Simulation of a fluidized-bed steam reformer

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Abstract—Considering improved heat transfer, uniformity of catalyst distribution and virtual elimination of diffusion limitations as the major advantages of fluidized beds, it seems preferable to employ such a reactor for reforming of natural gas, where the oxygen catalytically reacts with methane in order to provide the energy required for endothermic reforming reactions. In this study, a simulation model is developed in order the performance of the fluidized bed reactor of steam methane reforming to be predicted. A two phase model is adapted for this purpose. In this model, the bed is divided into several sections. At each section, the flow of the gas is considered as plug flow through the bubble phase and perfectly mixed through the emulsion phase. The results of the model are compared with the experimental data from a pilot scale fluidized bed reformer in terms of methane conversion at different operating conditions. Good agreement is found between the model predictions and the experimental data. The range of applicability of the model is also investigated in this study.

INTRODUCTION

Conventional fixed bed reformers suffer from some disadvantages such as low heat transfer rates, diffusional resistance in the catalyst pores and non-isothermicity. The highly endothermic steam methane reforming process is largely affected by the efficiency of heat input into the reformer; therefore, the choice of the reformer would be of prime importance. Apart from the improved heat transfer, catalyst bed uniformity and high mass transfer rate could be considered as advantages of using fluidized beds. There are, however, some uncertainties associated with the scale-up of fluidized bed reformers. In fact, this is the main obstacle, which impedes their widespread usage in industry. Thus, there is a real need to develop a comprehensive model for modeling the fluidized bed reformers. Since both physical and chemical phenomena coexist in fluidized beds, the corresponding submodels are required to be considered in the modeling. These submodels are the hydrodynamic and the reaction submodels. The hydrodynamic sub-model describes the properties of bubble and emulsion phases and their interactions.

The hydrodynamics of fluidized beds are the most complex among the gas-solid contactors such as fixed and moving beds. All the various available hydrodynamic models are categorized in three general groups: single, two, and three-phase models. In the two-phase concept of fluidization, the fluidized bed is divided into two phases, i.e., bubble and emulsion phase. Early two-phase models consider solid-free bubbles and the emulsion phase at minimum fluidizations. This assumption has been proved not to be satisfactory since the existence of solid particles in bubbles has been shown both experimentally and theoretically.1–3 The reaction sub-model explains the chemical reaction occurring in the bed.

Commercial process simulators that are widely employed in chemical process industries (CPI) are not generally strong in reactor modeling and only contain standard, ideal reactors, such as plug flow reactors (PFR) and Continuous stirred tank reactors (CSTR). Despite their wide applications in CPI, no model for fluidized bed reactors is offered in these simulators. In this work, standard reactor modules available in process simulators are combined in a logical way so that the fluidized bed steam methane reformers to be represented considering the reality of these reactors.
The details of such combination and the corresponding equations and expressions are detailed in the following sections.

**MODEL DESCRIPTION**

In this work, the fluidized bed steam reformer is modeled considering the hydrodynamics complexity and the reactions occurred within the bed. A combination of several ideal reactors is used for this purpose. The reactor is axially divided into several sections. In each section, two types of reactors are considered, i.e., a PFR reactor, which represents the gas flow through the bubbles, and a CSTR, which represents the gas flow through the emulsion. For the modeling purposes, it is assumed that at each section, the reactions and effluents mixing take place respectively. The $i^{th}$ stage of the model is illustrated schematically in Figure 1.

![Figure 1](image-url)  
**Figure 1.** Schematic diagram of the $i^{th}$ stage

The experimental methane reforming reactor from the literature is adapted for the model developed in this study. The reformed consists of the main body (OD = 114.3 mm, ID = 97.2 mm), tapered section, wind box (depth=25.4 mm, ID=88.9 mm), expanded section (OD = 219 mm, ID = 194 mm) and the distributor plate. The main body is surrounded by two semi-cylindrical electric heating elements each of 2.8 kW. At the autothermal conditions, the heat needed for the reaction was provided internally by the exothermic methane combustion. It is important to mention that the hydrodynamics of both phases is characterized by the dynamic two-phase (DTP) model and the bubbles reach their equilibrium size quickly above the distributor. Therefore, the bubble diameter is assumed to be constant along the bed height. The radial gradients along the bed are neglected and the axial profiles are only considered in this study.
**Governing equations**

The mass balance equations for both bubble and emulsion phase in each section of the reactor are as follows:

