

Preparation of a highly water-selective membrane for dehydration of acetone by incorporating potassium montmorillonite to construct ionized water channel

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HIGHLIGHTS

- Chitosan–potassium montmorillonite hybrid membrane was prepared.
- Ionic water channel was constructed by incorporated potassium montmorillonite.
- The hybrid membrane showed high separation factor.
- The possible transport mechanism for water in membrane has been proposed.

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ABSTRACT

Novel polymer–inorganic hybrid membrane was prepared by incorporating potassium montmorillonite (K⁺MMT) into chitosan (CS) for the dehydration of water–acetone mixture. The morphology, chemical and physical structure, as well as the hydrophilicity were characterized. The results showed that MMT uniformly dispersed in the chitosan matrix, formed hydrogen bonds with chitosan, and enhanced the thermal stability of the film. The membrane doped with 10 wt% of K⁺MMT exhibited the highest performance with separation factor of 2200, which was almost 8 times higher than 249 of pristine CS membrane at 50 °C for 5 wt% of water in the feed. The results suggested that the ionized water channel constructed by MMT intergallery in the hybrid membrane could greatly enhance the separation factor of CS membrane. Moreover, a plausible model for water transportation in the CS–K⁺MMT hybrid membranes was proposed. The results proved that K⁺MMT had a potential prospect in application for pervaporation.

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1. Introduction

Acetone is not only a widely used solvent, but also an important raw material for the production of methyl methacrylate and bisphenol-A (Shi et al., 2012), however, dewatering of acetone still remains a critical issue. Although acetone and water does not form an azeotrope (Shi et al., 2012), a strong reflux, a large column and high energy cost are required during the distillation process to obtain high purity acetone (Yeang et al., 2013). In this sense,

pervaporation, a membrane process for separating liquid mixture, has emerged as an energy-efficient alternative to distillation and other separation methods (Fu et al., 2014; Lee, 1993; Liu et al., 2011b; Qiao et al., 2006; van Baelen et al., 2005). It has been widely used in solvent dehydration due to its highly selective, economical, energy efficient and eco-friendly characteristics (Jiang et al., 2008; Pereira et al., 2005; van Baelen et al., 2005; Xianshe and Huang, 1996).

A good pervaporation membrane should possess both high permeation flux and excellent separation factor. In dehydration processes, excellent dehydration membrane should contain water-attractive functional groups so as to preferentially absorb water molecules and facilitate the transport of water. In order to enhance the permeation, many efforts have been devoted to establish water

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channel for facilitating the transport of water. In nature, there exists aquaporin (AQP) which comprises a family of water-transporting membrane proteins (Fujiyoshi et al., 2002). Aquaporin-1 (AQP1) is firstly identified as a water channel that contains selectivity filter and has only limited intrinsic water permeability (Tornroth-Horsefield et al., 2006). Within the selectivity filter, four bound waters are localized along three hydrophilic nodes, punctuating an otherwise extremely hydrophobic pore segment. The long hydrophobic pore and a minimal number of solute binding sites facilitate rapid water transport (Sui et al., 2001). Herein, bound water sites are one of the key factors in transporting water. Similarly, in pervaporation dehydration process, it has been also found that metal ions in membranes play an important role in separating water and organic solvent. This is because the metal ions can strongly bound water in their solvation shells (Niemöller et al., 1988), which is also called the hydrated layer. The water molecules organized around metal ions can lower the solubility of non-polar molecules and therefore induce the salting-out effect (Li et al., 2014). Because of the salting-out effect, polar water can transport through the hydrated layer, this is beneficial to enhancing the membrane's selectivity of water. Inspired by aquaporin, if unobstructed channels with hydrated metal ions (ionized water channel) could be constructed in the membranes, the separation performance of the membranes may be significantly improved.

To construct ionized water channel, the inorganic material with channels containing metal ions can be implanted into polymer matrix. On one hand, the ions in the intergallery channel can form hydrated layer with water which lower the solubility of non-polar acetone molecules (salting-out effect) and benefit the membrane's selectivity of water. On the other hand, the framework of the inorganic materials can prevent the polymer chains from dense packing and increase the spacing between polymer chains, which prevent the reduction of membrane permeability and avoid the trade-off effect (as shown in Scheme 1). Montmorillonite (MMT) is a typical ionic clay mineral from the smectite family, it consists of negatively charged host layer and hydrated cations cruised in the intergallery (Choudary et al., 2005; Ikeda et al., 2011; Joshi et al., 2009; Mitsudome et al., 2007; Yao et al., 2010). The metal ions in MMT interlayer usually include K^+ , Na^+ , Li^+ , Ca^{2+} and so on. Previous studies showed that the K^+ was more suitable to improve the performance of the membrane than other metal ions. Cabasso and Liu (1985) discussed the effect of different ions on the transport of alcohol/water mixture in Nafion membrane and it was found that separation factor increased in the following order: $K^+ > Na^+ > Li^+$. If potassium montmorillonite (K^+ MMT) is incorporated into the polymer, the hydrated ions in the intergallery can increase the selectivity of water; meantime, the nanosheets inhibit the polymer chains dense packing.

