International Journal of Engineering Research and Development
e-ISSN: 2278-067X, p-ISSN: 2278-800X, www.ijerd.com
Volume 11, Issue 11 (November 2015), PP.19-31

Measurement and Correlation of Diffusion Coefficient In Hexane – Air System

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Abstract:- The main objective of this study is to find out experimentally the effect of air flow rate on diffusivity of Hexane. Diffusivities of vapours are most conveniently determined by the method developed by WlNKELMANN in which liquid is allowed to evaporate in a vertical glass tube over the top of which a stream of vapour-free gas is passed, at a rate such that the vapour pressure is maintained almost at zero. The experiment was conducted at different flow rates of air varying from 0 to 10 lph. The Diffusion coefficient of Hexane at different flow rates of air are calculated and compared with the proposed model developed from the Gilliland equation.

The proposed model equation derived from Gilliland's equation is $D_f / D_0 = k1\ Q + c$. The parameter 'k1' and 'c' remains nearly constant for the range of flowrate of air (0 – 10 lph) . The value of 'k' and 'c' are 0.044 and 2.251 respectively. The experimental Diffusivity value at different flowrates are compared with the proposed model and the percentage error is around 2% in the flowrate of 0 – 10 lph. The results indicates that the proposed model can be used to predict the Diffusion coefficient at different flowrates of air at 32^0C and 101.3 kPa.

Keywords:- Diffusivity, Gilliland equation, Diffusion coefficient, Hexane-air system.

I. INTRODUCTION

Estimation of liquid diffusivities is of considerable interest in understanding the mechanism of solute transport in liquids specially in liquid liquid extraction, distillation, crystallization, absorption and chemical reaction.

If a solute is partitioned between two immiscible solvents, there is diffusion in liquids which was clarified by Thomas Graham, even though he has failed to put his experiments on a quantitative basis. Following graham's work, Fick has described diffusion on the same mathematical basis as Fourier's law of heat conductance or ohm's law of electrical conduction. This analogy is still a useful pedagogical tool.

Fick has defined the total one dimensional flux J_1 as

$J_1 = - D (\partial c_1 / \partial x)$

where c_1 is the concentration of the solute and x is the distance. The term D, which Fick calls "the constant depending on the nature of the substances" is of course, the diffusion coefficient.

Diffusion coefficient for liquids are atleast one thousand times smaller than those in gases. Theoretical estimation of diffusion in liquids are much less accurate than those for gases.

The various theoretical approaches to diffusion in liquid mixtures have helped to provide a greater insight into the diffusion process. The intermolecular forces, the shape and size of molecules are all important factors. However, these attempts have not been very successful when complex interactions between the molecules in the liquid state occur. In view of the difficulties in formulating the exact theory, model approaches such as cell model, free volume model and hard sphere model have been resorted too.

However, correlations based on experimental measurements predicts diffusivities better than those by theoretical predictions. Owing to the complex nature of liquids, correlations based on semi theoretical considerations also do not always give satisfactory predictions on account of complex molecular interactions as in high viscous solutes / solvents and also association tendencies in the solute – solvent systems.

II. SEMI EMPIRICAL CORRELATIONS

There are number of semi empirical correlations reported for the prediction of diffusivities in liquids. Most of the correlations do not take association or complex formation between solute and solvent molecule into consideration. Recently some of the investigators have studied systems involving simple binary solute-solvent interaction.

A. Studies which do not involve association or complex formation covering low viscous solvent systems

1)Scheibel's Correlation : Scheibel (25) has proposed the following correlation for prediction of liquid diffusivities at infinite dilution.

$D_{AB} = KT / \eta_B V_A^{0.33}$ ----- (3.1)

Where K is given by

$K = 8.2 \times 10^{-8} [1 + (3V_B / V_A)^{0.66}]$

Except for water as a solvent, if $V_A < V_B$, use $K = 25.2 \times 10^{-8}$

For benzene as a solvent if $V_A < 2V_B$, use $K = 18.9 \times 10^{-8}$

For other solvents if $V_A < 2.5V_B$, use $K = 17.5 \times 10^{-8}$.

2)Reddy and Doraiswamy : The following correlation has been suggested by Reddy and Doraiswamy (23) for prediction of diffusion coefficient in liquids.

$D_{AB} = k' M_B T / \eta_B (V_A V_B)^{0.33}$ --- (3.2)

Where $k = 10 \times 10^{-8}$ if $V_B / V_A = \leq 1.5$

$k = 8.5 \times 10^{-8}$ if $V_B / V_A = \geq 1.5$

3)Lusis and Ratcliff : To estimate the diffusion coefficient in organic solvents, Lusis and Ratcliff (15) have proposed the following correlation

$D_{AB} = 8.52 \times 10^{-8} T [(1.4(V_B / V_A)^{0.33} +(V_B/V_A)] / \eta_B (V_B)^{0.33}$ ---(3.3)

This equation is not recommended for systems where there are strong solute – solvent interactions such as formation of association complexes.

