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Viscosity B-coefficient for Viscous Flow of Some Amino Acids During the Passage of Alternating Current at Different Voltages

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ABSTRACT

Densities and viscosities of Glycine, DL-alanine, L-valine and L-serine in aqueous solutions have been measured experimentally under the influence of alternating currents at different voltages. The results obtained have been used to calculated the apparent molar volume, $V_{2\varphi}$, relative viscosity, η_{rel}, free energy of activation of viscous flow of solvent, $\Delta\mu_1^{o\#}$ and solute, $\Delta\mu_2^{o\#}$. The partial molar volume, $V^{\circ}_{2\varphi}$, at infinite dilution and Jones- Dole coefficient, B, were calculated by least-square method. These results have been discussed in terms of solute-solute, solute solvent interactions and structure making/breaking ability of solutes in the given solution. The trends of variations of viscosity values with voltages have been ascribed to the solute-solvent interactions operative in the solutions. The periodic changes in the magnitudes and direction of electric field lead to decrease the solute-solvent interactions. This effect seems to be responsible for a decrease in viscosity.

Key words: Viscosity, alternating current, glycine, alanine, valine, serine

INTRODUCTION

Amino acids are the fundamental structural units of proteins. Thermodynamic properties of these model compound in aqueous electrolytes media provide information about solute-solvent and solute-solute interactions that can be of great help in understanding the effects of electrolytes on biomolecules (Von-Hippel and Schleich, 1969; Romero *et al.*, 1999; Tamura and Gekko, 1995; Bai and Yan, 2003; Badaryani and Kumar, 2002).

Very recently, systematic efforts have been made to investigate the volumetric properties of amino acids in concentrated electrolytic solutions (Bardayani and Kumar, 2003). It was found that some electrolytes increase the apparent molar volume of amino acids. This increase was attributed the interactions of the ions of the electrolytes and zwitterionic head groups of amino acids.

Viscosity and its derived parameters provide valuable information regarding the shape and size of these molecules. Such measurements on the dipolar ions, particularly of the amino acids, have been carried out by a number of workers (Owaga *et al.*, 1984; Wadi and Goyal, 1992). However, the effect of passage of alternating current through aqueous solutions of amino acids at different voltages has not been studied.

In the present study, we report densities, ρ and viscosities, η of Glycine, DL-alanine, L-valine and L-serine from these measurements, a number of thermodynamic parameters namely, apparent molar volume, $V_2\varphi$, relative viscosity, η_{rel}, partial molar volume, $V^{\circ}_2\varphi$, at infinite dilution and

Jones-Dole coefficient, B, Free energy of activation of viscous flow of solvent, $\Delta\mu_1^{o\#}$ and solute, $\Delta\mu_2^{o\#}$, respectively, have been calculated.

MATERIALS AND METHODS

The amino acids glycine, DL-alanine, L-valine and L-serine used in this study were of high purity (mass fraction = 0.99) and were procured from Merk and Sigma, respectively and were used without further purification. Triply distilled water with a specific conductance less than $6\times10^{-8}\ \Omega^{-1}\ \mathrm{cm}^{-1}$ was used to prepare amino acids solutions. These solutions were kept in air tight bottles to avoid contamination and evaporation. The concentrations of these solutions were in the range of 0.01-0.1 mol L^{-1}. Mettler digital balance of readability ±0.01 mg was used for mass measurements. Solution densities were measured to $\pm3\times10^{-6}\ \mathrm{g\ cm}^{-3}$.

The densities (ρ) of solutions were measured using an Ostwald-sprengel type pycnometer of total volume of $8\times10^{-6}\ \mathrm{m}^3$ and capillary internal diameter of about 0.1 cm. Reproducibility in the density measurements was better than $\pm3\times10^{-2}\ \mathrm{kg\ m}^{-3}$. A thermostatically controlled well-stirred water bath controlled to ±0.01°C was used, viscosity measurements were carried out using a suspended level Ubbelohde viscometer with a flow time of approximately 300 sec for distilled water at 298.15 K. The measurements were made in the viscometer fitted with two platinum electrodes. The tip of the platinum wire in the capillary tube was located below the second mark for timing the flow of liquid in the viscometer. The power source as a generator and amplifier, connected to the apparatus through a step-up transformer and a resistance box. The current and voltage wave-forms were displaced on an S1-15 oscilloscope with a U2 pre-amplifier. The viscometer was placed within a thermostat. The average of at least three readings reproducible within ±0.1 sec was used as the final efflux time. The estimated viscosities have uncertainity within ±0.003 mPa.s. The viscosity values of water at 298.15 K was taken from the literature for calibration purpose (Kestin *et al*., 1978).

