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Permeance of pure vapours in porous γ-Al_2O_3/α-Al_2O_3 ceramic membrane

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ABSTRACT

The permeances of pure vapours including water, methanol, ethanol, cyclohexanol and cyclohexanone across porous alumina ceramic membrane were measured at the temperature ranges of 200–350 °C and in the trans-membrane pressure ranges of 0.005–0.050 MPa. Water molecules adsorbed on the surface of alumina membrane through hydrogen bond and this favoured to transport of water molecules across the membrane by surface diffusion. However, alcohols adsorbed on the surface of alumina membrane through chemical adsorption and this had a disadvantage for transport of alcohol molecules across the membrane. In terms of adsorption strength, cyclohexanol adsorbed more strongly on the surface of alumina than that of methanol and ethanol. Permeance of cyclohexanol at 350 °C was higher than that at 300 °C since high temperature favoured to desorption of cyclohexanol molecules from the surface of alumina membrane, leading to reduce of blockage of absorbed cyclohexanol molecular. Only at higher temperature, when the adsorption was eliminated, the permeate behaviour of alcohols vapours was similar to that of hydrogen and nitrogen, which decreased with increase of permeate temperature. If cyclohexanol, prior to cyclohexanone, was adsorbed on the surface of the porous alumina, the adsorbed cyclohexanol molecules would form a blockage for gas-phase transport of cyclohexanone across the membrane. This greatly enhanced the permselectivity for hydrogen/cyclohexanone, which was at least 17 and was much higher than the theoretical Knudsen diffusion value of 7.

1. Introduction

Compared with conventional separation methods such as distillation, absorption, adsorption and crystallisation, membrane separation has displayed many advantages, for example, high efficiency and saving energy [1,2]. Especially, membrane separation systems offer higher flexibility when treating feed fluctuations due to their natural modular character. Therefore, membrane-based separation processes have been progressively gaining acceptation in large-scale applications during the last decades. In terms of membrane materials, membranes can be classified in two types, *i.e.*, organic membrane such as polymeric membrane, and inorganic membrane such as palladium membrane belonged to dense membrane, and ceramic membrane belonged to porous membrane. Under mild conditions, organic membrane has a highly efficient separation factor except for unresistance for solvent. As for the later, porous ceramic membrane has some unique advantages such as technical resistance, chemical inactivity, non-swelling, thermal stability, and easy cleaning [3,4]. Thus the ceramic membrane is widely used in separation process and as membrane reactor at high temperature.

Last decades of years, transport of pure gas (*e.g.* H_2, N_2) or gas mixtures in porous inorganic membranes was intensively studied and utilised well in many separation processes. Gas transport across porous membrane mainly depends on Knudsen diffusion and viscous diffusion, and only to very small porous membrane with less than 1 nm pores, molecular sieving occurs. However, the transport mechanisms of vapours across porous inorganic membrane are more complicated. For example, surface adsorption should be considered to explain the permeation mechanism of vapours of alcohols and ketones and will produce a blockage for gas-phase transport. But sometimes, surface diffusion for the adsorbed molecules contributed much to the total flux across membrane.

Sawamura *et al.* utilised mordenite membrane for separating water from water–methanol–hydrogen mixtures at the temperature ranges of 150–250 °C [5]. The separation factor of water/hydrogen and water/methanol were 49–156 and 73–101, respectively. High separation factors were obtained since water pre-adsorbed in the micropores of mordenite hindered the permeation of hydrogen and methanol. However, under lower temperature, 75–80 °C, Sekulic *et al.* studied alcohol dehydration using silica and doped silica membrane and water was easier to permeate through the membrane than alcohol [2]. For example, 10% water in feed side became 89% in permeate side through silica membrane. It is obvious that as for above two examples, alcohol adsorption took place so as to transport across membrane by means of surface

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diffusion, and capillary condensation of water vapour occurred so as to transport across membrane by means of liquid diffusion.

Assabumrungrat and White measured permeances of acetone and isopropanol through a porous alumina membrane and modified previous vapour permeate model according to their results [6]. By similar means, they tested vapour perdmeances of methanol and ethanol, respectively [7]. Under their experimental conditions, no capillary condensation occurred, and experimental values were less than predicted values due to strong adsorption of alcohols leading to blockage effect.

However, the knowledge on the vapour transport across porous inorganic membrane at high temperatures is still quite limited. Thus this prompted us to a further research. In the work presented here, we investigated that the vapours displayed the behaviour for transporting through porous inorganic membrane under pressurised conditions at high temperatures. The influence of operation parameters such as permeate pressure, temperature on permeance was discussed. Furthermore, the stability of membrane was addressed.

