

Studies of intermolecular force coefficient (σ_{imf}^0) for methyl derivatives of urea in aqueous solutions with Friccohesity, a new physicochemical function, from 293.15 to 303.15 K^ψ

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Both surface tension (γ) and viscosity (η) for 0.0050 to 0.2560 kg mol⁻¹ urea, methylurea and dimethylurea solutions have been measured with Survismeter for Friccohesity (σ /Sm⁻¹), derived from frictional and cohesive forces. The limiting (σ^0) and slope values S_σ of σ at infinite dilution, $m = 0$, are determined from σ vs m mol kg⁻¹ fit for intermolecular forces coefficient (σ_{imf}^0) and influence of temperature, K. The S_σ values project an effect of concentrations on pairwise interactions of ureas. And decrease in σ_{imf}^0 and S_σ values from urea to dimethylurea focus contribution of -CH₃ group to hydrophobic interactions over the hydrophilic, due to -NH₂ and >CO- groups of urea. Substitution of H by -CH₃ is noted to weaken hydrogen bonding strength of >CO-.

The η emphasize frictional and γ the cohesive forces of attraction being fundamental transport properties¹⁻⁴ useful in material and civil engineering, pharmaceuticals, soaps and detergents, paper and textile and many others. Since inception a vacancy has been felt to measure both the functions with one instrument and fit in suitable relation to derive another property known as σ_{imf}^0 which has never been the focus of experimental measurements for liquid mixtures as no mixing is ideal. Even mixing of ideal gases does cause interactions due to van der Waals and surface forces. Thus a Survismeter is devised and an equation formulated by author himself referred to as Man Singh equation for σ_{imf}^0 , which is fundamental property like MP, BP and density. Hence σ_{imf}^0 is measured to illustrate a state of net intermolecular forces of ureas due to shift in interactions from hydrophilic to hydrophobic. Newton⁵ derived internal friction from (dv/dx) velocity gradient for homogenous solutions (Newtonian) and Stoke⁶ applied the hydrodynamic law on the viscous flow like-wise Gibson and Jacob developed falling sphere method followed by Ostwald viscometer, Einstein⁷ and Huggin⁸ calculated specific viscosity for polymers. Thus η and γ functions have been dealt separately and no common relation was attempted, thus σ resolves micro-level changes of intermolecular forces in combine. Wilhelmy, duNouy based on capillary pull and Jaeger, Sugden, Ferguson on

drop method have worked and Poisson-Raleigh gave their correction to capillary, Hakines and Brown⁵ emphasized the drop size with the surface force. There is very thin line of separation between η and γ , thus a Survismeter⁹ measured σ values, as usual⁵ methods engage laboratory infrastructure with manifold use of time and chemicals. Fundamentally the Friccohesity is derived from fric of frictional and cohesity of cohesive forces and σ_{imf}^0 measures the heteromolecular forces from hydrophilic to hydrophobic system.

Results and discussion

The surface tension and viscosity of benzene, acetic acid, ethylacetate, glycerol were measured and along with literature^{9,10} given in Table 1 with ± 0.05 dyne cm⁻¹ and ± 0.0001 milipoise accuracy.

The regression constants of ρ and η data with m are given in Table 2, and of σ vs m in Table 3 and the F and E^* obtained from B values with Arrhenius equation in Table 4.

The σ vs m are plotted in Fig. 1 with linear relation and its regression constants σ^0 and S_σ in Fig. 2 (a and b). The σ values for dimethylurea are found higher than of other ureas at each K with larger and lower scattering around lower and higher concentrations, respectively. The σ also investigates the dipole moment as heteromolecules

^ψDedicated to Professor H. C. Gaur on his 80th birthday.

