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Hydrodynamic Characteristics of a Novel Circulating Fluidized Bed Steam Reformer Operating in the Fast Fluidization Regime*

Moataz Bellah M. Mousa, Seif-Eddeen K. Fateen, and Essam A. Ibrahim

Abstract

Circulating Fluidized Bed Steam Reformers (CFBSRs) represent an important alternative for the production of syngas for the Fisher-Tropsch (FT) process and for hydrogen production. Most research regarding this novel CFBSRs did not consider its hydrodynamic characteristics. In this work, the riser Computational Fluid Dynamics (CFD) simulations were investigated using two phase Eulerian-Eulerian approach coupled with kinetic theory of granular flow with k-epsilon model to describe the turbulence of each phase. The model equations were solved via the commercial CFD package FLUENT, which uses the finite volume numerical approach. Cold flow simulations were carried out under the fast fluidization regime and results were validated qualitatively against available experimental data. The radial segregation of the catalyst, the velocity distribution of both phases and other characteristics of the flow were captured by the simulation. This work showed that for operation under high density and high flux conditions, solids flux should be higher than 300 kg/m²s and inlet void fraction lower than 85%.

KEYWORDS: Computational fluid dynamics, fluent, fast fluidization, steam reforming, hydrogen production, circulating fluidized bed reactor, two-phase flow

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Introduction

Syngas, a direct product of steam reforming, formed of carbon monoxide, carbon dioxide and hydrogen, has many uses in fuels industry, most importantly in its use in Fischer-Tropsch (FT) processes to produce liquid fuels, e.g. Gas To Liquid (GTL) Processes. The more valuable component is hydrogen, which can be separated from the mixture for its own important uses.

Steam reforming of hydrocarbons (methane as well as higher hydrocarbons) is the main process for production of syngas/H₂ (Christensen, 1996; Czernik et al., 2000; Elnashaie and Elshishini, 1993; Froment, 2000; Hou et al., 2000; Phillips et al., 1969; Rostrup-Nielsen, 1977; Rostrup-Nielsen, 1973; Twigg, 1989; Wang et al., 1997; Xu and Froment, 1989). Industrial steam reforming is commonly carried out in fixed bed reactors made up of a large number of tubes packed with nickel catalyst particles and surrounded top/side fired furnaces to supply the required energy for the endothermic reactions. However, these designs are not very efficient and not suitable for higher hydrocarbons. New designs are continuously proposed for reformers to overcome accompanying mass transfer, equilibrium limitations and catalyst deactivation problems. One of the most important and successful improvements was the Bubbling Fluidized Bed Membrane Steam Reformer (BFBMSR) (Adris et al., 1991; Adris et al., 1994, 1997) which was recently followed by a novel better design proposed by Chen et al. (Chen et al., 2003a; Chen et al., 2003b, c). The novel design is a Circulating Fluidized Bed Membrane Steam Reformer (CFBSR), which is commonly known as 3rd generation steam reformers, which resembles in principle the modern Fluidized Catalytic Cracking (FCC) units.

The basic structure of this novel configuration is schematically shown in Figure 1. It consists of three main parts: a riser, where steam reforming takes place; a gas-solid separator to separate the catalysts from the product gas stream; and the downer where the recycled catalyst is regenerated by the introduction of air to burn carbon formed on the catalyst. Burning carbon off the catalyst in the downer supplies necessary heat for the endothermic reforming reactions and through careful design auto-thermicity can be achieved. Inside the riser, nickel reforming catalyst is fluidized where high inlet gas velocity can allow higher productivity than prior reformers. The riser contains palladium-based hydrogen permselective membrane tubes to continuously remove produced hydrogen using sweep gas as low value stream. The presence of such membranes "breaks" the equilibrium limitations present due to the reversibility of the reactions (Chen and Elnashaie, 2004a, b; Chen et al., 2004). The novel design preserves the main advantage of the BFBMSR of the high reaction effectiveness factor (~1) because of the use of powdered catalyst in both cases.
In general, CFBSRs can be operated under different fluidization regimes; the most common is Fast Fluidization (FF) and this investigation will concentrate on this regime.

