



Adjusting carrier microenvironment in CO₂ separation fixed carrier membrane



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ABSTRACT

An ion-exchange material – Poly(diallyldimethylammonium carbonate) (PDDACA) containing mobile carbonate has been prepared and it has been blended in a fixed carrier membrane material – polyvinylamine (PVAm) containing abundant amine-group to adjust the carrier microenvironment of the fixed carrier membrane. It makes the mobile carrier cruise around the fixed carrier. On one hand, the CO₂ can be transported continuously in the swing area of fixed carrier through the mobile carriers, the passivity of fixed carrier can be overcome and the utilization efficiency of fixed carrier can be enhanced. On the other hand, a loose and low crystalline system will be obtained by blending two polymers, the number of effective mobile carrier will be increased with mobile carriers unlocking from the crystalline area, and the loose and low crystalline system will provide a good environment for the movement of mobile carriers. These are conducive to enhance the performance of blend membrane in comparison with single polymer membrane. The CO₂ permeance and CO₂/N₂ selectivity of PVAm-PDDACA membrane, which concurrently contains fixed amine carriers and movable carbonate carriers, can reach 909 GPU and 102 at 0.11 MPa, respectively. The performance of PVAm-PDDACA membrane is enhanced obviously in comparison with PVAm membrane and PDDACA membrane.

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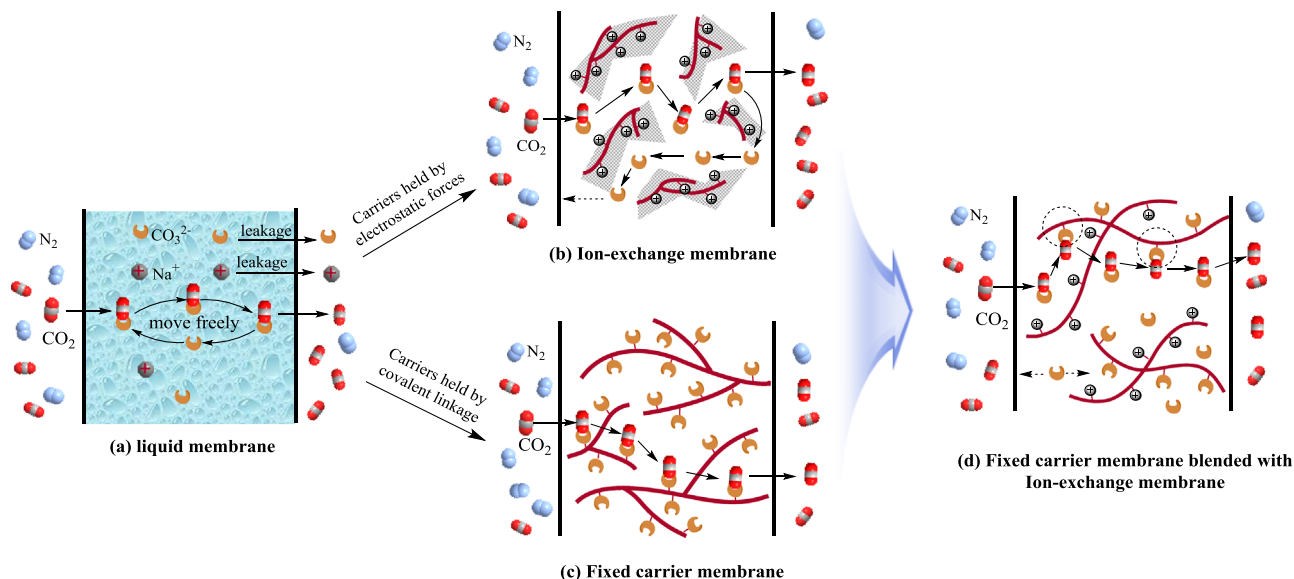
1. Introduction

CO₂ is the main greenhouse gas, which is responsible for global warming and causing extreme weather [1–4]. Meanwhile, CO₂ is the important source of carbon which can be transformed into energy and other chemical products. Separation and capture of CO₂ from flue gas and other sources have been considered as an ideal way for greenhouse gas emission control and reutilization of CO₂ as carbon resources [5–7]. Compared with other technologies, membrane-based CO₂ separation attracts more attention because it offers some advantages of low-energy consumption, simplicity of operation and environmental friendliness [8–13]. Among various membranes, the facilitated transport membrane has been intensively investigated, because the CO₂ permeance and

selectivity of the membrane could be simultaneously improved through the reversible reactions between CO₂ and reactive carriers (e.g. carbonate, amine group and carboxylate) in the membrane [14–18]. The facilitated transport membrane originates from liquid membrane and the liquid membrane usually possesses excellent gas separation performance because the carriers of liquid membrane can move freely and actively transport CO₂ like a boat (Scheme 1(a)). However, the poor stability of carriers in liquid membrane (carrier loss) seriously limits its application [19]. In order to prevent the loss of the carriers, the carriers are held either by electrostatic forces in ion-exchange membrane or by covalent linkage in fixed carrier membrane. The carriers in ion-exchange membrane can still move as well as actively meet and transport CO₂ like in the liquid membrane. In theory, the ion-exchange membrane should possess good gas separation performance like the liquid membrane (Scheme 1(b)). However, the charged polymeric matrix of ion-exchange membrane usually has strong polarity and high cohesive energy density which easily induce oriented crystallization [20–22]. This will lead to the phenomenon