Table 1. Experimental and literature data of surface tension (dyne cm⁻¹) and viscosity (milipoise) of solvents measured with the Survimeter at two temperatures. Δ is the difference between the two values

Systems	<i>T</i> (K)	Surface tension		Exp. - Lit. Δ	Viscosity		Exp. - Lit. Δ
		Lit.	Exp.		Lit.	Exp.	
Carbon tetrachloride	298.15	26.15	26.76	0.61	0.88	0.881	0.001
Methanol	298.15	22.28	22.51	0.23	0.547	0.545	-0.002
	293.13	22.55	22.98	0.43			
Ethyl ether	298.15	16.50	16.26	-0.24	0.222	0.224	0.002
Benzene	298.15	27.50	27.40	0.10			
	293.15	28.87	27.85	-1.02	0.601	0.602	0.001
Cyclohexane	298.15	23.82	24.12	0.30	0.98	0.979	-0.001
Acetic acid	293.15	27.42	27.16	-0.26			
Ethanol	293.15	22.40	22.75	0.35	1.06	1.061	0.001
Glycerol	298.15	64.00	64.01	0.01	1.49	1.489	-0.001

Lit.^{9,10} (the literature) and Exp. (experimental) values. Δ = Exp. - Lit.**Table 2.** Limiting density ($\rho^0/10^3$ kg m⁻³), viscosity ($\eta^0/\text{Nsm}^{-2} = 0.1$ kg m⁻¹s⁻¹ SI unit) and surface tension (γ^0/Nm^{-1} , SI unit) with slopes at infinite dilution, MU = methylurea, DMU = dimethylurea. Units : $S_d/10^3$ mol m⁻³, S_η/Nsm^{-2} mol, S_γ/Nm^{-1} mol kg⁻¹, S'_γ/Nm^{-1} mol kg⁻¹

Urea	ρ^0	S_d	η^0	S_η	γ^0	S_γ	S'_γ
293.15	0.99828	0.01037	0.99922	0.04318	72.71922	3.48884	-26.54166
298.15	0.9971	0.01004	0.89213	0.03566	71.9302	3.30798	-25.5666
303.15	0.99573	0.00915	0.79797	0.03844	71.15559	0.5181	0.5351
MU							
293.15	0.99827	0.00787	0.95501	0.36654	73.66501	-16.826	55.649
298.15	0.99712	0.00766	0.89131	0.10593	72.92856	3.26439	-26.24396
303.15	0.99537	0.00789	0.79653	0.10113	72.4113	-4.26209	-0.80623
DMU							
293.15	0.99824	0.00785	1.0142	0.26653	71.90332	-7.65048	-0.59596
298.15	0.99677	0.00716	0.90994	0.16965	71.12805	-9.28276	20.78895
303.15	0.99562	0.00529	0.81526	0.17059	70.16846	2.78667	-23.90933

Table 3. The Friction coefficient (σ^0/Sm^{-1}) and its slope S_σ . U-urea, MU-methylurea, DMU-dimethylurea

System	σ^0 at $m = 0$					
	293.15		298.15		303.15	
	σ^0	S_σ	σ^0	S_σ	σ^0	S_σ
U	0.13690	0.01221	0.12337	0.01053	0.11162	0.00448
MU	0.13669	0.02249	0.12157	0.02039	0.10950	0.02062
DMU	0.14076	0.05375	0.12769	0.03141	0.11546	0.02906

cause molecular dipole (μ_m) moment. For latter the solution is filled between the plates of capacitor to determine capacitating power. The dipoles align in a direction of the field decreasing its strength and increase the capacitance. A linear increase in the σ values with m proves enhancement of heteromolecular forces between ureas and water with water structure breaking. The ρ and η show

linear relation with m but γ polynomial with generation of internal pressure due to stronger heteromolecular forces with decrease in cohesive pressure (Table 2) due to -CH₃ groups with the 3×10^{-5} kg mol⁻¹ and 1×10^{-5} kg mol⁻¹ decreases in ρ^0 from methylurea to dimethylurea and from urea to methylurea, respectively.

