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Comparison of High and Low Concentrations of Hydrogen Sulfide Removal from Methane Using Commercial Polyphenylene Oxide Hollow Fiber Membrane

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Abstract

In this study, removal of high concentration of hydrogen sulfide from H_2S/CH_4 compares with those of low concentration. The experiments based on membrane separation using commercial polyphenylene oxide (PPO) hollow fiber membrane. It was observed that in the concentration range of 101 to 5008 ppm H_2S in CH_4, the trends of permeability of hydrogen or methane are not similar in the whole range. In the concentrations of 101, 198, and 968 ppm H_2S in methane, plasticization is often defined as the increase in H_2S-permeance with feed pressure, due to dilation of the glassy polymeric matrix; while for the H_2S feed concentrations of 401 and 3048 ppm, the competitive sorption effects dominant, and for H_2S concentration of 5008 ppm the balance between these two effects plays an important role for explanation of its trend. This diversity, results different trends of separation factors and ideal selectivities. In the range of 101 to 5008 ppm H_2S in methane, the effect of temperature on separation factor is nearly negligible. Moreover, in the range of 101 to 401 ppm H_2S in CH_4, ideal selectivity is decreased as temperature increased; while in the range of 968 to 5008 ppm, this parameter is increased by increasing the temperature.

Keywords: Membrane gas separation, Natural gas sweetening, Mixed gas permeation, Polyphenylene oxide membrane, Hydrogen sulfide

Introduction

Hydrogen sulfide occurs naturally in the environment (e.g., in volcanic gases, marshes, swamps, sulfur springs, decaying organic matter). It is produced by living organisms, including human beings, through the digestion and metabolism of sulfur-containing materials. Hydrogen sulfide is also a byproduct of many industrial processes, such as paper manufacturing, sewage treatment, landfills, or etc. This gas is also found in petroleum and natural gas. Oil or natural gas is considered sour if it has a high percentage of hydrogen sulfide. Natural gas can contain up to 28 percent hydrogen sulfide gas, consequently, it may be an air pollutant neat petroleum refineries and in oil and gas purification of natural gas and refinement of crude oil [1]. Atmospheric releases of hydrogen sulfide represent the most significant public health for the geothermal energy industry [2]. As mentioned before, oil or natural gas is considered sour if it has a high percentage of hydrogen sulfide. It has been found that Iranian raw natural gas have H_2S contents from 66.2 ppm to 3.27 mol.% in different gas fields.

Removal of hydrogen sulfide from sour gas wells or facilities (that it calls gas sweetening) may occur in a number of ways. 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, scalability, and smaller footprint. Commercialization of membranes for natural gas sweetening started about 30 years ago [3]. Due to the toxic and corrosive properties of hydrogen sulfide, it should be removed from natural gas to the level of 2 to 4 in order to meet the pipeline and other equipments' specifications.

Among the glassy polymers, polyphenylene oxide (PPO) possesses excellent separation properties that make it suitable candidate for gas separation process. This polymer is a good option for a wide range of industrial gas separation applications. The PPO is a linear amorphous thermoplastic with glass transition temperature (T_g) ranging of 212 to 218^{o}C. Because of the phenyl rings, PPO is hydrophobic in nature and has excellent resistance to water, acids, bases, alcohols, and steam. It has been reported that, amongst all glassy polymers PPO shows one of the highest permeabilities to gases [4-6]. The high permeability has been attributed to the absence of polar groups in the main chain of PPO [4]. One of the important factors of governing the separation properties of any industrial application of membrane is the presence of other contaminants in the stream.

Hydrogen sulfide removal by membranes is an important constraint in system selection affecting the economics of the process [7-9]. It is pointed out that most existing commercial membranes are not capable of economically reducing concentration of H_2S from 5000 ppm or more to 4 ppm, and that a hybrid system including absorption would be the preferred process [7-10]. It should be mentioned that the cost of the hybrid process is independent of concentration of