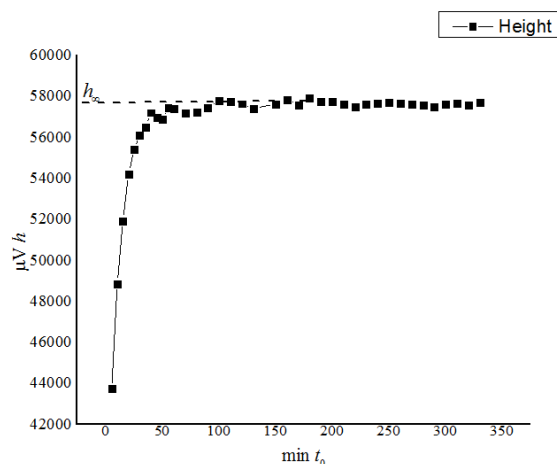


Fig. 4. Diffusion band (plot of sample peaks height, h , against time, t_0 , from the beginning of the experiment) for the evaporation of liquid, at 313.15 K and 101325 Pa.

Each sample peak which has been shown earlier [8], produced by two reversals in RF-GC, is symmetrical and its maximum height h from the ending baseline is given by

$$h \cong 2c(l', t_0) \quad (3)$$

Thus, the concentration of the vapors of an evaporating liquid, $c(l', t_0)$ at $x = l'$ and time t_0 is proportional to the height or the area of the experimentally obtained sampling peaks, and it is interrelated with the rate coefficient for the evaporation process, K_G , the diffusion coefficient of the vapor into the carrier gas, D , and the geometrical details of the diffusion column through the relation [8]:

$$c(l', t_0) = \frac{K_G D c_0}{V(K_G L + D)} \left\{ 1 - \exp \left[-2(K_G L + D)t_0 / L^2 \right] \right\} \quad (4)$$

where L is the length of the diffusion column and v the volumetric flow rate of the carrier-gas. Figure 5 represents the sampling of the above-mentioned process against time. It becomes obvious that after a period of time, which is characteristic of each particular interaction system, a steady-state situation is achieved. From this plot, at long times an infinity value for the peak height h_∞ can be obtained. This infinity h_∞ value is used for the linearization of the resulting relation [8]:

$$h_\infty = \frac{2K_G D c_0}{[v(K_G L + D)]} \quad (5)$$

Using the former approximation, one obtains [8]:

$$\ln(h_\infty - h) = \ln h_\infty - \left[\frac{2(K_G L + D)}{L^2} \right] t_0 \quad (6)$$

Thus, at long enough times, for which Eq. 4 was derived, a plot of $\ln(h_\infty - h)$ vs. t_0 is expected to be linear, and from its slope $-2(K_G L + D) / L^2$ a first value of K_G can be calculated from the known value of L and a literature or theoretically calculated value of D . [9] – [11]

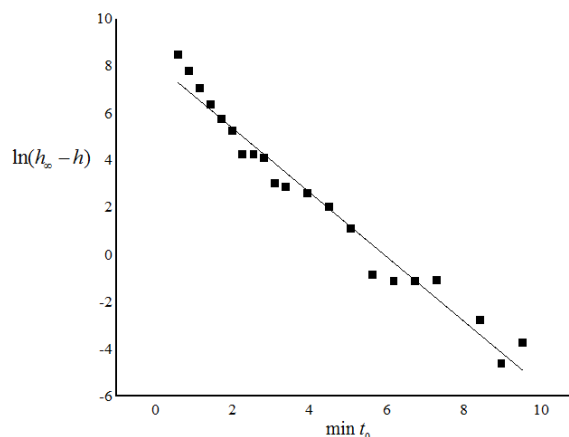


Fig. 5. Example of plot for the diffusion of liquid vapor into carrier gas at 313.15 K and 101325 Pa ($v = 1.00 \text{ cm}^3 \text{ s}^{-1}$)

The value of K_G can now be used to plot small time period data according to Eq. 14 of Ref. 9, which is substituted now for $c(l', t_0)$ in Eq. 2. After rearrangement, logarithms are taken and there results:

$$\ln \left[\left(\frac{L}{2t_0^{1/2}} + K_G t_0^{1/2} \right) \right] = \ln \left[\frac{4K_G c_0}{v} \left(\frac{DL}{\pi} \right)^{1/2} \right] - \frac{L^2}{4D} \frac{1}{t_0} \quad (7)$$

Now a plot of the lefthand side of this relation versus $\frac{1}{t_0}$ will yield a first approximation experimental value for D

from the slope $\frac{-L^2}{4D}$ of this new linear plot

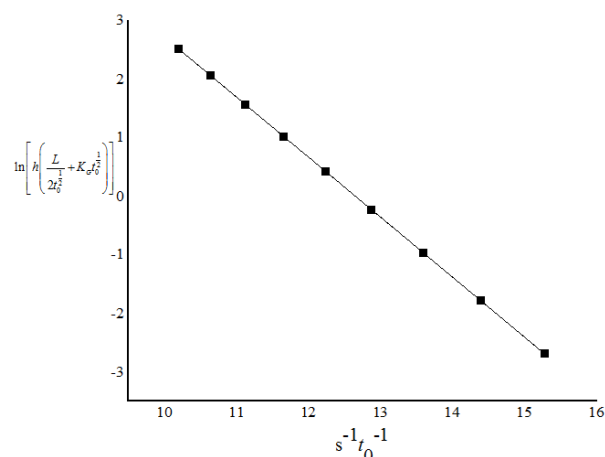


Fig. 6. Data from evaporation of liquid vapor into carrier gas at 323.15 K and 101325 Pa ($V = 1.00 \text{ cm}^3 \text{ sec}^{-1}$), plotting according to Equation 7

