CFD-PBE Numerical Simulation of CO₂ Capture Using MgO-based Sorbent

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1. Introduction

Presently, despite of the growing use of the natural gas in the United States, coal is still the major source used for electric power generation and most of the industrial CO₂ emission results from the combustion of coal. By cost effective capturing of the CO₂ before it is emitted to the atmosphere and then storing it, coal can continue to be used without affecting economic growth, while reducing carbon emissions to the atmosphere. Therefore, carbon capture and sequestration (CCS) is one of the key technologies needed to mitigate carbon dioxide emission from industrial sources and power plants.

Development of a regenerable dry sorbent-based technology such as sorption enhanced water gas shift (SEWGS), for use in an efficient power plant design (i.e., modern IGCC power plants) has gained a lot of attention in recent years. SEWGS is a high-pressure and high-temperature dry sorbent process that combines CO₂ capture with the equilibrium controlled WGS reaction that in turn increases CO₂ capture. If the required CO₂ removal is high, as it is the case in power plant applications, multiple reactors operating at progressively lower temperatures are required. However, if one of the Syngas products, either H₂ or CO₂, can be continuously removed from the reactor, i.e. by CO₂ capture, equilibrium will be shifted and the conversion of CO to CO₂ can be accomplished in a single reactor. Therefore, a SEWGS process for continuous separation of

CO₂ or H₂ is desirable. The sorbents augment or replace the CO conversion catalysts currently used in WGS reactors and improve overall WGS thermal efficiency. The major advantages of this high temperature sorbent include eliminating or reducing the amount of WGS catalyst required to fully shift the Syngas to CO₂ and H₂ and eliminating Syngas cooling/reheating that is necessary for current CO₂ separation systems (DOE/NETL, 2010).

Using a similar concept, Hassanzadeh and Abbasian (2010), and Zarghami and Abbasian (2012, 2013) have developed a regenerative high temperature CO₂ capture process that is potentially capable of removing over 98% of CO₂ from a simulated coal gas mixture at the IGCC conditions using a highly reactive and mechanically strong MgO-based sorbents. In addition, the sorbent exhibited some catalytic activity for the water-gas-shift (WGS) reaction at 300°C, increasing hydrogen concentration from 37% (inlet) to about 70% in the reactor exit. By modeling of the sorbent/catalyst performance in a packed bed, they showed that hydrogen concentration in the simulated coal gas mixture can be improved significantly. Their results indicate that both CO₂ capture and enhancement of hydrogen production can be carried out in a single unit (Hassanzadeh, 2007). Their proposed regenerative carbon capture process is outlined in Figure 1.

It has been shown that use of the circulating fluidized beds ensures continuous CO₂ removal processes based on the dry sorbent concepts (Yi et al., 2007). Circulating fluidized beds (CFB) reactors are among the most important devices in chemical and energy industries. One of the well-established applications of CFB reactors is in Fluid Catalytic Cracking (FCC) applications with more than seven decades history and more than 400 units in operation worldwide today (Chen, 2011). Gasification of coal and biomass, synthesis of Olefin from Methanol and chemical-looping are among relatively new applications of CFB reactors (Sundaresan, 2011). Using solid particles for CO₂ capture in a CFB reactor allows continuous carbon dioxide

removal in a relatively compact unit. The basic configuration of a CFB reactor consists of a riser where the particles are transported by the gas flow, a cyclone to separate gas and solid at the top of the riser, a standpipe (down-comer) to return the separated solid to the riser inlet and a flow controlling device (e.g. L-valve) to control the solid flow. Depending on the application, other vessels or reactors can be added to the system which makes the system more complex to design. For instance in processes that include a regenerable sorbent or catalyst a second fluidized bed reactor can be added between the down-comer and the L-valve to serve as a regenerator reactor.

Shimizu and coworkers (Shimizu et al, 1999) were among the first groups who proposed the concept of a dual fluidized bed for carbon dioxide capture using CaO solid sorbents for post-combustion applications. Following their work, application of CFB for carbon dioxide capture were studied by Abanades et al. (2007), Charitos et al. (2010), Abbasi and Arastoopour (2011), Nikolopoulos et al. (2012), and Breault and Huckaby (2013). The same concept is applicable to pre-combustion carbon capture applications using solid sorbents. Despite of the long history of operation that has led to a wealth of design and operational experience with these systems, confidence to design and build commercial plants without significant levels of pilot scale testing at various intermediate scales is still lacking. This is due to lack of complete understanding of the origin and nature of the inherently complex flow structures observed in these devices, and uncertainties as to how they change upon scale-up (Sundaresan, 2011).

Another challenge in the deployment of these novel technologies is the fact that the majority of the promising technologies for the CO₂ capture are still in lab or bench scales. To successfully scaled-up these processes, a powerful tool, i.e. Computational fluid dynamics (CFD), is needed to fill the gap between lab/bench scale and large scales needed for demonstration. CFD has been proven to be a cost effective tool for conducting virtual experiments, prototype testing, and

parametric studies. Analysis using CFD complements and reduces physical testing, and it can result in a significant time and cost savings (Mukhopadhyay et al., 2004; Arastoopour, 2013). Therefore, Computational Fluid Dynamics (CFD) and rigorous numerical modeling are essential to shed light on the complex behavior and flow structure in these systems and provide an attractive option to study and design of the proposed fluidization process required for continuous CO₂ removal using solid sorbents in a systematic and economically feasible way.

