Density and Viscosity study of Solvents, Mixture of Solvents with or without additives: A Mini Review

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Abstract

In the present review article we report here with the behavior of solvent, solvent mixture in presence and absence of additives with respect to their densities and viscosity. The paper also describe theories related with solvent behavior and volumetric study of solvents.

Key words: densities, viscosities, volumetric, solvent mixture.

Introduction

Volumetric measurements often yield valuable information regarding molecular interactions in the pure liquids as well as liquid mixtures. A familiar approach is the hypothesis that there is a direct correlation between the viscosity and the thermodynamic behavior of the solution. Very often it is essential to know the viscosity of the liquid mixtures to solve some practical problems.

The physical properties of a binary mixture such as viscosity and density are important, from practical and theoretical points of view to understand liquid theory. Their properties are extremely useful for the design of many types of transport (heat, mass, *etc.*) and process equipments in chemical industries and fluid flow. However, there seems to be no reliable theoretical method to estimate these viscosities. In addition many viscosity - composition curves are not simple functions of composition. Therefore viscosities of most systems can not be predicted from theory, but must be determined experimentally. From the survey of literature it indicates that partial molar volume and viscosity data give valuable information regarding solute-solvent and solute-solute interactions. Studies on viscosity of ionic solutions are of great help in characterizing the structure properties of solutions.

Poiseuille found that some salts increases the viscosity of water whereas other decreases the viscosity. Sprung appears to have been the first to measure viscosities of solution of $BaCl_2$ and found that it increased the viscosity of water. Arrhenius made some viscosity measurements on solutions and found that the change in viscosity caused by addition of a salt was roughly proportional to the concentration at low concentrations, but increased more rapidly than the concentration at moderate concentrations (1 normal). He proposed a new formula for the relationship between viscosity and concentration. *i.e* $\eta = A^c$, where, η is the relative viscosity compared to that of water at the same temperature and A is a constant for any given salt and temperature.

Applebey studied dilute solutions (c=0.00724). He made the suggestion that when a salt is dissolved in water there are two different effects on viscosity (1) a depolymerization of triple water molecules, $(H_2O)_3$, to form single molecules, which tend to diminish the viscosity, (2) an increase in viscosity due to the presence of the ions at the salt and the un-ionized molecules of the salt. The work of Merton on cesium nitrate appears to be more accurate and extensive study of a salt which diminishes the viscosity of water. Cox & Wolfenden from the study of the pair of electrolytes KCl-NaCl & KNO_3 - $NaNO_3$, have found that the difference in the values of 'B' of such pairs is connected with the difference in the individual character of the sphere of solvation of the cation & anion. It has already been suggested that in a mixed solvent the number of molecules of each species constituting the sphere of solvation depends not only on the character of the ion, but also on the specific properties of both kind of molecules present in the solvent.

The value of 'B' for different electrolytes differs widely. The value of B could be determined by the lyotropic number and entropy of hydration. 'B' is likely to be dependent on the nature of solvation sphere of ion which in mixed solvents is bound to be formed by both the component of solvent system.

From the point of view of the free volume approach, it is not surprising that those systems that show large positive viscosity deviations, tend also to show large negative deviations from volume additivity. If it is true that free volume is necessary for flow, then shrinking on mixing, which would appear to reduce the free volume, should be associated with an increase in viscosity. Correct use of this argument requires data for all the solutions over a range of temperature, which are not available, for which the relative excess volume, defined as percentage was used by :

$$v_r^E = 100 \left(\frac{V - V_1 x_1 \quad V_2 x_2}{V_1 x_1 + V_2 x_2} \right)$$

$$= 100 \left(\frac{(x_2^{-1} - 1) M_1 / M_2 + 1}{\rho(x_2^{-1} - 1) M_1 / M_2 \rho_1 + 1 / \rho_2} - 1 \right)$$

(1)

where, V is the molar volume, ρ, M & x are density, molecular weight and mole fraction respectively and the subscripts 1 & 2 refer to the two components. Values of the (negative) excess volume are approximately proportional to the product of the mole fractions, $x_1 x_2$.

