

# **Study of flow and interaction parameters of Aspirin in NS, DNS, RL, and pure water system at 298.15K, 303.15K, 308.15K, 310.15K and 313.15K.**

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**ABSTRACT:** The measurements of densities and viscosities of drug aspirin in pure water NS (Normal Saline), DNS (Dextrose with Normal Saline) and RL (Ringer lactate) systems have been made at 298.15K, 303.15K, 308.15K, 310.15K and 313.15K using a bi-capillary pycnometer and Ubbelohde viscometer. The density and viscosity data have been analyzed and used to explain the flow and molecular interaction parameters of drug aspirin in aqueous solutions as well as in presence of additives. Jones-Dole equation, Masson equation, Roots equation and Moulik equation have been verified at different temperatures. The drug interact with various ions, molecules, biological membranes present in the biological system is an important phenomenon and can be understood with the evaluated parameters.

**Keywords:** Aspirin, density, viscosity, partial molar volume, B-coefficient.

## **I.INTRODUCTION**

In solution chemistry the interpretation of molecular interactions are based on the theories like Van't Hoff, Gibbs, Debye-Hückel and Onsagar. The simple physical properties like density and viscosity play important role to explain the molecular interaction in aqueous solutions. Drugs which alter the pain sensitivity or removes pain from body parts are called pain killers or analgesics.

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 thereby affects the rapid absorption. Surender Singh<sup>1</sup> showed rapid absorption of aqueous sodium salicylate in plasma than in aqueous solution.

Acetylsalicylic acid belongs to class of drug referred to as non-steroidal anti-inflammatory drugs. Aspirin is clinically useful as analgesics, anti-inflammation, antipyretic, anti-thrombolytic and anti-rheumatic. Acetylsalicylic acid act by inhibiting the cyclooxygenase (COX) activity resulting in decrease synthesis of prostaglandin, leukotriene and thromboxane precursors which is the ubiquitous enzyme that catalyze the initial step in the synthesis of prostanoids .

It is useful in the treatment of pain, inflammation, and rheumatics' conditions. It is also useful in preventing formation of thrombus in certain cardiovascular diseases.<sup>2</sup> Importance of such drugs create an interest to study flow and interaction parameters at different temperatures. Such information play important role in pharmaceutical and medicinal chemistry.

The concentration dependence of the thermodynamic properties is a measure of solute-solute interaction and in the limit of infinite dilution these parameters serve as a measure of solute-solvent interactions<sup>3</sup>. In solution, the solute

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changes the water structure, as a result it also changes the solution viscosity. The variation of viscosity of solution with solute concentration is related to the size of the solute, nature of the solute as well as solute-solvent interaction.<sup>4</sup>

The drug-water molecular interaction and their temperature dependence play an important role in the understanding of drug action.<sup>5</sup> Transport property measurements are a powerful tool to study the behavior of various solutes in solutions. Drug actions i.e. drug reaching the blood stream, its extent of distribution, its binding to the receptors and finally producing the physiological action, all depend on various physicochemical properties chiefly decided by various interactions like ionic or covalent, charge transfer, hydrogen bonding, ion-dipole interactions, or hydrophobic interactions. The transport property measurements throw light on solute-solvent interactions which correlate with the structure making-breaking property of the solutes and solvents.<sup>6</sup>

The parameters like apparent molar volume, density, viscosity A and B coefficient and Jone-Dole parameters are useful to focus the solute-solvent interactions and to understand different biochemical reactions at 310.15 K i.e. at body temperature. It also enables to enrich the data at various composition and temperatures.

## II. EXPERIMENTAL

### A. Materials:

Aspirin of high purity was obtained re-crystallized and then used. De-ionized water with a specific conductance of  $< 10^{-6}$  S.cm<sup>-1</sup> was used for the preparation of solutions at room temperature in a molarity range ( $4 \times 10^{-3}$  to  $1.45 \times 10^{-2}$ ) mol.L<sup>-1</sup>. The precision of balance used was  $\pm 1 \times 10^{-5}$ g.

### B. Density measurements:

The bi-capillary pycnometer is calibrated by measuring the density of triple distilled water. The different concentrations of aspirin were prepared in pure water in NS, DNS and RL. The density measurements were carried out at various temperatures. The uncertainties in the reported densities are of  $\pm 1.13 \times 10^{-4}$ g.cm<sup>-3</sup>.

### C. Viscosity measurements:

The solution viscosities are measured with an uncertainty of  $\pm 2.4 \times 10^{-4}$  mPa.s by using Ubbelohde viscometer. The viscosity measurements were performed at T = (298.15 to 313.15) K. The temperature of thermostat was maintained with an accuracy of  $\pm 0.01$  K. The flow time will be measured at the accuracy of  $\pm 0.01$  s.

