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Apparent molar volumes of Aspirin in water at temperatures from 298.15 to 313.15 K

Bhujbal Ravindra C

Department of Chemistry, Hon. B. J. College, Ale, Pune-412411. (M. S.) India,Affiliated to University of Pune, Maharashtra State, India.
Email : rcbhujbal@gmail.com | Mobile: 9890460746

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ABSTRACT
Apparent molar volumes (ϕ_v) and viscosity B-coefficients for aspirin solutions in pure water system have been determined from density (ρ) and viscosity (η) measurements at 298.15 to 313.15 K using a pycnometer and Ubbelohde viscometer respectively. Eight different concentrations ranging from 0.0040 to 0.0145 M were prepared. The apparent molar volumes were calculated from the density data. In addition, the concentration dependence of the apparent molar volumes was examined using Masson's equation. The Jones-Dole equation was used to analyze viscosity data to obtain viscosity 'A' and 'B' coefficients. The drug interacts with various ions or molecules or biological membranes present in the biological system are an important phenomenon. The parameters derived from these equations have been interpreted in terms of solute-solute and solute-solvent interactions.

Keywords: Aspirin, density, viscosity, B-coefficient.

INTRODUCTION

The partial molar volume and the related volumetric parameters of drug compounds in dilute aqueous solutions at different temperatures and pressures have been investigated by several authors [1-4]. It is well known that physicochemical characterization of drugs plays a crucial role in all the stages associated to design and development of pharmaceutical dosage forms, especially those intended to parenteral administration [5].

In this context, as a contribution to generation and systematization of physicochemical information about drugs' aqueous behavior, the main goal of this study was to evaluate the effect of concentration and temperature on the apparent molar volume of drugs in water. With that purpose, an interpretation in terms of solute-solvent interactions based on the corresponding volumetric behavior was developed.

Drugs of the analgesics, antipyretics, and anti-inflammatory class include a heterogeneous group of compounds. Many of these agents affect pain, fever and inflammation and are referred to as the non-steroidal anti-inflammatory drugs (NSAIDS).

Non-narcotic analgesics have three important properties namely analgesics, antipyretics and anti-inflammatory (e.g. Aspirin and paracetamol). The non-narcotics (salicylates) are called aspirin like or Non-steroidal Anti-inflammatory Drugs (NSAIDS). These drugs have common mechanism of inhibiting the cyclooxygenase (COX), the key enzyme responsible for biosynthesis of Prostaglandins (PG).

Bio-pharmaceutics is the study of factors influencing the extent and rate of absorption. The rate and amount of drug absorption depends on biological and physicochemical factors. During their way to site of action, drug molecules have to cross one or more membranous barrier, which are lipoidal in nature and have different size of pores.

Physicochemical factors include lipid solubility, salt complexation, dissolution rate, Viscosity and drug stability in GIT. Lipid soluble drugs more unionized and easily absorbed Na and K salts of weak acid have higher absorption rate than acids.

All the drugs in any solid dosage form or suspension when administered will first change into drug solution in body fluids. So, dissolution rate is important factor affecting the rate of absorption. When a drug is more rapidly or completely absorbed from solution, it is very likely that its absorption will be dissolution limited.

Viscosity limits the dissolution rate and there by affect the rapid absorption. Eg. Aqueous Solution of Na-Salicylate showed its rapid appearance in plasma while the same drug in suspension form failed to reach the target as quickly as with aqueous solution [6].

The study of the volumetric behavior of electrolytes in solution provides information useful to elucidate ion–ion, ion–solvent, and solvent–solvent interactions. The concentration dependence of the apparent and partial molar volumes can be used to study ion–ion interactions, whereas the partial molar volumes at infinite dilution provide information on ion–solvent interactions. The data reported here were obtained by performing density measurements on aqueous solutions of aspirin.

METHODOLOGY

Materials:
Aspirin of high purity was recrystallized and then used. Deionized water with a specific conductance of $< 10^{-6}$ $S.cm^{-1}$ was used for the preparation of solutions at room temperature in a molarity range (4.0×10^{-3} to 1.45×10^{-2}) $mol.L^{-1}$. The precision of balance used was ± 1×10^{-5}g.