Robart A Staris[1] studied the viscosity of mixtures of water with a number of organic liquids and of some mixtures of two organic

liquids and analysed interms of its deviation from "ideality", expressed as

$$\ln H = \ln\eta - \left(x_1 \ln\eta_1 + x_2 \ln\eta_2 \right) \quad (2)$$

It is shown to represent approximately the behavior of the mixtures of two organic liquids. It fails with the water-organic mixtures. The viscosity behavior of dilute aqueous solutions of electrolytes has been studied in detail and numerous empirical relations have been reparted to explain the variation of viscosity with concentration. The relative viscosity, ηr, *i.e.* the viscosity of solution with respect to solvent, is generally used in most of the empirical relations. For dilute solutions ($c \leq 0.1$ M), the theoretical relation :

$$\eta_r = 1 + 2.5\phi \quad (3)$$

Where ϕ denotes the volume fraction and is equal to cV, V being the molar volume of electrolyte in solution. On the other hand the Jones-Dole semi-empirical relation gives :

$$\eta_r = 1 + A\sqrt{C} + B\,c \quad (4)$$

the constant A is identified as ion-ion interaction and the constant B is identified as viscosity B-coefficient dealing with ion-solvent interactions in solution. These two relations have undergone considerable modifications for concentrated solutios ($c \geq 0.1$M). In many case it has been seen that the Jones-Dole equation is the most appropriate equation for calculating the viscosity of dilute aqueous solutions of electrolytes. This equation is used for concentrated solutions in the following form[2].

$$\eta_r = 1 + B\,c \quad (5)$$

Unlike the viscosity behavior of dilute solutions, representation of viscosity at concentrations > 0.1M by one general equation becomes difficult. Vand, Thomas and Moulik[3] have extended the limiting extension of Einstein to higher concentrations and have advanced some useful relations which have been tested for their general validity in higher concentrations of limited range by Moulik. Based on the Eyring theory of absolute rate for viscous flow of liquids, Goldsack and Franchetto and Behera[4] successfully explained the variation of viscosity with concentration for aqueous solutions of electrolytes of alkali metal halides and non-electrolytes respectively. B.Sahu and B. Behera[5], developed a general equation to explain the concentration dependence of viscosity of concentrated aqueous solutions of electrolytes of the form

$$\eta_r = 1 + 2.5\phi + k_1\phi^2 + k_2\phi^3 + k_3\phi^4 \quad (6)$$

Where k's are constant

The viscosity B-coefficient of Jones-Dole equation is known to be sensitive to the nature of solute-solvent interactions *i.e.* it is related to the modification of micro-viscosity of the solvent in the neighbor-hood of the solute particle as compared to the bulk viscosity. A structure-making solute is expected to have a positive B-coefficient in the given solvent where as a structure-breaker may have a less positive or altogether negative B-coefficient[6]. However,a large solute may always posses a positive B-coefficient (irrespective of the nature of its interaction with the solvent) due to the "obstruction effect", *i.e.* a bending of the solvent streamlines round a large solute particle. The sign and magnitude of the temperature coefficient, dB/dT, rather than B, therefore often provides a better index of solute-solvent interactions. This observation is attributed to an increased thermal mobility of water molecules at higher temperature which causes the structure promotion by a solute more difficult (i.e. B is rendered less positive),

so that dB/dT becomes negative. A structure-breaker on the other hand, exerts a less disruptive influence on the structure of the solvent (which is already broken) at an elevated temperature and hence the corresponding dB/dT is positive. These considerations are obviously independent of the size of the solute particle. S.K. Mandal & S.K. Sanyal[7] determined viscosity B co-efficiient of phenol, resorcinol, p-cresol and benzyl alcohol in aqueous medium at 30,35 and 40°C.