2. Experimental

2.1. Membrane tubes

The asymmetric alumina membrane consists of a support and a skin layer. Porous α-Al_2O_3 tube (avg. pore size = 0.1 μm, i.d. = 9 mm, o.d. = 13.5 mm, Fusan Institute of Ceramics) used as the support was cleaned successively in anhydrous ethanol, and distilled water each for 20 min at least five times, and dried at 120 °C overnight. The top layer γ-Al_2O_3 with pore diameter of about 4 nm was prepared on the outer surface of the support by a sol–gel technique [8,9]. The length of γ-Al_2O_3 porous section is 30 mm and the total length of tube is 400 mm. The other part of the membrane except for γ-Al_2O_3 porous section was sealed using superior glaze under the calcination for 20 min at 950 °C. This membrane was used to test permeances for gases or vapours.

2.2. Gas permeance test

The gas permeances through porous inorganic membrane were measured in a dead-end permeate unit. Experimental setup was shown in Fig. 1(a). Nitrogen or hydrogen gas from a gas cylinder was firstly heated up by passing it through a preheating section. It was then directly fed into the membrane module. The module consisted of a stainless-steel casing (i.d. = 21 mm and o.d. = 27 mm), inside which the ceramic tube was placed and sealed to the stainless-steel casing using swagelok fittings and Teflon tape wrapped around the membrane. Tightness at the joint between the fitting and the membrane was very important to make a good seal. The membrane module was placed in an oven, so that the system was operated under isothermal conditions. The temperature in the membrane module was measured by a type-K

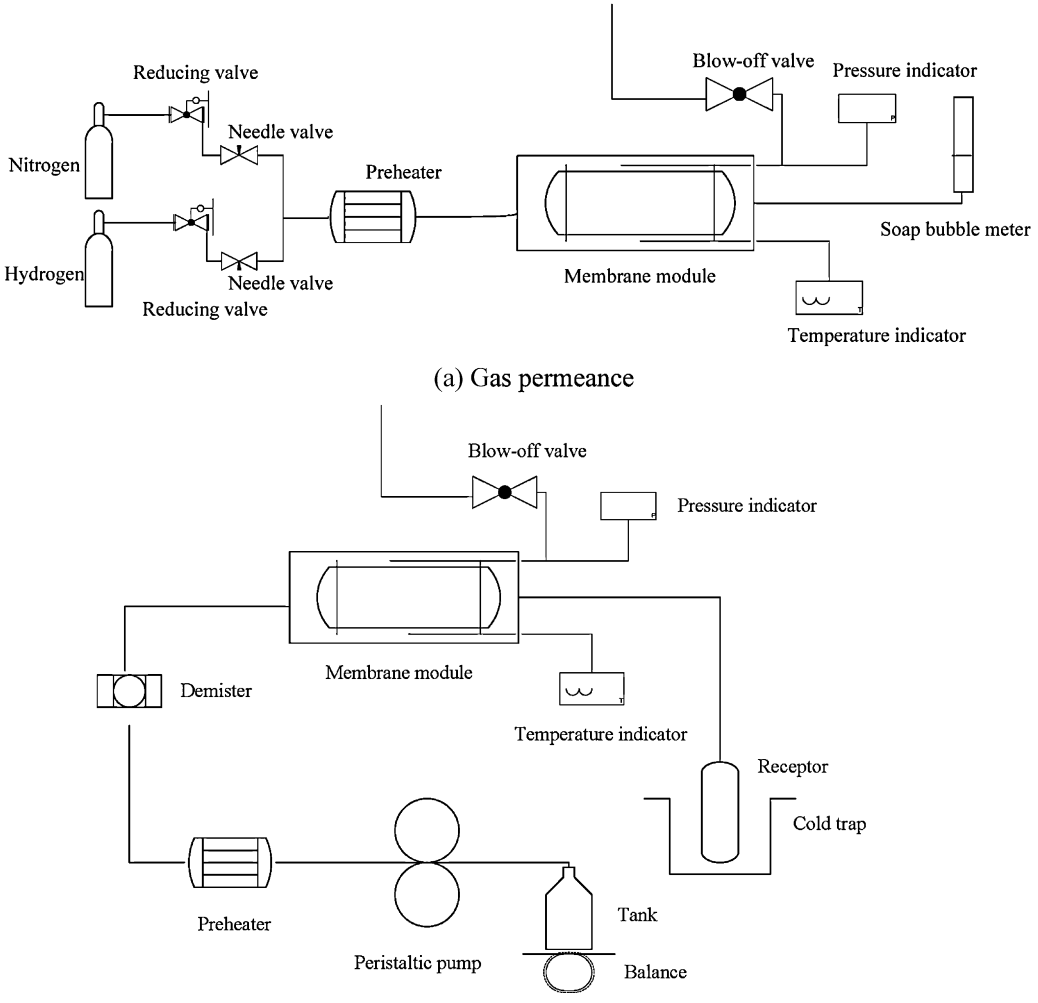

Fig. 1. Experimental setup.