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Solute–solute and solute–solvent interactions of L-histidine and L-arginine in aqueous-streptomycin sulphate solutions at different temperatures: A physicochemical study

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ABSTRACT

The densities, ρ of L-histidine and L-arginine in water and in aqueous-streptomycin sulphate [1% (0.0069 mol·kg^{-1}) and 2% (0.0137 mol·kg^{-1}) streptomycin sulphate in water] mixed solvents were measured at temperatures 293.15, 298.15, 303.15, 308.15, 313.15 and 318.15 K; and ultrasonic speed, u and viscosities, η of these solutions were measured at temperatures 298.15, 303.15, 308.15, 313.15 and 318.15 K and at atmospheric pressure. The ρ, u and η data have been used to calculate apparent molar volume, V_φ, limiting apparent molar volume, V°_ϕ, apparent molar compressibility, $K_{s,\phi}$, limiting apparent molar compressibility, $K^\circ_{s,\phi}$, transfer volume, $V^\circ_{\phi,tr}$, transfer compressibility, $K^\circ_{s,\phi,tr}$, and partial molar expansibility, E°_ϕ. The viscosity data have been used to determine Falkenhagen coefficient, A, Jones–Dole coefficient, B, free energy of activation of viscous flow per mole of solvent, $\Delta\mu^{\circ\#}_1$ and per mole of solute, $\Delta\mu^{\circ\#}_2$. The calculated parameters have been discussed in terms of various solute–solute and solute–solvent interactions prevailing in these solutions. The structure making/breaking ability of the amino acids in the aqueous-streptomycin sulphate solution is also discussed.

1. Introduction

Most biochemical processes occur in aqueous medium; therefore studies on physicochemical properties of biomolecules like amino acids, sugars and drugs in aqueous solution provide useful information which help in understanding the complex mechanism of molecular interactions. Thermodynamic and physicochemical measurements are useful in understanding the solute–solvent and solute–solute interactions in solutions [1–5]. When certain solutes are added to protein solutions they cause stabilization and destabilization to the proteins. Sometimes protein conformation is affected by these added co-solutes due to solvent effects or their direct binding. But due to complicated structure of proteins the study of protein–drug interactions is somewhat difficult [6–10]. Therefore, for a better understanding of the hydration behaviour of proteins, one useful approach is to study simpler model compounds such as amino acids. Several physicochemical properties of constituent amino acids in aqueous and mixed aqueous solutions have been used by various researchers to investigate solute–solvent interactions [11–16].

In our body drug–protein interactions play a very vital role in metabolic pathways or the biological processes occurring inside. So the characterization of drugs in aqueous and non-aqueous solutions had been a subject of interest because their activities involve interaction with biological membranes [17–21]. Thus, the properties of amino acids in aqueous-drug solutions are essential for understanding the chemistry of biological systems. Despite the importance of the subject, only a few physicochemical studies of amino acids in aqueous-drug solutions have been reported [22–26]. Therefore, we planned to carry out the volumetric, ultrasonic and viscometric studies of L-histidine and L-arginine in aqueous solutions of antibacterial drug streptomycin sulphate (chemical formula: $C_{42}H_{84}N_{14}O_{36}S_3$) to investigate interactions in these systems. The structure of streptomycin sulphate is given in Fig. 1.

In the present article, we report the densities, ρ of L-histidine and L-arginine in water and in aqueous-streptomycin sulphate [1% (0.0069 mol·kg^{-1}) and 2% (0.0138 mol·kg^{-1}) streptomycin sulphate in water] mixed solvents at temperatures 293.15, 298.15, 303.15, 308.15, 313.15 and 318.15 K; and ultrasonic speed, u and viscosities, η of these solutions at temperatures 298.15, 303.15, 308.15, 313.15 and 318.15 K and at atmospheric pressure. These experimental data have been used to calculate the apparent molar volume, V_ϕ, limiting apparent molar volume, V°_ϕ and the slope, S_v, transfer volume, $V^\circ_{\phi,tr}$, apparent molar compressibility, $K_{s,\phi}$, limiting apparent molar compressibility, $K^\circ_{s,\phi}$ and the slope, S_k, transfer volume, $V^\circ_{\phi,tr}$, transfer compressibility, $K^\circ_{s,\phi,tr}$, limiting apparent molar expansivity, E°_ϕ, Falkenhagen coefficient,

