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Diffusion patterns of gas flow through clay alumina supported DDR Zeolite Membrane

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Abstract

Generation of clean hydrogen energy involves separation of H_2 from CO_2 mixture by a Deca Dodecasil Rhombohedral (DDR) zeolite membrane. Permeance, permeability coefficient and selectivity of such a membrane contribute to the membrane efficiency. These parameters are usually controlled by the gas transport properties of the membrane which is directly related to diffusion mechanisms of gaseous components through the membrane pores. In this study, transport phenomena namely the Viscous, Knudsen and Molecular Sieving have been evaluated to analyze the contribution of each flux to the individual H_2 and CO_2 and their gas mixture through the zeolite membrane. To study the flow properties of the supported membrane, a simple, two-parameter, steady state model was formulated based on the analytical solution of the dusty gas model for both H_2 and CO_2. Maximum percentage deviation between the experimental data and the model predictions was 6%. This reasonably low value validates the proposed model. By and large good agreement between calculated and experimental fluxes confirms the computational and theoretical premises of the study.

Introduction

The global demand for clean and efficient energy has resulted in increased concern for hydrogen economy as a potential long term solution to the energy crisis. Hydrogen has been identified as the next generation clean fuel [1,2]. However, it is not available in nature in pure form. Hydrogen can be produced from SMR process which primarily consists of highly endothermic reaction followed by the water–gas shift reaction [3].

$$CH_4 + H_2O \rightarrow CO + 3H_2 \ (\Delta Hr = -41 \ KJ/mol) \ (i)$$

$$CO + H_2O \rightarrow CO_2 + H_2 \ (ii)$$
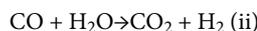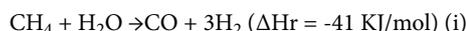
This leads finally to the formation of a mixture of H_2, CO_2, and subsequently H_2 is removed from the CO_2 dominated reaction mixture. Among many processes, membrane based separation process has currently be an emerging technique for purification of H_2. Because of their high thermal and chemical stabilities, inorganic micro porous membranes also serve good for high temperature H_2 separation. Inorganic membranes especially dense palladium and silica membranes are being used for separation of H_2 and CO_2 and other gases [4-8]. Deca-dodecasil 3R (DDR) framework, with pore aperture of 0.36×0.43 nm has been reported to be an excellent membrane material for H_2 separation from its mixture with light gases like CH_4, CO_2, N_2 etc. [9-11].

After the pioneering work of Gies et al. [12,13] on the synthesis of DDR zeolite network, different studies were reported on the process and sol feedstock optimization in order to improve the physico-chemical properties of the same [14,15]. Irrespective of several efforts, the time required for the synthesis of DDR membranes typically varies in the range of 25 to 42 days. Recently, Bose et al. [16] have reported a sonication mediated hydrothermal synthesis method with much reduced processing time. Regarding the performance characteristics of DDR membrane, a huge volume of work is available in literature, not only for H_2 separation but also for other light gases, which include CO_2/CH_4, propane/ propene, CO_2/N_2 etc. [17]. H_2/CO_2 perm selectivity, as high as 33 was reported in case of DDR membranes, structurally modified by chemical vapour deposition of tetraethylorthosilicate [18]. In another report, the variation of selectivity for different gas mixtures, like H_2/CO, CO_2/CO etc. have been studied a wide temperature range of 303-773 K using an all-silica, tubular DDR membrane [19]. Corresponding selectivity values were always recorded to be below five, which is much lower that their ideal counterpart. Performance of a rapidly synthesized DDR membrane has been also reported in case of H_2 separation from a HI decomposition mixture [20]. Related corrosion studies have also confirmed the stability of the DDR membrane under HI environment. Recently, in a review article [21], Muhammad et al. have summarized the challenges and future direction of research with DDR membranes in reference to CO_2 separation from natural gas mixture. It is suggested to explore the microwave-assisted synthesis of DDR zeolite in order to reduce the abnormally long processing time.

Different mathematical models of membrane based gas separation processes are primarily focused in finding the diffusivities of different light gases, both in mixed as well as in pure state. Maxwell-Stefan (MS) equation has been established to be a feasible approach in this regard [22,23]. In literature [24,25], the efficacy of MS equation in predicting the permeate fluxes of CO_2/CH_4, CO_2/N_2 and N_2/CH_4 gas mixtures across a SAPO-34 membrane has been reported. They have observed a strong dependence of MS diffusivities on the respective gas loading, which was further characterized through the Reid-Ehrlich parameter.

Among various types of inorganic membrane, zeolite coated membranes offer the potential of a breakthrough development which obviates several advantages.

Zeolite is a special class of porous inorganic materials with well-defined inter-crystalline pores. They have superior characteristics of thermal, mechanical, chemical and high pressure stability. Small pore zeolite membranes can separate light gases based on molecular sieving effects. Decadodecasil 3R (DDR) framework, with a pore diameter 0.36×0.43 nm, is considered as small pore sized zeolite. DDR Zeolite contains cylindrical pores having diameter of 0.36×0.43 nm. The study on mechanism of separation processes both experimentally and computationally is an important area for evaluation of the performance of the membrane.

In this paper, the contribution of different flux regimes on the gas mixture of H_2 and CO_2 transported through the alumina supported DDR zeolite membrane has been assessed. The transport mechanism of the gas transport in mass transfer boundary layer for rectangular geometry. The contribution of different fluxes are computed to estimate the amount of net flux of H_2 gas molecules as the permeate component. Established physical formalisms of gaseous flow through porous medium and semi-empirical formulations have been utilized in the computational scheme.

