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Henry's constant of carbon dioxide-aqueous deep eutectic solvent (choline chloride/ethylene glycol, choline chloride/glycerol, choline chloride/malonic acid) systems

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ABSTRACT

In this study, we present a new set of Henry's constant data for the system carbon dioxide-aqueous deep eutectic solvent (DES) (20 to 80 wt% DES) at $T = (303.15, 308.15, \text{and } 313.15)$ K. The DESs used were choline chloride-based: ethaline (choline chloride/ethylene glycol), glyceline (choline chloride/glycerol), and maline (choline chloride/malonic acid). A differential Henry's coefficient model was used to describe the behaviour of Henry's constant, and correlate it with temperature and concentration of DES in the aqueous DES solution. The correlation was found satisfactory such that the proposed model can be used in engineering calculations with reasonable accuracy.

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1. Introduction

A deep eutectic solvent (DES) is an ionic solvent that can be prepared by forming a complex between an ammonium salt and a hydrogen-bond donor (HBD), the product (DES) being one with melting point that is much lower than the melting point of the ammonium salt and HDB precursors [1]. In addition, DESs have low vapour pressure and low flammability, non-toxic, non-reactive with water, and biodegradable. It is for these reasons that DESs have great potential to be environment-friendly. Consequently, DESs have gained attention because, as recent studies have claimed, such solvents can be viable and low-cost alternatives to room-temperature ionic liquids in a variety of possible applications such as in biocatalysis [2–4], pharmaceuticals [5], electrochemistry [6,7], biodiesel purification [8–11], carbon dioxide absorption [12,13], and other chemical and industrial processes. For any future application of DESs, the solvent's physical properties will be indispensable in the design of the corresponding processes. In the literature, so far, only the following properties are available, and they are scant: density [14–18], surface tension [19], molar heat capacity [5,20], conductivity [1,21,22], and vapour pressure [23]. To date, there are no reported data on Henry's constant for the system CO_2-aqueous DES. Such data may be important particularly when the application for which a process design is to be made is absorption of carbon dioxide using DES as solvent.

In this study, we present Henry's constant, a thermophysical property that is related to gas solubility in liquid, for the system CO_2-aqueous DES. The DESs considered are those that have been prepared from choline chloride (as ammonium salt) and HBDs: ethylene glycol (ethaline), glycerol (glyceline), or malonic acid (maline). Since density data are required in the estimation of the Henry's constant, densities of aqueous maline systems in the high concentration range, which has not yet been reported in the literature, were also measured and presented. Values of the Henry's constant were then correlated with temperature and composition using a differential Henry's coefficient model.

2. Experimental

2.1. Chemicals

All DESs used were commercially available from Scionix Ltd. (United Kingdom): ethaline (1 mole choline chloride: 2 moles ethylene glycol), glyceline (1 mole choline chloride: 2 moles glycerol), and maline (1 mole choline chloride: 1 mole malonic acid). Each of these DESs has purity >98 wt%, and were used without further purification.

High purity distilled water, which was deionised (resistivity = 18.3 MΩ · cm) using a Barnstead Thermolyne water purifica-

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tion module (model EASYpure LF) was used in the experiments. We degassed the solutions prior to measurement by vacuum or using an ultrasonic bath (Branson model 3510). The aqueous solutions were prepared by weight using a digital analytical balance (Mettler-Toledo model AL204) having an accuracy of ±1 · 10^{-4} g.

2.2. Measurement of Henry's constant

In the experimental determination of Henry's constant, we used the solubility apparatus similar to those described in the studies of Al-Ghawas *et al.* [24] and Haimour [25]. The experimental setup, procedure, and method of analysis used were essentially the same as those presented in our previous works [13,26,27]. The density values used in the estimation of the Henry's constants were calculated using correlations we proposed in an earlier work for predicting densities of aqueous Glyceline and aqueous ethaline solutions [16]. For aqueous maline systems, we measured the densities of solutions in the high concentration range (50 to 100 wt%) to add to those data (10 to 40 wt%), which we had reported in a recent work [28]. The overall estimated uncertainty of the measured solubility data was ±3%.

2.3. Measurement of density

We used the Anton Paar (DMA 5000) vibrating-tube densimeter to measure the density of the samples. Details of this instrument, as well as the validation tests, were described in our previous works in which we used the same setup [16]. It was calibrated using air and degassed deionized water. The estimated uncertainties of the temperature and density measurements were ±0.01 K and ±0.1 kg · m^{-3}, respectively.