The motivation behind this study originates from the fact that rigorous CFD simulations of poly-disperse flows must include modeling of distribution and variation in particulate phase properties caused by particulate processes such as nucleation, growth, agglomeration, breakage and particularly chemical reactions. A computationally attractive way of computing the temporal and spatial changes in the particulate phase property distribution function is utilizing the population balance equation (PBE). Finite size domain complete set of trial functions method of moments (FCMOM) is a novel solution method among the “method of moment” family for population balance equation solution (Strumendo and Arastoopour, 2008). One of the objectives of this study is to deploy a two-way coupled Computational Fluid Dynamics-Population Balance Model (CFD-PBM) in simulations and design of the high temperature and high pressure regenerative process for CO₂ removal using MgO-based solid sorbents in a circulating fluidized bed reactor. The coupled method is based on the “Finite size domain Complete set of trial functions Method of Moment” (FCMOM) (Abbasi and Arastoopour, 2013). Another objective of this paper is to numerically assess the performance of the MgO-based sorbent (Abbasi et al., 2013 and 2014) in the proposed regenerative carbon dioxide capture and SEWGS process. For this purpose, we adopted a bench scale circulating fluidized bed reactor design from the National Energy Technology Laboratory (NETL) as our baseline (Breault et al., 2013; Clark et al., 2013).

2. Numerical Model

Carbon dioxide sorption takes place in the riser section of the CFB reactor; therefore, investigation of its hydrodynamics and its impact on the gas-solid mixing, conversion and selectivity is of a greater interest. In the present study, our focus is on the numerical simulation of the carbonator part that excludes the returning system. This approach has been used by many authors for similar processes (Benyahia et al., 2000; Koksai and Hamdullahpur, 2005; Abbasi and Arastoopour, 2011; Shuai et al., 2011, and Breault and Huckaby, 2013).

In order to obtain the solid circulation rate, we used the results of a full-loop cold flow simulation of the NETL C2U CFB reactor. The simulation were performed, as a part of an earlier study (Abbasi, 2013) using a 3D Eulerian-Eulerian CFD model and the results were validated against the reported experimental data. The full-loop simulations provided invaluable information on the hydrodynamics, pressure drop and most importantly the solid circulation rate of the system. However, the experiments were conducted at low pressure and low temperature conditions while our goal is to use the same reactor configuration for pre-combustion CO₂ capture at high pressure and high temperature (i.e. 425°C and 20 atm). Because of the effect of temperature and pressure on the viscosity and density of the fluidizing gas, a set of preliminary studies were performed to find the required inlet gas velocity at the target pressure and temperature that provides the same solid inventory in the riser. The details can be found elsewhere (Abbasi, 2013) and the results are reported in Table 1.

Having the solid circulation rate from the full-loop model at high pressure and high temperature, in this work our focus is on the simulations of the reactive flow in the riser and assessment of

theMgO-based sorbent performance in the adsorber reactor, using the two-way coupled CFD-PBM based on the FCMOM approach as illustrated in Figure 2 (Abbasi and Arastoopour, 2013).

2.1. Computational model

To develop a coupled CFD-PBE model, it is necessary to implement and solve the population balance equation in the CFD codes. Therefore, a set of transport equations based on the moments of the property distribution function was added to a commercial CFD code, ANSYS Fluent, for this purpose.

The original NETL C2U system is a bench scale circulating fluidized bed that unit uses a solid sorbent to separate CO₂ from a gas stream.

The riser part of the bench scale CFB reactor has an overall height of 3.35m. CO₂ reach gas enters the bottom of the carbonator and mixes with fresh sorbent. In the cold experiments with inert sorbent, fluidizing gas is air. Mean particle diameter is 185 microns with particle density of 2480 kg/m³. The sorbent particles mix with the coal gases adsorbing CO₂ into the particle through chemical reaction. The CO₂ laden particles flow up the riser, turn and flow into the cyclone. In a reacting C2U, CO₂-free gas is separated from particles in the cyclone and exits the system, and the CO₂ laden particles pass through a loop-seal and enter the regenerator where CO₂ is released from the sorbent-particles by heating up the spent sorbent. The CO₂-lean gas exits the C2U system and the regenerated sorbent particles continue through the loop to the next loop-seal. The fresh sorbent particles pass through the loop-seal to the adsorber and the process continues.

Figure 3 shows the geometry of the carbonator and the riser section of the reactor used in simulations. The operating and inlet conditions were set using the values presented in Table 1

and the pressure boundary condition was used at the outlet. Furthermore, at the wall no slip condition for the gas phase, and partial slip condition for the solid phase, based on Johnson and Jackson (1987) with specular coefficient of 0.2 were used. Restitution coefficient was set to 0.9. Spatial discretization of the governing equations was performed using a first-order upwind scheme along with a first-order and implicit temporal discretization. Pressure-velocity coupling was accomplished using the Phase Coupled SIMPLE algorithm. The time step of $10^{-4}s$ ensures convergence within 60 iterations per time step. In addition, four moments of the solid density distribution were tracked using the population balance approach.

The computational domain consists of approximately 87,000 cells with the same resolution used in the full-loop CFB simulations. As discussed by Nikolopoulos et al. (2012) grid resolution is a very important factor in numerical simulations of a CFB reactor when Eulerian two-fluid-model is employed. Since, grid independence studies require significant computational time in three-dimensional full-loop simulations, many researchers (Zhang and VanderHeyden, 2001; Ge et al., 2008; Nikolopoulos et al., 2012) have set a criteria based on the dimensionless grid size (i.e. ratio of the equivalent cell size to the particle size) that ensures grid independent results and to resolve the meso-scale structure (e.g. clusters) in the system. The smallest reported value for the dimensionless grid size is 21 reported by Nikolopoulos et al. (2012). On the other hand, Sundaresan (2011) reports that the validity of simulations with dimensionless grid size ratios less than 10 are questionable as well, due to a limit that non-continuum nature of particles imposes to the system. The average dimensionless grid size in our simulations is equal to 26, which according to Nikolopoulos et al. (2012) is sufficient to resolve the meso-scale structures in the system and provide a grid-independent solution.

