

Apparent molar volume and modeling of volumetric properties of ionic liquid aqueous solutions 1-butyl-3-methylimidazolium chloride [Bmim⁺][Cl⁻] at various temperatures

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Received: August 20th, de 2013. Received in revised form: May 5th, 2014. Accepted: May 26th, 2014

Abstract

Densities of the aqueous solutions of ionic liquid 1-butyl-3-methylimidazolium chloride [Bmim⁺][Cl⁻] were determined using a vibrating tube densitometer Anton Paar DMA 5000 at a temperature range between (283.15 - 218.15) K. The apparent molar volumes V_{ϕ} of aqueous chloride 1-butyl-3-methylimidazolium were calculated and adjusted to the Pitzer ion interaction model, obtaining the limiting apparent molar volumes V_{ϕ}^0 and Pitzer volumetric parameters $\beta_{MX}^{(0)V}$, $\beta_{MX}^{(1)V}$, C_{MX}^V at temperatures of 283.15, 288.15, 293.15, 298.15, 303.15, 313.15 and 318.15K, proving that this model adequately represents the experimental volumetric data below an ionic strength of 0.6018 mol / kg. Finally the limiting apparent molar expansibilities ϕ_E^0 were calculated from the limiting apparent molar volumes at various temperatures and the results are discussed in terms of the interactions occurring in solution.

Keywords: apparent molar volume, Pitzer parameters, density, limiting expansibilities, ionic liquid.

Volumen molar aparente y modelamiento de propiedades volumétricas de soluciones acuosas del líquido iónico cloruro de 1-butil-3-metilimidazolio [Bmim⁺][Cl⁻] a varias temperaturas

Resumen

Se determinaron las densidades de las soluciones acuosas del líquido iónico Cloruro 1-butil-3-metilimidazolio [Bmim⁺][Cl⁻] utilizando un densímetro de tubo vibrador Anton Paar DMA 5000 entre 283,15 y 218,15 K. Se calcularon y ajustaron los volúmenes molares aparentes V_{ϕ} del cloruro de 1-butil-3-metilimidazolio acuoso al modelo de interacción iónica de Pitzer y se obtuvieron los volúmenes molares aparentes límites V_{ϕ}^0 y los parámetros volumétricos de Pitzer $\beta_{MX}^{(0)V}$, $\beta_{MX}^{(1)V}$, C_{MX}^V a las temperaturas de (283.15, 288.15, 293.15, 298.15, 303.15, 313.15 y 318.15)K; comprobándose que este modelo representa adecuadamente los datos volumétricos experimentales por debajo de una fuerza iónica de 0,6018 mol/kg. Finalmente a partir del volumen molar aparente límite determinado a varias temperaturas, se calcularon las expansibilidades molares aparentes límites ϕ_E^0 y los resultados se discutieron en términos de las interacciones que ocurren en solución.

Palabras clave: Volumen molar aparente, parámetros de Pitzer, densidad, expansibilidades límites, líquido iónico.

1. Introduction

Although the scientific literature and patents reserve the term "Ionic Liquid" to designate the compounds that are exclusively composed of ions that are liquid at moderate temperatures (e.g. lower than 100°C) [1], some of them, as is

the case of 1-butyl-3-methylimidazolium chloride [Bmim⁺][Cl⁻], are solid at room temperature.

It is well known that the liquid mixtures of different compounds form solutions that do not usually behave ideally. The interpretation of no ideality is a very interesting topic, therefore, nowadays a large number of contributions have been

made, especially related to the modeling of volumetric properties.

However, there is a compelling reason for studying mixed ionic liquids, this is due to the fact that these individual compounds tend not to show, at the same time, a greater selectivity and extraction capacity compared to that of conventional organic solvents [2-7] in several processes such as: The recovery of solvents (acetone, ethanol, and butanol) from fermentation broth [2-4], the extraction of antibiotics [2-5], and the elimination of organic pollutants from aqueous waste streams [5].