Several theories have been put forward relating the viscosity of pure components to their mixtures. Eyring's theory, one of the earliest theories of liquid viscosity is due to Eyring & co-workers that may be written as -

$$\eta = (hN/M) \exp. (\Delta G^*/RT) \qquad (7)$$

Where η is viscosity, M is the molecular weight, T is the absolute temperature and h, N & R are Plank's constant, Avogadro's number and the gas constant respectively. ΔG^* represents the free energy of activation for viscous flow. The excess free energy of activation, ΔG^{*E} is given by the difference between the free energy of activation of the mixture and the free energy of activation of the ideal mixture. Thus eqn. (7) can be written as,

$$\Delta^*G^E = RT (\ln \eta M - x_1 \ln \eta_1 M_1 - x_2 \ln \eta_2 M_2) \quad (8)$$

Where η & M are the viscosity of mixture and the average molecular weight of the components in the mixture respectively η_1, x_1 & M_1 represent the viscosity, mole fraction & the molecular weight of the ith component. This theory has provided a foundation for numerous correlations. However, the search for satisfactory analytical correlations applicable generally, or atleast to groups of chemical binary systems has, not yet produced the answer. Some semiemperical correlations have been developed with limited sources.

On the basis of the absolute reaction rate theory of Eyring, McAllister derived the following cubic relation for the kinematic viscosity of a liquid mixture.

$$\ln\eta = x_1^3 \ln\eta_1 + 3x_1^2 x_2\ln\eta_{12}$$
$$+ 3x_1 x_2^2 \ln\eta_{21} + x_2^3 \ln\eta_2 + D \qquad (9)$$

Where ,

$$D = -\ln (x_1 + x_2 M_2/M_1) + 3x_1^2 x_2 \ln(\tfrac{2}{3} + M_2/3M_1)$$
$$+ 3x_1 x_2^2 \ln(\tfrac{1}{3} + 2M_2/3M_1) + x_2^3 \ln(M_2/M_1) \qquad (10)$$

Equation (10) is applicable only if the ratio of radii of two components in the mixture is smaller than 1.5. Also, eqn.(10) contain two adjustable parameters, namely $\ln\eta_{12}$ & $\ln\eta_{21}$, which could be determined by a least square method. The term D can be easily calculated from a knowledge of the composition of the mixture and of the molecular weight of the components. The selection of a cubic equation is justified.

The less complex, three parameter Auslander equation has the following form :

$$X_1(x_1 + B_{12}x_2) (\eta - \eta_1) + A_{21} x_2(B_{21}x_1 + x_2)$$
$$(\eta - \eta_2) = 0 \qquad (11)$$

Where,

A_{21}, B_{12} & B_{21} are the parameters representing binary interactions & could be calculated from a least squares fit of the experimental data.

Heric proposed the following relation

to correlate the binary viscosity data.

$$\ln\eta = x_1 \ln\eta_1 + x_2 \ln\eta_2 + x_1 \ln M_1 + x_2 \ln M_2$$
$$- \ln(x_1 M_1 + x_2 M_2) + \Delta_{12} \quad (12)$$

Where, $\Delta_{12} = \alpha x_1 x_2$, is a function representing departure from a non- interacting system and $\alpha_{12} = \alpha_{21}$ is the interaction parameter. Heric expressed α_{12} or α_{21} as a linear function of composition.

$$\alpha_{12} = \beta_{12}' + \beta_{12}'' (x-x_2) \quad (13)$$

The co-efficient β_{12}' & β_{12}'' could be determined from a weighted least square method.

Quite often viscosity data were also fitted to an empirical equation of the form

$$\eta = \eta_1 x_1 + \eta_2 x_2 + x_1 x_2 [a + b(x_1-x_2) + c(x_1-x_2)^2 + \ldots] \quad (14)$$

Where a, b & c represent the co-efficient to be evaluated.

