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Excess Molar Volumes, Apparent Molar Volumes, Partial Molar Volumes of Methionine, An Amino Acid, in Water + Ethanol And Water + Methanol Solutions at 298.15 K

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Abstract

Methionine is an amino acid that is extremely important for human health. To better understand the biochemical events occurring in the human body, the excess molar properties of methionine and aqueous ethanol and aqueous methanol mixtures were determined at 298.15 K. Interactions between components in solutions are explained. Negative deviations from the ideal state have been observed in methionine solutions due to hydrogen bonds, dipole interactions, charge-transfer interactions.

Keywords: *Methionine; excess molar volume; excess partial molar volume; apparent molar volume*

1. Introduction

Methionine is an amino acid found in many proteins present in our bodies and food (Figure 1). It is not just a building block for proteins but contains some unique properties. The most important of these properties is its ability to be converted into sulfur-containing molecules [1]. Sulfur-containing molecules play an important role in protecting tissues, modifying DNA, and maintaining the proper functioning of cells [2,3,4[. Of the amino acids used to make protein in the body, only methionine and cysteine contain sulfur. Methionine also plays a critical role in initiating the process of making new proteins in cells. It occurs continuously as old proteins break down [5]. For example, it helps muscles produce new proteins after a muscle-damaging workout [6,7].

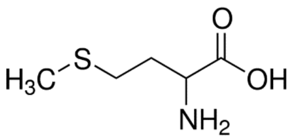

Figure 1. Methionine structure

Zhang et al. (2016) observed that the solubility of methionine in water is higher than in organic solvents, and the solubility decreases in water + acetone < water + ethanol < water + methanol in water + organic solvent mixtures [8]. According to the like dissolves like principle, they stated that the solubility of the amino acid, which has a polar structure, increases due to the increase in the polarity of organic solvents. Tyunina et al. (2020) reported that electrostatic interactions and the formation of hydrogen bonds were effective on the volumetric properties of methionine aqueous solutions [9]. El-Dossoki (2018) observed that the molal solubility of methionine decreases as the mole fraction of methanol and ethanol increases due to the salt effect [10]. He also determined that the apparent molar volume of DL-methionine in water and methanol-water and ethanol-water solutions increased as the mole fraction of methanol and ethanol increased. He pointed out the inverse proportion between volume and density as the reason for this.

The excess molecular properties of amino acid solutions are very useful for understanding the conformational stability and unfolding behaviour of protein molecules. Some researchers have used thermodynamic methods to investigate the interactions of amino acids with organic molecules or salts in aqueous solutions [11,12].

Excess properties can be defined as the difference between the ideal volumetric properties of the mixture and the observed volumetric properties as a result of the molecular interactions between the components that make up the mixture and in the solution [13]. The excess volumes (V^E) are defined as the difference of volume of real solutions and volume of pure components (ideal solution).

Excess thermodynamic properties, which can also be understood as deviations from ideal thermodynamic behaviours, provide a better understanding of the behaviour of components in chemical and biochemical events, and their roles in production and application processes. In general, the excess properties result from three types of interactions between the constituent molecules of liquid mixtures [14-15].

(a) Positive effects: Physical interactions consisting of dispersion forces or weak dipole-dipole interaction,
(b) negative effects: chemical or specific interactions, including charge transfer, H-bonds, and other complex formation interactions,

(c) Structural effects: These are the structural contributions arising from the differences in the size and shape of the component molecules in the mixture depending on the structure of their molecules.

In this study, excess molar volumes, apparent molar volumes, partial molar volumes and excess partial molar volumes of the binary mixture of methionine + methanol and methionine + ethanol in water at 298.15 K, which is one of the most important building blocks of proteins, were investigated.

2 Materials

Amino acids; methionine of 99.5% purity, methanol and ethanol of 99% purity were obtained from Merck Company. Some physicochemical properties of the components are given in Table 1.

Using twice-distilled water with a conductivity of 0.038 µS/cm, 10 cm^3 of alcohol solutions were prepared between 0-1 mole fractions by increasing the volume by 0.5 cm^3. To alcohol solutions, amino acid was added until saturation. The sample tubes were mixed with a magnetic stirrer overnight in a thermostatic water bath at 298.15±0.1 K by closing the mouth with a Teflon cap. Then, the solution was left to rest overnight and the supernatant was separated by filtration. The filtered solutions were kept in a thermostatic water bath at 298.15±0.1 K and their densities were measured by Anton Paar DMA 4500 Denismeter. Experiments were performed in 4 repetitions.