The FF regime is characterized by the well-known Core-Annulus structure where an annular structure of down-flowing solids is observed near the column walls while the particles load decreases considerably around the column center.

The remainder of the paper is organized as follows: A comparison between CFBSR and a similar steam reforming reactor is presented in the following section. Next, the previous attempts at modeling CFBSR are presented followed by a description of the scope and novelty of this work. The details of the CFD model are then introduced followed by the presentation of results and their discussion. The conclusions of this work are summarized at the end of the paper.
Comparison between CFBSR and the Oxy-Fuel Mass Recirculation Process

An oxy-fuel hydrogen generation process was recently introduced in the literature (Budzianowski, 2010a, b). This oxy-fuel recirculation process shares some similarities and some differences with CFBSR.

The oxy-fuel mass recirculation process is similar to the CFBSR as both processes require hydrogen and carbon monoxide separation. Both designs have carbon monoxide separation ex-situ. In-situ hydrogen separation, which is included in CFBSR design, generally offers more benefits.

An obvious difference between both systems is the reaction chamber design itself. In our case it utilizes a circulating catalyst, compared to a fixed bed wall coated catalyst in the oxy-fuel mass recirculation process. This difference leads to many pros for the suggested CFBSR over the other design. Catalyst circulation allows for continuous catalyst reactivation and thus much less need for shutdown. Furthermore, the fluidization state allowed for overcoming all the challenging mass transport limitations reported by in the suggested oxy-fuel mass recirculation process publications. The effectiveness factor in CFBSR is at least order of magnitude higher than that at the reactor of oxy-fuel mass recirculation process.

Finally, we need to shed some light on the relative efficiency of both designs. Although the reported reactor length for the oxy-fuel mass recirculation process is 2mm compared to the 2m length of the CFBSR, the used gas flow rate in our simulations (~10-2Kg/s) was 6 orders of magnitudes higher than that used in the oxy-fuel mass recirculation process simulations (~10-8 Kg/s). This huge difference in flow rates should reflect difference in intended applications and reactor length efficiency, where CFBSR gives higher conversion per unit length and actually that was expected due to the higher effectiveness factor.

Previous Modeling Attempts for CFBSR and Scope of This Work

In all the work done by Chen et al. for modeling the CFBSR (Chen and Elnashaie, 2004a, b, 2005; Chen et al., 2003a; Elnashaie et al., 2005), an ideal one-dimensional plug flow model was used with no axial dispersion. The velocity of the gas and solid phases were determined from the minimum fluidization velocity and from the range of FF regime (Kunii and Levenspiel, 1997), with no slip between the two phases (class I models as defined by (Harris and Davidson, 1994). This model assumes plug flow in the axial direction and complete radial mixing. These assumptions will affect the accuracy of the model predictions since the hydrodynamics is an important phenomenon that should be taken into account.
in modeling CFBSRs. Computational Fluid Dynamics (CFD) is a powerful tool to investigate these effects.

The modeling of the complex hydrodynamics and reaction behavior inside the CFBSR is to be performed in stages. The scope of this work is limited to the simulation of the cold case without membranes, which constitutes the first stage for complete understanding of the flow behavior inside the reactor. This stage includes the flow characterization under different fluidization regimes. The following stages would include the coupling between the flow behavior with the reaction kinetics. Since those two phenomena occur at different time scales, our preliminary attempts at modeling them simultaneously showed that larger computational power and longer simulation times were needed. The inclusion of the membranes will also affect the flow behavior. Both of these features will be investigated in our future work.

This work focuses on the development of a two-dimensional CFD model for the riser part (without membranes) of the novel design of the CFBSR and the numerical solution of this multiphase flow system is done using the commercial CFD package FLUENT 6.3.2. The dimensions of the riser were taken appropriately for the small-scale production of hydrogen for fuel cell applications (Chen and Elnashaie, 2004b; Chen et al., 2004).

The originality of this work lies in the CFD simulation of a novel hydrogen steam reformer, which enhances the understanding of the complicated flow behavior inside the reactor riser. The hydrodynamic behavior inside the riser will be impacted by the inclusion of the material transport through the membranes and the inclusion of the chemical reactions. However, the scope of this work is limited to the quantitative description of the cold case in the fast fluidization regime as a first step for the complete description of the reactive riser. To our knowledge, no published results are available for the simulation of the fluidized-bed riser designed for the hydrogen steam reforming.

