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Shortcut Methods for Simulating Separation of Multicomponent Gas Mixtures in Membranes

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Abstract- In this study shortcut mathematical models are presented for multicomponent systems with cross flow, co-current and counter current flow configurations. Cross flow mode is solved numerically in elements defined by differential stage cut and by assuming applicability of complete mixing model in the elements, which makes the model converge in short times. A simple yet robust method is proposed for solving co-current and counter current flow modes, which does not require any trial and error (except for counter current rating problems) or using dimensionless variables, and pressure drop calculations can be incorporated to the model easily. The presented models are applicable for systems with any number of components even for binary systems with no modifications required. The performance of the proposed models are validated by experimental data for co-current and counter current in differential stage cuts.

Keywords*:* Multicomponent gas separation, membrane, cross flow, co-current, counter current.

1. Introduction

Gas separation using membranes is a promising alternative to the conventional energy-intensive separation methods such as distillation. Lower capital and operating costs, lower foot print, high turn down ratios and adaptability to variations in feed are some of the characteristics that make membrane technology attractive. Similar to any other process, mathematical modeling should be carried out for designing these systems, as understanding the effects of different process parameters and flow configurations on performance of the membranes is the determining step in successful integration of these unit operations to the main processes. Several methods are developed for modeling binary systems, which is rarely encountered in actual separation problems, but when it comes to the multicomponent systems there are only a few models available, which either require iterative procedures or using dimensionless variables.

The ideal flow patterns for gas separation in membranes are complete mixing, cross flow, co-current and counter current flow. In deriving the equations for different models it is assumed that the membrane operates in isothermal condition, the Fick's law is applicable, pressure of the permeate side is constant and the permeabilities are pressure independent. In complete mixing mode the membrane is treated as a continuous stirred tank, which means the composition is uniform in the both feed and permeate sides along the membrane, and the effect of feed side pressure drop is also neglected in this model. Starting from the Fick's first law and assuming applicability of ideal gas law the rate equation can be written as follows:

$$y_p V_p = \frac{P_{Ma} A}{t}(p_h x_o - p_l y_p) \tag{1}$$

Where y_p and x_o are the concentrations of component *a* in permeate and retentate respectively, *t* is the membrane thickness, P_{Ma} is the permeability of the component *a*, and p_h and p_l are the pressures at high and low pressures sides respectively. The same equation can be written for species *b*, and by

dividing them, the following equation can be obtained, which can be used to calculate y_p for a binary system as shown by Geankoplis (2003):

$$\frac{y_p}{1-y_p} = \frac{\alpha^*[x_o - (p_l/p_h)y_p]}{(1-x_o) - (p_l/p_h)(1-y_p)} \tag{2}$$

For multicomponent systems it is required to use an trial and error procedure to find the composition of the permeate by guessing the concentration of one of the components in the permeate. Complete mixing model is straight forward, but it does not consider the actual driving force along the membrane to determine the permeation rates so it lacks accuracy, and the effect of pressure drop cannot be incorporated into it as well. The cross flow, co-current and counter current modes represent the actual operating conditions of the membranes more closely as the driving force in those models are not approximated as it is done in complete mixing mode.

2. Modeling

2.1. Cross Flow Mode

The cross flow model assumes no mixing in the permeate side nor on the high pressure side, and local permeation is just a function of local driving force. The high pressure stream has plug flow, but the permeate is pulled into vacuum and is perpendicular to the surface. The cross flow mode was first solved by Weller and Stern (1950) analytically for binary systems without considering the effect of pressure by introducing a set of dimensionless parameters. Shindo et al. (1985) solved this flow pattern by integrating the governing differential equation over differential membrane area.

In this work cross flow system is solved by dividing the membrane into differential elements and by choosing the elements small enough so that the complete mixing model's equations be applicable in each element. Solving the complete mixing mode in multicomponent systems requires a trial and error procedure, which starts with guessing the concentration of one of the components in the permeate side, and determining the area of the element accordingly. The composition of the permeate then can be calculated using the rate equations of other components, and the sum of the fractions of all the components in the permeate side should be equal to unity. This method can be used to solve the cross flow system by dividing the module into differential elements, and in this work a new approach is taken to determine the length of the differential element and that is by defining a differential stage cut in each step. This makes it possible to control the stage cut in order to minimise the error caused by complete mixing model's equations. The differential stage cut method converges faster as it determines the suitable step size based on stage cut and not the membrane area, and the pressure drop calculations can be carried out by an additional iteration in each step after determining the area.

2.2. Co-current and Counter Current Modes

In co-current mode the feed and the permeate are parallel to the membrane's surface, and the local driving force is affected by the concentrations of the diffusant in both feed and permeate side. A few methods are proposed in the literature for solving this model for multicomponent systems. Shindo et al. (1985) proposed a method for solving this flow mode which involved integrating the governing differential equation over differential membrane area using dimensionless variables by ignoring the effect of the pressure drop. Pan (1986) proposed a model for co-current and counter current modes by assuming applicability of cross flow driving force due to asymmetric structure of the membrane, and neglecting back diffusion from bulk due to presence of the porous support. Their proposed approach required a trial and error shooting method to obtain the solution in case of considering the effect of the pressure drop.

