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Ternary diffusion coefficients of DEG and LiBr in aqueous glycol–salt system ($DEG + LiBr + H_2O$)

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ABSTRACT

The ternary diffusion coefficients of diethylene glycol (DEG) and LiBr in aqueous glycol–salt solution containing $DEG + LiBr + H_2O$ were measured using the Taylor dispersion method. Measurements were done for temperatures of 303.15, 308.15, and 313.15 K. The investigated ternaries of the aqueous glycol–salt system contained total solute concentrations of 10, 15, and 20 wt%. The ternary diffusion coefficients, *i.e.* main diffusion coefficients (D_{11} and D_{22}) and the cross-diffusion coefficients (D_{12} and D_{21}), were correlated to temperature and concentration. A modified equation based on that proposed by Batchelor (1983) for a mixture of hard spheres in a continuum solvent was used to correlate the present diffusion coefficient data. Calculations yield satisfactory results.

1. Introduction

Liquid desiccants find important applications in dehumidification systems utilized in industrial and agricultural processes and in daily usage products (Chung and Luo, 1999; Jain and Bansal, 2007). These are materials, which due to their strong affinity to water vapor, can remove the latter from moist air and thus can be used in dehumidifiers and air conditioners. Liquid desiccant systems may offer advantages over the conventional vapor compression cooling systems and electric dehumidifiers since they utilize lower energy to effect operation due to desiccant regeneration (Chung and Luo, 1999; Dai and Zhang, 2004; Fumo and Goswani, 2001). In addition, they can use low-grade energy resources efficiently; thus, promotes the use of renewable energy and cheaper fuels, *i.e.* solar energy and waste heat (Dai and Zhang, 2004; Fumo and Goswani, 2001).

In industrial dehumidifiers, the most commonly used liquid desiccants are aqueous solutions of halide salts, such as LiCl, LiBr, and $CaCl_2$, and glycols such as diethylene glycol (DEG), triethylene glycol (TEG), tetraethylene glycol (T_4EG), propylene glycol (PG), and dipropylene glycol (DPG) (Al-Farayedhi *et al.*, 1999; Chung and Luo, 1999; Oberg and Goswami, 1998; Patil *et al.*, 1990; Yadav and Kaushik, 1995). Though such systems have been proven to be effective media for industrial applications, the search for advanced liquid desiccants has been continuing over the last years. In some studies, it was found that organic desiccant solutions glycol/salt/water formed from (DEG, PG, TEG, T_4EG, or DPG) and (LiCl, LiBr, or $MgCl_2$) can be considered as potential desiccants for dehumidification systems due to lower vapor pressures they exhibit when compared with conventionally used desiccants (Chen *et al.*, 2009, 2010; Chung and Luo, 1999; Tsai *et al.*, 2009).

To design the application of these ternary mixtures for industrial dehumidification systems, the understanding of the mechanism of mass transfer within the system, *i.e.* multicomponent diffusion and the availability of mass transfer properties will be vital. One of the important mass transport properties for any mixture is the diffusion coefficient. This parameter is necessary in order to calculate accurate transfer coefficients and other transport properties. Thus, the availability of such will be useful in process and equipment design to assess the effectiveness of the desiccant and dehumidification process as well. To date, however, literature data are limited to binary diffusivities of aqueous glycol, binary and ternary diffusivities of aqueous electrolyte solutions and ternary diffusivities of aqueous nonelectrolyte solutions (Byers and King, 1966; Cussler, 1976; Deng and Leaist, 1991; Fernández-Sempere *et al.*, 1996; Gosting and Akeley, 1952; Harned and Hildreth, 1951; Harned and Nuttall, 1949; Leaist, 1990; Leaist and Hao, 1993; Leaist and Kanakos, 2000; Leaist *et al.*, 1998; Merliss and Colver, 1969; Ribeiro *et al.*, 2001; Ribeiro *et al.*, 2002; Ribeiro *et al.*, 2006; Ribeiro *et al.*, 2007; Ternstrom *et al.*, 1996; Vergara *et al.*, 2004). Data are not yet available on ternary diffusion coefficients of studied system, DEG and LiBr in aqueous solutions, and present

