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Volumetric and Viscometric Studies of Some Amino Acids in Aqueous Solutions of Cadmium Chloride at T = (288.15 to 318.15) K and at Atmospheric Pressure

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Supporting Information

ABSTRACT: The apparent molar volumes, apparent molar adiabatic compressibilities, and viscosities of glycine, DL-α-alanine, DL-α-amino-*n*-butyric acid, L-valine, and L-leucine, in aqueous solutions of cadmium chloride, $CdCl_2$, at different concentrations have been determined in the temperature range (288.15 to 318.15) K from density, sound velocity, and time flow measurements, using a vibrating-tube digital densimeter, ultrasonic multifrequency interferometer, and Ubbelohde type capillary viscometer attached with an automatic viscosity measuring unit, respectively. From these data, partial molar volumes V^{o}_{2}, partial molar adiabatic compressibilities, $K^{o}_{S,2}$, viscosity B-coefficients, and the corresponding transfer parameters ($\Delta_t V^{o}_{2}$, $\Delta_t K^{o}_{S,2}$, and $\Delta_t B$), hydration numbers, n_H, and side chain group contributions, dB/dT have been calculated. Transfer parameters are positive and increase with the concentration of $CdCl_2$.

INTRODUCTION

Studies on the solubility and stability of proteins have generated a great interest for a long time, but because of complications involved in dealing with these complex molecules, various low molecular weight model compounds are generally taken for investigations.[1] Therefore, the physicochemical properties of amino acids, peptides, and their derivatives which mimic some specific aspects of protein structure in aqueous solutions have been extensively studied to gain a better understanding of solute−solvent interactions and their role in the conformational stability of proteins.[2] The completely unfolded, random coil, form of a denatured protein is of particular interest in the discussion of the thermodynamic stability of protein because it constitutes an ideal reference state.[3,4]

Salt solutions are known to produce remarkable effects on the conformation and properties of proteins.[5] The interactions between solvent and various constituent groups of a protein such as the amino acid side chain and the peptide backbone group play a central role in the structure, the conformation, and the function of proteins in aqueous solutions.[6] It has been established that metal ions play crucial roles in various biological processes. They are generally involved in enzyme regulation, stabilization of structure of reactive molecules, transportation to transmembrane channels, and so forth.[7] Transition metal ions (zinc, copper, iron, manganese, and cobalt) play a vital role in life systems, because of their natural presence in vitamins, enzymes, and proteins. Zinc ions are very important for a number of biological functions in a human body. Cadmium ions are mainly found in the biological systems in the form of Zn/Cd metallothionein (cysteine−rich protein) complex. Metallothionein are the only biomolecules known to normally contain cadmium. Therefore cadmium ion is found in association with proteins whose functions are probably involved with essential trace element (zinc) metabolism.[8] The effect of ions of salt on the solution behavior of amino acids can be studied through the volumetric and viscometric properties. Such studies on the solution behavior of amino acids are of great relevance because all biological fluids are not pure water and additives have a significant impact on protein stability and functions. These data are quite important for interpreting the hydration behavior of peptides and proteins using the additivity scheme. Further the temperature dependence of these properties is helpful in quantifying the kosmotropicity of amino acids in salt solution.[4] Therefore, in this paper we report the densities, sound velocities, and viscosities of glycine, DL-α-alanine, DL-α-amino-*n*-butyric acid, L-valine, and L-leucine, in aqueous $CdCl_2$ solutions of (0.1, 0.5, 1.0, and 1.5) mol·kg^{-1} at (288.15, 298.15, 308.15, and 318.15) K. From these data, various transfer parameters $\Delta_t V^{o}_{2}$, $\Delta_t K^{o}_{S,2}$, and $\Delta_t B$, hydration numbers, n_H, and side chain group contributions have been calculated.

EXPERIMENTAL SECTION

Glycine (G-7126, 0.99 mass fraction), DL-α-alanine (A-7502, 0.99 mass fraction), DL-α-amino-*n*-butyric acid (A-1754, 0.98 mass fraction), L-valine (V-0500, 0.99 mass fraction), and L-leucine (L-8000, 0.98 mass fraction) were obtained from Sigma Chemical Co. and were dried for 24 h in a vacuum oven before use. Analytical grade cadmium chloride monohydrate procured from CDH having purity 0.98 mass fraction was used as such after drying for 72 h in a vacuum desiccator at room temperature. Deionized, doubly distilled degassed water with specific conductance less than $1.29 \cdot 10^{-6}$ $\Omega^{-1} \cdot cm^{-1}$ was used for the preparation of all solutions. The solutions were prepared on the mass basis by using a Mettler balance having an accuracy of ± 0.01 mg. The densities of the solutions were measured by using

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Table 1. Partial Molar Volumes, V_2^o, of Some Amino Acids in Aqueous $CdCl_2$ from T = (288.15 to 318.15) K

	$V_2^o \cdot 10^6$ ($m^3 \cdot mol^{-1}$)			
amino acid	m_B/mol·kg^{-1} = 0.1	0.5	1.0	1.5
		T/K = 288.15		
glycine	44.11 ± 0.02	44.85 ± 0.01	45.63 ± 0.02	46.08 ± 0.01
DL-α-alanine	60.88 ± 0.01	61.93 ± 0.02	62.72 ± 0.01	63.15 ± 0.01
DL-α-amino-*n*-butyric acid	75.31 ± 0.04	75.92 ± 0.01	76.58 ± 0.01	77.40 ± 0.01
L-valine	90.41 ± 0.01	90.99 ± 0.02	91.76 ± 0.01	92.57 ± 0.01
L-leucine	106.91 ± 0.01	107.02 ± 0.01	107.09 ± 0.01	107.21 ± 0.01
		T/K = 298.15		
glycine	45.18 ± 0.02	46.13 ± 0.01	46.85 ± 0.01	47.56 ± 0.01
DL-α-alanine	61.84 ± 0.01	62.61 ± 0.01	63.70 ± 0.01	64.22 ± 0.01
DL-α-amino-*n*-butyric acid	76.54 ± 0.02	77.23 ± 0.01	78.50 ± 0.01	78.95 ± 0.01
L-valine	91.29 ± 0.01	92.06 ± 0.01	93.15 ± 0.01	93.73 ± 0.01
L-leucine	107.80 ± 0.01	107.96 ± 0.01	108.06 ± 0.01	108.22 ± 0.01
		T/K = 308.15		
glycine	46.03 ± 0.02	47.09 ± 0.02	48.04 ± 0.01	48.81 ± 0.01
DL-α-alanine	62.93 ± 0.01	63.56 ± 0.01	64.82 ± 0.01	65.29 ± 0.01
DL-α-amino-*n*-butyric acid	78.00 ± 0.02	78.63 ± 0.01	79.81 ± 0.01	80.42 ± 0.02
L-valine	92.31 ± 0.01	93.16 ± 0.01	94.48 ± 0.01	94.81 ± 0.01
L-leucine	108.57 ± 0.01	108.88 ± 0.01	109.01 ± 0.01	109.35 ± 0.02
		T/K = 318.15		
glycine	47.29 ± 0.03	48.17 ± 0.01	49.23 ± 0.01	50.10 ± 0.01
DL-α-alanine	63.81 ± 0.01	64.71 ± 0.01	65.46 ± 0.01	66.26 ± 0.01
DL-α-amino-*n*-butyric acid	78.54 ± 0.02	79.40 ± 0.01	80.13 ± 0.01	80.88 ± 0.01
L-valine	92.99 ± 0.01	94.32 ± 0.01	95.11 ± 0.01	95.73 ± 0.01
L-leucine	109.59 ± 0.01	109.85 ± 0.01	109.98 ± 0.02	110.41 ± 0.02

vibrating-tube digital densimeter (model DMA 60/602, Anton Paar, Austria).[9] The calculated uncertainty value in the experimentally determined densities on average is $3.7 \cdot 10^{-3}$ kg·m^{-3}. The measured values of densities of aqueous solutions of sodium chloride at 298.15 K agree well with literature values.[10−12]

