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EFFECT OF TEMPERATURE AND CONCENTRATION ON THE VISCOSITY OF AQUEOUS SOLUTIONS OF 3-AMINOPROPANOIC ACID, 4-AMINOBUTANOIC ACID, 5-AMINOPENTANOIC ACID, 6-AMINOHEXANOIC ACID

EFECTO DE LA TEMPERATURA Y LA CONCENTRACIÓN EN LA VISCOSIDAD DE SOLUCIONES ACUOSAS DE ÁCIDO 3-AMINOPROPANOICO, ÁCIDO 4-AMINOBUTANOICO, ÁCIDO 5-AMINOPENTANOICO, ÁCIDO 6-AMINOHEXANOICO

EFEITO DA TEMPERATURA E A CONCENTRAÇÃO NA VISCOSIDADE DAS SOLUÇÕES AQUOSAS DE ÁCIDO 3-AMINOPROPANOICO, ÁCIDO 4-AMINOBUTANOICO, ÁCIDO 5-AMINOPENTANOICO, ÁCIDO 6-AMINOHEXANOICO

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ABSTRACT

In this work we present the effect of temperature on the viscosities of aqueous solutions of 3-aminopropanoic acid, 4-aminobutanoic acid, 5-aminopentanoic acid and 6-aminohexanoic acid as a function of concentration. The experimental measurements were done from 293.15 K to 308.15 K. At each temperature the experimental data were fitted to the Tsangaris-Martin equation and the *B* viscosity coefficient was determined. The dependence of the *B* coefficients on the number of carbon atoms of the amino acids is linear, so the contribution of polar and apolar groups was established. The results are interpreted in terms of amino acid hydration.

Key words*:* Amino acid; viscosity; hydrophobic hydration; solute-solvent interactions.

RESUMEN

En este trabajo se presenta el efecto de la temperatura sobre las viscosidades de soluciones acuosas de acido 3-aminopropanoico, acido 4-aminobutanoico, acido

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5-aminopentanoico, acido 6-aminohexanoico en función de la concentración. Las determinaciones experimentales se realizaron en un intervalo de temperatura entre 293,15 K hasta 308,15 K. A cada una de las temperaturas trabajadas, los datos experimentales se ajustaron a la ecuación de Tsangaris-Martin y se determinó el coeficiente *B* de viscosidad. La dependencia del coeficiente *B* con el número de carbonos del aminoácido es lineal lo cual permitió establecer la contribución de los grupos polares y apolares del aminoácido. Los resultados se interpretan en términos de la hidratación de los aminoácidos.

Palabras clave: aminoácidos; viscosidad; hidratación hidrofóbica; interacción soluto-solvente.

RESUMO

Neste trabalho é apresentado o efeito da temperatura sobre as viscosidades das soluções aquosas de ácido 3-aminopropanoico, ácido 4-aminobutanoico, ácido 5-aminopentanoico, ácido 6-aminohexanoico em função da concentração. As determinações experimentais foram feitas em um intervalo de temperatura entre os 293,15 K até 308,15 K. Para cada uma das temperaturas trabalhadas, os dados experimentais foram ajustados à equação de Tsangaris-Martin, e o coeficiente *B* de viscosidade foi determinado. A dependência do coeficiente *B* com o número de carbonos do aminoácido é linear e permitiu estabelecer a contribuição dos grupos polares e apolares do aminoácido. Os resultados foram interpretados em termos da hidratação dos aminoácidos.

Palavras-chave: aminoácidos; viscosidade; hidratação hidrofóbica; interação soluto-solvente.

INTRODUCTION

Amino acids have been considered model compounds that can give useful information to understand the behavior of proteins and the role of solvent structure in the denaturation process. For this reason and due to their applications, the properties of amino acids in aqueous solution including their viscometric properties have been extensively studied (1-18). The information has been used to provide an insight about hydrophobicity, hydration properties and solute-solvent interactions. However most of these studies refer to naturally occurring small α-amino acids at 298.15 K and very few data are reported in literature for α,ω-amino acids specially at temperatures different from 298.15 K (5, 8, 15, 16).

