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A multicomponent transport model for dehydration of organic vapors by zeolite membranes

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Abstract

The permeation of gases and vapors through zeolite membranes is usually described by the adsorption–diffusion model, which is similar to the solution–diffusion model for polymeric membranes. According to this type of model, a permeating component first adsorbs into zeolite micropores and then diffuses through the zeolite pores due to its chemical potential gradient. However, multicomponent transport is complicated by the need to account for coupling effects, and these effects should be considered in any rigorous transport model. A general multicomponent transport model was developed herein based on the Maxwell–Stefan approach to describe dehydration of organic vapors by zeolite membranes. Separation of an ethanol–water vapor mixture by a silicalite membrane was selected to demonstrate the application of the method. The mixture adsorption behavior was expressed in terms of the extended Langmuir model, and concentration dependent diffusivity of each component was recovered from permeation as well as adsorption data from the literature. The permeability and selectivity values predicted by the multicomponent transport model conformed to the experimental data. The results indicated that the kinetic coupling has a very minor contribution in transport and can be easily ignored, while the equilibrium coupling has a major role to transport through the silicalite membrane.

Keywords: Zeolite membrane; Silicalite; Dehydration; Coupling; model; Maxwell–Stefan

1. Introduction

Zeolites are inorganic materials which consist of hydrated alumina silicates. These materials are composed of micropores and have a crystalline structure with molecular sieving characteristics. They also show excellent mechanical, chemical and thermal stability during separation processes. These properties render them suitable materials

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for separation of gaseous and organic vapor mixtures. The dehydration of organic mixtures is of great importance for the production of pure organic products. Several articles have been presented in the literature dealing with good separation properties of water/organic vapor mixtures employing zeolite membranes [1,2].

The understanding of the transport mechanism within the membrane is the first step for proper design, application and development of zeolite membranes. This requires properly describing diffusional as well as the equilibrium aspects of multicomponent transport through zeolite membranes. Multicomponent transport through membranes is usually complicated by the need to account for the kinetic and equilibrium coupling effects. The Maxwell–Stefan formulation of multicomponent transport offers a solid framework for the theoretical investigation of the extent of such couplings in a specific membrane system. A distinct advantage of the Maxwell–Stefan formulation is that the binary Maxwell–Stefan diffusivities retain their physical significance and can be used directly in the description of complex multicomponent system [3].

The transport and separation characteristics of zeolite membranes for separating light hydrocarbons and other gaseous mixtures were previously investigated through the application of the Maxwell–Stefan theory by several authors [4–6]. We have also presented in our recent paper an application of the Maxwell–Stefan formulation to describe the separation behavior of liquid ethanol–water mixtures by a PDMS membrane in dialysis and pervaporation [7].

The main purpose of this study was to develop a generic mechanistic model based on the Maxwell–Stefan formulation to describe dehydration of water/organic vapor mixtures using zeolite membranes. The developed model is a predictive model capable of describing separation performance of multicomponent vapor mixtures using only pure component adsorption and permeation data. The system chosen to validate the developed transport model was the dehydration of ethanol/water vapor mixtures by a silicalite membrane. The single and binary adsorption and permeation data reported by Nomura et al. [8] were used for this purpose.

2. Transport model description

For a ternary system containing permeating components 1 and 2 and the membrane, p, the molar flux of the two permeating components is expressed as [9]:

$$\begin{pmatrix} N_1 \\ N_2 \end{pmatrix} = -C_t \begin{bmatrix} B_{11} & B_{12} \\ B_{21} & B_{22} \end{bmatrix}^{-1} \begin{bmatrix} \Gamma_{11} & \Gamma_{12} \\ \Gamma_{21} & \Gamma_{21} \end{bmatrix} \begin{pmatrix} \dfrac{\partial x_1}{\partial z} \\ \dfrac{\partial x_2}{\partial z} \end{pmatrix} \quad (1)$$

where the elements of equilibrium matrix are defined as follows:

$$\Gamma_{ij} = \frac{x_i}{RT}\left(\frac{\partial \mu_i}{\partial x_j}\right) \quad (2)$$

and the elements of multicomponent diffusivity matrix are:

$$B_{11} = \frac{x_2}{Đ_{12}} + \frac{x_P}{Đ_{1P}} \qquad B_{12} = -\frac{x_1}{Đ_{12}}$$
$$B_{11} = -\frac{x_2}{Đ_{12}} \qquad B_{22} = \frac{x_1}{Đ_{12}} + \frac{x_P}{Đ_{2P}} \quad (3)$$

where x_i is the mole fraction of component i in the membrane. $Đ_{1P}$ and $Đ_{2P}$ are binary component 1–silicalite and component 2–silicalite Maxwell–Stefan diffusivity, respectively, which are determined from Fick diffusivity and pure component adsorption isotherms which are explained below. $Đ_{12}$ is the binary componet 1–component 2 vapor