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## Dehumidification of air using liquid membranes with ionic liquids



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#### ABSTRACT

This paper presents application of the hygroscopic ionic liquids, such as 1-ethyl-3-methylimidazolium ethylsulfate, 1-ethyl-3-methylimidazolium dicyanamide and 1-ethyl-3-methylimidazolium tetra-fluoroborate, for the membrane-based air dehumidification. Liquid membranes, supported by the hydrophobic porous membrane, were prepared and their dehumidification efficiency was evaluated in terms of the water recovery rate and water vapor permeability. The performance of supported liquid membranes, composed of ionic liquid, was higher when compared to that, containing triethylene glycol. Ionic liquids showed recovery of water from 80%RH humid air as high as 10 g/h.

The permeabilities of water vapor through the ionic liquids were 26,000–46,000 Barrer and the separation factor to the air was over 1000. These measured permeabilities were compared to that predicted by the solution diffusion mechanism, which was supported by the adsorption, diffusivity and viscosity measurements of the liquids. It was demonstrated that solution–diffusion mechanism well describes transport of air through the ionic liquid membrane and with further considerations can be applied for the description of water vapor transport in ILs.

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#### 1. Introduction

Room temperature ionic liquids (ILs) are organic salts that remain in the liquid state at room temperature. The unique properties of these liquids are negligible vapor pressure, low flammability, and high thermal and chemical stabilities over a wide temperature range [1]. These properties enable ILs to replace organic solvents in areas of chemistry such as organic synthesis, electrochemistry, spectroscopy, and mass spectrometry. The number of applications of ILs is increasing because of their tunable physicochemical properties and today's commercial availability.

During the last decade, many new kinds of ILs and their combinations have been developed and tested for different types of solvent-based separation processes, such as an extraction solvent in sample preparation [2], in liquid-liquid extraction and solidphase microextraction [3,4]. The dehumidification performance of 1-ethyl-3-methylimidazolium tetrafluoroborate ([emim][BF<sub>4</sub>]) liquid as an absorbent in a counter-flow dehumidifier has been reported by Luo et al. [5]. Ionic liquids have also been intensively studied as separation materials in supported liquid membranes (SLM), in which the IL is held inside the pores of a porous support (e.g., a porous solid membrane) by means of capillary forces. The efficiency of these ionic liquid-SLM have been mainly investigated for gas separation [6–8], focusing specifically on the separation of

http://dx.doi.org/10.1016/j.memsci.2015.10.069 0376-7388/© 2015 Elsevier B.V. All rights reserved. CO<sub>2</sub> from a gas mixture.

The dehumidification of air is required in various industrial processes. Currently, due to energy savings, membrane-based dehumidification has attracted researcher's attention. When compared to conventional dehumidification methods, membrane dehumidification has a potential for the development of the technology that would be more effective from economic and technical point of view. The ways to improve the process is to study the effect of the membrane material on the air dehumidification performance. So far, among the supported liquid membranes, only the triethylene glycol (TEG) membrane has been investigated for the dehumidification of air [9,10].

There may be several requirements for applying a liquid to the membrane dehumidification of air as follows:

- 1. The vapor pressure of the liquid should be low to avoid any evaporation loss;
- 2. The liquid should have low viscosity for a high permeation rate;
- 3. The affinity of the liquid towards water vapor should be high in order to achieve a high adsorption;

Previous research by Cuadrado-Prado et al. [11] demonstrated that several kinds of ILs have a comparatively high adsorption of water. Therefore, these hygroscopic ionic liquids are expected to be suitable materials for air dehumidification.

Based on the previously stated requirements as mentioned above for the liquid membrane material, this study evaluated three kinds of hydrophilic ionic liquids, which are hygroscopic by nature

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and commercially available. The main objective of the present study was to investigate the dehumidification efficiency of these ionic liquids as liquid membrane materials. The sorption of water vapor by the ILs and the water recovery ability of the liquid membranes with the ILs were experimentally investigated. Also, since water absorption by TEG is the most common process used for gas dehydration, we compared its dehumidification performance with the efficiency of the ionic liquids.

#### 2. Experimental

#### 2.1. Materials

Three ionic liquids, 1-ethyl-3-methylimidazolium ethylsulfate ([emim][ESU]), 1-ethyl-3-methylimidazolium dicyanamide ([emim][DCA]) and [emim][BF<sub>4</sub>] were used as the liquid membrane materials. The physical properties of these ILs and TEG are presented in Table 1. The ILs, [emim][ESU] and [emim][DCA], were purchased from the Merck Group, Japan. [emim][BF<sub>4</sub>], triethylene glycol (TEG) and aluminum oxide nanoparticles (40–50 nm powder) were supplied by Wako Pure Chemical Ind., Japan.

