Viscosities of L-Phenylalanine, L-Leucine, L-Glutamic Acid, or L-Proline + 2.0 mol·dm^{-3} Aqueous NaCl or 2.0 mol·dm^{-3} Aqueous $NaNO_3$ Solutions at T = (298.15 to 328.15) K

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Viscosity coefficients of L-phenylalanine, L-leucine, L-glutamic acid, or L-proline in 2.0 mol·dm^{-3} aqueous NaCl or 2.0 mol·dm^{-3} aqueous $NaNO_3$ systems have been determined as a function of molal concentrations of amino acids at temperatures of (298.15, 303.15, 308.15, 313.15, 318.15, 323.15, and 328.15) K. Viscosity coefficients vary linearly with an increase in molal concentration of the amino acids. The trends of variation of viscosity values with an increase in molal concentration of L-phenylalanine, L-leucine, L-glutamic acid, and L-proline in 2.0 mol·dm^{-3} aqueous NaCl and 2.0 mol·dm^{-3} aqueous $NaNO_3$ solutions and with an increase in temperature have been ascribed to the solute−solvent interactions operative in the solutions. The computed Jones−Dole *B*-coefficients have been also interpreted in terms of solute−solvent interactions.

Introduction

The study of protein−salt interactions is useful to understand several biological processes occurring in physiological media such as blood membranes and cellular fluids in living organisms. Since proteins are large complex molecules, the direct estimation of protein−salt interactions is difficult. The study of amino acid/peptide−salt interactions may reduce the degree of complexity involved in studying the protein−salt interactions. A number of workers have studied the viscometric properties of amino acids and peptides in aqueous salt solutions.[1−5] Structure-making and -breaking effects of an electrolyte on solvent can be determined by various parameters resolved from viscosity studies. The Jones−Dole viscosity *B*-coefficients depend on both the size of the solute and the solvent−solute interactions. A number of researchers have determined the viscosity *B*-coefficient values of amino acids and peptides in aqueous media[6−12] and in aqueous electrolytes.[4,12−16]

Viscosities of L-alanine, L-proline, L-valine, and L-leucine in 2.0 mol·dm^{-3} aqueous KCl and 2.0 mol·dm^{-3} aqueous KNO_3,[17] and L-histidine, L-glutamic acid, L-tryptophan, and glycylglycine in 2.0 mol·dm^{-3} aqueous KCl and 2.0 mol·dm^{-3} aqueous KNO_3 solutions,[18] and L-leucine, L-asparagine, and glycylglycine in 1.5 mol·dm^{-3} aqueous NaCl, $NaNO_3$, and KNO_3 solutions[19] and in 1.5 mol·dm^{-3} KCl, 1.0 mol·dm^{-3} KNO_3, and 0.5 mol·dm^{-3} K_2SO_4 have been measured for several molal concentrations of amino acids and peptides at different temperatures of (303.15, 308.15, 313.15, 318.15, and 323.15)[20] K in our laboratory. The results have been discussed in terms of solute−solvent and solute−solute interactions.

This study focuses on the measurements of viscosity coefficients of L-phenylalanine, L-leucine, L-glutamic acid, or L-proline + 2.0 mol·dm^{-3} aqueous NaCl or 2.0 mol·dm^{-3} aqueous $NaNO_3$ solutions as functions of amino acid concentration and temperatures of (298.15, 303.15, 308.15, 313.15, 318.15, 323.15, and 328.15) K. The Jones−Dole viscosity *B*-coefficients have been also computed from the viscosity data. The obtained viscosity coefficients and viscosity *B*-coefficients results have been discussed in terms of solute−solvent interactions operative in the systems.

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Materials and Methods

The amino acids L-phenylalanine, L-leucine, L-glutamic acid, and L-proline and the salts sodium chloride and sodium nitrate with minimum mass fraction purities of 0.99 used in this study were purchased from SRL (India) and E. Merck (India), respectively. The amino acids were recrystallized twice in ethanol + water, dried at T = 383.15 K, and kept in a vacuum desiccator over P_2O_5 for at least 72 h before use. The salts were recrystallized twice in triply distilled water, dried at T = 423.15 K for at least 3 h, and then kept over P_2O_5 in a vacuum desiccator at room temperature for a minimum of 48 h prior to their use. Stock solutions of 2.0 mol·dm^{-3} aqueous NaCl and 2.0 mol·dm^{-3} aqueous $NaNO_3$ were prepared in triply distilled water and used as solvents for the preparation of amino acid solutions. The specific conductivity of triply distilled water used was less than $1.0 \cdot 10^{-6}$ S·cm^{-1}. All of the solutions were stored in special airtight bottles to avoid the exposure of solutions to air and evaporation.