2.2. CFD Model

The CFD multiphase model is the well-known Two-fluid-model (Gidaspow, 1994 and Arastoopour 2001) which is based on the Eulerian-Granular approach that treats both the dispersed and continuous phases as interpenetrating pseudo-continua. It uses a single pressure field for all phases. Therefore, the pressure term is the same regardless of the phase. Different phases can mix and interpenetrate into one another but the volume of a phase cannot be occupied by the other phases. Volume fractions of the phases are assumed to be continuous functions of space and time and their sum is equal to one.

The interaction between the mean flow of both phases is modeled through the interaction term which includes a drag force, and virtual mass effect when acceleration of secondary phase relative to the primary and additional lift forces exist. The drag model that was used in our work was Syamlal and O'Brien (1993) drag model (eq. T-19). Their proposed drag law accounts for increased volume loading through the use of a drag coefficient that depends on volume fraction-dependent terminal velocities ($v_{r,s}$) for falling particles in settling or fluidized beds (eq. T-20). The Syamlal and O'Brien drag expression contains adjustable parameters that can be used to tune the drag to match the theoretical minimum fluidization velocity to experimentally observed values. The minimum velocity at which the drag between particles and the fluid just balances the weight of the particle bed is called the minimum fluidization velocity. The parameter p in Equation (T-21) is related to the minimum fluidization velocity through the velocity–voidage correlation and the terminal Reynolds number, Re_t . The parameter p is changed until the following criterion is met (Vejahati et al., 2009):

$$\left[U_{mf}^{experimental} - Re_{ts} \frac{\varepsilon_g \mu_g}{d_s \rho_g} \right] \xrightarrow{Minimum} 0$$

Where:

$$Re_{ts} = \left(\frac{\sqrt{23.04 + 2.52 \sqrt{4 \frac{Ar}{3}} - 4.8}}{1.26} \right)^2$$

$$Ar = \frac{(\rho_s - \rho_g) d_s^3 \rho_g g}{\mu_g}$$

$$v_{r,s} = \frac{A + 0.06 B Re_{ts}}{1 + 0.06 Re_{ts}}$$

$$Re_t = v_{r,s} Re_{ts}$$

Also the parameter q in Equation (T-21) is adopted to guarantee the continuity of the velocity–voidage correlation at $\varepsilon_g = 0.85$ as follows:

$$q = 1.28 + \frac{\log(p)}{\log(0.85)}$$

It should be noted that the original drag correlation was derived for homogeneous gas-solid flows and the adjustment reduces the drag to partially account for the heterogeneous gas-solid structure in the fluidized bed; however the adjustment should not alter the behavior of the drag correlation when voidage approaches 1. In that limit, the single sphere drag coefficient should be recovered (Vejahati et al., 2009).

In turbulence flows, interaction between gas turbulence and particle phase fluctuations becomes important, although it has a secondary effect in the dense or relatively dense flows. Gas turbulence effect is more likely to be localized to the very dilute zones and at the dense/dilute interface where it plays a significant role in entrainment of particles into the dilute region

(Sundaresan, 2011). In this study we neglected the interaction between gas turbulence and particle fluctuations.

The basic equations for the two-fluid-model, the continuity and momentum equations for each phase are derived from the general transport theorem. As there are some unknowns in the governing equations for each phase, the closure of the equation is possible by use of averaging techniques and introducing the constitutive equations based on the flow behavior; for instance the kinetic theory may be used for particle phase flow and Newtonian flow model for the gas phase. Appendix A summarizes the governing equations of the Eulerian-Granular Two-fluid-model. The assumptions of the two-fluid-model include isothermal condition for the system, ideal gas behavior of the gas phase, and uniform and constant size for the particles in the solid phase.

2.3. Population balance model

Experimental data in the literature (Yi et al., 2007) on fresh, carbonated and regenerated sorbent particles in the CO₂ capture process using solid sorbents have shown that the sorbent particles size distribution essentially remains constant during carbonation and regeneration processes. However, the mass of sorbent particle changes during both sorption (mass gain) and regeneration processes (mass loss), that leads to the changes in the density of particles. Consequently, the particles go through a structural change (e.g. change in porosity distribution). To account for the temporal and spatial evolution of the sorbent density distribution a population balance approach can be used.

The population balance equation (PBE) is a balance equation based on the number density function $f(\xi; \mathbf{x}, t)$, where ξ and $\mathbf{x}$ are internal and external coordinates, respectively. PBE

accounts for the spatial and temporal evolutions of the number density function in a single control volume. Depending on the system of interest, the number density function $f(\xi; \mathbf{x}, t)$ may have only one internal coordinate (i.e., particle density) or multiple coordinates such as particle size and density. The system of our interest is a univariate system with the particle density being the only internal coordinate.

For an inhomogeneous particulate system without diffusion, the governing equation becomes (Randolph and Larson, 1988):

$$\frac{\partial f(\xi; \mathbf{x}, t)}{\partial t} + \frac{\partial}{\partial x_i} [v_p(\mathbf{x}, t) f(\xi; \mathbf{x}, t)] + \frac{\partial}{\partial \xi_j} \left[\frac{\partial \xi_j}{\partial t} f(\xi; \mathbf{x}, t) \right] = h(\xi; \mathbf{x}, t) \quad (1)$$

The terms on the left-hand side are accumulation term, convective term with respect to the external coordinate, and convective term with respect to the internal coordinate, respectively.