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Scheme 1. Schematic of CO₂ transport mechanisms in different membranes: (a) Na₂CO₃ liquid membrane; (b) ion-exchange membrane; (c) fixed carrier membrane; (d) fixed carrier membrane blended with ion-exchange membrane.

that part of the carriers will be locked in the crystalline area and the efficiency of carriers cannot be fully expressed. Simultaneously, the movement of the rest free carriers will be hindered by the crystalline area (Scheme 1(b)). Thus, the performance of ion-exchange membrane is usually not as high as expected. In contrast, the polymer chain of fixed carrier membrane is loose and flexible. It is easier to make the fixed carrier membrane get excellent permeability [23,24]. But the selectivity of fixed carrier membrane is not very high, because the carriers in the fixed carrier membrane are covalently bonded to the membrane matrix and can only vibrate in a confined space to wait the arrival of CO₂ (Scheme 1(c)). Most importantly, the fixed carrier membrane is easy to be modified by physical or chemical method, and the material of fixed carrier membrane is cheap and readily obtained. Therefore, the fixed carrier membrane is widely favorable [14].

Here we propose to adjust the carrier microenvironment of fixed carrier membrane by blending the fixed carrier membrane material with some ion-exchange membrane material, which will make a certain amount of mobile carrier cruise around the fixed carrier. On the one hand, the passivity of fixed carrier can be overcome, the fixed carrier do not need to passively wait for CO₂ entering into its swing area, the CO₂ can be transported in the swing area of fixed carrier through mobile carrier and the utilization efficiency of fixed carrier can be enhanced. On the other hand, the fixed carrier material is beneficial to improve the efficiency of the mobile carrier of ion-exchange material. The neat arrangement of two kinds of polymer can be disrupted by the interpenetrating of two kinds of polymer, thus, the crystallization of ion-exchange material can be decreased and the part of carbonate ions fettered by crystallization can be liberated [25,26]. Meanwhile, the loose and flexible polymer chains provide a relaxed environment for the free movement of mobile carrier. These are conducive to enhance the efficiency of the ion-exchange membrane. Thus, it is reasonable to speculate that the blend membrane will combine the advantage of the fixed carrier membrane and ion-exchange membrane.

In this paper, the fixed carrier membrane material polyvinylamine (PVAm) containing abundant anime carrier and the ion-exchange membrane material Poly(diallyldimethylammonium carbonate) (PDDACA) containing high efficient mobile carbonate carrier have been prepared, respectively. Moreover, the PVAm has been blended with some PDDACA to fabricate PVAm-PDDACA

membrane for CO₂ separation, the effects on the performance of the PVAm-PDDACA membrane and the stability of the PVAm-PDDACA membrane have been investigated.

2. Experimental

2.1. Material

N-vinylformamide (NVF) from Aldrich was distilled under vacuum and stored at $-15\text{ }^{\circ}\text{C}$. 2, 2'-Azobis (2-methylpropion-amidine) dihydrochloride (AIBA) from Aldrich was recrystallized from ethanol and stored at $-15\text{ }^{\circ}\text{C}$. Poly(diallyldimethylammonium chloride) (PDDA, 20 wt% aqueous solution, M.W. 400,000–500,000), sodium hydroxide ($>97\%$), ammonium bicarbonate ($>99\%$) and Amberlite 717 were purchased from Aladdin Reagent Co., Ltd. Ethanol and hydrochloric acid were of analytical grade and obtained from Tianjin Kewei Chemical Reagent Co. Ltd. (China). The polysulfone ultra-filtration membrane with an average cutoff molecular weight of 6000 was supplied by Vontron Technology Co., Ltd. (China).

2.2. Synthesis of PVAm and PDDACA

PVAm was synthesized according to a method in the reference [27]. The synthetic procedure is shown in Scheme 2 and described in details as follows: 10 g (0.14 mol) NVF monomer and a certain amount of AIBA were dissolved in 40 ml deionized water. Then the synthesis of PNVF was carried out in a flask equipped with a stirrer at $50 \pm 2\text{ }^{\circ}\text{C}$ for 5 h under nitrogen atmosphere. After polymerization, a colorless transparent viscous gel-like liquid was obtained. A certain concentration of hydrochloric acid was added into the flask and acidic hydrolysis was carried out at $70 \pm 2\text{ }^{\circ}\text{C}$ for 2 h. Then, the solution was poured into a large quantity of ethanol to precipitate a white deposit. The deposit was dried in a vacuum oven at $40\text{ }^{\circ}\text{C}$ for 48 h. The dried product, which is PVAm·HCl polymer, was dissolved in a certain amount of deionized water. The polymer solution was mixed with excessive strongly basic anion exchange resin (Amberlite 717) and was further stirred for 2 h. Finally, the PVAm aqueous solution was obtained by vacuum filtration of the mixture.

The PDDACA was obtained by anion exchange treatment of commercial PDDA. The synthesis pathway of PDDACA is depicted

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