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# Vapor permeation modeling of multi-component systems using a poly(dimethylsiloxane) membrane

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## Abstract

This study examined the simultaneous removal and recovery of volatile organic compounds (VOCs) from permanent gaseous streams using a vapor permeation (VP) technique. The VOC fluxes in multi-component feed systems were predicted using a poly(dimethylsiloxane) (PDMS) membrane in the VP process. The investigated vapors included methanol, toluene, *m*-xylene, iso-pentane, and water. The solution-diffusion model derived from Fick's first law was used in establishing the mathematical models to describe the vapor permeation flux as a function of the downstream pressure and vapor activity level. The upstream sorption concentration was predicted using the multi-component UNIQUAC equations. The vapor diffusion coefficient was determined employing Long's model. The results demonstrated that the estimated vapor permeation fluxes were in good agreement with the experimental data. No diffusivity coupling terms were needed in the mass transfer equations for the VP of multi-component feed systems. Therefore, the diffusivity parameters established from single-component vapor systems can be extended to multi-component feed systems. This finding allows the simplification of the mass transport modeling.
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*Keywords:* Vapor permeation; Multi-component feed; Organic vapors; Modeling; Poly(dimethylsiloxane) (PDMS)

## 1. Introduction

Many industrial processes, such as petroleum refining, printing, metal cleaning, painting, gluing and coating, produce waste air streams containing organic solvents. These volatile organic compounds (VOCs) represent significant environmental pollution as well as economic loss problems. According to Baker et al. [1] solvents such as toluene, xylene, perchloroethylene, trichloroethane, ethyl alcohol, methyl alcohol and acetone, are the major sources of solvent emissions. Several techniques (including activated carbon adsorption [2,3], solvent absorption [4], thermal or catalytic combustion [5,6], condensation [5], etc.) have been proposed for reducing such emitted solvents in industry. Some methods show drawbacks in terms of efficiency and cost. Some treatments require regeneration and encounter the disadvantage of changing the disposal problem into a wastewater and/or dumping ground issue [7].

Vapor permeation (VP), a membrane-based technology, is an effective way to separate and recover volatile organic compounds from gaseous streams [1]. In this process, a vapor-laded stream is fed into a vapor permeation unit containing a membrane. Some vapor components (usually organic ones) have high affinity with the membrane material in the VP module and pass through the membrane into a permeate stream at a higher rate than permanent gases, such as nitrogen and oxygen. The retentate becomes a vapor-lean stream. The vapors are enriched and concentrated on the permeate side and can be recovered by condensation [8]. Many researches [7,9–11] have demonstrated that the VP process is economically feasible for separating and recovering organic vapors from air or other gaseous streams. Although inorganic membranes have been investigated in this application [12–15], polymeric membranes – especially poly(dimethylsiloxane) (PDMS) – are the most commonly used membrane materials for the separation of organic vapors from permanent gases [1]. The PDMS membrane yielded higher flux than the other membrane materials. For example, it demonstrated higher permeability than a poly(ether block amide) composite supported on a microporous poly(vinylidene fluoride) substrate [16] by one order of magnitude.

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The mass transfer mechanism in a VP process is the so-called solution-diffusion model [17]. A permeant transport takes place in three steps: (1) solution of permeants at the upstream membrane surface; (2) diffusion of the sorbed components across the membrane matrix; (3) desorption of the permeants from the membrane into the vapor phase at the permeate side [17]. The third step usually has negligible mass transfer resistance compared with the first two steps [18]. Therefore the sorption and diffusivity properties determine the vapor permeability.

Some researches have focused on organic vapor flux (or permeability) of single component through polymers [1,7,10] in a VP process. The effects of operating condition and process design on vapor permeability and selectivity were reported [19–23]. However, multi-component permeation behavior is still limited. Leemann et al. [9] found out that the permeability and selectivity of toluene–ethyl acetate vapor mixtures were less than the single vapor systems by ∼10%. Deng et al. [24] showed that acetone vapor permeance was higher with a moistened feed (i.e. containing water vapor) but the selectivity was lower. Baertsch et al. [25] studied the VP permeability through a silicalite-zeolite membrane fed with binary and ternary aromatic vapors. They found that the faster penetrants were slowed to rates similar to the slower permeating molecules and no separation was obtained. Gales et al. [26] separated acetone–ethyl acetate–ethanol mixture from nitrogen in a hollow fiber module consisting of PDMS and polyetherimide (PEI) sub-layer. They developed a numerical model to predict total flux and found that the calculated values were higher than the experimental ones.