The sound velocities were measured using a multifrequency ultrasonic interferometer (model M-82, Mittal Enterprises), with a maximum uncertainty of 0.5 m·s^{-1} in velocity. The temperature of the solution was maintained within 0.01 K using a constant temperature circulator bath (model MVF 25 Julabo/Germany) for velocity measurements. The sound velocity data of aqueous solutions of glycine at 298.15 K have been compared with literature values,[13−15] and good agreement has been observed.

Solution viscosities were measured by a suspended level Micro Ubbelohde viscometer, which was placed in a water thermostat (model MC 31A Julabo/Germany) having a temperature stability within 0.01 K. Flow time measurements were performed using an automatic viscosity time measurement unit (SCHOTT AVS 350) with a resolution of 0.01 s. The average of at least six readings was used as the final efflux time. The measured viscosities of aqueous solutions of glycine at 298.15 K have been compared with literature values which agree well with the literature values.[16,17] The viscosities for water at different temperatures were taken from the literature.[18] The measured viscosity has an uncertainty of ± 0.001 mPa·s.

RESULTS AND DISCUSSION

Apparent Molar Volumes. The density data measured for the studied amino acids at m_B (molality of $CdCl_2$ in water) = (0.1, 0.5, 1.0, and 1.5) mol·kg^{-1} aqueous $CdCl_2$ solutions at (288.15, 298.15, 308.15, and 318.15) K are given in Table S1 of the Supporting Information. The densities of the studied amino acids in aqueous solutions of $CdCl_2$ decrease with temperature and the decrease in density is relatively more at higher concentration of $CdCl_2$. Further the densities of amino acid solutions increase with the increase of the concentration of aqueous $CdCl_2$ solutions. The apparent molar volumes, $V_{2,\phi}$, of amino acids have been calculated using the following relation:

$$V_{2,\phi} = (M/\rho) - [(\rho - \rho_o)/(m_A \cdot \rho \cdot \rho_o)] \quad (1)$$

where M and m_A are, respectively, the molar mass and the molality of amino acids, and ρ and ρ_o are the densities of solution and the solvent, respectively. The uncertainty occurring due to various measured quantities in $V_{2,\phi}$ range from $0.007 \cdot 10^{-6}$ to $0.002 \cdot 10^{-6}$ m^3·mol^{-1} for the lower ($m_A \leq 0.0451$) and higher concentration ranges, respectively, over the studied temperature range. However, in the case of L-valine and L-leucine the uncertainty in $V_{2,\phi}$ is slightly higher because a very dilute concentration range has been studied. The uncertainty values in $V_{2,\phi}$ (for L-valine and L-leucine) range from ($0.033 \cdot 10^{-6}$ to $0.005 \cdot 10^{-6}$) m^3·mol^{-1} for the lower ($m_A \leq 0.0123$) and higher concentration ranges, respectively.

The apparent molar volume data found to be adequately represented by the linear equation:

$$V_{2,\phi} = V_2^o + S_v m_A \quad (2)$$

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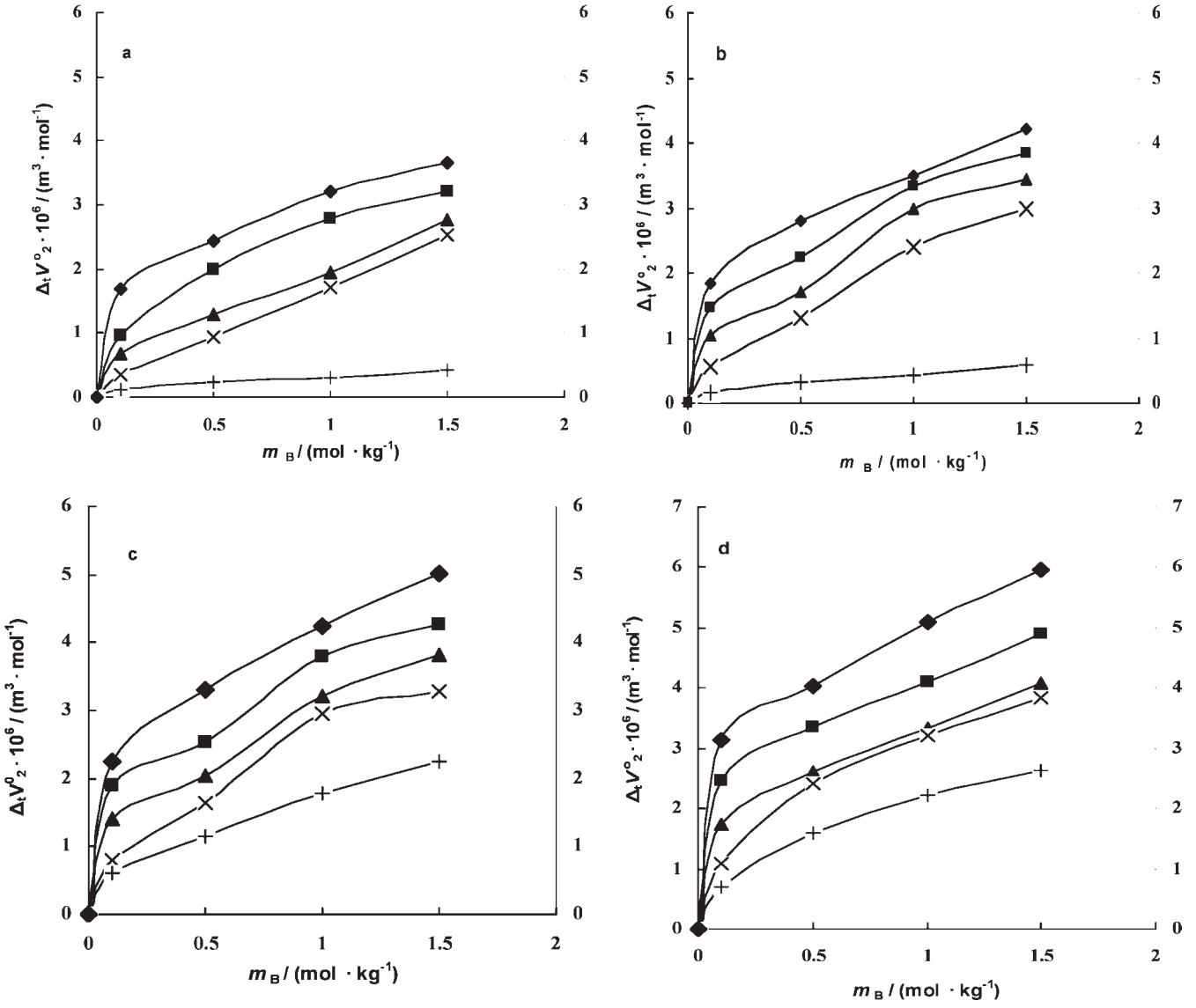