The six-seven compositions in the range (0.0145M to 0.0040M) of solutions of aspirin were prepared in NS, DNS, RL and pure water.

### D. Data evaluation:

The apparent molar volume,  $\phi_v$ , was obtained from the observed densities using the following equation<sup>7-10</sup>

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

Where  $M_2$ , C,  $\rho$  and  $\rho_0$  are the molar mass of the aspirin, concentration (mol.L<sup>-1</sup>), and the densities of the solution and the solvent, respectively.

The apparent molar volume  $\phi_v$  was plotted against the square root of concentration ( $C^{1/2}$ ) in accordance with the Masson's equation<sup>11</sup>

$$\phi_v = \phi_v^0 + S_v C^{1/2} \quad (2)$$

Where  $\phi_v^0$  is the limiting apparent molar volume and  $S_v$  a semi-empirical parameter which depends on nature of solvent, solute, and temperature. Its value for large organic solutes is not of much significance<sup>5</sup>.

The observed viscosities for the aqueous solutions of aspirin were plotted in accordance with Jones-Dole equation<sup>12</sup>

$$\frac{\eta_r - 1}{C^{1/2}} = A + BC^{1/2} \quad (3)$$

Where  $\eta_r = (\eta/\eta_0)$  and  $\eta$ ,  $\eta_0$  are viscosities of the solution and solvent respectively, C is the molar concentration. The linear plots for  $(\eta_r - 1)/C^{1/2}$  versus  $C^{1/2}$  were expected and 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<sup>13</sup> and the B-coefficient,

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solute-solvent interactions. The interactions can also be evaluated from following models such as Moulik<sup>14</sup> and Root<sup>15</sup> equation

$$\eta_r^2 = M + KC^2 \tag{4}$$

$$(d - d_0)/C = A - B C^{1/2} \tag{5}$$

### III. RESULTS AND DISCUSSION

In table 1, the values of the densities, apparent molar volumes and viscosities of aspirin in NS, DNS, RL and pure distilled water are reported. In all sets the densities and viscosities of solutions increases with increase in concentration of solution. At higher temperatures the densities and viscosities becomes smaller. The apparent molar volumes  $\phi_v$ 's are calculated by using equation 1. The linear dependence of  $\phi_v$  on concentration is observed for all the concentrations investigated. Figure 1-4 shows plots of apparent molar volume  $\phi_v$  of aspirin against square root of concentration over the temperature range 293.15 K to 313.15K for different solvent systems..

The partial molar volume  $\phi_v^0$  have been calculated from the intercept of linear plots using equation 2. The  $\phi_v^0$  values are listed in table 2. The  $\phi_v^0$  values for set 2 and 3 are positive, while that for set 1 and 4 are negative. The  $\phi_v^0$  value provides information regarding the ion-solvent interactions, drug hydrophobicity and hydration properties. The lower negative  $\phi_v^0$  values suggest stronger hydrogen bonding<sup>5</sup>.

The  $\phi_v^0$  values increases with rise in temperature for all sets performed<sup>16</sup>, except set 3. The positive values of  $S_v$  obtained from the slope indicate strong inter and intra molecular interactions for set 1 and 4. The magnitude of  $S_v$  do not show specific trend with temperature. The strong solute-solvent interactions reflects weak solute-solute interactions.

**Table 1:** Density, apparent molar volume and viscosity of aspirin in NS, DNS, RL and pure distilled water at different temperatures.

SET 1 in NS					SET 2 in DNS				
T /K	C/mole/lit	$\rho$ /(gm/cc)	$\eta$ / mPa.s	$\phi_v$ /cm <sup>3</sup> mol <sup>-1</sup>	T /K	C/mole/lit	$\rho$ /(gm/cc)	$\eta$ / mPa.s	$\phi_v$ /cm <sup>3</sup> mol <sup>-1</sup>
298.15	0.0055	1.00502	0.90366	-103.12	298.15	0.0055	1.01005	0.87155	2728.059
	0.007	1.00509	0.90558	-52.515		0.007	1.0124	0.88415	1853.452
	0.0085	1.00519	0.90716	-23.289		0.0085	1.01365	0.89547	1413.855
	0.01	1.00529	0.9088	-2.8302		0.01	1.01595	0.90562	1003.641
	0.0115	1.00541	0.91086	10.5583		0.0115	1.01811	0.91567	712.3232
	0.013	1.00561	0.91335	14.7244		0.013	1.02011	0.9301	500.2467
	0.0145	1.00581	0.91485	18.0287		0.0145	1.02273	0.95348	290.3091
303.15	0.0055	1.00366	0.8055	-114.15	303.15	0.0055	1.00795	0.83998	2808.955
	0.007	1.00372	0.80623	-59.718		0.007	1.01052	0.85051	1885.81
	0.0085	1.00382	0.80736	-29.192		0.0085	1.01285	0.86149	1316.088
	0.01	1.0039	0.80836	-5.8281		0.01	1.01533	0.87657	902.6164
	0.0115	1.0041	0.81011	1.02747		0.0115	1.01701	0.89085	665.0241
	0.013	1.00416	0.81156	17.0483		0.013	1.01877	0.90101	476.2438
	0.0145	1.0043	0.81231	24.2485		0.0145	1.02061	0.91874	321.127
308.15	0.0055	1.00199	0.72609	-123.44	308.15	0.0055	1.00684	0.76336	2699.801