Although there is much literature on aqueous solutions of α-amino acids, data on the viscosities of solutions of α,ω-amino acids are limited in particular at temperatures different from 298.15 K. In order to extend the understanding on the behavior of aqueous solutions of amino acids and as continuation of our previous experimental studies on viscosity of the aqueous mixtures of these solutes (11, 13, 14), we report here new experimental viscosity data for aqueous solutions of 3-amino propanoic acid, 4-amino butanoic acid, 5-amino pentanoic acid and 6-amino hexanoic acid as a function of concentration at 293.15, 298.15, 303.15 and 308.15 K. The amino acids were

chosen to examine the effect of increase in the number of CH_2 groups.

The behavior of the viscosity of solutions is a consequence of the different interactions between solute and the solvent molecules. For this reason it depends on the nature of solvent and solute nature, the size and shape of the latter, concentration, and is affected by external factors such as temperature. Several industrial applications are very sensitive to flow behavior and are specially affected by the behavior of viscosity with temperature. In this regard, information about the flow properties of aqueous solutions of α,ω-amino acids is important. Of particular interest is the behavior of the relative viscosity as a function of concentration, since it gives important information about solute hydration.

In this work the relative viscosity results were used to determine the *B*-coefficients of amino acids at the selected temperatures. The dependence of the *B* coefficient with temperature was used to discuss the effect of the hydrocarbon chain on water structure and the hydration properties of the solutes. A comparison between the *B* coefficients of *α*-amino acids and *α,ω*-amino acids is presented. The results show that the coefficients for *α,ω*-amino acids are smaller than the corresponding values for *α*-amino acids at the same temperatures suggesting that solute–solvent hydrophobic interactions are larger for *α*-amino acids due to the more exposed aliphatic chain to the solvent.

MATERIALS AND METHODS

The materials used in this work were: 3-aminopropanoic acid (β-alanine) Merck purity ≥ 99 %, 4-aminobutanoic acid (γ-aminobutyric acid) Merck purity ≥ 98 %, 5-aminopentanoic acid (δ-aminovaleric acid) Aldrich purity ≥ 97 % and 6-aminohexanoic acid (ε-aminocaproic acid) Merck purity ≥ 99 %. The amino acids were dried under vacuum at room temperature and kept in desiccator at least 48 h before use. Water was doubly distilled and deionized according to literature and degassed before use obtaining a product with conductivity less than 2 μS m^{-1} (19, 20). All solutions were prepared by weight using a Mettler balance AT 261 dual range with sensitivity of 10^{-5} g in the lower range.

Viscosity η was determined using two Ubbelohde viscometers with efflux times *t* near 500 s for water that were calibrated with water at the selected temperatures. Reproducibility of efflux times was in all cases better than 0.05 %. Efflux times were determined at 293.15, 298.15, 303.15 and 308.15 K for the amino acid aqueous solutions. All measurements were carried out in a constant temperature bath with temperature controlled to ± 0.005 K. Reported values are the average of at least three independent measurements in each viscometer. Densities ρ of the solutions at the selected temperatures were taken from a previous work (21). They were measured at the same temperatures using pycnometers of the Wood-Brusie type, in a constant temperature bath with temperature controlled to ± 0.01 K. Density values are the average of three independent measurements and uncertainty in the density measurements is estimated to be $\pm 5 \cdot 10^{-5}$ g cm^{-3}

The viscosity data were obtained from the general equation:

$$\eta = \alpha\rho t - \beta\rho / t \quad [1]$$

where α and β are the viscometer constants obtained by calibration with water at each temperature, ρ is the density and *t* the efflux time. The relative viscosities $\eta_r = \eta/\eta_o$ were calculated from the solution and solvent viscosities respectively.