Figure 1. Partial molar volume of transfer, $\Delta_t V_2^o$, of some amino acids vs m_B, molalities of cadmium chloride: ♦, glycine; ■, DL-α-alanine; ▲, DL-α-amino-*n*-butyric acid; ×, L-valine; +, L-leucine at (a) 288.15 K; (b) 298.15 K; (c) 308.15 K; (d) 318.15 K.

where V_2^o is the apparent molar volume at infinite dilution, also known as the partial molar volume, and S_v is the experimental slope; the values are summarized in Table 1 along with standard deviations.

Partial molar volumes of transfer, $\Delta_t V_2^o$ at infinite dilution from water (data reported earlier[9]) to aqueous $CdCl_2$ solutions have been calculated as follows:

$$\Delta_t V_2^o = V_2^o(\text{in aqueous } CdCl_2 \text{ solutions}) - V_2^o(\text{in water}) \quad (3)$$

The V_2^o values for the studied amino acids in aqueous solutions of cadmium chloride are higher than the values in water,[9] which result in positive $\Delta_t V_2^o$ values. These values increase with the increase of the molality of cadmium chloride (Figure 1), and the increase is sharp in the lower concentration range. The transfer values also increase with temperature in all cases. The $\Delta_t V_2^o$ values vary almost linearly after $m_B \approx 0.1$ mol·kg^{-1}.

A comparison of the present transfer volumes of amino acids (Table S2 of the Supporting Information) with the literature data[9,19−23] (in the presence of various electrolytes) in the concentration range of $m_B \approx$ (0.1 to 1.25) mol·kg^{-1} at temperatures of (288.15 to 308.15) K reveals that in general the transfer volumes, $\Delta_t V_2^o$, are positive in the presence of various electrolytes and the $\Delta_t V_2^o$ values increase with the concentration of cosolute. It may be noted that the $\Delta_t V_2^o$ values in general decrease with an increase in the size of nonpolar side chain of amino acids except in case of $MgCl_2$ where the reverse is true. From the overall comparison it appears that the interactions between the amino acids and the various electrolytes are highly specific both to the concentration and the nature of the cation of the cosolutes as anion is common in all cases. Generally the magnitude of $\Delta_t V_2^o$ values is higher in case of divalent cations in comparison to monovalent cations. Thus, the size and charge density of the cation, ionic strength, and nonpolar side chain of the amino acids have the strong bearing on the transfer volumes.

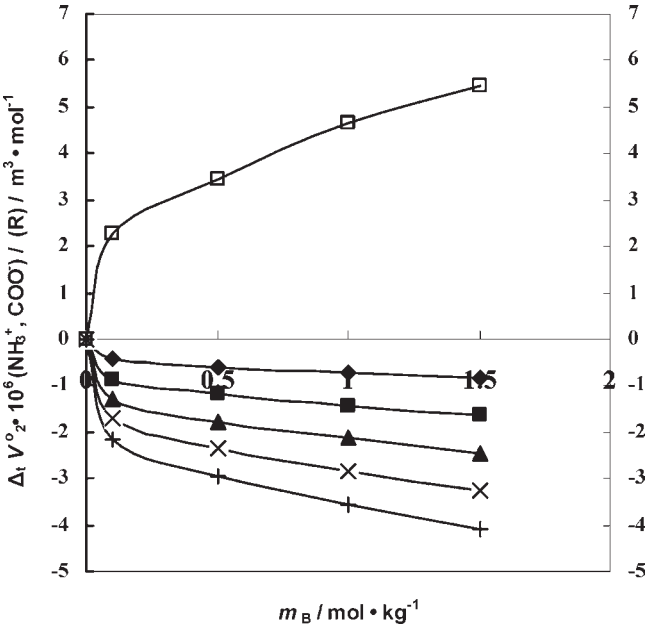

Figure 2. $\Delta_t V_2^o(NH_3^+, COO^-)/\Delta_t V_2^o(R)$ vs m_B, molalities of cadmium chloride: □, NH_3^+, COO^-; ♦, $-CH_2$; ■, $CHCH_3$; ▲, $-CHCH_2CH_3$; ×, $-CHCH(CH_3)_2$; +, $-CHCHCH_2(CH_3)_2$ at 298.15 K.

As the Cd^{2+} ions can form bonds with more than one amino acid molecules, thus it is very difficult to rationalize the volumetric behavior of the presently studied systems. However few comments can be made in terms of some gross interactions. According to the cosphere overlap model,[24] the ion-charged group interactions contribute positively, while ion−nonpolar group interactions contribute negatively to $\Delta_t V_2^o$ values. Therefore, the presently observed positive $\Delta_t V_2^o$ values in the presence of $CdCl_2$ on the whole indicate that the ion-charged group interactions dominate over the ion−nonpolar group interactions in these solutions.

The limiting partial molar volume V_2^o of solute can be represented according to the modified equation of Shahidi and Farrell[25] as follows:

$$V_2^o = V_{vw} + V_{void} - V_{shrinkage} \quad (4)$$

where V_{vw} is the van der Waals volume, V_{void} is the volume associated with void or empty space, and $V_{shrinkage}$ is the volume of shrinkage that arise from the electrostriction of the solvent. Assuming that the values of V_{vw} and V_{void} remain almost same in both water and aqueous $CdCl_2$ solutions, the positive $\Delta_t V_2^o$ values may be attributed to a decrease in the shrinkage volume in the presence of $CdCl_2$ solutions. This may be due to stronger interactions between amino acids and ions of the cosolute (Cd^{2+}, Cl^-), which increases the apparent molar volume of the studied amino acids in the presence of $CdCl_2$ and thus exhibits positive $\Delta_t V_2^o$ values.