Table 1 Molecular weight and densities of pure components in mixtures

Component	M, g/mol	Density, 298.15 K
Methionine	149.21	Powder
Methanol	32.04	0.7048
Ethanol	46.07	0.7875
Water	18.02	0.9968

3 Methodology

3.1 Excess Molar Properties

The excess molar volume of a liquid solution is defined by the following equation [16-17].

$$V^E = V - \sum_{i=0} x_i V_i^0 \tag{1}$$

where V is the molar volume of the solution, xi is the mole fractions of components, and V_i^0 is the molar volumes of pure components, respectively. Eq. (1) can be written in terms of density as Eq. (2). The excess molar volumes, calculated from the density data in Eq. (4), are listed in Table 3 and plotted in Figure 3.

$$V^E = \sum_{i=1} X_i M_i \left(\frac{1}{\rho} - \frac{1}{\rho_i}\right) \tag{2}$$

in which M_i is the molar masses of components, ρ is the density of the mixture, and ρ_i represent the densities of pure components, respectively.

The excess molar properties (V^E) can be correlated using the Redlich–Kister equation [16-20]:

$$V^E = X_i(1 - X_i) \sum_{i=1} A_i (2X_i - 1)^i \tag{3}$$

The values of the coefficients A_i were calculated by the method of least squares along with the standard deviation $\sigma(V^E)$. The coefficients A_i are adjustable parameters for a better fit of the excess functions.

$$\sigma(V^E) = \left[\frac{\sum_{i=1}^{i=n}\left(V_{cal,i}^E - V_{exp,i}^E\right)}{n}\right]^{1/2} \tag{4}$$

where n is the number of parameters, V_{expt} and V_{cal} are the experimental and calculated parameters, respectively.

3.2 Partial Volume and Excess Partial Volume

To have more knowledge about interactions solute and solvent, the values of partial molar volumes of binary mixtures have been calculated using the following equations [18-20]:

$$V_i = \left(\frac{\partial V}{\partial n_i}\right)_{P,T,n_j} \tag{5}$$

Differentiation of Eq. (3) and combination with Eq. (5), gives the following equations for the partial molar volume of pure components

$$V_1 = V^E + V_1^0 + (1 - X_1)(\partial V^E/\partial X_1)_{T,P} \tag{6}$$

$$V_2 = V^E + V_2^0 - X_1(\partial V^E/\partial X_1)_{T,P} \tag{7}$$

where V_1^0 and V_2^0 are the molar volume of the pure component. Combination of Eqs. (3), (6) and (7) leads to Eqs. (8) and (9).

$$V_1 = V_1^0 + (1 - x)^2 \sum_{i=0} A_i(1 - 2x)^i - 2x(1 - x)^2 \sum_{i=1} A_i i(1 - 2x)^{i-1} \tag{8}$$

$$V_2 = V_2^0 + x^2 \sum_{i=0} A_i(1 - 2x)^i + 2x^2(1 - x) \sum_{i=1} A_i i(1 - 2x)^{i-1} \tag{9}$$

Values of the partial molar volumes at infinite dilution, V_i^∞, were obtained by the linear extrapolation of corresponding partial molar volumes using Eq. (8) and Eq. (9). Extrapolation of V_i to x_i = 0 results in V_i^∞. The excess partial molar volumes at infinite dilution, V_i^E, were calculated using the following relations [18-20]:

$$V_i^E = V_i - V_i^0 \tag{10}$$

The partial properties at infinite dilution are of interest since, at the limit of infinite dilution, the (solute + solute) interactions disappear. The values of the partial molar volume at infinite dilution provide information about (solute + solvent) interaction, independent of the composition effect. Setting (x = 0) in Eq. (8) and (x = 1) in Eq. (9) leads to Eq. (11) and Eq. (12). Eq. (11) and (12) give V_1^∞ and V_2^∞ (the partial molar volumes at infinite dilution) for the component (1) and (2), respectively [18-20].

