

Comparison of Densities and Surface tension with Literature Data for Pure Components at 293.15, 298.15, 303.15 and 313.15 k.

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Abstract: The study of thermodynamics and transport properties of liquid and liquid mixture is an important part of liquid chemistry. Chiefly, liquid particle movement, surface tension, viscosity, liquid crystals are the vital part of study in liquid chemistry. The transport properties (viscosity, thermal conductivity and diffusion coefficient) of liquids and gases (fluids) are important for the most efficient engineering design of many processes in the oil, chemical and biotechnological industries. They characterize the response of a fluid to changes in its temperature, speed of flow or composition. Present thesis entitled "Critical Analysis of Thermodynamic and Transport Properties of Liquid and Liquid Mixture", is divided into five chapters.

Keywords:- liquid particle movement, liquid chemistry, Liquid mixture, Solid state.

Introduction

Solid state is a recognized field of science in which many advances have been achieved over the past decades. But in recent years the liquid state has been subject of intense study. However, due to characteristics of liquid state, i.e., the strong interactions of particles and their state of disorder, the theoretical analysis has lagged far behind theories of gaseous and crystalline states. The knowledge of the thermodynamic and physical properties of multi component systems is essential for design calculations involving separations, heat transfer, mass transfer and fluid flow. Traditional theories of solutions dwell exclusively on two aspects of liquid mixtures. One of these is the entropy associated with dispersion of the low molecular species or of their constituent elements in case of complex molecules, among one another. A lattice model often serves as the device for estimating this combinatorial entropy. The other aspect relates to the interactions between neighbouring molecules and in particular, to the difference in interactions between unlike and like neighbour pairs. Treatment of the properties of liquid mixtures has progressed little beyond the level of interpretation possible within the framework supported by these two considerations alone [1, 2]. The equilibrium properties of a liquid are strongly dependent on what may be loosely called its local structure, often expressed in terms such as packing density, free volume or more exactly in terms of the radial distribution function. In as such as, this local structure depends on the forces between molecules and volume of the molecules; in general, it will change with the composition. This change in turn will be reflected in the thermodynamic properties of the mixture. Contributions of this nature have either been



ignored altogether, or correction to a state of null volume change on mixing has been adopted as a means of compensating for the effects referred [2]. It will be apparent, however that, adjustment of one thermodynamic quantity (e.g., volume in this manner) will not in general, affect a simultaneous correction of others (e.g., the free energy of that part of it relating to the local structure) to their linearly interpolated values. The choice of volume as the property to be conserved is arbitrary and there is assurance that nullity of volume change obviates consideration of other characteristic properties of the liquid. More sophisticated treatments [3-4] of liquid mixtures derived their conceptual basis from-

(a) the cell model for liquids and (or) from –

(b) the postulations of a universal form for the intermolecular potential expressed by

$\in_{ij} \in \stackrel{*}{\in}_{ij} \phi(\mathbf{r}_{ij} = \mathbf{r}^{*}_{ij})$

where, r_{ij} is the distance between centres of molecules i & j, $\in_{ij}^{*} \& r_{ij}^{*}$ are characteristic parameter for the pair, and $\phi(r/r^{*})$ is the universal function of its agreement.

The intermolecular energy is usually discussed on the basis of the familiar Lennard-Jones potential operating between the molecular centres. Even in comparatively simple polyatomic molecules, the acentric distribution of polarizable electrons necessitates fairly drastic modification of this potential. Kihara[3], Hamann and Lambert[4] and Pitzer[5,6] have offered modified intermolecular potentials for polyatomic (globular) molecules, especially those approximating spherical symmetry in form.

An extension [7] of these considerations which suggests itself, as a basis for treating the intermolecular energy in liquid consists in integrating the interactions of elements of the core of one molecule with the elements of all surrounding molecules, these latter being treated as occupying the space outside the cavity reserved for the molecule in questions.

The result obtained departs markedly from the form of the intermolecular energy according to the theory of Lennard-Jones and Devonshire.

In the limit of very large particles such that the range of intermolecular interactions, attractive as well as repulsive, is small compared to the molecular diameter and to the distance between boundaries of the domains of neighbouring molecules, the intermolecular energy can then be treated as arising effectively from interactions between the surfaces of adjoining molecules. The calculations support an account of the intermolecular energy on this basis, and they indicate further that this manifestly approximation should be satisfactory even for small polyatomic molecules. Certainly, this approximation, which has a long history of usage in solution theory, is preferable to central force potentials, the Lennard-Jones type of virtually all molecules. It offers the additional advantage to treatment of non-spherical molecules and of mixtures of molecules deferring in size. There are two principal reasons for the great amount of experimental and theoretical work on the properties of liquid mixture1. The first is that they provide one way of studying the physical forces acting between the two molecules of different species and second for the study of mixtures, is the appearance of new phenomenon (Interactions) which are not present in pure liquids.

