Simulation for CO₂ capture using tubular dual-phase membrane module

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Abstract. Dual-phase membrane is a newly developed membrane that is capable of capturing carbon dioxide (CO₂) from flue gas at high temperature up to 823 K. To date, the researches on CO₂ capture using dual-phase membrane are performed experimentally. However, the gas separation performance of the scale-up tubular dual-phase membrane module is scarcely studied. Therefore, the potential application of the dual-phase membrane module remains as a challenge. The design of membrane module and its implementation in actual operating conditions can be analyzed beforehand by using Computational Fluid Dynamics (CFD) simulation. In this paper, the hydrodynamic profile of the gas flowing inside a tubular dual-phase membrane module was studied to investigate its potential for industrial application. CFD simulation of gas mixture consisting of CO₂ and nitrogen (N₂) that flowing through the membrane module was performed at 823 K. Among the parameters investigated are the absolute pressure, concentration of CO₂ and gas velocity within the tubular membrane module. The inlet mass flow rate was set at 0.00448 kg s⁻¹ and the total volume of the membrane module was 0.031 m³. Based on the simulation, 0.09 bar of pressure drop was observed when the feed gas stream passed across the membrane zone to the outlet zone. There was about 86 % of CO₂ recovery with the CO₂ concentration decreased from 20 mol % to 3.3 mol %. Besides, the membrane stage cut was around 0.17 with 83 % of the gas leaving the membrane module at the retentate side. The simulation results give reliable statement over the separation efficiency of CO₂ and flow pattern in membrane module. The scale-up performance of single membrane module has been predicted through the simulation based on the experimental data.

Keywords: Dual-phase membrane; CO₂ separation; Membrane module; CFD simulation; Industrial application.

1. Introduction

According to a global statistic reported in 2013 [1], nearly 50% of the total carbon dioxide (CO₂) emission is resulted from burning of fossil fuel for power generation. This has led to serious environmental issues and hence, approaches to separate and capture CO₂ rather than releasing them to the environment has become desirable in the industrial sector [2]. Membrane technology has been brought to industrial application as an alternative method for flue gas purification due to its high CO₂...
capture efficiency, simplicity in process design, low energy consumption and scale-up flexibility [3, 4].

In year 2005, the first dual-phase membrane that showed promising potential for CO$_2$ separation at high temperature was reported by Lin and coworkers [5]. It was a dense dual-phase membrane consisting of molten carbonate phase infiltrated in a porous inorganic solid support that exhibited an infinite selectivity for CO$_2$ over N$_2$ at temperatures above 500°C. The underlying principle is that at high temperature, CO$_2$ can permeate through the membrane as carbonate-ion (CO$_3^{2-}$) under the CO$_2$ partial pressure gradient [6-9]. In year 2018, a novel type of dual-phase membrane based on molten hydroxide liquid phase contained in a nanoporous solid support, termed as hydroxide/ceramic dual-phase membrane (HCDP), was developed by Céron et al. [10] to separate CO$_2$ at flue gas temperature ranged from 250 °C to 650 °C. This membrane shows exceptionally high CO$_2$ selectivity and permeability, which is about 25 times more effective than the previously reported dual-phase membranes. With the advancement of the dual-phase membrane, it can be a promising technology to be retrofitted in the existing power plants for an effective CO$_2$ separation.

Computational fluid dynamics (CFD) is a numerical analysis of fluid mechanics system involving fluid flow, mass and heat transfer inside a defined flow geometry by means of computer-based simulation [11, 12]. CFD can be used as a powerful CAE (computer-aided engineering) tool in industrial research and design to analyze the performance of new processes and equipment. Thus far, theoretical studies of dual-phase membrane with experimental validation were widely presented in the literature. However, CFD simulation for scaling up the dual-phase membrane module for industrial application has not yet been reported. Making an actual prototype of the membrane for experimental testing has always been costly and time-consuming, considering the large scale of equipment in a power plant. Hence, a performance prediction has always been difficult, whereby consumers are refrained from implementing the technology. Compared to the experimental approaches, CFD simulation can be carried out at reduced cost and flexibility to control of the variables, at the same time procures reliable results [12]. The CFD techniques offer the capacity to study the dual-phase membrane at conditions over the limit of experiments to generate different trend analysis that permit understanding of the feature of the result. Hence, CFD analysis is an useful tool in investigating the feasibility and performance of a membrane module for power plant application. In this paper, the hydrodynamic profile of the gas flowing through the tubular dual-phase membrane module will be investigated using CFD simulation based on the reported CO$_2$ permeance data of dual-phase membrane.