The viscosity measurements were carried out using a suspended Ubbelohde type viscometer. A thoroughly cleaned and perfectly dried viscometer filled with the test solution was placed vertically in the glass-walled paraffin bath thermostat. The thermostat used for measurements of viscosity values was maintained at a desired temperature (± 0.01 K) for about 30 min prior to recording of readings at each temperature of study. After attainment of thermal equilibrium, efflux times of flow were recorded with an electronic watch with the resolution of 0.01 s. The average of at least four readings reproducible within 0.1 s was used as final efflux time. The viscosity values of water used as a reference liquid at different temperatures were taken from the literature.[21] The densities required for the calculation of viscosity values of the solutions were taken from our earlier reported studies.[22]

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Table 1. Viscosities η as Functions of Molality m and Temperature T

m	$\eta/10^{-4}$ (Pa·s)						
mol·kg^{-1}	T/K = 298.15	T/K = 303.15	T/K = 308.15	T/K = 313.15	T/K = 318.15	T/K = 323.15	T/K = 328.15
			L-Phenylalanine in 2.0 mol·dm^{-3} Aqueous NaCl Solution				
0.0000	10.850	9.761	8.816	8.070	7.426	6.760	6.257
0.0187	11.033	9.942	8.977	8.177	7.508	6.841	6.355
0.0374	11.236	10.127	9.082	8.264	7.584	6.915	6.394
0.0562	11.403	10.224	9.187	8.360	7.663	7.009	6.486
0.0751	11.648	10.484	9.262	8.478	7.737	7.123	6.566
0.0941	12.034	10.816	9.356	8.571	7.828	7.187	6.594
			L-Phenylalanine in 2.0 mol·dm^{-3} Aqueous $NaNO_3$ Solution				
0.0000	10.364	9.367	8.519	7.777	7.167	6.559	6.043
0.0180	10.561	9.523	8.652	7.897	7.257	6.653	6.195
0.0362	10.631	9.601	8.702	7.992	7.316	6.694	6.271
0.0543	10.772	9.741	8.841	8.084	7.379	6.763	6.337
0.0726	10.945	9.867	8.956	8.207	7.466	6.849	6.406
0.0909	11.056	9.949	9.046	8.269	7.554	6.918	6.509
0.1094	11.305	10.043	9.140	8.434	7.638	6.998	6.562
0.1280	11.512	10.268	9.255	8.477	7.775	7.075	6.630
0.1465	11.626	10.426	9.438	8.586	7.887	7.189	6.768
			L-Leucine in 2.0 mol·dm^{-3} Aqueous NaCl Solution				
0.0000	10.850	9.761	8.816	8.070	7.426	6.760	6.257
0.0186	10.933	9.810	8.908	8.162	7.423	6.782	6.320
0.0374	11.112	9.882	8.997	8.233	7.529	6.895	6.375
0.0562	11.152	10.042	9.084	8.301	7.597	6.988	6.443
0.0750	11.377	10.213	9.158	8.392	7.679	7.078	6.499
0.0940	11.621	10.473	9.268	8.493	7.753	7.176	6.553
0.1130	11.800	10.643	9.447	8.575	7.825	7.270	6.594
			L-Leucine in 2.0 mol·dm^{-3} Aqueous $NaNO_3$ Solution				
0.0000	10.364	9.367	8.519	7.777	7.167	6.559	6.043
0.0182	10.509	9.554	8.592	7.879	7.208	6.624	6.132
0.0364	10.623	9.683	8.711	7.969	7.332	6.703	6.202
0.0547	10.716	9.774	8.804	8.049	7.411	6.763	6.280
0.0731	10.805	9.854	8.843	8.127	7.481	6.797	6.323
0.0915	10.906	9.971	8.951	8.182	7.545	6.852	6.378
0.1101	10.990	10.056	9.062	8.283	7.619	6.898	6.422
0.1287	11.078	10.125	9.166	8.369	7.704	6.955	6.479
0.1474	11.179	10.234	9.283	8.458	7.802	7.033	6.531
			L-Glutamic Acid in 2.0 mol·dm^{-3} Aqueous NaCl Solution				
0.0000	10.850	9.761	8.816	8.070	7.426	6.760	6.257
0.0093	10.862	9.755	8.852	8.114	7.410	6.762	6.293
0.0186	10.939	9.815	8.903	8.156	7.459	6.800	6.322
0.0280	11.008	9.864	8.959	8.202	7.497	6.837	6.359
0.0373	11.038	9.909	8.978	8.239	7.542	6.874	6.388
0.0467	11.077	9.966	9.026	8.286	7.589	6.920	6.424
0.0561	11.137	10.026	9.076	8.335	7.629	6.967	6.454
			L-Glutamic Acid in 2.0 mol·dm^{-3} Aqueous $NaNO_3$ Solution				
0.0000	10.364	9.367	8.519	7.777	7.167	6.559	6.043
0.0091	10.419	9.409	8.616	7.838	7.209	6.580	6.078
0.0182	10.455	9.469	8.684	7.881	7.255	6.620	6.122
0.0272	10.524	9.562	8.738	7.932	7.295	6.683	6.157
0.0364	10.570	9.609	8.823	7.983	7.390	6.743	6.218
0.0455	10.652	9.682	8.904	8.046	7.462	6.796	6.261
0.0546	10.724	9.780	8.973	8.103	7.543	6.846	6.317
			L-Proline in 2.0 mol·dm^{-3} Aqueous NaCl Solution				
0.0000	10.850	9.761	8.816	8.070	7.426	6.760	6.257
0.1893	11.417	10.259	9.324	8.512	7.822	7.106	6.591
0.3854	12.223	10.961	9.878	9.043	8.271	7.552	6.981
0.5881	13.074	11.713	10.586	9.692	8.792	7.984	7.529
0.7988	14.125	12.651	11.472	10.306	9.463	8.527	7.869
1.0166	14.865	13.683	11.976	10.784	9.830	9.083	8.383
1.2426	16.320	14.548	12.996	11.715	10.774	9.643	8.878
1.4782	17.291	15.516	13.810	12.396	11.416	10.168	9.418
1.7234	18.447	16.474	14.731	13.182	12.000	10.771	9.856
			L-Proline in 2.0 mol·dm^{-3} Aqueous $NaNO_3$ Solution				
0.0000	10.364	9.367	8.519	7.777	7.167	6.559	6.043
0.1841	11.095	9.951	9.134	8.281	7.694	6.986	6.448
0.3748	11.844	10.652	9.619	8.743	8.065	7.324	6.785
0.5725	12.625	11.327	10.216	9.272	8.502	7.744	7.165
0.7773	13.450	11.999	10.835	9.866	9.001	8.194	7.548
0.9898	14.344	12.802	11.530	10.464	9.595	8.705	8.019
1.2110	15.431	13.705	12.241	11.133	10.152	9.206	8.498
1.4404	16.531	14.711	13.199	11.891	10.933	9.791	9.041
1.6789	17.797	15.818	14.139	12.749	11.575	10.472	9.608