III. RESULTS AND DISCUSSION

TABLE I: RATE COEFFICIENTS FOR THE EVAPORATION OF METHANOL, K_G , AND DIFFUSION COEFFICIENTS OF ITS VAPORS INTO NITROGEN, D_{FOUND} , UNDER THE EFFECT OF VARIOUS AMOUNT OF SURFACTANT TRITON X-100, AT 313.15 AND 101 325 PA

Monolayer Thickness of Triton X-100	$10^2 K_G$ (cm s ⁻¹)	Retardation of k_c , %	$10^3 D_{\text{found}}$ /cm ² s ⁻¹	$10^3 D_{\text{lit}}$ /cm ² s ⁻¹	$10^3 D_{\text{lit}}$ /cm ² s ⁻¹	Deviation,%	Deviation,%
0	96.69 \pm 0.20	-	181.8 \pm 0.04 ^a	184.31	200.58	1.4*	9.3 [#]
1	31.75 \pm 0.05	67.2	189.23 \pm 0.02 ^a	184.31	200.58	2.6*	5.7 [#]
2	20.71 \pm 0.14	78.6	188.07 \pm 0.02 ^a	184.31	200.58	2.0*	6.2 [#]
3	17.31 \pm 0.07	82.1	183.14 \pm 0.02 ^a	184.31	200.58	0.6*	8.7 [#]
4	12.30 \pm 0.31	87.3	189.46 \pm 0.02	184.31	200.58	2.7*	5.5 [#]
	Mean values		186.35 \pm 0.02			(1.9*) ^c	(7.1 [#]) ^c
	Precision, %		99.9 ^b				

^aUncertainty obtained from the standard error of the k_c and D values, estimated from the slopes of the linear plots of Eqs. 20 and 21 of Ref. 8 respectively.

^bPrecision determined from the mean value and the standard error of the experimentally obtained diffusion coefficients.

^cMean deviation of the experimental diffusion coefficients from the respective predicted^{*}[11] and experimental[#][15] literature values, D_{lit} .

Table I summarizes the results for the evaporation of methanol under the influence of the surfactant Triton X-100 ranging from 0 to 4 monolayers. The diffusion coefficients which are determined from this experiment were compared with the predicted values from the Fuller-Giddings equation [11] and experimentally obtained values from previous experimental work [15] which was using a different method. The deviation of the diffusion coefficients of the current experiment from the predicted and experimental literature values, D_{lit} are illustrated in the last column and are based on the following equation:

$$\text{Deviation}(\%) = \left[\frac{(D_{\text{found}} - D_{\text{theoretical/literature}})}{D_{\text{found}}} \right] \times 100$$

Based on the diffusion coefficient values, D_{found} , ones can be assured that they are independent of the addition of surfactant, as expected [3]. The mean deviation of the methanol + Triton X-100 liquid mixtures experimental obtained by RF-GC, D_{found} , from the predicted and literature values are 1.9 and 7.1 % respectively. The D_{found} values falls between the predicted and literature ones. The total reproducibility of this method is determined 99.9% for the case of the liquid stated.

The uncertainty in the determination of the vaporization values, k_c , varies from 0.2 to 2.5% in this case. Based on the percentages, we can conclude that the values experimentally obtained by use of RF-GC can be used to reliably predict the effect of Triton X-100 in the evaporation rates of methanol.

We can conclude that the k_c values in the presence of various amounts of Triton X-100 corresponding to the formation of an adsorbed monolayer at the surface of the methanol; demonstrating that Triton X-100 does make the evaporation rates decrease. The evaporation rate retards by a high percentage, i.e. 67.2%; with just one monolayer of TritonX-100 being applied on the surface of the methanol.

IV. CONCLUSION

The present method gives an alternative from the previous method [15], since the former method can be used in simultaneously determining the evaporation rates and diffusion rates of the liquid under study. The present method can be considered accurate since the results show that the diffusion in the current experiment deviates from the predicted theoretical value by less than 2.0% (based on mean values); as compared from the works by Anikar H.J. et. al. [15]. Also, the use of RF-GC has value because of its precision and experimental simplicity.

The evaporation rates found by the current experiment are absolute and as a result they can be related to the physical properties of the evaporating species.

We can obviously extend the use of RF-GC to other topics that are related to the environmental, such as investigation of water evaporation (sea, lake, river), the effect of ionic and zwitterionic surfactants on the evaporation rates of liquid pollutants; as well as experiments with surfactants under steering conditions in order to investigate their durability[3].

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Upon study, he was appointed as residential assistance for PERMATAPintar Program in Universiti Kebangsaan Malaysia. He flew away to Thailand and Singapore to give the speech on his research. His diligence was rewarded with a degree in Master of Science in Physical/ Environmental Sciences in 2011. Before graduation, he was offered to join Malaysian Agricultural Research and Development Institute (MARDI). There onwards, his interest of research in science and business grew. This was further encouraged by the recognition and support from MARDI, where he was appointed as Technical Officer for MARDI Kuala Linggi Incubator Program. This project leads him to train SMEs for essential oil production. In MARDI Headquarters Serdang, he is responsible to lead Essential Oil Analysis Unit under Food and Agricultural Analysis Laboratory Program, Technical Service Centre. He is also the gatekeeper for Malaysian Herbal Authentication Centre (MHAC).

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