$$V_1^\infty = V_1^0 + \sum_{i=0} A_i(-1)^i \tag{11}$$

$$V_2^\infty = V_2^0 + \sum_{i=0} A_i \tag{12}$$

Rearrangement of Eqs. (11) and (12) leads to Eqs. (13) and (14) for the excess partial molar volumes of components at infinite dilution,

$$V_1^{E,\infty} = \sum_{i=0} A_i(-1)^i \quad (13)$$

$$V_2^{E,\infty} = \sum_{i=0} A_i \quad (14)$$

Eq. (13) and Eq. (14) represent the partial molar volumes of components in solutions at infinite dilution, respectively.

3.3 Apparent Molar Volume

The apparent molar volume (V_φ) is a very useful parameter to understand the interactions between ion-solvent, ion-ion and solvent-solvent (structural) molecules that occur in mixtures. The Redlich–Kister equation and its derivatives do not always provide the best representation of properties of either component at infinite dilution in the other component. Instead of using the Redlich–Kister equation, we have also considered another approach, which may be more convenient and accurate, by calculating the partial molar volume at infinite dilution through apparent molar volumes. Apparent molar volumes of components, $V\phi$, were calculated from experimental data using the following relations [18-20].

$$V_{\emptyset 1} = \frac{(V - x_2 V_2^0)}{x_1} = \frac{M_1}{\rho} + \frac{(\rho_2 - \rho) x_2 M_2}{x_1 \rho \rho_2} \quad (15)$$

$$V_{\emptyset 2} = \frac{(V - x_1 V_1^0)}{1 - x_1} = \frac{M_2}{\rho} + \frac{(\rho_1 - \rho) x_1 M_1}{x_2 \rho \rho_1} \quad (16)$$

where M_i, m, ρ_i and ρ_i^0 are molecular weight, molality, densities of components and pure components, respectively. Extrapolation of $V_{\emptyset i}$ to $x_i = 0$ give the values of the limiting apparent molar volume ($V_{\emptyset,i}^0$). The excess apparent molar volumes at infinite dilution, $V_{\emptyset,i}^E$, were also calculated by equations similar to Eqs. (10). The apparent molar volumes of $V_{\phi 1}$ and $V_{\phi 2}$ can be calculated from Eq. (17) and Eq. (18).

$$V_{\emptyset 1} = \frac{V - (1-x) V_2^0}{x} \quad (17)$$

$$V_{\emptyset 2} = \frac{V - x V_1^0}{1-x} \quad (18)$$

where V_1^0 and V_2^0 are the molar volumes determined from the experimental densities by M_i/ρ_i, and V is the molar volume of the solution determined by the following equation:

$$V = V^E + (x_1 V_1^0 + x_2 V_2^0) \quad (19)$$

The combination of Eq. (1), Eq. (17), and Eq. (18) lead to:

$$V_{\emptyset 1} = V_1^0 - \frac{V^E}{x} \quad (20)$$

$$V_{\emptyset 2} = V_2^0 - \frac{V^E}{1-x} \quad (21)$$

Simple graphical or analytical extrapolation of $V_{\phi 1}$ to $x = 0$ and $V_{\phi 2}$ to $x = 1$ leads to the partial molar volumes at infinite dilution, V_1^∞ and V_2^∞, respectively. Further, the limiting partial molar volume is expressed as $V_i^\infty = V_i^{E,\infty} + V_i^0$ from which the $V_i^{E,\infty}$ can be derived. It can be observed that the procedures of obtaining $V_i^{E,\infty}$ values by Eq. (13) and (14) or from the extrapolated $V_{\phi 1}$ values of Eq. (20) and Eq. (21) lead to comparable magnitudes of V_i^∞ values.

Partial molar volumes were calculated at infinite dilution from excess molar volumes using a method based on extrapolation of reduced volume. This method was obtained by rearrangement of Eq. (20) and division by *(1-x)*.

$$\frac{V^E}{x(1-x)} = \frac{V_{\emptyset i} - V_i^0}{1-x} \quad (22)$$

Linear extrapolation of the ‘‘reduced volume” represented by $V^E/x(1-x)$ to $x = 0$ and $x = 1$ leads to the desired V_1^∞ and V_2^∞, respectively. Thus, the methods of obtaining V_1^∞ and V_2^∞ by way of Eqs. (14) and (15), extrapolation of $V_{\phi i}$ to $x = 0$ or $V^E/x(1-x)$ to $x = 0$ are all satisfactory, giving equally approximately equal values of partial molar volumes at infinite dilution.