T.M. Aminabhav et.al.[8] determined the density & viscosity for six binary solvent mixtures at 20°C and compared the experimental data with literature values and fitted to several empirical relations. Viscosity of liquid mixtures provide information for the elucidation of fundamental behavior of liquid mixtures, help in correlation of mixture viscosities with those of pure components & provide a basis for the selection of physico-chemical methods of analysis. Nigam and Mahl[9] have shown that the sign & magnitude of excess viscosity, η^E & Grunberg & Nissan characteristic parameter (d) can be used to predict the degree & the type of interaction.

S.L. Oswal & A. Venkateswara Rao[10] reported density & viscosity measurements for six binary liquid mixtures of triethylamine, triprophylamine and tributylamine with cyclohexane & benzene. T.M. Aminabhavi et. al.[11] reported the densities viscosities & refractive indices in binary mixtures of methylacetoacetate, with esters such as methyl acetate, ethyl acetate, etc. K. Purna Chandra Rao[12] determined viscosities of pure components & binary liquid mixtures of N- N-Dimethylfor-mamide, & N, N-Dimethylacetamide with ethyl acetate, n-propyl acetate, etc. at 303.13k.

A.B. Bilkis[13], S.K. Biswas & M. Alamgir reported the measurements of density and viscosity of binary system of ethylene glycol-water, ethylene glycol-dioxane and ethylene glycol-acetone.B.Vijaykumar et.al.[14] reported the measurements of density and viscosity of pure formamide and its binary mixtures with ethylene glycol, diethylene glycol, etc. as a function of composition over the entire range at 35°C. P.S. Nikam et.al.[15] reported density & viscosity studies of aniline with alcohols (C5-C10) and the effect of chain length of alcohols on these properties of binary mixtures of aniline with alcohols. (C5,C6, C7, C8, C9 & C10). Ramkrishna Pramanik & Sanjib Bagchi[16] studied and reported the density & viscosity of ethanol and 1-octanol binary system.

It is well known that the reaction medium plays an important role in determining reactivity which is reflected in thermodynamic, transport and spectral properties[17]. In order to gain insight into the mechanism of such interactions, thermodynamic studies involving one or more solutes in mixed solvent systems are highly useful. Studies on the apparent molar volumes of electrolytes and dependence of

viscosity on concentration of solutes and temperature of solutions have been employed as a function of studying ion-ion and ion-solvent interactions[18]. It has been found by a number of workers[19] that the addition of electrolyte could break or make the structure of a liquid. Since, the viscosity is a property of the liquid which depends upon the intermolecular forces, the structural aspect of the liquid can be inferred from the viscosity of solutions at different concentration and temperatures.

A. Chaudhari *et.al.*[20] reported the measurement of density and viscosity data of some mineral salts like ammonium nitrate, potassium nitrate, *etc.* in 30% mass tetrahydrofuran and water mixtures at different temperatures. Amalendu Pal and Harish Kumar[21] reported the viscosities and excess molar volume as a function of composition for binary liquid mixtures of dipropylene glycol monomethyl ether and of dipropylene glycol monobutyl ether with 1-propanol & 2-propanol. Bhoj Bahadur Gurung *et.al.*[22] measured and reported density and viscosity of pure benzene and its binary mixtures with carbontetrachloride and chloroform as a function of composition over the entire range at different temperatures.

The volumetric & viscometric studies of electrolytes at infinite dilution in various mixed solvent system have contributed to our knowledge about electrolyte-non-electrolyte-water interactions. By examining the viscosity B co-efficient and $V^{\circ}\phi$ of ions as function of size, nature, temperature and composition of the mixed solvent, it is possible to study the effect of these parameters on ion-water interactions, with the hope of obtaining a better understanding of the interactions in solutions.