4)King. Hsuch and Mao : The following correlation has been suggested by king, Hsuch and Mao (11) incorporating latent heat of vaporization as correlating parameter.

$D_{AB} = 4.4 \times 10^{-8} T (V_B / V_A)^{0.16} (\Delta H V_B / \Delta H V_A)^{0.5}$ --- (3.4)

However, the above equation was found to be inadequate for applicability to viscous and aqueous solvent systems.

5)Laddha – Smith : Laddha – Smith (13) correlation for the estimation of liquid diffusivities was based on the hydrodynamic theory of diffusion. Stokes – Einstein equation suggests that for dilute solution

$D\eta / T = k / 6\pi N r_A$ ---- (3.5)

Where r_A is the radius of the solute particle. If the radius of the diffusing solute particle is not influenced by the solvent media, the value of r_A may be computed by the following equation

$r_A = [(3\pi / 4) (V_A / N)]^{0.5}$ ---- (3.6)

where V_A = the molar volume of the solute

As the experimental analysis of diffusivity data for various liquid system indicates that the product $D\eta / T (V_A)^{0.33}$ is not constant, it seems apparent that the radius of the diffusing solute particle is influenced by the solvent species. Hence the dependency of r_A on molecular volumes of the solute species may be expressed by the following equation.

$r_A = [(3\pi /4) (V_A / N)]^{0.33} f (V_B / V_A)$ ---- (3.7)

$\alpha [(3\pi /4) (V_A / N)]^{0.33} (V_B / V_A)^{\delta}$ ---- (3.8)

where α and δ are constants.

The following generalized relationship results for the estimation of liquid diffusivity by substituting the value of r_A from the above equation into the Stokes – Einstein equation, we have

$[D_{AB} \eta_B / T] (V_A)^{0.33} = (k N / 6 \pi)^{0.33} (4 \pi / 3)^{0.33} (V_B / V_A)^{\delta}$ --- (3.9)

since the product $(k N / 6 \pi)^{0.33} (4 \pi / 3)^{0.33}$ is constant, this equation reduces to

$[D_{AB} \eta_B / T] (V_A)^{0.33} = \alpha_0 (V_B / V_A)^{\delta}$ --- (3.10)

where $\delta = 0.1604$ and ηB is the viscosity of the solvent.

For solutes other than water α_0 was found to be 1.892×10^{-7}

However, laddha and smith have analysed the applicability of equation (3.10) To systems with water as solute (a highly associating solute) and gave a new value for α_0 on the basis of data analysis as follows

$[D_{AB}\ \eta_B / T]\ (V_A)^{0.33} = 1.06 \times 10^{-7}(V_B / V_A)^{0.1604}$ ---- (3.11)

B. Studies involving high viscous solvent Systems

Lusis (14), based on an intensive study of predictive method for diffusion coefficients in high viscous solvents, found that no correlation suggested so far was adequate enough. He has proposed a correlation for diffusivities in viscous liquids, which is also applicable for associated and hydrogen bonding liquids. He contended that latent heats of vaporization of both solute and solvent are just enough to explain association. The correlation proposed is

$D_{AB} = 5.2 \times 10^{-8}\ (T/ \eta_B)\ (\eta_B\ \Delta H_B / \eta_A\ \Delta H_A)^{0.25}$ --- (3.12)

Where ΔH_A and ΔH_B are the latent heats of vaporization of solute and solvent at the temperature of diffusion.

C. Studies incorporating effect of solvent association

Wilke and chang (32) introduced an association parameter for the solvent in their correlation. Only for a few solvents they have given the association parameter and have not defined the association parameter for most of the solvents. Also they have not taken into consideration solute – solvent interactions or association among solute molecules. The wilke – chang correlation is

$D_{AB} = 7.4 \times 10^{-8}\ (\varphi\ M_B)^{0.5}\ T / \eta_B\ (V_A)^{0.6}$ --- (3.13)

Where φ is the association parameter of solvent

Φ = 2.6 for water as the solvent

Φ = 1.9 for methanol as solvent

Φ = 1.5 for ethanol as solvent

Φ = 1.0 for non associating solvents

D. Studies incorporating solute – solvent interaction

Ratcliff and Lusis (22) have shown that solute-solvent association complex formation in organic liquid mixtures can lead to large errors in the prediction of diffusivities by existing correlations. They have considered a simple case where the complex consists of one molecule of solute and one of solvent. With the help of the ideal solution model of an associated liquid, it was shown that a significant improvement can be made in the predictions, if proper account is taken of complex formation. A complex would be formed by hydrogen bonding in organic mixtures where one component contains a donor atom but no active hydrogen atoms (as in ethers, ketones, esters, aldehydes etc.) while the other contains an active hydrogen atom but no donor atoms (as in chloroform and alcohols). For example in the solute – solvent system, acetone – chloroform, the following association complex is possible between one molecule of acetone containing the donor atom and one molecule of chloroform containing the active hydrogen atom. The following correlation is given by Ratcliff and Lusis