The uncertainties in viscosity measurements and molal concentration values of solutions have been found to be within 0.002 mPa·s and 0.0002 mol kg^{-1}, respectively.

Results and Discussion

The measured viscosity values of L-phenylalanine/L-leucine/L-glutamic acid/L-proline + 2.0 mol·dm^{-3} aqueous NaCl/2.0 mol·dm^{-3} aqueous $NaNO_3$ systems as functions of amino acid concentration and temperatures of (298.15, 303.15, 308.15, 313.15, 318.15, 323.15, and 328.15) K are listed in Table 1. The viscosity value of 2.0 mol·dm^{-3} aqueous NaCl solution ($10.850 \cdot 10^{-4}$ Pa·s) is larger than the value of 2.0 mol·dm^{-3} aqueous $NaNO_3$ solution ($10.364 \cdot 10^{-4}$ Pa·s) at $T = 298.15$ K. Similarly, the viscosity values of the investigated systems, amino acid + 2.0 mol·dm^{-3} aqueous NaCl solutions, are larger than the corresponding values of the amino acid + 2.0 mol·dm^{-3} aqueous $NaNO_3$ solutions. The viscosity values of the systems studied (amino acid + 2.0 mol·dm^{-3} aqueous NaCl/2.0 mol·dm^{-3} aqueous $NaNO_3$) show an increasing trend with an increasing concentration of amino acid in solution. This trend of variation of η values may be attributed to an increase in the ion−zwitterion interactions, $Na^+/Cl^-/NO_3^-/NH_3^+/COO^-$, and with zwitterion−zwitterion interactions an increase in the number of amino acid molecules/zwitterions in solutions which in turn may cause more frictional resistance to the flow of solutions. The viscosity values of all of the systems under investigation exhibit a decreasing trend of variation with an increase in temperature. An increase in temperature may increase the kinetic energy of molecules, which in turn may decrease the ion−ion (Na^+, Cl^-, Na^+, NO^{3-}), zwitterion−ion (COO^-, Na^+, NH_3^+ $-Cl^-/-NO_3^-$), and zwitterion−zwitterion interactions.