RESULTS AND DISCUSSION

The measured viscosity values for the amino acid solutions as functions of amino acid concentration, temperature and applied voltages are listed in Table 1-4. Direct comparisons of viscosity results are not possible as no report of viscosity values in literature is available under similar conditions for the systems studied. Viscosities of amino acids solutions (ranging from 0.01 to 0.1 M) at constant temperature and different voltages have been found to be increasing with an increase in concentration of amino acid in solutions. This may be attributed to an increase in solute-solvent interactions with successive increase in the number of amino acid molecules/ zwitterions in solutions, which may, in turn cause more frictional resistance to the flow of solutions. The viscosity values of amino acid solutions at a given concentration decrease with an increase in both applied voltages and current. The periodic changes in the magnitudes and direction of electric field may have caused the increase in kinetic energy of molecules in the solution, which in turn, decrease the solute-solvent interactions. This may cause the rapid movement of molecules into the empty sites. Such a decrease in interactions seems to be responsible for the decrease in viscosity. The passage of direct current does not produce this effect.

The densities of amino acid solutions as a function of concentrations are listed in Appendix A. The values of increase with increase ñ in concentration of amino acids in all the systems under investigation. This may be attributed to strong intermolecular forces, which in turn, leads to a decrease in intermolecular free path.

Table 1: Viscosity of some amino acids under the influence of 25 volt at different concentrations in presence of alternating current at 298.15 K and time of application current (60 sec)

C mol^{-1}	Glycine	DL-alanine	L-valine	L-serine
0.01	0.7834	0.7866	0.7853	0.7930
0.03	0.7875	0.7952	0.7942	0.8056
0.05	0.7933	0.8048	0.8289	0.8314
0.07	0.8028	0.8154	0.8388	0.8433
0.09	0.8104	0.8231	0.8461	0.8550
0.10	0.8131	0.8253	0.8481	0.8570

Table 2: Viscosity of some amino acids under the effect of 50 volt at different concentrations in presence of alternating current at 298.15 K and time of application current (60 sec)

C mol^{-1}	Glycine	DL-alanine	L-valine	L-serine
0.01	0.7589	0.7606	0.7648	0.7597
0.03	0.7622	0.7648	0.7732	0.7647
0.05	0.7674	0.7725	0.7861	0.7734
0.07	0.7741	0.7818	0.7953	0.7817
0.09	0.7848	0.7903	0.8035	0.7910
0.10	0.7844	0.7988	0.8275	0.7977

Table 3: Viscosity of some amino acids under the effect of 100 volt at different concentrations in presence of alternating current at 298.15 K and time of application current (60 sec)

C mol^{-1}	Glycine	DL-alanine	L-valine	L-serine
0.01	0.7576	0.7606	0.7665	0.7589
0.03	0.7616	0.7656	0.7715	0.7636
0.05	0.7655	0.7735	0.7833	0.7724
0.07	0.7851	0.7933	0.8244	0.7861
0.09	0.7843	0.8002	0.8456	0.7987
0.10	0.7860	0.8012	0.8470	0.7995

Table 4: Viscosity of some amino acids under the effect of 150 volt at different concentrations in the presence of alternating current at 298.15 K and time of application current (60 sec)

C mol^{-1}	Glycine	DL-alanine	L-valine	L-serine
0.01	0.7098	0.7127	0.7157	0.7117
0.03	0.7123	0.7166	0.7221	0.7152
0.05	0.7166	0.7245	0.7362	0.7235
0.07	0.7254	0.7333	0.7710	0.7431
0.09	0.7312	0.7421	0.7950	0.7568
0.10	0.7332	0.7489	0.7970	0.7575

The apparent molar volumes $V_{2,\varphi}$ were calculated from ρ data using the following equation:

$$V_{2,\varphi} = \frac{M}{\rho + 10^3 (\rho_o - \rho) m \rho \rho_o} \tag{1}$$

where, M is the relative molar mass of the solute (kg mol^{-1}), m is the molality (mol kg^{-1}) of the solution, ρ_o and ρ the densities (10^{-3} kg m^{-3}) of solvent and solution, respectively. The calculated values of $V_{2,\varphi}$ are included in Appendix B. In all cases, the dependence of the apparent molar volume on concentration is linear in the concentration range considered. The data were fitted by least squares to the relation:

$$V_{2,\varphi} = V^{\circ}_{2,\varphi} + S_v m \tag{2}$$

where, $V^{\circ}_{2,\varphi}$ is limiting value of the apparent molar volume (equal to the partial molar volume at infinite dilution), S_v the experimental slope. The values of $V^{\circ}_{2,\varphi}$ and S_v obtained for amino acid solutions at different concentrations are reported in Appendix C together with the standard errors. It is evident that the values of the slope S_v for all amino acids at different concentrations are positive indicating that solute-solvent interactions operative in the solutions increase with increasing concentrations.

