Computational Fluid Dynamics Simulation of CO₂ Adsorption On Nanoporous Activated Carbon: Effect of Feed Velocity

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ARTICLE INFO

Article history:
Received 25 July 2014
Received in revised form 8 July 2014
Accepted 15 September 2014
Available online 17 October 2014

Keywords:
Computational Fluid Dynamics (CFD); Nanoporous media; Fixed-bed adsorption

ABSTRACT

The understanding of fluid flow inside a fixed-bed adsorption column is crucial as mass transfer in the bed is influenced by the column hydrodynamics. In this study, a numerical model has been developed to simulate the adsorption of chemical species of CO₂/CH₄ mixture inside the geometry containing solid adsorbent particles, namely nanoporous activated carbon. The laboratory-scale geometry consists of a High-Pressure Volumetric Analyzer (HPVA) adsorption chamber, where in this study the effect of heat transfer within the bed is assumed negligible. The Computational Fluid Dynamics (CFD) simulations are performed using ANSYS Fluent® 14.0 to investigate the effect of flow rate on CO₂ adsorption capacity through the variation of the inlet feed velocities. Simulation results show that the amount of gas adsorbed inside the column varies inversely with the increase of inlet velocity, which is in agreement with the experimental data. In addition, pressure gradient across the domain shows an increment trend as the flow rate of the gases increases.

INTRODUCTION

The anthropogenic emission of carbon dioxide is the main issue of concern for today’s world. The harmful effect of greenhouse gases poses an actual threat to the sustainability of ecosystem and of environment (Rodrigo Serna-Guerrero, 2010). In current stage, fossil fuels supply more than 85% of the world energy demand and are primary source of energy (RavichandarBabarao, 2010). The extraordinary use of fossil fuels for the advancement of human society over the past two centuries has affected adversely on the composition of Earth’s atmosphere. In addition to CO₂ capture to address environmental concerns, the removal of CO₂ in applications including air purification and natural gas treatment is also required. Regarding natural gas (NG), the amount of CH₄ and CO₂ in NG generally varies from 70-90% and 0-20% respectively, while the CO₂ is considered as an impurity and the removal of CO₂ becomes necessary as higher content of it in NG leads to the reduction in heating value and pipeline corrosion problems (Nouh, 2010).

After the enforcement of Kyoto protocol in 2005, carbon capture and storage (CCS) techniques from flue gases were taken into consideration with special attention towards the subject (Fengsheng, 2009). Several different approaches have been proposed to remove CO₂ from flue gases on large scale. The four main approaches that are currently in use for the separation process are cryogenic distillation, membrane purification, absorption with liquids and adsorption using solids (Sunho Choi, 2009). The ability of solid sorbents to reduce the energy demand of capture processes due to potentially higher loading capacities and lower heats of sorption is advantageous. Solid sorbents are most effective when they have a large surface area-to-mass ratio and a preferential interaction with CO₂ (Jennifer Wilcox,).

The type of adsorbent used in this work is activated carbon derived from agricultural waste, which is palm mesocarp fibre (PMF). The major advantage of using activated carbon from agrowaste is that Malaysia is the world’s top producer of palm oil and a huge amount of solid wastes such as empty fruit bunches (EFB) and palm oil kernel are discarded during the post-production process (Saad Hashim Khalil, 2011). Activated carbons are sorbents with a highly developed porosity, especially micropores and mesopores, that find enormous applications in various medical, industrial and scientific fields (Saad Hashim Khalil, 2011).