$\frac{\partial \xi_j}{\partial t}$ is the flux in ξ -space (Marchisio et al., 2003), or, in other words, the growth rate of the internal variable ξ (e.g., density). Moreover, it is assumed that the particles are convected with the same phase velocity $v_p(\mathbf{x}, t)$; therefore, the particle velocity is independent of the internal property. In all the solution methods based on the method of moments (MOM), the transport equation (PBE) is written in terms of the lower-order moments of the number density function, f , in a closed form (Marchisio et al., 2003). The i^{th} moment of the number density function is defined as:

$$\mu_i = \int_0^\infty \xi^i \cdot f(\xi; \mathbf{x}, t) d\xi \quad (2)$$

Using FCMOM, the solution of the PBE is sought in the finite interval between the minimum and maximum values $[\xi_{min}, \xi_{max}]$ of the particle property instead of in the $[0, \infty]$ range. In general form, the evolution of the number density function is tracked imposing moving boundary conditions. Details of the method could be found elsewhere (Strumendo and Arastoopour, 2008; Abbasi and Arastoopour, 2013).

In the formulation of PBM for CO₂ sorption process using solid sorbents, ξ_{min} and ξ_{max} can be considered as the density of fresh sorbent and the density of the sorbent with complete conversion, respectively. To derive the final governing Population Balance Equation accounting for the temporal and spatial evolution of the sorbent density distribution, the following assumptions were made:

- 1- Uniform and constant particle size distribution.
- 2- Density of the sorbent particles is defined as a function of MgO conversion.
- 3- Density distribution function is defined in the range of $[\xi_{min}, \xi_{max}]$ and then using a coordinate transformation is transformed to $[-1, +1]$.
- 4- Constant maximum sorbent density, ξ_{max} , corresponding to the fully reacted sorbent.
- 5- Variable minimum sorbent density, ξ_{min} , corresponding to the fresh sorbent. The rate of change is related to the rate of reaction.
- 6- The change in the density due to the chemical reaction is modeled as a growth process.

Considering above mentioned assumptions, the final form of the moments' transport equations becomes:

$$\begin{aligned}
\frac{\partial \mu_i}{\partial t} + \frac{\partial}{\partial x_j} [v_{p,j} \mu_i] = & - \frac{2}{(\xi_{\max} - \xi_{\min})} \cdot \{ [G'_{+1} \overline{f'_{+1}} - (-1)^i \cdot G'_{-1} \overline{f'_{-1}}] - i \cdot \int_{-1}^1 G' \overline{f'} \cdot (\bar{\xi})^{i-1} \cdot d\bar{\xi} \} - \\
& \{ [\overline{f'_{+1}} - (-1)^i \cdot \overline{f'_{-1}}] - i \cdot \mu_{i-1} \} \cdot \frac{1}{(\xi_{\max} - \xi_{\min})} \cdot \left(\frac{d\xi_{\min}}{dt} \right) - \\
& \{ [\overline{f'_{+1}} - (-1)^{i+1} \cdot \overline{f'_{-1}}] - (i+1) \cdot \mu_i \} \cdot \frac{1}{(\xi_{\max} - \xi_{\min})} \cdot \left(-\frac{d\xi_{\min}}{dt} \right) - \\
& \{ [\overline{f'_{+1}} - (-1)^i \cdot \overline{f'_{-1}}] - i \cdot \mu_{i-1} \} \cdot \frac{v_{p,j}}{(\xi_{\max} - \xi_{\min})} \cdot \left(\frac{\partial \xi_{\min}}{\partial x_j} \right) - \\
& \{ [\overline{f'_{+1}} - (-1)^{i+1} \cdot \overline{f'_{-1}}] - (i+1) \cdot \mu_i \} \cdot \frac{v_{p,j}}{(\xi_{\max} - \xi_{\min})} \cdot \left(-\frac{\partial \xi_{\min}}{\partial x_j} \right)
\end{aligned} \tag{3}$$

Moments evolution equations must be coupled with the moving boundary conditions providing the governing equations for $\xi_{\min}(t, x)$. There is also a source term due to the reaction and $\xi_{\min}(t, x)$ is convected because of the convective particle movement in the presence of spatially inhomogeneous conditions. The definition of the moving boundary conditions for $\xi_{\min}(t, x)$ is based on the evaluation of the following:

- (a) The spatial derivatives of $\xi_{\min}(t, x)$.
- (b) The velocity $v_{p,\min}(t, x)$ of the particles whose densities are equal to $\xi_{\min}(t, x)$.

The moving boundary condition for $\xi_{\min}(t, x)$ is then given by

$$\frac{\partial \xi_{\min}}{\partial t} + \nabla \cdot v_{p,\min} \cdot \xi_{\min} = G \tag{4}$$

where G is the growth rate of the particle density due to the reaction.

2.4. Reaction kinetics

To describe the heterogeneous reaction between the sorbent and CO₂ and to define the growth rate of the particle density due to the reaction, it was assumed that the particles can be described (see Figure 4) by two distinct zones representing the product layer with higher density, ξ_2 , and the inner core (fresh sorbent) with a lower density, ξ_1 .

The average density of the particle, ξ , can be written in the form of:

$$\xi = (\xi_1 - \xi_2) \left(\frac{r_c}{r} \right)^3 + \xi_2 \quad (5)$$

by considering the classical shrinking core model for the reaction we have

$$\left(\frac{r_c}{r} \right)^3 = 1 - X \quad (6)$$

where, X is the overall conversion. Therefore, the growth rate of the particle density, G , is

$$G = \frac{\partial \xi}{\partial t} = (\xi_2 - \xi_1) \frac{\partial X}{\partial t} \quad (7)$$

$\frac{\partial X}{\partial t}$ is provided by the heterogeneous reaction kinetic model between CO₂ and sorbent based

on a two-zone variable diffusivity shrinking core model, as described by Abbasi et al. (2013):

$$\frac{dX}{dt} = - \frac{\frac{3}{r_p} \frac{k_s}{N_{MgO}^o} (C_b - C_e) (1 - X)^{\frac{2}{3}}}{1 + \frac{k_s}{D_g} r_p (1 - X)^{\frac{1}{3}} (1 - \sqrt[3]{1 - X})} \quad (8)$$

where, C_b is the CO_2 concentration in the bulk gas, C_e (Define This) is the equilibrium CO_2 concentration, D_g is the product layer diffusivity of CO_2 , r_p is the radius of the particle, k_s is the surface reaction constant rate at each zone, and N_{MgO}^o is the initial number of moles of MgO per unit volume. In the above model, the dependence of the gas diffusion coefficient D_g , through the porous product layer with respect to conversion was assumed to follow an exponential decay function (Abbasi et al., 2013).