It is well established fact that (Frank and Wen, 1957) when a molecule with non polar and polar parts dissolves in water at room temperature, the water adjacent to the non polar part becomes more ice-likes structure than normal and this effect increases with size of the non-polar part of molecule. Thus $V^{\circ}_{2,\varphi}$ values reflect the net structural effects of the charged groups and the hydrophobic CH_2 groups in the solvent. The values of $V^{\circ}_{2,\varphi}$ for all the amino acids studied in water vary linearly (average correlation coefficients R = 0.9997) with the number of carbon atoms in their alkyl chains at given temperatures.

The Viscosities (η) of the solutions were determined from the following equation:

$$\eta = \frac{(kt - 1/t)}{\rho} \tag{3}$$

where, t is the efflux time, ρ is the density of the solution and k and l are constants for the viscometer used the studied solutions at different voltages under the effect of AC which are collected in Table 1-4.

The relative viscosities $\eta_r(\eta/\eta_o)$ where η and η_o are the viscosities of solution and solvent respectively) of the studied solutions. The B coefficients were calculated by using the following Jones-Dole equation by least squares method:

$$\eta_r = \eta/\eta_o = 1 + B\,C \tag{4}$$

where, C is the molarity (calculated from molality) of the solution. The B- coefficients thus obtained are given in Table 5.

The values of viscosity B-coefficients for all amino acids studied in water at different temperatures and voltages are positive indicating that the solute-solvent interactions are strong.

Thermodynamic activation parameters for viscous flow of amino acid in water evaluated using the relation proposed by (Feakins *et al.*, 1974) as follows:

$$B = \frac{(V^{\circ}_{1,\varphi} - V^{\circ}_{2,\varphi})}{1000 + (V^{\circ}_{1,\varphi}/1000)(\mu^{\circ}{}_2^{\#} - \mu^{\circ}{}_1^{\#})/RT} \tag{5}$$

where, $V^{\circ}_{1,\varphi}$ and $V^{\circ}_{2,\varphi}$ are the partial molar volumes of solvent and solute at infinite dilution, respectively, $\Delta\mu_1^{\circ\,\#}$ the free energy of activation per mole of the solvent and $\Delta\mu_2^{\circ\#}$ is the free energy of activation per mole of the solute. $\Delta\mu^{\circ}{}_1^{\#}$ and $\Delta\mu^{\circ}{}_2^{\#}$ were calculated from the following equations:

Table 5: The values of B's coefficient of some amino acids in different voltages under the effect of AC at 298.15 K and time of application current (60 sec)

	B ($cm^3\ mol^{-1}$)			
Amino acids	25 volt	50 volt	100 volt	150 volt
Glycine	0.4307	0.415	0.746	0.3986
DL-alanine	0.4307	0.561	0.675	0.5772
L-valine	0.9588	0.827	1.390	1.4750
L-serine	0.5600	0.565	0.665	0.8429

Table 6: Activation free energies $\Delta\mu^{o\#}_2$ for viscous flow of amino acids in water in different voltages and 50 Hz at 298.15 K and time of application current (60 sec)

Amino acids	25 volt	50 volt	100 volt	150 volt
Glycine	29.97±0.029	30.01±0.047	32.29±0.059	39.48±0.042
DL-alanine	57.15±0.037	57.25±0.054	57.87±0.020	57.86±0.062
L-valine	94.91±0.048	95.62±0.051	97.21±0.021	98.20±0.033
L-serine	52.48±0.038	54.82±0.039	54.38±0.041	55.26± 0.045

$$\Delta\mu^{\circ}{}_1^{\#} = RT\ln(\eta_o V^{\circ}{}_{1,\varphi}/hN_A) \tag{6}$$

$$\Delta\mu^{\circ}{}_2^{\#} = \frac{\Delta\mu^{\circ}{}_1^{\#} + RT}{V^{\circ}{}_{1,\varphi}[1000B - (V^{\circ}{}_{1,\varphi} - V^{\circ}{}_{2,\varphi})} \tag{7}$$

where, N_A is the Avogadro's number, R and h the gas and Planck's constant and T is the absolute temperature. The calculated $\Delta\mu_2^{o\#}$ values at different voltages are given in Table 7.