$D_{AB} = 1 / (1 + k)\ (D_{A1} + kD_{A1B1})$ ---- (3.14)

Where D_{A1} is the diffusion coefficient of monomer given by

$D_{A1} = 8.52 \times 10^{-10}\ (T / \eta B)\ (1 / VB)^{0.33}\ [1.4\ (VB/VA)^{0.33} + VB/VA]^{0.33}$

D_{A1B1} is the diffusion coefficient of complex given by

$D_{A1B1} = 8.52 \times 10^{-10}\ (T / \eta_B)\ (1 / V_B)^{0.33}$

And $k = (1 - \gamma_A) / \gamma_A$ where γ_A = Activity Coefficient

In a recent work kuppuswamy and laddha (12) have investigated the diffusivities of sample solute – solvent complex forming systems by using the model of Ratcliff and Lusis given by the equation (16) but used different equations for defining D_{A1} and D_{A1B1} on the basis of Laddha – Smith (13) η_B model which is

$D_{A1} = 2.0 \times 10^{-7}\ (T / \eta_B)\ (1 / V_B)^{0.33}\ (V_B / V_A)^{0.16}$ ---- (3.15)

$D_{A1} = 2.0 \times 10^{-7}\ (T / \eta_B)\ [1 / (V_A + V_B)]^{0.33}\ [V_B / (V_A + V_B)]^{0.16}$ ---- (3.16)

They found that prediction by the use of the above model were in better agreement with the experimental values as compared to Ratcliff and Lusis (22) predictions.

Karthikeyan and Laddha (10) have suggested the following correlation for highly viscous solutes forming simple solute – solvent complexes on the basis of Lusis (14) model, but with slightly varying constants.

$D_{AB} = 5.625 \times 10^{-8}\ (T / \eta_B)\ [\eta_B\ \Delta HB/ \eta_A\ \Delta H_A]^{0.2225}$ ---- (3.17)

E. Conclusions from Literature Review

The above literature review indicates that little attention has been paid towards studying systems, where solute association affect diffusivity values, as for example in the case of acetic acid solute where association may occur.

Hence it would be of interest to study diffusivities of solute systems which exhibit solute association, and to check the applicability of the correlation of Laddha – Smith (3.13) for water as solute (a highly associating solute) and that of Lusis. A new attempt at deriving a model to take into account the solute association will be useful.

III. EXPERIMENTAL SET UP AND PROCEDURE

Diffusivities of vapours are most conveniently determined by the method developed by WINKELMANN in which liquid is allowed to evaporate in a vertical glass tube over the top of which a stream of vapour-free gas is passed, at a rate such that the vapour pressure is maintained almost at zero. If the apparatus is maintained at a steady temperature, there will be no eddy currents in the vertical tube and mass transfer will take place from the surface by molecular diffusion alone. The rate of evaporation can be followed by the rate of fall of the liquid surface, and since the concentration gradient is known, the diffusivity can then be calculated.

IV. RESULTS AND DISCUSSIONS

A. Determination of Diffusion Coefficient

Stefan's law :

$N_A = - D\, dC_AC_T / dy\, C_B$

Modifying Stefan's law we get the following equation,

$N_A = DC_T(C_{A1} - C_{A2}) / (y_2 - y_1)\, C_{Bm}$

From the above equation the rate of mass transfer is given by:

$N_A = DC_AC_T / L\, C_{Bm}$

Where C_A is the saturation concentration at the interface and L is the effective distance through which mass transfer is taking place. Considering the evaporation of the liquid:

$N_A = \rho_L\, dL / M\, dt$

Where ρ_L is the density of the liquid.

$\rho_L\, dL / M\, dt = DC_AC_T / L\, C_{Bm}$

integrating and putting $L = L_0$ at $t = 0$:

$L^2 - L^2_0 = 2MDC_AC_T\, t / \rho_L\, C_{Bm}$

L_0 will not be measured accurately nor is the effective distance for diffusion, L, at time t.

Accurate values of ($L - L_0$) are available, however, and hence :

$(L - L_0)(L - L_0 + 2L_0) = 2MDC_AC_T\, t / \rho_L\, C_{Bm}$

OR $\quad t/(L - L_0) = \rho_L\, C_{Bm}(L - L_0) / 2MDC_AC_T + \rho_L\, C_{Bm}\, L_0 / MDC_AC_T$

If 's' is the slope of a plot of $t / (L - L_0)$ against $(L - L_0)$, then

$s = \rho_L\, C_{Bm} / 2MDC_AC_T$ (or)

$D = \rho_L\, C_{Bm} / 2MC_AC_T\, s$

Table I. Effect of flowrate of

Time(min)	**Volume (m^3)**	**Time / Volume (min/m^3)**
0	0.00	0.00
20	0.20	100.00
65	0.50	130.00
80	0.60	133.33
95	0.70	135.71
110	0.80	137.50
125	0.90	138.89
145	1.00	145.00
185	1.20	154.17
225	1.40	160.71
245	1.45	168.97
300	1.50	200.00
320	1.60	200.00
350	1.75	200.00