II. Material & Methods

A theoretical in hand prediction of various thermodynamic properties of liquids and liquid mixtures has an advantage, for there are numerous instances where it is impractical to measure various physical and thermodynamic properties at all external conditions of interests. But, for to verify the validity of various theoretical predictions at varying conditions of compositions and temperature it is necessary to have a check in the form of experimental findings. A careful examination of differences between theoretical predictions and experimental findings, excluding minimal experimental error, gives a very clear insight regarding the molecular interactions which exist and may exist. A large difference between the two results clearly points out the possibility of those interactions, which may not have been considered in a particular theoretical approach. Thus, apart from revealing the nature of interactions which may exist in a particular liquid



mixture, the experimental findings also pave a scientific and precise path for the development of various theories which predict various physical and thermodynamic properties. Experimental Technique and Procedure

2.1 Density Measurements

The pycnometer was used to ensure the accurate measurements of density of liquids and every care was taken to minimize the errors. The double distilled water was used to calibrate the pycnometer. The Climbed pyknometer, (fig.2.1) similar to that of Parker and Parker [73], with minor modifications was used for the measurement of density. It consisted of a bulb of about of 26 cm. The stems were made of capillary tube of uniform bore of about 1 mm. In order to prevent any loss of liquids due to evaporation, the open ends of the stems were closed with Teflon caps. The caps had small orifices to ensure that the pressure of inside capillary was equal to atmospheric pressure. The two stems of the pycnometer were divided into fifty equal parts. Marks were made at each of the two stems at equal levels. The liquid level in each stem was observed with reference to the marks. The total length of unfilled capillary was kept near 10 cm., to ensure minimal loss of vapours on account of diffusion. The well cleaned and dried pycnometer was accurately weighed. The liquid was introduced into the weighed pycnometer with the help of a hypodermic syringe until levels of liquids in both the stems nearly corresponded with the marks on the stems. The pycnometer was kept immersed vertically in a thermostat maintained at required temperature for about an hour. This enabled the unfilled parts to be drained completely and allowed liquid-vapour equilibrium to be established. The amount of liquid at the initial filling was adjusted to allow for expansion (or contraction) due to difference of temperature between the bath and surroundings. The difference of heights of liquids between the marks and the lowest point in the meniscus in both the limbs was measured when liquid levels were stationary. The pycnometer was removed from the bath, wiped with clean damp cloth and kept into the balance for about 15 minutes before weighing. Accuracy in the density measurements was found to be $\pm 2 \times 10^{-4}$ g.cc⁻¹.



Figure 1: Diagram of Pycnometer.

2.2 Calibration of the Pycnometer

The Pycnometer was calibrated before the measurements. Double distilled water was used as standard solvent. The (X) divisions i.e. the total height with capillaries of Pycnometer, was plotted against volume (V) at 25° C. Volume (V) is indirectly calculated from equation V=mX + C, after determining the mass of water every time. Calibration constants were evaluated by the method of least square. The experimental error was estimated by calculating densities of pure liquids from the calibration constants.



In the equation V=mX +C, 'C' is the volume of bulb without liquid and 'X' is the total number of divisions on the two capillaries. The calibration constants were evaluated by the following equations using the least square fit method. The points which lie away from the mean straight line (when V is plotted against X) were neglected.

Densities were then calculated using the well-known expression $\rho=M/V$. The volume was evaluated at 25^oC by using standard densities for water (m₂-m₁) for every X value. Precision of density measurements was confirmed by checking densities of pure components with that of literature value at (25 ± 0.01) ^oC. It was found that experimental values were in good agreement with literature values (Table-1&2).