Rate of carbonation reaction can be calculated from the rate of sorbent conversion, obtained from the two zone variable diffusivity shrinking core model which gives the dX/dt in the following correlation:

$$r_{cbn} = \frac{y_{MgO}}{MW_{sorbent}} \frac{dX}{dt} \quad (9)$$

for the water gas shift reaction

$$r_{WGS} = k_{WGS} \left(y_{CO} y_{H_2O} - y_{CO_2} y_{H_2} / K_{e,WGS} \right) \quad (10)$$

where, y_i is the mole fraction of species i in the gas mixture, $K_{e,WGS}$ is the WGS reaction equilibrium constant, k_{WGS} is the forward reaction rate constant. In the temperature range of 315° to 480°C , the following expression provides a good estimate for the WGS equilibrium (Moe, 1962):

$$K_e = \exp \left(\frac{4577.8}{T} - 4.33 \right) \quad (11)$$

where T is WGS reaction operating temperature in K .

Hassanzadeh (2007) proposed the following expression for k_{wGS} as a function of MgO conversion and reported the model parameters as shown in Table 2.

$$k_{wGS} = k_{wGS}^0 \exp(-\beta \cdot X) \quad (12)$$

The reaction kinetic model was implemented into our CFD code through a supplemental C code (User-defined-function). This code calculates the overall rate of reaction based on eq.9 and eq.10 and provides the source term (R_j) in the species conservation equations (T-6 and T-7). The model has been validated against TGA and packed bed experimental data in previous studies (Abbasi et al., 2013; Abbasi et al., 2014).

3. Simulation results and discussion

The first set of simulations focused on the carbonation reaction at the elevated pressure and temperature. The effects of solid circulation rate and gas residence time on the CO₂ capture performance were investigated. After finding the optimum solid circulation rate and gas residence time (i.e. inlet gas velocity), water gas shift reaction was also added to the system to study the performance of the sorbent in the sorption enhanced water gas shift (SEWGS) process described earlier. The rate of the carbonation reaction and the water gas shift reaction were added to the model using the variable diffusivity shrinking core model (Abbasi et al., 2013) and the equilibrium reaction rate, respectively, as described. The Coupled CFD-PBM algorithm was also used to account for the variation of particle density due to the chemical reaction between CO₂ and the MgO-based solid sorbents in the carbonator reactor.

A summary of all the simulations performed at different solid circulation rates and gas inlet velocities are presented in Table 3. These variables were varied within a range that ensures the

fast fluidization regime in the riser. To study the effect of solid circulation rate, the inlet solid mass flow rate increased by factors of 5 and 10 while the inlet gas velocity was kept constant at the baseline value. Furthermore, at the inlet solid mass flow rate of 220 g/s (Case 1), the inlet gas velocity was decreased by 25% and 35% (Case 3 and Case 4) to investigate the effect of gas residence time on CO₂ removal. In these simulations a 50/50 (mole fraction) mixture of CO₂ and N₂ was used as the inlet gas.

3.1. Effect of solid circulation rate. The effect of solids circulation rate on the removal of CO₂ from the 50/50 (mole fraction) mixture of CO₂ and N₂ is presented in Figure 5. It can be seen that as the solid mass flow rate is increased, the CO₂ exit mole fraction decreases. At baseline condition (44 g/s), a 30% CO₂ removal was achieved. At 5 times higher solid mass flow rate (Case 1), removal increased to 40% and after that even with 10 times higher solid mass flow rate (Case 2) no increase in CO₂ removal was observed. Since there is no difference in CO₂ removal by further increasing the solid inlet mass flow rate, it suggests that the process is controlled by the reaction rate and an improvement is expected by decreasing the gas residence time. Figure 6 and Figure 7 also support this conclusion.

Figure 6 shows the contours of instantaneous solid volume fraction, CO₂ mole fraction and reaction rate at $t=20s$ for solid circulation rate of 220 g/s. The solid volume fraction contours show a very dense and well mixed solid phase in the carbonator and a dilute region in the riser. Reaction rate contours (Part of Figure 6) also shows that most of the CO₂ capture takes place in the carbonator with very little reaction in the riser. Figure 7 compares the solid volume fractions at different solid circulation rates. Solid phase in the carbonator becomes denser by increasing the inlet solid mass flow rate; however, by further increasing the solid mass flow rate to 440 g/s solid hold-up in the carbonator remains constant and the additional solid bypasses via the riser.

2. Effect of gas residence time. The effect of gas residence time was studied by changing the inlet gas velocity and keeping the solid circulation rate constant at 220 g/s. Three cases with inlet gas velocities of baseline (0.15 m/s), 25% and 35% lower gas velocities (0.1125 m/s and 0.0975 m/s) were investigated. As expected and is shown in Figure 8, CO₂ removal increases by increasing the gas residence time, providing the gas and solid a longer contact time and more efficient process.