A linear relationship has been observed between V_2^o of the amino acids and the number of carbon atoms, n_C, in their alkyl side chains, which is represented as:

$$V_2^o = V_2^o(NH_3^+, COO^-) + n_C V_2^o(CH_2) \quad (5)$$

where $V_2^o(NH_3^+, COO^-)$ and $V_2^o(CH_2)$ represent the zwitterionic end groups and methylene group contributions to V_2^o, respectively, and their values are listed in Table S3. The zwitterionic end groups $V_2^o(NH_3^+, COO^-)$ contribution for amino acids increases with the increase in the concentration of cadmium chloride (Table S3 of the Supporting Information). The values of $V_2^o(NH_3^+, COO^-)$ in the presence of $CdCl_2$ are more as compared to the value in water.[9] Similarly, the contributions of the (NH_3^+, COO^-) group and R groups (R = $-CH_2$, $-CHCH_3$, $-CHCH_2CH_3$, $-CHCH_2CH(CH_3)_2$) to $\Delta_t V_2^o$ values (calculated by an equation analogous to eq 3) of amino acids from water to aqueous $CdCl_2$ solutions have been calculated and depicted in Figure 2 (only representative plot at 298.15 K is given). The $\Delta_t V_2^o(NH_3^+, COO^-)$ values are positive and increase sharply initially, and thereafter almost linear behavior is observed with the molality of $CdCl_2$. The contributions of R groups to $\Delta_t V_2^o$ are negative and decrease slightly with the increase in concentration of $CdCl_2$ at all temperatures. Further, the negative contribution of R groups increases with the increase in the size of the nonpolar side chains of amino acids.

The number of water molecules, n_H, bound to the amino acids were calculated at (288.15, 298.15, and 308.15) K by using the method reported by Millero et al.[26,27]

$$n_H = V_{elect}/(V_e^o - V_b^o) \quad (6)$$

where V_e^o is the molar volume of electrostricted water and V_b^o is the molar volume of bulk water as already reported.[9] From the data given in Table 2, it may be seen that the n_H values for the studied amino acids are less in the presence of $CdCl_2$ as compared to their values in water,[9] and these values decrease with further increase in concentration of $CdCl_2$. The n_H values also decrease with the increase in temperature. These observations indicate that the $CdCl_2$ exert a dehydration effect on amino acids.

A comparison of present results with those reported in the presence of zinc chloride[9] shows that V_2^o values for the studied amino acids in $ZnCl_2$ come out to be large than in $CdCl_2$ and thus result in higher $\Delta_t V_2^o$ values in the case of $ZnCl_2$ as compared to $CdCl_2$. This may be rationalized in terms of the size of the metal ions. The Zn^{2+} ion having a smaller size (0.74 Å) as compared to the Cd^{2+} ion (0.95 Å) may interact strongly with amino acids. Further the hydration numbers for studied amino acids in aqueous solution of $CdCl_2$ are large than in the presence of $ZnCl_2$ which shows that $ZnCl_2$ exerts more dehydration effect than $CdCl_2$ in these systems.

Apparent Molar Adiabatic Compressibilities. The adiabatic compressibilities, K_S, were calculated from the measured sound velocities (Table S4 of the Supporting Information) and densities as follows:

$$K_S = 1/U^2\rho \quad (7)$$

where U is the sound velocity. The sound velocities of the studied amino acids in aqueous solutions of $CdCl_2$ increase with temperature as well as with the concentration. The apparent molar adiabatic compressibilities, $K_{S,2,\phi}$, for amino acids in water and in aqueous solutions of $CdCl_2$ at (288.15, 298.15, 308.15, and 318.15) K (Table S4 of the Supporting Information) were calculated using the following relation:

$$K_{S,2,\phi} = (K_S M/\rho) - [(K_S^o \cdot \rho - K_S \cdot \rho_o)/(m_A \cdot \rho \cdot \rho_o)] \quad (8)$$

where K_S and K_S^o are the adiabatic compressibilities of solution and solvent, respectively. The values of uncertainty in the determination of $K_{S,2,\phi}$ come out to ($0.729 \cdot 10^{-15}$ and $0.474 \cdot 10^{-15}$) $m^3 \cdot mol^{-1} \cdot Pa^{-1}$ at lower and higher concentrations, respectively, for the studied amino acids. At infinite dilution, the apparent molar adiabatic compressibility becomes equal to the partial molar adiabatic compressibility, $K_{S,2}^o$ ($K_{S,2,\phi} = K_{S,2}^o$). The $K_{S,2}^o$ values have

Table 2. Hydration Numbers, n_H, of Studied Amino Acids in Aqueous $CdCl_2$ Solutions from T = (288.15 to 308.15) K

m_B	n_H					
mol·kg^{-1}	T/K = 288.15	298.15	308.15	288.15	298.15	308.15
		Glycine			DL-α-Alanine	
0.1	2.68 ± 0.01	2.05 ± 0.02	1.47 ± 0.02	3.75 ± 0.01	3.00 ± 0.02	2.21 ± 0.02
0.5	2.42 ± 0.03	1.74 ± 0.03	1.20 ± 0.03	3.38 ± 0.01	2.77 ± 0.01	2.05 ± 0.01
1.0	2.15 ± 0.03	1.52 ± 0.03	0.96 ± 0.03	3.11 ± 0.03	2.44 ± 0.03	1.73 ± 0.03
1.5	2.00 ± 0.03	1.31 ± 0.03	0.77 ± 0.03	2.97 ± 0.03	2.28 ± 0.03	1.62 ± 0.03
		L-Valine			L-Leucine	
0.1	4.01 ± 0.01	3.26 ± 0.02	2.43 ± 0.02	5.90 ± 0.01	4.92 ± 0.02	3.87 ± 0.02
0.5	3.81 ± 0.02	3.02 ± 0.02	2.22 ± 0.02	5.87 ± 0.02	4.87 ± 0.02	3.79 ± 0.02
1.0	3.54 ± 0.01	2.69 ± 0.03	1.89 ± 0.03	5.84 ± 0.02	4.84 ± 0.02	3.76 ± 0.02
1.5	3.27 ± 0.01	2.52 ± 0.03	1.81 ± 0.03	5.80 ± 0.01	4.79 ± 0.04	3.67 ± 0.04

Table 3. Partial Molar Adiabatic Compressibilities, $K^o_{S,2}$ of Some Amino Acids in Water and in Aqueous $CdCl_2$ Solutions from T = (288.15 to 318.15) K

