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Transient reacting flow simulation of spouted fluidized bed for coal-direct chemical looping combustion with different Fe-based oxygen carriers

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HIGHLIGHTS

- High fidelity CFD/DEM simulations of a spouted fluidized bed CLC fuel reactor.
- Chemical reactions successfully integrated into the CFD/DEM solver.
- Improved fluidization using Fe_2O_3 dispersed on $MgAl_2O_4$ as oxygen carrier.
- Reacting flow simulations for methane reported for the first time in the literature.

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ABSTRACT

Coal-direct chemical-looping combustion (CD-CLC) is a next generation combustion technology that shows great promise as a solution for the need of high-efficiency low-cost carbon capture from fossil fueled power plants. To realize this technology on an industrial scale, the development of high-fidelity simulations is a necessary step to develop a thorough understanding of the CLC process. In this paper, simulations of multiphase flow in a CD-CLC process with chemical reactions are performed using the ANSYS Fluent CFD software. Discrete Element Method (DEM) provides the means for tracking the motion of individual metal oxide particles in the CD-CLC system as they react with the fuel and combine with CFD for capturing the solid–gas multiphase hydrodynamics. The overall results of the coupled CFD/DEM simulations using Fe-based oxygen carriers reacting with gaseous CH_4 demonstrate that chemical reactions have been successfully incorporated into the CFD/DEM approach. The simulations show a strong dependence of the fluidization performance of the fuel reactor on the density of bed material and provide important insight into selection of a right oxygen carrier for the enhanced performance. This work provides the foundation for future simulations of CD-CLC systems using solid coal as fuel, which will be crucial for successful deployment of CD-CLC technology from the laboratory scale to pilot and industrial scale projects.

1. Introduction

The relationship between the global surface temperature of the earth and the concentration of CO_2 in the atmosphere was discovered by Arrhenius as early as 1896 [1]. Since then, CO_2 levels in the atmosphere have increased by almost 30% compared to the pre-industrial times. The primary contributor to this large increase in CO_2 levels is carbon emissions from power plants burning fossil fuels, which currently remain the world's main energy source. Addressing carbon emissions from power plants has been an active area of research. In recent years, several technologies to capture CO_2 emissions from fossil fueled power plants have been demonstrated. These technologies can be broadly categorized as pre-combustion capture such as the integrated gas combined cycle (IGCC), post-combustion capture such as sorbent-based absorption, and oxy-fuel combustion. However, each of these technologies requires a separate process to isolate CO_2 from the other gases, thereby incurring a significant energy penalty, which reduces the plant efficiency and can lead to an increase in the cost of electricity.

One technology that has shown great promise for high-efficiency low-cost carbon capture is chemical-looping combustion (CLC). The CLC process utilizes dual fluidized bed reactors – an air

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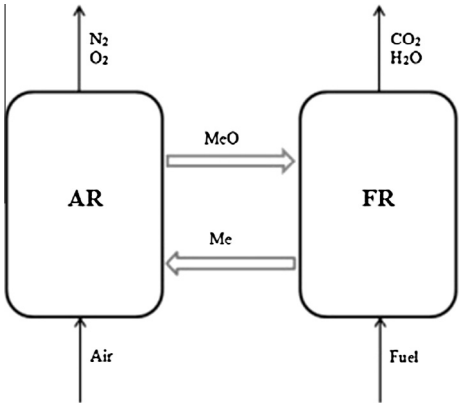

Fig. 1. Schematic diagram of the CLC process, AR = Air Reactor, FR = Fuel Reactor.

reactor and a fuel reactor, and a metal oxide oxygen carrier that circulates between the two as shown in the schematic of Fig. 1. The main advantage of the CLC process is that the combustion of fuel in the fuel reactor takes place in the absence of air using oxygen provided by the metal oxide oxygen carrier (OC), which gets reduced in the process; the flue stream from the fuel reactor is not contaminated by other gases such as N_2. This provides a high-purity CO_2 stream available for capture at the fuel reactor outlet without the need for expensive separation process. The reduced OC particles are pneumatically transported to the air reactor where they are re-oxidized by oxygen from the air to complete the loop. The only energy cost of separation associated with CLC is the energy cost of solid recirculation; research has shown that this is only about 0.3% of the total energy released by the CLC process [2]. This is considerably lower than the benchmark for pre-combustion technologies for carbon capture such as oxy-fuel combustion where the oxygen separation process can consume about 15% of the total energy. Thus, CLC holds the answer as the next-generation combustion technology due to its potential to allow CO_2 capture with little to no effect on the efficiency of the power plant. Several studies about the energy and exergy of CLC system suggest that power plant efficiencies greater than 50% can be achieved along with nearly complete CO_2 capture [3–5].