In this work a shortcut method is proposed which is based on assumption of constant concentration of the feed and permeate in differential element. In this method the composition of the permeate and its flow rate can be determined by solving the rate equations simultaneously in each element. By rearranging Eq. (1) in the differential element it can be written:

$$y_i = \frac{p_h x_i}{p_l + Vt/(dAP_{Ma})} \qquad (i = 1,..,n) \tag{3}$$

$$\sum_{i=1}^{n} y_i = 1 \tag{4}$$

Where n is the number of components. By solving the above system of equation in each element it is possible to determine the local composition and flow rate of the permeate and by integrating over the entire module the overall compositions and flow rates can be determined for the permeate and retentate streams. It should be noted that the composition of the feed entering the next step can simply be calculated by setting up the component mass balance for each gas.

Counter current flow can be solved similarly to co-current flow, but in this flow pattern the calculations should start from the retentate side and continue backward to the membrane's inlet. Solving the counter current flow requires a trial an error procedure as the composition and flow rate of the retentate should be guessed. According to Kowali et al. (1992) the performance of the two flow configurations are close to each other so the results of cross flow modes can be considered as a good starting point in solving these problems.

3. Results

The proposed method was validated using the experimental results provided by Pan (1986) for co-current and counter current flow modes for a mixture of H_2, N_2, Ar and CH_4, and the simulations results are presented in Figs. 1-4.

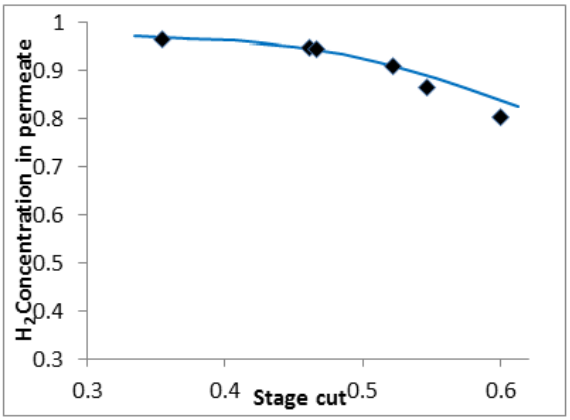

Fig. 1. Calculated concentration of H_2 in the permeate compared to the experimental data for co-current model.

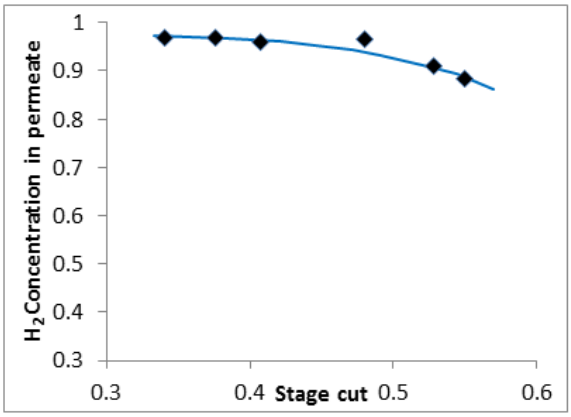

Fig. 2. Calculated concentration of H_2 in the permeate compared to the experimental data for counter current model.

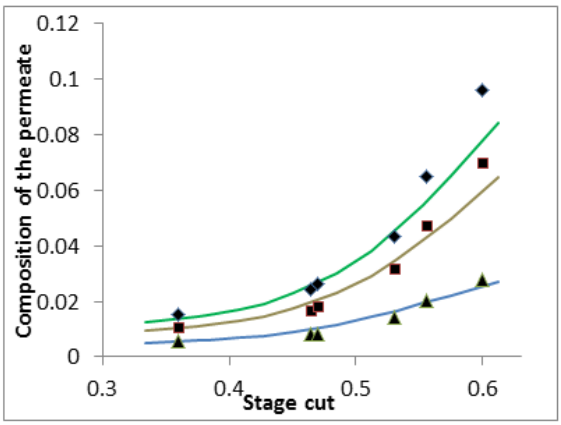

Fig. 3. Calculated concentration of N_2 ◆, Ar▲ and CH_4■ in the permeate compared to the experimental data for co-current model.

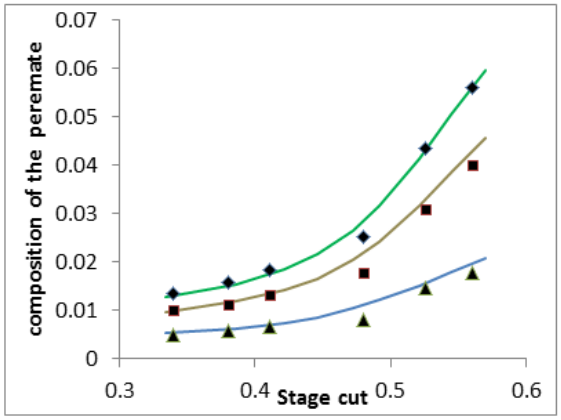

Fig. 4. Calculated concentration of N_2 ◆, Ar▲ and CH_4■ in the permeate compared to the experimental data for counter current model.

As can be seen the presented method demonstrates satisfactory performance over a wide range of stage cuts, with some deviations in higher stage cuts which is normally seen in all of the available models. The proposed approach is a simple yet robust method for solving plug flow problem as the calculations are straight forward, and the pressure drop calculations can easily be incorporated to the solution without the need for further iterations for the co-current flow.

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