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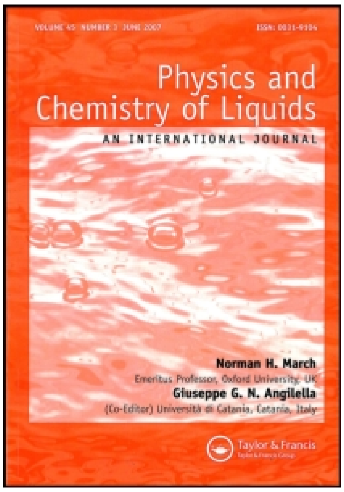

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Properties of methanol and acetone in dilute aqueous solutions

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Properties of methanol and acetone in dilute aqueous solutions

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The apparent molar volume Φ_v of methanol and acetone in dilute aqueous solutions was determined from density measurement at 293 K. The limiting apparent molar volumes Φ_v^0 at infinite dilution of these organic liquids were calculated by Masson's equation. The variation of Φ_v with concentration was also discussed in terms of liquid structure. Viscosity coefficient A and B for both liquids were determined by Jone–Dole equation. This liquid exhibits strong solute–solvent interactions. Grunberg–Nissan constant d_{12} was also calculated from the viscosity data, which indicates the formation of associating clusters in the solution. A new modified Jone–Dole equation was also applied with the experimental results, which was applied for both liquids as solute in dilute aqueous solutions.

Keywords: apparent molar volume; modified Jone–Dole equation; Grunberg–Nissan constant

1. Introduction

In solution chemistry, some information on the important water–solute interaction is forthcoming from the knowledge of the apparent molar volume. In fact, in highly dilute aqueous solutions, the two properties apparent molar volume and viscosity are needed to build up such knowledge of inter-molecular interactions. An understanding of structural and dynamics of water molecules in different environments is also important in such aqueous solutions. In this study, our focus is on dilute aqueous solutions, which can give general features of solution structures. Hence viscometric and volumetric studies of methanol and acetone were carried out in highly dilute aqueous media.

Alcohols may be considered as derivatives of water in which one hydrogen atom is replaced by an alkyl group. The presence of an unshared electron in the oxygen outer shell imparts an important feature in the structure of alcohols. Alcohols become water-like in their physical properties as their ratio of hydroxyl in the C–H bond is increased. Methanol is the first member of class of alcohols, which is widely used as solvent and is a principal raw material in chemical industry. The nature of acetone can be best understood in terms

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of partial negative charge of the acetone dipole being concentrated on a single atom, the oxygen atom, compared to the positive charge being spread over the carbonyl groups, which could hinder the close approach of an anion. For both the solvent methanol and acetone the solvation of cation is greatest [1].

Volumetric properties of alcohols have been extensively studied [2–5]. In these studies it has been concluded that the lower alcohols like methanol get dissolved in the cavities available in the water structure; Kaulgud *et al.* [6] observed the change in the slopes of the plots C (molar concentration) *versus* Φ_v (at nearly $2\,\mathrm{mol\,dm^{-3}}$), which indicates that the structural properties of aqueous methanol solutions and the nature of the interactions present in it still require investigation in lower concentration ranges. Secondly, aqueous methanol solution has been used by various workers in conductometric or viscometric studies for different solutes as a solvent [7–11] and results were analysed in terms of solute–solvent/solute–solute/solvent–solvent interactions without considering the direct impact of co-solute methanol towards the behaviours of solvent. Similar studies were also reported by different workers with acetone in place of methanol [12–17] in conductometric or viscometric studies.

A survey of the literature showed that the organic liquids have been used in certain compositions with water as solvents for other solutes. The most widely used organic liquids are 1,4-dioxane [18,19], dimethyl sulphoxide [20,21], dimethyl formamide [22,23], acetonitrile [9,24–26], ethanol [27], 2-methoxy ethanol [28], etc. Water–organic liquid systems have been also used for volumetric studies during interaction studies of solutions [29–33].