Experimental

Materials

The chemical reagents used are colloidal silica, structure directing agent 1-admantanamine and ethylene diamine. Colloidal silica (Ludox HS -30 Sigma) is procured from Aldrich, India. Structure directing agent 1-admantanamine is procured from Aldrich, India and also ethylene diamine is procured from Merck, India. The gas cylinders which includes for CO_2 and H_2, purchased from BOC scientific, India.

Synthesis of DDR zeolite membrane

Figure 1 depicts the flowchart for the synthesis of the DDR zeolite membrane. The molar composition of the sol used for the synthesis was silica: 1-adamantanamine: ethylene diamine: water = 1:0.5:4:100. Two reactant mixtures were prepared respectively by suspending measured amount of colloidal silica and deionised water (DI water) in a glass beaker (mixture-1). The solution was stirred with a magnetic stirrer (SCHOTT Instruments GmbH, Germany) at 200 rpm for 30 minutes. In another mixture (mixture-2), 1- adamantanamine was mixed with ethylene diamine and the calculated amount of water was added. All these procedures were carried out at room temperature (30). The resulting mixture was sonicated for 3 hours. The support substrate was seeded with DDR zeolite seed crystals and dried at 100 for 24 hours. The seeded substrates were vertically placed in an autoclave. The autoclave was filled with the reaction mixture having the above mentioned composition and same synthesis method. After synthesis, the zeolite coated membrane was washed thoroughly with deionized water until the pH of the washing liquid became neutral. The synthesized membrane was calcined in air at 650 for 5 hours to remove the organic compounds. Indigenous clay alumina tube of outer diameter 10 mm, Inner diameter 7 mm, and thickness 3 mm was used as support for synthesis of the DDR membrane.
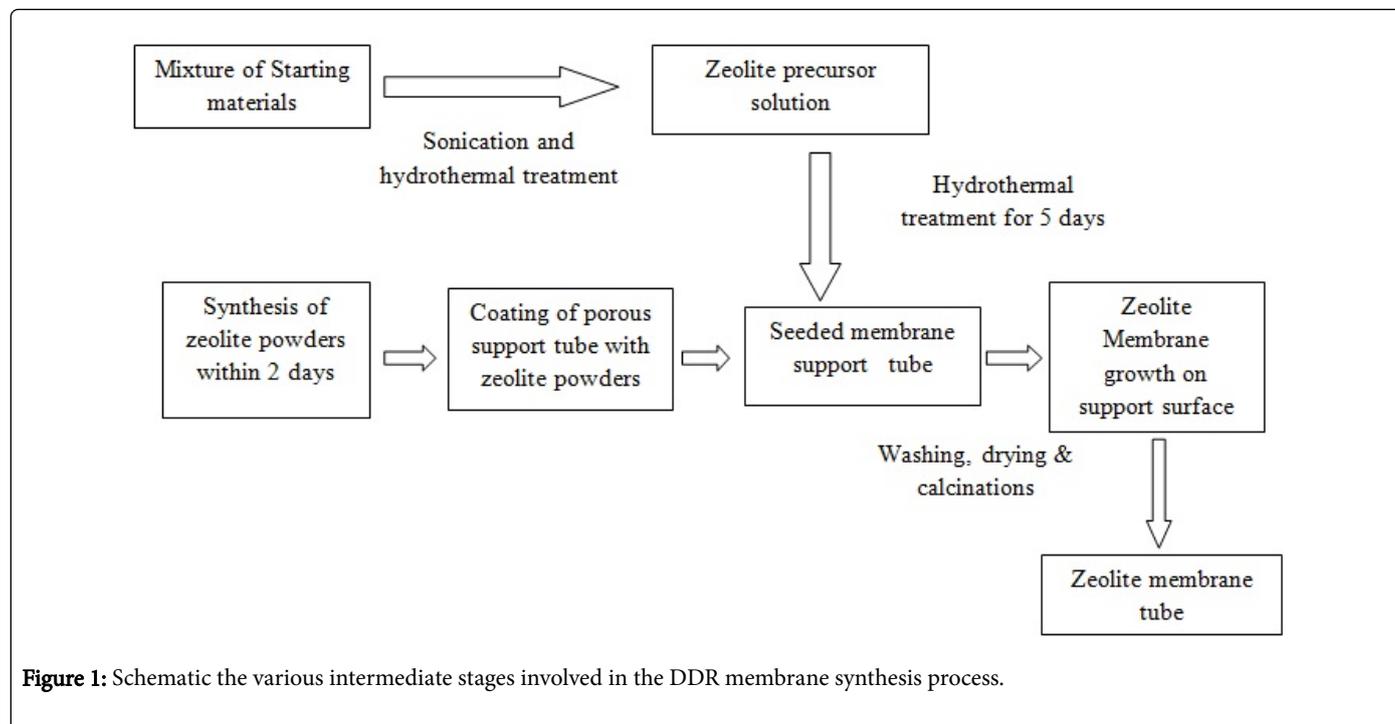
Figure 1: Schematic the various intermediate stages involved in the DDR membrane synthesis process.

Gas permeation experiment

Single gas and gas mixture permeation were measured by a specially designed permeation bench maintained developed in our laboratory. The DDR membrane was encapsulated in a SS membrane holder fitted with a single feed inlet along with two separate outlets, one for retentate and another permeate stream. The set up was completely sealed between two silicon O-rings to avoid gas leakage. As shown in Figure 2, the feed gas was delivered to the module through individual mass flow controllers fitted in the feed lines of both the gas cylinders.