CFD modeling

Since the CFBSR operates under dense conditions of voidage, $\varepsilon_p$, it is a four-way coupling problem according to Elghobashi's classification (Elghobashi, 1991). The following interactions need to be modeled to accurately describe the system:

- Gas phase flow affects the catalyst particles through drag and turbulence.
- Catalyst particles influence gas flow by reducing its mean momentum and suppressing its turbulence.
- Catalyst particles interact together due to collisions through particle pressure and viscous stresses.
- Catalyst particles collision with the walls of the riser.
In this work, the Eulerian-Eulerian multiphase modeling was used to describe the flow inside the CFBSR. In such modeling type, the phases are considered to be continuous and fully interpenetrating. The interpenetrating phases can exchange mass, momentum and energy. Each phase is described by a set of conservation equations derived from single flow equations and averaged either in time (Drew, 1983) and space (Ahmadi, 1987; Harlow and Amsden, 1975) or using ensemble averaging (Enwald et al., 1996). This leads to equations nearly similar to the single phase ones but with additional terms to account for the property exchange among phases. These equations are averaged with the introduction of the volume fraction, $\alpha$, representing the volume occupied by each phase in a control volume.

Beside the conservation equations, many constitutive (closure) equations are needed either to model exchange factors between phases or to model rheological flow properties for solids. The latter is commonly modeled using the Kinetic Theory of Granular Flow (KTGF) which is based on the granular temperature that is equivalent to thermal temperature in kinetic theory of gas flow. The granular temperature ($\Theta$) measures particles kinetic energy oscillation due to collisions and fluctuating motions of the particles and is equal to one third of the square of velocity variations.

The key to accurate modeling lies to a large extent in the closure laws used. A considerable number of models have been developed for this purpose but the most suitable ones are shown next. The suitability was examined through many trial cases of the different closure laws based on numerical stability and description of flow physics. The balance equations (conservation and constitutive) described next are those used by the commercial CFD package FLUENT (Fluent Inc., USA) and are reported in its documentation (Fluent, 2006).

**Eulerian - Eulerian model with KTGF**

The rate of mass accumulation per unit volume and the net rate of convective mass flux are related by the continuity equation of phase “q”

$$\frac{\partial}{\partial t} (\alpha_q \rho_q) + \nabla \cdot (\alpha_q \rho_q \mathbf{v}_q) = 0$$

(1)

While the species balance equation for phase “q” is given by

$$\frac{\partial}{\partial t} (\alpha_q \rho_q \mathbf{x}_q) + \nabla \cdot (\alpha_q \rho_q \mathbf{v}_q \mathbf{x}_q) = \nabla \cdot (\alpha_q \rho_q \mathbf{D}_k \nabla \mathbf{x}_q)$$

(2)
The momentum balance for phase “q” relate the transient and convective momentum transfer with the stress-strain tensor \( \tau_q \) for phase q, the pressure of all phases and inter-phase momentum exchange term as follows

\[
\frac{\partial}{\partial t}(\alpha_q \rho_q v_{-q}) + \nabla \cdot (\alpha_q \rho_q v_{-q} v_q) = \nabla \cdot \tau_q - \alpha_q \nabla p + \alpha_q \rho_q g + F_q
\]  

(3)

F_q represents the inter-phase momentum exchange term between phase "q" and the other phase present in the system and is equal to \( K_{gs}(v_g - v_s) \). \( K_{gs} \) is an inter-phase exchange coefficient which was modeled using (Syamlal and O'Brien, 1989; Syamlal et al., 1993) model.

**Energy Equation**

The energy balance equation for phase q is

\[
\frac{\partial}{\partial t}(\alpha_q \rho_q h_q) + \nabla \cdot (\alpha_q \rho_q v_q h_q) = -\alpha_q \frac{\partial p_q}{\partial t} + \nabla \cdot v_q + Q_{pq}
\]  

(4)

The rate of energy transfer between phases is assumed to be a function of the temperature difference, for example rate of heat transfer from gas to solid is

\[
Q_{gs} = h_{gs}(T_g - T_s)
\]  

(5)

and

\[
h_{gs} = \frac{6k_g \alpha_s \alpha_g \text{Nu}_g}{d_s^2}
\]  

(6)

FLUENT uses the following Nusselt number correlation suggested by Gunn (Gunn, 1978). In addition to mass, momentum and heat conservation equations for the solid phase, a fluctuation of kinetic energy equation was also solved by applying KTGF to consider the energy fluctuation of solid particles.

