An Efficient Numerical Approach for the Separation of Gases Using Membrane in a Multicomponent Gas Mixture

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***Abstract*—An efficient, stable and fastest numerical method is predicted by doing comparison between different numerical methods which are used to solve cross and co-current flow model in multicomponent membrane gas separation. Considering cross and co-current flow model numerical methods such as Bogacki–Shampine method, Dormand–Prince method, Adams-Bashforth-Moulton method, numerical differentiation formulas, modified Rosenbrock formula of order 2, Trapezoidal rule with free interpolant and Trapezoidal rule with backward difference formula of order 2 are observed. To solve cross and co-current flow model stiff and non-stiff numerical methods are implemented and the characteristics of each method are discussed briefly. The stability and computational speed of considered numerical methods are investigated for the selection of best numerical method. The results obtained from recommended numerical method are compared with experimental and numerical results available in literature. The numerical results show good agreement with literature values.**

***Index Terms*—Cross flow, co-current flow, numerical comparison, membrane gas separation.**

I. Introduction

Membrane gas separation occurs is appeared as attractive technology in chemical engineering. The membrane gas separation expose to many industrial and chemical processes, like, e.g. hydrogen recovery, Air separation, natural gas dehydration, etc. A common feature of all those processes is that a gas mixture at a high pressure is fed to the feed side of the membrane, while the permeate gas at a lower pressure is removed from the permeate side of the membrane. An appropriate modeling method to describe membrane gas separation in multicomponent gas mixture has been developed in the late 1950s by the important articles of [1, 2]. The reader is referred to [3, 4] for the detailed overview on mathematical modeling of multicomponent membrane gas separation.It is clear that for correct analysis of this membrane gas separation flow processes, high computational effort is necessary. Maximum mathematical models proposed in the past were comprehensive models are based on ordinary differential equation, whose numerical values generally showed good agreement to experimental results [3-8]. The aim in this article is to explore the influence of different numerical methods on the dynamic and stationary behavior of a membrane gas separation model. The cross and co-current flow process is considered in paper for the comparison of different numerical methods in single membrane gas separation unit.This process will be illustrated by a mathematical model comprising primarily stage cut, membrane area permeation and rejection of component. The paper is prepared along these lines: After a short explanation of the focused cross and co-current model and there main equations, several selected numerical methods for the numerical simulation of the models will be described.The behavior of cross and co-current flow patterns is shown in results. The significance of a correct numerical simulation and the conclusions reached from these studies are described in the last part of the paper.

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II. Model Derivation of Cross and Co-current Flow

Fig. 1 and Fig. 2 described the cross and co-current flow pattern in a single permeation unit. The feed Q^f with specific flow rate enters the unit and is divided into two streams [4]: Q^p on the permeate side and Q^0 leaving on the reject side. These streams have mole fractionsx_i^f,$y_i^p$and x_i^o respectively. Hence

$$\sum_{i=1}^{n} x_i^f = 1 \tag{1}$$

$$\sum_{i=1}^{n} x_i^o = 1 \tag{2}$$

$$\sum_{i=1}^{n} y_i^p = 1 \tag{3}$$

We can write overall mass balance as

$$Q^f = Q^p + Q^o \tag{4}$$

Also, we can write component balance as

$$Q^f x_i^f = Q^p y_i^p + Q^o x_i^o \tag{5}$$

The stage cut is defined as

$$\Phi = \frac{Q^p}{Q^f} = \frac{Q^f - Q^o}{Q^f} \tag{6}$$

The permeabilities of different gases in a specific membrane are arranged in descending order, i.e.

$K1>K2>\ldots>Ki>Ki+1>\ldots>$Kn and dA represents the small differential volume.

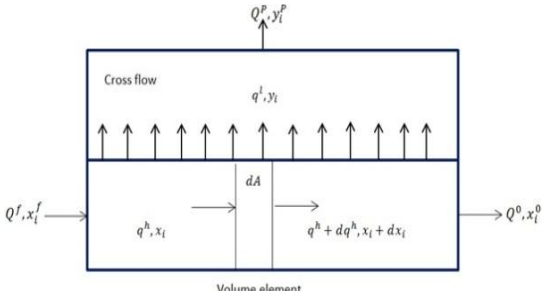

Fig. 1. Cross flow.