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	0.007	1.00206	0.72675	-68.39		0.007	1.00923	0.77754	1824.681
	0.0085	1.00213	0.72771	-32.771		0.0085	1.01135	0.78286	1289.537
	0.01	1.0022	0.72812	-7.8375		0.01	1.01409	0.79498	854.213
	0.0115	1.00229	0.72896	8.85282		0.0115	1.01601	0.80429	602.2885
	0.013	1.00272	0.72989	-4.454		0.013	1.01895	0.81874	331.6535
	0.0145	1.00326	0.73122	-22.591		0.0145	1.02097	0.83981	179.1546
310.15	0.0055	1.00134	0.70172	-152.64	310.15	0.0055	1.00589	0.73115	2735.672
	0.007	1.00141	0.70279	-91.313		0.007	1.0082	0.74476	1863.846
	0.0085	1.0015	0.70382	-53.984		0.0085	1.01111	0.7533	1230.536
	0.01	1.00158	0.70516	-26.853		0.01	1.0129	0.76668	896.9958
	0.0115	1.00162	0.70631	-3.3199		0.0115	1.01576	0.78001	559.2696
	0.013	1.00171	0.70714	10.9346		0.013	1.01801	0.78922	345.4719
	0.0145	1.00262	0.70794	-34.34		0.0145	1.02024	0.79473	177.2601
313.15	0.0055	0.99961	0.66465	-54.477	313.15	0.0055	1.004	0.69226	2846.196
	0.007	1.00005	0.66614	-67.096		0.007	1.00707	0.70208	1843.773
	0.0085	1.00026	0.6679	-48.156		0.0085	1.00921	0.7154	1302.523
	0.01	1.00026	0.6691	-13.863		0.01	1.01201	0.72099	858.8757
	0.0115	1.00035	0.67077	3.64438		0.0115	1.01455	0.73366	553.1505
	0.013	1.00056	0.67193	7.86552		0.013	1.01695	0.74146	328.5461
	0.0145	1.00167	0.67295	-50.96		0.0145	1.01955	0.75535	136.8752

**Table 1:** Continued....

SET 3 In RL					SET 4 In D.W.				
T /K	C/mole/lit	$\rho$ /(gm/cc)	$\eta$ / mPa.s	$\phi_v$ /cm <sup>3</sup> mol <sup>-1</sup>	T /K	C/mole/lit	$\rho$ /(gm/cc)	$\eta$ / mPa.s	$\phi_v$ /cm <sup>3</sup> mol <sup>-1</sup>
298.15	0.0055	1.00175	0.89785	453.228		0.0055	0.99958	0.89824	-282.494
	0.007	1.0023	0.89919	316.272		0.007	0.9999	0.89957	-229.09
	0.0085	1.00281	0.90113	232.344		0.0085	1.00033	0.90074	-207.513
	0.01	1.00373	0.90273	132.727		0.01	1.00052	0.90172	-168.338
	0.0115	1.00447	0.90446	74.6991		0.0115	1.0007	0.90334	-138.511
	0.013	1.00493	0.90555	51.5305		0.013	1.00092	0.90526	-118.653
	0.0145	1.00562	0.90705	17.3448		0.0145	1.0011	0.90651	-100.136
303.15	0.0055	1.00038	0.8077	471.9481	303.15	0.0055	0.89824	0.80696	-273.744
	0.007	1.00071	0.80878	362.2962		0.007	0.89957	0.80865	-205.007