For mixed gas experiments, the gases were premixed in a cyclone type gas mixer located in the immediate upstream position relative to the membrane module. The retentate and the permeate flow rates were measured by two soap bubble flow meters (Zentech, India). Additionally, the gas compositions of both the streams were determined by a gas chromatograph (Model–Trace GC ultra, serial no: 20092814, Thermo scientific, Germany).
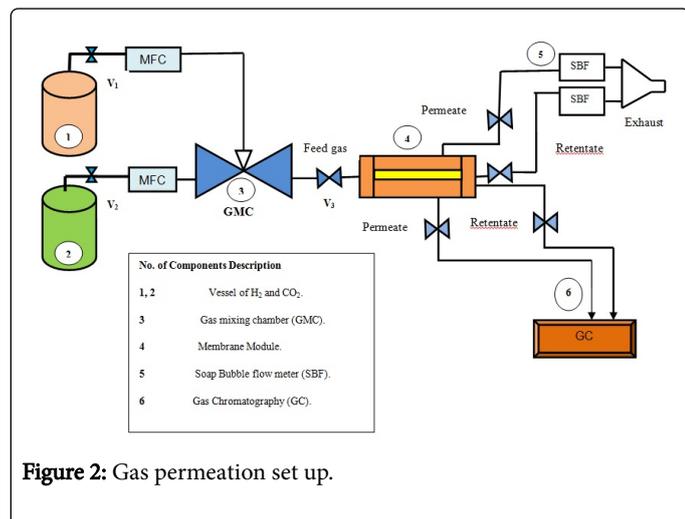
Figure 2: Gas permeation set up.

Theoretical model development

It is the most generalized approach of developing the diffusion patterns through DDR zeolite coated ceramic supported membrane. Figure 3 represents the schematic of feed, permeate and retentate flow directions of the DDR zeolite membrane along with its structural dimensions. In the present study, DDR zeolite coated ceramic membrane module consists of two layers, namely (1) zeolitic layer (2) Non zeolitic layer. The zeolitic pores, that are smaller and the relatively bigger non zeolitic pores. Different transport mechanisms are able to contribute to the total flux through the zeolite membrane such as activated diffusion through zeolitic crystals and Knudsen and viscous mechanisms through non zeolitic pores. Within the non zeolitic pores, the flux is regulated by the thermal vibration of gas molecules that impinge on the pore walls with the result that the gas molecules either diffusion or get sieved to the pore wall. In general, viscous mechanism dominates when the membranes contain defects or relatively larger pores. When the stream of (H_2-CO_2) gas mixture strikes zeolitic surface, the molecular size of carrier gas being comparable to zeolitic pore size are considered to be unable to escape from the force field of pore wall and consequently get sieved by the zeolitic pores. While the H_2 with a relatively smaller molecular diameter compared to that CO_2 mostly passes through the zeolitic pores.

The main objective of the proposed model is to simulate the steady state variations of the major process variables, namely (1) Total permeate flux of the gas component (2) Knudsen diffusivity of individual gases (3) Mean defect radius of the pore size distribution (4) Defect flux of the individual component of gas (5) zeolitic flux under different parametric conditions. Therefore simultaneous equations are to be necessarily formulated. The assumption of the model formulation as follows
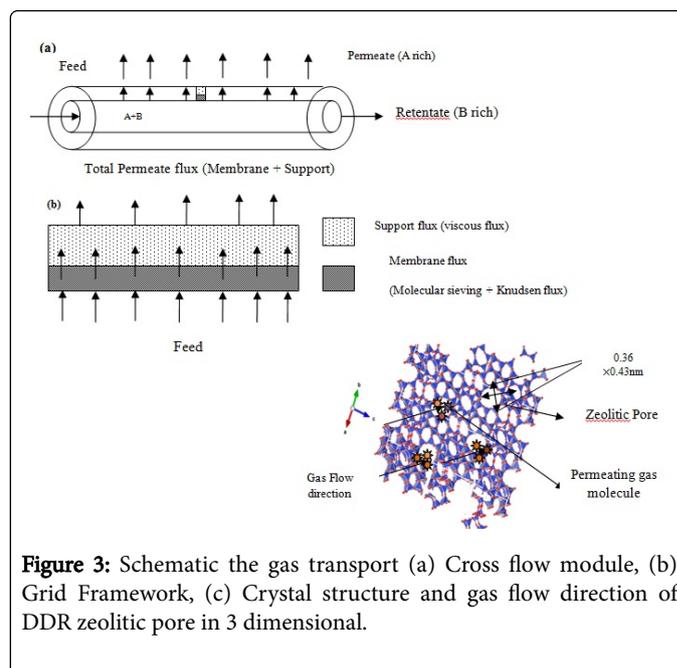
Figure 3: Schematic the gas transport (a) Cross flow module, (b) Grid Framework, (c) Crystal structure and gas flow direction of DDR zeolitic pore in 3 dimensional.

The flow is assumed to be linear as the Reynolds no was evaluated to be less as 32.

Negligible pressure drop from feed to the retentate side. This may be justified in terms of the respective pressure drop according to Hagen-Poiseuille equation. The calculation indicates that the feed to retentate pressure drop is less than 5% of the TMP.

- Isothermal operation.
- Variation of membrane surface and permeate concentrations over length of the module have been neglected.
- Variation of axial feed velocity with radial coordinate has been neglected.
- Axial diffusion of species has been neglected.
- Diffusion through the support layer has been neglected with the comparison of membrane layer diffusion.