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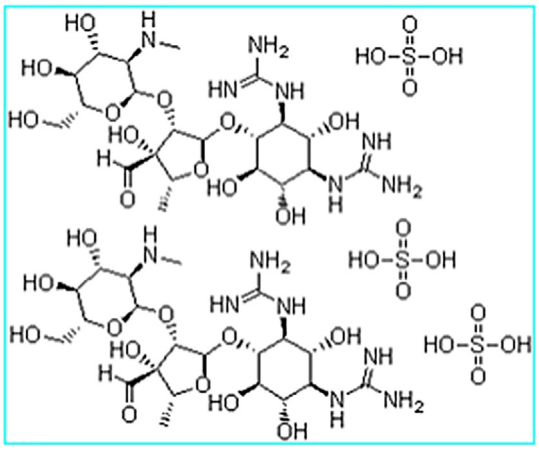

Fig. 1. Structure of streptomycin sulphate (chemical formula: $C_{42}H_{84}N_{14}O_{36}S_3$).

A, Jones–Dole coefficient, *B*, free energies of activation of viscous flow per mole of solvent, $\Delta\mu_1^{\circ\#}$ and per mole of solute, $\Delta\mu_2^{\circ\#}$. The results have been interpreted in terms of solute–solvent and solute–solute interactions in these systems.

2. Experimental

2.1. Chemicals

L-Histidine (SRL India, purity >99%) and L-arginine (SRL India, purity >99%) were recrystallized from ethanol–water solutions, and dried in vacuum at room temperature for 24 h. Thereafter, these chemicals were stored over P_2O_5 in desiccator before use. The drug streptomycin sulphate (SRL India, purity >99%) was used as such without further purification, except drying in oven for 24 h. The purity of the purified chemicals was checked by performing gas chromatography analysis using Shimadzu Gas Chromatograph (Model: GC-2010 Plus). The final purities and other specifications of the chemicals used are given in Table 1. The aqueous-drug solutions [1% (0.0069 mol·kg^{-1}) and 2% (0.0138 mol·kg^{-1}) streptomycin sulphate in water] were prepared using triple distilled water (conductivity less than $1 \cdot 10^{-6}$ S·cm^{-1}). These drug solutions were used as solvents to prepare solutions of eight different molal concentrations of L-histidine (ranging from 0 to 0.16 mol·kg^{-1}) and L-arginine (ranging from 0 to 0.2 mol·kg^{-1}). The weighings were done on an electronic balance (Model: GR-202R, AND, Japan) with a precision of ± 0.01 mg. All the solutions were prepared with care and stored in special airtight bottles to avoid contamination and evaporation. The uncertainty in the molality of the solutions was estimated within $\pm 1 \cdot 10^{-4}$ mol·kg^{-1}.

2.2. Equipments and procedures

The densities of the solutions were measured by using a single-capillary pycnometer (made of Borosil glass) having a bulb capacity of ~10 mL. The capillary, with graduated marks, had a uniform bore and could be closed by a well-fitting glass cap. The marks on the capillary were calibrated by using triple distilled water. The densities of pure water used in the calibration at required temperatures were taken from the literature [27]. The uncertainty in density measurements was within $\pm 2 \cdot 10^{-2}$ kg·m^{-3}. The ultrasonic speeds in the solutions were measured using a single-crystal variable-path multifrequency ultrasonic interferometer (Model: M-81DS, Mittal Enterprises, India) having stainless steel sample cell (with digital micrometer) operating at 3 MHz. The uncertainty in ultrasonic speed measurements was within ±0.3 m·s^{-1}. The viscosities of the solutions were measured by using Ubbelohde type suspended level viscometer. The viscometer

Table 2
Densities, ρ/kg·m^{-3} of solutions of L-histidine and L-arginine in streptomycin sulphate + water (1 and 2% streptomycin sulphate, w/w in water) solvents as functions of molality, *m* of L-histidine/L-arginine at different temperatures and at atmospheric pressure.