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	0.0085	1.00156	0.81072	230.29		0.0085	0.90074	0.81016	-178.254
	0.01	1.00248	0.8128	130.8995		0.01	0.90172	0.81211	-143.457
	0.0115	1.00331	0.81401	65.24755		0.0115	0.90334	0.81329	-118.611
	0.013	1.00356	0.81625	59.27282		0.013	0.90526	0.81456	-103.361
	0.0145	1.00417	0.81863	29.75596		0.0145	0.90651	0.81595	-93.3448
308.15	0.0055	0.99872	0.73242	463.6665	308.15	0.0055	0.99668	0.7264	-296.142
	0.007	0.99913	0.73284	344.3493		0.007	0.9968	0.72732	-211.092
	0.0085	0.99978	0.73475	238.9166		0.0085	0.99709	0.72842	-176.179
	0.01	1.00082	0.7368	126.1247		0.01	0.99735	0.72912	-148.722
	0.0115	1.0015	0.73845	74.05231		0.0115	0.99769	0.73051	-135.426
	0.013	1.0019	0.74184	55.52907		0.013	0.99772	0.73144	-101.209
	0.0145	1.00273	0.74286	11.19135		0.0145	0.998	0.73277	-91.4166
310.15	0.0055	0.99807	0.69865	433.1212	310.15	0.0055	0.99595	0.69963	-298.193
	0.007	0.99841	0.69985	330.3384		0.007	0.99608	0.70048	-214.125
	0.0085	0.99906	0.70091	227.3416		0.0085	0.99645	0.70181	-188.154
	0.01	0.99978	0.70225	148.24		0.01	0.9967	0.70265	-157.893
	0.0115	1.00078	0.70311	65.41271		0.0115	0.99712	0.70415	-150.408
	0.013	1.00096	0.70476	64.81038		0.013	0.99726	0.70575	-122.968
	0.0145	1.00186	0.70628	14.65067		0.0145	0.99735	0.70705	-97.7333
313.15	0.0055	0.99692	0.66303	439.0707	313.15	0.0055	0.99458	0.66089	-249.048
	0.007	0.99733	0.66428	324.9852		0.007	0.99502	0.66172	-220.122
	0.0085	0.99798	0.66605	222.8829		0.0085	0.99523	0.66286	-174.134
	0.01	0.99902	0.66697	112.3465		0.01	0.99563	0.66402	-161.092
	0.0115	0.99963	0.66865	68.09913		0.0115	0.99583	0.6649	-133.924
	0.013	0.99982	0.67044	66.42411		0.013	0.99593	0.66621	-105.273
	0.0145	1.00064	0.67154	21.57513		0.0145	0.9962	0.66708	-94.3663

**Table 2:** The flow and interaction parameters of aspirin solutions in the temperature range (298.15 to 313.15K).

	T /K	Masson equation		Roots equation		Jone-Dole equation		Moulik equation	
		$\phi v^0$	Sv	A	B	A	B	M	K
Set 1	298.15	-270.5	2538	0.451	-2.546	-0.397	3.797	0.982	136.3
	303.15	-304.2	2844	0.485	-2.850	-0.676	4.966	0.956	95.37
	308.15	-263.5	2304	0.443	-2.305	-0.559	4.119	0.954	72.62

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	310.15	-341.5	2948	0.521	-2.945	-0.489	4.088	0.975	97.05
	313.15	-134.2	1036	0.314	-1.034	-0.306	3.301	0.990	137.3
Set 2	298.15	6218	-50640	-6.190	51.87	-4.202	30.85	0.685	717.8
	303.15	6323	-51885	-6.287	53.06	-3.190	25.82	0.794	878.2
	308.15	6361	-53131	-6.315	54.24	-3.002	24.80	0.807	905.3
	310.15	6439	-53876	-6.389	54.96	-2.994	24.68	0.813	839.4
	313.15	6694	-56373	-6.641	57.44	-2.838	23.71	0.823	851.8
Set 3	298.15	1113	-9421	-0.937	9.452	-0.293	2.679	0.983	112.1
	303.15	1174	-9930	-0.996	9.950	-0.503	4.066	0.966	145.7
	308.15	1167	-9918	-0.987	9.915	-0.774	5.650	0.942	164.0
	310.15	1090	-9179	-0.909	9.174	-1.018	6.406	0.916	108.9
	313.15	1079	-9108	-0.897	9.092	-0.748	5.288	0.943	136.2
Set 4	298.15	-565.2	3920	0.743	-3.909	-0.13	1.090	1.007	103.5
	303.15	-534.9	3800	0.712	-3.784	0.022	1.144	1.014	123.3
	308.15	-574.6	4138	0.751	-4.113	0.00	0.964	1.008	96.98
	310.15	-558.9	3878	0.735	-3.852	0.005	1.156	1.011	121.1
	313.15	-501.7	3432	0.678	-3.405	0.030	0.914	1.012	107.0

The viscosities ( $\eta$ ) of solutions are listed in table 1. The  $\eta$  values increase with concentration and decrease with rise in temperature. This suggests the existence of molecular interactions occurring in the system. The viscosity data have been analyzed by using Jones –Dole equation 3. Where  $\eta$  and  $\eta_0$  are the viscosity of solute and solvent, respectively. The linear plots were observed for Jones Dole, Massons, Roots and Moulik equations. (figure 1-4). The flow and interaction parameters are reported in table no. 2.