	$K^o_{S,2}\cdot 10^{15}$ (m^3·mol^{-1}· Pa^{-1})				
amino acid	m_B/mol·kg^{-1} = 0.0	0.1	0.5	1.0	1.5
	T/K = 288.15				
glycine	−31.10[a] ± 0.04, −31.66[b], −31.30[c], −28.34[d]	−27.36 ± 0.03	−26.56 ± 0.03	−26.11 ± 0.02	−25.46 ± 0.05
DL-α-alanine	−30.20 ± 0.01, −31.01[b], −30.40[c], −25.96[d]	−25.56 ± 0.02	−25.17 ± 0.02	−24.23 ± 0.01	−23.81 ± 0.02
DL-α-amino-*n*- butyric acid	−29.40 ± 0.01, −34.40[c], −40.37[e]	−24.30 ± 0.02	−23.86 ± 0.04	−22.63 ± 0.04	−22.08 ± 0.01
L-valine	−39.00 ± 0.01, −38.90[b], −37.80[c]	−36.07 ± 0.01	−35.01 ± 0.01	−34.56 ± 0.03	−34.22 ± 0.02
L-leucine	−43.50 ± 0.04, −43.95[b], −43.00[c]	−42.02 ± 0.02	−40.25 ± 0.01	−39.32 ± 0.01	−38.90 ± 0.01
	T/K = 298.15				
glycine	−26.40 ± 0.03, −26.50[b], −26.60[c], −26.38[d], −27.00[f]	−22.50 ± 0.01	−21.65 ± 0.02	−20.90 ± 0.02	−20.34 ± 0.03
DL-α-alanine	−25.60 ± 0.01, −25.16[b], −25.10[c], −23.91[d], −25.03[f]	−20.59 ± 0.01	−19.99 ± 0.01	−19.19 ± 0.02	−18.65 ± 0.02
DL-α-amino-*n*- butyric acid	−25.00 ± 0.01, −27.10[b], −29.95[d], −25.55[g], −25.56[h]	−18.82 ± 0.01	−18.16 ± 0.02	−17.72 ± 0.02	−17.19 ± 0.02
L-valine	−29.78 ± 0.02, −29.82[b], −28.80[c], −30.62[f]	−26.20 ± 0.02	−25.44 ± 0.01	−24.43 ± 0.02	−24.04 ± 0.02
L-leucine	−31.90 ± 0.03, −31.59[b], 30.50[c], −31.78[f]	−29.16 ± 0.03	−28.25 ± 0.01	−27.52 ± 0.01	−26.91 ± 0.02
	T/K = 308.15				
glycine	−23.60 ± 0.01, −23.59[b], −23.15[d]	−18.92 ± 0.02	−18.51 ± 0.01	−17.92 ± 0.02	−17.17 ± 0.04
DL-α-alanine	−20.90 ± 0.01, −23.87[b], −20.25[d]	−14.88 ± 0.04	−14.11 ± 0.01	−13.91 ± 0.01	−13.42 ± 0.1
DL-α-amino-*n*- butyric acid	−19.40 ± 0.01, −19.34[e]	−12.86 ± 0.02	−12.27 ± 0.02	−11.18 ± 0.01	−10.86 ± 0.03
L-valine	−23.71 ± 0.02, −23.56[b]	−19.65 ± 0.01	−18.89 ± 0.03	−18.23 ± 0.02	−17.55 ± 0.02
L-leucine	−22.20 ± 0.01, −21.70[b]	−18.68 ± 0.01	−18.08 ± 0.01	−17.26 ± 0.01	−16.69 ± 0.03
	T/K = 318.15				
glycine	−21.80 ± 0.03, −21.56[b]	−16.55 ± 0.03	−15.95 ± 0.03	−15.72 ± 0.02	−14.74 ± 0.03
DL-α-alanine	−18.00 ± 0.02, −17.85[b]	−11.44 ± 0.02	−10.99 ± 0.01	−10.24 ± 0.03	−9.64 ± 0.1
DL-α-amino-*n*- butyric acid	−17.10 ± 0.02	−9.89 ± 0.01	−9.23 ± 0.03	−8.45 ± 0.03	−8.09 ± 0.01
L-valine	−18.03 ± 0.03, −18.23[b]	−13.62 ± 0.01	−12.73 ± 0.03	−12.12 ± 0.03	−11.25 ± 0.01
L-leucine	−15.30 ± 0.03, −16.35[b]	−11.44 ± 0.02	−11.04 ± 0.01	−10.19 ± 0.02	−9.34 ± 0.04

[a] Present work. [b] Ref 15. [c] Ref 28. [d] Ref 29. [e] Ref 30. [f] Ref 26. [g] Ref 31. [h] Ref 14.

been calculated using the following equation:

$$K_{S,2,\phi} = K^o_{S,2} + S_K m_A \qquad (9)$$

where S_K is the experimental slope. The $K^o_{S,2}$ values for studied amino acids at (288.15 to 318.15) K in pure water (Table 3) agree well with the literature data.[14,15,26,28−31] The $K^o_{S,2}$ values in water as well as in aqueous $CdCl_2$ solution (Table 3) increase at (288.15 and 298.15) K in the order: L-leucine < L-valine < glycine < DL-α-alanine < DL-α-amino-*n*-butyric acid, but at (308.15 and 318.15) K, the order is: L-valine < L-leucine < glycine < DL-α-alanine < DL-α-amino-*n*-butyric acid, which may be considered as a decreasing order of hydration for the studied amino acids. From this it is obvious that with the introduction of methyl group, that is, from

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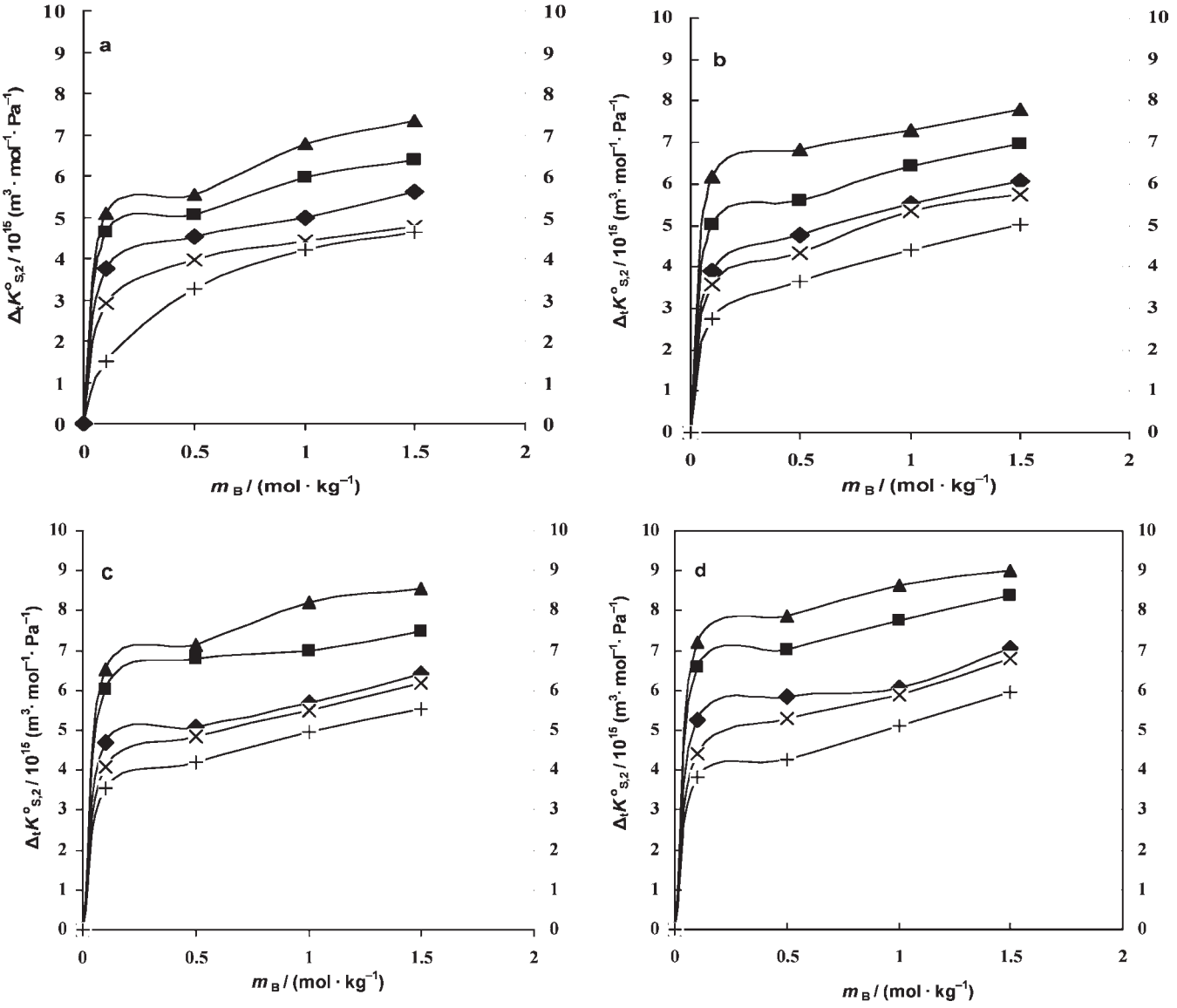