The latter depict development of hydrophobic interac-

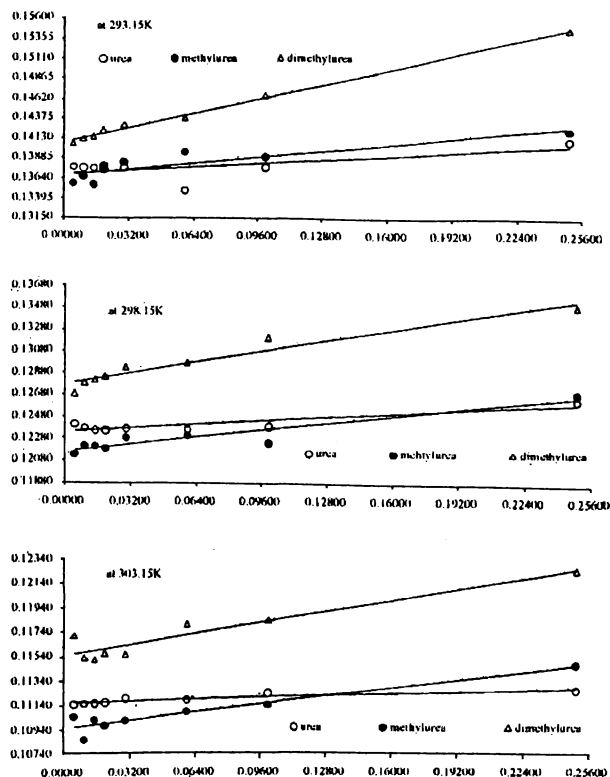


Fig. 1. Represent the plot of Friccohesity (σ , Sm^{-1}) vs m of ureas at three temperatures, Y-axis consists Friccohesity values and X-axis the m .

tions due to weaker intermolecular force. The temperatures from 293.15 to 298.15 and from 298.15 to 303.15 K are termed as lower and higher, respectively. The $1.18 \times 10^{-3} \text{ kg mol}^{-1}$ and $1.37 \times 10^{-3} \text{ kg mol}^{-1}$ decreases in ρ of urea from 293.15 to 298.15 and from 298.15 to 303.15 K, respectively elucidate larger weakening in intermolecular forces due to electrostriction of water. For per 5°C increase in temperature from 293.15 causes less weakening in electrostriction of urea rather than from 298.15 K due to weaker hydrogen bonding. The 115×10^{-3} and $1.75 \times 10^{-3} \text{ kg m}^{-3}$, and 1.47×10^{-3} and $1.15 \times 10^{-3} \text{ kg m}^{-3}$ decreases in ρ values for methylurea and dimethylurea from 293.15 to 298.15 and 298.15 to 303.15 K, respectively, are noted. Here methylurea with larger decrease proves abnormal behavior with drastic reorientation of molecular forces while dimethylurea with weakening in interaction. Larger weakening for methylurea due to single $-\text{CH}_3$ with much asymmetry to the molecular motions represent a reverse effect of K on σ of methylurea and dimethylurea at lower temperatures. The methylurea at lower and dimethylurea at higher K pro-

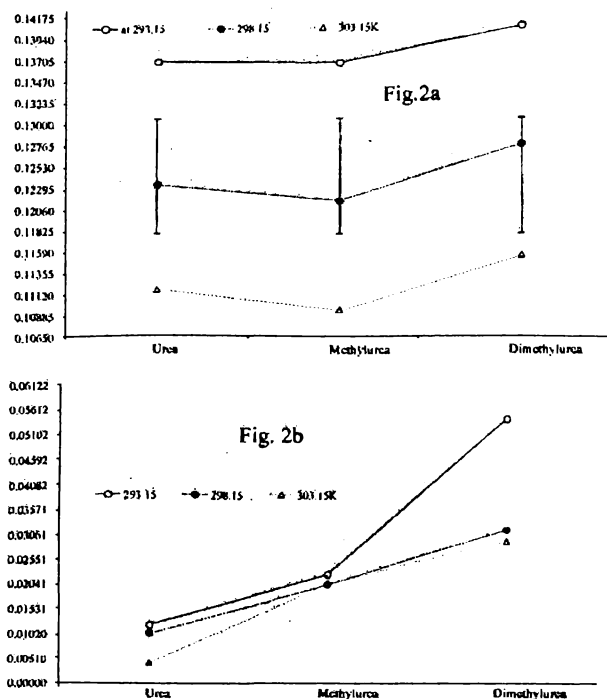


Fig. 2. (a) The plot of the σ_{imf}^0 against m of ureas at three temperature, Y-axis consists σ_{imf}^0 and X-axis ureas; (b) its slope values on Y-axis.