2. Methodology
The simulation was performed using ANSYS Fluent 19.2 to investigate the potential performance of the tubular dual-phase membrane module for CO$_2$ separation. ANSYS Workbench 19.2 was used to set up the geometry, and the corresponding computational mesh was created using the meshing tool. The simplified geometry consisted of three parts, including the inlet, porous membrane and outlet zones. Typically, the general workflow of CFD simulation includes three main stages, which can be categorized as pre-processing, solving and post-processing.

2.1. Pre-processing
The three-dimensional fluid flow system was created using ANSYS Design Modeler. After the geometry was created, a computational mesh for the flow volume was generated. The geometry was meshed with structured quadrilateral elements with adaptive sizing, generating the resultant meshing structure of the geometry model from coarse to fine mesh size.

2.2. Solving
The CFD simulation of the membrane geometry was conducted using ANSYS FLUENT Solver. The first step was to choose a solver based on the characteristic of fluid flow, which was the numerical methods for ANSYS to solve the governing integral equations for the conservation of mass and
momentum. In the simulation, the cell zone conditions for inlet and outlet zone were set as fluid zone. Meanwhile, the condition for membrane zone was set as porous zone with mass source term. The other boundaries were the wall of the membrane module. Before starting calculation, the flow field was initialized by setting initial values for the flow variables. Standard Initialization was chosen, and the values set of the inlet zone was used to initialize the flow field for solutions. The simulation was run until it reached to convergence.

2.3. Post-processing
A completely defined fluid flow system consists of a geometry, computational mesh, CFD setup setting, solution and results. The post-processing steps involved the display of the simulation results in ANSYS FLUENT, which included the contours of absolute pressure, concentration of species (CO$_2$) and velocity of fluid flow throughout the system (membrane module).

3. Results and discussion
3.1. Geometry and dimension of tubular dual-phase membrane module
Figure 1 shows the geometry of the proposed tubular membrane module. A binary mixture of gas consisting of carbon dioxide (CO$_2$) and nitrogen (N$_2$) was fed into the membrane module through the inlet. Attributed to the characteristics of dual-phase membrane, CO$_2$ was captured by the membrane when the feed gas permeated through the membrane layer. The retentate gas with low composition of CO$_2$ then exited the membrane module through the outlet. The outer diameter of feed inlet, membrane and retentate outlet sections were 20 cm, 17 cm and 8 cm, respectively.

![Figure 1. Schematic diagram of tubular membrane module.](image)

The tubular membranes were arranged orderly in the membrane zone of the module as shown in Figure 2. Particularly, membrane arrangement will affect the porosity and resistance of the gas flowing across the membrane module. The selective membrane layer was at the outer side of the tubular membrane. Therefore, the outer membrane diameter was used to calculate the effective surface area of membrane.
The porosity of membrane, $\gamma$, was determined using Equation (1).

$$\gamma = \frac{D^2 \pi}{4L^2 \sin \theta}$$  \hspace{1cm} (1)

where $D$ is membrane outer wall diameter (m); $L$ is the sum of the membrane outer diameter and spacing between membrane (m) and $\theta$ is angle between adjacent membrane (º).

Meanwhile, the resistivity of the membrane, $R_x$ (1/m²), was estimated using Equation (2).

$$R_x = \frac{12}{s^2}$$  \hspace{1cm} (2)

where $s$ is spacing between membrane (m).