Figure 2 shows the variation of  $(\eta_r - 1)/C^{1/2}$  against square root of concentration over the temperature range 293.15 K to 313.15K for different sets performed. 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<sup>17,18</sup>. The strong interaction immobilizes the neighboring solvent molecules and presents large obstruction to viscous flow of solution thereby increasing viscosity. Thus the present system behaves as structure maker.

The parameters obtained from Masson equation, Root’s equation, Jones-Dole equation, Moulik equation are listed in table 2. The observed values of A are negative and of B are positive. The positive values of B at all temperatures indicate water structuring<sup>17</sup>. M & K values are positive, M shows small magnitudes than K.

**IV. CONCLUSIONS**

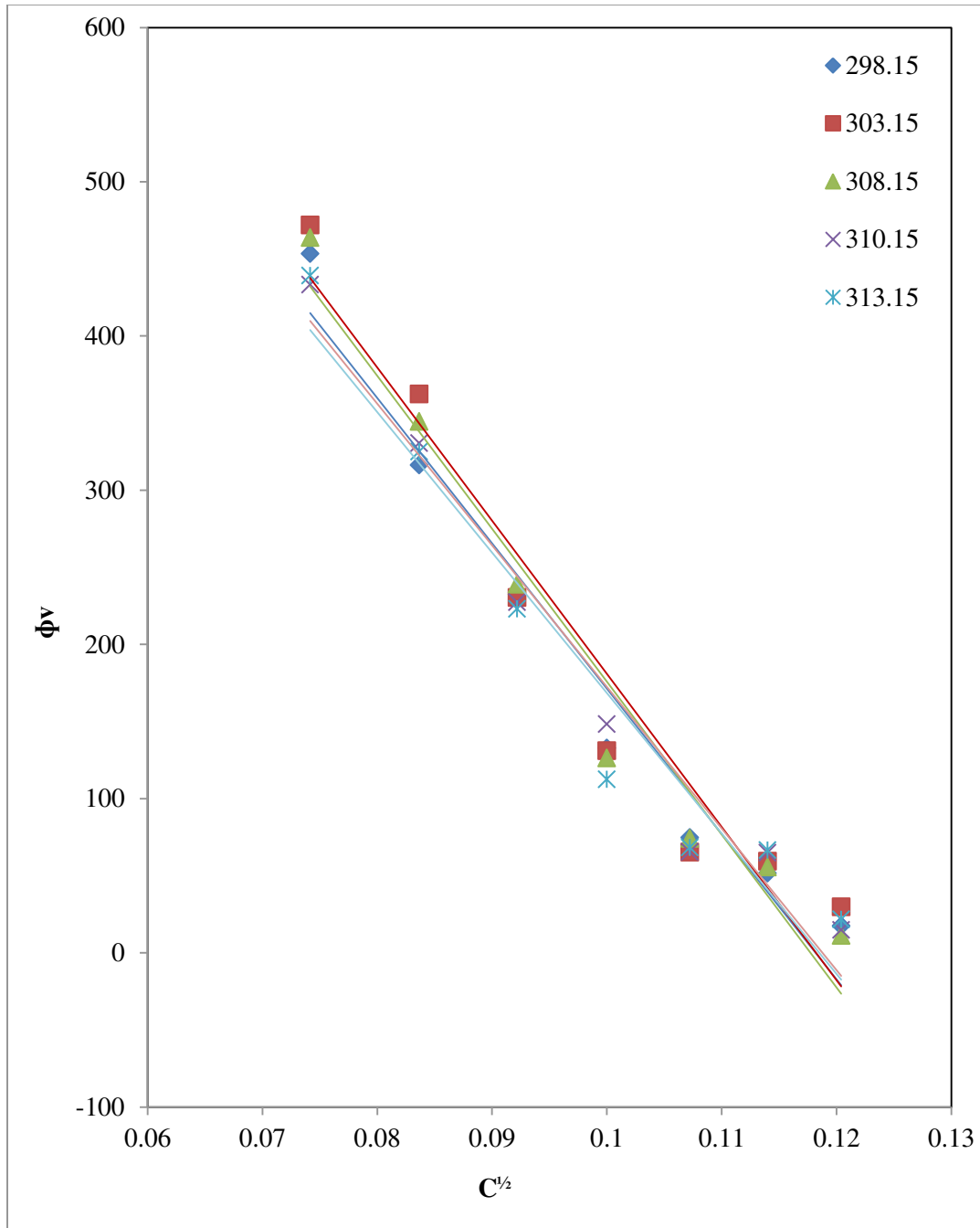
Study of flow and interaction parameters of aspirin in presence of additives such as NaCl, KCl, Dextrose, Sodium lactate etc. at different temperatures suggests that

