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Development of a multicomponent mass transport model for predicting CO_2 separation behavior from its mixture with natural gas and hydrogen using zeolite membranes

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ABSTRACT

CO_2 removal from sour natural gas for the purpose of gas sweetening and from synthesis gas to obtain pure hydrogen is an important process in gas and oil refining industries. In this study, modeling of CO_2 separation from natural gas and synthesize gas by zeolite membrane has been investigated. This model is based on adsorption–diffusion model which uses the modified Maxwell–Stefan formulation for expression of fluxes equations. This model is capable to predict mixture fluxes and mixture separation selectivity using only pure component adsorption and permeation experimental data. The model developed in this study is also a full coupled model considering correctly the kinetic and thermodynamic contribution in component fluxes. The results of modeling indicated that the thermodynamic aspect has a major role to transport through the zeolite membranes and can not be ignored for both components, while the kinetic aspect has a major contribution in transport of strongly adsorbed species (CO_2). Finally the results of fluxes and separation selectivity estimated by the model showed a good agreement with experimental data.

Keywords: Zeolite membrane; CO_2; Hydrogen; Natural gas; Modeling; Maxell-Stefan

1. Introduction

Separation of CO_2 from natural gas and synthesis gas is an important process because CO_2 reduces energy content of natural gas. Development of effective separation technologies for removal of CO_2 from natural gas is still a challenge for researchers. The technology most widely used for CO_2 removal from natural gas is amine absorption, but amine plants are complex and costly [1]. Membrane plants using CO_2-selective cellulose acetate membranes were installed in 1980s, and currently the largest membrane facility for CO_2 removal is 700 million scfd [2]. The main problems that limit the use of polymeric membranes are their poor performance stability at high pressure and in the presence of highly sorbing components [3].

Microporous inorganic membranes have been studied for gas separation due to their superior thermal, mechanical and chemical stability and high pressure resistance compared to conventional polymeric membranes. Zeolite membranes have the potential to eliminate the chemical instability of polymeric membranes against such components. Falconer and co-workers [4–9] used SAPO-34 membranes to separate CO_2 and CH_4 mixtures and found that the highest selectivity for CO_2 was 270 at 253 K [9]. However, the CO_2 selectivity and flux through these membranes were significantly reduced by the presence of water [6] in natural gas. It can be explained knowing that the SAPO-34 zeolite is hydrophilic in nature and has

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a strong affinity to water adsorption. To prevent the negative effect of water adsorption on the purification of natural gas, the zeolite membranes should be hydrophobic.

Some investigators [10–13] have used different techniques to determine the adsorption and permeation properties of CH_4 and CO_2 on silicalite-1 membrane. However, the theoretical aspect of such separations still remains an area for more research due to the complex phenomena occurring within the zeolite membrane matrix which should be explained by a sophisticated model. Bakker et al. [14] proposed a surface-diffusion model in combination with activated gas diffusion to explain the temperature dependence of single-component fluxes (permeances). This model could correctly predict the temperature dependence of the single-component permeation.

In this study of separation process of CO_2–CH_4 mixture by silicalite-1 membrane and CO_2–H_2 mixture by SAPO-34 membrane were investigated. Adsorption isotherms of components mixture on silicalite-1 and SAPO-34 membranes were described with extended Langmuir adsorption model. Matrix of multicomponent diffusion coefficients determined by use of binary diffusion coefficients based on Maxwell–Stefan formulation. Finally, the predicted multicomponent fluxes and selectivities were compared against the experimental data available in the literature.

2. Modeling

2.1. Model structure

The model developed is based on adsorption-diffusion theory which considers a diffusing component is first adsorbed into zeolite micropores surface and then diffuses through it due to the pressure gradient between two sides of zeolite membrane. General equation of Maxwell-Stefan for a ternary system containing permeating components 1 and 2 and the membrane *m*, was used for predicting component fluxes [15]:

$$\begin{pmatrix} N_1 \\ N_2 \end{pmatrix} = -\varepsilon\rho \begin{bmatrix} q_1^{sat} & 0 \\ 0 & q_2^{sat} \end{bmatrix} \begin{bmatrix} B_{11} & B_{12} \\ B_{21} & B_{22} \end{bmatrix}^{-1} \begin{bmatrix} \Gamma_{11} & \Gamma_{12} \\ \Gamma_{21} & \Gamma_{22} \end{bmatrix} \begin{pmatrix} \frac{\Delta\theta_1}{l} \\ \frac{\Delta\theta_2}{l} \end{pmatrix} \tag{1}$$

where [B] is kinetic matrix and [Γ] is thermodynamic matrix, these matrixes are calculated from Eqs. (2) and (3), repectively [16].

