Immobilized Liquid Membrane for Propylene-Propane Separation

Maryam TakhtRavanchi, Tahereh Kaghazchi, and Ali Kargari

***Abstract*—**Separation of propylene-propane mixture using immobilized liquid membrane was investigated. The effect of trans-membrane pressure and carrier concentration on membrane separation performance was studied. It was observed that for 30:70 (vol. %) propylene-propane mixture, at pressure of 120kPa and carrier concentration of 20wt. %, a separation factor of 474 was obtained.

***Keywords*—**Facilitated Transport, Immobilized Liquid Membrane, Propylene-Propane Separation, Silver Nitrate.

I. Introduction

LIGHT olefins such as ethylene and propylene are produced in great quantities in petrochemical plants. An important step in the manufacture of olefins is large-scale separation of the olefin from the corresponding paraffin. Currently, this separation is carried out by distillation, which is highly energy-intensive due to the cryogenic temperatures required for the process. Distillation columns are often up to 100 meter tall and typically contain over 200 trays. With reflux ratios greater than 10, a very high energy input is required for the distillation process [1], [2].

Membrane technology has been proposed as an alternative approach to the conventional distillation process for olefin/paraffin separation [3]. Polymeric membranes [4]–[7] and facilitated transport membranes [8]-[11] have been developed for olefin/paraffin separation. A detailed description of their performance for this specific separation can be found in literature [12]-[15].

In Fig. 1 a schematic diagram of facilitated transport mechanism for olefin-paraffin separation in an immobilized liquid membrane is depicted.

In this paper, the performance of an immobilized liquid membrane system for the separation of propylene-propane mixture was investigated.

M. TakhtRavanchi is with Amirkabir University of Technology, Tehran 15875-4413 Iran (e-mail: ravanchi@aut.ac.ir)

T. Kaghazchi is with Amirkabir University of Technology, Tehran 15875-4413 Iran (corresponding author to provide phone: +98-21-64543152; fax: +98-21-66405847; e-mail: kaghazch@aut.ac.ir)

A. Kargari is with Amirkabir University of Technology, Tehran 15875-4413 Iran and Iranian Research Organization for Science and Technology (IROST), Tehran, 15815-3538 Iran (e-mail: ali_kargari@yahoo.com)

The authors gratefully acknowledge the financial support received in the form of a research grant (research project No.: 83111149) from the National Petrochemical Company- Research and Technology center (NPC-RT), Iran.

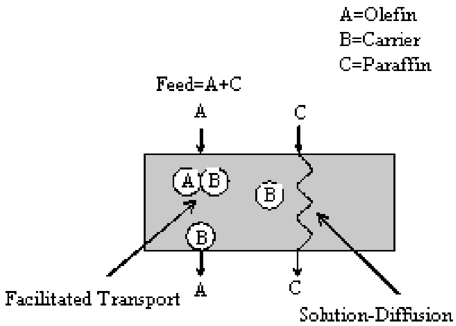

Fig. 1 Schematic diagram of the facilitated transport mechanism in an immobilized liquid membrane

II. Experiments

A. Feed Gases

Industrial grade propylene from Tabriz Petrochemical Company (99.74 mole %) and industrial grade propane from Tehran Refinery Complex (99.79 mole %) were used as feed gases and pure nitrogen was used as sweep gas.

B. Carrier Solution

Silver Nitrate ($AgNO_3$, GR Pro Analysis) which was purchased from Merck Co., was used as the carrier of propylene. An aqueous solution of silver nitrate was prepared by dissolving silver nitrate in deionized water.

C. Flat Sheet Membrane Module

Polyvinilydene diflouride (PVDF) flat sheet membranes (Durapore from Millipore, filter diameter 142mm) were used as the support of the liquid membrane. After being immersed in the carrier solution, the membrane filter was sandwiched between two compartments of the module. Once prepared, the membrane filter could be used for 3-4 weeks with no change in separation and permeation properties.