The single membrane area for the tubular membrane, $a$ (m²), was estimated based on Equation (3).

$$a = L^2 \sin \theta$$  \hspace{1cm} (3)

Then, the effective membrane surface area per cubic, $A$ (m²/m³), was calculated based on Equation (4).

$$A = ND\pi$$  \hspace{1cm} (4)

where $N$ is the number of tubular membrane per cubic.

$N$ was estimated based on Equation (5).

$$N = \frac{1}{a}$$  \hspace{1cm} (5)

Table 1 tabulates the values used for the variables stated from Equations (1) to (5).
Table 1. Dimension and configuration of tubular membrane module for dual-phase membrane.

| Dimension                        | Unit | Value       |
|----------------------------------|------|-------------|
| Module shell length              | m    | 1           |
| Module shell diameter            | m    | 0.2         |
| Membrane shell thickness         | m    | 0.03        |
| Volume of membrane layer         | m$^3$| 0.0177      |
| Membrane wall thickness, $d$     | m    | 0.002       |
| Membrane outer wall diameter, $D$| m    | 0.01        |
| Spacing between membrane, $s$    | m    | 0.002       |
| Angle between adjacent membrane, $\theta$ | ° | 60         |
| Single membrane area, $a$        | m$^2$| 1.247×10^{-4} |
| Number of tubular membrane per cubic, $N$ | -  | 8019 |
| Effective surface area per cubic, $A$ | m$^2$/m$^3$ | 250 |
| Porosity, $\gamma$              | -    | 0.63        |
| Resistance across membrane material at x-direction, $R_x$ | 1/m$^2$ | 3×10$^6$ |

3.2. Meshing of membrane module in ANSYS Fluent

The membrane module consisted of three layers in concentric tubular configuration. The outermost layer formed the inlet (feed) zone, the middle layer was the membrane zone, while the innermost layer was the outlet (retentate) zone. After the creation of geometry, the module was meshed with hexahedral cells of element size 10 mm with adaptive sizing. The geometry consisting of three regions contained altogether 58622 nodes and 47475 elements. This overall mesh count was considerably high for a simple geometry, which aimed to obtain a good mesh resolution without corrupted cells on the wall surface. Figure 3 shows the meshing of the membrane module in cross section view.

![Figure 3. Cross section view of the meshed membrane module using element size of 10 mm.](image)

3.3. Simulation conditions

For this simulation, the feed stream consisted of two species, which were CO$_2$ and N$_2$, forming a binary mixture of gas. Since the selectivity of CO$_2$/N$_2$ is over 1000 for dual-phase membrane [10], the permeance of N$_2$ was ignored in the simulation. The module was configured to separate CO$_2$ from the mixture and therefore required high permeance of CO$_2$. It is noted that, a User Defined Function was developed to capture CO$_2$ at the membrane zone. The value of CO$_2$ permeance was set at 8.9×10^{-8} mol m$^{-2}$s$^{-1}$Pa$^{-1}$ based on the reported permeance of dual-phase membrane with thickness of 2000 μm [10]. The operating temperature was 823K and the composition of feed gas consisted of 20 % CO$_2$ and 80 % N$_2$.

Table 2 shows the simulation conditions. The temperature at the feed side was 823 K to resemble the operation condition for a dual-phase membrane in a power plant. The absolute pressure was 1.1 bar.
at the inlet side. The feed inlet was set to pressure-inlet. Thus, the inlet gas velocity, flow rate and gas viscosity were determined using ANSYS Fluent default function. Besides, a turbulence model (Transition SST) and SIMPLE solver were selected for the simulation.

| Parameter         | Unit   | Value  |
|-------------------|--------|--------|
| Temperature       | K      | 823    |
| Inlet CO₂ concentration | mol % | 20     |
| Inlet N₂ concentration | mol % | 80     |
| Inlet pressure    | bar    | 1.1    |
| Inlet gas velocity| m/s    | 1.1067 |
| Inlet mass flow rate | kg/s  | 0.00448 |
| Gas Viscosity     | kg/m·s | 1.72×10⁻⁵ |
| Fluid model       | -      | Transition SST |
| Solver            | -      | SIMPLE  |