2. Experimental

Deionised water was distilled twice with a small quantity of alkaline potassium permanganate. Finally, water was distilled in Corning glass apparatus with sulphuric acid. The specific conductance of prepared distilled water for the study was of the order $2 \times 10^{-6}\,\Omega\,\mathrm{cm}^{-1}$. The methanol and acetone grade BDH (British Drug House Chemicals, Ltd.) were used and solutions were prepared by dissolving the calculated volume of the liquids for desired strength in distilled water. Density measurements of solutions were made using pyknometer, having a capacity of $15\,\mathrm{cm}^3$. The pyknometer was calibrated with distilled water having densities $0.9972\,\mathrm{gm\,cm^{-3}}$ and $0.9942\,\mathrm{gm\,cm^{-3}}$ at 298 K and 308 K, respectively. A thermostatically controlled water bath, capable of maintaining the temperature constant ±0.01°C, was used during the studies. All weighings were made on a single-pan digital balance, Citizen, made with an accuracy of 0.0001 gm. The density values were reproducible $\pm 0.0002\,\mathrm{gm\,cm^{-3}}$. Viscosities were measured using an Ubbelohode-type capillary viscometer (PSL, England), which was calibrated with doubly distilled water by measuring efflux time using a stop watch with a resolution of 0.01 s. The average of at least four readings reproducible within an accuracy of 0.01 s was taken as the final efflux time.

3. Viscometric studies

For both the organic liquids, namely methanol and acetone, the values of η/η_0 at different concentrations of liquid in aqueous solution were observed and are given in Table 1.

Table 1. Variation of η/η_0 with concentration of acetone and methanol at temperature 293 K.

	Acetone			Methanol	
S. no.	Concentration (mol dm^{-3})	η/η_0	S. no.	Concentration (mol dm^{-3})	η/η_0
1	0.000		1	0.000	–
2	0.086	1.011	2	0.156	1.020
3	0.172	1.022	3	0.311	1.038
4	0.257	1.033	4	0.466	1.053
5	0.343	1.045	5	0.621	1.068
6	0.426	1.057	6	0.776	1.083
7	0.514	1.070	7	0.930	1.098
8	0.599	1.083	8	1.084	1.114
9	0.684	1.097	–	–	–
10	0.769	1.110	–	–	–
11	0.854	1.123	–	–	–

Notes: η = viscosity of solution. η_0 = viscosity of water.

Table 2. Values of A and B coefficient of the Jone–Dole equation for aqueous methanol and aqueous acetone at temperature 293 K.

	Acetone			Methanol	
S. no.	$\sqrt{C}$	$(\eta/\eta_0 - 1)/\sqrt{C}$	S. no.	$\sqrt{C}$	$(\eta/\eta_0 - 1)/\sqrt{C}$
1	0.2932	0.0375	1	0.3949	0.0500
2	0.4147	0.0530	2	0.5577	0.0681
3	0.5069	0.0651	3	0.6826	0.0776
4	0.5856	0.0768	4	0.7880	0.0862
5	0.6526	0.0871	5	0.8809	0.0942
6	0.7169	0.0976	6	0.9643	0.1016
7	0.7739	0.1060	7	1.0411	0.1095
8	0.8270	0.1173	–	–	–
9	0.8769	0.1254	–	–	–
10	0.9241	0.1331	–	–	–

Notes: $A = -0.011(\mathrm{dm}^{3/2}\,\mathrm{mol}^{-1/2})$. $A = 0.017(\mathrm{dm}^{3/2}\,\mathrm{mol}^{-1/2})$. $B = 0.153$ $(\mathrm{dm}^3\,\mathrm{mol}^{-1})$. $B = 0.088$ $(\mathrm{dm}^3\,\mathrm{mol}^{-1})$. Linearity = 0.995. Linearity = 0.997. C = molar concentration (mol dm^{-3}).