P.S. Nikam *et.al.*[23] have determined densities of ammonium sulphate, potassium sulphate and aluminium sulphate in aqueous DMF at different concentrations in the temperature range 298.15 to 313.15K. From these data, apparent molar volumes ($V\phi$) have been derived and analyzed using Masson equation. The limiting apparent molar volume ($V^{\circ}\phi$) and slope (S_V) are interpreted interms of solute-solvent and solute-solute interactions respectively. The structure making/breaking capacities of electrolytes have been ascertained from the equation of L.G. Hepler. Recently, M.L. Parmar and D.K. Dhiman[24] have reported studies on the determination of partial molar volumes of some mineral salts from density measurements in aqueous medium at different concentrations and temperatures. The results have been interpreted in terms of ion-solvent and ion-ion interactions.

Reena Gupta and Mukhtar Singh[25] measured density & viscosity of ternary systems involving maltose, alkali metal halides and water at different temperatures. The density and viscosity data have been analyzed by applying Jones-Dole equation. The structure making and structure breaking capacities of halides in the presence of maltose have been ascertained from the temperature dependence. Anwar Ali *et.al.*[26] presented the density, viscosity & refractive index data for urea and D-glucose in glycine and water mixed solvent as a function of urea/glucose concentration at 308K. The experimental data obtained was analysed interms of solute-solvent and solute-solute interactions.

S. Senthil Raja & T.R. Kubendran[27] reported density & viscosity data for 1,4-

dioxane + carbon tetrachloride, carbon tetrachloride + butanol & 1,4-dioxane + butanol at 303.15, 308.15 & 313.15 K. The experimental data was used to calculate deviations in viscosity, and excess molar volume of the mixtures. M.L. Parmar & M.K. Guleria[28] determined relative viscosities for the solutions of oxalic acid & its salts like ammonium oxalate, sodium oxalate & potassium oxalate at different concentration in water and in binary aqueous mixture of tetrahydrofuran. The data have been evaluated using the Jones-Dole equation and the obtained parameters have been interpreted interms of solute-solute and solute-solvent interactions.

S.K. Lomesh *et.al.*[29] reported molar volume, viscosity and molar conductance of copper sulphate in water, aqueous sodium chloride and dextrose from density, viscosity and conductance data. The experimental data obtained was analysed in terms of Jones-Dole equation, Walden product and interpreted in terms of solute-solvent interactions and structure making/breaking behavior of copper sulphate.

A thorough knowledge of transport & thermodynamic properties of multi component liquid mixture is essential in many industrial applications. Therefore, the viscosities of multi component liquid mixtures are required in many chemical engineering calculations. Dominguez and co-workers[30] have carried out studies relating to thermodynamic and transport properties of binary and ternary mixtures involving butanols , n-hexane, 1-chlorobutane, 1-butylamine at 298.15 & 313.15 K. Surabhi Varshney & Mukhtar Singh[31] carried out viscometric studies on molecular interactions in ternary liquid mixture of ethane 1,2-diol and pyridine with some polar & non-polar solvents. From the density and viscosity data, the viscosity deviations and the excess thermodynamic properties have been calculated. On the basis of there values of excess thermodynamic properties, molecular interactions between the mixing components have been discussed.

Physiological & biological activities are medium dependent. Thus study of their interactions and physical characterization is essential. As per molecular modeling data, their interaction with water could influence actual biological processes and hence estimation of parameters such as density, volume and viscosity become necessary. Man Singh[32] measured density and viscosity of citric acid & disodium hydrogen orthophosphate at various strengths and at different temperatures. Varsha Sharma *et.al.*[33] explained viscosity results of uranyl soaps in dimethyl formamide in terms of equations proposed by Einstein, Vand, Moulik & Jones-Dole.

Partial molar volumes of electrolytes provide valuable information about ion-ion, ion-solvent & solvent-solvent interactions[34-35]. This information is of fundamental importance for understanding the reaction rates and equilibria involving dissolved electrolytes. The structural interactions of non-ionic solutes with ionic ones in different solvents are important in many fields of chemistry. The studies of such interactions of non-electrolytes with electrolytes in solution are very significant and useful for investigating their physico-chemical behavior[36-37]. Vishnu *et.al.*[38-39] have studied aqueous & non-aqueous ternary system containing polyhydroxy compounds and electrolytes like alkali halides.

