



Layer-by-layer assembly of carbide derived carbon-polyamide membrane for CO₂ separation from natural gas

Abdelrahman Awad^a, Isam H. Aljundi^{a, b, *}

^a Chemical Engineering Department, King Fahd University of Petroleum and Minerals, Dhahran 31261, Saudi Arabia

^b Center of Excellence in Nanotechnology, King Fahd University of Petroleum and Minerals, Dhahran 31261, Saudi Arabia



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ABSTRACT

Carbide-derived-carbon (CDC) have garnered increasing attention because it has shown potential for various applications including gas separation. In this article, we report for the first time the fabrication of CDC/polyamide composite membrane for CO₂/CH₄ separation. Different loadings of CDC were embedded in the polyamide layer to develop CDC/Polyamide mixed matrix membranes (MMMs) by the interfacial polymerization reaction of piperazine (PIP) and isophthaloyl chloride (IPC). The morphology and structural properties of the fabricated membranes, as well as the CDC nanoparticles, were examined by SEM, FT-IR, TGA, XRD, and N₂ adsorption analysis. The characterization results confirmed the successful incorporation of the CDC nanoparticles into the rough polyamide layer. Gas permeation measurements of the fabricated CDC/PA membranes demonstrated an 88% and 49% enhancement in CO₂ permeance and CO₂/CH₄ selectivity compared to the neat polyamide membrane. The CDC nanoparticles disrupted the polyamide matrix which resulted in a higher free volume to transport gases. In addition, MMMs were assembled layer-by-layer and their permeation tests revealed that building more than one selective layer on top of the polysulfone support increased the CO₂/CH₄ selectivity and decreased the CO₂ permeance. MMMs with 10 selective layers showed the best separation performance with a CO₂/CH₄ selectivity of 24.

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1. Introduction

Current demographic trends suggest that the world population will grow by 24.6% and reach 9.1 billion by 2040 [1]. With this population growth, the demand for clean energy is certain to increase rapidly. The estimated consumption of natural gas (NG) in 2014 was 3393 billion cubic meters and this is projected to expand by 45% by 2040 [1]. Even though NG is considered as a green energy source compared with other fossil fuels, it contains impurities such as CO₂ and H₂S. Large amounts of CO₂ in raw NG affect the selling price, damage pipelines and equipment, and lead to pipeline blockages during the transportation of liquefied natural gas (LNG) over long distances [2]. Therefore the amount of CO₂ in NG must be reduced to the limit specified by transportation and distribution companies (<2%) [3]. Since the importance of clean NG is increasing, the search for a new effective, robust and efficient process for natural gas purification has been accentuated in recent

research studies.

CO₂ capture methods using conventional absorption [4–8], adsorption [9], and cryogenic [10] processes have been shown to have serious disadvantages. On the other hand, membrane separation is simple, cheap, energy efficient and has a better environmental impact. These advantages make membrane gas separation a competitive technology for CO₂ separation [11–14]. Over the past few decades, membrane based CO₂ separation has been extensively investigated and developed using a large number of materials (polymeric, inorganic and hybrid) and different process schemes [12–15]. Polymeric membranes are cheap, mechanically stable and easy to prepare in different modules. Nevertheless, the tradeoff limitation and the issue of plasticization are a challenge for using polymeric membranes in industry. Although inorganic membranes overcome the trade-off limit in small scale production, much larger costs are incurred when manufacturing inorganic membranes. Moreover, improvements in reproducibility are needed for large-scale production [14].

Recently, (MMMs) have been known to exceed the Robeson's tradeoff limits and to be highly processable and cheap compared to inorganic membranes [16–18]. It can be considered as a modified

* Corresponding author. Chemical Engineering Department, King Fahd University of Petroleum and Minerals, Dhahran 31261, Saudi Arabia.

E-mail address: aljundi@kfupm.edu.sa (I.H. Aljundi).

organic membrane in which either micro or nanoparticles of inorganic fillers are embedded into an organic continuous matrix to form a heterogeneous membrane. Over the past few decades, tremendous advancements in MMMs for CO₂ separation have been made by using different inorganic fillers such as zeolites [19–23], MOFs [24–28], silica [29–31], metal oxides [32,33], new nanomaterials [34–37], and carbon materials [38–42]. Obviously, MMMs provide a new route to benefit from the advantages of inorganic materials. However, the inorganic fillers are not completely utilized because some of the incorporated nanoparticles end up in the nonselective substrate layer. Also controlling the distribution of fillers and the filler/polymer compatibility are considered as constraints in MMMs [17].

In this regard, thin film composite (TFC) and thin film nanocomposite (TFN) membranes are types of membranes that have a selective layer prepared by interfacial polymerization (IP) reaction [43]. TFC/TFN membranes can provide a high level of separation performance by precisely controlling the distribution of inorganic nanoparticles so they can be deposited only within the skin of the top selective layer during the IP reaction [43]. The IP reaction is defined as a type of step-growth polymerization in which the polymerization reaction occurs at an interface between an aqueous solution containing one monomer (polyamine) and an organic solution containing a second monomer (polyacyl halides). The IP process produces polyamide (PA) membranes with the following advantages: the possibility of producing an ultrathin selective skin layer, the availability of a large number of monomers that can be used, and more importantly, their defect-free nature and ease of scale-up to the commercial modulus [44]. However, most, if not all, of the currently developed TFC/TFN membranes are focused on reverse osmosis [45–48] nano-filtration [49], and the pervaporation process [50]. The development of TCN/TFN membranes using IP for the application of gas separation still lags behind and needs further study.