#### 2.2. Adsorption measurements

The adsorption measurements of the water vapor by the ILs were performed at 25 °C and atmospheric pressure. Each IL sample was dried and then placed in a sample basket. The sample liquid in the basket has a depth of L=1-2 mm, which is the diffusion length from the liquid surface. Various saturated salt solutions were placed in the measuring chamber to control the relative humidity of the air. The mass change in the sample under constant humidity was continuously measured by a magnetic suspension balance (FMS-S type, Rubotherm, Germany). The continuous measurement for one or two days could obtain the adsorption equilibrium data at each humidity condition. If this unsteady-state adsorption curve was compared to the theoretical diffusion equation, an apparent diffusion coefficient of water vapor in the IL could be determined.

#### 2.3. Viscosity measurement

The viscosity of the ionic liquids and TEG was measured by a viscometer (SV-10, A&D Instruments, Ltd., Japan). The temperature of the sample was maintained at  $25 \pm 0.1$  °C by an external temperature controller. The results are shown in Table 1.

#### 2.4. Membrane module and liquid membranes

The flat-membrane module consisted of a hydrophobic

#### Table 1

Physical properties of the ILs and TEG (25 °C).

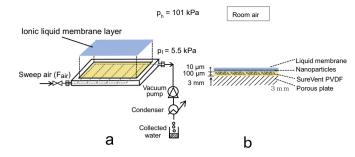


Fig. 1. Membrane module: (a) module; (b) components of the membrane.

microporous membrane (SureVent PVDF, Millipore, pore size 0.1  $\mu$ m, porosity 70%, 100  $\mu$ m thickness), which were placed on a polypropylene porous plate, and an aluminum frame [10] as shown in Fig. 1. The membrane permeation area was A=676 cm<sup>2</sup> (26 cm  $\times$  26 cm). The frame had an inlet port for sweep air feed and an outlet port for the permeate. To prepare a liquid membrane, 0.8 g of the IL was mixed with 0.2 g of Al<sub>2</sub>O<sub>3</sub> nanoparticles. The Al<sub>2</sub>O<sub>3</sub> nanoparticles were used as a supporting material of the liquid membrane. The obtained paste was directly applied under trans-membrane conditions on the surface of the SureVent PVDF membrane. The hydrophobic SureVent PVDF with the surface tension of 21 mN/m [10] allows the ionic liquid with a greater surface tension to be supported on the surface of the microporous membrane under a vacuum mode permeation condition.

The thickness of the liquid membrane layer, which in this study is associated with the membrane thickness,  $\delta$ , was calculated based on the amount of ingredient material used in the membrane preparation. The obtained thicknesses of the [emim][ESU], [emim] [DCA] and [emim][BF4] membranes are presented in Table 1. The TEG-SLM was prepared in the same way as the ionic liquid-SLMs.

#### 2.5. Dehumidification test

The experimental setup for the air dehumidification tests can be found in our previous report using a liquid membrane with the TEG [10]. The only difference in the setup of the present study is that the membrane module was placed in a chamber with a volume of 0.097 m<sup>3</sup> to maintain a constant air relative humidity for the experiment. The humidity in the chamber was controlled by an air circulation system using a water bubbler and a temperature controlled bath. All the permeation tests were conducted at room temperature and atmospheric pressure.

On the permeate side of the membrane module, a vacuum pump (N843.3 AN.22E type, KNF, Germany) with a power of 300 W and flow rate of 43 L/min (at atmospheric pressure) was placed. The permeate side pressure ( $p_l$ ) was in the range of 4.5–6 kPa. Sweep air with a flow rate ( $F_{air}$ ) of 600 cm<sup>3</sup>/min was

	[emim][ESU]	[emim][DCA]	[emim][BF <sub>4</sub> ]	TEG
Chemical structure	$H_3C$ $N$ $H_3C$ $N$ $CH_3$ $O$ $-S$ $O$ $CH_3$	H <sub>3</sub> C N CH <sub>3</sub> CN	H <sub>3</sub> C N CH <sub>3</sub> BF <sub>4</sub>	H0~ <sup>0</sup> ~0 <sup>0</sup>
(g/mol)	236.3	177.2	198.0	150.2
Viscosity, $m_B$ (mPa s)	106.7	20.7	39.6	44.6
Density, $r_B$ (kg/m <sup>3</sup> )	1239	1080	1294	1125
$P_{W}$ , Water permeability (kmol m/(s m <sup>2</sup> kPa)	$1.56 \times 10^{-11}$	$1.85 \times 10^{-11}$	$1.20 \times 10^{-11}$	$0.92 \times 10^{-11}$
Adsorption of N <sub>2</sub> at 1 atm (kg-N <sub>2</sub> /kg-IL)	_	0.000017 [26] *	0.000020 [27]	-
$D_{AB}$ by Morgan (Eq. (4)) (m <sup>2</sup> /s)	_	$1.60 \times 10^{-9}$	$7.91 \times 10^{-10}$	-
$(N_2)$ (Water)	$8.14\times10^{-10}$	$2.71 \times 10^{-9}$	$1.34 \times 10^{-9}$	-
Membrane thickness (µm)	10.3	11.5	9.9	12.8

\* Estimated from original data at 40 °C.

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