4. Result and Discussion

In this study, the densities, apparent molar volumes, partial molar volumes and excess molar properties of saturated solutions of methionine in aqueous solutions of different concentrations of methanol and ethanol at a constant temperature of 298.15 K were investigated. The densities of aqueous mixtures of amino acid over the entire range of compositions at 298.15 are shown in Table 2 and Figure 2.

Table 2. Densities and Excess Molar Volumes of Methionine + Ethanol + Water) and (Methanol + Water) Solutions at 298.15 K.

	Densities, (g/cm^3)		V^E, (cm^3/mol)	
x	Methanol + Water	Ethanol + Water	Methanol + Water	Ethanol + Water
0.00	0.9964	0.9983	0.0000	0.0000
0.05	1.0079	0.9951	-0.6118	-0.6359
0.10	1.0141	0.9901	-1.1343	-1.2238
0.15	1.0143	0.9837	-1.5474	-1.7660
0.20	1.0085	0.9765	-1.8410	-2.2658
0.25	0.9973	0.9687	-2.0135	-2.7269
0.30	0.9818	0.9606	-2.0715	-3.1528
0.35	0.9635	0.9524	-2.0306	-3.5466
0.40	0.9438	0.9442	-1.9161	-3.9098
0.45	0.9245	0.9359	-1.7630	-4.2420
0.50	0.9070	0.9276	-1.6136	-4.5397
0.55	0.8925	0.9190	-1.5135	-4.7954
0.60	0.8819	0.9100	-1.5035	-4.9969
0.65	0.8755	0.9002	-1.6096	-5.1255
0.70	0.8728	0.8892	-1.8329	-5.1549
0.75	0.8726	0.8766	-2.1397	-5.0486
0.80	0.8727	0.8617	-2.4549	-4.7577
0.85	0.8696	0.8440	-2.6535	-4.2159
0.90	0.8587	0.8227	-2.5449	-3.3335
0.95	0.8340	0.7970	-1.8311	-1.9866
1.00	0.7877	0.7661	0.0000	0.0000

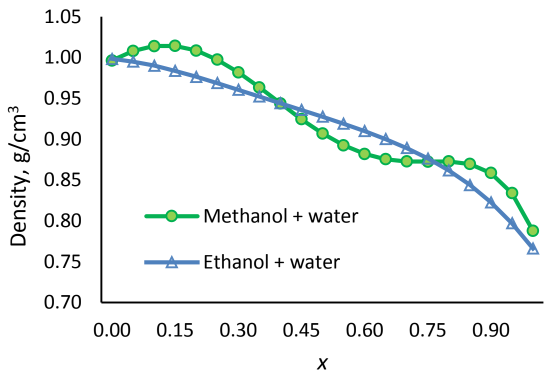

Figure 2. Densities of solutions of methionine + (ethanol +water) and (methanol + water) at 298.15 K.

As can be seen from Table 2, the solubility of amino acids decreases as the alcohol concentration decreases. Therefore, densities of binary mixtures of methionine and alcohol decrease as methanol and ethanol mole fraction increase. The density of the ethanol solution decreases slightly more than the methanol solution. However, the densities of the binary mixtures were found to be close to each other due to the strong interaction between functional groups in the side chain of the polar methionine and both alcohols.

The excess molar properties of methionine in aqueous alcohol solutions deviate from the ideal state. Chemical, physical, structural interactions between solvent-solute and components are the cause of this deviation. As can be seen in Table 2 and Figure 3, the excess molar volume of the binary mixture of methionine with ethanol and methanol showed a negative deviation. Negative deviation in excess molar property is in the direction of increasing alcohol fraction. The negative deviation of excess molar volume in ethanol solution is greater and more regular because ethanol is more polar than methanol and forms stronger H-bonds. The excess molar volume in methanol solutions shows irregularly negative deviations. The tendency of methionine to dissolve in the more polar water is deactivated as the mole fraction of methanol increases. Negative deviations in the excess molar volume of a binary mixture of methionine and both alcohol solutions indicate a strong hydrogen bond and chemical effects between the solvent and solute. Thus, it was understood that chemical interactions including charge transfer, H-bonds are effective in binary mixtures of methionine and alcohol solutions.