No. of Divisions X(cm.)	Weight of WaterM (g)	Volume of Water V (ml)
49	67.9055	29.2394
77	67.9515	29.2856
73	67.9484	29.2824
71	67.9436	29.2776
64	67.9293	29.2634
57	67.9121	29.2460
77	67.9516	29.2857
65	67.9335	29.2675
51	67.9157	29.2496

 Table 1: Parameters of the Calibration Constants for the pycnometer at 298.15K

Weight of Pcynometer = 38.7538 g. m= 0.001647 C= 29.15948

Table 2: Experimental and Literature Values of Densities of Pure Components at 298.15 k

	ρ (g/cc)	
Liquids	Exp	Lit*
Acetonitrile	0.7811	0.7795
Formamide	1.1290	1.1292
N- Methylacetamide (NMA)	0.9512	0.9524
N,N-Dimethylformamide (DMF)	0.9501	0.94383
N,N-Dimethylacetamide (DMA)	0.9502	0.93634

2.2 **Surface Tension Measurement**

2.2.1 **Jaeger's Method**

If spherical bubble is produced in a liquid, the excess pressure inside the bubble is $P=2\sigma/r$, where r is the radius and σ is the surface tension of the liquid. The relation is used by Jaeger (fig.2.2) in determining the surface tension of liquid and liquid mixture, by observing the pressure necessary to produce a bubble in the liquid. For this purpose, a Wolf's bottle is taken to produce small pressure which is measured by the manometer. This is connected to a vertical tube, the lower end of which has fairly narrow orifice of radius (r) dipped into the liquid, kept in a double walled jacket having one outlet and one inlet, attached to the thermostatic bath for temperature control. By opening the stop-cock, water is allowed to trickle in the wolf bottle which increases the pressure and bubble is formed. The bubble bursts off as pressure further



increases. Then another bubble is formed. The manometer registers a pressure difference (H) which is maximum when the bubble is completely formed.



Figure 2: Principle of Jaeger's Method.

2.2.2 Ultrasonic Studies

An ultrasonic interferometer was used to determine sound velocity in liquid mixtures. It is a simple and direct device to determine the ultrasonic velocity in liquids with a high degree of accuracy. In order to measure the sound velocity, a crystal controlled ultrasonic interferometer supplied by M/s. Mittal Enterprises, New Delhi, operating at a frequency of 2 MHz was used. Many workers [7-8] have used this technique. Velocity at 200 MHz with double crystal interferometer was measured by Hunter and Dardy[4]. The moveable crystal is a detector of ultrasonic velocity. Sound velocity measurements were carried out with other types of interferometers in which the resonance of liquid, filling the cavity was studied. Dobbs and Finegold[85] used a barium titanate cylinder filled with the experimental liquid, plated in such a way that one half of this acted as a receiver. Fort and Moore [86] measured ultrasonic velocity of fourteen binary mixtures using a single crystal interferometer working at 200 MHz. The interferometer (fig.3.3) was similar to that of Hubbard and Loomis [87] but incorporated a barium titanate crystal instead of a quartz crystal thereby avoiding the necessity for high voltage and consequent insulation difficulties.

The principle used in the determination of velocity (c) is based on the accurate determination of the wave length (λ) in the medium. A quartz crystal fixed at the bottom of the cell produces ultrasonic waves of known frequency. These waves are reflected by a moveable metallic plates kept parallel to the quartz crystal. If the separation between these plates is exactly a whole multiple of the sound wave length, standing





waves are formed in the medium. This acoustic resonance gives rise to an acoustic electrical reaction on the generator driving the quartz crystal and the anode current of the generator becomes maximum.



Figure 3: Diagram of Ultrasonic Interferometer.

2.3 Ultrasonic Measurements

The measuring cell was connected to the output terminal of the generator through a shielded cable. Before switching on the generator, the cell was filled with the experimental liquid. The ultrasonic waves moved normal from the quartz crystal till they were reflected back from the moveable plate and the standing waves were found in the liquid in between the reflector plate and the quartz crystal.

The gold-plated quartz crystal having frequency 2 MHz and micrometer with least count of 0.001 cm. was used. The distance travelled by the micrometer between the two successive maxima was just half of the wavelength and for each set of reading'd' for 20 maxima were recorded for the sake of convenience. Thus, distance gave the value of wavelength, with the help of the following relation,

Table 3: Experimental and Literature	Values of Ultrasonic velocity	of Pure Components at 29	98.15 k

	U (m/s)		
Liquids	Exp	Lit*	
Acetonitrile	1290.1	1326.3	
Formamide	1601.0	1626.3	
N- Methylacetamide (NMA)	1401.0	1431.6	
N,N-Dimethylformamide (DMF)	1465.2	1496.4	
N,N-Dimethylacetamide (DMA)	1451.3	1501.3	



2.4 Viscosity Measurements

The viscosity of various liquids is measured by using a precalibratedubbelohde capillary viscometer¹¹³. This method is based on the measurement of time, where time taken for a given quantity of liquid to pass between two points is taken as an indication of the viscosity. For a capillary viscometer, the viscosity is given by the following equation which also takes into account Hagenback- Couette correction.