$$B_{ii} = \frac{1}{\text{Đ}_{im}} + \sum_{\substack{i=1 \\ j\neq i}}^{n} \frac{\theta_j}{\text{Đ}_{ij}} \qquad B_{ij} = -\frac{\theta_i}{\text{Đ}_{ij}}, \; i \neq j. \tag{2}$$

$$\Gamma_{ij} = \frac{q_i}{p_i} \frac{\partial p_i}{\partial q_j}\bigg|_{T,P,\Sigma} \tag{3}$$

where $\theta_i = q_i / q_i^{sat}$ is the fractional coverage, q_i the coverage, p_i the partial pressure of i and Đ_{im} is MS diffusivity for component *i* in membrane. Eq. (1) is the working transport equation for the multicomponent permeation through the membrane. Consequently, the evaluation of the kinetic (diffusivity) and thermodynamic matrix elements must be the next task.

2.2. Binary diffusivities

Fick's diffusion coefficients, D_{im}, can be calculated by having the adsorption and permeation data from the following equations:

$$\pi = D_{im} \cdot S, \; S = \frac{q}{p}, \qquad D_{im}(m^2 \cdot s^{-1}) = \frac{\Pi \cdot L \; (mol \cdot m \cdot m^{-2} \cdot s^{-1} \cdot Kpa^{-1})}{\frac{q}{p} \cdot \rho\varepsilon \; (mol \cdot m^{-3} \cdot Kpa^{-1})} \tag{4}$$

Then MS binary diffusivity Đ_{im} then can be calculated from the Fick diffusivities as follows:

$$\text{Đ}_{im} = D_{im}\,\Gamma^{-1} = D_{im} \frac{\partial \ln q_i}{\partial \ln p_i} = D_{im} \cdot \frac{p_i}{q_i} \cdot \frac{\partial q_i}{\partial p_i} \tag{5}$$

2.3. Determination of thermodynamic matrix

The elements of multicomponent thermodynamic matrix are defined by Eq. (3). For the ternary system, the elements of thermodynamic matrix can be defined as:

$$\Gamma = \begin{bmatrix} \frac{q_1}{p_1} \cdot \left(\frac{\partial p_1}{\partial q_1}\right) & \frac{q_{sat,2}}{q_{sat,1}} \cdot \frac{q_1}{p_1} \cdot \left(\frac{\partial p_1}{\partial q_2}\right) \\ \frac{q_{sat,1}}{q_{sat,2}} \frac{q_2}{p_2} \cdot \left(\frac{\partial p_2}{\partial q_1}\right) & \frac{q_2}{p_2} \cdot \left(\frac{\partial p_2}{\partial q_2}\right) \end{bmatrix} \tag{6}$$

In order to calculate the elements of thermodynamic matrix, the adsorption isotherm for gas mixture is required. The extended langmuir model usually gives a reasonable approximation for estimating the adsorption isotherm of mixture using only binary adsorption parameters:

$$q_1 = \frac{q_1^{sat} b_1 p_1}{1 + b_1 p_1 + b_2 p} \text{ and } q_2 = \frac{q_2^{sat} b_2 p_2}{1 + b_1 p_1 + b_2 p_2} \quad (7)$$

where q^{sat} and b are pure component adsorption parameters. In order to evaluate the partial derivatives in Eq. (6), it is required to solve Eq. (7) simultaneously to derive an explicit expression for p_1 and p_2 in terms of q_1 and q_2 which results in:

$$p_1 = \frac{q_{sat,2} q_1}{b_1 q_{sat,1} q_{sat,2} - b_1 q_{sat,2} q_1 - b_1 q_{sat,1} q_2},$$
$$p_2 = \frac{q_{sat,1} q_2}{b_2 q_{sat,1} q_{sat,2} - b_2 q_{sat,1} q_2 - b_2 q_{sat,2} q_1} \quad (8)$$