As shown in Figure 8 decreasing the inlet gas velocity by 35% leads to an additional 20% CO₂ removal, reaching to 60% CO₂ removal. Figure 9 presents contours of instantaneous CO₂ mole fraction at $t=20\text{ sec}$, for different gas residence times. Similar to previous cases, most of the carbon capture takes place in the lower part of the carbonator. The improving effect of longer gas residence time is evident by comparing the concentrations of CO₂ at the exit which decreases by reducing the inlet gas velocity and higher gas residence time in the reactor. However, even at the lowest inlet gas velocity (0.0975 m/s) the outlet CO₂ concentration is far from the equilibrium limit that is the lowest achievable concentration at this operating condition. Further decreasing the inlet gas velocity is not possible due to the change in the behavior of the fluidized bed, not being at turbulent/fast fluidization regime.

3.3. Effect of reaction on the solid phase density distribution. To account for the variation of particle phase density due to the chemical reaction between the MgO and CO₂, the coupled CFD-PBE model was utilized. In this case it is assumed that the solid sorbent entering the reactor has a uni-modal density distribution representing a partially regenerated sorbent as shown in Figure 10 in dimensionless form. The probability density function spans between a minimum density (i.e., 2480 kg/m³) corresponds to the fresh sorbent and a maximum density (i.e., 2830 kg/m³) corresponds to the fully reacted sorbent. The mean density value at the solid inlet is 2569

kg/m^3 . Density growth rate, G , in equation 3 and 4 was calculated based on the carbonation reaction rate according to equation 7 and treated as a linear growth term. The mean density value was calculated based on the ratio of the second moment of the distribution function to the first moment of the distribution function (μ_1/μ_0).

Figure 11 shows contours of the time averaged minimum and mean solid density in the reactor. The minimum density contours of the solid (Figure 11) provides valuable information about the location of the fresh sorbent front. Based on equation 4, the minimum density boundary (fresh sorbent front) moves with the reaction rate, and as the reaction rate is significantly higher in the lower part of the reactor (see Figure 6), the minimum density values rise faster in this region. However, the mean density values, calculated based on the moments of the distribution are increasing along the height of the reactor.

Although in this specific case the change in the density of the sorbent is not significant and has very small effect on the hydrodynamics of the system, it can become important in the cases where the sorbent has a significantly higher reaction rate or higher residence time in a different reactor design. In addition, the coupled CFD-PBE model is a valuable tool for design and optimization of the reactor and can be useful in calculation of the regeneration rate and the rate of sorbent make-up to the system. Another application of such a model is studying of the effect of particle size distribution on the reactor performance and in the cases that the particle attrition and particle breakage exists. It also has many applications beyond the scope of this study where the changes in the phase properties are more significant with strong effect on the hydrodynamics of the system.

3.4. Sorption enhanced water gas shift (SEWGS) in the carbonator. SEWGS is a high-pressure and high-temperature regenerative dry sorbent process that combines CO₂ capture with the WGS reaction. WGS is an equilibrium controlled reaction in which carbon monoxide reacts with water vapor to form carbon dioxide and hydrogen. If CO₂ can be continuously removed from the reactor, (i.e., by CO₂ capture), the WGS reaction will be shifted toward production of more H₂ as a clean source of energy. Therefore, a SEWGS process for continuous separation of CO₂ or H₂ would be desirable. In this section, the applicability of the MgO-based sorbent for such processes is examined.

Hassanzadeh (2007) evaluated the same MgO-based dry sorbent in a SEWGS reaction in packed-bed reactor at 350 °C and showed that the sorbent is able to remove more than 90% of the CO₂ and to increase the Hydrogen mole fraction from 37% *mol* (dry basis) in the inlet to 55% *mol* in the outlet. He also showed that a mixture of the sorbent and a commercial WGS catalyst will be able to produce a relatively pure stream of Hydrogen, by assuming that mixing the sorbent with the commercial WGS catalysts will increase the water gas shift reaction rate (reported in Table 2) by 10 times. In this study the same assumption was made and performance of the sorbent was evaluated for a SEWGS process. The inlet gas is a simulated Syngas with a composition of 20% *mol* CO₂, 20% *mol* H₂O, 30%*mol* CO, and 30%*mol* H₂. Initially, the reactor contains only steam (H₂O). The operating condition corresponds to the Case 1 in Table 3 except the temperature was at 350 °C.

Figure 12 shows the gas composition (wet basis) in the reactor outlet that corresponds to about 50% CO₂ removal and H₂ mole fraction at the outlet being around 65% *mol*. Since the carbonation reaction is limited by equilibrium, and in this case due to the lower concentration of CO₂, the reaction has less driving force to proceed, the total amount of CO₂ adsorbed by the

sorbent is less than the amount adsorbed in the cases with 50% inlet CO₂ mole fraction. It should be noted that this is not the adverse effect of the water gas shift on CO₂ removal, but the difficulty of removing CO₂ from a more dilute mixture.

Figure 13 shows the instantaneous Syngas composition (mol% wet basis) in the reactor at $t=20$ sec that matches the values reported in Figure 12. As the reaction proceeds along the reactor, and mainly in the lower part, concentration of CO₂ decreases due to the absorption by the sorbent and it shifts the water gas shift reaction toward producing more Hydrogen. CO concentration drops from 30% mol to about 20% mol and CO₂ and H₂O reach to about 10% mol concentration. This composition corresponds to 50% CO₂ removal.

Previous studies on the same sorbent in a lab scale packed bed reactor showed that this sorbent has the potential and required capacity to meet the 90% CO₂ removal goal set by the United States Department of Energy (DOE). However, our simulations show that the rate of the reaction is not sufficiently high to remove 90% of CO₂ in the current circulating fluidized bed reactor. Since most of the CO₂ capture takes place in the lower part of the reactor and also the process is limited by the rate of the reaction, increasing the contact time between the solid and the gas would potentially improve the efficiency of the process. This could be achieved by an optimum design of the lower part of the bed that provides a better gas-solid contact time utilizing the developed CFD model.