Steady state permeates gaseous flux: The molar permeate flux of individual component species may be expressed as a variation of transmembrane pressure difference as:

$$N_i(Total) = \frac{1}{\tau}\int_{(r_i > r_z)}^{\infty} N_i n k(r_i) a_i(r_i) \eta(r_i) dr_i + N_i v + N_i z \qquad (1)$$

Where, $N_{link}(r_i)$ is the permeation flux through a non zeolitic pore radius. r_i which is larger than the zeolitic pore. Here $a_i(r_i)$ is the cross sectional area of each pore radii. Gas transport through non zeolitic pores is governed by Knudsen and viscous flow mechanisms. The dusty gas model has been widely used to describe gas phase transport in many different porous systems. This model, which describes mass transport in multicomponent systems, can be simplified for simple gas flow as a linear combination and viscous contribution [26]. Generally Knudsen flow plays an important role in H_2/CO_2 separation in membranes with pore sizes higher than 0.5 nm.

If we assume one dimensional flow, Knudsen flux through a porous medium is expressed as:

$$N_{ink} = -\frac{1}{R_g T} D_{ikn} \frac{dP_i}{dx} \qquad (2)$$

Where, D_{ikn} is the Knudsen diffusivity and is described as:

$$D_{ikn} = \frac{4}{3} M_0 \sqrt{\frac{8000 R_g T}{\pi M_W}}$$

Where, M_0 is Knudsen structural parameter. If we assume the pores are cylindrical open ended and non-intersecting.

Knudsen diffusion would be expressed as:

$$D_{ikn} = \frac{2}{3} r_i \sqrt{\frac{8000 R_g T}{\pi M_W}} = 97 r_i \sqrt{\frac{T}{M_W}} \qquad (4)$$

$\langle r_i \rangle$ is the integral mean defect radius of pore size distribution. The equation becomes:

$$r_i = \int_{r_{imin}}^{r_{imax}} r_i \eta(r_i) dr_i \qquad (5)$$

Where, R_g is the gas constant and T is absolute temperature. M_W is the molecular weight of the individual gas. The rationale is related to the fact that the Knudsen diffusivity is much higher than zeolitic crystal diffusivities. The equation becomes that:

$$N_{ink} = \int_{r_i > r_z}^{\infty} N_{ink}(r_i) a_i(r_i) \eta(r_i) dr_i \qquad (6)$$

Where, $\eta(r_i)$ is the pore size distribution. It can be represented as:

$$\eta(r_i) = \frac{a_i(r_i) n(r_i)}{A_i} \times n(r_i)$$

is number of the non zeolitic pores which are present to the selective layer. A_i is the total area is related to the zeolitic layer and non zeolitic layer.

Knudsen number: $(Kn = \frac{\lambda}{r})$

Indicates, whether Knudsen diffusion and viscous flow dominates. For K_n numbers between 0.01 to 10, both Knudsen and viscous flow dominates [27]. Viscous flow is dominant when the pore size of the membrane is larger relative to the zeolite pore. In the case, molecules can easily pass through the membrane without any serious interactions to the pore wall. However, it is not being considered as a selective flow in zeolite membrane.

If we assume one dimensional flow, viscous flux is expressed as:

$$N_{iv} = -\frac{1}{R_g T} \frac{B_0 P_m}{\mu} \frac{dP_i}{dx} \qquad (7)$$

Where, B_0 and P_m are the permeability constant and mean pressure between feed and permeate side respectively. Assuming non intersecting cylindrical pores B_0 can be defined as the following

$$B_o = \frac{r_i^2}{8} \qquad (8)$$

Based on the dusty gas model through non zeolitic pores. It can be expressed as a combination of Knudsen and viscous flux

$$N_{inz} = -\frac{1}{R_g T} \left[D_{ikn} \frac{dP_i}{dx} + \frac{B_o P_m}{\mu} \frac{dP_i}{dx} \right] \qquad (9)$$

μ is the viscosity and is calculated based on 'Wilke model' [28]. Wilke model is an empirical correlations derived from simple kinetic theory that might be easily extended to multi component systems. By using above mentioned equations and integrating over membrane thickness, we can easily show that non zeolitic flux can be expressed as below

$$N_{inz} = N_{ink} + N_{inv} = \frac{1}{\Delta x} 97(r_i) \sqrt{\frac{T}{M_W}} \frac{\Delta P_i}{R_g T} + \frac{1}{\Delta x} \frac{r_i^2 P_m}{8\mu} \frac{\Delta P}{R_g T} \qquad (10)$$

Where, the first and second terms represent Knudsen and viscous flows respectively. We have assumed cylindrical and non-intersecting pore in the model. However, in reality they are not perfect. It is also assumed that the dominant part of the non zeoltic gas transport happens in free transport process of cylindrical shape with radii. The molar permeate flux through zeolitic crystals is often referred to as zeolitic diffusion. It is independent of transmembrane pressure difference. Macroscopic and molecular dynamics are the two major methods used to describe diffusion mechanism. Based on our experimental data, macroscopic model is the best choice to describe the diffusion through zeolite crystals. In this study, we have used macroscopic model (multiscale model) to describe mass transfer through zeolite membranes. Intra crystalline mass transfer through zeolite occurs with a combination of diffusion and sieving mechanisms. Thus, total flux through the membrane can be estimated as:

$$N_{iTotal} = \beta N_{iz} + (1 - \beta) N_{inz} \qquad (11)$$

Where, $\beta = A_Z / A_t \times (1-\beta)$ is the fraction of cross sectional area that corresponds to the defects or non-zeolite pores. A_Z and A_t are the zeolite open area and total permeable area of the membrane respectively. In this paper, the flux has been simulated by considering the Knudsen diffusion, viscous flow through the non-zeolitic pores of membrane and zeolitic diffusion. We have considered the behaviour of the gas molecule is incompressible. So that condensation permeation mechanism is ignored in this study because the vapour pressure for light gases H_2 and CO_2 considered in the study decreases in the pores and is not significant enough to cause condensation.