Differentiating Eq. (8) with respect to q_1 and q_2 gives the partial derivatives required in thermodynamic matrix and therefore, the elements of the thermodynamic matrix are defined as:

$$\Gamma_{11} = \frac{q_1}{p_1} \frac{q_{sat,1} q_{sat,2}^2 b_1 - q_{sat,1} q_{sat,2} b_1 q_2}{(b_1 q_{sat,1} q_{sat,2} - b_1 q_{sat,2} q_1 - b_1 q_{sat,1} q_2)^2} \quad (9)$$

$$\Gamma_{12} = \frac{q_{sat,2}}{q_{sat,1}} \cdot \frac{q_1}{p_1} \cdot \frac{q_{sat,1} q_{sat,2} b_1 q_1}{(b_1 q_{sat,1} q_{sat,2} - b_1 q_{sat,2} q_1 - b_1 q_{sat,1} q_2)^2} \quad (10)$$

$$\Gamma_{21} = \frac{q_{sat,1}}{q_{sat,2}} \cdot \frac{q_2}{p_2} \cdot \frac{q_{sat,1} q_{sat,2} b_2 q_2}{(b_2 q_{sat,1} q_{sat,2} - b_2 q_{sat,1} q_2 - b_2 q_{sat,2} q_1)^2} \quad (11)$$

$$\Gamma_{22} = \frac{q_2}{p_2} \cdot \frac{q_{sat,1}^2 q_{sat,2} b_2 - q_{sat,1} q_{sat,2} b_2 q_1}{b_2 q_{sat,1} q_{sat,2} - b_2 q_{sat,1} q_2 - b_2 q_{sat,2} q_1{}^2} \quad (12)$$

2.4. Determination kinetic matrix elements

A main advantage of Maxwell-Stefan approach for describing multicomponent transport is that the binary diffusion coefficients determined from pure component permeation experiments retain their physical significance and can be directly used in the description of multicomponent diffusion process. The elements of kinetic matrix were introduced by Eq. (2). For the system under study, components 1 and 2 refer to carbon dioxide and methane, respectively, and m refers to the membrane.

The MS diffusivity for the binary system of component-membrane, Đ_{im}, were calculated from Eq. (5) whereas to calculate component-component binary MS diffusivity, Đ_{12}, Eq. (14) proposed originally by Krishna [17] was used.

$$\text{Đ}_{12} = \text{Đ}_1^{\theta_1/(\theta_1+\theta_2)} . \text{Đ}_2^{\theta_2/(\theta_1+\theta_2)} \quad (13)$$

3. Results and discussions

The generic model was developed which is applicable to all gas separation processes using a zeolite membrane. The data required for the model application are only the pure component adsorption and permeation values. With such data on hand, it is possible to predict the performance of the membrane for mixed gas permeation including the flux of individual components and separation factors.

3.1. Model results for CO_2–CH_4 mixture separation

3.1.1. Single component adsorption

Adsorption step was described by the Langmuir model. The Langmuir type adsorption is considered to arise from sorption into zeolite micropores where chain mobility and packing are highly restricted. The amount of loading (microvoid occupancy) for different adsorbing molecules is different. Weidong zhu et al. [18] reported the adsorption isotherms data of the pure components CO_2(1) and CH_4(2) on silicalite-1 crystals at the temperature range of 303–408 K. The experimental adsorption data were fitted by the Langmuir adsorption model:

$$\theta_i = \frac{q_i}{q_i^{sat}} = \frac{b_i P}{1 + b_i P} \quad (14)$$

Figs. 1 (a) and (b) show the experimental adsorption data and the fitted model isotherm for CO_2 and CH_4, respectively.

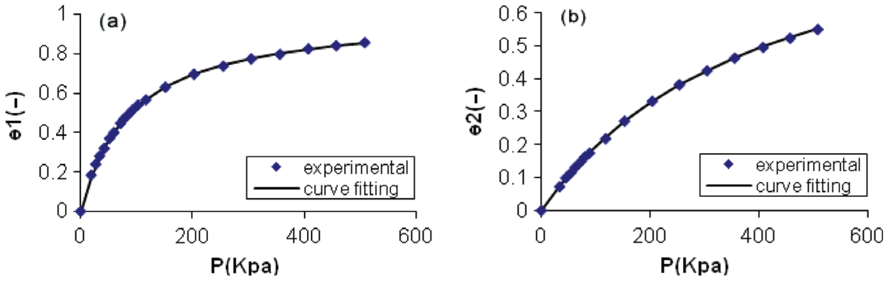

Fig. 1. (a) Adsorption isotherm of pure CO_2(1), (b) Adsorption isotherm of pure CH_4(2), through the silicalite-1 membrane as a function of the total feed pressure at 303 K [18].