It has been established that the volumetric behavior of solutions of electrolytes and non-electrolytes may provide useful information about the solute-solvent and solute-solute interactions. In this sense and because ionic liquids can be considered ionic compounds; since they exhibit a similar behavior to the inorganic salts in aqueous solutions, they dissociate partially or completely in water, and are exclusively formed by hydrophobic or hydrophilic ions depending on the structure of the cation and/or anion; their mixtures with other substances are attractive systems for such analysis. That is why expansibilities of solute, partial molar volumes and apparent molar volumes have shown to be very useful tools in the interpretation of molecular interactions occurring in solution [8].

On the other hand, it is well known that due to their structure and ionic interactions, ionic liquids and their mixtures exhibit unique properties. Strong ion-ion interactions present in ionic liquids lead to highly organized three-dimensional supramolecular polymer networks of cations and anions joined by hydrogen bonds and/or Coulomb interactions; where the force of ion-ion interaction depends on the structure of ionic liquid and can greatly affect the ability of the individual components (anions or cations) to interact with dissolved species [9].

However, despite its interest and importance, the information about the physico-chemical properties of mixtures of ionic liquids with molecular solvents is very limited, even though this information is extremely important for different technological processes, such as the design of solvents and other processes previously mentioned in the preceding paragraphs [10].

The present work is oriented towards obtaining a set of precise measurements of density and apparent molar volumes of [Bmim +] [Cl-] in an aqueous environment, the allowable range of solubility and the range of temperature (283.15-318.15) K every 5 degrees. At the same time we seek to predict and correlate these volumes with the model of the Pitzer ion interaction.

2. Experimental part

Reagents used were: 1-butyl-3-methylimidazolium chloride (fraction of mass > 0.99), purchased from the trading house Across Organic (99% purity) and doubly distilled and deionized water (conductivity less than 2 μS) in accordance with the recommendations of the literature [11, 12].

The solutions were prepared gravimetrically using a balance (OHAUS Explorer model) with a sensitivity of ± 1 × 10⁻⁴ g, in tightly closed bottles to prevent loss by evaporation of some of the components. The experimental uncertainty

estimate for the molalities was ± 0.0002. Experimental densities in the diluted region of aqueous solutions of [Bmim+][Cl-] were measured using a vibrating tube digital densimeter Anton Paar DMA 5000, in the range of temperature (283.15-318.15) K and at an atmospheric pressure of 0.10 MPa. The cell of the DMA was calibrated with dry air and ultra-pure water at atmospheric pressure. The samples were thermostated and controlled at ± 0.001 K. The uncertainty values of certain densities were ± 0.2 × 10⁻⁵ g/cm³.

Since the [Bmim+][Cl-] is a solid at the conditions of temperature and pressure in which the data were taken, it was impossible to obtain densities and viscosity information about this pure IL. Therefore densities of the binary mixtures [Bmim+][Cl-] + H₂O at 308.15 K were compared to the density data previously reported in literature [13], which showed to be similar, as shown in Fig. 1.

3. Results and discussion

The experimental values of densities, ρ (·cm⁻³ g), measured at different temperatures and molalities, (mol · kg⁻¹), are shown in Table 1.

The values obtained for the V_{ϕ} apparent molar volumes and molal concentrations (m) [Bmim+][Cl-] ranging from 283.15 to 318.15 K are shown in Table 2.

The apparent molar volume V_{ϕ} (cm³ · mol⁻¹) of a solute with molar mass, M_2 (g · mol⁻¹), is given by

$$V_{\phi} = \frac{M_2}{\rho} + \frac{1000(\rho_0 - \rho)}{\rho_0 \rho m_2} \quad (1)$$

Where ρ_0 is the density of pure water and m is the molality of [Bmim+][Cl-] in (mol · kg⁻¹)

The apparent molar volume of [Bmim +][Cl-] in an aqueous solution, can be expressed by the Pitzer ion interaction model, which is an extension of the theory limit of Debye-Hückel which applies to very low concentrations; according to this model, ions of the same charge tend to stay away from each other and therefore short range forces would have very few consequences. Contrary to ions of opposite charge, which approach as close as possible, being affected by short range forces. This model is described by the following equations. [14-17]

$$V_{\phi} = \bar{V}_B^0 + \nu |z_M z_X| \left(\frac{A_V}{2b} \right) \ln(1 + b\sqrt{I}) + (2 \nu_X \nu_M RT) [B_{MX}^1 + (\nu_M z_M) m^2 C_{MX}^1] \quad (2)$$