Results and Discussion

Simulation algorithm

Equation:1 constitutes four major equations among them for the mentioned process variables, namely (1) Total permeate flux of the gas component (2) Knudsen diffusivity of individual gases (3) Mean defect radius of the pore size distribution (4) Defect flux of the individual component of gas (5) zeolitic flux under different parametric conditions. The individual mathematical structure clearly indicates

that they are a set of simultaneous nonlinear partial differential equations of the form

$$\{f_i(N_{itotal}, N_{inz}, N_{iz}, D_{ikn}, r_i)\} = 0$$

For, i =1, 5. Therefore in the present study, we have applied the "Newton Raphson" method with a suitable tolerance to solve them. However, the equation includes some process parameters like α etc. Through the effective permeabilities of individual component, the selectivity ($\alpha \equiv P_A/P_B$) can be determined by pure gas based on experimental data. The defect radii $\langle r_i \rangle$ is identified to be unknown parameters of the proposed model. The defect radii $\langle r_i \rangle$ was considered to be the adjustable parameter of the model. In order to evaluate the respective values, global objective functions of the following form have been introduced.

$$E(D) = \frac{1}{n} \sum_{i=1}^{n} \left(1 - \frac{J_{ij}^{model}}{J_{ij}^{exp}}\right)^2 \qquad (12)$$

Where, n is the total number of data points marked by different applied transmembrane pressure. It is evident from the definition that the objective function, E(**D**) actually represents the sum of the normalized square error between the experimental and model predicted values over the entire parametric space of the present study and it was minimized to obtain the parameter vector, **D**=[r_i]T. The objective function E(**D**) was minimized upon adjusting parameters to obtain the least model prediction with respect to the experimental data points at different TMP and feed concentration of individual gases. A small tolerance limit of 0.0001 for E(**D**) was used to stop criterion of "Newton Raphson" method.

$$N_{inz} = N_{inzo}, N_{iz} = N_{izo}, N_{iv} = 0$$

Were, the initial value/guess used in the present study. The steady state simulation scheme is shown in Figure 4.

Figure 5a and 5b represents the FESEM micrograph of surface morphology and cross sectional view of synthesized DDR membrane on clay alumina support. The thickness membrane layer is ~ 25 μm and support is 3 mm. The Figure clearly shows the pore structure of the support layer is in the micron size range and membrane layer contains only intracrystalline pores. Miachon et al. have proposed that the intracrystalline area of the membrane increases with temperature which results in an increase in non-zeolitic flux [29]. However, in many studies, membrane structure is assumed to be homogeneous and defect free [30].
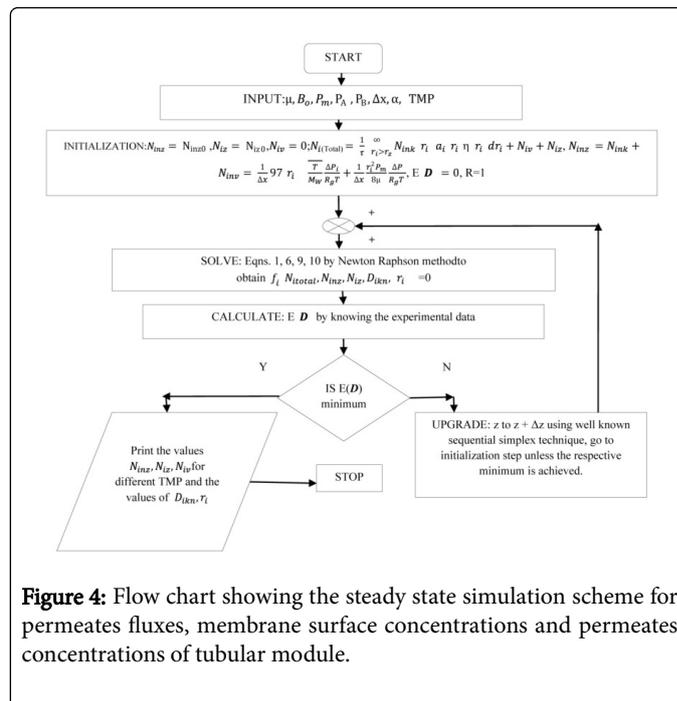
Figure 4: Flow chart showing the steady state simulation scheme for permeates fluxes, membrane surface concentrations and permeates concentrations of tubular module.
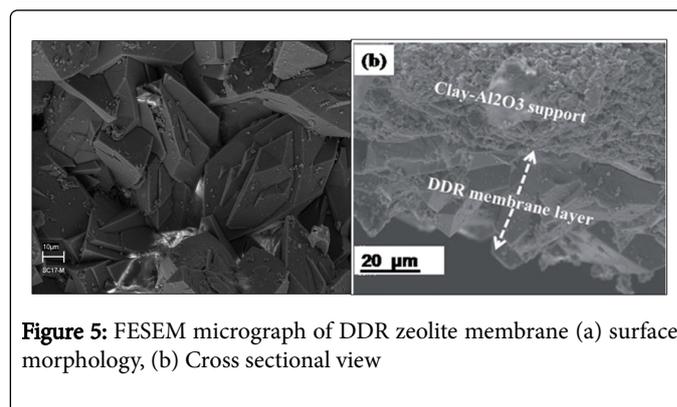
Figure 5: FESEM micrograph of DDR zeolite membrane (a) surface morphology, (b) Cross sectional view