Figure 3. Partial molar adiabatic compressibility of transfer, $\Delta_t K^o_{S,2}$, for some amino acids vs m_B, molalities of cadmium chloride: ♦, glycine; ■, DL-α-alanine; ▲, DL-α-amino-*n*-butyric acid; ×, L-valine; +, L-leucine at (a) 288.15 K; (b) 298.15 K; (c) 308.15 K; (d) 318.15 K.

glycine to DL-α-alanine to DL-α-amino-*n*-butyric acid, the overlap of cospheres of hydrophilic (COO^- and NH_3^+) with that of the hydrophobic group (nonpolar side chain of amino acids) decrease their hydration ability in the above order. But with a further increase in the size of nonpolar side chain (as in case of L-valine and L-leucine) the effect of hydrophobic chain dominates which is responsible for large negative value of compressibilities in these cases. Further the reversal of the above order for L-valine and L-leucine at higher temperatures indicates the peculiar temperature effect in the case of amino acids having a large nonpolar side chain.

The $\Delta_t K^o_{S,2}$ values (calculated by an equation analogous to eq 3) for the studied systems in aqueous $CdCl_2$ solutions are illustrated in Figure 3. The $\Delta_t K^o_{S,2}$ values are positive and increase with the increase in the concentration of $CdCl_2$ as well as with temperature in all cases. The $\Delta_t K^o_{S,2}$ values increase in the order L-leucine < L-valine < glycine < DL-α-alanine < DL-α-amino-*n*-butyric acid at all concentrations of $CdCl_2$ as well as with temperature. The positive $\Delta_t K^o_{S,2}$ values may be attributed to the dominance of the ionic interactions occurring in these solutions. Because of these interactions, the less compressible water present in their hydration shells comes out into the bulk water (which is more compressible), thus exhibiting positive $\Delta_t K^o_{S,2}$ values. Therefore, the presently observed positive $\Delta_t K^o_{S,2}$ values for the studied amino acids throughout the concentration range of $CdCl_2$ also supports the conclusions drawn from the volumetric studies.

The hydration numbers, n_H, of amino acids were calculated at (288.15, 298.15, 308.15, and 318.15) K, by using the method reported by Millero et al.[26]

$$n_H = \frac{-K^o_{S,2}(\text{elect})}{K^o_S V^o_1} \tag{10}$$

where K^o_S is the compressibility of bulk water or bulk solvent, V^o_1 is the molar volume of bulk water or bulk solvent, and $K^o_{S,2}$(elect) is the electrostriction partial molar compressibility. The n_H values

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Table 4. Viscosity *B*-Coefficients and Temperature Coefficients, d*B*/d*T*, of Some Amino Acids in Water and in Aqueous $CdCl_2$ Solutions from *T* = (288.15 to 318.15) K

	$B \cdot 10^3$ ($m^3 \cdot mol^{-1}$)				
amino acids	m_B/mol·kg^{-1} = 0.0	0.1	0.5	1.0	1.5
		T/K = 288.15			
glycine	0.136[a] ± 0.002, 0.132[b], 0.129[c]	0.142 ± 0.002	0.145 ± 0.002	0.150 ± 0.003	0.154 ± 0.003
DL-α-alanine	0.268 ± 0.001, 0.269[b], 0.259[c]	0.279 ± 0.002	0.282 ± 0.002	0.288 ± 0.002	0.292 ± 0.002
DL-α-amino-*n*-butyric acid	0.358 ± 0.002, 0.379[b], 0.322[c]	0.373 ± 0.003	0.375 ± 0.004	0.383 ± 0.004	0.386 ± 0.004
L-valine	0.484 ± 0.002, 0.487[b]	0.503 ± 0.004	0.509 ± 0.003	0.513 ± 0.003	0.516 ± 0.003
L-leucine	0.544 ± 0.004, 0.540[b]	0.568 ± 0.003	0.573 ± 0.004	0.577 ± 0.004	0.587 ± 0.004
		T/K = 298.15			
glycine	0.142 ± 0.002, 0.146[b], 0.143[c], 0.1427[d]	0.150 ± 0.004	0.151 ± 0.002	0.155 ± 0.002	0.158 ± 0.002
DL-α-alanine	0.255 ± 0.007, 0.258[b], 0.253[c]	0.263 ± 0.003	0.267 ± 0.004	0.272 ± 0.007	0.276 ± 0.006
DL-α-amino-*n*-butyric acid	0.335 ± 0.004, 0.352[b]	0.348 ± 0.005	0.351 ± 0.005	0.353 ± 0.006	0.360 ± 0.005
L-valine	0.426 ± 0.006, 0.447[b]	0.442 ± 0.007	0.448 ± 0.008	0.451 ± 0.007	0.455 ± 0.006
L-leucine	0.532 ± 0.007, 0.487[b]	0.552 ± 0.003	0.554 ± 0.008	0.559 ± 0.006	0.566 ± 0.006
		T/K = 308.15			
glycine	0.146 ± 0.002, 0.146[b], 0.148[c]	0.151 ± 0.002	0.153 ± 0.002	0.156 ± 0.002	0.160 ± 0.004
DL-α-alanine	0.255 ± 0.002, 0.250[b], 0.247[c]	0.261 ± 0.002	0.264 ± 0.002	0.268 ± 0.001	0.271 ± 0.002
DL-α-amino-*n*-butyric acid	0.336 ± 0.001, 0.336[b], 0.322[e]	0.347 ± 0.004	0.350 ± 0.005	0.354 ± 0.002	0.358 ± 0.002
L-valine	0.420 ± 0.003, 0.418[b]	0.434 ± 0.002	0.439 ± 0.002	0.441 ± 0.002	0.444 ± 0.002
L-leucine	0.482 ± 0.003, 0.483[b]	0.499 ± 0.004	0.503 ± 0.003	0.507 ± 0.006	0.510 ± 0.003
		T/K = 318.15			
glycine	0.150 ± 0.003, 0.152[f]	0.154 ± 0.002	0.155 ± 0.002	0.157 ± 0.001	0.160 ± 0.003
DL-α-alanine	0.249 ± 0.002, 0.245[f]	0.254 ± 0.003	0.257 ± 0.001	0.259 ± 0.004	0.263 ± 0.002
DL-α-amino-*n*-butyric acid	0.305 ± 0.002, 0.307[f]	0.313 ± 0.003	0.315 ± 0.002	0.320 ± 0.003	0.324 ± 0.005
L-valine	0.398 ± 0.002	0.409 ± 0.003	0.413 ± 0.004	0.416 ± 0.006	0.417 ± 0.006
L-leucine	0.435 ± 0.003	0.449 ± 0.004	0.453 ± 0.005	0.454 ± 0.002	0.458 ± 0.003