Air(0 lph) on Diffusivity of Hexane at 32°C and 101.3 kPa

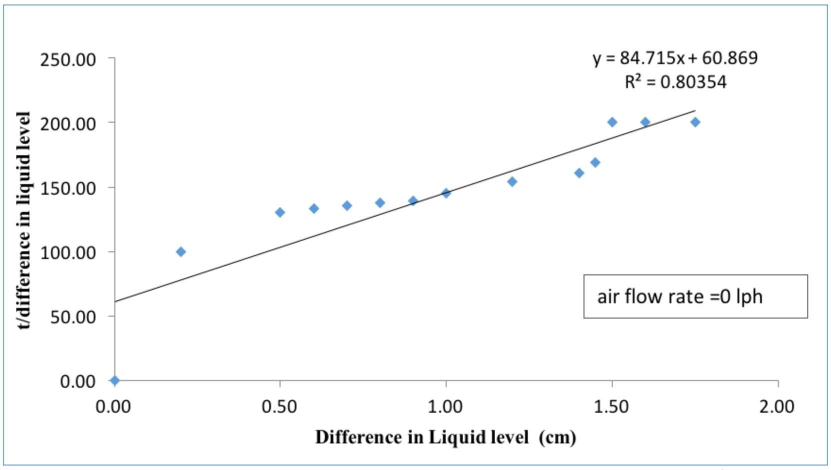


Table II. Effect of flowrate of air(2 lph) on Diffusivity of Hexane at 32°C and 101.3 kPa

Time(min)	Volume(m^3)	Time/Volume (min/m^3)
0	0.00	0.00
60	0.90	66.67
100	1.20	83.33
125	1.30	96.15
130	1.30	100.00
175	1.60	109.38
295	2.00	147.50
340	2.30	147.83
390	2.45	159.18

Fig.2. Effect of flowrate of air(2 lph) on Diffusivity of Hexane at 32°C and 101.3 kPa

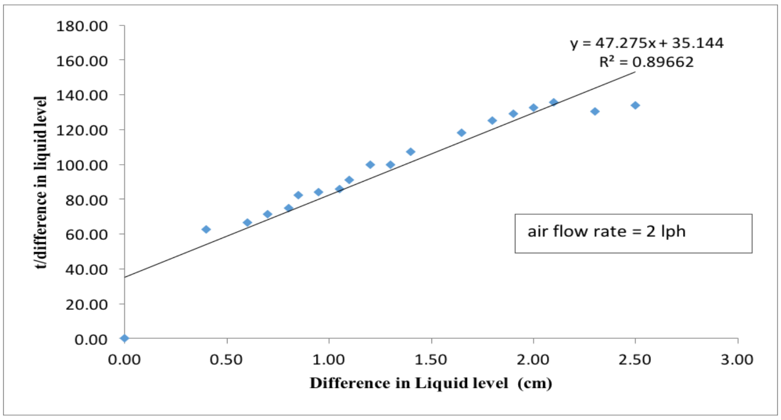


Table III. Effect of flowrate of air(3 lph) on Diffusivity of Hexane at 32°C and 101.3 kPa

Time (min)	Volume (m^3)	Time/Volume(min/m^3)
0	0.00	0.00
23	0.45	51.11
33	0.50	66.00
53	0.70	75.71
73	0.90	81.11
83	1.00	83.00
103	1.20	85.83

123	**1.30**	**94.62**
138	**1.40**	**98.57**
158	**1.50**	**105.33**
178	**1.60**	**111.25**
193	**1.70**	**113.53**
223	**1.90**	**117.37**
243	**1.95**	**124.62**
288	**2.10**	**137.14**
308	**2.25**	**136.89**
368	**2.40**	**153.33**

Fig.3. Effect of flowrate of air(3 lph) on Diffusivity of Hexane at 32°C and 101.3 kPa

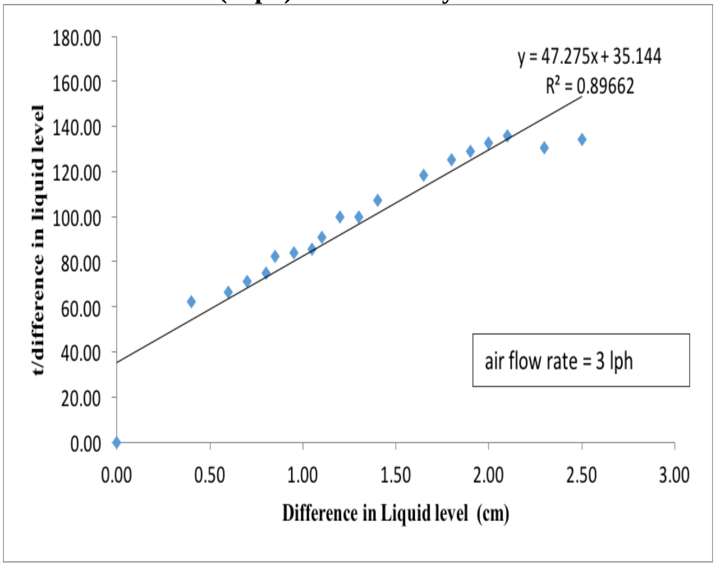


Table IV. Effect of flowrate of air(4 lph) on Diffusivity of Hexane at 32°C and 101.3 kPa