4. Conclusion

This work established a base-line design for the circulating fluidized bed reactor for both CO₂ capture process and SEWGS process using an MgO-based solid sorbent. The proposed

circulating fluidized bed reactor ensures a continuous CO₂ capture and provides a very good mixing and heat transfer between the solid and the gas.

Some preliminary parametric studies was performed that showed the capacity of the current process for a maximum of 60% projected CO₂ removal which is still far from the equilibrium limit. Although the results prove the concept of the process, there are still vast opportunities for further investigations and optimization of the reactor design. The CFD model can be used to find the optimum reactor design, specifically the geometry of the carbonator, to maximize the gas-solid contact and hence CO₂ removal. A better reactor design could increase the overall performance of the reactor toward capturing CO₂ up to the equilibrium limit of this reaction.

It was also demonstrated that the coupled CFD-PBE model based on the FCMOM approach is a valuable tool for design and optimization of the reactor and can be useful in calculation of the regeneration rate and sorbent make-up in the regenerative solid-based process for carbon capture. The model also provides a powerful tool to study the importance of other polydispersities in the system such as particle size distribution and pore size distribution. Overall, this research study has opened interesting research areas in the multi-phase flow area, which was not possible to investigate by other models assuming uniform properties of the reacting phases in a system.

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6. Nomenclature

Ar	Archimedes number, -
C_b	CO ₂ concentration in gas, $mole/m^3$
C_D	drag coefficient
C_e	Equilibrium CO ₂ concentration, $mole/m^3$
C_i	concentration at reaction interface, $mole/m^3$
D_g	Product layer diffusivity, m^2/s
d_p	Particle diameter
E_{mi}	coefficients in the moment equations for the FSE
$\overline{f}$	dimensionless number density function
G	growth rate
h	net rate of introduction of new particles in PBE
i	moment order, -
K	constant in the growth rate law
k_s	Reaction rate constant, m/s
K_{WGS}	WGS equilibrium constant
MW	Molecular weight, $kg/kmole$
N_{MgO}^o	Initial number of moles of MgO, $mole/m^3$
p_s	Solid phase pressure
Re	Reynolds number (-)
Re_p	particle Reynolds number (-)
t	time (s)
v_k	k^{th} phase velocity (m/s)
v_p	particulate phase velocity (m/s)
x	external coordinate vector in PBE

Greek symbols

$\bar{\xi}$	Dimensionless internal coordinate in PBE
β_{kj}	drag coefficient between k phase and j phase
ε_k	k^{th} phase volume fraction,-
μ_i	i^{th} moment of the number density function
μ_k	k^{th} phase viscosity, $kg/m\ s$
ζ	internal coordinate in PBE
ρ_k	k^{th} phase density, kg/m^3
τ_k	k^{th} phase stress tensor, Pa
θ	Granular temperature,

Appendix A

Table A.1. Summary of two-fluid-model governing equations

Conservation of mass:

- Gas phase:

$$\frac{\partial}{\partial t}(\varepsilon_g \rho_g) + \nabla \cdot (\varepsilon_g \rho_g \vec{v}_g) = \dot{m}_g \quad (\text{T-1})$$

- Solid phase:

$$\frac{\partial}{\partial t}(\varepsilon_s \rho_s) + \nabla \cdot (\varepsilon_s \rho_s \vec{v}_s) = \dot{m}_s \quad (\text{T-2})$$

$$\varepsilon_g + \varepsilon_s = 1 \quad (\text{T-3})$$

Conservation of momentum:

- Gas phase:

$$\frac{\partial}{\partial t}(\varepsilon_g \rho_g \vec{v}_g) + \nabla \cdot (\varepsilon_g \rho_g \vec{v}_g \vec{v}_g) = -\varepsilon_g \nabla P + \nabla \cdot \tau_g + \varepsilon_g \rho_g g - \beta_{gs}(\vec{v}_g - \vec{v}_s) \quad (\text{T-4})$$

- Solid phase:

$$\frac{\partial}{\partial t}(\varepsilon_s \rho_s \vec{v}_s) + \nabla \cdot (\varepsilon_s \rho_s \vec{v}_s \vec{v}_s) = -\varepsilon_s \nabla P + \nabla \cdot \tau_s - \nabla p_s + \varepsilon_s \rho_s g + \beta_{gs}(\vec{v}_g - \vec{v}_s) \quad (\text{T-5})$$

Conservation of species;

- Gas phase:

$$\frac{\partial}{\partial t}(\varepsilon_g \rho_g y_j) + \nabla \cdot (\varepsilon_g \rho_g \vec{v}_g y_j) = R_j \quad (\text{T-6})$$

- Solid phase:

$$\frac{\partial}{\partial t}(\varepsilon_s \rho_s y_i) + \nabla \cdot (\varepsilon_s \rho_s \vec{v}_s y_i) = R_j \quad (\text{T-7})$$

Conservation of solid phase fluctuating energy;

$$\frac{3}{2} \left[\frac{\partial}{\partial t}(\varepsilon_s \rho_s \theta) + \nabla \cdot (\varepsilon_s \rho_s \theta \vec{v}_s) \right] = (-p_s \bar{I} + \tau_s) : \nabla \vec{v}_s + \nabla \cdot (\kappa_s \nabla \theta) - \gamma_s \quad (\text{T-8})$$

Table A.2. Summary of two-fluid-model constitutive equations

Gas phase stress:

$$\tau_g = \varepsilon_g \mu_g \left[\nabla \vec{v}_g + (\nabla \vec{v}_g)^T \right] - \frac{2}{3} \varepsilon_g \mu_g (\nabla \cdot \vec{v}_g) I \quad (\text{T-9})$$