3.4. **Hydrodynamics profile in membrane module**

The pressure drop of gas stream across a membrane module is one of the important aspects that affect the performance of a tubular membrane module. Figure 4 shows the pressure profile in three regions of the membrane module. The inlet zone showed the greatest pressure, which decreased toward the middle part of the module. The outlet of membrane module demonstrated the lowest pressure. There was an average pressure drop of 0.09 bar within the membrane module as the feed stream passed across the membrane zone to the outlet zone. The main cause of the decrease in pressure was the viscous and frictional resistance within the tubular membranes.

![Figure 4. Absolute pressure along the membrane module in X-Z direction.](image)

Figure 5 illustrates the mole fraction of CO₂ in the membrane module. It can be observed that there is a decrease in CO₂ concentration from the inlet zone to the outlet zone. The decrease of CO₂ concentration is due to the capture of CO₂ through molten electrolytes contained in the tubular dual-phase membrane [13]. As observed from Figure 5, the membrane module was capable of reducing the concentration of CO₂ from 20 mol % to 3.3 mol %, which contributed to 86 % recovery of the CO₂.
At temperature of 823K, the molten electrolyte of dual-phase membrane absorbed CO$_2$ to form CO$_3^{2-}$. The dissolved CO$_3^{2-}$ ions transported across the membrane via the carbonate mechanism. On the other hand, the solubility of N$_2$ was greatly less than that of CO$_2$[10]. Hence, N$_2$ molecules were not permeated through the dual-phase membrane, but remained to form the retentate gas stream.

Figure 6 shows the velocity magnitude of gas flowing within the membrane module. Referring to Figure 6, the inlet gas velocity of 1.1067 m/s was increased to 2.1201 m/s at the outlet. Meanwhile, membrane zone demonstrated the lowest gas velocity of 0.1405 m/s. This is mainly due to the higher resistance caused by the tubular membrane layer.

Based on the simulation, the outlet mass flow rate achieved a stable value of 0.0028 kg/s. As compared to the inlet mass flow rate of 0.00448 kg/s. The membrane stage cut or fraction of feed permeated for the simulation condition was 0.17. Around 83 % of gas was left the membrane module as retentate.

Subsequently, one of the parameters, which was the velocity magnitude of the gas, was selected to conduct the grid independence testing. Element with smaller size of 5 mm was used to run the simulation and the cross section view of the meshed membrane module is shown in Figure 7 (left). Meanwhile, Figure 7 (right) shows the insignificant change in the velocity magnitude of the gas flowing within membrane module by using the meshing with refined element. This has indicated that the meshing with the element size of 10 mm is sufficient to simulate the hydrodynamic profiles results for the membrane module in the study.
Figure 7. (Left) Cross section view of the meshed membrane module using smaller element size of 5 mm. (Right) Velocity magnitude of gas flowing within membrane module in X-Z direction using element size of 5 mm.

4. Conclusion
In this paper, the scale-up performance of dual-phase membrane module for CO₂ capture was studied via Computational Fluid Dynamics simulation using ANSYS Fluent 19.2. A geometry of the membrane module was built and meshed to appropriate number of cell elements. The solver of ANSYS Fluent enabled the simulation of CO₂ transport in the membrane module at different regions. In overall, the absolute pressure, CO₂ concentration and velocity profile within the membrane module have been simulated based on the experimental data of CO₂ permeance. Based on the simulation results, high CO₂ recovery up to 86% and low membrane stage cut of 0.17 are expected with the design of the tubular membrane module. Nevertheless, the feasibility of the dual-phase tubular membrane module for large-scale industrial application still requires further improvement as compared to the commercialized hollow fiber membrane module as it needs larger size of the membrane module. For future recommendation, the diameter of the dual-phase membrane can be further reduced to optimize the separation performance of the module for industrial application. The compatibility of dual-phase membrane to high temperature application offers a potential outlook for CO₂ capture in the power plant, which contributes to about half of the overall CO₂ emission.

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