Density measurements:
The pycnometer was calibrated by measuring the densities of triple distilled water. The densities of distilled organic liquids like acetone, alcohol, benzene, carbon tetra chloride, aniline, and nitrobenzene were evaluated with respect to density of water. The density was measured with an uncertainty of ± 1.48 $\times10^{-4}g.cm^{-3}$.

Viscosity measurements:
The solution viscosities were measured with an uncertainty of ± 2.4×10^{-4} mPa.s by using Ubbelohde viscometer. The viscosity measurements were performed at 25, 30, 35, 37 and 40°C. The temperature of thermostat is maintained to desired temperature, by using demerstat with an accuracy of ± 0.1 K. The flow time will be measured at the accuracy of ± 0.01 s.

The different compositions (0.0145M to 0.0040M) of solutions of aspirin were prepared in pure water. The

viscosities were measured at 25, 30, 35, 37 and 40°C for seven different concentrations.

DATA EVALUATION

The apparent molar volumes, ϕ_v, were obtained from the density results using the following equation[7-10]

$$\Phi_v = \frac{1000(\rho_0 - \rho)}{C\rho_0} + \frac{M_2}{\rho} \qquad (1)$$

where M_2, C, ρ and ρ_0 are the molar mass of the aspirin, concentration (mol.L^{-1}), and the densities of the solution and the solvent, respectively.

The apparent molar volumes (ϕ_v) were plotted against the square root of concentration ($C^{½}$) in accordance with the Masson's equation[11]

$$\phi_v = \Phi_v^0 + S_v.C^{½} \qquad (2)$$

Where Φ_v^0 is the limiting apparent molar volume and S_v a semi-empirical parameter which depends on the nature of solvent, solute and temperature.

The viscosity results for the aqueous solutions of drugs were plotted in accordance with Jones-Dole equation [12]

$$\frac{\eta_r - 1}{C^{½}} = A + BC^{½} \qquad (3)$$

Where $\eta_r = (\eta/\eta_o)$ and η, η_o are viscosities of the solution and solvent respectively, C is the molar concentration. The linear plots for $(\eta_r - 1)/C^{½}$ versus $C^{½}$ were obtained for the aspirin. The B-coefficients were obtained from the linear plots using the least-square fitting method. The A- coefficient reflects solute-solute interaction[13] and the B-coefficient reflect the solute-solvent interactions.

RESULTS AND DISCUSSION

The values of the densities (ρ) and apparent molar volumes (ϕ_v) of aspirin solution in pure water at 298.15, 303.15, 308.15, 310.15 and 313.15K temperature are shown in Table 1. In all sets the densities and ϕ_v values of solutions increases with increase in concentration of solution. Figure 1 shows the linear plots of ϕ_v vs $C^{½}$ for aspirin solutions in pure water at different temperatures. Masson's parameters Φ_v^0 and S_v were obtained from linear plots are reported in table 2. The Φ_v^0 values are negative and S_v values obtained are positive for the systems studied furnish important information regarding the solute-solvent and solute-solute interactions.

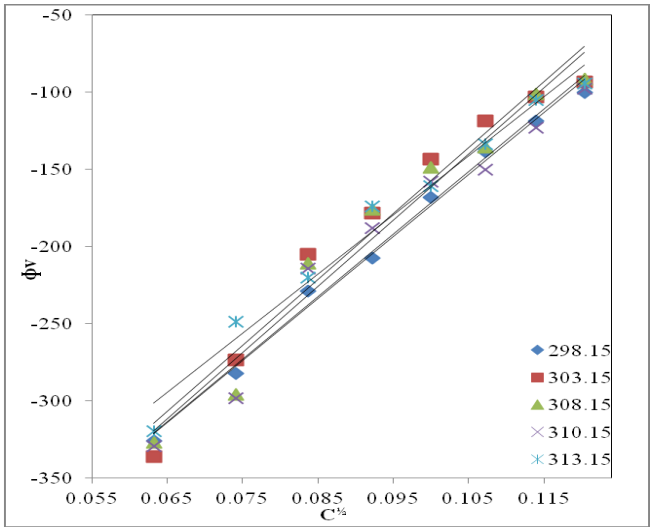

Fig 1: Plot of Φ_v (cm^3.mol^{-1}) Vs $C^{½}$ (mol$^{1/2}$.dm$^{-3/2}$) for aspirin solutions in pure water at temperature range T= (298.15 to 313.15K).