Solid phase stress:

$$\tau_s = \varepsilon_s \mu_s \left[\nabla \vec{v}_s + (\nabla \vec{v}_s)^T \right] + \varepsilon_s \left(\xi_s - \frac{2}{3} \mu_s \right) \nabla \cdot \vec{v}_s I \quad (\text{T-10})$$

Collisional dissipation of solid fluctuating energy;

$$\gamma_s = 3(1 - e^2) \varepsilon_s^2 \rho_s g_0 \theta \left(\frac{4}{d_p} \sqrt{\frac{\theta}{\pi}} \right) \quad (\text{T-11})$$

Radial distribution function;

$$g_0 = \left[1 - \left(\frac{\varepsilon_s}{\varepsilon_{s,\max}} \right)^{1/3} \right]^{-1} \quad (\text{T-12})$$

Solid phase pressure:

$$p_s = \varepsilon_s \rho_s \theta [1 + 2g_0 \varepsilon_s (1 + e)] + p_{s,fr} \quad (\text{T-13})$$

$$p_{s,fr} = Fr. \frac{(\varepsilon_s - \varepsilon_{\min})^n}{(\varepsilon_{\max} - \varepsilon_s)^p}, \quad (\text{T-14})$$

$$\varepsilon_{\min} = 0.53 \leq \varepsilon_s \leq \varepsilon_{\max} = 0.63, \quad Fr = 0.1 \varepsilon_s, n = 2, p = 3$$

Solid phase shear viscosity;

$$\mu_s = \frac{4}{5} \varepsilon_s \rho_s d_p g_0 (1 + e) \sqrt{\frac{\theta}{\pi}} + \frac{10 \rho_s d_p \sqrt{\pi \theta}}{96(1 + e) g_0 \varepsilon_s} \left[1 + \frac{4}{5} g_0 \varepsilon_s (1 + e) \right]^2 + \mu_{fr} \quad (\text{T-15})$$

$$\mu_{fr} = \frac{p_{fr} \sin \phi}{\sqrt{4 \Pi_{2D}}} \quad (\text{Schaeffer, 1978}) \quad (\text{T-16})$$

Solid phase bulk viscosity:

$$\xi_s = \frac{4}{3} \varepsilon_s \rho_s d_p g_0 (1 + e) \sqrt{\frac{\theta}{\pi}} \quad (\text{T-17})$$

Conductivity of the fluctuating energy:

$$\kappa_s = \frac{150 \rho_s d_p \sqrt{\theta \pi}}{384(1 + e) g_0} \left[1 + \frac{6}{5} \varepsilon_s g_0 (1 + e) \right]^2 + 2 \rho_s \varepsilon_s^2 d_p (1 + e) g_0 \sqrt{\frac{\theta}{\pi}} \quad (\text{T-18})$$

(Gidaspow et al., 1991)

Drag coefficient :

$$\beta_{gs} = \frac{3\varepsilon_g \varepsilon_s \rho_g}{4v_{r,s}^2 d_s} C_D \left(\frac{Re_s}{v_{r,s}} \right) (\vec{v}_g - \vec{v}_s) \quad (\text{T-19})$$

where $v_{r,s}$ is the particle terminal velocity

$$v_{r,s} = 0.5(A - 0.06Re_s) + \sqrt{(0.06Re_s)^2 + 0.12Re_s(2B - A) + A^2} \quad (\text{T-20})$$

with A and B has a form of

$$A = \varepsilon_g^{4.14} \text{ and } B = \begin{cases} P\varepsilon_g^{1.28}, & \varepsilon_g \leq 0.85 \\ \varepsilon_g^q, & \varepsilon_g > 0.85 \end{cases} \quad (\text{T-21})$$

and p and q are the adjustable parameters.

FIGURES

Figure 1. Regenerative carbon capture process concept based on metal-oxide solid sorbent (Hassanzadeh, 2007).

Figure 2. CFD-PBE Coupling Algorithm (Abbasi and Arastoopour, 2013)

Figure 3. Computational domain and geometry of the carbonator reactor

Figure 4. Schematic of the reacting particle

Figure 5. Effect of solid circulation rate on CO₂ removal

Figure 6. Contours of instantaneous solid volume fraction, CO₂ mole fraction and reaction rate at $t=20$ sec, for solid circulation rate of 220 g/s

Figure 7. Contours of instantaneous solid volume fraction at $t=20$ sec, for the different solid circulation rates.

Figure 8. Effect of inlet gas velocity on CO₂ removal

Figure 9. Contours of instantaneous CO₂ mole fraction at $t=20$ sec, for different inlet gas velocities.

Figure 10. Particle density distribution function at the inlet

Figure 11. Contours of the time averaged minimum and mean density of the solid phase after 20 Sec.

Figure 12. SEWGS outlet gas composition at 350°C.

Figure 13. Contours of the instantaneous Syngas composition (% *mol*) at $t=20$ sec going through the SEWGS reaction.

TABLES

Table 1. Baseline high pressure and high temperature operating conditions

Parameter		value	
Pressure		20 atm	
Temperature		425 °C	
Inlet gas velocity		0.15 m/s	
Inlet solid rate		0.044 kg/s	
Inlet gas composition		CO ₂ capture	SEWGS
	CO ₂ mole fraction	50 %	20%
	H ₂ mole fraction	0%	30%
	CO mole fraction	0%	30%
	H ₂ O mole fraction	0%	20%
	N ₂ mole fraction	50%	Balance

Table 2. WGS Reaction Model Parameters at Different Operating Temperatures

Model Parameter	350°C	400°C	425°C
k_{WGS0} , mol/g.min	0.0003	0.001	0.002
β	3.5	3.5	3.5

Table 3. Summary of simulation cases

Case	inlet gas velocity (m/s)	Solid circulation rate (g/s)
Baseline	0.15	44
Case 1	0.15	220
Case 2	0.15	440
Case 3	0.1125	220
Case 4	0.0975	220