Partial molar volumes of components for all compositions can be calculated by using the Redlich-Kister coefficients in Eq. (3) at 298.15 K, and the results are shown in Table 3. The parameters A_i in Eq. (3) are reported in Table 4 along with standard deviations, σ, calculated by using Eq. (4). The partial molar volumes at infinite dilution show a good agreement between the calculation method, Eq. (11) and Eq. (12), and the linear extrapolation method of Eq. (8) and (9). As can be seen in Table 3, partial molar volumes in ethanol + water and methanol + water mixtures were calculated as close to each other. From these values, it is understood that there are stronger hydrogen bonds and dipole interactions between the sulfur in the structure of the amino acid and water molecules.

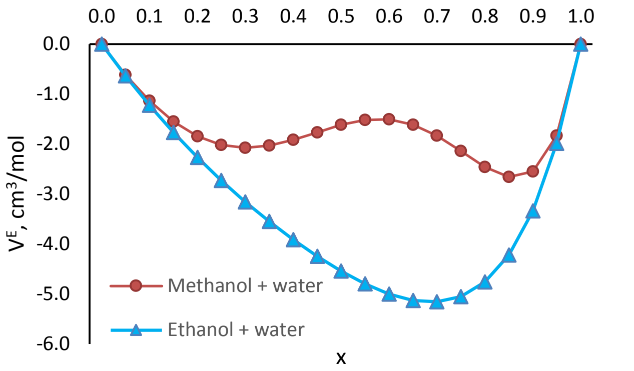

Figure 3. L-Excess molar volumes of methionine + (ethanol+water) and methionine + (methanol + water) solutions at 298.15 K.

From Eq. (13) and Eq. (14), excess apparent molar volumes values of methanol + water (1) and ethanol + water (2) solutions of methionine in infinitely dilute solutions were found V_1^{∞}=149.73 cm^3/mol and V_2^{∞}=149.50 cm^3/mol at 298.15 K, respectively. Similarly, it was found to be V_1^{∞}=144.74 cm^3/mol and V_2^{∞}=149.50 cm^3/mol from the linear extrapolation of Eq. (20) and Eq. (21). However, it can be said that the value found by interpolation is more reliable.

The change in volumetric properties of a binary mixture of methionine + alcohol can be explained by chemical interactions that occur in the form of charge-transfer interactions, hydrogen bonds and dipole-dipole interactions. Charge transfer bonds are formed between the electron donor amine group and the electron acceptor sulfur and carboxyl groups in the methionine structure. Strong hydrogen bonds and dipole-dipole interactions occur between the N, S atoms in the side chain molecules of methionine and the -OH group of the water molecule. Dipole-dipole interactions occur due to the dipole moment difference. The dipole moments of water, ethanol and methanol molecules are 1.85 D, 1.67 D and 1.69 D, respectively [21,22].

Table 3. Apparent Molar Volumes and Partial Molar Volumes of Methionine + (Ethanol + Water) and (Methanol + Water) Solutions at 298.15 K.