duce slightly lower decrease with stronger hydrophobic interactions. The two $-\text{CH}_3$ show fewer decrease in σ with K while methylurea with weaker hydrophobic interactions larger decreases with larger effect on hydrophilic interactions at higher K. The S_d values are noted as urea $>$ methylurea $>$ dimethylurea proving stronger urea-urea interaction over methylurea-methylurea and dimethylurea-dimethylurea. Thus stronger $-\text{NH}_2$ and $-\text{OC} <$ hydrophilic interactions between urea molecules with subsequent weakening of the $\text{NH} \cdots \text{OC} <$ hydrogen bonds for methylurea and dimethylurea due to $-\text{CH}_3$. The decreases in η^0 and ρ^0 values with K except dimethylurea and methylurea at 293.15 and 293.15 K respectively prove loosening in the hydrogen bonds. The viscous forces for dimethylurea increase and for methyl urea the increase is slightly larger than earlier due to asymmetric structure. It infers that K decreases viscous as well as surface forces. Around 0.10709 and $0.09416 \text{ kg m s}^{-1}$ a decrease in η^0 , and 0.0637 and $0.09478 \text{ kg m s}^{-1}$ increase and 0.10426 and 0.09468 further decrease for urea, methylurea and dimethylurea from 293.15 to 298.15 and 298.15 to 303.15

K, respectively, depict many motions and torque for methylurea with partially hydrophobic and hydrophilic interactions.

Table 4. The frequency factor F and activation energy of viscous from ($E^*/\text{kJ mol}^{-1} \text{K}^{-1}$), for systems evaluated from Arrhenius equation fitting flow times

	F	E^*
U	3560.00002	-6.55087
MU	6103.51477	-7.11842
DMU	2878.12731	-6.35648

The higher F and E^* values¹¹ (Table 4) also support such action of dimethylurea with weakening hydrophobic interaction with K. From urea to methylurea a 0.09421 decrease in η^0 and 0.01498 cP increase for methylurea to dimethylurea at 293.15 K. But from 298.15 to 303.15 the decrease is slightly lower whereas the $-\text{CH}_3$ causes much weakening in frictional forces of ureas at lower K. The S_η values are reported as S_η (methylurea) > S_η (dimethylurea) > S_η (urea) at 293.15 while S_η (dimethylurea) > S_η (methylurea) > S_η (urea) at 298.15 and 303.15 K (Table 2). The latter shows stronger torque for methylurea and dimethylurea due to hydrophobic interactions than of hydrophilic. Thus hydrophilic interactions support mild pairwise interactions over hydrophobic. The methylurea > urea > dimethylurea at 293.15 and 298.15 while methylurea > dimethylurea > urea at 303.15 K sequences of γ^0 values show stronger cohesive forces for methylurea. Hence larger is an element of asymmetry stronger is the frictional and cohesive forces. A lower decrease in γ^0 values with $-\text{CH}_3$ and K proves mild weakening of cohesive forces due to weight of the molecule and weakening in hydrogen bonding. But the forces are higher for methylurea forming the drop of higher weight with polynomial relation against molality, m . The S_γ values for urea are positive which decrease with K, negative for methylurea at 293.15 and 303.15, while positive at 298.15 K. The negative values predict weakening and positive strenthenig of cohesive, respectively.

Intermolecular force coefficient (σ_{imf}^0) :

