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Gases separation by ZSM-5 based membranes

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Abstract

Present work analyses the effect produced by the presence of ZSM-5 zeolite in an alumina tube upon the permeation rate of different gases (nitrogen, oxygen, carbon dioxide and normal-butane). A comparison between experimental data of permeation through the membrane with and without ZSM-5 zeolite was performed with an increase in the resistance to transport more evident for n-butane. Also the influence of temperature upon permeation has been analyzed.

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Keywords: Gas separation; membrane; ZSM-5; mass transfer

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1. Introduction

Gas separation from a complex stream is an important process employed widely throughout the chemical and other industries. In example the separation of air into oxygen and nitrogen, carbon dioxide capture by gas-liquid absorption or the removal of volatile organic compounds from effluent streams. However membrane-based gas separation is becoming increasingly popular due to its different inherent advantages in comparison with the more traditional procedures [1, 2]. These advantages include low capital and operating costs, lower energy requirements and generally an increase in the easily of operation than in example gas absorption processes (with important cost in solvent regeneration).

A membrane may be simply defined as a solid interphase between two bulk phases (gas or liquids) [3]. In the last few years, the importance of the use of membranes for separation processes has grown quickly taking into account the important advantages previously described and the experience in solid phase synthesis. Nowadays membranes are being employed in a wide range of uses at laboratory or industrial levels, such as microfiltration, ultrafiltration, nanofiltration, reverse osmosis and electrodialysis. The separation in membranes equipment is produced by means of differences in solutes' permeability when they flowing through the membrane. The more common classification of membranes for gas separation is based in their nature: porous inorganic and dense polymeric.

2. Materials and methods

The ZSM-5 zeolite was synthesized by hydrothermal crystallization following the method proposed by the International Zeolite Association (IZA) [4]. It consists in a previous seeding gel preparation by mixing water, sodium hydroxide and tetrapropylammonium hydroxide. The next step was the addition of silicic acid in portions and under stirring. The resulting mixture was stirred for one hour at room temperature and the mixture was aged at 100 ºC for 16 hours. This gel was used to synthesize the zeolite.

The synthesis gel was produced mixing water, sodium hydroxide and sodium aluminate. Silicic acid was added to the previous mixture again in small portions under stirring and this mixture was stirred for 1 hour at room temperature. The seeding gel was added to this mixture and shook again for 1 hour.

The crystallization process was performed in a PTPF lined stainless steel autoclave at 180 ºC for 40 hours in the absence of stirring. The product was recovered by filtration and washed with distilled water.

To produce the zeolite-based membrane using the previous obtained zeolite, the procedure consists in the production of the gel and the alumina tube coating using this gel. Then a crystallization process was performed producing the zeolite crystallization on the inner tube surface. These tubes were washed with water, dried and calcined.

The alumina tube coating using the ZSM-5 gel was performed by slip coating procedure that consists in filling the tube in vertical position during a certain period of time. It was developed using both tube ends. This procedure was performed by triplicate to guarantee a suitable coating. The solvent infiltrates in the alumina support producing the material accumulation. The tube ends were maintained in contact with the synthesis gel during a long time to obtain a suitable coating to avoid gas phase bypass.

The permeation experiments were performed using the experimental set-up shows in figure 1 (a hollow fiber contactor). The feed gas was admitted to the inner side of membrane, and the total pressure on each membrane side was atmospheric. The permeation rate was measured using a difference between the feed and outlet of gas flow-rate values. An □-alumina membrane (commonly used as support in gas separation membranes) and the zeolite-modified tube were used to evaluate the effect of these kinds of support upon gas separation and for this reason different gases have been used.

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