The values of η/η_0 obtained for both liquids (solute) at different concentrations were utilised for the determination of inter-molecular interactions in aqueous solution. The interaction parameters were obtained with the help of Jone–Dole Equation (1).

$$\frac{(\eta/\eta_0 - 1)}{\sqrt{C}} = A + B\sqrt{C}, \tag{1}$$

where A and B are the coefficients for the ion–ion and ion–solvent interactions, respectively, and C is the molar concentration. The value of A and B were obtained from the intercept and slope of the linear plots of $(\eta/\eta_0 - 1)\sqrt{C}$. The obtained values of intercept and slope for both the solute methanol and acetone are given in Table 2

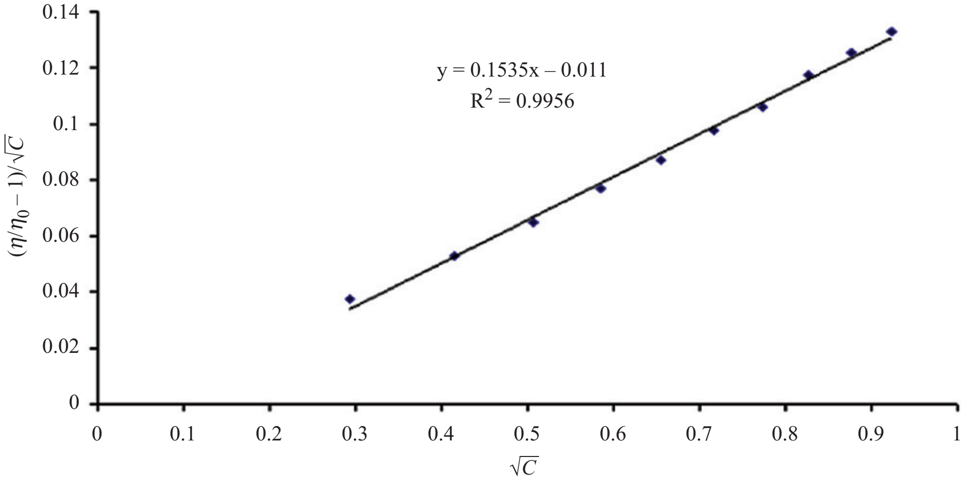

Figure 1. Jone–Dole plot $\sqrt{C}$ *versus* $(\eta/\eta_0 - 1)/\sqrt{C}$.

Table 3. Values of A_x and B_x coefficient of the modified Jone–Dole equation for aqueous methanol and aqueous acetone at temperature 293 K.

	Acetone			Methanol	
S. no.	$\sqrt{n_s/n_w}$	$(\eta/\eta_0 - 1)/\sqrt{n_s/n_w}$	S. no.	$\sqrt{n_s/n_w}$	$(\eta/\eta_0 - 1)/\sqrt{n_s/n_w}$
1	0.0395	0.2785	1	0.0532	0.3760
2	0.0560	0.3928	2	0.0753	0.5047
3	0.0688	0.4792	3	0.0924	0.5732
4	0.0795	0.5657	4	0.1071	0.6350
5	0.0891	0.6397	5	0.1200	0.6913
6	0.0979	0.7146	6	0.1318	0.7434
7	0.1060	0.7826	7	0.1427	0.7985
8	0.1126	0.8614	–	–	–
9	0.1208	0.9000	–	–	–
10	0.1278	1.017	–	–	–

Notes: $A_x = -0.059$. $A_x = 0.145$. $B_x = 7.985$. $B_x = 4.569$. Linearity = 0.988. Linearity = 0.996. C = molar concentration (mol dm^{-3}).

with tabulated data. The linear coefficients were calculated for both the sets of data. The calculations were carried out with the help of the computer Jone–Dole equation and the validity of the Jone–Dole equation was checked graphically. The representative graph in the case of methanol is given in Figure 1.