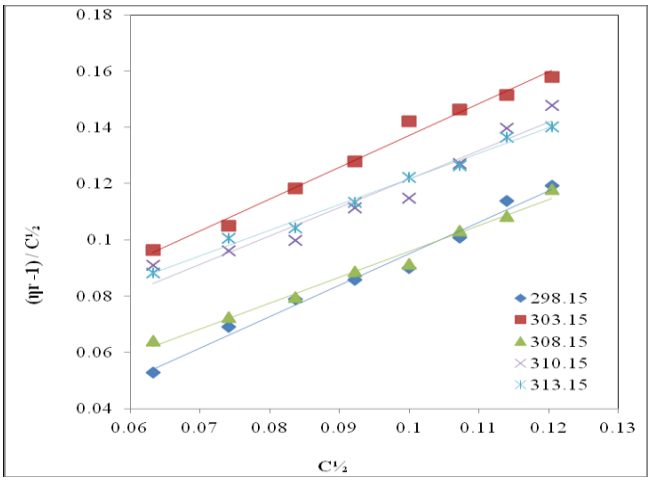

Fig2: Plot of $(\eta_r - 1)/C^{½}$ versus $C^{½}$ for aspirin solutions in pure water at different temperatures.

The values of the viscosity and relative viscosities of aspirin in pure distilled water at 298.15, 303.15, 308.15, 310.15 and 313.15K temperature are given in Table 3.

In all sets the viscosities of solutions increases with increase in concentration of solution, while viscosity decreases with increase in temperature. Figure 2 shows the variation of $(\eta_r-1)/C^{1/2}$ against square root of concentration at different temperatures in pure water.

'A' is constant independent of concentration and 'B' is Jones-Dole coefficient represents measure of order and disorder introduced by solute into the solution; positive 'B' coefficient shows strong alignment of solvent towards solute and is related to the effect of the ions on the structure of water[14]. The Jones-Dole parameters are given in Table 4. The positive values of 'B' at all temperatures indicate water structuring[15]. The values of B-coefficient decreases with increase in temperature as shown in fig.3 to indicate structure promoting tendency of the compound[16].

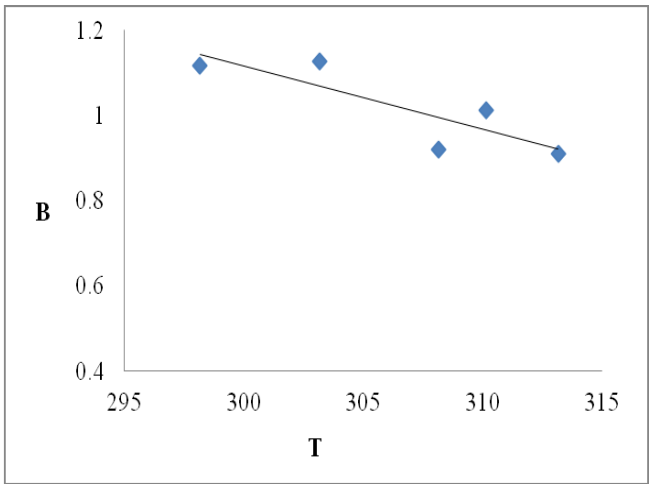

Fig 3: Plot of B-coefficient versus temperature for aspirin solutions in pure water at different temperatures.

Table 1: Densities and apparent molar volumes of aspirin solution in pure water at different temperatures.