The transport equation for the granular temperature, derived from kinetic theory, takes the form (Ding and Gidaspow, 1990). The constitutive equations for granular flow can be divided into two subsections: Granular temperature closures and KTGF models including viscosities, solids pressure and other rheological factors for granular phase. The definitions of those equations are summarized in Table 1 below.
The different models and constitutive equations contain many parameters as maximum packing limit ($\alpha_{s,max}$), the radial distribution function ($g_{0,ss}$) and solid-solid restitution coefficient ($e_{ss}$). The maximum packing limit is a limit set on the maximum packing of solids at any grid point to prevent unphysical clustering in the simulations and is usually derived from the case of a packed bed, which represent the maximum physical packing limit. Its common values are 0.60 (Benyahia et al., 2000), 0.63 (Yang, 2003) and 0.64 (Jiradilok et al., 2006) with 0.63 being the commonly used value in the literature and also used in this work. The radial distribution function is a correction factor that is used to account for the increase in probability of collisions between particles in dense cases. Thus, it can be observed that at very dilute conditions "$g_0$" approaches one as the distance between solids goes to infinity and at very dense regions "$g_0$" goes up to infinity. The correlation used for unit-size cases was suggested by (Lun and Savage, 1986).

<table>
<thead>
<tr>
<th>Table. 1. Summary of the used E-E model</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Stress-Strain Tensor</strong></td>
</tr>
<tr>
<td>$\tau_g = \alpha_g \mu_{g,\text{eff}} (\nabla v_g + \nabla v_g^T) + \alpha_g \left( \lambda_g - \frac{2}{3} \mu_g \right) \nabla \cdot v_g \mathbb{I}$ (6)</td>
</tr>
<tr>
<td>$\mu_{g,\text{eff}} = \mu_g + \mu_{g,t}$ (7)</td>
</tr>
<tr>
<td><strong>Solids phase closures</strong></td>
</tr>
<tr>
<td><strong>Solids Pressure</strong></td>
</tr>
<tr>
<td>$p_s = 2 \rho_s (1 + e_{ss}) \alpha_s^2 g_{0,ss} \Theta_s$ (8)</td>
</tr>
<tr>
<td><strong>Solids Shear viscosity</strong></td>
</tr>
<tr>
<td>$\mu_{s,\text{eff.}} = \mu_{s,\text{col}} + \mu_{s,\text{kin}} + \mu_{s,\text{friction}} + \mu_{s,\text{turb.}}$ (9)</td>
</tr>
<tr>
<td>$\mu_{s,\text{col}} = \frac{4}{5} \alpha_s \rho_s d_s s_{g,ss}(1 + e_{ss}) \left( \frac{\Theta_s}{\pi} \right) \frac{1}{2}$ (10)</td>
</tr>
<tr>
<td>$\mu_{s,\text{kin}} = \frac{\alpha_s \rho_s d_s \sqrt{\Theta_s \pi}}{6(3 - e_{ss})} \left[ 1 + \frac{2}{5}(1 + e_{ss})(3e_{ss} - 1) \alpha_s g_{0,ss} \right]$ (11)</td>
</tr>
<tr>
<td>$\mu_{s,\text{fr}} = \frac{p_s \sin \phi}{2 \sqrt{T_{2D}}}$ (12)</td>
</tr>
</tbody>
</table>
Solids Bulk Viscosity

\[
\dot{\lambda}_s = \frac{4}{3} \alpha_s \rho_s d_s g_{0,ss} (1 + e_{ss}) \left( \frac{\Theta_s}{\pi} \right)^{\frac{1}{2}}
\]