Permeation mechanism through the membrane depends on many parameters such as interaction between gas molecules and membranes pore wall and also experimental operating conditions such as temperature and pressure [31]. Pore diameter, pore wall structure, interconnection structure of channels and molecule-molecule interaction also affect mass transport mechanism across the membrane [32]. Figure 6-8 describe the variation of molar steady state permeate flux with transmembrane pressure. It can be observed that the transport model reasonably fits with its experimental data. It represents the steady state permeate flux of individual component and mixed gas as a function of transmembrane pressure at different bulk concentration. For hydrogen separation from the mixture of H_2-CO_2, the effect of adsorption process is negligible and the separation process is mainly controlled by diffusion and molecular sieving. Hanebath et al. have assumed only Knudsen diffusion for H_2 through silicate-1 membranes. They have modelled the flux through zeolite based on a combination of Knudsen, activated and surface diffusion. They have also used 'Dusty gas model' to simulate the flow mechanism through stainless steel support of their MFI membrane. Markovic et.al also reported gas separation through porous glass membranes with relatively narrow pore size [33,34]. In this experiment, viscous flow

increases by the increasing of transmembrane pressure. In general, viscous mechanism dominates when the membrane contains defects or relatively large pores and the separation process is performed at high pressure conditions. Knudsen flux is constant with the variation of transmembrane pressure difference. With no pressure difference between feed and permeate side, only zeolite and Knudsen flux contributions have assumed to be present while there is no viscous flux [35]. In these above references, it is observed that Knudsen flux is independent of transmembrane pressure. Molecular sieving phenomena decrease sluggishly with the variation of transmembrane pressure. In Figure 6, it is observed that the steady state permeates flux of pure CO_2 decreases with different transmembrane pressure and feed concentration. The same trend is observed with H_2 and mixture gas described in Figures 7 and 8 respectively. The same analyses are valid for different gases for different kinds of (Viscous flow, Knudsen flux, Molecular sieving) transport mechanism of through the membrane. Lower zeolitic contribution is observed in CO_2 permeate flux modelling. This might be because of lower diffusivity of CO_2 compared with H_2. Some deviations are observed at higher pressure especially for CO_2. One of the reasons for this behaviour might be under estimation of defect flow at high pressure conditions. As evident, the permeate flux is observed to increase with transmembrane pressure. The total permeate flux exhibits an increasing trend with feed concentration. However, it is tending towards saturation for all H_2,

CO_2, and $H_2:CO_2$. Trend is observed with respect to feed concentration, though the respective profile is slightly nonlinear. It may be attributed that the deviations at lower transmembrane pressure between the experimental values are considerably minimized for the gas mixture as the movement of the relatively small H_2 gas molecule is retarded by that of the larger CO_2 molecules. The diffusion rate of gases having smaller kinetic diameter generally becomes significantly higher through the larger pore of zeolite. The molecular kinetic diameters of H_2 and CO_2 are 0.29 and 0.33 nm respectively, the kinetic diameter of CO_2 being close to the pore size of DDR zeolite. The difference in molecular size between H_2 and CO_2 causes difference in the diffusion rates through the DDR zeolite membrane. Respective model predictions for the total permeate fluxes were close to the experimental values for all the different data point as indicated in Figure 6-8. The mean value of the normalized standard deviation, defined as:

$$\bar{\sigma}_j = \sqrt{\frac{\sum_{i=1}^{n}\left(1 - \frac{J_{ij}^{model}}{J_{ij}^{exp}}\right)^2}{n-1}}$$