	Apparent molar volumes, V_ϕ, (cm^3/mol)				Partial Molar Volume, V_i (cm^3/mol)		Excess partial Molar Volume, V_i^E (cm^3/mol)	
x	Methanol + Water[a]	Ethanol + Water[b]	Methanol + Water[c]	Ethanol + Water[d]	Methanol + Water	Ethanol + Water	Methanol + Water	Ethanol + Water
0.00	147.97	147.43	149.75	149.46	149.73	149.46	-0.01	0.00
0.05	146.18	147.93	115.58	147.87	149.74	149.47	1.70	-0.47
0.10	145.23	148.72	123.59	146.40	149.74	149.47	2.61	-1.23
0.15	145.20	149.74	132.12	145.01	149.75	149.47	2.64	-2.20
0.20	146.09	150.91	140.77	143.64	149.75	149.47	1.80	-3.33
0.25	147.82	152.20	149.20	142.23	149.75	149.46	0.14	-4.56
0.30	150.29	153.55	157.16	140.71	149.75	149.46	-2.22	-5.86
0.35	153.31	154.95	164.37	139.02	149.75	149.46	-5.12	-7.20
0.40	156.68	156.38	170.60	137.10	149.75	149.46	-8.34	-8.57
0.45	160.14	157.84	175.63	134.88	149.75	149.46	-11.65	-9.96
0.50	163.40	159.33	179.28	132.29	149.75	149.46	-14.77	-11.39
0.55	166.18	160.90	181.44	129.27	149.75	149.46	-17.43	-12.89
0.60	168.28	162.58	182.15	125.78	149.75	149.46	-19.44	-14.50
0.65	169.57	164.45	181.56	121.80	149.75	149.46	-20.68	-16.29
0.70	170.12	166.59	180.04	117.35	149.75	149.46	-21.20	-18.34
0.75	170.16	169.12	178.07	112.62	149.75	149.46	-21.24	-20.76
0.80	170.15	172.19	176.29	108.09	149.75	149.46	-21.23	-23.69
0.85	170.78	175.98	175.44	105.20	149.75	149.46	-21.84	-27.33
0.90	173.06	180.76	176.43	108.82	149.75	149.47	-24.01	-31.90
0.95	178.45	186.87	180.45	145.04	149.75	149.46	-29.17	-37.75
1.00	189.42	194.77	189.42	194.77	149.75	149.53	-39.68	-45.24

[a] (Eq. 15), [b] (Eq. 16), [c] (Eq. 20), [d] (Eq. 21)

Tablo 4. The Values of Redlich-Kister Coefficients for The Studied Binary Mixtures of at 298.15 K and at Under The Atmospheric Pressure

Mixture	Metanol + Water	Ethanol + Water
A_0	0,5908	0,1995
A_1	-0,5940	-0,0924
A_2	-0,0172	-0,2116
A_3	0,0064	0,0415
A_4	-0,0005	-0,0027
A_5	0,0000	0,0001
σ	7,7400	15,07

5. Conclusion

In this study, the intermolecular interactions of methionine in aqueous alcohol solutions at 298.15 K were tried to be understood from the density, apparent molar volume, partial molar volume and excess molar properties. Since methionine, which has a strong polar structure, has strong functional groups such as sulfur, carboxyl, amine, and strong interactions between water molecules, the effect of increasing alcohol concentration on these interactions was weak. As the ethanol concentration increases, the volumetric changes are slightly more pronounced compared to methanol. Hydrogen bonds, dipole interactions and charge transfer complexes are stronger than structural interactions in the molecular behaviour of amino acids in solutions.

The volumetric properties of amino acids in water and alcohol are extremely important for understanding their interactions with the structures surrounding the proteins. Understanding the solvent-soluble, solute-soluble interactions of amino acids, which are the building blocks of proteins, is important in terms of understanding the behaviour of amino acids in biochemical reactions. As it can be understood from the volumetric properties of methionine, depending on the chemical structure of biological systems, interactions deviating from the ideal state occur with the amino acid due to physical and chemical effects such as hydrogen bonds, dipole interactions, charge-transfer interactions. These interactions, which occur due to these chemical and physical effects, can trigger biological events depending on the environment. For this reason, volumetric properties will give an idea for a better understanding of the behavior of compounds such as methionine, which have an important role in biological events, in solutions.

Nomenclature

A_i *Redlich–Kister equation Coefficients*
M_i *Molar masses of components,*
ρ_i *Densities of pure components*
ρ *Density of the mixture,*
$\sigma(V^E)$ *Standart deviation*
V *Molar volume of the solution*
V_ϕ *Apparent molar volume*
$V_{\emptyset,i}^E$ *Excess apparent molar volumes at infinite dilution,*
V_i^0 *Molar volumes of pure components*
V_i^E *Excess partial molar volume,*
V_i^∞ *Partial molar volumes at infinite dilution*
V_i^0 *Molar volume of the pure component*
V_{exp}^E *Experimental excess molar volume*
V_{cal}^E *Calculated excess molar volume*
x_i *Mole fractions of components*
x *Mole fractions of the mixture*

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