A great deal of early research in the area of chemical-looping combustion has focused primarily on the use of gaseous fuels such as natural gas and syngas. However, since coal is more abundant than natural gas and is projected to remain one of the dominant fossil fuels in the near future, the use of coal for CLC has garnered significant interest in recent years. One way to utilize coal in a CLC process is to first gasify the coal into syngas in a standalone gasifier and then inject the freshly-converted syngas into the fuel reactor. To ensure the absence of nitrogen and other gases in the syngas, the gasification must be carried out with oxygen instead of air, which requires an additional air separation unit. As such, this approach introduces the inefficiencies associated with oxy-fuel combustion and similar technologies. From the perspective of the CLC process, this scenario is identical to the one that uses gaseous fuel. The alternate approach, known as coal-direct chemical-looping combustion (CD-CLC), is to inject pulverized coal directly into the fuel reactor, which eliminates the need of a separate gasifier and reduces the complexity of the power plant. The solid–solid reaction rate of coal with the metal oxide in CD-CLC is negligible [6]. It has been proposed to first gasify the coal in the fuel reactor with CO_2 or H_2O as the fluidized agent and then react the oxygen carrier with the products of gasification [7]; this process is known as coal-direct chemical-looping combustion with in situ gasification.

The work of Leion et al. [8] has identified that the rate of fuel conversion in the CD-CLC process is limited by the char gasification step. The agglomeration between oxygen carrier and coal ash is another concern as it has been reported to reduce the reactivity of the metal oxide particles [9]. These concerns are addressed by utilizing a spouted fluidized bed fuel reactor with relatively large diameter particles, unlike in CLC using gaseous fuels, which can use a bubbling fluidized bed for the fuel reactor. According to Geldart's powder classification [10], the larger diameter particles correspond to Group D or spoutable particles. In a spouted fluidized bed, a high velocity jet of pulverized coal and the fluidizing agent is injected at the center of the fluidized bed to induce strong circulation rates for the solid particles and enhance the solid–gas mixing. The increased friction from the mixing of solids can also serve to slough off the ash build-up on the metal oxide particles and restore reactivity [11].

Although laboratory-scale experiments of CLC systems are common in the literature, numerical studies using CFD have been limited. Initial CFD studies in the field demonstrated the capability of computational methods to model a multiphase gas/solid system and were not based on any particular experiment [12,13]. Later, the work of Mahalatkar et al. [14,15] based on a single reactor setup showed that CFD simulation is able to match the reaction mechanics inside a CLC fuel reactor with reasonable accuracy. However, the single reactor setup "cannot be operated with solid fuels and the design and operation of the hot gas switching system is problematic" [16]. In order to design a CLC system for operation using solid coal given its likelihood to remain a dominant fossil fuel in the near future, the use of the dual fluidized bed setup with a spouted bed fuel reactor is suggested. The single reactor simulations that exist in literature do not provide any information about the circulation of oxygen carrier inside a dual fluidized bed setup.

In order to develop a credible simulation for an interconnected dual fluidized bed configuration typical of a CD-CLC setup with particulate metal oxides and fuel present, it is critical to accurately capture the solid circulation and separation as a result of the solid–gas two-way coupling and the solid–solid interaction. This is achieved by employing the Lagrangian particle-based model or discrete element method (DEM) where the trajectory of each individual particle is resolved based on a force balance calculation for the particle. The particle tracking is coupled with the CFD solution for the fluid phase by considering the interaction between the particle and the fluid separately for each particle. The computational cost of DEM depends on the number of particle collisions, which increases dramatically as the number of particles increases. Owing to the high computational cost, particle-based models for CLC simulation are scarce in the literature to date. A few cold-flow simulations conducted using the coupled CFD/DEM model have proven capable in accurately matching the particle dynamics of various laboratory scale fluidized bed experiments using relatively large particles [17–19]. However, no work has been done in the literature on integrating the chemical reactions into a CFD/DEM model. All simulations on the chemical reactions aspect of CD-CLC have focused either on Eulerian simulations [14,15] or more recently, on process simulations from an energy balance perspective using software such as Aspen Plus [20–22]. It is crucial to combine the solid particle dynamics and chemical reactions into one credible model for the whole CD-CLC fuel reactor that can be used to investigate various aspects of CD-CLC including reactor design, inlet jet velocity, and the physical properties of the oxygen carrier in order to achieve a setup optimized for maximum power generation.

In this paper, the transient cold-flow simulations of Zhang et al. [19] are extended to incorporate a chemical reaction between the metal oxide and the gaseous fuel. The coupled CFD/DEM model is employed for unsteady tracking of individual oxygen carrier parti-