Time (min)	**Volume (m^3)**	**Time/Volume (min/m^3)**
0	0.00	0.00
15	0.20	75.00
45	0.40	112.50
65	0.60	108.33
75	0.70	107.14
85	0.80	106.25
100	1.00	100.00
125	1.10	113.64
145	1.20	120.83
175	1.50	116.67
245	1.75	140.00
270	1.85	145.95

Fig.4. Effect of flowrate of air(4 lph) on Diffusivity of Hexane at 32°C and 101.3 kPa

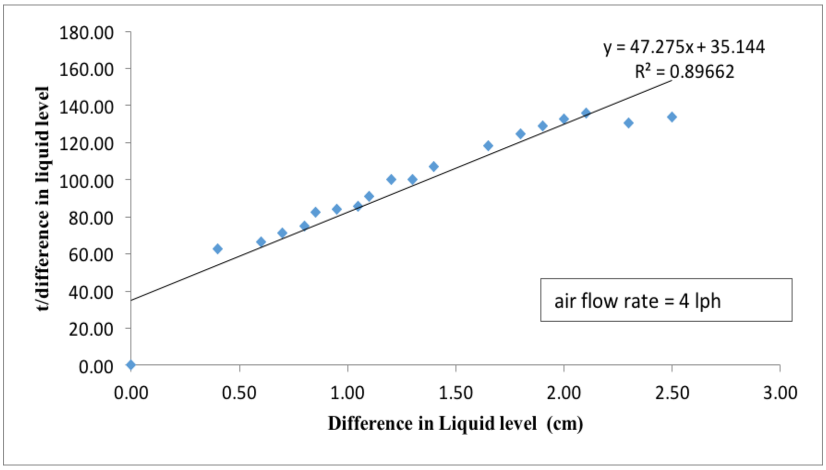


Table V. Effect of flowrate of air(5 lph) on Diffusivity of Hexane at 32°C and 101.3 kPa

Time(min)	Volume(m^3)	Time/Volume (min/m^3)
0	0.00	0.00
45	0.60	75.00
70	0.80	87.50
95	1.00	95.00
105	1.10	95.45
115	1.15	100.00
140	1.30	107.69
170	1.45	117.24
180	1.50	120.00
210	1.60	131.25
225	1.65	136.36
245	1.80	136.11
290	2.00	145.00
330	2.20	150.00

Fig.5. Effect of flowrate of air(5 lph) on Diffusivity of Hexane at 32°C and 101.3 kPa

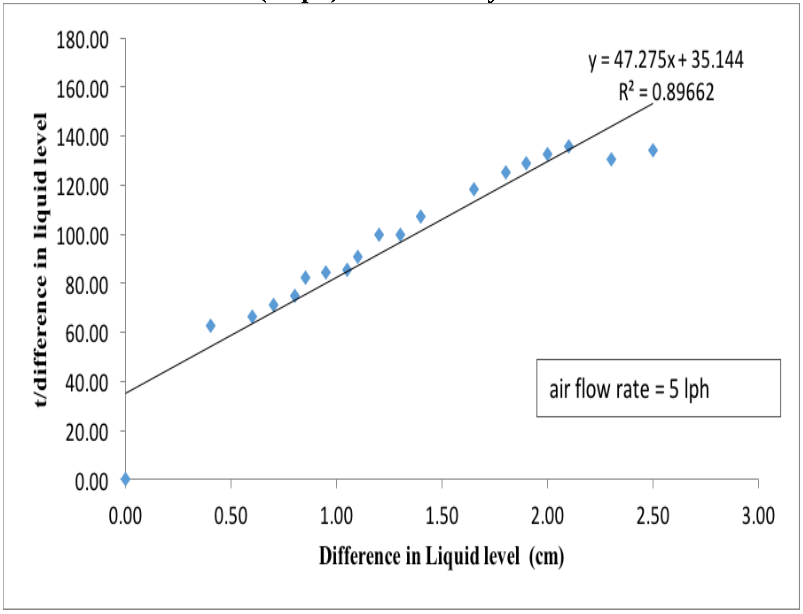


Table VI. Effect of flowrate of air(6 lph) on Diffusivity of Hexane at 32°C and 101.3 kPa