D. Experimental Apparatus and Procedure

The schematic diagram of the experimental setup is shown in Fig. 2. All tubing used to connect all parts of the setup was stainless steel (AISI 316). The experimental procedure is as follows. Propylene and propane, after passing through mass flow controllers (Brooks Instruments, model 5850S), were mixed and entered the humidifier. The humidified feed passes

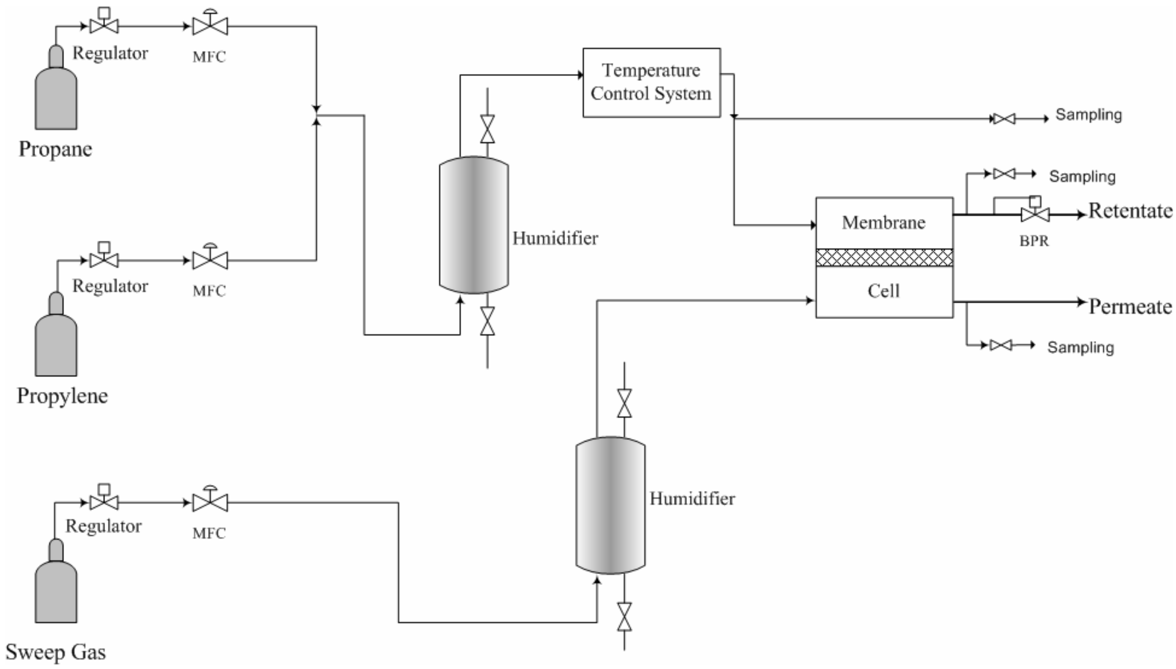

Fig. 2 Schematic diagram of the supported liquid membrane system

through a temperature control system and enters the membrane cell. A combination of a heater and a cooler were used as the temperature control system. The feed gas was introduced to the upper compartment of the cell and the sweep gas, nitrogen, was supplied to the lower compartment. The main product, permeate, was collected from the lower compartment and the secondary product, retentate, was collected from the upper compartment. A back pressure regulator (BPR, Tescom, Germany) was used on the retentate line to control the pressure of the system. During all experiments, sweep gas was at atmospheric pressure. The experiments were conducted at room temperature (298±5K). All the experimental data were obtained after an initial permeation period of 4-6 hr.

E. Analysis

The gas composition was determined by a Gas Chromatograph (Agilent 6890N) equipped with a Flame Ionization Detector (FID, Agilent Technologies Inc. column, HP Al/S, 0.53 mm in diameter, and 50 m in length).

III. Results and Discussion

The effect of trans-membrane pressure and carrier concentration on membrane performance is shown in Figs. 3-5. Different mixtures of propylene-propane were used as feed gas.