The model parameters recovered from a nonlinear fit to the pure components adsorption data, were listed in Table 1. The extracted saturation capacity for CO_2 is higher than that of CH_4 which indicates a stronger affinity between CO_2 and silicalite-1 sorbent.

3.1.2. Extent of kinetic coupling

Weidong zhu et al. [18] also reported single component flux and permeance of CH_4 and CO_2 through silicalite-1 membrane as a function of feed pressure. With the values of the parameters ($\varepsilon = 0.35$, $L = 2 \times 10^{-5}$ and $\rho = 1760$ kg m^{-3}), the binary diffusivities were calculated using Eqs. (4) and (5). Then the elements of kinetic matrix were calculated by Eq. (2). Table 2 compares cross terms values in kinetic matrix with the main term values. The results obtained from developed model showed that the value of the cross-element for component 1(CO_2) is about 35% of the main term, and for component 2(CH_4) is only 7% of the main term. The results indicated that kinetic coupling has a major contribution in transport, in special, for more strongly adsorbed component, CO_2.

Table 1
Estimated parameter values obtained from fitting of the adsorption data for silicalite-1 by the Langmuir model at 303K

No	Component	q^{sat} = (*mol·kgmembrane*$^{-1}$)	b(*Kpa*$^{-1}$)
1	CO_2	3.0569	11.42×10^{-3}
2	CH_4	2.7343	2.42×10^{-3}

Table 2
Average values of the main and cross-terms in kinetic matrix for 50/50 mixture of CO_2/ CH_4 through the silicalite-1 membrane at 303 K

p_i(*Kpa*)	θ_1	θ_2	$B_{11} \times 10^9$	$B_{12} \times 10^9$	$B_{21} \times 10^9$	$B_{22} \times 10^9$
50	0.3375	0.0715	0.227	0.067	0.014	0.229
75	0.4203	0.0891	0.207	0.074	0.015	0.211
100	0.479	0.1015	0.188	0.074	0.016	0.196
125	0.5229	0.1108	0.171	0.073	0.016	0.184
150	0.5569	0.118	0.157	0.071	0.015	0.173

3.1.3. Extent of thermodynamic coupling

The investigation using the model developed shows that the thermodynamic coupling has also a major contribution to transport through the membrane. Figs. 2(a) and (2b) show the significance of the thermodynamic cross-terms in comparison with the main terms. In general, the results obtained from the model indicated that the flux of both components can be thermodynamically affected by the presence of the other component.

3.1.4. Prediction of mixture permeation and selectivity

The model developed in this study can be validated by the prediction of mixture permeation fluxes. Fig. 3(a) shows a comparison between the experimental data with the model predicted values. Experimentally, both components fluxes slightly increase with pressure, and the model predicts the same trend. The permeation selectivity for CO_2 over CH_4 at different pressures was calculated using the Eqs. (1) and (15). Fig. 3(b) also shows the predicted mixture selectivity at 303 K slightly decreases with total feed pressure which is in agreement with the experimental data.

$$\alpha_{CO_2} = \frac{N_1}{N_2} \tag{15}$$

The model predictions were almost in good agreement with the experimental data for CO_2(1) and CH_4(2) .

3.2. Model results for CO_2–H_2 mixture separation

3.2.1. Single component adsorption

Mei Hong et al. [19] reported the adsorption isotherms data of the pure components CO_2(1) and H_2(2) on SAPO-34 membrane at the 295 K. The experimental adsorption data were fitted by the Langmuir adsorption model (Fig. 4 (a) and (b)).

By fitting of Langmuir adsorption model with pure components experimental data, the parameters of model were determined and listed in Table 3.

