

461d CO₂/C₂H₆ Separation Using Solubility Selective Membrane Materials

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Carbon dioxide is an impurity which must be removed from natural gas streams in a process commonly known as natural gas conditioning. Ethane is a major component of natural gas streams, and it forms a minimum pressure azeotrope with carbon dioxide which hinders carbon dioxide separation [1]. At 293K, the carbon dioxide/ethane azeotrope has a carbon dioxide mole fraction of 0.7 [2]. Traditional techniques (e.g., chemical absorption using amine solutions and adsorption using molecular sieves), for breaking the carbon dioxide/ethane azeotrope are capital intensive and require complex process control [3].

This study investigates the effectiveness of membranes to break the carbon dioxide/ethane azeotrope. Membrane technology has been successfully implemented in natural gas processing facilities and has been shown to be effective in separating carbon dioxide from methane [1]. The membrane material selected to break the carbon dioxide/ethane azeotrope should have a high carbon dioxide/ethane selectivity and high carbon dioxide permeability.

From previous work in our laboratory, crosslinked poly(ethylene oxide) [XLPEO] shows high carbon dioxide permeability and high carbon dioxide/ethane selectivity. The pure gas permeability and solubility of carbon dioxide and ethane in XLPEO has been measured at temperatures ranging from 253K to 308K and over a pressure range of 0 to 15 atmospheres. The permeability of carbon dioxide and ethane increase as temperature increases, while the gas solubility increases as temperature decreases. At 253K, the permeability of carbon dioxide increases strongly with increasing carbon dioxide partial pressure, since carbon dioxide strongly plasticizes XLPEO. These strong plasticizing effects are not observed at 253K for ethane.

Mixed gas permeation experiments were conducted using a gas mixture containing 45.3 mol % carbon dioxide and the balance ethane. The mixed gas results show carbon dioxide strongly plasticizes XLPEO films at lower temperatures. Slight plasticization of the film is seen at 283K and 308K. Carbon dioxide plasticization decreases the mixed gas carbon dioxide/ethane selectivity, relative to pure gas values, because the ethane permeability is enhanced by the plasticization effects of carbon dioxide. The mixed gas selectivity increases as temperature decreases. At 253K and 10 atmospheres pressure the carbon dioxide permeability is 83 Barrers and the mixed gas carbon dioxide/ethane selectivity is 13. The pure gas and mixed gas carbon dioxide permeabilities and carbon dioxide/ethane selectivities were plotted on a Robeson plot and were found to be lie close to the upper bound.

The performance of the membrane material was simulated using a computer model developed in our laboratory [4]. The mixed gas experimental conditions and results were used as parameters for the simulation. It was established that XLPEO is effective in breaking the carbon dioxide/ethane azeotrope. When the feed stream to the membrane module contains 45.3 mol% carbon dioxide and the balance ethane, 85% ethane recovery is achieved at an ethane purity of 78%, for a one pass separation at 253K.

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