1. The values of  $\phi_v^0$  at all temperatures are negative for set 1 and 4, suggest the weak ion-solvent interactions in aqueous solution, that may have the implication for the permeation of those molecules through biological membranes.
2. The  $S_v$  values are positive for set 1 and 4 suggesting strong ion-ion interactions.
3. The positive values of Jones-Dole coefficient B at all temperatures indicates water structuring behavior.
4. In aqueous solution the values of partial molar volume are positive/ negative and decrease with the extent of hydrogen bonding. Lower the partial molar volume value, stronger is the hydrogen bond. Positive values of B suggesting strongly hydrated solute indicating structure promoting tendency i.e. kosmotropes (structure makers).

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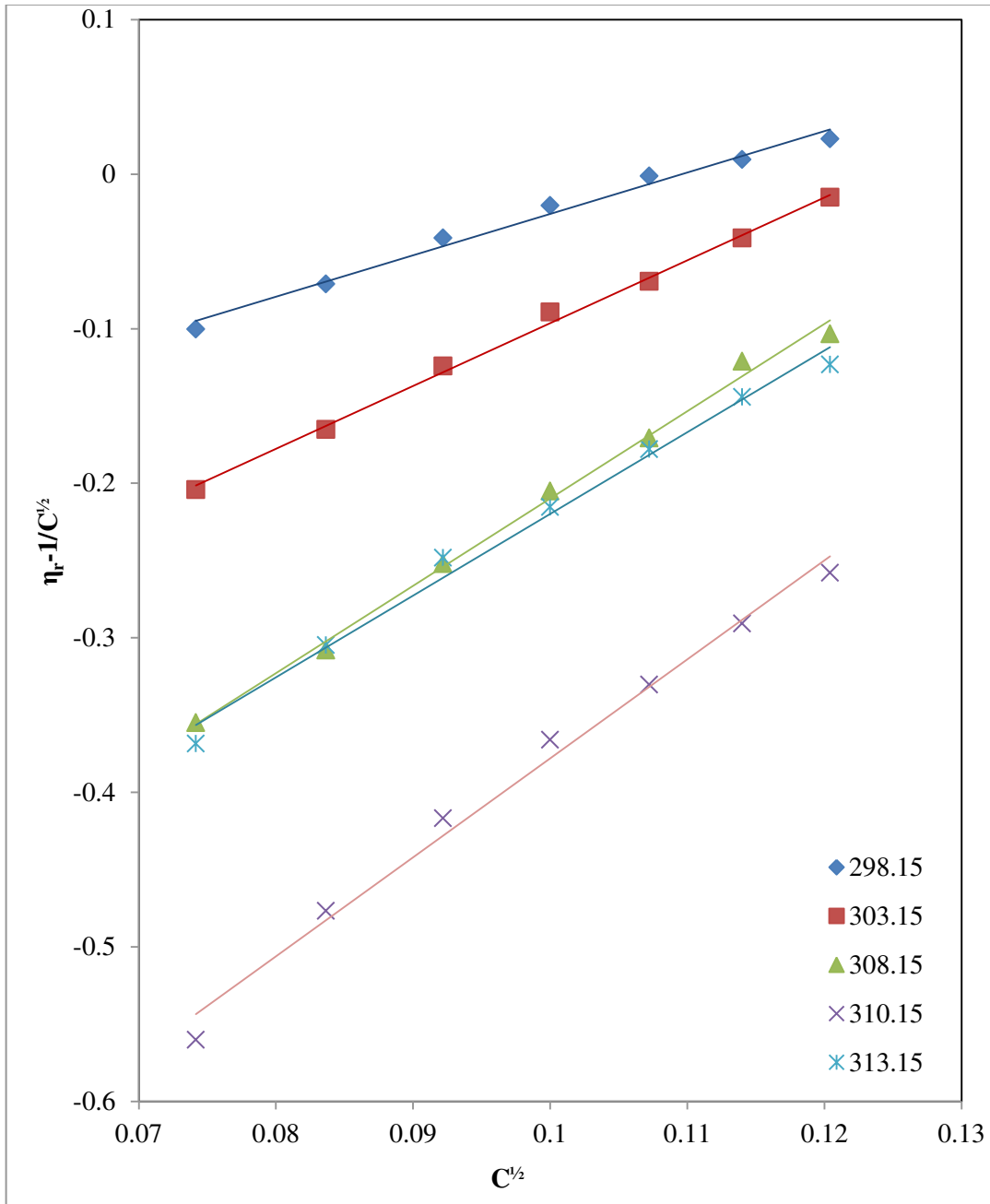