The objective of this study is to estimate the VOC vapor flux through a PDMS membrane in the presence of water or other VOC vapors using the solution-diffusion theory. The investigated vapors included methanol, toluene, *m*-xylene, iso-pentane, and water. Toluene and iso-pentane are the major constituents in gasoline and serve as good model vapors for gasoline recovery applications using VP. In practical situations the gasoline vapor is accompanied with high water vapor concentrations. Therefore the effect of water activity on the performance of toluene and iso-pentane was carried out in this study. *m*-Xylene and methanol are chosen to represent non-polar and polar co-existing vapors in the feed streams. The solubility and diffusivity parameters established in previous papers [27,28] were used into Fick's first law [29] to establish the mathematical model describing the vapor permeation flux. The predicted results were compared with the experimental data to validate the mass transport model.

## 2. Experimental

### 2.1. Membrane preparation

The dense PDMS membrane was prepared from two-component ingredients (KE-1310, both from Shin-Etsu Co., Japan) according to a previous paper [30]. The polymer solution was cast and dried at 80 °C for 6 h prior to use. A digital thickness gauge (model 345, Elcometer Instrument Ltd., England) was used to measure the membrane thickness at 10 locations and the average was reported.

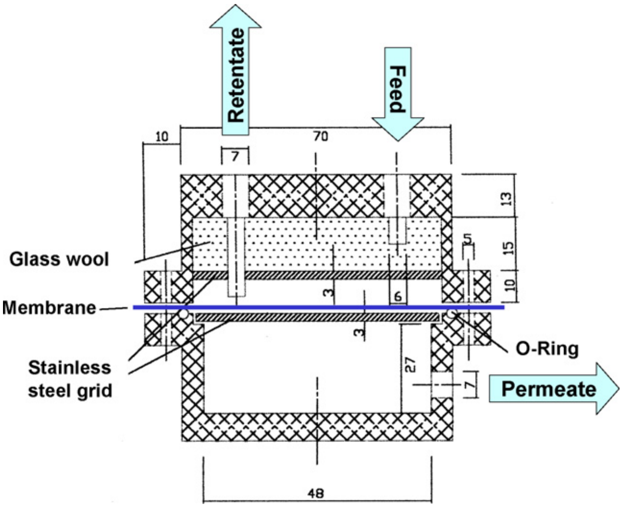

Fig. 1. Cross-sectional view of the membrane module designed for vapor permeation (dimension in mm).

### 2.2. Vapor permeation measurement

### 2.2.1. Membrane module

Fig. 1 shows a cross-sectional view of the membrane module, made of stainless steel 316. A PDMS membrane of known thickness was mounted inside the module. The effective membrane surface area was 19.63 cm$^{-2}$. A stainless grid, upstream of the membrane, was used to hold glass wool, which helped the inlet gas to distribute evenly toward the membrane surface. The grid underneath the membrane was used to reinforce the mechanical strength so that the membrane did not rupture when a vacuum was applied at the downstream side of the membrane. The temperature around the membrane module was controlled with a thermostat at 25 °C.

### 2.2.2. Generating vapors

Fig. 2 demonstrates the apparatus setup for the VP experiments. The organic vapor was generated from a bubbler placed into a thermostatic oven, which was maintained at 25 °C. To generate vapors at various activities, the vapor stream exiting from the bubbler was diluted with nitrogen gas and thoroughly mixed prior to being fed into the membrane module. Gas flow meters (Restek Corp., Bellefonte, PA, USA) and mass flow controllers (model 5850E, Brooks Inst., Hatfield, PA, USA) were used to achieve the desired flow rates, resulting in various vapor activity levels in the feed streams. Different flow rate combinations of purging nitrogen and diluting gas were used but the total flow rate was fixed. A mixer, consisting of an expanded Pyrex tube filled with glass wool, was installed to allow thorough mixing of the vapor and the diluting nitrogen gas. The vapors investigated included methanol (MeOH), toluene (Tol), *m*-xylene (Xyl), iso-pentane (Pen), and water. One to three bubblers containing different solvents were linked parallel to generate single or multiple vapors as a feed stream. The dashed lines in Fig. 2 represent the additional flow paths of gaseous streams from the bubblers.

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