Granular Temperature Closures

Granular Energy diffusion coefficient

\[
k_{th} = \frac{15 d_s \rho_s \alpha_s \sqrt{\Theta_s / \pi}}{4(41 - 33 \eta)} \left[ 1 + \frac{12}{5} (4 \eta - 3) \eta^2 \alpha_s g_{0,ss} + \frac{16}{15 \pi} (41 - 33 \eta) \eta \alpha_s g_{0,ss} \right]^{-\frac{1}{2}}
\]

\[
\eta = \frac{1}{2} (1 + e_{ss})
\]

Collision dissipation energy

\[
\gamma_{es} = \frac{12 (1 - e_{ss}^2) g_{0,ss} \alpha_s^3 \Theta_s^3}{d_s \sqrt{\pi}} \rho_s \alpha_s^2 \Theta_s^2
\]

Turbulence model

In comparison to single-phase flows, the number of terms to be modeled in the momentum equations in multiphase flows is large, and this makes the modeling of turbulence in multiphase simulations extremely complex.

In the \((k-\varepsilon)\) turbulence model, the continuous phase turbulence is modeled using transport equations for turbulent kinetic energy and turbulent energy dissipation rates. The transport equation for the turbulent kinetic energy, \(k\), for gas phase include terms that represent the interphase exchange turbulent momentum transfer for the gas and solid phases. The default values for the constants that appear in the turbulent energy dissipation were used due to the lack of better values fitted to experimental data.

Finally, the turbulent viscosity is written in terms of the turbulent kinetic energy and turbulent dissipation rate as follows

\[
\mu_{t,g} = \rho_g C_{\mu} \frac{k_g^2}{\varepsilon_g}
\]

The previous equations were solved to calculate value for the eddy viscosity, \(\mu_{t,g}\), this value is then added to gas dynamic viscosity giving \(\mu_{g,eff}\) which is present in the gas stress tensor. Thus the effective fluid phase shear viscosity is, \(\mu_{g,eff} = \mu_g + \mu_{t,g}\).
The same approach was applied for the solid phase where a turbulent component of the solids viscosity could also be calculated by solving two extra transport equations for turbulent kinetic energy and dissipation of the solids phase, then the effective solids viscosity is, $\mu_{s,eff} = \mu_s + \mu_{s,t}$.

**Solution procedure**

The riser was simulated as a 2-D axisymmetric case (Ranade, 1999). The main justifications for using symmetric assumption are the use of common inlet and common outlet for both gas and catalyst, modeling the developed zone of the riser only, and to decrease the computational load by solving lower grid size. The grid was generated with total number of 36,000 cells and was non-uniformly distributed in the radial direction; more cells were placed near the wall. An equally spaced grid was applied for the axial direction. A close view for how the grid was spaced is shown in Figure 2.

In order to ensure numerical stability, a Courant number, $C$, has to be less than 1 (Ferziger and Peric 2002). $C$ is a function of the smallest cell dimension and largest instantaneous velocity:

$$C = \frac{u_{max} \Delta t}{\Delta z_{min}}$$

Time steps of 1E-5 to 1E-4 which correspond to courant numbers of 0.3-0.9 at various conditions were used.

The simulations were started with time step of 1E-5 and then increased gradually in intervals of 5E-6 to reach 1E-4. Increasing in time step was done over long periods of time and after ensuring that results were stabilized at the prior time step value. A relative error between two successive values of $10^{-3}$ as the convergence criterion for each scaled residual component was enough to achieve good results. The typical computational time for this simulation was 12–16 days on a 2 GHz Quad core workstation.

The finite volume approach was used to solve the non-linear coupled partial differential equations and the commercial software FLUENT (ANSYS, USA) was used to provide this numerical solution. The phase-coupled SIMPLE (PC-SIMPLE) (Patankar, 1980; Vasquez and Ivanov, 2000) algorithm was used, which is an extension of the SIMPLE (Semi-Implicit Method for Pressure-Linked Equations) algorithm to multiphase flows, to deal with pressure–velocity coupling. In this algorithm, the coupling terms were treated implicitly and formed part of the solution matrix. The Inter-Phase Slip Algorithm (IPSA) method was also used to deal with the coupling between the continuity and the velocity; it uses the Partial Elimination Algorithm (PEA) developed by Spalding to decouple the
drag forces. Moreover the Full Elimination Algorithm (FEA) (Benyahia et al., 2002; Benyahia et al., 2000) was used to assist convergence because of the high coupling between momentum and energy equations through exchange terms "inter-equation coupling" (Dartevelle, 2005) for both phases, thus affecting the numerical scheme. In addition, to improve computational speed, algebraic multi-grid cycles were used for many variables, especially pressure and granular temperature, both of which have shown very slow convergence. The solution time was greatly reduced by using V and W cycles for slow-converging variables. The work of Benyahia et al (Benyahia et al., 2002; Benyahia et al., 2000) showed that the final results are not sensitive to initial conditions.