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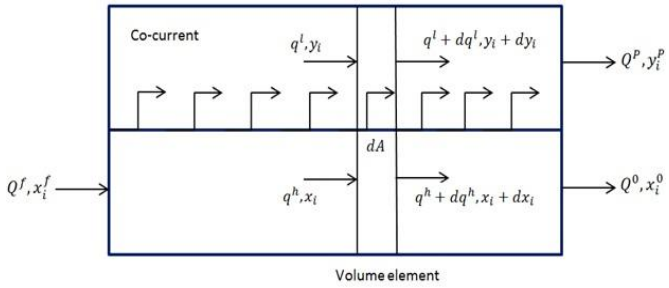

Fig. 2. Co-current flow.

Introducing the dimensionless quantities [4]

$$Pr = P_l/P_h \tag{7}$$

$$\gamma_i = K_i/K_1 \tag{8}$$

$$\bar{q}^h = q^h/Q^f \tag{9}$$

$$\bar{A} = \frac{AK_1P_h}{\delta Q^f} \tag{10}$$

where γ_i separation is factor, $\bar{q}^h$ shows the dimensionless flow rate and $\bar{A}$ represents dimensionless area. Governing equations by using these dimensionless variables appeared as

$$\frac{d\bar{q}^h}{d\bar{A}} = -\sum_{i=1}^{n}[\gamma_i(x_i - Pry_i)] \tag{11}$$

$$\frac{dx_i}{dA} = \frac{x_i\left[\sum_{j=1}^{n}\gamma_j(x_j - Pry_j)\right] - \gamma_i(x_i - Pry_i)}{\bar{q}^h} \tag{12}$$

These equations have the following boundary conditions

$$x_i|_{\bar{A}=0} = x_i^f \tag{13}$$

$$\bar{q}^h|_{\bar{A}=0} = 1 \tag{14}$$

x_i is variable mole fraction.

In the form of dimensionless quantities, the model equations can be written as [4]:

For cross flow

$$y_i = f(x_1, x_2, x_3, \dots, x_n, Pr = f(\{x_i\}, Pr) \tag{15}$$

Co-current flow

$$y_i = \begin{Bmatrix} \frac{x_i^f - \bar{q}^h x_i}{1-\bar{q}^h}, & \bar{q}^h \neq 1 \\ f(\{x_i\}, Pr), & \bar{q}^h = 1 \end{Bmatrix} \tag{16}$$

III. Numerical Methods

On the basis of criteria mentioned in [9] the following, Bogacki–Shampine (BS) method, Dormand–Prince (DP) method, Adams-Bashforth-Moulton (ABM) method, numerical differentiation formulas (NDF), modified Rosenbrock formula of order 2 (MRF2), Trapezoidal rule with free interpolant (TR-FI) and Trapezoidal rule with backward difference formulaof order 2 (TR-BDF2) are selected. These methods are used to solve and compare the coupled ordinary differential equations of cross and co-current flow in a single permeation unit.

A. Solution algorithm

The solution algorithm for membrane gas separation with cross and co-current flow is

- Input: Feed composition (x_i^f), permeabilities of i^{th} component (K_i), membrane thickness (δ), feed flow rate (Q^f), feed pressure (P_h) and permeate pressure (P_l).
- Calculate pressure ratio (Pr) and permeabilities ratio (γ_i) using equations (7) and (8) respectively.
- Calculate y_i (initial) using any above mentioned solver until the equation (3) is satisfied. Use $x_i = x_i^f$ and P=P_h
- Calculate $\frac{d\bar{q}^h}{d\bar{A}}$ and $\frac{dx_i}{dA}$ by with the help of boundary condition given for cross and co-current flow separately. After each step update the value of y_i with the new values ofx_i. Proceed solving based on update values until A=1
- Calculate the mole fraction of i^{th} component in permeate by using equation (15) and (16) for cross flow and co-current flow respectively.
- Finally calculate permeate flow rate (Q^p) and stage cut (Φ) from equation (4) and (6) respectively.

IV. Results and Discussion

The process conditions for the separation of a four component mixture are given [2] and [3] are used to check the stability of various numerical methods. The model of cross and co-current flow is solved by various numerical methods by using step size of 0.1 and tolerance level of 10^{-9}. TableI shows the summarized numerical behavior of all methods in permeate streams. All model shows stable behavior except NDF in reject streams. In permeate stream three numerical methods Dormand–Prince method, ABM method and modified Rosenbrock formula of order 2 show stable behavior in all components of gas stream while other numerical models have inconsistent numerical behavior. Table II shows CPU time elapsed by different numerical methods to solve the cross and co-current flow model for membrane gas separation. ABM method used least time of 0.247755 and 0.354268 seconds to solve the co-current and cross flow flow model respectively while numerical Trapezoidal rule with free interpolant take 0.622862 and 0.877675 seconds respectively to show the slowest behavior.