	dB/d$T \cdot 10^3$ ($m^3 \cdot mol^{-1} \cdot K^{-1}$)				
amino acid	m_B = 0.0	0.1	0.5	1.0	1.5
glycine	0.0005	0.0003	0.0004	0.0002	0.0002
DL-α-alanine	−0.0006	−0.0008	−0.0008	−0.0009	−0.0009
DL-α-amino-*n*-butyric acid	−0.0002	−0.0018	−0.0018	−0.0019	−0.0019
L-valine	−0.0026	−0.0029	−0.0030	−0.0030	−0.0031
L-leucine	−0.0037	−0.0041	−0.0041	−0.0042	−0.0044

[a] Present work. [b] Ref 16. [c] Ref 33. [d] Ref 34. [e] Ref 35. [f] Ref 36.

calculated from eq 10 are given in the Table S5 (Supporting Information), and the calculated values for the studied amino acids agree well with literature values.[15,26] The n_H values are less in aqueous $CdCl_2$ solutions as compared to their values in water, and the values almost remain constant over the concentration range of $CdCl_2$ studied. However, the hydration numbers decrease with an increase in temperature.

Viscosity. The viscosities, η, of the solutions were determined from flow time, *t*, using the equation:

$$n/\rho = at - (b/t) \qquad (11)$$

where *a* and *b* are viscometer constants and ρ and *t* are the density and flow time of solution, respectively. The absolute viscosities of solvent (η_o) and solution (η) are given in Table S6 of the Supporting Information. For all of the amino acids studied, the viscosities increase regularly with an increase in the concentration of aqueous $CdCl_2$ solutions and decrease with temperature. The relative viscosities, η_r ($\eta_r = \eta/\eta_o$), were fitted by the method of least-squares analysis to obtain the viscosity *B*-coefficients using the Jones−Dole equation as follows:[32]

$$n_r = 1 + BC \qquad (12)$$

where *C* is the molarity ($mol \cdot L^{-1}$), which is calculated from the density data. The viscosity *B*-coefficients along with their standard deviations are summarized in Table 4. The values for viscosity *B*-coefficients for studied amino acids at (288.15 to 318.15) K in pure water agree well with the literature data.[16,33−36] It may be noted that, at any given temperature, viscosity *B*-coefficients increase in the following order (Table 4): glycine < DL-α-alanine < DL-α-amino-*n*-butyric acid < L-valine < L-leucine. Further viscosity *B*-coefficient values decrease with increase in the temperature for the studied amino acids except in case of

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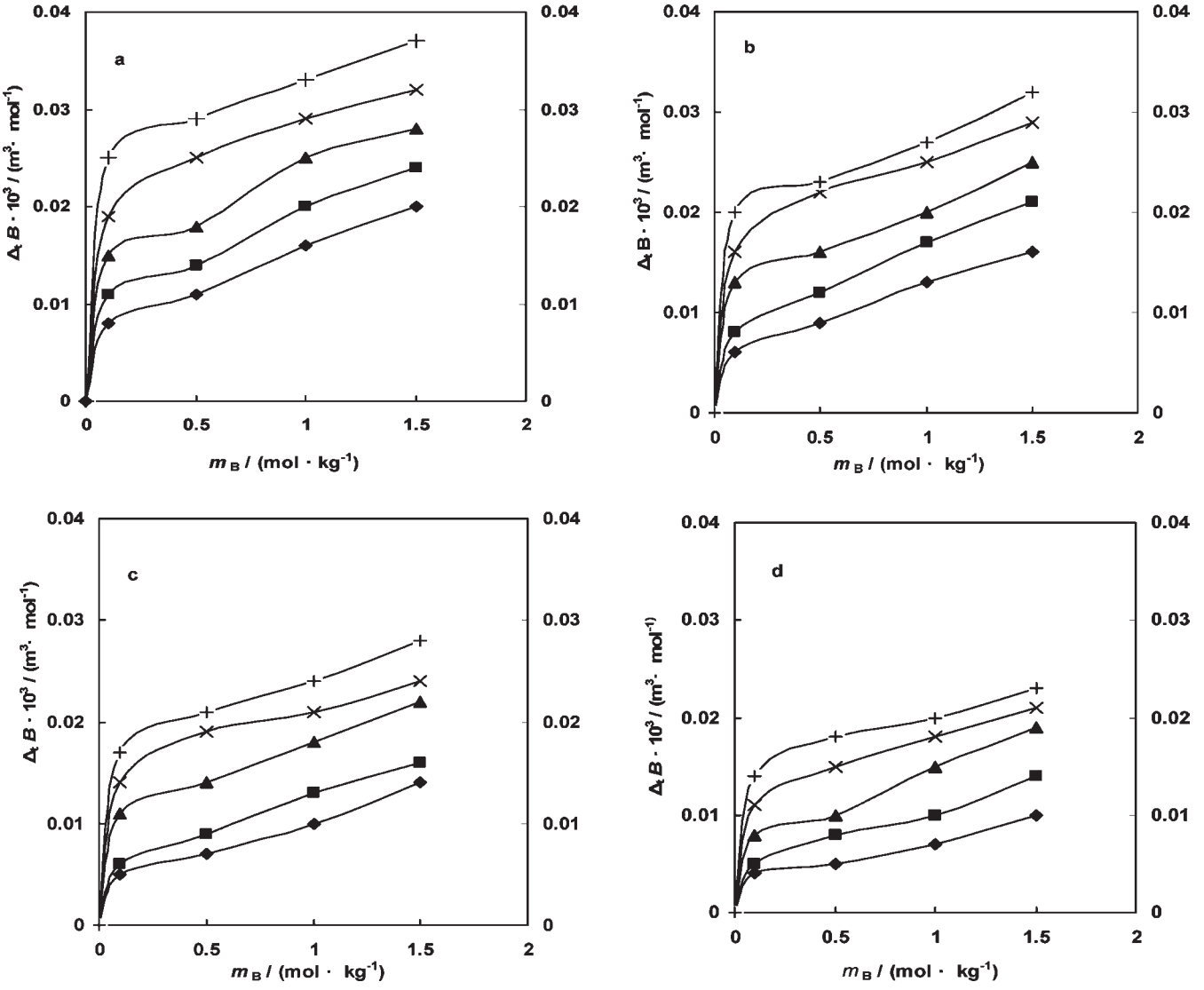

Figure 4. Viscosity *B*-coefficients of transfer, $\Delta_t B$, of some amino acids vs m_B, molalities of cadmium chloride: ♦, glycine; ■, DL-α-alanine; ▲, DL-α-amino butyric acid; ×, L-valine; +, L-leucine at (a) 288.15 K; (b) 298.15 K; (c) 308.15 K; (d) 318.15 K.

glycine where these values increase which may be due to more hydrophilic character in the case of glycine. The dB/dT is a better criterion for determining the solute effect on the structure of the solutions as compared with the *B*-coefficients.[24] The positive values of dB/dT indicate that the solute is structure breaker, whereas negative values indicate that solute is a structure maker. Therefore, the presently observed negative values of dB/dT for the studied amino acids in water and in aqueous solutions of $CdCl_2$ indicate that all studied amino acids except glycine act as a structure maker in water as well as in an aqueous $CdCl_2$ solution.