For both the solutes using the values of concentration, n_s and n_w were calculated and the data obtained are recorded in Table 3. From the linear plots of $\sqrt{n_s}/n_w$ *versus* $\eta/\eta_0/\sqrt{n_s/n_w}$, A_x and B_x values for acetone were evaluated from the intercept and slope, respectively (Figure 2), in order to test the validity of modified Jone–Dole Equation (2).

$$\frac{(\eta/\eta_0) - 1}{\sqrt{n_s/n_w}} = A_x + B_x\sqrt{\frac{n_s}{n_w}}. \tag{2}$$

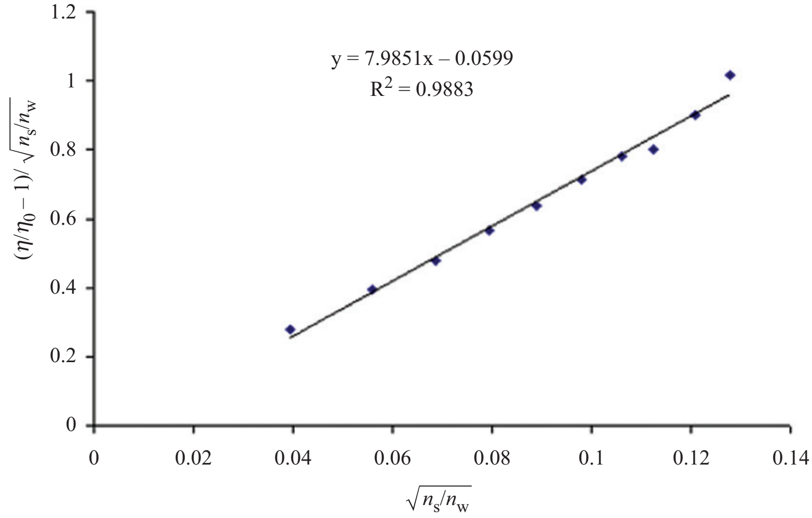

Figure 2. Modified Jone–Dole plot $\sqrt{n_s/n_w}$ *versus* $(\eta/\eta_c - 1)/\sqrt{n_s/n_w}$.

The linearity of the plot was also calculated with the help of computer using regression analysis. The values of linearity are given at the bottom of Table 3.

The values of viscosities of aqueous liquid solutions under study have been calculated to the Grunberg and Nissan [34] logarithmic relation Equation (3) between the viscosity of liquid solutions and pure components.

$$\ln = X_1 \ln \eta_1 + X_1 X_2 d_{12}, \tag{3}$$

where d_{12} is a constant which is regarded as the measure to the strength of interactions between solute and solvent molecules. X represents the mole fraction of the components η_1 and η_2 represents the viscosity of the components 1 and 2 in pure state and η is the viscosity of solutions. The d_{12} values are given in Tables 4 and 5.

4. Results and discussion

4.1. *Volumetric studies*

Though we have undertaken the conductometric and viscometric studies as our subject of investigation, in the case of organic liquids, the conductance of solution remained nearly constant with the change of concentration. It was therefore thought to undertake the volumetric study of the solutions at the same concentration in order to obtain a comparative account of experimental results.

For both the organic liquids (solutes), namely methanol and acetone, the densities of aqueous solution at different concentrations were measured. The values of the densities obtained are listed in Table 6 at different concentrations.

The partial molar volume may be defined in change in volume at constant temperature and pressure, when one mole of solute is added to a large volume of solvent and is identical

Table 4. The values of d_{12} obtained by Grunberg–Nissan equation for acetone in water at 293 K.

S. no.	Mole fraction X_1 of solute	Mole fraction X_2 of solvent	Viscosity of solution	d_{12} Constant of Grumberg–Nissan equation
1	0.00156	0.99843	1.011	8.1950
2	0.00313	0.99687	1.022	8.1405
3	0.00469	0.99531	1.033	5.1135
4	0.00629	0.99371	1.045	8.2075
5	0.00784	0.99215	1.057	8.2886
6	0.00950	0.99049	1.070	8.3546
7	0.01112	0.98887	1.083	8.4189
8	0.01275	0.98724	1.097	8.5237
9	0.01440	0.98559	1.110	8.5264
10	0.01607	0.98393	1.123	8.5134

Table 5. The values of d_{12} obtained by Grunberg–Nissan equation for methanol in water at 293 K.