Nowadays, special carbon materials with a high surface area and nano-size pores are playing a significant role in the advancement of new technologies. With the advent of the nanotechnology revolution, carbon nanomaterials have garnered increasing attention from scientific researchers because they have shown potential for various applications including gas separation and storage. Carbide-derived carbon is a tunable carbon material synthesized by extracting metals from metal carbide by leaching in supercritical water, halogenation at high temperatures, vacuum decomposition, and other methods. Back in the 1960s, carbide-derived carbon was produced as a byproduct from the metal chloride manufacturing process according to Eq. (1):



where Me is the metal [51].

According to the preparation method and conditions, amorphous or nano-crystalline structures can be synthesized [52]. Due to the layer-by-layer extraction of metal from the template metal carbide, precise control over the nanoparticles' properties (pore size and shape, surface chemistry and surface area) could be achieved by changing the operating conditions, composition and structure of the carbide precursor. Gogotsi and his co-workers [53] produced CDC with a pore size distribution comparable to the pore size of zeolite materials and narrower than certain carbon nanotubes (CNTs) and activated carbon. Ranjan et al. [54] synthesized TiC-CDC at 800 °C and reported a methane gas uptake of 46 cm³/g for the prepared TiC-CDC nanoparticles which is higher than the gas uptake of both activated carbon (less than 35 cm³/g) and some MOFs (less than 10 cm³/g).

The use of carbide-derived carbon as a membrane was

investigated by Hoffman et al. [55]. They fabricated an amorphous membrane structure and their permeation results demonstrated nitrogen permeability of 67 Barrer. In another work, Hoffman et al. [56], reported on CO₂ and CH₄ sorption analysis at 25 °C of Ti₃SC₂-CDC prepared at 600 °C. The results demonstrated an adsorption of 125 and 55 cc/g for the CO₂ and CH₄; respectively. The differences in the gas sorption were attributed to the interactions between the carbon surface and gas molecules as well as the chemical affinity of gases on the CDC surface, rather than to the molecular sieving effect. Therefore, CDC offers many advantages in separation processes over polymers, zeolite, and porous carbon and provides higher gas sorption capacity.

Despite the attractive characteristics and the enormous potential of CDC for gas separation the area has not been well-studied for this application neither as an inorganic membrane nor as a nanofiller to fabricate MMMs. Therefore, this work is an attempt to introduce new nanomaterials as a filler to delve deeper into TFN membranes for the application of CO₂ separation. The objective of this research is to prepare, characterize, and evaluate the performance of CDC/PA multi-layer MMMs. The separation properties of the fabricated membranes were evaluated using the pure gases of CO₂ and CH₄ in a constant volume variable pressure apparatus. Furthermore, the effect of CDC loading and operating conditions (T, P) on the separation performance were studied.

2. Experimental

2.1. Materials

All chemicals were purchased from Sigma-Aldrich except methanol and *n*-hexane which were obtained from Scharlau. Polysulfone pellets (Mw of 35000) were used to fabricate the membrane support. *N,N*-dimethylacetamide (DMAC) with a purity of 99.9% was used as a less volatile solvent, and inhibitor-free tetrahydrofuran (THF) with a purity of 99.9% was the primary volatile solvent, while absolute ethanol was selected as a non-solvent additive. PIP and IPC with a purity of 99% were used as the monomers for the IP reaction. Titanium Carbide Nano powder (TiC, 99 + %) purchased from USA Research nanomaterials Inc., USA. CO₂ and CH₄ gases with a purity of 99.999% were purchased from Abdullah Hashim Company, KSA.

2.2. Synthesis of CDC nanoparticles

Following the reported procedure by our group [57], titanium carbide in a quartz boat was inserted in a quartz tubular furnace and heated at a rate of 10 °C/min to the desired temperature, while continuously purging with Argon to create an air/oxygen free closed system. When the furnace temperature reached the desired set point, pure chlorine gas was introduced at a flow rate of (10–13 cm³/min) for 3 h. After chlorination, the post treatment was carried out with hydrogen gas at the same final temperature for 1 h to remove the remaining chlorine from the CDC which enhances the surface area and micro-pore volume of the nanoparticles. Then, the furnace was purged with argon gas to cool it down to an ambient temperature.

2.3. Membranes preparation

2.3.1. Polysulfone support

PSF support was fabricated by a dry/wet phase inversion technique. Prior to membrane preparation, the PSF polymer pellets were dried overnight at 100 °C in a vacuum oven in order to completely remove the moisture from the polymer. Dry PSF pellets were dissolved in a mixture of DMACs and THF, then ethanol was

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