For, j = 1 to m, was found to be as small as 0.01

Figure 6: Contribution of the different transport mechanism to total CO_2 flux as a function of different applied transmembrane pressure at 303.15 K across the DDR zeolite membrane.
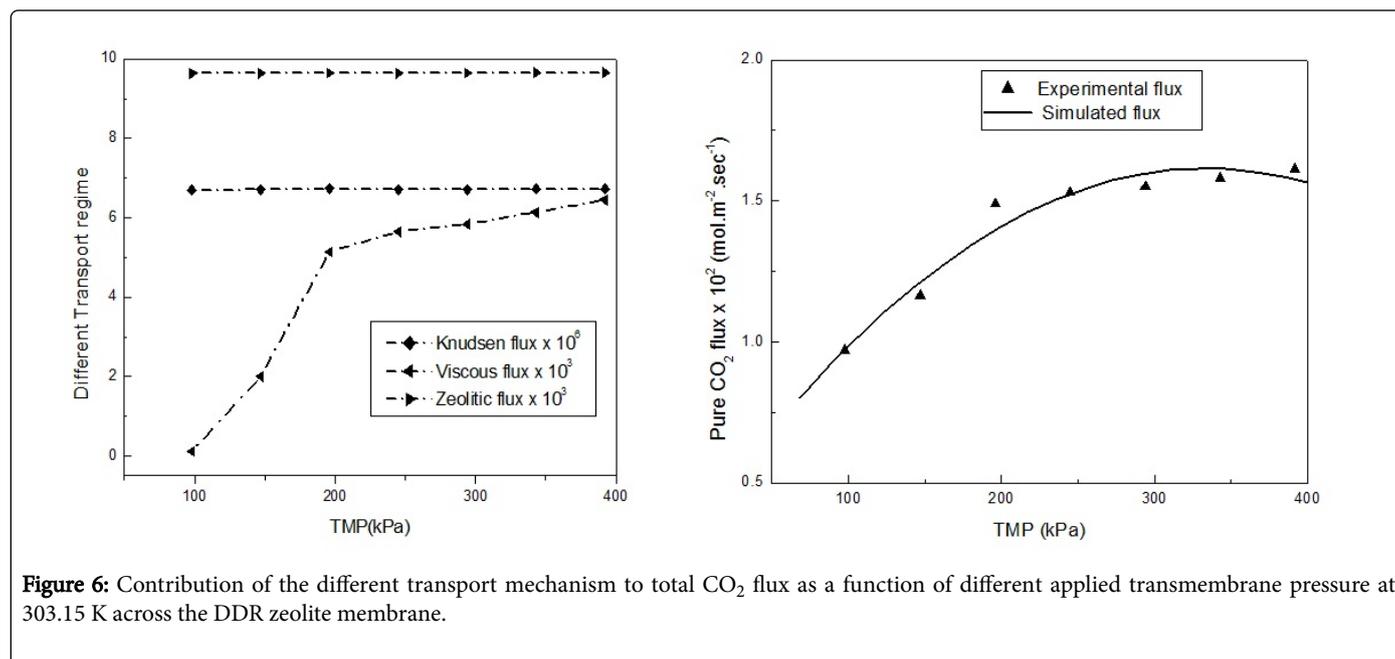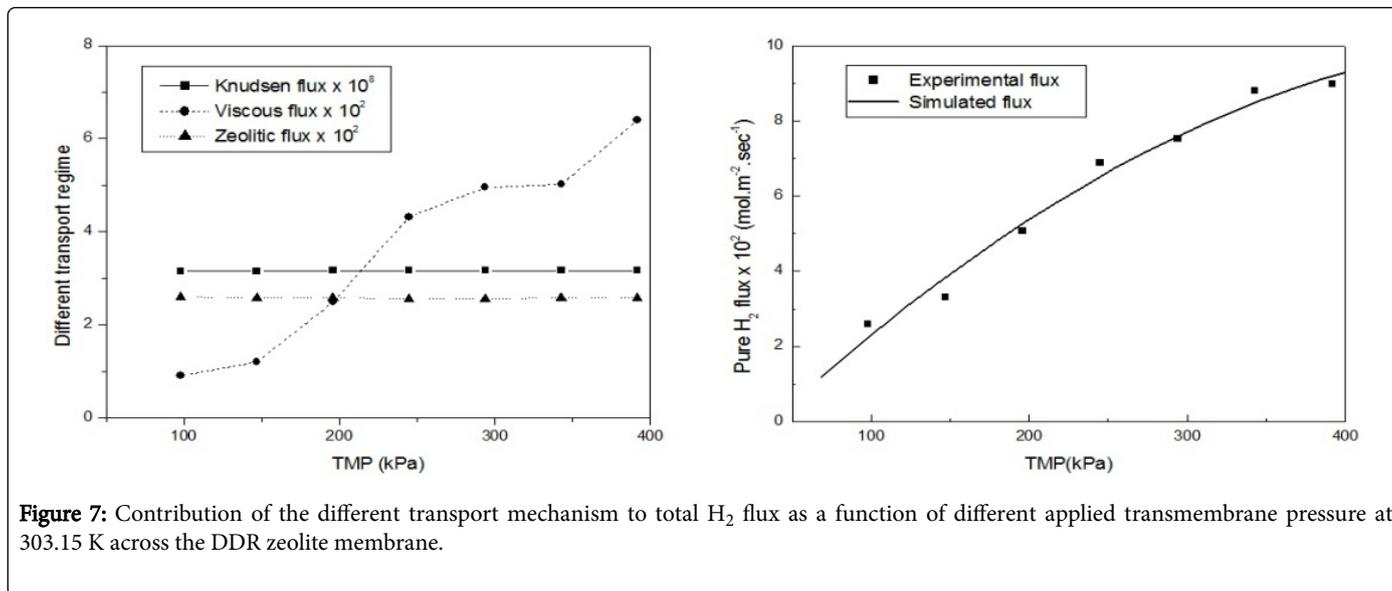
Figure 7: Contribution of the different transport mechanism to total H_2 flux as a function of different applied transmembrane pressure at 303.15 K across the DDR zeolite membrane.
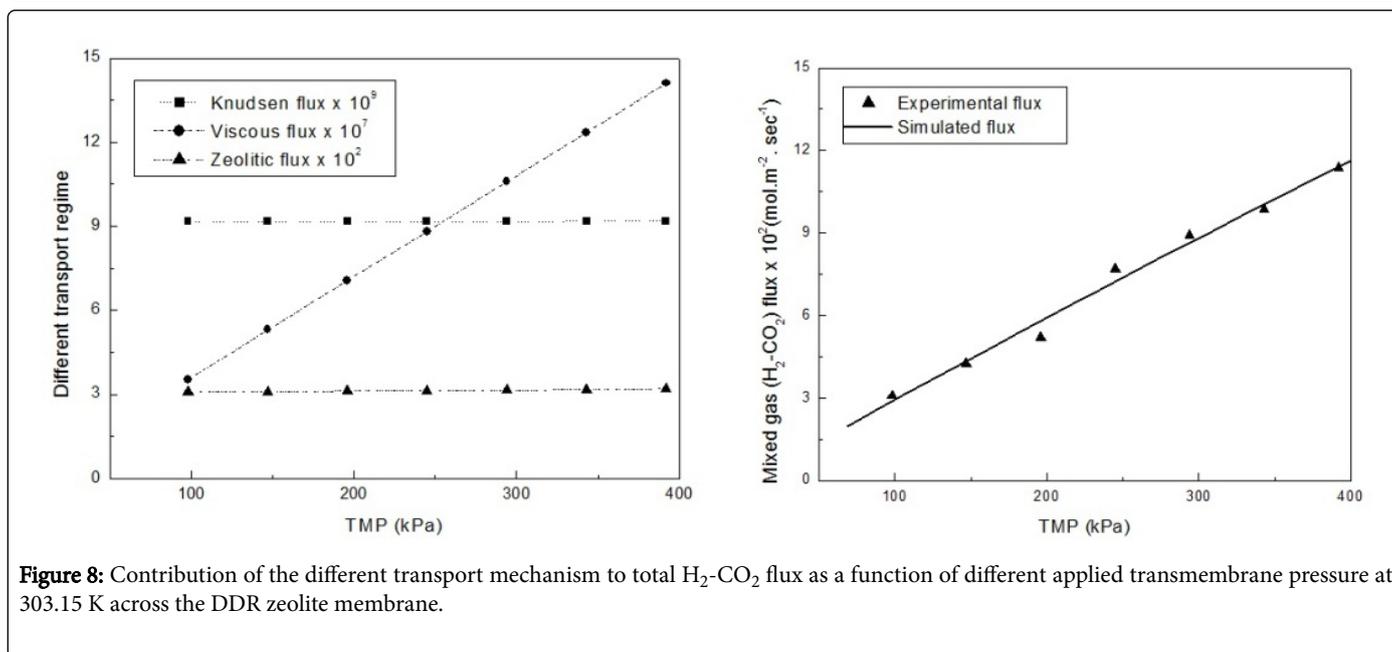
Figure 8: Contribution of the different transport mechanism to total H_2-CO_2 flux as a function of different applied transmembrane pressure at 303.15 K across the DDR zeolite membrane.