RESULTS AND DISCUSSION

Tables 1-4 show the experimental results for density and viscosity of the aqueous solutions of 3-aminopropanoic acid, 4-aminobutanoic acid, 5-aminopentanoic acid, and 6-aminohexanoic acid as a function of molality at 293.15, 298.15, 303.15 and 308.15 K. The density data were obtained adjusting the values reported in literature from a previous work at the same temperatures considered in this work (21).

The concentration dependence of the viscosity of aqueous solutions of α,ω-amino acids exhibits the usual behavior. At all temperatures, solution viscosity increases with amino acid concentration and the effect is more pronounced as the length of the hydrocarbon chain becomes larger. From Tables 1-4 it can be seen that for each amino acid, the viscosity decreases as temperature increases.

The relative viscosity values were calculated and adjusted by least-squares to a second order equation as proposed by Tsangaris-Martin (3).

$$\eta_r = 1 + Bm + Dm^2 \quad [2]$$

In this equation *B* and *D* are empirical coefficients; however it has been suggested that the *B* coefficient depends on the size, shape and charge of the solute molecule and is sensitive to solute solvent interactions (4, 15-18). The effect of temperature on the viscosity of the solutes in aqueous solution is well described by a polynomial second order equation. Table 5 shows the values of the *B* coefficients of α,ω-amino acids in aqueous

Table 1. Density and viscosity for 3-aminopropanoic acid in aqueous solution at 293.15, 298.15, 303.15 and 308.15 K.

m	293.15		298.15 K		303.15		308.15	
	ρ	*η*	*ρ*	*η*	*ρ*	*η*	*ρ*	*η*
mol kg^{-1}	10^{-3} kg m^{-3}	10^{3} Pa s	10^{-3} kg m^{-3}	10^{3} Pa s	10^{-3} kg m^{-3}	10^{3} Pa s	10^{-3} kg m^{-3}	10^{3} Pa s
0.99013	1.02690	1.31020	1.02551	1.12019	1.02392	1.04220	1.02221	0.91220
0.7258	1.01940	1.21456	1.01807	1.05330	1.01656	0.96240	1.01487	0.84020
0.6308	1.01663	1.16882	1.01532	1.01198	1.01384	0.91943	1.01216	0.82410
0.4944	1.01271	1.11266	1.01144	0.96770	1.00999	0.87522	1.00834	0.78710
0.3492	1.00847	1.08555	1.00723	0.94120	1.00583	0.84967	1.00419	0.76404
0.2437	1.00554	1.05684	1.00432	0.92214	1.00295	0.82844	1.00132	0.74836
0.0938	1.00113	1.01941	0.99995	0.89480	0.99862	0.80286	0.99701	0.72917
0.0662	1.00032	1.01012	0.99914	0.88120	0.99782	0.79874	0.99621	0.72604

Table 2. Density and viscosity for 4-aminobutanoic acid in aqueous solution at 293.15, 298.15, 303.15 and 308.15 K.

m	293.15		298.15 K		303.15		308.15	
	ρ	η	ρ	η	ρ	η	ρ	η
mol kg^{-1}	10^{-3} kg m^{-3}	10^{3} Pa s	10^{-3} kg m^{-3}	10^{3} Pa s	10^{-3} kg m^{-3}	10^{3} Pa s	10^{-3} kg m^{-3}	10^{3} Pa s
0.99013	1.0252	1.3624	1.0239	1.2123	1.0224	1.08623	1.0206	0.9970
0.7212	1.0184	1.2622	1.0171	1.1056	1.0156	0.99410	1.0139	0.8942
0.6194	1.0156	1.2034	1.0143	1.0704	1.0128	0.94011	1.0111	0.8529
0.3919	1.0093	1.1231	1.0081	0.9968	1.0067	0.89086	1.0050	0.8246
0.2703	1.0059	1.0934	1.0047	0.9605	1.0033	0.87224	1.0016	0.7803
0.2540	1.0055	1.0664	1.0043	0.9558	1.0029	0.85345	1.0013	0.7764
0.1046	1.0013	1.0320	1.0002	0.9142	0.9988	0.81514	0.9972	0.7419
0.0715	1.0004	1.0222	0.9993	0.9055	0.9979	0.80697	0.9963	0.7344