Figure 2. Enlarged section of the grid pattern used in this work.

The boundary equations applied for the solid tangential velocity and granular temperature at the wall are

\[
v_{s,w} = - \frac{6 \mu_s a_{s,max}}{\sqrt{3} \pi \rho_s \alpha_s g_{0,ss} \sqrt{\Theta_s}} \frac{\partial v_{s,w}}{\partial n} \tag{19}
\]

\[
\Theta_{s,w} = - \frac{k_s \Theta_s \partial \Theta_{s,w}}{\gamma_{s,w}} + \frac{\sqrt{3} \pi \rho_s \alpha_s v_{s,ss}^2 g_{0,ss} \Theta_s^{3/2}}{6 a_{s,max} \gamma_{s,w}} \tag{20}
\]

Where \( \gamma_{s,w} \) is the collisional dissipation at wall and equals to

\[
\gamma_{s,w} = \frac{\sqrt{3} \pi (1 - e_{sw}^2) \rho_s \alpha_s g_{0,ss} \Theta_s^{3/2}}{4 a_{s,max}} \tag{21}
\]

\( \phi \) is the specularity coefficient; a measure of the fraction of tangential collisions which transfer momentum to the wall. It is equivalent to one minus the
tangential restitution coefficient (Ranade, 2002) and was previously estimated to
range from 0.5 to 0.6 (Johnson and Jackson, 1987; Ranade, 1999). Recently,
(Almuttahar and Taghipour, 2008) reported that a specularity coefficient of near
zero (which corresponds to free-slip at the wall) gave better agreement with
experimental results in a high density CFB which agreed with Ranade's findings
(Ranade, 1999) that decreasing the specularity coefficient leads to a flatter solids
velocity profile. The sensitivity of the results of some cases to the specularity
coefficient is examined in this study.

The operating conditions and dimensions of the reactor simulated in this
work are given in Table 2.

Table 2. Operating conditions and dimensions used in current work

<table>
<thead>
<tr>
<th><strong>Riser</strong></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Inner diameter of the riser (m)</td>
<td>0.0978</td>
</tr>
<tr>
<td>Height of the riser (m)</td>
<td>2.0</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th><strong>Inlet components composition (%)</strong></th>
</tr>
</thead>
<tbody>
<tr>
<td>CH₄ (Kmol/h)</td>
</tr>
<tr>
<td>H₂O (Kmol/h)</td>
</tr>
<tr>
<td>CO₂ (Kmol/h)</td>
</tr>
<tr>
<td>H₂ (Kmol/h)</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th><strong>Operating conditions</strong></th>
</tr>
</thead>
<tbody>
<tr>
<td>Temperature (K)</td>
</tr>
<tr>
<td>Pressure (Kpa)</td>
</tr>
<tr>
<td>Density (Kg/m³)</td>
</tr>
<tr>
<td>Viscosity (Pa.s)</td>
</tr>
</tbody>
</table>

**Results and discussion**

**Catalyst properties**

Chen and his co-workers (Chen et al., 2003a; Chen et al., 2003b, c) used steam
reforming catalyst with the following properties for the novel CFBSR:

1- Catalyst particle diameter of 186 μm to ensure an effectiveness factor of unity
   without falling in the Geldart's type C particles range, which are difficult to
   fluidize.

2- Catalyst density equal to 2835 kg/m³ based on the work of (Elnashaie and
   Elshishini, 1993).
Inlet catalyst volume fraction of 0.2, based on (Kunii and Levenspiel, 1997). This falls into the range of Geldart type B particles as shown in Figure 3.