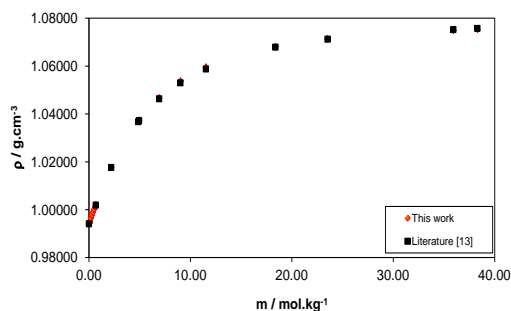


Figure 1. Comparison of experimental densities [Bmim+][Cl-] + H₂O in (g cm⁻³) with densities reported in literature [13] to 308.15 K
Fuente: Los autores

Table 1.

Values of the experimental densities in ρ ($g \cdot cm^{-3}$) solutions [Bmim⁺][Cl⁻] + H₂O from 283.15 to 318.15 K

T/K	283.15	288.15	293.15	298.15	303.15	308.15	313.15	318.15
m_{Li}								
	$\rho/g \cdot cm^{-3}$							
0.0099	0.99991	0.99931	0.99840	0.99723	0.99581	0.99401	0.99240	0.99030
0.0198	1.00005	0.99942	0.99851	0.99733	0.99590	0.99410	0.99249	0.99039
0.0305	1.00020	0.99955	0.99863	0.99744	0.99601	0.99420	0.99259	0.99048
0.0500	1.00047	0.99979	0.99885	0.99765	0.99621	0.99439	0.99278	0.99066
0.0795	1.00089	1.00015	0.99919	0.99798	0.99652	0.99469	0.99306	0.99094
0.1006	1.00119	1.00042	0.99944	0.99822	0.99675	0.99491	0.99328	0.99114
0.2005	1.00260	1.00174	1.00070	0.99944	0.99792	0.99606	0.99437	0.99221
0.3002	1.00403	1.00314	1.00204	1.00078	0.99920	0.99733	0.99557	0.99339
0.4065	1.00555	1.00463	1.00350	1.00218	1.00059	0.99867	0.99678	0.99462
0.5053	1.00695	1.00602	1.00488	1.00350	1.00187	0.99988	0.99789	0.99575
0.6018	1.00831	1.00738	1.00623	1.00482	1.00312	1.00106	0.99897	0.99683

Fuente: Los autores

Table2.

Values of the apparent molar volumes $V_{\phi} / cm^3 \cdot mol^{-1}$ solutions [Bmim⁺][Cl⁻] + H₂O between 283.15 to 318.15 K

T/K	283.15	288.15	293.15	298.15	303.15	308.15	313.15	318.15
$m / mol \cdot kg^{-1}$								
	$V_f / cm^3 \cdot mol^{-1}$							
0.0099	160.63	163.11	164.03	165.00	165.85	166.42	167.70	167.92
0.0198	160.67	163.00	163.97	164.88	165.71	166.35	167.37	167.75
0.0305	160.62	162.92	163.87	164.75	165.60	166.25	167.00	167.56
0.0500	160.55	162.77	163.68	164.56	165.35	166.03	166.51	167.27
0.0795	160.46	162.53	163.46	164.28	165.01	165.72	165.85	166.91
0.1006	160.38	162.35	163.32	164.10	164.79	165.50	165.60	166.69
0.2005	160.13	161.43	162.50	162.93	163.70	164.42	164.86	165.74
0.3002	159.82	160.99	161.84	162.43	163.11	163.68	164.45	165.09
0.4065	159.57	160.53	161.26	161.86	162.52	163.13	164.11	164.62
0.5053	159.36	160.17	160.81	161.45	162.12	162.82	163.88	164.32
0.6018	159.15	159.88	160.46	161.09	161.79	162.55	163.68	164.10