Much interest and attention is devoted to modeling and simulation of kinetic and equilibrium adsorption phenomenon in the fixed bed adsorption column in order to observe it at laboratory scale. For this purpose, several mass transfer models are proposed throughout the literature such as pore-diffusion model, linear
driving force (LDF) and quadratic driving force (QDF) (Ruthven, 1994). Computational fluid dynamics (CFD) is used to model hydrodynamics, mass and heat transfer phenomena for the design and optimization of process equipment. CFD simulation is an appropriate tool to be used when the process performance is dictated by fluid dynamics (Nouh, 2010). The objective of this study is to determine the hydrodynamics and adsorption phenomena for the CO₂/CH₄ mixture in the fixed-bed adsorption column filled with nanoporous activated carbon through CFD approach by taking into account the effect of feed velocity on the amount of gas adsorbed throughout the column. Concentration factor is calculated by obtaining the ratio of released amount of gas (C_{out}) in the bed at specific time to the inlet gas concentration (C_{in}). The effect of feed velocity on the amount of gas adsorbed is taken by plotting the concentration factor along the height of the column at the above mentioned flow rates.

**CFD Model and Flow Setup:**

The dynamic behavior of the fixed bed adsorption column is modeled using integrated CFD model, where the mass transfer is considered to be convective-diffusive. In this work, the inlet concentrations of CO₂/CH₄ are set at (20/80)\%, respectively, as the typical value found in the practical natural gas treatment system. In addition, considering the low concentration of CO₂ and the small length-scale of the actual HPVA column investigated, an isothermal column operation is thus assumed. This assumption simplifies the model as the mass balance could be described with the absence of energy balance.

The following assumptions are made to formulate the gas adsorption mechanism in this system:

1) The adsorption between CO₂ and CH₄ is assumed to be competitive.
2) Heat transfer within the bed is negligible.
3) The mass transfer is represented by linear driving force (LDF) model.
4) The mass transfer coefficient takes into account the external fluid film resistance and macropore diffusion.
5) The porous media domain is utilized where the porosity is considered as uniform throughout.

The model used for the flow dynamics is the modified Darcy’s law combined with laminar Navier-Stokes equations. This model, called ‘Darcy-Brinkman-Forchheimer’ model has been extensively used to simulate flow in porous media and its validity is discussed in the literature (Nield, 1991; Vufai, 1995).

Multiple equations are solved by the ANSYS Fluent® 14.0 solver, which includes:

The continuity equation,

\[ \nabla \cdot U = 0 \]

The momentum equation,

\[ \frac{\partial (\rho U)}{\partial t} + \nabla \cdot (\rho U U) = -\nabla p + \rho \nabla \cdot \tau + \rho f \]

For fluid-particle friction forces, \( F \),

\[ F = -\frac{\mu d}{K} \beta \rho U^2 \]

For fluid particle friction forces, ‘\( K \)’ is the permeability of porous media and ‘\( \beta \)’ is the non-Darcy term or inertial resistance coefficient. In a fixed bed of narrow sized spheres, it is recommended to calculate ‘\( K \)’ and ‘\( \beta \)’ from the Ergun’s law (Augier, 2008), which are given in the expressions below.

\[ K = \frac{\varepsilon d^2}{150(1-\varepsilon)} \]

\[ \beta = 1.75 \frac{1-\varepsilon}{\varepsilon d_3} \]

The literature suggests that for cylindrical vessels with D/d>2 and bed height of H>20d, the bed porosity can be approximated by the expression as follows (Pushnoy, 2006; Ali Qasim, 2014):

\[ \varepsilon = \frac{A}{D} + B \]

Where A, B and n are constants, dependent on the shape of particle whereas D and d are vessel diameter and particle diameter respectively.

The general mass balance equation for an adsorption packed bed column is:

\[ D_{\alpha} \frac{\partial^2 C_i}{\partial x_i^2} + u \frac{\partial C_i}{\partial x_i} + \frac{\partial C_i}{\partial t} + \frac{1-\varepsilon}{\varepsilon} \frac{dq_i}{dt} = 0 \]

\( dq_i/dt \) is based on the LDF model expressed as:

\[ \frac{dq_i}{dt} = (C_{inh} - C_i) K_i \alpha \]

The intraparticle concentration, \( C_i \), for adsorbent particle is described by multi-component Langmuir model (Suzuki, 1989).

\[ q_i = \frac{q_i b_i C_s}{1 + b_i C_s + b_n C_n} \]
Where, A= CO$_2$; B= CH$_4$; $b_i$ is equilibrium constant.

