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Modeling and simulation of inorganic shell and tube membranes for vapor permeation

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Abstract

Basic features of a simulation tool developed to enable tailor made design of shell and tube modules for vapor permeation are demonstrated. The predictive model describes a ceramic membrane module using the resistance-in-series model, that accounts for concentration polarization and support layer contributions. Using ethanol dehydration as base case, a parametric study is carried out to demonstrate the effects associated with changes in variables such as feed flow rate, feed pressure, module feed side, ideal separation factor and tube diameter. From the outcome of these simulations, basic rules to qualitative predict the performance of vapor permeation modules are suggested. © 2006 Elsevier B.V. All rights reserved.

Keywords: Vapor permeation; Inorganic membranes; Shell and tube; Module performance; Ethanol dehydration

1. Introduction

Separations of azeotropic mixtures, such as alcohol/water and recovery of solvents, are typical examples of bulk chemical processes where combining membranes with distillation has proved to be a technically and economically attractive alternative for common hybrid processes [1-5]. As in some of these processes a saturated mixture at near azeotropic composition leaves the top of the column, an evident choice is to apply vapor permeation instead of pervaporation, providing a certain degree of superheating to the membrane feed to avoid the possibility of condensation in the membrane tubes [1,6].

Ethanol dehydration is a typical industrial application where pervaporation combined with distillation is an already established technology [2,7,8]. Vapor permeation is more suitable than pervaporation for alcohol dehydration, because the vapor leaving the top of the column can be the feed stream for the membrane unit. Membranes with high flux are a better option for this purpose, since the permeate stream can be recycled back to the distillation column to recover the permeated ethanol. Unfortunately for all applications a decline in flux has been

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observed with time [9–11]. Currently, the stability of the high flux performance is a main concern of the ceramic membrane manufacturers. Namely a prerequisite for successful implementation of membranes in bulk chemicals separation is achieving a rather high and stable flux.

For the configuration in which the membrane is coupled directly with the distillate stream, the membrane feed pressure is the same as in the top of the column, therefore the driving force for mass transport through the membrane can only be altered by column pressure and/or superheating temperature. This may well imply operation at pressures and temperatures well above that utilized with well established polymeric membranes [12]. Such a trend led to increased interest for development of suitable inorganic membranes. Unfortunately, the manufacture of ceramic membranes is intrinsically more expensive and complicated than for polymers, which is an additional argument for pushing toward a flux as high as possible.

However, concerning the design/rating of vapor or liquid mass transfer equipment there is always a strong relation between the hydrodynamics imposed by geometry and the mass transfer performance of the contacting device. In other words, a designer should be able to minimize intrinsic membrane limitations and find the operation at the most favorable conditions. Regarding the associated complexities this is not an easy task, which nevertheless could be easier if a reliable design tool would be available.

This study introduces a model that includes the relation between design/operating variables and the performance of a vapor permeation module. Using the dehydration of ethanol as base case, a parametric study is carried out to determine the effects associated with the changes in some variables and the mass transfer resistances. From the outcome of these simulations, basic rules to improve the performance of vapor permeation modules are suggested and the validity of the rules are demonstrated by some examples.

2. Model description

The mathematical model is comprised by a membrane mass transfer model and a shell and tube module model. The first accounts for the mass transported from the feed to the permeate side in a differential of length and the second includes the geometry and hydraulic conditions and integrates lengthwise the complete module. In the present model, the pressure drop is calculated using established engineering models [13]. The assumptions of the model are: (a) steady state, (b) binary system, (c) isothermal operation and (d) no back-mixing effects considered.

The latter is justified by the fact that the values of Bodestein number encountered in this study were well above those implying a significant role of back-mixing [14].

2.1. Membrane mass transfer

Regarding the mass transfer resistance, an inorganic membrane may be defined as a permselective barrier or interface between two phases [15], with a very thin (selective) layer which determines the separation. Because the selective layer thickness is in the nanometer range a thicker layer is necessary to give mechanical strength. The top (selective) layer must be a defect free surface, since a few defects can reduce significantly the selectivity without having much influence on the flux. A recent study [16] has indicated that the support layer could have a significant effect for membranes with high flux. Another resistance to mass transfer is that associated with the feed side boundary layer caused by the change in composition of the most permeable component. Each of individual resistances is described in greater detail in what follows.

2.1.1. Selective layer

The transport properties of the different components through the membrane are described by the permeance, Π_i , which is defined as the transport flux per unit driving force. Usually it is determined in laboratory scale membrane characterization experiments with pressure, temperature and feed composition varied over the range of interest. The corresponding mass transfer resistance can be expressed as

$$\frac{1}{k_{\rm m}} = \frac{1}{\Pi_i RT} \tag{1}$$

where *R* is the gas constant and *T* is the absolute temperature.

The temperature dependence of the permeance is usually described using an Arrhenius type equation. However, for the purposes of this study a constant experimental value for the permeance is used.

2.1.2. Support layer

The support layer must have an open porous network to minimize the mass transfer resistance. For porous membranes, the resistance can be governed by different mechanisms depending on the size of the molecules and other support characteristics. The well known Knudsen number, Kn, which relates the membrane pore diameter with the mean free path of the molecule, determines the type of transport mechanism through the pores. For the support layer of the membrane considered in this study the calculated Kn is between 10 and 100, asserting the validity of the Knudsen regime in this layer. Knudsen diffusivity is defined as

$$D_{\rm Kn} = \frac{d_{\rm p}}{3} \sqrt{\frac{8RT}{\pi \rm MW}} \tag{2}$$

where d_p is the mean pore diameter of the support layer and MW is the molecular weight of the permeation species. The corresponding mass transfer resistance can be expressed as

$$\frac{1}{k_{\rm sl}} = \frac{\delta}{D_{\rm Kn}} \tag{3}$$

where δ is the thickness of the support layer.

2.1.3. Boundary layer

During membrane operation the component to be separated permeates at higher rate than the other components in the feed, which implies a certain degree of depletion in the immediate vicinity of the selective layer. This phenomenon known as concentration polarization affects the membrane flux adversely, because it causes a significant reduction in the driving force. In fact this is a typical mass transfer resistance which can be described using appropriate expressions for the mass transfer coefficient, k_{bl} :

$$Sh = \frac{k_{\rm bl} \cdot d_{\rm h}}{D_{\rm ab}} = C_1 R e^{C_2} S c^{C_3} \left(\frac{d_{\rm h}}{l}\right)^{C_4} \tag{4}$$

where d_h is the (hydraulic) diameter, D_{ab} the diffusion coefficient and C_i are constants of the Sherwood correlation that depend on the hydraulic conditions. The values of the constants C_1 , C_2 , C_3 and C_4 are 1.62, 0.33, 0.33 and 0.33 for laminar flow (Re < 2100) and 0.04, 0.75, 0.33 and 0 for turbulent flow [15], respectively.

2.1.4. Overall mass transfer resistance

According to resistance-in-series approach, the overall mass transfer coefficient is defined as

$$k_{\rm ov} = \left[\frac{1}{k_{\rm m}} + \frac{1}{k_{\rm bl}} + \frac{1}{k_{\rm sl}}\right]^{-1}$$
(5)

where the subindices m, bl and sl stand for the membrane, boundary layer and the support layer, respectively.

Since resistance of the selective layer is assumed constant, and that of support layer appears to be practically constant at the

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