S. no.	Mole fraction X_1 of solute	Mole fraction X_2 of solvent	Viscosity of solution	d_{12} Constant of Grumber–Nissan equation
1	0.00282	0.99717	1.020	7.5752
2	0.00564	0.99435	1.038	7.1874
3	0.00848	0.99152	1.053	6.5580
4	0.01134	0.98866	1.068	6.4042
5	0.01421	0.98579	1.083	6.2336
6	0.01708	0.98291	1.098	6.1083
7	0.01997	0.98002	1.114	6.0545

Table 6. The variation of densities with concentration of acetone and methanol at temperature 293 K.

	Acetone			Methanol	
S. no.	Concentration (mol dm^{-3})	Density (gm cm^{-3})	S. no.	Concentration (mol dm^{-3})	Density (gm cm^{-3})
1	0.000	0.9982	1	0.000	0.9982
2	0.086	0.9957	2	0.156	0.9973
3	0.172	0.9968	3	0.311	0.9964
4	0.257	0.9961	4	0.466	0.9956
5	0.343	0.9954	5	0.621	0.9947
6	0.428	0.9947	6	0.776	0.9938
7	0.514	0.9940	7	0.930	0.9930
8	0.599	0.9933	8	1.084	0.9921
9	0.684	0.9926	–	–	–
10	0.769	0.9919	–	–	–
11	0.854	0.9912	–	–	–

Table 7. Values Φ_v of the Masson equation for aqueous methanol and aqueous acetone at temperature 293 K.

	Acetone			Methanol	
S. no.	$\sqrt{C}$	Φ_v	S. no.	$\sqrt{C}$	Φ_v
1	0.2932	66.3042	1	0.156	37.8666
2	0.4147	66.3042	2	0.311	37.8852
3	0.5069	66.3359	3	0.466	37.6764
4	0.5856	66.3279	4	0.621	37.7332
5	0.6542	66.3422	5	0.776	37.7673
6	0.7169	66.3359	6	0.930	37.6884
7	0.7739	66.3450	7	1.084	37.7244
8	0.8270	66.3519	–	–	–
9	0.8769	66.3572	–	–	–
10	0.9241	66.3615	–	–	–

Notes: $\Phi_v^0 = 66.27$. $\Phi_v^0 = 37.86$. $S_v = 0.091$. $S_v = -0.168$. C = molar concentration (mol dm^{-3}).

to the apparent molar volume at infinite dilution. The apparent molar volume Φ_v is given by the standard expression [35] in Equation (4).

$$\Phi_v = \frac{M}{d_0} + 1000(d - d_0), \tag{4}$$

where M is the molar mass of solute, d and d_0 are densities of solution and pure solvent, respectively, and C is the molarity of solute.

For both the organic liquids, methanol and acetone, Φ_v values were calculated at different concentrations (for methanol, 0.10–1 mol dm^{-3}) and (for acetone, 0.2–4 mol dm^{-3}). The values obtained are given in Table 7. The Φ_v *versus* $\sqrt{C}$ curve is given as an illustrative example for methanol in Figure 3. These plots are straight lines and show that Masson's Equation (5) is valid within the concentration range studied.

$$\Phi_v = \Phi_v^0 + S_v\sqrt{C}, \tag{5}$$

where Φ_v^0 is the apparent molar volume at infinite dilution (intercept) and S_v is the slope of the plot and C is the molar concentration.

Alcohols have physical and chemical properties of similar to water and are capable of forming the hydrogen bond. Alcohols having lower molecular weight are soluble in water and can act as an acid. The equilibrium in aqueous solution is given below. The value of pka = 20 for acetone is higher than water and alcohols of lower molecular weight.

$$H_2OH^+ + OH^-(\text{pka} = 15.8), \tag{6}$$

$$R{-}O{-}HR{-}O{-}H + H^+(\text{pka} = 18.0), \tag{7}$$

$$R{-}O^- + H_2OR{-}O{-}H + OH^-. \tag{8}$$

Volumetric properties of aqueous solutions of alcohols have been studied [2]. It was observed that there was a loss of volume of alcohols on dissolution in water. Keeping in the mind the peculiar behaviour of alcohols, we have investigated the volumetric and viscometric properties of methanol and acetone in very dilute aqueous solutions.