All measurements were performed in three to five replicates.

3. Results and discussion

3.1. Density

The density of maline and its aqueous solutions at (50 to 80) wt% DES were measured at temperatures from (303.15 to 323.15) K, and the values are presented in table 1. These data would serve as additions to those we recently reported for aqueous maline solutions at (10 to 40) wt% DES [28]. We likewise present figure 1 to describe the dependence of the density on DES concentration and temperature. The density decreased with temperature, and increased with DES concentration, as would be expected. Such is a typical behaviour observed for densities of many aqueous DES systems [16,18]. To represent the density of maline as a function of temperature, we used the linear relation

$$\rho = a_0 + a_1 T, \tag{1}$$

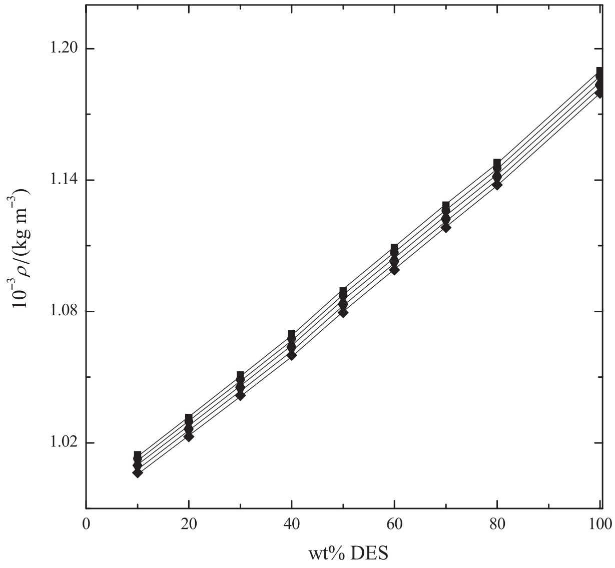

FIGURE 1. Plot of the density of aqueous maline solutions as a function of temperature: ■, 303.15 K; ●, 308.15 K; ▲, 313.15 K; ▼, 318.15 K; ◆, 323.15 K; data for (10 to 40) wt% DES, taken from [28]; lines, calculated.

where ρ is the density of the DES, T is the temperature, and a_0, a_1 are empirical parameters obtained by fitting equation (1) using the experimental data. They have values $a_0 = 1345.1$ and $a_1 = -0.5120$. For the aqueous maline solutions, we combined the experimental results obtained in the present work with those we reported earlier [28], and used a Redlich–Kister-type equation to model the temperature and composition dependence of the density (or molar volume) data. The equation has the form:

$$V^E = x_1 x_2 \sum_{j=0}^{n} A_j (x_1 - x_2)^j, \tag{2}$$

where V^E is the excess molar volume of the aqueous solution containing DES (1) and water (2), x_1, x_2 are the mole fractions of components 1 and 2, and A_j is the temperature-dependent parameter defined as

$$A_j = a_{j,0} + a_{j,1} T, \tag{3}$$

where $a_{j,i}$'s are empirical constants. The experimental V^E of each aqueous solution is expressed as

$$V^E = V_m - \sum_{i=1}^{2} x_i V_i, \tag{4}$$

where V_i is the molar volume of component i, and V_m is the molar volume of the solution defined in equation (5).

$$V_m = \rho^{-1} \sum_{i=1}^{2} x_i M_i. \tag{5}$$

Here ρ is the density of the aqueous solution, and M_i is the molar mass of component i. The values of $a_{j,i}$'s in equation (3) were obtained by fitting the equation along with equation (2) using the

TABLE 1
Density, ρ, of aqueous maline solutions at T = (303.15 to 323.15) K.

	$\rho/(\text{kg} \cdot \text{m}^{-3})$				
DES wt%	T = 303.15 K	T = 308.15 K	T = 313.15 K	T = 318.15 K	T = 323.15 K
50.0	1089.4	1087.1	1084.6	1082.0	1079.8
59.9	1109.2	1106.9	1104.4	1101.7	1099.0
70.0	1128.5	1126.0	1123.5	1120.9	1118.3
80.0	1148.0	1145.5	1142.9	1140.3	1137.8
pure	1189.9	1187.3	1184.7	1182.1	1179.7

Uncertainties: $u(\rho)$ = ±0.1 kg · m^{-3}; u(wt%) = ±0.1%; $u(T)$ = ±0.1 K.

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