Fuente: Los autores

Where the Ionic interaction parameters, are given by

$$B_{MX}^I = \left(\frac{\partial B^{Ex}}{\partial P} \right)_{T,I} = \beta_{MX}^{(0)V} + \frac{2\beta_{MX}^{(1)V}}{\alpha^2 I} [1 - (1 + \alpha\sqrt{I})] \quad (3)$$

$$\beta_{MX}^{(0)V} = \left(\frac{\partial \beta_{MX}^{(0)V}}{\partial P} \right)_{T,I} \quad (4)$$

$$\beta_{MX}^{(1)V} = \left(\frac{\partial \beta_{MX}^{(1)V}}{\partial P} \right)_{T,I} \quad (5)$$

$$C_{MX}^V = (z_M z_X)^{-1/2} C_{MX}^{\phi V} \quad (6)$$

$$C_{MX}^{\phi V} = \left(\frac{\partial C_{MX}^{\phi V}}{\partial P} \right)_{T,I} \quad (7)$$

$$A_V = 2A_{\phi} RT \left[3 \frac{\partial \ln D}{\partial P} + \left(\frac{\partial \ln V_{\phi}^0}{\partial P} \right)_{T,I} \right] \quad (8)$$

The numerical values recommended for coefficients b and α are as follows: b=1.2 kg^{1/2} mol^{-1/2} for electrolytes of all types of charge; α 1=2.0 kg^{1/2} mol^{-1/2} type 1:1 1:2 and 2:1 electrolytes; α 1=1.4 kg^{1/2} mol^{-1/2} y α 2=12 kg^{1/2} mol^{-1/2} for type 2:2 electrolytes; the coefficient $\beta_{MX}^{(2)V}$ is assumed to be equal to zero for electrolytes of all charges, except type 2:2 electrolytes. The coefficients α 1, α 2 y b, are accepted as independent of temperature, which leads to equation 2 to be expressed in the following way for the [Bmim⁺][Cl⁻].

$$V_{\phi, MX} = \bar{V}_{MX}^0 + \left(\frac{A_V}{1.2} \right) \ln(1 + 1.2\sqrt{I}) + 2RTm \left\{ \beta_{MX}^{(0)V} + \beta_{MX}^{(1)V} \left(\frac{1}{2I} \right) [1 - (1 + 2\sqrt{I}) \exp(-2\sqrt{I})] + mC_{MX}^V \right\} \quad (9)$$

Rearranging the equation above, we obtain the right work equation for the system under study.

$$Y = \frac{1}{2RT} \left(V_{\phi, MX} + \left(\frac{A_V}{1.2} \right) \ln(1 + 1.2\sqrt{I}) \right) = \frac{\bar{V}_{MX}^0}{2RT} + m\beta_{MX}^{(0)V} + \frac{m\beta_{MX}^{(1)V}}{2I} [1 - (1 + 2\sqrt{I}) \exp(-2\sqrt{I})] + m^2 C_{MX}^V \quad (10)$$

Or by the following way

$$Y = \frac{1}{2RT} \left(V_{\phi, MX} + \left(\frac{A_V}{1.2} \right) \ln(1 + 1.2\sqrt{I}) \right) = a_0 + m\beta_{MX}^{(0)V} + m y^1 \beta_{MX}^{(1)V} + m^2 C_{MX}^V \quad (11)$$

Where Y is the function to correlate from experimental data, and it is given by:

$$y^1 = [1 - (1 + 2\sqrt{I}) \exp(-2\sqrt{I})] / 2I \quad (12)$$

and a_0 by

$$a_0 = \frac{\bar{V}_{MX}^0}{2RT} \quad (13)$$

Table 3.

Pitzer ion interaction parameters values a_0 , $\beta_{MX}^{(0)V}$, $\beta_{MX}^{(1)V}$, C_{MX}^V , and standard deviation, which are obtained by the equation 11 fixing from 283.15 to 318.15 K