Facilitated transport is a combination of two processes: absorption (on the feed side) and stripping (on the permeate side). Increasing the pressure is in favor of absorption and decreasing the pressure is in favor of stripping. Thus, the more the pressure on the feed side, the more the absorbed propylene on the feed side. Due to the pressure difference between feed side and permeate side, the complexed propylene is decomposed on the permeate side. Therefore, the more the trans-membrane pressure, the more the driving force for separation. Hence, more propylene was transported across the membrane and separation factor was increased.

In facilitated transport membranes, propylene permeation occurs via two mechanisms: Fickian diffusion and facilitation transport. In the absence of carrier (Ag^+) in the membrane, propylene was permeated only via Fickian diffusion. When carrier was added to the membrane system, propylene was permeated via Fickian diffusion and facilitated transport. Based upon facilitated transport mechanism, when more carriers are available in the membrane, more propylene can be transported along the membrane thickness and this will cause membrane to have a better separation performance.

In an immobilized liquid membrane with a constant carrier concentration and at constant trans-membrane pressure, when a few concentration of propylene was in the feed stream, the separation factor was higher in comparison with the case when a large concentration of propylene was in the feed stream. The reason is that, in the former case, most of the propylene molecules can react with carrier molecules and can transport across the membrane via facilitated mechanism, but in the latter case, as more propylene molecules were in the membrane, all of them cannot react with the carrier molecules and cannot transport across the membrane via facilitated mechanism. Hence, separation factor in the former case is greater than in the latter case.

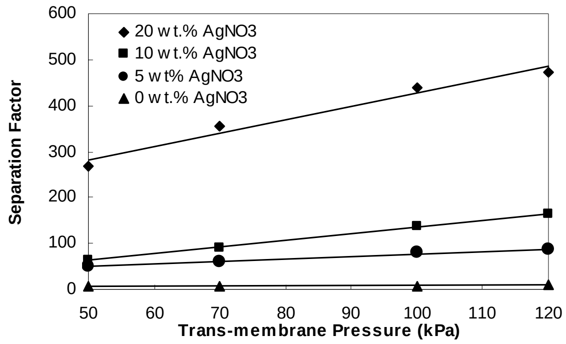

Fig. 3 Performance of membrane system for the separation of 30:70 vol.% propylene-propane mixture

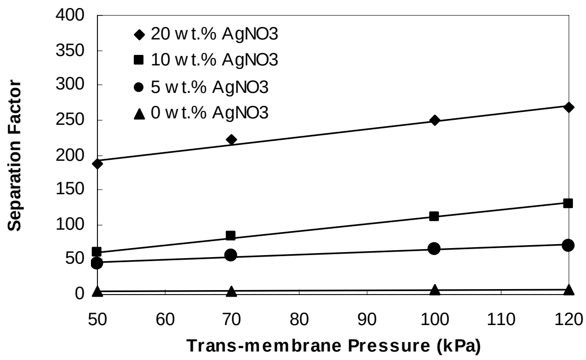

Fig. 4 Performance of membrane system for the separation of 50:50 vol.% propylene-propane mixture

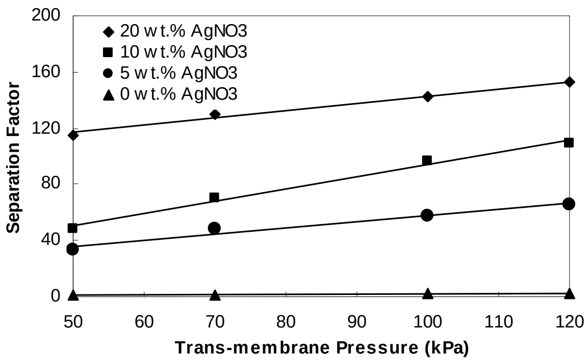

Fig. 5 Performance of membrane system for the separation of 70:30 vol.% propylene-propane mixture

IV. Conclusion

A series of experiments were carried out on the separation of propylene-propane mixtures using immobilized liquid membrane containing aqueous solution of silver nitrate ($AgNO_3$). The influence of trans-membrane pressure and carrier concentration on membrane performance was investigated. It was found that increasing the trans-membrane pressure and carrier concentration is in favor of separation factor and a more purified product was obtained.