Table 3. Density and viscosity for 5-aminopentanoic acid in aqueous solution at 293.15, 298.15, 303.15 and 308.15 K.

m	293.15		298.15 K		303.15		308.15	
	ρ	η	ρ	η	ρ	η	ρ	η
mol kg^{-1}	10^{-3} kg m^{-3}	10^{3} Pa s	10^{-3} kg m^{-3}	10^{3} Pa s	10^{-3} kg m^{-3}	10^{3} Pa s	10^{-3} kg m^{-3}	10^{3} Pa s
0.9577	1.0236	1.4688	1.0222	1.3752	1.0206	1.1911	1.0190	1.0427
0.7240	1.0175	1.3453	1.0161	1.2120	1.0146	1.0742	1.0129	0.9432
0.6237	1.0149	1.2793	1.0136	1.1539	1.0121	1.0136	1.0104	0.8995
0.4805	1.0111	1.1953	1.0099	1.0782	1.0084	0.9496	1.0068	0.8441
0.3320	1.0071	1.1210	1.0059	1.0090	1.0044	0.8924	1.0028	0.7954
0.2393	1.0047	1.0812	1.0035	0.9704	1.0020	0.8613	1.0004	0.7695
0.0938	1.0008	1.0290	0.9996	0.9173	0.9982	0.8195	0.9966	0.7359
0.0662	1.0000	1.0205	0.9989	0.9082	0.9975	0.8126	0.9959	0.7305

solution obtained in this work at 293.15, 298.15, 303.15 and 308.15 K.

α,ω-amino acids in aqueous solution show relatively large positive *B* coefficients that increase with the number of CH_2 groups. This behavior indicates that the magnitude of the coefficients is a consequence of the increase in the hydrophobic character of the amino acid as the hydrocarbon chain length increases.

Even though the effect of temperature is small, the observed trend shows a de-

Table 4. Density and viscosity for 6-aminohexanoic acid in aqueous solutions at 293.15, 298.15, 303.15 and 308.15 K.

m	293.15		298.15 K		303.15		308.15	
	ρ	η	ρ	η	ρ	η	ρ	η
mol kg^{-1}	10^{-3} kg m^{-3}	10^{3} Pa s	10^{-3} kg m^{-3}	10^{3} Pa s	10^{-3} kg m^{-3}	10^{3} Pa s	10^{-3} kg m^{-3}	10^{3} Pa s
0.9577	1.0220	1.7255	1.0206	1.4210	1.0190	1.3024	1.0172	1.1901
0.7368	1.0168	1.5233	1.0155	1.2676	1.0140	1.1522	1.0122	1.0455
0.6163	1.0139	1.4006	1.0126	1.1918	1.0111	1.0724	1.0094	0.9788
0.4860	1.0105	1.2926	1.0093	1.1160	1.0078	0.9989	1.0061	0.8902
0.3440	1.0072	1.1895	1.0060	1.0407	1.0046	0.9300	1.0029	0.8337
0.2507	1.0048	1.1299	1.0036	0.9955	1.0022	0.8892	1.0006	0.7965
0.0938	1.0010	1.0443	0.9999	0.9268	0.9986	0.8288	0.9969	0.7442
0.0662	1.0003	1.0312	0.9992	0.9157	0.9979	0.8192	0.9963	0.7363

Table 5. *B* coefficients of 3-aminopropanoic acid, 4-aminobutanoic acid, 5-aminopentanoic acid, and 6-aminohexanoic acid in aqueous solution at 293.15, 298.15, 303.15 and 308.15 K.