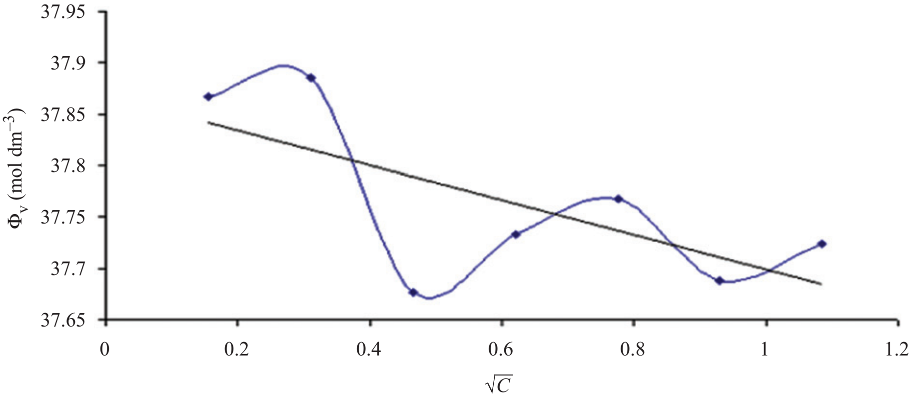

Figure 3. Masson plot $\sqrt{C}$ *versus* Φ_v.

The apparent molar volumes Φ_v of acetone and methanol calculated from density data are presented in Table 7. Results for methanol are nearly in agreement with the reported [6] values. The values of apparent molar volume Φ_v were used for applicability of Masson's Equation (5). The plot of $C^{0.5}$ *versus* Φ_v was not linear. The variation of Φ_v for methanol and acetone with $\sqrt{C}$ followed a cyclic trend, having maxima and minima with an increase in $\sqrt{C}$, which attends to zero at higher values of $\sqrt{C}$. These types of trend resemble the X-ray refraction data on liquids in radial distribution function, which is a measure of average particle density as a function of distance from an arbitrary origin. The fluctuations at lower concentrations of alcohols were also observed by Kaulgud [6].

The B-coefficient for methanol and acetone calculated with the help of the Jone–Dole equation were positive, suggesting that both are 'structure makers'. The negative value of S_v for the methanol and very low value of S_v for acetone justify the negligible ion–ion interaction in aqueous solution. The higher value of S_v for acetone may due to its tendency to form dipole. B-coefficients are also known as a measure of the order or disorder introduced in the solvent structure due to the ion or dipole formed by the solute, which suggest that strong solute–solvent interactions were present in the dilute solutions. The value of A is very small for methanol and negative for acetone, which confirms the conclusion drawn from volumetric results that in the study ion–ion or solute–solute interactions were very weak [36]. The viscosity data obtained in the study were in the good agreement with the modified Jone–Dole equation used in case of sulphuric acid. In this case, the ratio of B_x/B was 51.5 for methanol and 52.0 for acetone, which is nearly the same as observed in the case of sulphuric acid – 53.2.

The value of the Grunberg–Nissan constant d_{12} in the case of acetone decreases initially with an increase of the concentration of acetone and reaches a minimum value at mole fraction 0.0047; after that its value increases continuously. This indicates that the strength of interaction between solute and solvent molecule decreases due to the mutual interactions of solute at lower concentrations. The strength of solute–solvent interaction increased with increasing the concentration of acetone. In the case of methanol, the value of the Grunberg–Nissan constant followed a continuous decrease with increasing concentration of methanol. This indicates the decrease in strength of solute–solvent interactions with the increase in alcohol concentration. It is suggested that the molecules of alcohol form associating clusters through inter-molecular hydrogen bonding [37] due to the presence of hydroxyl group which results in a decrease in the Grunberg–Nissan constant.

It has been concluded that both the solutes methanol and acetone were accumulated in the cavities of water molecules. Ultimately, these results in the loss of volume or decrease in density on dissolution in water at lower concentration, the Φ_v value attaining maxima due to the effect of solute–solvent interactions, which leads to a regular trend at higher concentration of solute as is evident from the variation of the Grunberg–Nissan constant, methanol and acetone act as 'structure makers'.

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