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Removal of hydrogen sulfide from methane using commercial polyphenylene oxide and Cardo-type polyimide hollow fiber membranes

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Abstract−The performance of commercially available poly (2,6-dimethyl-1,4-phenylene oxide) (PPO) and Cardo-type polyimide (PI) hollow fiber membranes was investigated in removing hydrogen sulfide from methane in a series of bench-scale experiments. It was observed that in the concentration range of hydrogen sulfide in methane from 101 to 401 ppm, the methane permeability decreased in the presence of hydrogen sulfide for Cardo-type polyimide hollow fiber membranes, whereas the PPO membrane performance was not affected. The separation coefficients of hydrogen sulfide/methane were 6 and 4 for PI and PPO membranes, respectively. Effects of temperature on the performance of PI and PPO membranes were investigated. It was observed that the permeabilities of both components of the mixture increased by increasing temperature, whereas the selectivities remained constant.

Key words: Polyphenylene Oxide Membrane, Polyimide Membrane, Natural Gas Sweetening, Hydrogen Sulfide, Hollow Fiber

INTRODUCTION

Natural gas resources around the world contain different levels of acid gases, mainly hydrogen sulfide and carbon dioxide, as impurities. Samples of Iranian raw natural gas have H_2S contents from 66.2 ppm to 3.27 mol% in different gas fields. Due to the toxic and corrosive properties of this gas, it should be removed from natural gas in order to meet the pipeline specifications.

While absorption processes are the main treatment for the removal of acidic gas from natural gas, polymeric membranes have gained momentum during the past few decades.

The advantages of membranes compared with the competing processes are their lower energy and capital costs as well as operation simplicity, easy scale up, and smaller carbon footprint. Commercialization of membranes for natural gas sweetening started around 1979-80 [1].

Although a significant number of studies have been focused on the removal of CO_2 from CH_4, the permeation behavior of H_2S in membranes has been investigated to a lesser extent. The main reason for this is the high toxic and corrosive properties of this gas. In fact, only a few researches have been carried out on the separation of H_2S from CH_4, notably the studies reported by Stern [2-7], Baker [8,9], and Klass and Landahl [10], in which the H_2S/CH_4 selectivities have been reported. Most results presented in these researches were related to cellulose acetate (CA) [3,8], polyimide [6], polyurethane [5,7] and PEBAX [7,8] membranes.

Among the glassy polymers, Cardo-type polyimide (PI) and polyphenylene oxide (PPO) possess excellent separation properties that make them suitable candidates for gas separation. The polyimide hollow fiber membranes that were developed for CO_2/N_2 separation by "Research Institute of Innovative Technology for the Earth" (RITE, Japan) proved to be good candidates for CO_2/CH_4 separation due to their high selectivities in comparison with other glassy membranes [11].

PPO has also been proven a suitable candidate for a wide range of industrial gas separation applications. It is a linear amorphous thermoplastic with glass transition temperature (T_g) ranging from 212 to 218 °C. Because of the presence of the phenyl rings, PPO is hydrophobic and has excellent resistance to water, acids, alcohols, steam and bases. It has been reported that, among all glassy polymers, PPO shows one of the highest permeabilities to gases [12-16]. The high permeability has been attributed to the absence of polar groups in the main chain of PPO [16]. An important factor governing the separation properties of any industrial application of membrane is the presence of other contaminants in the stream.

Bhide and Stern [3] presented the effect of H_2S on the performance of CA membranes in removing acid gases from CH_4. They indicated that at concentrations greater than 1% H_2S in a tertiary mixture of CH_4-CO_2-H_2S, not only CO_2, but also H_2S shows plasticization effects. The group showed that the combined effects of the two gases reduced the selectivities of the membranes.

Lee et al. [17] studied the effect of H_2S impurity on the performance of the CA membrane and observed that in the presence of water vapor the plasticization effect of H_2S is magnified mainly in terms of permeation velocity. They proposed that membrane treatment of gases containing both H_2S and water vapor should be avoided,

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