ABM is observed as most stable and efficient method for the separation of gases through membrane using cross and co-current flow model. To validate the model the results obtained by using ABM model are compared with experimental results and simulation results reported by other researchers.Experimental results reported by Kaldis et al. [10] are used to compare the results of ABM model. A polyimide hollow fiber membrane module is used for the separation of a gas mixture of four components i.e. H_2, CH_4, C_2H_6, and CO_2. The effective membrane area of 10 cm^2 was used. H_2S was used instead of CO_2 in the experiments due to safety considerations and handling problems. It must be stated that

in the Polyimide membranes the permeabilities of CO_2 and H_2S are relatively the same.In the experiment, the pressure value was keep constant at 20 bar and the effect of stage cut on reject and permeate composition was examined by varying feed flow rate from 5 to 30 Nl•hr−1. The reject side acts as a source from which the components are passing through the membrane. So, the permeation level decreases with gradual decrease in reject. Permeate stream comprises H2 and CO2 mostly, and hydrocarbons are present in minor quantity. The stage cut marginally effect their concentrations. 93% of permeate streams is enriched with hydrogen. Carbon dioxide concentrations vary from 6.5 to 9.5% in permeate stream, while methane and ethane total concentration is less than 0.5% in permeate stream. Figs. 3-6 show the reject and permeate mole fraction of hydrogen, carbon dioxide, methane and ethane respectively obtained using above mentioned numerical methods.

TABLE I: NUMERICAL BEHAVIOR OF DIFFERENT METHOD IN PERMEATION STREAM.

Method	Numerical Behavior			
	H_2	N_2	O_2	CH_4
Bogacki–Shampine	Stable	Unstable	Unstable	Unstable
Dormand–Prince	Stable	Stable	Stable	Stable
Adams-Bashforth-Moulton	Stable	Stable	Stable	Stable
Numerical differentiation formulas	Unstable	Unstable	Unstable	Unstable
Modified Rosenbrock formula of order 2	Stable	Stable	Stable	Stable
Trapezoidal rule with free interpolant	Stable	Unstable	Unstable	Stable
TR-BDF2	Unstable	Unstable	Stable	Unstable

TABLE II: TIME ELAPSED BY DIFFERENT NUMERICAL METHODS.

Numerical method	Co-current flow	Cross flow
Bogacki–Shampine	0.258900	0.363276
Dormand–Prince	0.304146	0.437841
Adams-Bashforth-Moulton	0.247755	0.354268
Numerical differentiation formulas	0.622262	0.870155
Modified Rosenbrock formula of order 2	0.502016	0.810065
Trapezoidal rule with free interpolant	0.622284	0.877675
TR-BDF2	0.477841	0.762346

TABLE III: COMPARISON OF MOLE FRACTION OF PERMEATION RESULTS CALCULATED BY ABM MODEL WITH OTHER MODELS AT STAGE CUT = 0.5.

Gas	Co-current flow	Cross flow	S.P Kaldis model [10]	M. Peer model [11]
Hydrogen (H_2)	0.92	0.91	0.93	0.92
Carbon dioxide (CO_2)	0.079	0.074	0.073	0.075
Methane (CH_4)	0.0061	0.0054	0.002066	-
Ethane (C_2H_6)	0.0003	0.0002	0.002066	-

Since the flow rate acts as a driving force to permeate the gas mixture through the membrane. So, with the increase in membrane area, the reject flow decreases and its tendency of permeation decreases. Hydrocarbons methane and ethane are the least permeable components in this case. Their graph shows different variation in comparison with the hydrogen and carbon dioxide. Here the permeate streams going to be increased while the rejected streams are increasing. Since these are the least permeable components so the membrane does not allow them to be passed out. With the continuous permeation hydrogen and carbon dioxide, space produces at the reject side or the reject flow rate is becoming enriched with the least permeable components. At the permeate side, the streams going to be increased slightly, and they can act as minor fractions in the permeate side. The model is used to simulate the membrane gas separation process.. The effect of stage cut on reject and permeate compositions is shown in these Figs. The values of cross and co-current flow calculated by ABM model are approximately over. The concentrations of hydrocarbons are low due to due to their small amounts present in the permeate stream. The results obtained by ABM model are compared with the experimental results reported in literature [10]. Table III shows the comparison of other researcher's model and experimental data with results predicted in this model. We can observe the good agreement between experiment data and model results.