Figure 3. Type of particles used in the work of (Chen et al., 2003a; Chen et al., 2003b, c)

The use of catalyst of Geldart "B" type shall lead to excessive circulation of catalyst inside the riser. To investigate the effect of using catalyst with these properties in the riser, we solved a case with those catalyst properties with the same operating conditions shown in Table 2 and Table 3. The circulation pattern was clear and excessive, as expected, and shown in Figure 4, which presents the catalyst velocity vectors at section of the riser.

Figure 4. Excessive catalyst circulation inside riser in case of using catalyst of Geldart's type "B" particles. Velocity was averaged at the period between 20 to 40s.
Figure 5 shows that this circulation pattern is repeated four times all over the riser height. This excessive circulation of catalyst is not desired for catalytic reactors with high reaction rates, due to the fast catalyst deactivation (carbon deposition) and non-uniform catalyst residence time distribution occurs. For those reasons the used catalyst in the CFBSR was selected to be of Geldart's "A" type.

For catalyst of Geldart's "A" type (Figure 6), smaller size pellets should be used and of lower density if possible. In the next CFD simulations, catalyst particle diameter was set to 105µm based also on the work of (Adris et al., 1994), but using particles obtained from lower mesh size screening. Also, the catalyst density was set to 1990 kg/m³ based on that used by (Wesenberg, 2006). The inlet conditions for the FF regime are shown in Table 3.

<table>
<thead>
<tr>
<th>Operating condition</th>
<th>FF</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gs (kg/m²s)</td>
<td>1500</td>
</tr>
<tr>
<td>Gg (Kg/m²s)</td>
<td>3.4</td>
</tr>
<tr>
<td>£s</td>
<td>0.2</td>
</tr>
</tbody>
</table>

Table 3. Summary of used inlet conditions for FF

Figure 5. Contours of catalyst volume fraction all over the riser height. Velocity was averaged at the period between 20 to 40s.
The catalyst volume fractions for the FF case were nearly constant near the centre of the riser, with higher variations observed towards the riser walls as shown in Figure 7 for the enlarged section between 0.75m and 1.25m. This catalyst accumulation near walls and continuous formation and break down of catalyst clusters (red colors in Figure 7) are common features for FF flows.

Figure 8 shows the common core-annulus flow pattern, which characterizes the FF regime. The catalyst flows downwards at the walls, while a dilute gas-catalyst flow is taking place in the core of the riser with maximum axial velocity.

Fig.6. Properties of new proposed catalyst for efficient flow inside CFBSR

The Catalyst volume fraction axial profile for the FF case (Figure 9) shows the familiar S-shape profile reported by (Lim et al., 1995) for dense fast fluidized beds. As expected the average volume fraction through the riser was high due to the used high catalyst flux in the FF case (1500 kg/m².s).

Better contact between gas phase and catalyst usually results in better catalytic reactor performance, thus radial and axial mixing play important role in the CFBSRs. For very fast catalytic reactions like steam reforming better distribution of catalyst inside the riser is more important than long contact time. As in this type of high rates of reactions the side reactions forming the deactivating carbon are also fast and therefore catalyst gets quickly deactivated especially under the absence of mass transfer limitations in CFBSRs ( where the effectiveness factor, \( \eta \sim 1 \)). Therefore long residence time is not beneficial. Also reactor performance is highly affected by temperature distribution inside the riser since the main reaction is highly endothermic and the water-gas shift reaction (one of the main side reactions ) is exothermic thus any presence of cold spots
will favor (from thermodynamic equilibrium point view, but not from kinetics point of view) the side reaction and affect reactor yield and selectivity, therefore affecting the composition of the product syngas (increases CO₂ and H₂ on the expense of CO). Based on this simple preliminary analysis, the optimization of hydrodynamic conditions for CFBSR will be based on following criteria:

1- Axial mixing (Homogeneity of catalyst distribution in the axial direction)
2- Radial mixing (Homogeneity of catalyst distribution in the radial direction)
3- Catalyst residence time and residence time distribution (RTD)