The dimethylurea > urea > methylurea order depicts a sequence of the strength of σ values with a larger contribution of $-\text{CH}_3$ group. The η magnitude depends on molecular size, length and strength of forces¹²⁻¹⁴. The liquid emerges from a capillary with appreciable kinetic energy for hydrodynamic sphere. Thus Stoke's law⁶ manifests force of attraction holding droplets tending to be-

come sphere with least surface area due to mutual cohesion. The η values define electrostatic and γ cohesive forces of attraction, thus the σ defines the state of electrostatic and dynamics of intermolecular interaction with higher γ values and stronger cohesion. Thereby, σ estimates wetting, detergent, emulsification and capillary rise. For greater accuracy, Poisson-Raleigh formulated a drop formation very slowly at jet due to σ , the quanta of solution that eventually falls off is a function of γ and ρ with kinetic energy flow. Thus σ more accurately predicts binding forces during development of the drop. As the forces are applied on circumference, the lowermost tip of the capillary as well as on the liquid layer being formed and such divisions of the forces being associated with transport properties have never been accounted for either measurement or interpretation. Thereby the studies of σ upto some extent is attempted to focus them in combine. For example around 0.00 to 0.032 kg mol⁻¹ (Fig. 1), the dimethylurea > urea > methylurea sequence of σ signify stronger forces of dimethylurea. The σ values show less precision around 0.0 to 0.015 kg mol⁻¹ and the higher around 0.015 to 0.250 kg mol⁻¹ concentrations. Urea in dilute solutions predicts higher activity with stronger hydrophilic interactions, due to $-\text{NH}_2$ and $>\text{CO}-$ groups. Further two $-\text{CH}_3$ groups cause stronger hydrophobic interactions with bulk water rather than broken. The dimethylurea > methylurea > urea order of σ values around 0.064 to 0.256 kg mol⁻¹ at each K infers stronger intermolecular forces for methylurea than of urea, the $-\text{CH}_3$ of methylurea enhances the methylurea-methylurea interaction due to stronger intermolecular force for hydrophobic interactions rather than of hydrophilic. It confirms cage formation by bulk water around methylurea and dimethylurea. Thus bulk water is more structured rather than of destabilized water around $-\text{NH}_2$ and $>\text{CO}-$ due to hydrogen bonding. Thus substitution of H by $-\text{CH}_3$ weakens hydrogen bond forming strength of $>\text{CO}-$ group (Table 3), the σ for dimethylurea remains same, thus the $-\text{CH}_3$ stabilize an effect of concentration on the intermolecular forces. The σ values for methylurea are higher around 0.180 and 0.128 kg mol⁻¹ (Fig. 1) at 298.15 and 303.15 K. It reflects stronger σ_{imf}^0 for urea and methylurea before and after these concentrations, respectively. Thus $-\text{NH}_2$ and $>\text{CO}-$ cause stronger binding forces around lower concentrations whereas in case of methylurea weaken. But stronger forces around 0.180 and 0.128 kg mol⁻¹ reflect a critical transition in the behavior of urea and methylurea with composition. It shows an inability

of $-\text{CH}_3$ to withstand an influence of concentration on pairwise interactions where the σ effectively undergo changes around lower concentration. The σ values of urea and methylurea are higher by for 298.15 and 303.15 while very low for 298.15 K with much effect of $-\text{CH}_3$ on interacting forces (Fig. 1) at lower K and concentrations. The σ values for dimethylurea, urea and methylurea are considerably higher at each K, and values of σ^0 (Table 3) are listed as dimethylurea > urea > methylurea at each K. It predicts stronger σ_{imf}^0 for dimethylurea with stronger hydrophobic interaction than those of hydrophilic. Thus a substitution of H from $-\text{NH}_2$ by single $-\text{CH}_3$ weakens the hydrogen bonding and hydrophilic interaction of urea. It produces lower water-urea interactions with K, the σ^0 values decrease as $293.15 > 298.15 > 303.15$ K (Fig. 2a). The 0.01353, 0.01512 and 0.01307 decreases in respective values from 293.15 to 298.15 for urea, methylurea and dimethylurea and 0.01175, 0.01207 and 0.01203 Sm^{-1} from 298.15 to 303.15 K, respectively are noted. Comparatively larger decreases at lower temperatures show faster reorientation in molecular motions generating weaker frictional and cohesive forces. However urea at lower temperatures produces comparatively lower decrease for per 5°C increase with weaker solute-solvent interactions with K with larger decrease in forces for 293.15 to 298.15 than of 298.15 to 303.15 K. The decrease in σ^0 for methyl derivatives remains same, thus a substitution by $-\text{CH}_3$ at infinite dilution matters less. Likewise the S_σ values decrease as $293.15 > 298.15 > 303.15$ and increase with $-\text{CH}_3$ (Fig. 2b). It reveals weakening in forces with concentration due to disruption of electrostatic of water. Increase in S_σ with $-\text{CH}_3$ and decrease with K show enhancement of force coefficient with larger decrease at 303.15 K. These prove that the σ^0 and S_σ (Figs. 2a and b) directly measure molecular interaction and resolves micro-changes in the molecular forces. The σ values were fitted to Andrade equation for frequency factor F which is noted as methylurea > urea > dimethylurea. An element of asymmetry in methylurea is high causing millions of molecular motions for optimization. While urea due to symmetry causes less motions and dimethylurea comparatively gains symmetry and mass thus has least values of F with lower motions. But dimethylurea > urea > methylurea sequence of E^* proves that larger sized dimethylurea need higher E^* with stronger torsional forces for Newtonian flow.