It is observed that B-coefficients of the studied some amino acids vary linearly with the number of carbon atoms in their alkyl chains that given temperatures and voltages. This linear relation can be represented by:

$$B = B(NH_3^+, COO^-) + n_c B(CH_2) \tag{8}$$

where, n_c is the number of C atoms in the alkyl chain of amino acid. The regression of B data using Eq. 8 gives B (NH_3^+, COO^-) and B (CH_2) values as respective contributions of (NH_3^+, COO^-) and the CH_2 groups are listed in Table 7. These results clearly reveal the structural features of these solutions. The positive dB/dV values for (NH_3^+, COO^-) groups confirm that these charged groups are structure-breakers while the hydrophobic CH_2 groups with negative dB/dV values are structure makers (Belibagli and Ayranci, 1990).

It is clear from Table 6 that values of $\Delta\mu^{\circ}{}_2^{\#}$ are positive indicating a stronger solute-solvent interaction. In other words, the formation of the transition state is less favoured. This means that the formation of the transition state is accompanied by the rupture and distortion of the intermolecular forces in solvent structure. It may be pointed put also that $\Delta\mu^{\circ}{}_2^{\#}$ values at constant temperature or voltage increase in the following order:

Glycine<L-serine<DL-alanine<L-valine

Table 7: Contributions of the zwitterionic (NH_3^+, COO^-) and CH_2 groups of amino acids to the viscosity B-coefficient

Group	B ($dm^3\ mol^{-1}$)	B ($dm^3\ mol^{-1}$)	B ($dm^3\ mol^{-1}$)	B ($dm^3\ mol^{-1}$)
Voltages	25	50	100	150
NH_3^+, COO^-	0.024±0.027	0.052±0.029	0.074±0.026	0.089±0.023
CH_2	0.120±0.008	0.103±0.009	0.088±0.007	0.077±0.007

The increase in $\Delta\mu^\circ_2{}^\#$ values from glycine to L-valine can be attributed to the difference in interactions of different alkyl chains of amino acids with water. Thus, the results inferred from $\Delta\mu^\circ_2{}^\#$ are consistent with those inferred from $V^\circ_{2,\varphi}$.

As for B-coefficient, the $\Delta\mu^\circ_2{}^\#$ also vary linearly with n_c:

$$\Delta\mu^\circ_2{}^\# = \Delta\mu^\circ_2{}^\# (NH_3^+, COO^-) + n_c\, \Delta\mu^\circ_2{}^\# (CH_2) \tag{9}$$

The regression of $\Delta\mu^\circ_2{}^\#$ data using equation gives $\Delta\mu^\circ_2{}^\#$ (NH_3^+, COO^-) and $\Delta\mu_2^{\circ\#}$ (CH_2) values as respective contributions of (NH_3^+, COO^-) and (CH_2) groups are given in Table 7.

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APPENDIX

Appendix A: The tabulation of measured data of m vs ρ and apparent molar volume for all systems studied is presented below in Table A. Table A: Tabulation of measured data m vs. ρ for the systems studied at different temperatures in absence of AC at 298.15 K

Concentration (m $mol^{-1}\ kg^{-1}$)	Glycine ($\rho/10^3\ kg\ m^{-3}$)	DL-alanine ($\rho/10^3\ kg\ m^{-3}$)	L-Valine ($\rho/10^3\ kg\ m^{-3}$)	L-serine ($\rho/10^3\ kg\ m^{-3}$)
0.01	0.9980	0.9976	0.9979	0.9983
0.03	0.9993	0.9987	0.9985	0.9995
0.05	1.0004	0.9997	0.9970	1.0014
0.07	1.0019	1.0013	1.0009	1.0042
0.09	1.0034	1.0028	1.0018	1.0056
0.1	1.0040	1.0035	1.0028	1.0067

Appendix B: Experimental slope, S_v, of some amino acids at 298.15 K

Amino acid	$S_v\ 10^6$ ($m^3\ mol^{-1}$)
Glycine	0.90±0.04
DL-Alanine	0.74±0.07
L-Valine	0.40±0.2
L-Serine	1.30±0.2

Appendix C: The partial molar volume, $V^\circ_{2,\varphi}$, at infinite dilution for aqueous amino acids solutions at different temperatures in absence of AC

Amino acids	Glycine	DL-alanine	L-Valine	L-Serine
$V^\circ_{2,\varphi}$	43.50±0.1	60.70±0.1	90.90±0.1	60.90±0.1
$10^{-6}\ m^3\ mol^{-1}$	[43.39[b]]	[62.62[b]]		[60.80[b]]

[a]Values in parentheses represent standard deviation, [b]Belibagli and Ayranci (1990)

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