References

[1] M. Takht Ravanchi, T. Kaghazchi, and A. Kargari, "Application of Membrane Separation Processes in Petrochemical Industry: A Review", *Desalination*, to be published.

[2] R. B. Eldridge, "Olefin/Paraffin Separation Technology: A Review", *Ind. Eng. Chem. Res.*, vol. 32, pp. 2208-2212, 1993.

[3] R. W. Baker, *Membrane Technology and Applications*, England: John Wiley & Sons Ltd, 2004, ch1.

[4] O. M. Ilinitch, G. L. Semin, M. V. Chertova, and K. I. Zamaraev, "Novel polymeric membranes for separation of hydrocarbons", *J. Membr. Sci.*, vol. 66, pp. 1-8, 1992.

[5] R. L. Burns and W. J. Koros, "Defining the challenges for C3H6/C3H8 separation using polymeric membranes", *J. Membr. Sci.*, vol. 211, pp. 299–309, 2003.

[6] K. Tanaka, A. Taguchi, J. Q. Hao, H. Kita, and K. Okamoto, "Permeation and separation properties of polyimide membranes to olefin and paraffins", *J. Membr. Sci.*, vol. 121, pp. 197-207, 1996.

[7] J. H. Kim, B. R. Min, J. Won, S. H. Joo, H. S. Kim, and Y. S. Kang, "Role of polymer matrix in polymer-silver complexes for structure, interaction, and facilitated olefin transport", *Macromolecules*, vol. 36, pp. 6183-6188, 2003.

[8] Y. S. Kang, J. H. Kim, J. Won, and H. S. Kim, "Solid-State Facilitated Membranes for Separation of Olefins/Paraffins and Oxygen/Nitrogen", in *Materials Science of Membranes for Gas and Vapor Separation*, 1st ed. Y. Yampolskii, I. Pinnau, and B. Freeman, Eds. England: Wiley, 2006, pp. 391-410.

[9] Y. Yoon, J. Won, and Y. S. Kang, "Polymer electrolyte membranes containing silver ion for facilitated olefin transport", *Macromolecules*, vol. 33, pp. 3185-3186, 2000.

[10] J. H. Kim, S. M. Park, J. Won, and Y. S. Kang, "Unusual separation property of propylene/propane mixtures through polymer/silver complex membranes containing mixed salts", *J. Membr. Sci.*, vol. 248, pp. 171-176, 2005.

[11] Y. S. Kang, J. O. Won, B. S. Jung, H. H. Park, and S. G. Oh, "Solid State Polymer Electrolyte Facilitated Transport Membranes Containing Surfactants", U.S. Patent 6 645 276, 2003.

[12] Y. S. Kang, H. Ch. Park, J. O. Won, S. U. Hong, and T. M. Kwon, "Facilitated Transport Separation Membranes using Solid State Polymer Electrolytes", U.S. Patent 2001/0015334, 2001.

[13] W. S. Ho and D. C. Dalrymple, "Facilitated transport of olefins in Ag-containing polymer membranes", *J. Membr. Sci.*, vol. 91, pp. 13-25, 1994.

[14] J. D. Way and R. D. Noble, "Facilitated transport", in *Membrane Handbook*, 1st ed., W. S. Ho and S. S. Sirkar, Eds. New York: Van Nostrand, 1992, pp. 833-866.

[15] E. L. Cussler, "Facilitated and active transport", in: *Polymeric Gas Separation Membranes*, 1st ed., D. R. Paul and Y. P. Yampolskii, Eds. U.S: CRC Press, 1994, pp. 273-300.