1.1. Geometry and Meshing:

The HPVA column is investigated in the form of a vertical cylindrical column, with a geometry of 5 mm diameter and 50 mm height. The ICEM CFD. Figure 1 illustrates the domain geometry, which is created using ANSYS Design Modeler® and discretized to a number of computational nodes, which is developed via ANSYS ICEM CFD®. The number of nodes and elements in the mesh are 42400 and 44541 respectively in which 40095 elements are hexahedral.

Fig. 1: Domain geometry and mesh development.(a) Axial section ; (b) Radial section.

1.2. Boundary conditions and computational method:

Considering a three dimensional axisymmetric domain, the above set of governing equations which include conservation equations for mass and momentum, coupled with system mass balance are solved using ANSYS Fluent® 14.0. The mass transfer coefficients are modeled as user-defined source terms through the implementation in the User Defined Scalars (UDS) in the setup.

Table 1 shows the flow properties setup for the porous fluid domain, while Table 2 accounts for the properties of porous media.

Table 1: Flow properties inside domain.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fluids</td>
<td>CO$_2$ Ideal gas and CH$_4$ Ideal gas</td>
</tr>
<tr>
<td>Fluid Morphology</td>
<td>Continuous Fluids</td>
</tr>
<tr>
<td>Reference pressure</td>
<td>1 atm</td>
</tr>
<tr>
<td>Heat transfer model</td>
<td>Isothermal</td>
</tr>
<tr>
<td>Inlet flowrates</td>
<td>50 cm$^3$/min, 100 cm$^3$/min, 150 cm$^3$/min, 200 cm$^3$/min, 250 cm$^3$/min</td>
</tr>
</tbody>
</table>

Table 2: Properties of porous media.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Adsorbent Type</td>
<td>Activated Carbon (derived from Palm Mesocarp Fibre)</td>
</tr>
<tr>
<td>Particle size (dp)</td>
<td>250 µm</td>
</tr>
<tr>
<td>Porosity ($\varepsilon$)</td>
<td>0.40</td>
</tr>
<tr>
<td>Adsorbent Bulk Density</td>
<td>2100 kg/m$^3$</td>
</tr>
<tr>
<td>Bed Length</td>
<td>50 mm</td>
</tr>
<tr>
<td>Bed Diameter</td>
<td>5 mm</td>
</tr>
</tbody>
</table>

RESULTS AND DISCUSSIONS

3.1. Pressure Gradient inside domain:

The pressure gradient inside porous domain depends upon the factors of permeability and resistance coefficient along with viscosity, density and inlet velocity of the fluid. The model uses the modified Darcy equation and its validity is discussed by Zeng and Grigg (2006). Andrade et al. (1998) modeled gas flow in porous media based on the Reynolds number defined as:

$$Re = \frac{K \rho v}{\mu}$$  \hspace{1cm} (10)

For the above used approach, the emergence of critical Reynolds number is 0.01-0.1 (Andrade, 1998). The
comparison of pressure gradient values for the adsorbent of particle size of 250 µm at varying flow rates is done in the following Figure 2.

![Graph showing pressure gradient](image)

**Fig. 2: Pressure gradient across the column at various flow rates.**

Figure 2 shows that the pressure gradient increases as the fluid progresses along the column and attains a certain constant value after some height and decreases at the exit of the column and also the pressure drop increases as the fluid inlet velocity is increased.

### 3.2 Effect of feed velocity on bed CO₂ concentration factor:

Figure 3 shows the amount of gas having adsorbed along the height of the column at the given flow rates. It can be observed from the graph that more quantity of CO₂ is adsorbed in the adsorbent when the velocity is low, indicating adsorption capacity increased at lower feed velocity. The concentration factors can be calculated on the basis of the following graph and are reported in Figure 4.