Survismeter :

It consists (a) bulbs, (b) limbs and (c) capillaries, and

its sketch diagram is shown in Fig. 3. Bulb number 8 holds about $20 \times 10^{-3} \text{ dm}^3$ solution but $15 \times 10^{-3} \text{ dm}^3$ is filled for measurements and pressure equilibrium. It is connected to limb numbers 3 and 7 of $10 \times 10^{-3} \text{ dm}^3$ adjoining the lower ends of limbs 1, 2 and 4 with an upward flow of solution being sucked to the bulbs 6 and 5, and 9 of limb 4th and 2nd, respectively. Bulb number 6 of $10 \times 10^{-3} \text{ dm}^3$ allows flow within upper and lower marks made on capillaries. The bulb number 5 stabilizes the flow by resisting the pressure changes, bulb 9th is of $10 \times 10^{-3} \text{ dm}^3$, the solution is sucked to above upper mark and released to flow down dropwise through a vertical capillary attached to its lower tube. The drops formed due to cohesive forces at a lower end of the capillary extended to bulb 7, are counted, upper end of limb number 1 is kept open for pressure. The tubes marked with 1, 2, 3 and 4 are limbs, their upper ends are fitted with ground glass joints of \$5 (except tube number 3 with \$9). The \$ represents standard glass joint and numbers along with the \$ size, the stoppers are used to block upper ends of tubes. Limb numbers 1 and 3 are attached to bulb 8th and 7th and limb 2nd and 4th to bulb numbers 5 and 6 for pressure, and bulb number 9 controls air pressure on the

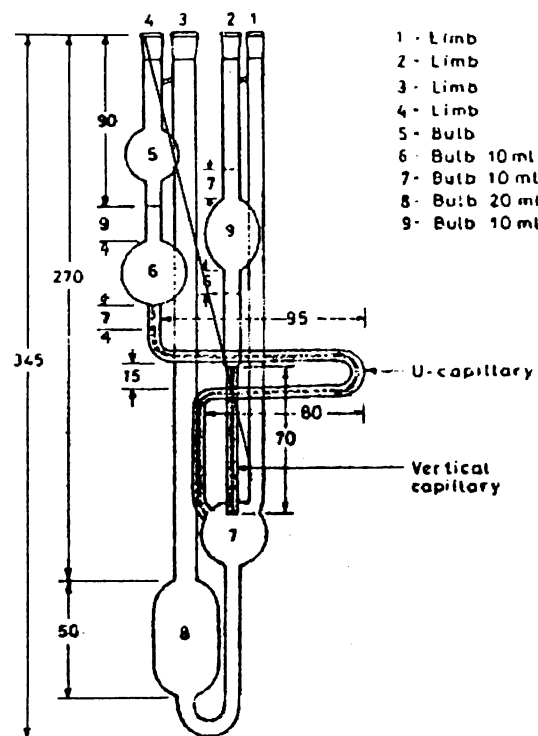


Fig. 3. Survismeter. All dimensions are in mm.

overhead of the top ends of limbs. The (c) represents two capillaries with inner diameter 0.5 mm, the U form, adjoining bulb numbers 6 and 7, and the vertical adjoining bulb numbers 9 and 7th, respectively. The U capillary nullifies effect of gravitational force on the downward flow.