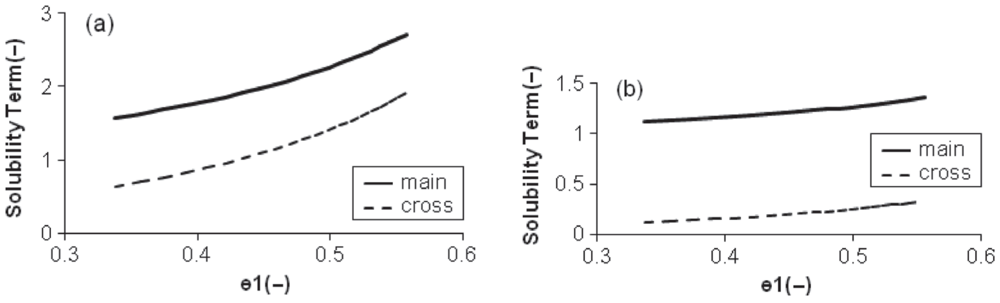

Fig. 2. Values of the main and cross-terms in thermodynamic matrix for 50/50 mixture of CO_2/CH_4 through the silicalite-1 membrane at 303 K for, (a) CO_2, (b) CH_4.

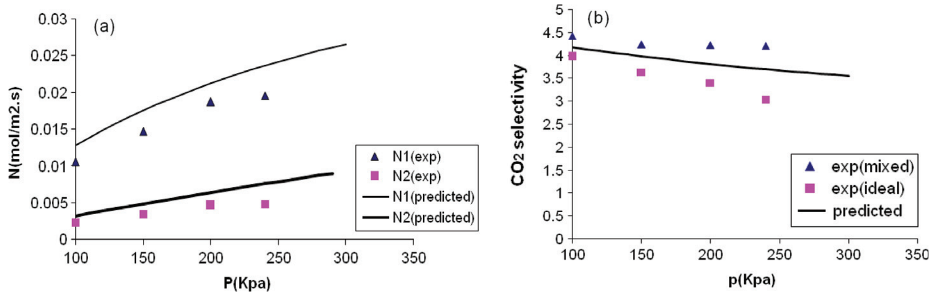

Fig. 3. (a) Comparison of experimental and model predicted fluxes of CO_2(1) and CH_4(2) in mixture through the silicalite-1 membrane as a function of the total feed pressure at 303 K [18], (b) Mixture permeation selectivity for CO_2 over CH_4.

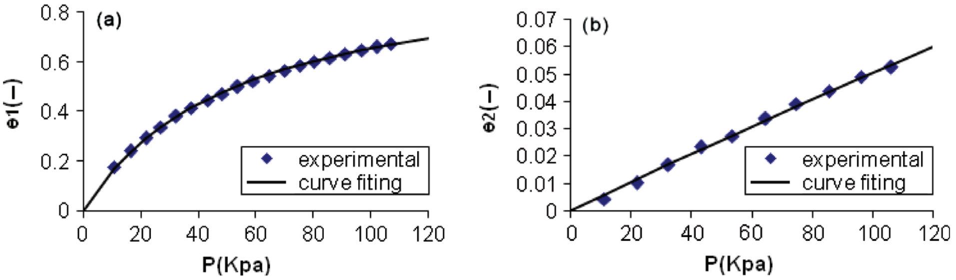

Fig. 4. (a) Adsorption isotherm of pure components on the SAPO-34 membrane as a function of the total feed pressure at 295 K, (a) pure CO_2(1), (b) pure H_2(2), [19].

Table 3
Estimated parameter values obtained from fitting of the adsorption data for SAPO-34 membrane with the Langmuir model at 295K

No	Component	q^{sat} = ($mol \cdot kgmembrane^{-1}$)	$b(Kpa^{-1})$
1	CO_2	18.8×10^{-3}	5.279
2	H_2	0.53×10^{-3}	0.731

Table 4
Average values of the main and cross-terms in kinetic matrix for 43/57 mixture of CO_2/H_2 at 295 K

$B_{22} \times 10^{11}$	$B_{21} \times 10^{11}$	$B_{12} \times 10^{11}$	$B_{11} \times 10^{11}$	$p_i(Kpa)$
1.55	0.05	1.47	2.63	86
1.90	0.07	1.83	2.78	172
2.29	0.08	2.23	3.25	258
2.68	0.10	2.61	3.79	344
3.57	0.13	3.51	5.24	559

3.2.2. Extent of kinetic coupling

Mei Hong et al. [19] also reported single component permeance of H_2 and CO_2 as a function of feed pressure. As previously mentioned the elements of kinetic matrix were calculated by Eq. (2). Table 4 compares cross terms values in kinetic matrix with the main term values. The results obtained from developed model revealed that the value of the cross-element for component 1(CO_2) is about 55% of the main term, and for component 2 (H_2) is about 3% of the main term. This table showed kinetic coupling has a major contribution in CO_2 transport, while plays a marginal rule in H_2 transport.