Comparative parameter analysis study with the variation transmembrane pressure: Lin and Burgraaf have shown that the ratio (β'/α') is a comparative parameter related to the medium pore size and different membranes [36]. The steady state permeate molar flux of a membrane having defect size distribution is

$$N_{inz} = N_{iv} + \int_{r_i > r_z}^{\infty} N_{inz}(r_i)\eta(r_i)dr_i \quad (13)$$

By using the corresponding equations for Knudsen, viscous and intercrystalline transport mechanisms.

$$N_{itatal} = \alpha'\left[P'\right] + \beta' \quad (14)$$

Where, the parameters α' and β' are expressed as:

$$\alpha' = \frac{r_i^2}{8\mu}\frac{1}{R_g T}$$

$$\beta' = 97\, r_i\sqrt{\frac{T}{M_W}}\frac{1}{R_g T}\frac{\Delta P_i}{\Delta x_i} + N_{iz}$$

$$\epsilon = 97\, r_i\sqrt{\frac{T}{M_W}}\frac{1}{R_g T}$$

Related to Knudsen contribution

$$\varphi = N_{iz}$$

represents the flux associated with zeolitic pore. In the analysis it is shown that ϵ is independent of pressure and membrane type. Because of the weak adsorption affinity of H2, its zeolitic flux is almost constant

at 303 K for applied different transmembrane pressure. Therefore, ϵ can be assumed to be independent of pressure. It may attributed Knudsen diffusivities are much higher than the zeolitic crystal cavities. Therefore, very small fraction of the membrane area is required to provide a large contribution of the defect flux. In particular, the model fit values for the essential parameter $\beta = Az/At$ fall into a range $0.9 < \beta < 1$

for the evaluated membranes in this work. This means that the factor Az/At essentially does not modify the magnitudes of ϵ termed as estimated. Hence, in general for normal quality zeolite membranes the zeolite open area over the total permeable area ratio (A_z/A_t) is a value the approximates to one [37]. The fractions of non-selective viscous flux pass through the relatively large defect pore. To characterize the membrane defect size we should plot steady state permeate molar flux versus P'. The linear function, its slope (α') and intercept (ß) is defined by the above mentioned equation. We have introduced another comparative parameter that could also be very valuable in characterization of non-uniform membrane. It can be easily shown that:

$$(\beta')^2 = \epsilon^2 + \varphi^2 + 2\epsilon\varphi \qquad (15)$$

The second term and third term of this expressions can be neglected in comparison of zeolitic flow is negligible at 303 K for H2. As a result, $(\beta')2=\varphi2$ and the equation becomes that:

$$\frac{(\beta')^2}{\alpha'} = \frac{97^2\left(\frac{1}{\tau}\right)^2\left(1-\frac{A_Z}{A_t}\right)^2 \frac{T}{M_W}\frac{1}{R_g^2 T^2}\frac{dP_i}{dx_i}}{\left(1-\frac{A_Z}{A_t}\right)\frac{r_i^2}{8\mu}\frac{1}{R_g T}\frac{dP_i}{dx_i}} \qquad (16)$$

$$\frac{(\beta')^2}{\alpha'} = 97^2\left(1-\frac{A_Z}{A_t}\right)\frac{8\mu}{R_g M_W} \qquad (17)$$

Right hand side of equation can be written as a constant ω which only depends on the viscosity and the molecular weight of the permeating gas multiplied by the defect area of the membrane [36,37].

$$\frac{(\beta')^2}{\alpha'} = \left(1-\frac{A_Z}{A_t}\right)\omega \qquad (18)$$

Thus $(\beta')^2/\alpha'$ is an additional parameter attributed to the defect area of each membrane and the corresponding tortuosity. This parameter characterizes the defect area and is proportional to $(1-A_z/A_t)$ and the $(\beta')^2/\alpha'$ values are calculated for the membranes.

Conclusion

The summary above reveals that reasonable values of gaseous flux can be obtained for a range of experimental conditions specifically in the range of pressure difference. By and large good agreement between calculated and experimental fluxes confirms the computational and theoretical premises of the study. These observations of gas flows through the zeolitic membrane may be applicable to other inorganic micro porous membranes which may be a part of future research. The work can be extended to large scale membrane modules for studies of gas flows from multi-component gaseous mixtures in order to create database for membrane separators design. Gas separation through ceramic supported membrane is a deeply complex process. In addition to the testing difficulties with experiments multicomponent transport modelling is complicated further as one should take into account different kinds interactions such as competitive adsorption and pore

blocking. Such a knowledge will require physico-chemical or empirical formalisms of the adsorption processes by the membrane that can be plugged into the present computational scheme. Presently such formalisms are scanty in literature and therefore these issues become subject of future research.

Acknowledgment

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