Molar Conc. of aspirin (C) mol/dm^3	Temperatures				
	298.15K	303.15K	308.15K	310.15K	313.15K
Densities (ρ), (g.cm^{-3})					
0.0040	0.99906	0.99776	0.99609	0.99536	0.99422
0.0055	0.99958	0.99819	0.99668	0.99595	0.99458
0.0070	0.99990	0.99839	0.99680	0.99608	0.99502
0.0085	1.00033	0.99874	0.99709	0.99645	0.99523
0.0100	1.00052	0.99893	0.99735	0.99670	0.99563
0.0115	1.00070	0.99913	0.99769	0.99712	0.99583
0.0130	1.00092	0.99938	0.99772	0.99726	0.99593
0.0145	1.00110	0.99966	0.99800	0.99735	0.99620
Apparent molar volumes (Φ_v) (cm^3.mol^{-1})					
0.0040	-325.80	-336.29	-326.78	-329.54	-319.82
0.0055	-282.49	-273.74	-296.14	-298.19	-249.05
0.0070	-229.09	-205.01	-211.09	-214.12	-220.12
0.0085	-207.51	-178.25	-176.18	-188.15	-174.13
0.0100	-168.34	-143.46	-148.72	-157.89	-161.09
0.0115	-138.51	-118.61	-135.43	-150.41	-133.92
0.0130	-118.65	-103.36	-101.21	-122.97	-105.27
0.0145	-100.14	-93.345	-91.417	-97.733	-94.366

Table 2: Masson's parameters Φ_v^0 ($cm^3.mol^{-1}$) and S_v ($cm^3 .mol^{-3/2} .L^{1/2}$) of aspirin solutions in pure water solvent system at different temperatures.

Temperature (K) →	**298.15**	**303.15**	**308.15**	**310.15**	**313.15**
Φ_v^0 ($cm^3.mol^{-1}$)	-575.3	-584.5	-591.1	-577.8	-543.6
S_v ($cm^3 .mol^{-3/2} .L^{1/2}$)	4017	4271	4394	4058	3829

Table 3 : Viscosities and relative viscosities of aspirin solution in pure water at different temperatures.

Molar Conc. of aspirin (C) mol/dm^3	Temperatures				
	298.15K	303.15K	308.15K	310.15K	313.15K
Viscosities (η) ($Nm^{-3}.s$)					
0.0040	0.89665	0.80560	0.72544	0.69868	0.65966
0.0055	0.89824	0.80696	0.72640	0.69963	0.66089
0.0070	0.89957	0.80865	0.72732	0.70048	0.66172
0.0085	0.90074	0.81016	0.72842	0.70181	0.66286
0.0100	0.90172	0.81211	0.72912	0.70265	0.66402
0.0115	0.90334	0.81329	0.73051	0.70415	0.66490
0.0130	0.90526	0.81456	0.73144	0.70575	0.66621
0.0145	0.90651	0.81595	0.73277	0.70705	0.66708
Relative viscosities (η_r)					
0.0040	1.00333	1.00609	1.00405	1.00576	1.00558
0.0055	1.00511	1.00779	1.00538	1.00713	1.00745
0.0070	1.00660	1.00990	1.00666	1.00835	1.00872
0.0085	1.00791	1.01179	1.00818	1.01026	1.01046
0.0100	1.00901	1.01422	1.00915	1.01147	1.01223
0.0115	1.01082	1.01570	1.01107	1.01363	1.01357
0.0130	1.01297	1.01728	1.01236	1.01594	1.01556
0.0145	1.01437	1.01902	1.01420	1.01781	1.01689

Table 4 : Jones-Dole parameters of aspirin solutions in pure water.

Parameters of Jones-Dole equation					
T (K)	298.15	303.15	308.15	310.15	313.15
A ($dm^{3/2}mol^{-1/2}$)	-0.016	0.024	0.003	0.020	0.030
B ($dm^3.mol^{-1)}$	1.118	1.128	0.920	1.013	0.911

CONCLUSION

In the present report, from volumetric and viscometric study of aqueous solutions of aspirin at different temperatures are systematically presented. The densities of pain killer solutions under investigation decrease with increase in temperature and increases with increase in concentration. It has been observed that there exist strong solute–solvent interactions in these systems, which increases with increase in pain killer concentration.

The negative Φ_v^0 and positive S_v values shows that aspirin will be considerably associated in presence of ions. Since S_v is measure of solute-solute interactions. S_v values do not change systematically with change in temperature, and hence it suggests that the solute-solute interactions are insensitive to change in temperature. The positive values of Jones-Dole coefficient 'B' indicates structure promoting tendency and strong interactions between solute and solvent. Positive values of 'B' suggesting strongly hydrated solute indicating structure promoting tendency i.e. kosmotropes (structure makers). The Jones-Dole and Masson's equations are found to obey the aspirin in pure water system.

Conflicts of interest: The authors stated that no conflicts of interest.

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