This method is particularly suitable for obtaining viscosity because the ratio of viscosities of liquid mixture and the pure liquid is proportional to their drainage time

S.No.	Component Liquids	Time (sec.)
1	Acetonitrile	67.3
10	Formamide	101.3
3	n-methylacetamide	97.4
4	Di-methylacetamide	81.4
	Di-methyl acetamide	92.3

Table 4: Parameters of the Calibration Constants for the Suspended Level Ubbelohde Viscometer at 25^oC

The measurements were carried out at $(25\pm0.01)^{0}$ C. Due care was taken to ensure that temperature should not only be constant but also uniform as the change of $\pm 0.01^{0}$ C in temperature causes approximately 0.02% change in viscosity value. A constant temperature was attained by maintaining a constant flow of water from a thermostat, through the glass jacket in which viscometer was kept. The time of flow for each liquid mixture, was measured at least 3-4 times until reproducible timing was attained. The time of flow of pure liquids was also measured to check the accuracy of the results. A correlation within \pm 0.0017 c.p. was recorded.

Table 5: Viscosity of Pure Liquid at 25^oC

S.No.	Component Liquids	Exp. (c.p.)	Lit. (c.p.) nm ⁻³ *
1	Acetonitrile	0.3626	0.3636
2	Formamide	3.3220	3.3240
3	n-methylacetamide	3.3022	3.3176
4	Di-methylacetamide	0.7655	0.7670
	Di-methyl acetamide	0.9437	0.9443

III. Result Analysis

Table 6 presents the comparison of experimental densities and surface tension of acetonitrile, formamide, N-methylacetamide (NMA), N,N-dimethyl acetamide (DMA) and N,N-dimethyl formamide (DMF) with literature values [66-68] at 293.15, 298.15, 303.15 and 313.15 K.

Table 6: Comparison of Densities and Surface tension with Literature Data for Pure Components at 293.15 , 298.15 ,303.15 and 313.15 k.

Liquids	Temperature (T)	Density (ρ) (g/cc)		Surface Tension (σ) (dyne/cm ²)	
	(K)	Exp	Lit*	Exp	Lit*
Acetonitrile					
	293.15	0.7965	0.7822	29.25	29.29
	298.15	0.7811	0.7795	28.23	28.25
	303.15	0.7733	0.7713	27.63	27.80
	313.15	0.7605	0.7615	27.20	





Formamide					
	293.15	1.1320	1.1334	58.22	58.35
	298.15	1.1290	1.1292	58.02	58.15
	303.15	1.1250	1.1241	57.78	
	313.15	1.1117		57.47	57.54
N- Methylacetamide (NMA)					
	293.15	0.9563	0.9568	34.13	34.58
	298.15	0.9512	0.9524	33.52	33.68
	303.15	0.9502	0.9500	33.05	33.67
	313.15	0.9405		32.53	32.17
N,N-Dimethylformamide (DMF)					
	293.15	0.9551	0.9487	36.62	36.76
	298.15	0.9501	0.9439	36.31	36.42
	303.15	0.9419	0.9412	35.90	
	313.15	0.9325	0.9310	35.53	34.40
N,N-Dimethylacetamide (DMA)					
	293.15	0.9623	0.9415	33.13	33.29
	298.15	0.9502	0.9363	32.43	
	303.15	0.9360	0.9317	31.91	32.43
	313.15	0.9221	0.9232	31.38	30.92

Table 7: Coefficients	of the Redlich -	KisterEquatiion	and Standard	Deviation	(σ) for Surf	ace te	nsion of
Binary Liquid Mixture	es at various tem	peratures					