Vijay Laxmi & Mukhtar Singh[40] measured viscosities & apparent molar volumes of solutions of uni-univalent & bi-univalent electrolytes (NaCl, KCl, NH_4Cl, etc.) and ($BaCl_2$, $MgCl_2$) in purely aqueous & aqueous thiourea solutions at different temperatures. These data have been used to calculate the constants of Jones-Dole & Masson's equations.

Grouping of solvents into classes is often based on the nature of the intermolecular forces, because the manner whereby solute & solvent molecules are associated with one another brings about a marked effect on the resulting properties. After the introduction of the concept of ionization power of solvent[41] much work has been devoted to the solvent effects on the rate and equilibrium processes[42]. Because of the close connection between liquid structure and macroscopic properties, determination of volumetric and viscometric properties are valuable tool to learn the liquid state[43]. On the other hand, reliable measurements of solvent properties over a wide range of composition, pressure and temperature is not often feasible; hence, prediction and correlation methods constitute a valuable option to overcome such difficulties[44].

Ankan Chaudhari *et.al.*[45] measured and reported the density & viscosity of binary mixtures of tetrahydrofuran with n-pentane, n-hexane & n-heptane as a function of the compostion at 288.15, 293.15 and 298.15K. From these data, excess thermodynamic properties were calculated. The results have been interpreted interms of possible molecular interactions existing between the component of these mixtures. The study of biomolecules plays a very important role in understanding the thermodynamic behavior of biochemical processes in living cells. Attwood *et.al.*[46] determined the apparent molar volume & adiabatic compressibility of aq. solution of phenothiazine and chlorpromazine drugs with regard to temperature dependence. Temperature increase caused linear increase in both apparent molar volume and compressibility of chlorpromazine in micellar form.

A. Kumar *et. al.*[47] measured densities & viscosities of triethylamine with methanol, ethanol & 1-propanol. Gibb's free energy for the activation flow & Grunberg-Nissan interaction parameters were computed. The results were discussed in the light of intermolecular interactions.Aminabhavi *et. al.*[48] studied densities, viscosities of binary mixture of methyl acetoacetate with aliphatic alcohols (C_1 - C_8). The variation of various excess properties with chain length & temperature was discussed. In another study, they analyzed the viscosity data using viscosity equations such as McAllister, Auslander & Heric[49].

Wen-Lu-Weng[50] reported densities & viscosities for the binary mixture of butylamine with aliphatic alcohols (C_4 - C_8). The negative excess molar volumes & viscosity deviations results revealed the presence of molecular interactions of the cross association between the alkanols & amine molecules. Su-chan-ku & Chein-Hsiun Tu[51] reported densities & refractive indices of bianary mixture of dimethyl ether of a glycol with ethanol at various temperatures.

Horsein A. Zarei[52] reported densities, excess molar volumes & Partial molar volumes of the binary mixtures of acetic acid + alcohols

(C_1-C_4) at 298.15 K. Pavel Hyncica *et. al.*[53] reported densities & Partial molar volumes of organic solutes in water at the temperature rang 298 to 573 K. Y. Leong *et.al.*[54] reported partial molar volumes of acetomitrile + water mixture over the temp range 273.15 to 318.5 K. Volumetric properties of binary mixtures are complex properties because they depend on only on solute - solute, solvent-solvent and solute-solvent interactions, but also on the structural effects arising from interstitial accommodation due to the difference in molar volume and free volume between components present in the solution. Knowledge of several properties, including densities at different temperature, is required for engineering design and for subsequent operations.

R.B. Torres *et.al.*[55] measured the densities & reported volumetric properties of binary mixtures of acetonitrile and alcohols at different temperatures.

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