T/K	a_0	$\beta_{MX}^{(0)V}$	$\beta_{MX}^{(1)V}$	C_{MX}^V	σ
	$bar.cm^3.mol^{-1}$	$kg.bar^{-1}.mol^{-1}$	$kg^2.bar^{-1}.mol^{-1}$	bar^{-1}	
283.15	3.41E-03	-3.81E-05	-1.48E-04	3.04E-06	3.76E-07
288.15	3.40E-03	-1.32E-04	-2.33E-04	1.12E-04	4.89E-07
293.15	3.36E-03	-1.26E-04	-2.28E-04	7.77E-05	1.58E-07
298.15	3.33E-03	-1.42E-04	-2.81E-04	1.22E-04	6.16E-07
303.15	3.29E-03	-1.92E-04	-2.14E-04	1.64E-04	5.37E-07
308.15	3.25E-03	-1.46E-04	-2.67E-04	1.20E-04	5.32E-07
313.15	3.21E-03	-1.69E-04	-3.54E-04	2.43E-04	2.35E-06
318.15	3.17E-03	-1.50E-04	-3.05E-04	1.66E-04	6.45E-07

Fuente: Los autores

Table 4.

Apparent residual molar volumes ΔV_ϕ at various temperatures.

T/K	283.15	288.15	293.15	298.15	303.15	308.15	313.15	318.15
$m / mol.kg^{-1}$	$\Delta V_\phi / cm^3.mol^{-1}$							
0.0099	0.05	0.04	0.03	0.05	0.08	0.04	0.48	0.15
0.0198	0.08	-0.01	0.01	0.00	0.02	0.04	0.26	0.07
0.0305	0.03	0.00	0.00	-0.02	0.02	0.04	0.03	-0.01
0.0500	-0.01	0.00	-0.04	-0.01	-0.01	0.02	-0.19	-0.08
0.0795	-0.03	0.01	-0.01	0.02	-0.04	0.02	-0.44	-0.10
0.1006	-0.06	0.02	0.03	0.06	-0.02	0.02	-0.43	-0.08
0.2005	-0.02	-0.14	-0.01	-0.19	-0.14	-0.14	-0.09	-0.05
0.3002	-0.06	0.03	-0.01	0.04	0.05	-0.12	0.22	0.04
0.4065	-0.03	0.08	0.01	0.07	0.08	-0.04	0.30	0.10
0.5053	0.01	0.05	0.01	0.06	0.05	0.06	0.15	0.07
0.6018	0.02	-0.05	0.00	-0.06	-0.07	0.06	-0.26	-0.08

Fuente: Los autores

According to this model, it is clear that the molar volume apparent pressure and constant temperature of each Ionic liquid in diluted solutions will depend only on the properties of the solvent and of the total concentration or the ionic strength of the solution.

Parameters, carry out a similar role to the first, second and third virial coefficients, that is why they characterize the forces of interaction of short range between the anion and cation of the ionic liquid. Both parameters of the molar volume of apparent limit shown in Table 3, were obtained by minimization of the function.

$$\sigma = \left(\sum_i^m \frac{(y_{exp,i} - y_{cal,i})^2}{n-p} \right)^{1/2} \quad (14)$$

Where n is the number of experimental points and p is the number of adjustable parameters. The correlated and predicted results from experimental data measured in this study are presented as the residual, $\Delta V_\phi = V_\phi^{exp} - V_\phi^{cal}$ in Table 4. An example of the behavior is illustrated in Fig. 2, the correlated and predicted results at temperature of 288.15 K. As previously observed, both results agree presenting a good precision.

$$V_\phi^0 = a + bT + cT^2 \quad (15)$$

Where T is the temperature in Kelvin degrees, and where a, b, and c are empirical constants dependent on the solute and solvent.

The partial molar expansibilities limits are obtained by differentiation of the above equation with respect to temperature

$$\phi_E^0 = \left(\frac{\partial V_\phi^0}{\partial T} \right)_p = bT + 2cT \quad (16)$$

In Table 5, the values of limiting apparent molar volumes V_ϕ^0 obtained by the methods of Redlich Meyer and Pitzer were reported:

Where the first ones were obtained by correlating the defined term using the relation $V_\phi - S_V \sqrt{m_2}$ in function of m_2 , which is in accordance with the equation of Redlich Mayer.