The viscosity *B*-coefficients of transfer, $\Delta_t B$, from water to aqueous $CdCl_2$ solutions have been calculated using an equation analogous to eq 3. The $\Delta_t B$ values for the studied amino acids in aqueous $CdCl_2$ solutions are positive. The magnitude of $\Delta_t B$ values decreases with the increase in temperature and increase with $CdCl_2$ concentration at all temperatures (Figure 4).

A comparison of present $\Delta_t B$ values (Table S2 of the Supporting Information) with the values reported in other electrolytes having common anion[19−21] shows that, at lower concentrations, that is, $m_B \approx 0.1$ mol·kg^{-1} the $\Delta_t B$ values for glycine and DL-α-alanine are greater in case of $CdCl_2$ than in case of $MgCl_2$, but at higher concentrations, that is, $m_B \approx$ (0.81 to 2.0) mol·kg^{-1}, the reverse trend has been observed. For L-leucine the $\Delta_t B$ values come out to be larger in case of $MgCl_2$ than $CdCl_2$ both at lower and higher concentrations. However, it may be noted that the $\Delta_t B$ values are negative in the case of $BaCl_2$ and KCl. These observations again indicate that the interactions in these systems are highly specific to both the concentration and the nature of the cation of the cosolute.

A linear relationship has been observed between the *B*-coefficients of the amino acids and the number of the carbon atoms, n_c, in their alkyl side chain by using an equation analogous to eq 5. The regression parameter, $B(NH_3^+,COO^-)$, the zwitterion end group contribution, and $B(CH_2)$, the methylene group contribution, to *B*-coefficients are listed in Table S7 of the Supporting Information and agree well with the literature data in water.[19,16] The $B(NH_3^+, COO^-)$ values in aqueous $CdCl_2$ solutions are large in comparison to their values in water. The $B(CH_2)$ methylene group values in water are slightly smaller than in aqueous $CdCl_2$ solutions, and these values remain almost

Table 5. Coefficients A_1 and A_2 for Some Amino Acids in Water and in Aqueous $CdCl_2$ Solutions from T = (288.15 K to 318.15) K

m_B / mol·kg^{-1}	$-A_1 \cdot 10^3$ / m^3·mol^{-1}	A_2	R^a	SD^b
	T/K = 288.15			
0.0	0.130 ± 0.03	6.5 ± 0.4	0.993	0.02
	0.128[c]			
0.1	0.151 ± 0.03	6.9 ± 0.4	0.988	0.01
0.5	0.113 ± 0.03	6.3 ± 0.4	0.972	0.03
1.0	0.163 ± 0.03	7.1 ± 0.4	0.995	0.02
1.5	0.169 ± 0.04	7.2 ± 0.4	0.997	0.02
	T/K = 298.15			
0.0	0.114 ± 0.01	6.0 ± 0.10	0.999	0.01
	0.079[c]			
0.1	0.138 ± 0.01	6.4 ± 0.10	0.999	0.02
0.5	0.144 ± 0.01	6.5 ± 0.10	0.999	0.03
1.0	0.149 ± 0.01	6.5 ± 0.10	0.999	0.02
1.5	0.155 ± 0.01	6.6 ± 0.10	0.999	0.01
	T/K = 308.15			
0.0	0.072 ± 0.02	5.2 ± 0.3	0.995	0.01
	0.055[c]			
0.1	0.096 ± 0.03	5.6 ± 0.3	0.996	0.01
0.5	0.102 ± 0.03	5.7 ± 0.3	0.996	0.02
1.0	0.106 ± 0.02	5.7 ± 2.8	0.997	0.02
1.5	0.108 ± 0.03	5.7 ± 0.3	0.997	0.01
	T/K = 318.15			
0.0	0.035 ± 0.03	4.5 ± 0.3	0.989	0.02
0.1	0.062 ± 0.03	5.6 ± 0.4	0.996	0.01
0.5	0.061 ± 0.05	4.8 ± 0.6	0.991	0.02
1.0	0.070 ± 0.04	4.9 ± 0.4	0.990	0.01
1.5	0.072 ± 0.04	4.9 ± 0.4	0.991	0.02

[a] Correlation coefficients. [b] Standard deviation of the fit. [c] Ref 16.

constant with the concentration of $CdCl_2$. It may also be noted that the $B(NH_3^+,COO^-)$ values increase with temperature and $B(CH_2)$ values decrease with temperature.

The viscosity B-coefficients for the studied amino acids in water and in aqueous $CdCl_2$ solutions also show a linear correlation with V_2^o as follows:

$$B = A_1 + A_2 V_2^o \quad (13)$$

The coefficients A_1 and A_2 obtained by least-squares analysis are given in Table 5. The A_2 values reflect the size and shape of solute.[16] It can be noted that as A_2 values are greater than 2.5; this indicates that amino acids are strongly solvated in aqueous $CdCl_2$ solutions.

CONCLUSIONS

The partial molar volume, V_2^o, partial molar adiabatic compressibility, $K^o_{S,2}$, viscosity B-coefficients, of glycine, DL-α-alanine, DL-α-amino-*n*-butyric acid, L-valine, and L-leucine in aqueous solution of $CdCl_2$ have been determined at (288.15 to 318.15) K. The various transfer values ($\Delta_t V^o_2$, $\Delta_t K^o_{S,2}$, and $\Delta_t B$) are positive in all cases and increase with the increases in the concentration of $CdCl_2$ as well as with temperature, except $\Delta_t B$ values which decrease with temperature. The variation of $K^o_{S,2}$ values from glycine to DL-α-amino-*n*-butyric acid, suggesting that the cosphere of hydrophilic (COO^- and NH_3^+) groups overlap destructively with that of a hydrophobic group (nonpolar side chain of amino acids) and thus decrease their hydration ability. However in the case of L-valine and L-leucine, the hydrophobic effect of nonpolar side chain starts dominating, and the $K^o_{S,2}$ values are more negative in these cases. Further, the negative value of dB/dT for the studied amino acids in water and in aqueous solutions of $CdCl_2$ indicate that the studied amino acids except glycine act as a structure maker in water as well as in aqueous $CdCl_2$ solution. It may be noted that these properties are model-dependent.

ASSOCIATED CONTENT

Supporting Information. Tables S1 to S7. This material is available free of charge via the Internet at http://pubs.acs.org.

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