Acetonitrile+Formamide						
	T (K)	A0	A1	A2	A3	Stddev
σ_{exp}	293.15	-5.2686	-11.1474	-28.9847	30.3061	0.3686
	298.15	-0.3307	-8.3370	-28.1552	34.5652	0.4973
	303.15	0.0278	-5.9953	-36.4166	47.6330	0.6672
	313.15	-57.6806	92.9073	31.3491	-137.2633	3.0627
Acetonitri	le+NMA					
σ_{exp}	293.15	17.6548	29.7436	-17.4971	-31.3344	0.2848
	298.15	23.8720	14.8862	-51.0542	16.0588	0.1315
	303.15	24.9271	22.3447	-47.0046	16.1655	0.3605
	313.15	15.5429	20.9382	-17.9296	-16.4929	0.1160
Acetonitri	le+DMF					
σ_{exp}	293.15	20.6928	28.6860	-21.6652	-33.9279	0.2501
	298.15	17.5303	33.1361	-9.5627	-44.2660	0.3710
	303.15	16.3748	41.9846	-2.7388	-65.0142	0.6727
	313.15	18.5570	23.4815	-19.1109	-23.4336	0.2177



Acetoni	itrile+DMA					
σ_{exp}	293.15	15.8101	39.8844	13.6810	-59.3757	0.7131
	298.15	17.8280	35.5942	6.7003	-42.9307	0.4090
	303.15	16.9877	50.2500	29.7536	-76.7784	0.9730
	313.15	15.4054	34.7928	13.0453	-39.4369	0.8306

Table 8: Parameters of McAllister Three Body and Four Body Interaction model and Standard deviation (σ) for Surface tension of Binary liquid Mixtures at Various Temperatures

		McAllister	3 body		
Acetonitrile+Fo	ormamide				
	T (K)	а	В	stddev	stddev
σ_{exp}	293.15	35.4202	48.2889	0.9219	0.4118
	298.15	38.3114	47.3894	0.8781	0.4675
	303.15	39.5438	44.6321	1.1027	0.7625
	313.15	34.1459	21.3215	2.9503	2.9688
Acetonitrile+N	MA				
σ_{exp}	293.15	42.9448	33.0786	0.7096	0.2031
	298.15	40.0890	35.6439	1.0562	0.0957
	303.15	44.1611	32.3605	0.9397	0.1987
	313.15	38.7541	31.8407	0.5825	0.1094
Acetonitrile+D	MF				
σ _{exp}	293.15	44.3425	35.8184	0.8156	0.3237
	298.15	44.4521	33.6076	0.6480	0.4784
	303.15	45.0725	32.0908	0.8432	0.8928
	313.15	41.5688	34.1532	0.6261	0.1865
Acetonitrile+D	MA				
σ _{exp}	293.15	44.7516	32.2687	0.3741	0.3145
	298.15	44.6010	31.6164	0.4340	0.3624
	303.15	48.1241	29.7207	0.4168	0.4094
	313.15	43.5316	29.6949	0.3998	0.4055

	McAllister 4 body			
acetonitrile+forman				
	T (K)	А	В	С
σ_{exp}	293.15	30.1902	50.9952	44.6793
	298.15	32.0701	51.5219	44.5294
	303.15	32.7080	50.6728	42.5901



	313.15	31.8112	27.6899	26.8989
acetonitrile+N	IMA			
σ_{exp}	293.15	34.2311	51.0448	26.1546
	298.15	30.1770	60.0895	24.2992
	303.15	32.9791	57.0027	23.4684
	313.15	31.5626	45.9733	25.7942
acetonitrile+D	OMF			
σ_{exp}	293.15	34.8498	53.2481	29.2371
	298.15	36.2747	46.4994	29.9356
	303.15	36.9058	44.5249	29.3900
	313.15	33.3097	47.9986	28.9509
acetonitrile+D	OMA			
σ_{exp}	293.15	41.4487	38.9932	30.6910
	298.15	40.8680	39.0838	29.6508
	303.15	45.8818	33.7703	31.3820
	313.15	41.3585	33.8148	30.3449

IV. Conclusion

Traditional theories of solutions dwell exclusively on two aspects of Liquid mixtures. One of these is the entropy associated with dispersion of the low molecular species, or of their constituent elements in case of complex molecules, among one another. A lattice model often serves as the device for estimating this "combinatorial" entropy. The other aspect relates to the interactions between neighbouring molecules and in particular, to the difference in the interactions between unlike and like neighbour pairs. Treatment of the properties of liquid mixtures has progressed little beyond the level of interpretation possible within the framework supported by these two considerations alone.

The equilibrium properties of a liquid are strongly dependent on what may be loosely called its local structure, often expressed in terms such as packing density, free volume, or more exactly in terms of the radial distribution function. In as such as, this local structure depends on the forces between molecules and volume of the molecules; in general, it will change with the composition. This change in turn will be reflected in the thermodynamic properties of the mixture.

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