**Figure 1:** Plot of  $\phi v$  versus  $C^{1/2}$  for aspirin solution in RL at different temperatures.

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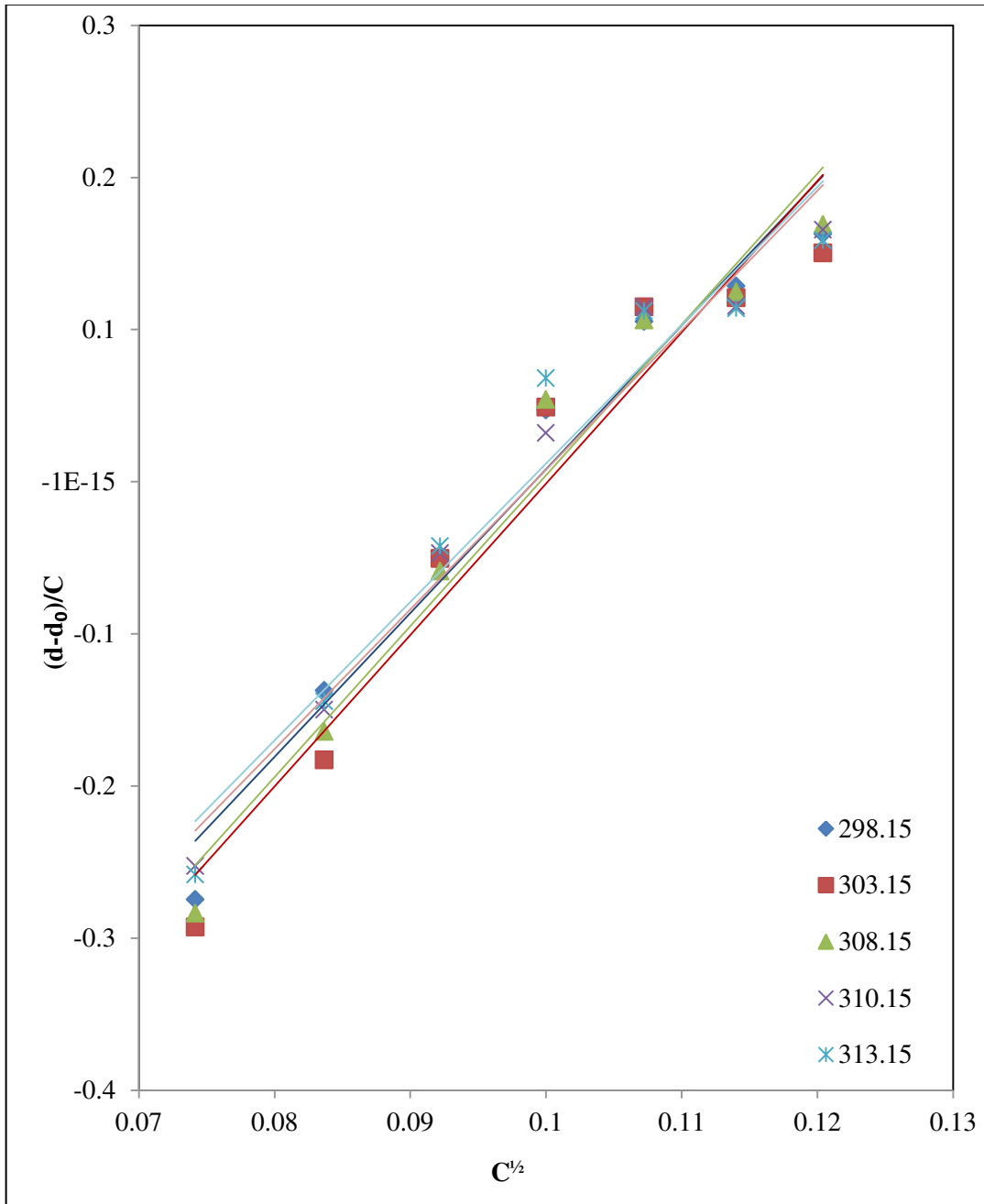
**Figure 2:** plot of  $(\eta_r - 1)/C^{1/2}$  versus  $C^{1/2}$  of aspirin solutions in RL at different temperatures.