Time (min)	Volume (m^3)	Time/Volume (min/m^3)
0	0.00	0.00
10	0.20	50.00
20	0.35	57.14
25	0.40	62.50
35	0.50	70.00
45	0.60	75.00
60	0.70	85.71
70	0.80	87.50
75	0.95	78.95
85	1.00	85.00
95	1.10	86.36
110	1.15	95.65
120	1.20	100.00
130	1.30	100.00
150	1.40	107.14
175	1.60	109.38
205	1.70	120.59
230	1.80	127.78
250	1.90	131.58
275	2.00	137.50
295	2.10	140.48
310	2.20	140.91
350	2.30	152.17
360	2.40	150.00

Fig.6. Effect of flowrate of air(6 lph) on Diffusivity of Hexane at 32°C and 101.3 kPa

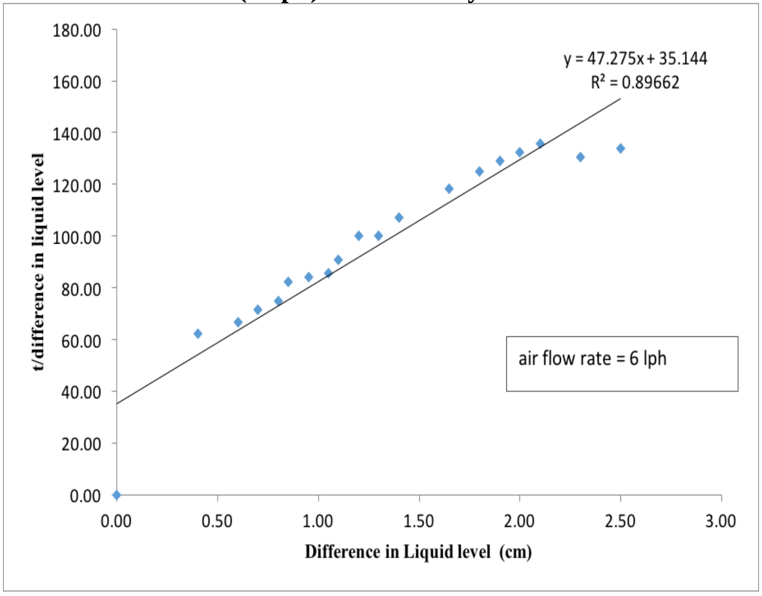


Table VII. Effect of flowrate of air(7 lph) on Diffusivity of Hexane at 32°C and 101.3 kPa

Time (min)	Volume (m^3)	Time/Volume (min/m^3)
0	0.00	0.00
30	0.40	75.00
40	0.50	80.00
50	0.60	83.33
60	0.70	85.71
70	0.80	87.50
95	1.00	95.00
110	1.10	100.00
130	1.25	104.00
140	1.30	107.69
150	1.40	107.14
170	1.70	100.00
185	1.55	119.35
200	1.60	125.00
215	1.70	126.47
230	1.80	127.78
260	1.90	136.84
275	2.00	137.50
327	2.10	155.71
350	2.30	152.17
370	2.45	151.02
430	2.60	165.38

Fig.7. Effect of flowrate of air(7 lph) on Diffusivity of Hexane at 32°C and 101.3 kPa

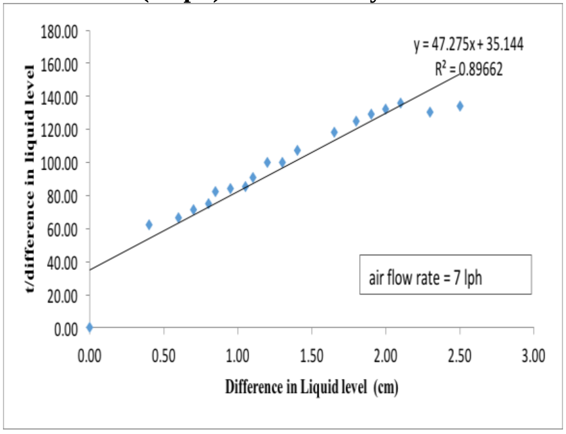


Table VIII. Effect of flowrate of air(8 lph) on Diffusivity of Hexane at 32°C and 101.3 kPa

Time(min)	Volume(m^3)	Time/Volume (min/m^3)
0	0.00	0.00
25	0.40	62.50
40	0.60	66.67
50	0.70	71.43
60	0.80	75.00
70	0.85	82.35
80	0.95	84.21
90	1.05	85.71
100	1.10	90.91

120	1.20	100.00
130	1.30	100.00
150	1.40	107.14
195	1.65	118.18
225	1.80	125.00
245	1.90	128.95
265	2.00	132.50
285	2.10	135.71
300	2.30	130.43
335	2.50	134.00

Fig.8. Effect of flowrate of air(8 lph) on Diffusivity of Hexane at 32^{o}C and 101.3 kPa