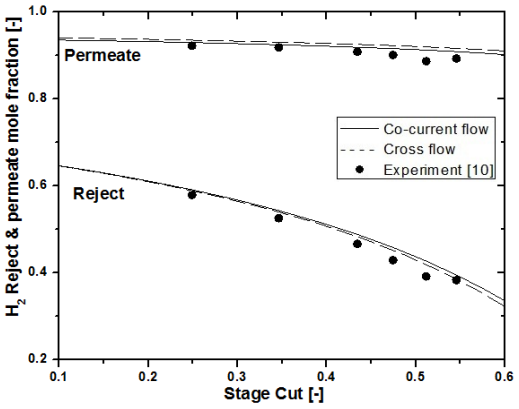

Fig. 3. Effect of stage cut on reject and permeate composition of H_2.

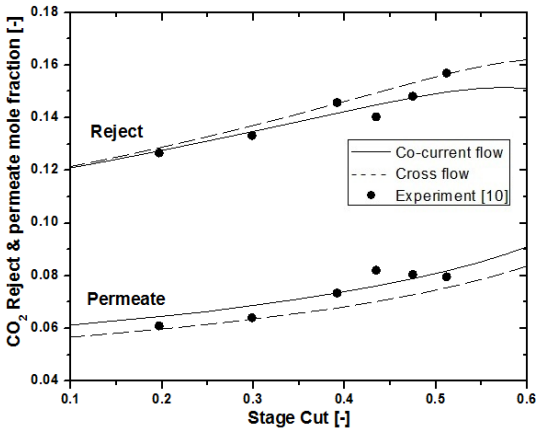

Fig. 4. Effect of stage cut on reject and permeate composition of CO_2.

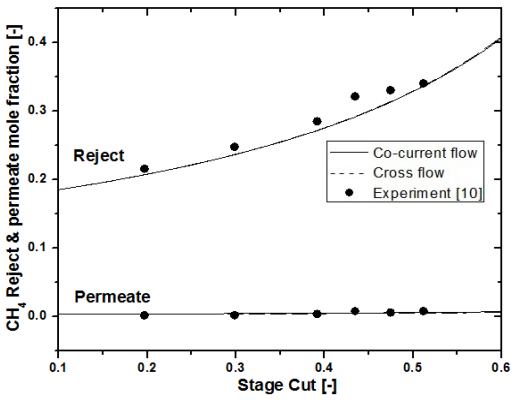

Fig. 5. Effect of stage cut on reject and permeate composition of CH_4.

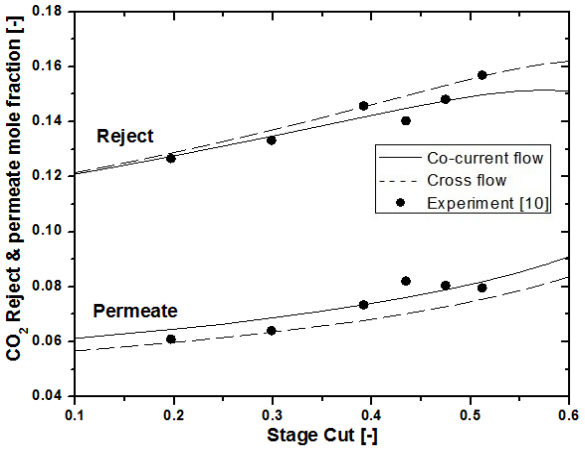

Fig. 6. Effect of stage cut on reject and permeate composition of C_2H_6.

V. Conclusion

The key emphasis of this study is the identification of proper numerical methods for the simulation of cross and co-current flow, since this is of main significance for an precise calculation of permeate and reject composition in a membrane gas separation processes. Therefore, different state of the art numerical methods of solving coupled differential equation with initial value problems are compared. The selected numerical method has an important role in the conclusion of computed behavior of membrane gas separation process. It is essential to have a deep knowledge of physical and chemical properties in order to describe the numerical results in an appropriate way.Several model parameters created problems during the simulation. These problems are fixed by adjusting the step size and tolerance level in MATLAB. Thus it can be summarized, that stable and fast behavior of ABM method can be identified as an efficient numerical method producing correct results and needing an adequate computational effort for the simulation of cross and co-current flow, where permeation composition is one of the targeting phenomena. Future work essentials to be focused on the advance expansion of the model proposed using multiple permeation unit and membrane modules in order to achieve an improved results of permeate and reject composition.

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