In recent years, polymer-clay hybrid membranes have received greater attention for pervaporation dehydration of organic solvent

due to their hydrophilic, thermal and mechanical properties. Adoor et al. (2006) incorporated Na^+ MMT into PVA matrix for pervaporation dehydration of aqueous mixtures of isopropanol and 1,4-dioxane, the separation factor was found to be increased. Bhat and Aminabhavi (2006) intercalated the NaAlg into partial Na^+ MMT galleries and the selectivity to water was increased, this result is ascribed to the spatial distribution, rearrangement of intercalating polymer chains and interfacial interaction between the clay layers and the polymers.

In this study, K^+ MMT was used as the inorganic filler with channels containing metal ions to enhance the water transportation and selectivity. Chitosan (CS) was chosen as the polymeric matrix due to its good film-forming property, high hydrophilicity as well as excellent chemical-resistant characteristic (Ma et al., 2013). Varied amount of K^+ MMT was incorporated into CS to prepare CS- K^+ MMT hybrid membranes. The physical and chemical structure were characterized. Separation performance of the hybrid membranes was evaluated by pervaporation dehydration of acetone. Mass transport mechanism of the hybrid membranes was discussed, especially the effect of hydrated layer of ions in the MMT intergallery on water selectivity. Aimed at optimizing the operation conditions of the as-prepared membranes, the effects of operation temperature and feed concentration on the separation performance were evaluated as well.

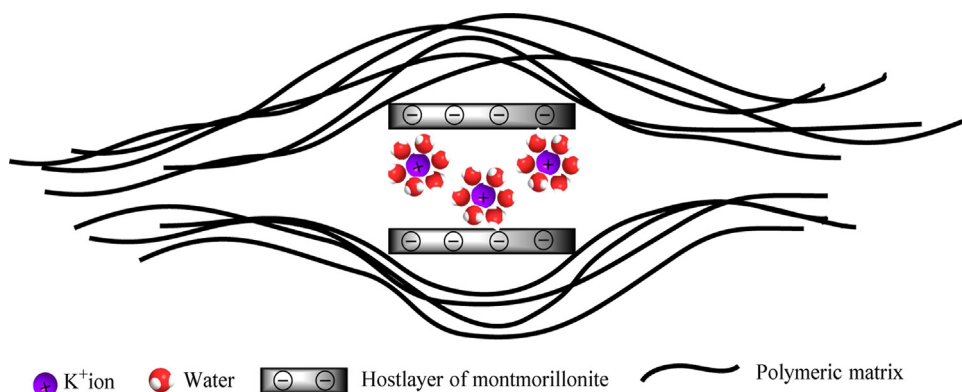
2. Experimental

2.1. Materials

The flat-sheet polyacrylonitrile (PAN) ultrafiltration membrane with a molecular weight cut-off of 100,000 was obtained from Shanghai MegaVision Membrane Engineering and Technology Co. Ltd. (Shanghai, China). Chitosan (viscosity of 400 mPa s) and K^+ MMT (specific surface area of 240 m²/g) was obtained from Aladdin Reagent (Shanghai) Co. Ltd. Acetic acid (≥ 99.5 wt%), glutaraldehyde (GA) (50 wt%), hydrochloric acid (36–38 wt%) and absolute acetone (≥ 99.7 wt%) were purchased from Tianjin Kewei Ltd. (Tianjin, China). All the reagents were of analytical grade and used without further purification. Deionized water was used in all experiments.

2.2. Membrane preparation

The flat-sheet PAN membrane was immersed into deionized water for two days to remove glycerol, and dried at room-temperature. K^+ MMT was dispersed in water by stirring for 2 days and sonicating for 30 min by cell rupture. Different amount of K^+ MMT solution was diluted to 100 ml by water. 2.0 g CS powder



Scheme 1. Schematic of montmorillonite implanted in polymeric matrix.

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