Solute	*T*	*B*
	K	kg mol^{-1}
3-aminopropanoic acid	293.15	0.1921
	298.15	0.1896
	303.15	0.1538
	308.15	0.1305
4-aminobutanoic acid	293.15	0.2904
	298.15	0.2701
	303.15	0.2539
	308.15	0.2254
5-aminopentanoic acid	293.15	0.3621
	298.15	0.3069
	303.15	0.2983
	308.15	0.2758
6-aminohexanoic acid	293.15	0.4604
	298.15	0.4136
	303.15	0.3788
	308.15	0.3521

crease of the coefficients with temperature. The first derivative of the *B* coefficient with temperature d*B*/d*T* has been used as indicative of the effect of solute on water structure (16-18). The negative sign has been attributed to a structure making effect while the positive sign to a structure breaking effect on water structure. In this study, all the solutes considered, shows a negative value for d*B*/d*T* suggesting that 3-aminopropanoic acid, 4-aminobutanoic acid, 5-aminopentanoic acid and 6-aminohexanoic acid have a water structure making character.

Table 6 presents a comparison between the *B* coefficients of α-amino acids and α,ω-amino acids including literature data.

For glycine, the coefficient increases with temperature, as has been reported by several authors who explain this behavior as a consequence of the breaking structure effect that this solute exerts on water structure (2, 4, 5, 16). For the other

Table 6. *B* coefficients of α-amino acids and α,ω-amino acids at 293.15, 298.15, 303.15 and 308.15 K.

Amino acid	*B* kg mol-1			
	293.15 K	298.15 K	303.15 K	308.15 K
Glycine	0.1339 [a]	0.143 [b] 0.143 [c]	0.137 [d]	0.153 [e] 0.148 [f]
2-aminopropanoic		0.251 [c] 0.259 [f] 0.2525[g]	0.24 [d]	0.241 [e] 0.251 [f]
3-aminopropanoic [h]	0,1921	0.1896	0.1538	0.1305
2-aminobutanoic [i]	0.3103	0.295	0.2793	
4-aminobutanoic [h]	0.2904	0.2701	0.2539	0.2254
2-aminopentanoic [i]	0.3993	0.371	0.3519	
5-aminopentanoic [h]	0.3621	0.3069	0.2983	0.2758
2-aminohexanoic [i]	0.4925	0.4543	0.4342	
6-aminohexanoic [h]	0.4604	0.4136	0.3788	0.3521

[a] Ref. (4); [b] Ref. (16); [c] Ref. (18); [d] Ref. (1); [e] Ref. (5); [f] Ref. (2); [g] Ref. (13); [h] This work; [i] Ref. (11).

amino acids, the behavior changes, the coefficient decreases with temperature and d*B*/d*T* shows a negative value at all temperatures suggesting that they have a water structure making character. The calculated values are small and do not follow a definite trend. Taking into account the experimental uncertainty, they are not reported and the discussion is referred only to the sign of the slope.

It is clear that in aqueous solutions, the coefficients for α,ω-amino acids are smaller than the corresponding values for *α*-amino acids at the same temperatures as is shown in Table 6. This suggests that solute–solvent hydrophobic interactions are larger for *α*-amino acids due to the more exposed aliphatic chain to the solvent (21).

At each temperature the *B* coefficient is a linear function of the number of carbon atoms of the hydrocarbon chain. These results show that it can be described in terms of the contributions of the charged and methylene groups, according to a simple additivity approach. This simple model is based on the dependence of the property on the number of carbon atoms or the number of methylene groups (11, 18, 22, 23), and can be represented by the following equation:

$$B = B_{(NH_3^+, COO^-)} + nB_{(CH_2)} \qquad [3]$$

Table 7 shows the behavior of the viscometric group contributions to the *B* coefficient with temperature. The observed change is very small and the general trend is to diminish as temperature in-

creases, following the observed behavior reported in literature for α-amino acids (11, 16).

Table 7. Methylene contribution to the *B* coefficient.

T	B_{CH_2}
K	**kg mol^{-1}**
293.15	0.0877
298.15	0.0709
303.15	0.0719
308.15	0.0715

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