Procedure : A standard procedure is used for cleaning and drying and calibrated as under.

Viscosity : About $15 \times 10^{-3} \text{ dm}^3$ solution of known viscosity is filled in 8th bulb through 3rd limb leaving the other ends of the limbs open. The Survimeter is mounted on SS stand and kept in thermostat. The solution is sucked upward from bulb number 8th through U capillary by glass syringe of $50 \times 10^{-3} \text{ dm}^3$ fitted with airtight plunger. One end of Teflon tube is attached with needle of syringe and the another to a movable stopper with hole. The stopper is fixed in a socket of 4th limb and pulling the plunger out sucks up solution till the bulb number 5th is filled up, thereafter the stopper is removed. During sucking, the socket of limb 3rd is kept open while of 2nd and 1st blocked with stopper and for down flow the syringe and stopper of limb number 1st are removed. The flow time within the marks of bulb number 6th is counted with 0.01 s electronic racer and η is calculated with eq. (1).

$$\eta = (\rho t / \rho_0 t_0) \eta_0 \quad (1)$$

The η and η_0 viscosities, t and t_0 flow times, and ρ and ρ_0 densities of solution and solvent, respectively.

Surface tension :

After flow time, the solution is sucked to the bulb number 9th above its upper mark blocking the socket of the limbs 4th and 1st. For dropwise flow from the bulb number 9th, the stoppers are removed followed by syringe and three concordant drop numbers are counted in the bulb 7th. A nut bolt fitted clip is used for air pressure on overhead of the limb number 2nd and surface tension is calculated with eq. (2).

$$\gamma/\gamma_0 = (n_0\rho)/(n\rho_0) \quad (2)$$

The γ and γ_0 surface tensions, n and n_0 number of drops, respectively. From eqs. (1) and (2), eq. (3) is derived.

$$(\eta/\gamma) = (\eta_0/\gamma_0)(tn/t_0n_0) \quad (3)$$

The $(\eta/\gamma) = \sigma/\text{Sm}^{-1}$ and $(\eta_0/\gamma_0) = \sigma_0$ for solvent, eq. (3) now becomes eq. (4), the Man Singh eq.

$$\sigma = \sigma_0[(t/t_0 \pm B/t)((n/n_0) + 0.0012(1 - w/w_0))] \quad (4)$$

The B/t is kinetic energy correction to viscous flow, $0.0012 \times 10^3 \text{ kg m}^{-3}$ air density and $(1 - w/w_0)$ buoyancy correction to w and w_0 the weights of solution and solvent, respectively. The γ is $w = mg = 2\pi r\gamma$, thus buoyancy correction is made. The B/t and kinetic correction (k) were obtained from $\eta = \rho(k - B/t)$ with known η and ρ values. The B/t and k values are found to be -0.1821×10^{-4} and 1.8977×10^{-4} and -0.20508×10^{-5} and 2.32647×10^{-5} at 298.15 and 303.15 K, respectively. Survimeter constant B with distilled water is calculated from $\eta/\rho = Bt - V/8\pi rLt$, the V total volume of water flows, r radius and L is length of capillary. The densities were measured with $20 \times 10^{-3} \text{ dm}^3$ bicapillary pycnometer and 0.01 mg Dhona balance model 100DS, the σ data were fitted in a first order linear equation given under.

$$\sigma = \sigma^0 + S_\sigma m \quad (5)$$

The σ^0 is limiting, and S_σ slope values plotted in Fig. 2(a and b).

Conclusion :

Thus symmetry and $-\text{CH}_3$ contribute their shares to σ_{imf}^0 , temperature and concentration and show many influences on solute-solvent and pairwise interactions. The $-\text{CH}_3$ groups enhance the hydrophobic interaction and σ_{imf}^0 with higher values.

The both σ^0 and S_σ display weaker effect at 303.15 K with similar effect of K and concentration. The $-\text{CH}_3$ groups weaken the hydrogen bonding strength of the $>\text{CO}-$ group and enhances hydrophobic interactions at each K.

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