



Carbon–Al₂O₃–Ag composite molecular sieve membranes for gas separation

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ABSTRACT

Phenolic resins loaded with two different inorganic fillers (boehmite (γ -AlO(OH)) and silver (Ag)) were used to prepare composite carbon membranes. Polymer solutions containing γ -AlO(OH) and AgNO₃ were prepared and the effect of Ag on the transport properties of the composite membrane was evaluated. The polymer solutions were coated on α -Al₂O₃ tubes and carbonized in a single dipping-drying-pyrolysis step. After pyrolysis at 550 °C, γ -AlO(OH) yielded γ -Al₂O₃, and Ag agglomerated forming spherical nanoparticles of 30 nm in diameter. Ag loading enhanced the carbon membrane performance for several gas pairs of interest, especially for C₃H₆/C₃H₈ separation, where the C₃H₆/C₃H₈ permselectivity increased from a maximum of 15 to 38.

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1. Introduction

Polymer membranes are successfully implemented in several industrial processes (Fritzsche and Kurz, 1990; Nunes and Peinemann, 2006). Their production is reproducible, can be made in flat sheet or hollow fiber configurations, and scale up is simple (Koros and Mahajan, 2000). However, polymer membranes are chemically and thermally unstable. Moreover, their performance for each gas separation is limited by an upper bound curve which relates permeability and selectivity, as reported by Robeson (1991, 2008).

Meanwhile, inorganic membranes such as micro- and mesoporous silica membranes (de Vos and Verweij, 1998), zeolites membranes (Coronas and Santamaria, 1999) and carbon membranes (Ismail and David, 2001) have emerged as potential candidates for gas separations. Carbon molecular sieve membranes (CMSM) are obtained from carbonization of polymeric precursors under a controlled inert atmosphere. Among the most used precursors are polyimides (Barsema et al., 2003c; Fuertes and Centeno, 1998; Tin et al., 2006; Yamamoto et al., 1997), polyacrylonitrile (PAN) (Linkov et al.,

1994; Song et al., 2009), cellulose (Campo et al., 2010a; Lie and Hagg, 2006) and phenolic resins (Centeno and Fuertes, 1999; Fuertes and Menendez, 2002). After carbonization, the resultant pore network comprises large pores (0.5–0.8 nm) interconnected by smaller pores (0.3–0.5 nm), called constrictions, responsible for molecular sieving (Koresh and Soffer, 1980). The separation properties exhibited by CMSMs result from this molecular sieving mechanism combined with selective micropore diffusion (Rao and Sircar, 1993), where the more strongly adsorbing species impair the passage of less-adsorbing ones. The main feature of CMSMs is that its pore size distribution (PSD) can be tailored to suit a desired application by optimizing the pyrolysis conditions and chemical composition of the precursor. These membranes present high permeabilities and excellent selectivities, overcoming the Robeson upper bound for polymers, and they are resistant to harsh thermal and chemical environments enabling numerous potential applications (Ismail and David, 2003). However, they are brittle, lack mechanical stability and production suffers from low reproducibility (Koros and Mahajan, 2000).

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