Figure 7. Transient distributions of catalyst volume fraction at enlarged section in the riser operating at FF regime between 0.75m and 1.25m
The quality of axial mixing can be examined by comparing the axial change in mean time catalyst volume fraction. For radial distribution of catalyst, it is more preferable to have a flatter profile of catalyst volume fraction over the riser diameter which reveals better radial homogeneity, more suitable for steam reforming reaction. In other words, while the core-annulus structure is not avoidable in all regimes, it will be better to have lower catalyst concentration near the wall and thus better radial catalyst distribution. This will help evading high reactants concentration near the centre of the riser. The radial profiles of the catalyst axial velocity at different elevations shown in Figure 10 illustrates a relatively steep profile for the FF regime, which may suggest low efficiency of the catalyst mixing in the radial direction and may result in negative impact on the reaction quality. Changing the fluidization regime to Dense Suspension Upflow (DSU) may offer a better radial catalyst mixing. A CFD model of the riser operating in the DSU regime will confirm this claim.

Figure 8. Enlarged section near the wall at 1m of the riser after 40 seconds of the flow showing catalyst velocity vectors in FF case
The characterizing feature of FF regime is back mixing in the annulus. This back mixing increases catalyst residence time and affects the RTD inside the riser. Although this increase in residence time is suitable for other types of reactions like coal combustion and cracking (Chang and Zhou, 2003; Jung and Gamwo, 2008), in steam reforming, the catalyst is rapidly de-activated by formed
carbon and hence quick regeneration is essential which occurs outside the riser. More over the unequal RTD of catalyst inside the riser due to this back mixing will lead to occurrence of cold spots inside the riser which will favor the water-gas shift side reaction thus affecting the reactor yield and selectivity.

**Conclusions**

This preliminary hydrodynamic study of this novel and promising CFBSR was carried out under FF conditions and revealed some of the hydrodynamic characteristics of this novel unit, which will be useful in its more accurate design and simulation than that based on chemical reaction engineering facts alone. This work showed that the circulating fluidized bed steam reformer must be operated under high density and high flux conditions, according to Zhu et al. (74) definition, using nickel/alumina catalyst of Geldart's “A” type. Thus, the suggested properties for the catalyst are mean particle size of 105 µm and particle density of 1990 kg/m³. Those properties should assure small enough particle size to approach unity effectiveness factor, but not too small to have hard fluidization characteristics.

This work also showed that for the operation under high density and high flux conditions, adequate gas properties and flow rate together with catalyst flux and inlet void fraction have to be employed. In general, solids flux should be higher than 300 kg/m²s and inlet void fraction lower than 85%.

Further studies are needed and are underway covering other regions of fluidization, such as Dense Suspension Upflow (DSU) in order to find the optimal operating conditions from a hydrodynamics points of view and combine it with the optimal conditions from a chemical reaction engineering point of view to exploit the synergetic effect between them. The following steps include the build-up of a highly efficient pilot plant for further development of the process and its model development for design and simulation.

**Notation**

- $C_D$: drag coefficient
- $D$: diffusivity, m²/s
- $d_s$: diameter of the solid-phase particles, m
- $F$: inter-phase momentum exchange term
- $h$: specific enthalpy, J/kg
- $K$: inter-phase exchange coefficient
- $k$: thermal conductivity
- $K$: turbulent kinetic energy
- $Nu$: dimensionless Nusselt number
$k_{\theta s}$  diffusion coefficient  

$P$  pressure, Pa  

$Q_{pg}$  intensity of heat exchange between both phases  

$v$  velocity vector, m/s  

$v_{r,s}$  terminal velocity of the solid phase, m/s  

$x$  mass fraction  

**Greek Letters**  

$\alpha$  volume fraction  

$\rho$  density, kg/m$^3$  

$\tau$  Stress-strain tensor  

$\gamma_{\theta s}$  collisional dissipation of energy  

$\phi_{gs}$  energy exchange between the gas and solid phases  

$\varepsilon$  turbulent dissipation rate  

$\sigma$  turbulent Prandtl number  

$\mu_t$  eddy viscosity  

$\gamma_{s,w}$  collisional dissipation at wall  

$\phi$  specularity coefficient  

**Subscripts**  

$g$  gas  

$p$  particle  

$q$  phase q  

$s$  solid  

**Superscripts**  

$i$  species i  

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