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Nomenclature

a	constant of Eq. (4) for a given system and carrier stream composition
A_1, A_2, A_3	determined parameters of Eq. (14)
b	constant of Eq. (4) for a given system and carrier stream composition
B_0	baseline detector voltage
B_1, B_2	determined parameters of Eq. (15)
B_1t	small linear drift in the baseline signal
$\bar{c}_1, \bar{c}_2$	concentration of the carrier stream
$\Delta c_1, \Delta c_2$	injected solution concentration $(\bar{c}_1 + \Delta c_1, \bar{c}_2 + \Delta c_2)$ difference to the concentration $(\bar{c}_1, \bar{c}_2)$ of the carrier stream
C	total molar density (kmol/m^3)
C_k	molar density of solute k (kmol/m^3)
D_1, D_2	two overlapping Gaussians centered on time t_R with variances $r^2L/24D_1U$ and $r^2L/24D_2U$
D_{11}	main diffusion coefficient of component 1 due to concentration gradient of component 1 (m/s^2)
D_{12}	cross-diffusion coefficient of component 1 due to concentration gradient of component 2 (m/s^2)
D_{21}	cross-diffusion coefficient of component 2 due to concentration gradient of component 1 (m/s^2)
D_{22}	main diffusion coefficient of component 2 due to concentration gradient of component 2 (m/s^2)
D_i^∞	diffusion coefficient at infinite dilution (m/s^2)
D_{ii}	main diffusion coefficient (m/s/^{2})
D_{ij}	cross-diffusion coefficient (m/s^2)
D_{ik}	ternary diffusion coefficient (m/s^2)
$-D_{ik}\bigtriangledown C_k$	molar flux of solute i produced by the gradient in the concentration of solute k
f_1	relative molar ratio defined as $\bar{c}_1/(\bar{c}_1 + \bar{c}_2)$
J_i	molar flux of solute i with respect to the volume-fixed plane
L	length of the tube (m)
n	refractive index profile
R_1, R_2	molar refractivities of the solutes
R_c	radius of the coil tube (mm)
Re	Reynolds number
Sc	Schmidt number
r	inner radius (mm)
r_c	internal radius of the capillary tube (mm)
t	retention time (s)
t_R	mean retention time (s)
T	absolute temperature (K)
U	mean speed of stream (m/s)
v	detector signal
v_{max}	height of the eluted slope peak relative to the baseline
x_j	mole fraction of component j

Greek symbols

α_1	fraction of the initial refractive index contributed by solute 1
λ	ratio between the radii of the component spheres
ϕ	volume fraction of the component
ω	defined as R_c/r_c

molecular theories may not be used solely to predict such parameters accurately (Ren *et al.*, 2005).

Thus, in this work, the ternary diffusion coefficients of DEG and LiBr in aqueous solutions containing DEG and LiBr were measured using the Taylor dispersion method. Measurements were done for temperatures of 303.2, 308.2, and 313.2 K and for various total glycol–salt concentrations of 10, 15, and 20 wt%, each having five different molar ratios. The obtained ternary diffusion coefficients, namely, main diffusion (D_{11} and D_{12}) and cross-diffusion (D_{21} and D_{22}), were reported as functions of both temperature and composition. The present diffusion coefficient data were correlated by a modified Batchelor (1983) equation which was originally proposed for a mixture of hard spheres in a continuum solvent.

2. Experimental

2.1. Chemicals

The glycol, DEG, has a minimum purity of 99.9% and was obtained from TEDIA Co., whereas, the salt, LiBr, was from Scharlau Chemie and has a minimum purity of 99%. A system consisting of $MgCl_2 + MgSO_4 + H_2O$ at 298.2 K having a total salt concentration of 0.40 kmol/m^3 was used as calibration solution. The salts of $MgCl_2$ (from Scharlau Chemie) and $MgSO_4$ (from Showa Chemical Co., Ltd.) have a minimum purity of 99% and 95%, respectively. All reagents were used without further purification. The liquid water used to prepare all aqueous solutions was Type I reagent-grade having a resistivity of 18.3 MΩ cm and total organic carbon of less than 15 ppb. This water was purified using our in-house water purification system (Barnstead Thermodyne, Easy Pure 1052). Mettler-Toledo (model AL204) digital balance with accuracy $\pm 1 \times 10^{-4}$ g was used in the preparation of the solutions.

2.2. Ternary diffusion coefficient measurement

A Taylor dispersion apparatus was used to measure the diffusion coefficient of the investigated aqueous glycol–salt system. The set-up had been described in our previous works (Chang *et al.*, 2005; Su *et al.*, 2007); so, only the most important details of the measurement are discussed here.

The diffusion tube, manufactured by Valco Instruments Inc., was 50.218 m with inside radius (R_c) of 0.254 mm. Arranged in helical stack of horizontal coils with radius (r_c) of 300 mm, the tube was placed in a constant-temperature bath equipped with a thermometer (Hart Scientific, model 1502) having a reading accuracy of ±0.01 K. A metering pump provided the constant (laminar) flow of 0.08–0.2 ml/min. The carrier fluid was made to flow in a six-point injection valve (Rheodyne, model 7725i) into a diffusion tube. The carrier fluid (solution) is the actual investigated solution.

To prevent the secondary flow in the helical coil, a flow rate of 0.18 ml/min was maintained, as suggested by Alizadeh *et al.* (1980), to ensure that $Re^2\omega^{-1}Sc \leq 100$, where Re and Sc are Reynolds and Schmidt number, respectively, and $\omega = R_c/r_c$. Through the six-way injection valve, 1–1.5 μL of solution (with