The viscosity data have been fitted to the Jones−Dole[23] equation of the form

$$\eta_r = \eta/\eta_o = 1 + Am^{1/2} + Bm \qquad (1)$$

where η_r is the relative viscosity of the solution, m is the molal concentration of solution, and η and η_o are the viscosities of solution and solvent, respectively. A, the Falkenhagen coefficient, represents the solute−solute interactions associated with the size and shape of solute, and B is a measure of structural modifications induced by the solute−solvent interactions.[23−25] The plots of $(\eta/\eta_o - 1)/m^{1/2}$ versus $m^{1/2}$ have been found to be linear at all temperatures of study in accordance with the Jones−Dole equation. The A- and B-coefficient values are the intercept and slope of the straight line, respectively, obtained from linear regression. The B-coefficient values have been given in the Table 2.

The positive B-coefficient values of L-phenylalanine, L-leucine, L-glutamic acid, and L-proline in 2.0 mol·dm^{-3} aqueous NaCl and 2.0 mol·dm^{-3} aqueous $NaNO_3$ solutions indicate a strong alignment of zwitterions with ions/water dipoles. The viscosity B-coefficients of L-phenylalanine, L-leucine, L-glutamic acid, and L-proline in 2.0 mol·dm^{-3} aqueous NaCl and 2.0 mol·dm^{-3} aqueous $NaNO_3$ solutions are larger than the corresponding values in water.[26−29] The observed larger B-coefficient values of L-phenylalanine, L-leucine, L-glutamic acid, and L-proline in 2.0 mol·dm^{-3} aqueous NaCl and 2.0 mol·dm^{-3} aqueous $NaNO_3$ solutions than those in aqueous medium may be attributed to Na^+-Cl^-, $Na^+-NO_3^-$, and COO^--Na^+, and $-NH_3^+-Cl^-/NO_3^-$ interactions in solution.

Conclusions

The viscosity coefficient values of amino acid + 2.0 mol·dm^{-3} aqueous NaCl/2.0 mol·dm^{-3} aqueous $NaNO_3$ systems have been found to be increasing with an increase in molal concentration of amino acids in solutions. The viscosity B-coefficients have been found to be positive for all of the systems. The viscosity coefficient and viscosity B-coefficient results have been interpreted in terms of the zwitterion−ion and zwitterion−water dipole interactions with a successive increase in the number of amino acid molecules or zwitterions in solution.

Table 2. Viscosity *B*-Coefficients of Amino Acids in 2.0 mol·dm^{-3} Aqueous NaCl and 2.0 mol·dm^{-3} Aqueous $NaNO_3$ Solutions at Different Temperatures

amino acid	salt	T / K	B-coefficient / $dm^3 \cdot mol^{-1}$
L-phenylalanine	NaCl	298.15	1.2390
		303.15	1.1465
		308.15	0.3785
		313.15	0.6228
		318.15	0.5476
		323.15	0.7325
		328.15	0.4358
	$NaNO_3$	298.15	0.9064
		303.15	0.7838
		308.15	0.7753
		313.15	0.7549
		318.15	0.7767
		323.15	0.6994
		328.15	0.5704
L-leucine	NaCl	298.15	0.9239
		303.15	1.1236
		308.15	0.6285
		313.15	0.5361
		318.15	0.6308
		323.15	0.8857
		328.15	0.4056
	$NaNO_3$	298.15	0.3710
		303.15	0.3273
		308.15	0.6301
		313.15	0.4971
		318.15	0.6668
		323.15	0.3991
		328.15	0.3878
L-glutamic acid	NaCl	298.15	0.5186
		303.15	0.6047
		308.15	0.5458
		313.15	0.5723
		318.15	0.6217
		323.15	0.6691
		328.15	0.4209
	$NaNO_3$	298.15	0.6284
		303.15	0.9630
		308.15	0.7881
		313.15	0.7060
		318.15	1.2075
		323.15	1.1579
		328.15	0.9455
L-proline	NaCl	298.15	0.4755
		303.15	0.4755
		308.15	0.4382
		313.15	0.4059
		318.15	0.4094
		323.15	0.3851
		328.15	0.3646
	$NaNO_3$	298.15	0.4442
		303.15	0.4342
		308.15	0.3950
		313.15	0.3944
		318.15	0.3620
		323.15	0.3590
		328.15	0.3483

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