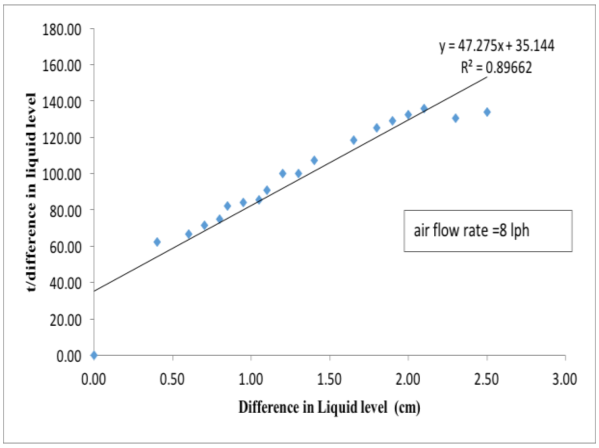


Table IX. Effect of flowrate of air(9 lph) on Diffusivity of Hexane at 32^{o}C and 101.3 kPa

Time (min)	**Volume (m^3)**	**Time/Volume (min/m^3)**
0	0.00	0.00
45	0.50	90.00
60	0.80	75.00
90	1.00	90.00
110	1.10	100.00
130	1.30	100.00
150	1.40	107.14
190	1.60	118.75
240	1.95	123.08
265	2.00	132.50
280	2.05	136.59
300	2.10	142.86
315	2.20	143.18
330	2.25	146.67
345	2.30	150.00
360	2.35	153.19
405	2.45	165.31

Fig.9. Effect of flowrate of air(9 lph) on Diffusivity of Hexane at 32^{o}C and 101.3 kPa

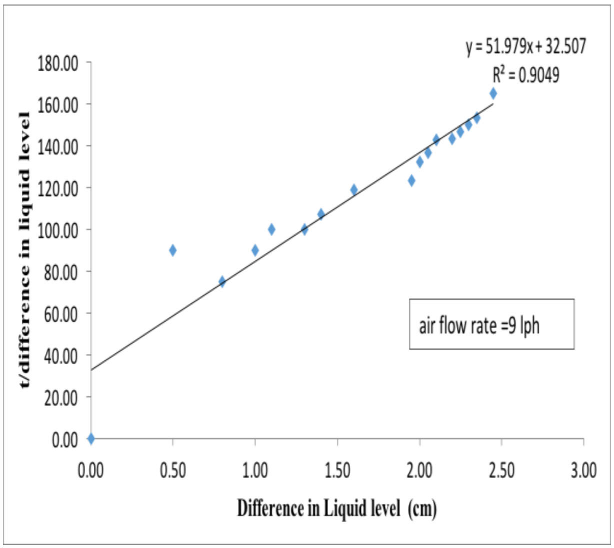


Table X. Effect of flowrate of air(10 lph)on Diffusivity of Hexane at 32°C and 101.3kPa

Time (min)	Volume (m^3)	Time/Volume (min/m^3)
0	0.00	0.00
20	0.40	50.00
50	0.70	71.43
60	0.80	75.00
70	1.00	70.00
80	1.10	72.73
95	1.20	79.17
115	1.35	85.19
140	1.50	93.33
225	2.00	112.50
260	2.25	115.56
285	2.40	118.75
325	2.60	125.00

Fig.10. Effect of flowrate of air(10 lph) on Diffusivity of Hexane at 32°C and 101.3 kPa

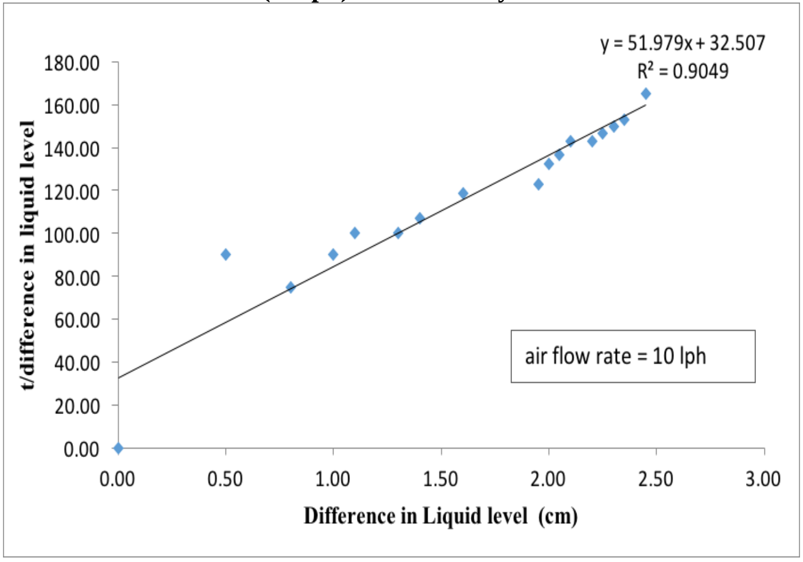


Table XI. Effect of Air flowrate on Diffusivity of Hexane

S.NO	Flow rate of air (lph)	Diffusivity (m^2/s)
1	0.0	3.20E-06
2	2.0	6.02E-06
3	3.0	7.47E-06
4	4.0	7.72E-06
5	5.0	7.76E-06
6	6.0	7.83E-06
7	7.0	8.09E-06
8	8.0	8.24E-06
9	9.0	9.23E-06
10	10.0	9.75E-06