m/mol · kg^{-1}	*T*/K					
	293.15	298.15	303.15	308.15	313.15	318.15
L-Histidine in 1% aqueous-streptomycin sulphate						
0.0000	1002.76	1001.29	999.81	998.34	996.86	995.39
0.0198	1003.86	1002.39	1000.92	999.45	997.98	996.51
0.0397	1004.97	1003.50	1002.03	1000.57	999.10	997.63
0.0598	1006.08	1004.62	1003.15	1001.70	1000.23	998.77
0.0799	1007.20	1005.74	1004.27	1002.82	1001.36	999.90
0.1000	1008.31	1006.86	1005.40	1003.95	1002.49	1001.04
0.1196	1009.40	1007.95	1006.49	1005.05	1003.59	1002.14
0.1402	1010.54	1009.09	1007.64	1006.20	1004.74	1003.30
0.1601	1011.64	1010.19	1008.75	1007.31	1005.86	1004.42
L-Histidine in 2% aqueous-streptomycin sulphate						
0.0000	1007.26	1005.43	1003.60	1001.77	999.94	998.11
0.0199	1008.33	1006.50	1004.68	1002.86	1001.04	999.21
0.0402	1009.42	1007.60	1005.78	1003.97	1002.15	1000.33
0.0598	1010.47	1008.66	1006.85	1005.04	1003.22	1001.41
0.0798	1011.54	1009.74	1007.93	1006.13	1004.32	1002.52
0.1004	1012.65	1010.85	1009.05	1007.25	1005.45	1003.65
0.1203	1013.72	1011.92	1010.12	1008.33	1006.54	1004.75
0.1399	1014.77	1012.98	1011.19	1009.40	1007.61	1005.83
0.1601	1015.86	1014.07	1012.28	1010.49	1008.71	1006.93
L-Arginine in 1% aqueous-streptomycin sulphate						
0.0000	1002.76	1001.29	999.81	998.34	996.86	995.39
0.0248	1003.97	1002.51	1001.04	999.57	998.10	996.64
0.0503	1005.22	1003.76	1002.30	1000.84	999.38	997.92
0.0752	1006.43	1004.98	1003.52	1002.07	1000.62	999.17
0.1001	1007.64	1006.19	1004.74	1003.30	1001.85	1000.41
0.1247	1008.83	1007.39	1005.94	1004.50	1003.06	1001.63
0.1502	1010.06	1008.63	1007.18	1005.75	1004.31	1002.89
0.1754	1011.27	1009.84	1008.41	1006.98	1005.55	1004.13
0.1997	1012.43	1011.02	1009.58	1008.16	1006.74	1005.33
L-Arginine in 2% aqueous-streptomycin sulphate						
0.0000	1007.26	1005.43	1003.60	1001.77	999.94	998.11
0.0248	1008.43	1006.61	1004.79	1002.97	1001.15	999.33
0.0499	1009.61	1007.79	1005.98	1004.17	1002.37	1000.56
0.0748	1010.77	1008.97	1007.17	1005.37	1003.57	1001.77
0.1002	1011.96	1010.17	1008.37	1006.58	1004.79	1003.01
0.1249	1013.11	1011.33	1009.54	1007.76	1005.98	1004.20
0.1502	1014.29	1012.52	1010.73	1008.96	1007.19	1005.42
0.1751	1015.44	1013.68	1011.91	1010.14	1008.38	1006.62
0.1998	1016.59	1014.83	1013.07	1011.31	1009.56	1007.81

Table 1
Specification of chemical samples.

Chemical name (CAS number)	Provenance	Purification method	Final mass fraction purity	Analysis method
L-Histidine (71-00-1)	SRL, India	Re-crystallization	>0.994	GC[a]
L-Arginine (7200-25-1)	SRL, India	Re-crystallization	>0.994	GC
Streptomycin sulphate (3810-74-0)	SRL, India	Re-crystallization	>0.99	GC

[a] GC = gas chromatography.