3.2.3. Extent of thermodynamic coupling

Figs. 5(a) and 5(b) shows the significance of the thermodynamic cross-terms in comparison with the main terms. The results obtained from the examination of the developed model for this system shows that the thermodynamic coupling has a major contribution to transport through the membrane and the flux of both components can be thermodynamically affected by the presence of the other component. However, this effect is higher for CO_2 which is more strongly adsorbed component.

3.2.4. Prediction of mixture permeation and selectivity

By using of Eq. (1), molar flux of CO_2 and H_2 in mixture was calculated. Permeances from flowing equation can be calculated as:

$$\Pi_i = \frac{N_i}{\Delta p_i} \qquad (16)$$

The permeation selectivities were calculated using the Eqs. (1) and (15). Since the experimental data for the

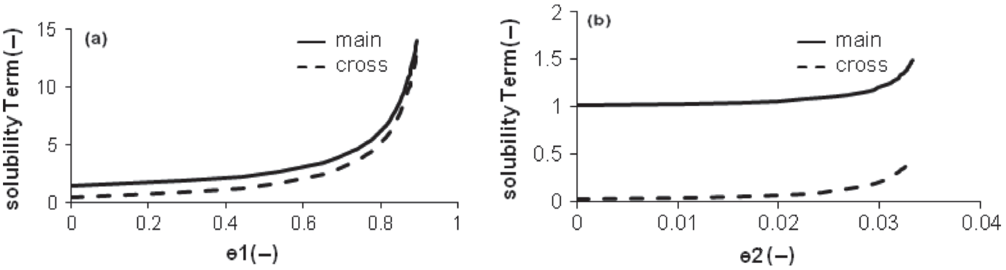

Fig. 5. (a) Values of the main and cross-terms in thermodynamic matrix for 43/57 mixture of CO_2/H_2 on SAPO-34 membrane at 295 K for (a) CO_2, (b) H_2.

Table 5
Comparison of experimental and model predicted permeances and separation selectivity of $CO_2(1)$ and $H_2(2)$ in mixture through the SAPO-34 membrane at 1600 Kpa pressure and 295 K [19]

Predicted	Experimental data	P = 1600Kpa
3.72×10^{-8}	3.39×10^{-8}	CO_2 Permeance $(mol \cdot m^{-2} \cdot s^{-1} \cdot pa^{-1})$
1.31×10^{-9}	1.57×10^{-9}	H_2 Permeance $(mol \cdot m^{-2} \cdot s^{-1} \cdot pa^{-1})$
22.61	21.59	CO_2/H_2 Separation selectivity(-)

mixture was available only in single pressure of 1600 Kpa, It was used to validate the model. Table 5 shows the predicted permeance and separation selectivity are in agreement with the experimental data.

4. Conclusion

In this study a general fully coupled model was introduced for the removal of CO_2 from its mixture with methane and hydrogen. This model was capable to predict mixture fluxes and separation selectivity using only pure component adsorption and permeation experimental data. The results for fluxes and separation selectivity estimated by the model provided a good agreement with experimental data. Also the results of this study indicated that the thermodynamic coupling has a major role to transport through the zeolite membranes and can not be ignored for both components while the kinetic coupling has a major contribution in transport mainly for strongly adsorbed component, i.e., CO_2.

Symbols

b — adsorption equilibrium constant (Pa^{-1})
B — kinetic correction factor (-)
D — Fick's diffusion coefficient ($\mathrm{m}^2 \cdot \mathrm{s}^{-1}$)
$Đ$ — Maxwell–Stefan diffusivity ($\mathrm{m}^2 \cdot \mathrm{s}^{-1}$)
l — thickness of the membrane (m)
N — Mole flux ($\mathrm{mol\ m}^{-2} \cdot \mathrm{s}^{-1}$)
p — pressure (Pa)
q — amount adsorbed ($\mathrm{mol} \cdot \mathrm{kgmembrane}^{-1}$)
q^{sat} — saturation amount adsorbed ($\mathrm{mol} \cdot \mathrm{kg}^{-1}$)

Greek symbols

α — separation selectivity (-)
ε — porosity in the support layer (-)
θ — fractional coverage (-)
ρ — zeolite density (kgm^{-3})
Γ — thermodynamic correction factor (-)

Subscripts

1, 2 — components 1, 2
i, j — components i, j
m — membrane
t — total

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