**Bubble phase:**

\[ C_{Ab(i-1)} U_b A_b - A_b \varepsilon_b \int_{z_{i-1}}^{z_i} r_{A(i)} dz - K_{be} (C_{Ab(i)} - C_{Ae(i)}) V_{bi} - C_{Ab(i)} U_b A_b = 0 \]  

(1)

**Emulsion phase:**

\[ C_{Ae(i-1)} U_e A_e - r_{A(i)} V_{Ci} + K_{be} (C_{Ab(i)} - C_{Ae(i)}) V_{ei} \delta_i - C_{Ae(i)} U_e A_e = 0 \]

(2)

**Volume of the i-th stage:**

\[ V_i = \frac{V_i}{n} \]

(3)

**Volume of bubble phase and emulsion phase in each stage:**

\[ V_{bi} = V_i \delta \]

\[ V_{ei} = V_i (1 - \delta) \]

(4)

**Volume of PFR and CSTR in each stage:**

\[ V_{Pi} = V_{bi} \varepsilon_b \]

\[ V_{Ci} = V_{ei} \varepsilon_e \]

(5)

In this model, number of divisions of the reactor is determined based on the experimental data. The hydrodynamic and mass transfer coefficients (bubble velocity and diameter, phase fraction, voidage, mass transfer coefficient), which are needed to solve the Equations (1) and (2), are obtained from the literatures.5–7

Two reforming and two combustion reactions are considered in the modeling as given below:

1. First reforming reaction (CH\(_4\) + H\(_2\)O ↔ CO + 3H\(_2\))

\[ r_1 = \frac{k_1 (P_{CH_4} P_{H_2O}^{0.5} / P_{H_2}^{1.25}) (1 - (P_{CO_2} P_{H_2}^{3} / K_{P_1} P_{CH_4} P_{H_2O}))}{(1 + K_{CO} P_{CO} + K_{H} P_{H_2}^{0.5} + K_{H_2O} P_{H_2O} / P_{H_2})^2} \]

(6)

2. Second reforming reaction (CO + H\(_2\) ↔ CO\(_2\) + H\(_2\))

\[ r_2 = \frac{k_2 (P_{CO} P_{H_2O}^{0.5} / P_{H_2}^{0.5}) (1 - (P_{CO_2} P_{H_2}^{3} / K_{P_2} P_{CO} P_{H_2O}))}{(1 + K_{CO} P_{CO} + K_{H} P_{H_2}^{0.5} + K_{H_2O} P_{H_2O} / P_{H_2})^2} \]

(7)

3. Third reaction equation (CH\(_4\) + 2H\(_2\)O ↔ CO\(_2\) + 4H\(_2\))

\[ r_3 = \frac{k_3 (P_{CH_4} P_{H_2O} / P_{H_2}^{1.25}) (1 - (P_{CO_2} P_{H_2}^{3} / K_{P_3} P_{CH_4} P_{H_2O}))}{(1 + K_{CO} P_{CO} + K_{H} P_{H_2}^{0.5} + K_{H_2O} P_{H_2O} / P_{H_2})^2} \]

(8)

4. Methane oxidation (CH\(_4\) + (2–\(\alpha\)/2) O\(_2\) ↔ \(\alpha\)CO + (1–\(\alpha\))CO\(_2\) + 2H\(_2\)O)

\[ r_4 = Ky_{CH_4} Y_{O_2}^n \]

(9)
RESULTS AND DISCUSSION

The number of theoretical sections was calculated to be 3 based on the “J factor”. The model equations were then solved simultaneously with the aid of MATLAB programming, for set of experimental data available as shown in Table 1. The results then have been compared with the experimental data.

<table>
<thead>
<tr>
<th>Set No.</th>
<th>Steam flow rate (mol/h)</th>
<th>Temperature (°C)</th>
<th>Pressure (MPa)</th>
<th>Oxygen flow rate (mol/h)</th>
<th>Methane flow rate (mol/h)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>80</td>
<td>650</td>
<td>0.55</td>
<td>12–22</td>
<td>40</td>
</tr>
<tr>
<td>2</td>
<td>80</td>
<td>650</td>
<td>0.35–0.6</td>
<td>18</td>
<td>40</td>
</tr>
<tr>
<td>3</td>
<td>80</td>
<td>575–675</td>
<td>0.5</td>
<td>18</td>
<td>40</td>
</tr>
<tr>
<td>4</td>
<td>60–120</td>
<td>650</td>
<td>0.5</td>
<td>18</td>
<td>40</td>
</tr>
</tbody>
</table>

