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Original Research Paper

Hydrodynamic characteristics in a pilot-scale cold flow model for chemical looping combustion

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ABSTRACT

Chemical Looping Combustion (CLC) in two interconnected fluidized beds, i.e., the air reactor and the fuel reactor has been recognized to be promising. As the CLC setup design is critical and sensitive to oxygen carrier (OC) materials, it is very much essential to investigate hydrodynamics in a specially fabricated cold model set up for the successful development and operational control of corresponding large-scale hot model. In this study, a pilot-scale cold flow model CLC system has been designed and tested. The riser and fuel reactor were operated at circulated fluidized bed and bubbling fluidized bed conditions, respectively and the control of solid circulation between two reactors was done by two loop seals operated in bubbling fluidized bed conditions. The effect of fluidization velocity in the riser on the voidage profiles, solid circulation rate, and pressure profiles were investigated using Indian ilmenite ($150-212 \mu m$) as OC. The stable operation of the system was established under various operational conditions. The results will be useful for the development of ilmenite based hot model CLC system. Moreover, the achievable variations of solid circulation rate in the present study in cold model setup will determine obtainable limit of extent of oxygen transport and thermal energy.

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1. Introduction

Carbon dioxide (CO₂) is recognized as one of the major green-50 house gas (GHG) responsible for global warming and climate 51 changes. According to the fifth assessment report of Intergovern-52 mental Panel on Climate Change (IPCC), anthropogenic emissions 53 54 of CO₂ from fossil fuel combustion and other industrial processes 55 contributed ~78% of the total GHG emission increase from 1970 56 to 2010. Fossil fuel-related CO₂ emissions reached 32 Gt/yr, in 2010, and grew further by about 3% between 2010 and 2011 [1]. 57 58 It is projected that if the current trend continues, CO₂ emissions from the energy sector will get almost doubled or even tripled by 59 2050 compared to the level in 2010 [1]. This global concern has 60 motivated an extensive research work towards developing more 61 62 economical and efficient process for CO₂ capture and sequestration 63 (CCS). Three possible approaches are envisaged for the CO₂ capture from fossil fuel based power plants: pre-combustion, post-64

combustion, and oxy-fuel combustion [2]. Recently, Chemical looping combustion (CLC), a promising oxy-fuel combustion technique has gained wide attention because it can integrate fossil fuel combustion, inherent CO₂ capture with high thermo-economic efficiency and low-cost. Unlike most other oxy-fuel combustion techniques, CLC negates the requirement of pure oxygen by introducing a suitable metal oxide as an oxygen carrier (OC) that transfers oxygen to the fuel reactor (FR). CLC technology is advantageous over the other combustion technologies as it avoids the direct contact between the air and fuel. An appropriate solid metal oxide as an OC is circulated between the two reactors and transfers the lattice oxygen of the carriers from air reactor (AR) to fuel reactor (FR) as shown in reactions (1) and (2).

$$(2n+m)\ Me_xO_y+C_nH_{2m}\rightarrow (2n+m)\ Me_xO_{y-1}+mH_2O+nCO_2$$

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(1)

$$Me_xO_{y-1} + 1/2 \ O_2 \rightarrow Me_xO_y \tag{2}$$

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where Me_xO_v is metal oxide OC and Me_xO_{v-1} is the reduced form of metal oxide OC. In the FR, the gaseous fuel such as natural gas, syn-

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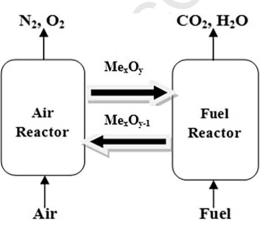
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Adecay factor for solid U_t terminal velocity of particles, m/sAcross section area of stand pipe, m² U_{pa} primary air velocity, m/sArarchimedes numberUsuperficial air velocity in riser, m/sCconstant U_s solid velocity in riser, m/sC_Ddrag coefficient Δz differential height, md_pparticle diameter, m Δz differential height, mDdiameter of riser, m $Greek letters$ fssolid friction factor δ_b bubble volume fraction in dense bedggravitational constant, m/s² ρ_p solid particle density (kg/m³)Gssolid circulation rate, kg/m².s ρ_g density of gas (kg/m³)hdzheight of bed in dense zone of riser above distributor, m ρ_b bulk density of material, kg/m³hszheight of bed in transport zone of riser above distributor, m ρ_b solid age at minimum fluidizationkdecay factor in splash zone in riser ε_{∞} voidage at minimum fluidizationk_{\infty}particle elutriation rate constant ε_{dz} voidage in infinite heightk_{\infty}particle elutriation rate constant ε_{dz} voidage in splash zoneAttime, s ε_{dz} voidage in transport zoneUmfminimum fluidization velocity of particles, m/s μ_g viscosity of air (kg/m-s)