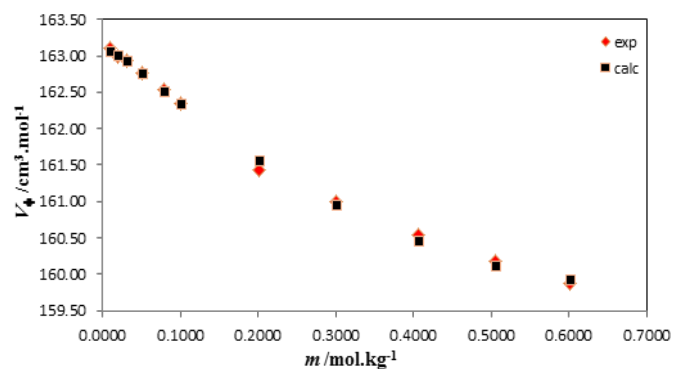


Figure 2. Apparent molar volumes of [Bmim⁺][Cl⁻] experimental and calculated using the Pitzer ion interaction model, at 288.15 K.

Fuente: Los autores

Table 5.

The molar volumes apparent limits and molar expansibilities apparent limit values ϕ_E^0 at various temperatures

T/K	$V_\phi^0 / \text{cm}^3.\text{mol}^{-1}$		$\phi_E^0 / \text{cm}^3.\text{mol}^{-1}.\text{K}^{-1}$
	Redlich Mayer	Pitzer	
283.15	160.44	160.43	0.3021
288.15	162.88	162.99	0.2621
293.15	163.91	163.92	0.2221
298.15	164.89	164.89	0.1821
303.15	165.71	165.69	0.1421
308.15	166.33	166.30	0.1021
313.15	167.70	167.18	0.0621
318.15	167.81	167.68	0.0221

Fuente: Los autores

$$V_\phi - S_V \sqrt{m_2} = V_\phi^0 - b_V m_2 \quad (17)$$

Here b_V is an empirical parameter characteristic of ionic liquid and m_2 is the molality of the ionic liquid and S_V is the theoretical slope limit of Debye Hückel, which depending on the temperature t ($^\circ\text{C}$) is given by the expression [18].

$$S_V = 1.4447 + 1.6799 \times 10^{-2}t - 8.4055 \times 10^{-6}t^2 + 5.5153 \times 10^{-7}t^3 \quad (18)$$

The reason for using the equation of Redlich-Mayer is essentially the same one that the Pitzer ion interaction model used, i.e., it is applied to analyze volumetric data of diluted electrolyte solutions.

While the second ones are obtained as a result of the correlation of the Pitzer ion interaction model, which are implicitly contained in the setting parameter a_0 , previously determined by this model.

The analysis of the results displayed in Table 5 shows that both results are very similar; the reason for this is possibly due to the fact that the S_V term of the Redlich Mayer equation depends on a variable of the Debye Hückel limiting law, which is characteristic of aqueous solvents and is also contained in the Pitzer ion interaction model, this makes both methods to converge at the same limit.

It can be verified that the values of the apparent ionic liquid ϕ_E^0 limiting expansibility coefficient $[[\text{Bmim}^+][\text{Cl}^-]]$ according to Table 5, decrease linearly with the increase of temperature with a slope equal to,

$$\frac{\partial \phi_E^0}{\partial T} = -0.008 \quad (19)$$

The fact that ϕ_E^0 linearly decreases with a temperature increase means that the second derivative of V_ϕ^0 with respect to temperature is also negative. In concordance with Hepler [18], it is evident that the studied ionic liquid behaves as a substance capable of disrupting the three-dimensional structure of water in the studied temperature range.

4. Conclusions

Densities of the binary system $[\text{Bmim}^+][\text{Cl}^-] + \text{H}_2\text{O}$ were determined using a vibrating tube densitometer Anton Paar

DMA 5000 at temperatures of K (283.15, 288.15, 293.15, 298.15, 303.15, 313.15, and 318.15). The experimental data were correlated and predicted through the use of the Pitzer ion interaction model. A good correlation can be observed between the predicted values and those measured experimentally in working conditions. Small standard deviations, σ , show that the Pitzer ion interaction model is appropriate for the representation of the volumetric properties of aqueous solutions of ionic liquids.

Acknowledgments

The authors thank the Universidad de Córdoba for the support provided for the realization of this work.

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