In Figure 2, the experimental data and the model predictions are plotted versus oxygen input flowrate for the changes in the methane conversion. As shown in the figure, the corresponding agreement was satisfactory. The irreversible reaction of methane to produce CO and/or CO$_2$ and H$_2$O can describe the reason of increasing the conversion of methane with the increase in the oxygen flowrate. In order to show the agreement of the results, the Pearson correlation coefficient was calculated. It was 0.9763 for this set of runs. Since the catalyst used in this work, slightly differs from the one the DTP model’s constants are evaluated for, the deviation seems reasonable.

![Figure 2. Variation of oxygen input flowrate](image)

Figure 3 compares the experimental and model results for methane conversion through the second set of runs. In this set, the methane conversion is decreased with an increase in the reactor pressure. This trend was expected as increasing pressure is thermodynamically unfavourable to the reaction system. The model prediction and the experimental data have good agreement.
In Figure 4, the predicted and the experimental data for third set of runs are compared. In this set, the methane conversion is increased with an increase in the reactor temperature. A rather close agreement is found between the experimental data and the model prediction. The same reason stated for the Figure 1 might be generalized for the deviation of calculated results from experimental ones in other this figure, as well.

The experimental and calculated data for the last set of runs are compared in Figure 5. In this set, the methane conversion is increased with an increase in the steam methane ratio. The results have similar trend with the experimental data and the comparison between the experimental data is quite satisfactory.
CONCLUSION

A model is adapted for a fluidized bed steam reformer. This model had been previously applied for simple first order reactions. In this work, however, the model is adapted for the multiple catalytic reactions of the steam reforming by developing the hydrodynamic and the reaction submodels. The hydrodynamic submodel is based on the two-phase concept of fluidization where the fluidized bed is divided into a number of equal-volume stages depending on the operating conditions and physical properties of the bed materials. In this model, in each stage, the bubble phase is considered as a PFR reactor and the emulsion phase is considered as a CSTR. The model developed in this work for the fluidized bed reformers can be easily introduced to the commercial process simulators since the corresponding sub-elements (PFR and CSTR modules) are already available in the simulators. The results obtained from the model indicate a good agreement with the experimental data. If the parameters of DTP model were evaluated specifically for the catalyst employed in this reactor, the model predictions would have been improved accordingly.

NOTATION

$A$  
cross sectional area, m$^2$

$C_A$  
concentration of component A, kmol/m$^3$

$H_a$  
Hata number, dimensionless

$I$  
dimensionless number ($H_aU_0/U_{mf}$)

$K$  
reaction rate constant

$K_1$  
constant, kmol/kgcat·s·kPa$^{0.25}$

$K_2$  
constant, kmol/kgcat·s·kPa

$K_3$  
constant, kmol/kgcat·s·kPa$^{0.25}$

$K_{CO}$  
adSORPTION coefficient, kPa$^{-1}$

$K_H$  
adSORPTION coefficient, kPa$^{-0.5}$

$K_{H2O}$  
adSORPTION coefficient, dimensionless

$K_{p1}$  
equilibrium constant, kPa$^2$

$K_{p2}$  
equilibrium constant, kPa$^3$

$K_{p3}$  
equilibrium constant, kPa$^2$

$K_{be}$  
mass transfer coefficient, s$^{-1}$

$P_A$  
partial pressure of component A, kPa

$m$  
constant, dimensionless

$n$  
constant, dimensionless

$r$  
reaction rate, kmol/m$^3$·s
\( U_b \) bubble velocity, m/s
\( U_e \) emulsion velocity, m/s
\( V_b \) bubble phase volume, m³
\( V_e \) CSTR volume, m³
\( V_c \) emulsion phase volume, m³
\( V_t \) total volume, m³
\( V_p \) PFR volume, m³
\( Y_i \) mole fraction of component \( i \), dimensionless

Greek symbols
\( \varepsilon_b \) bubble-phase voidage, dimensionless
\( \varepsilon_e \) emulsion-phase voidage, dimensionless
\( \delta \) bubble-phase fraction, dimensionless

Subscripts
\( b \) bubble phase
\( e \) emulsion phase
\( i \) section number

REFERENCES