![Graph showing CO₂ concentration](image)

**Fig. 3: Effect of feed velocity on CO₂ concentration across the adsorption column.**

The objective of this section is to study the influence of inlet feed velocity on packed bed adsorption process. In order to study the adsorption condition on CO₂ adsorption process, CO₂ concentration factor along the bed is obtained at different feed velocities. Figure 4 demonstrates the relation between the concentration factor at feed flow rates of 50 cm³/min to 250 cm³/min along the bed length with constant CO₂ concentration, where concentration factor is equal to \( \frac{C}{C_{in}} \). Based on Figure 4, the concentration factor is found to be reducing along the adsorption bed. At the entrance, the bed will adsorb most of CO₂ and get saturated faster. Since the region is saturated earlier, it increases the unadsorbed CO₂ and gives higher concentration factor at the entrance region. Moving along the bed, concentration factor decreases as a result of adsorption by the upstream bed region. Cavenati et al. (2006) observed the similar trend in their studies involving the CH₄/CO₂/N₂ gas mixture separation (Cavenati, 2006). Besides this, higher Reynolds number tends to reduce the concentration factor values along the column since high feed velocity reduces the gas residence time in the bed and increases the unadsorbed CO₂.
Fig. 4: Effect of feed flow rate on bed CO$_2$ concentration factor at 20% CO$_2$ concentration.

3.3 Effect of feed velocity on bed CH$_4$ concentration factor:
The concentration gradient inside the bed for CH$_4$ is shown in Figure 5, where the gas is adsorbed under different feed velocities.

Fig. 5: Effect of feed velocity on CH$_4$ concentration gradient at 80% CH$_4$ concentration.

The following Figure 6 presents the influence of feed flow rate on concentration factor for methane. Different feed velocities have been selected to study the process performance for a packed bed column with the specifications mentioned in Table 1 and Table 2. From Figure 6, it can be concluded that as the flow rate increases, higher product recovery in terms of CH$_4$ recovery can be achieved along the column. Higher feed velocity speeds up the transport of gas, thus, as Reynolds number increases, the gas leaves the column faster which leads to an increase of product recovery.

Fig. 6: Effect of feed flow rate on bed CH$_4$ concentration factor at 80% CH$_4$ concentration.
Conclusion:

In this study, CFD simulation is used to simulate transport and adsorption phenomenon of the CO$_2$-CH$_4$ fixed bed adsorption column. The convective-diffusive mass transfer model is based on LDF and thermal effects are neglected. The effect of feed flow rate along the length of the column is obtained by varying the flow rate for different values and observing its effect on the adsorption process. The study shows that the hydrodynamics within the bed significantly influences the performance of adsorption process.

Nomenclature:

- $F$: Fluid-particle friction forces (N/m$^3$)
- $K$: Permeability (m$^2$)
- $U$: Intersitial velocity (m/s)
- $U_{sl}$: Superficial Velocity (m/s) ($= \epsilon_i U$)
- $d_p$: Particle diameter (m)
- $C_{bulk}$: Bulk concentration (kg/m$^3$)
- $C_p$: Intraparticle concentration (kg/m$^3$)
- $K_i$: Mass transfer coefficient of species i (m/s)
- $q_i$: Average adsorbed phase conc. of i (kg/m$^3$)
- $q_{m}$: Maximum adsorbed conc. (kg/m$^3$)
- $b_i$: Equilibrium constant

Greek characters:

- $\epsilon$: Porosity
- $\mu$: Gas viscosity (Pa.s)
- $\rho$: Gas density (kg/m$^3$)
- $\beta$: Non-Darcy coefficient (m$^{-1}$)

ACKNOWLEDGMENT

The authors gratefully acknowledge the support of Universiti Teknologi PETRONAS for providing grant and facilities for the research.

REFERENCES