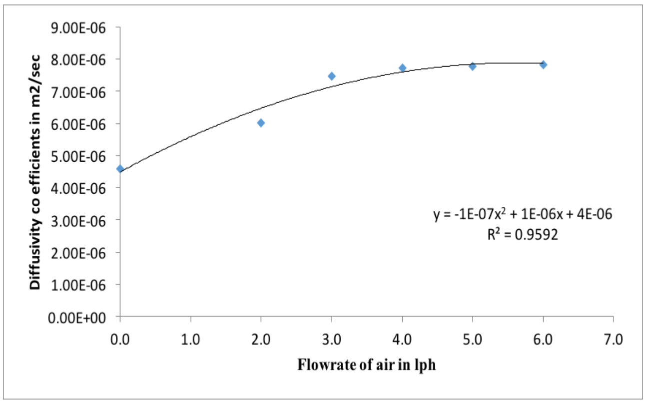


Fig.11. Effect of Air flowrate on Diffusivity of Hexane

V. PROPOSED MODEL EQUATION

Proposed Model equation (Modified Gilliland's) :

$\mathbf{D_f / D_0 = k_1 Q + c}$

where

D_f = Diffusivity at different flowrate of air (m^2/s)

D_0 = Diffusivity at no flowrate of air (calculated from Gilliland equation)

$D_0 = 4.3 \times 10^{-4} T^{1.5} \sqrt{((1/M_A)+(1/M_B))} / P(V_A^{1/3} + V_B^{1/3})^2$

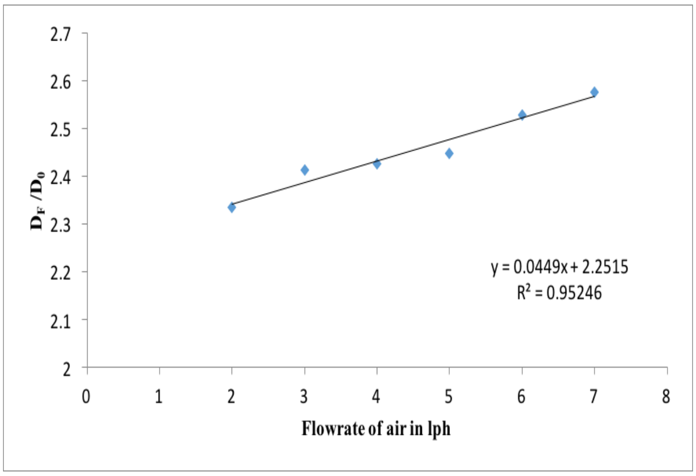
Q = flowrate of air (lph)

k_1 and c = constant

Table XII. Effect of Diffusivity ratios with air flowrate

Flowrate (lph)	Diffusivity (m^2/s)	D_f/D_0
3	**7.47E-06**	**2.33**
4	**7.72E-06**	**2.41**
5	**7.76E-06**	**2.42**
6	**7.83E-06**	**2.44**
7	**8.09E-06**	**2.52**
8	**8.24E-06**	**2.57**

Fig.12. Effect of Diffusivity ratios with air flowrate



From the graph: $K_1 = 0.044$, $C = 2.251$

Table XIII. Comparison of experimental values of Diffusion coefficient by the present proposed model

D_0	Q	k_1 (graph)	C (graph)	D_f (equation)	D_f(exp)	% error
3.20E-06	3	0.044	2.251	7.6256E-06	7.47E-06	2.082999
3.20E-06	4	0.044	2.251	7.7664E-06	7.72E-06	0.601036
3.20E-06	5	0.044	2.251	7.9072E-06	7.76E-06	1.896907
3.20E-06	6	0.044	2.251	8.048E-06	7.83E-06	2.784163
3.20E-06	7	0.044	2.251	8.1888E-06	8.09E-06	1.221261
3.20E-06	8	0.044	2.251	8.3296E-06	8.24E-06	1.087379

VI. CONCLUSIONS

The following conclusions are drawn based on the results of the present investigation involving the effect of flowrate of air on diffusivity of Hexane at 32^0C and 101.3 kPa. The diffusivity model equation was derived for predicting the diffusivity Coefficient of Hexane vapour from Stefan’s equation. The proposed model equation deriverd from Gilliland’s equation is $\mathbf{D_f / D_0 = k_1 Q + c}$ **.** The parameter ‘k_1’ and ‘c’ remains nearly constant for the range of flowrate of air (0 – 10 lph). The value of ‘k’ and ‘c’ are 0.044 and 2.251 respectively. The experimental Diffusivity value at different flowrates are compared with the proposed model and the percentage error is around 2% in the flowrate of 0 – 10 lph. The results indicates that the proposed model can be used to predict the Diffusion coefficient at different flowrates of air at 32^0C and 101.3 kPa.

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