gas from coal gasification, or solid fuel such as coal is oxidized by the OC to CO_2 and H_2O . The exit gas stream from the FR contains only CO_2 and H_2O , and almost pure CO_2 is obtained after condensation of H_2O . The metal or reduced metal oxide is then transferred to the AR where it gets regenerated, and thus the gas stream leaving the AR contains only nitrogen and unreacted oxygen [3]. The principle concept of the CLC is shown in Fig. 1.

93 The basic idea of CLC was first introduced by Lewis and Gilliland [4] in 1954 for the production of CO₂. Ishida et al. [5] were the first 94 95 to introduce the name of CLC in their thermodynamic study. The 96 basic design of CLC reactors based on circulated fluidized bed 97 (CFB) concept was introduced by Lyngfelt et al. [6]. However, the large-scale operation and successful commercialization of CLC is 98 still contingent upon the development of suitable OCs. The design, 99 capital cost, and operation of a CLC unit mainly depend on the 100 101 properties of OCs [7]. A successful OC material should have high reactivity, sufficient oxygen transport capacity, sufficient mechan-102 103 ical stability under repeated oxidation/reduction cycles, low tendency for agglomeration in fluidized bed reactors, environment 104 105 benignity, and low production cost. In the last decade, several dif-106 ferent metal-based OCs such as the oxides of copper, nickel, man-107 ganese, cobalt, iron-single or mixed metal oxide have been 108 investigated under different operating conditions in CLC system





[3,8]. These metal-based oxygen carriers have also been combined 109 with inert binders such as Al₂O₃, TiO₂, ZrO₂, etc. to improve the 110 reactivity, durability, and fluidization behavior of carriers. Another 111 recent development is the use of natural OCs based on mineral ores 112 and natural industrial waste or by-products from industry. The 113 benefits of natural OCs over the synthetic OCs are that they are 114 low cost, readily available, non-toxic and environmentally benign. 115 Recently, ilmenite (an iron-titanium mineral ore) has emerged as a 116 potential OC for solid fuel based CLC [3,8-10]. 117

In order to demonstrate the principle of new combustion tech-118 nology and commercial scale CLC system, it is also necessary to 119 know the design and operation of the reactor system. Several com-120 binations of two-reactor configurations (bench/pilot scale) have 121 been suggested, designed and tested in past in different opera-122 tional conditions using cold and hot prototype [3,11–17]. The com-123 bination of some innovative designs consist of four compartments 124 interconnected fluidized bed (IFB) reactor [15], IFB reactor with 125 alternative valve, periodically operated packed bed reactor, IFB 126 combining fluidized bed and moving bed, IFB combining two bub-127 bling bed, IFB combining riser and bubbling bed, IFB combining 128 riser and turbulent bed etc. Dual circulating fluidized bed (DCFB) 129 system where both the reactors are operated in fast fluidization 130 regime is also proposed and tested for CLC system to achieve better 131 gas-solid contact [14,17]. However, two IFB reactors consisting of a 132 fast fluidized bed air reactor and bubbling fluidized bed fuel reac-133 tor, respectively, are believed to be the most promising configura-134 tion for successful and stable long-term operation of CLC [6,11]. 135 Various types of non-mechanical devices such as L-valves, loop 136 seals, etc. have been widely used to facilitate the flow of solids 137 between the reactor units in the IFB configurations [12-19]. Gener-138 ally, the L-valve is used to achieve maximum operational flexibility 139 through solid circulation control; while loop seal is suggested to 140 minimize intermixing of exhaust gases in the solid circulation loop. 141 The effect of various geometrical and particle parameters on the 142 operation of the L-valve have been investigated by Knowlton and 143 Hirsan [20]. Geldart and Jones [21], and Yang and Knowlton [22] 144 have estimated the aeration rate, solids circulation rate, and pres-145 sure drop correlations in an L-valve. However, the L-valve may 146 experience flow problems at high temperatures of CLC if particles 147 flowing through the L-valve are Geldart group B particles that lie 148 near the AB boundary. It will not operate automatically over an 149 infinite range of solid flow rates [23]. Therefore, the L-valve is used 150

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