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**Figure 3:** plot of  $(d-d_0)/C$  versus  $C^{1/2}$  for aspirin solutions in RL at different temperatures.

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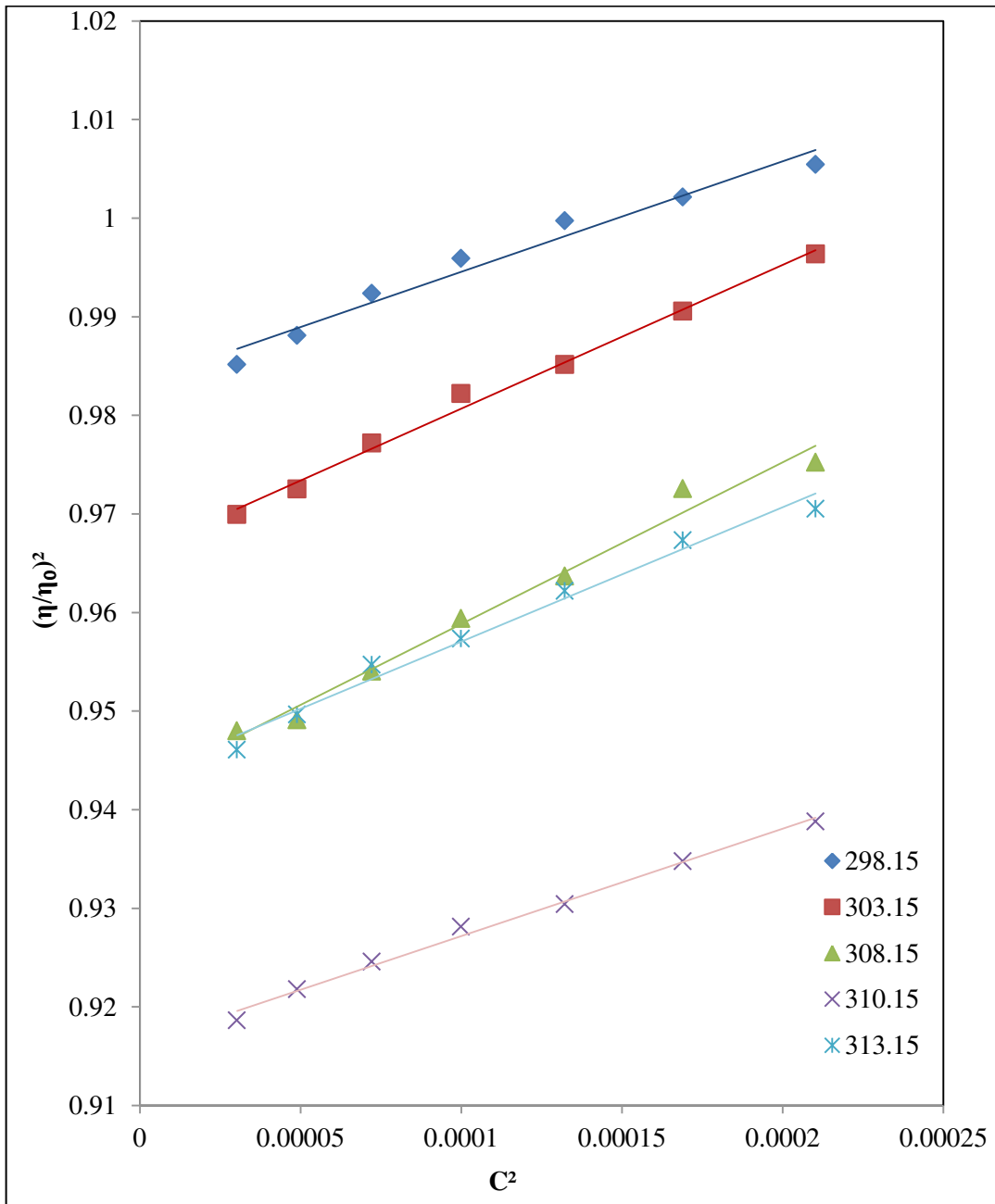


Figure 4: plot of  $(\eta/\eta_0)^2$  versus  $C^2$  for aspirin solutions in RL at different temperatures.

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