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Estimating the solids circulation rate in a 100-kW chemical looping combustor

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HIGHLIGHTS

- Experiments with a novel oxygen carrier were conducted by adding batches of fuel.
- The O_2 dip in the AR that followed after each fuel batch could be used to model circulation.
- By making several batch experiments at varying circulation, a general model was obtained.
- The circulation correlated strongly to the internal mass flow in the AR riser.
- The internal mass flow in the AR riser could be used to calculate the solids circulation.

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ABSTRACT

Chemical looping combustion (CLC) is a technology of CO_2 capture that can drastically reduce its cost. The solids circulation inside a 100-kW chemical looping combustor was investigated using a novel oxygen carrier called Sinaus by adding fuel batches to the fuel reactor. The decline and subsequent rise of oxygen concentration in the air reactor after each addition was used to determine the residence time of solids in the fuel reactor and adjacent vessels. The obtained residence time, in combination with the solids inventory, determined the solids circulation for a particular batch experiment. After performing a number of such experiments, the above circulation was correlated with other experimental data, revealing a good correlation between the solids flow at the top of the air reactor riser and the actual circulation obtained using batch tests. The relationship between global circulation, $\dot{m}$, and the mass flow in the air reactor riser, $\dot{m}_{riser}$, was found to be linear within the investigated interval, being described as $\dot{m} = 6.6 + 0.057\dot{m}_{riser}$. Although this correlation was valid only for the investigated reactor system, the approach used to obtain the solids circulation could be used to derive a similar correlation for any dual fluidized bed system.

1. Introduction

Carbon capture and storage is an economically attractive method of avoiding CO_2 emission into the atmosphere, with negative emissions being possible if biomass is used as fuel. Chemical looping combustion (CLC) is a technology that can drastically reduce the cost of CO_2 capture, featuring systems based on interconnected fluidized beds comprising metal oxide particles as oxygen carriers. These particles transport oxygen from combustion air to fuel, making CO_2 capture an inherent feature of the CLC process. Control of the chemical looping process requires knowing the circulation of solids between two principal interconnected beds, i.e., the air (AR) and fuel (FR) reactors.

Although Ishida et al. were the first to recognize the potential of CLC as a CO_2 capture technology potentially exhibiting no energy penalty (Ishida and Jin, 1999), the concept of chemical looping for the purpose of CO_2 production was patented already in 1954 by Lewis and Gilliland (Lewis et al., 1954). The first CLC pilot based on interconnected fluidized beds was constructed and operated in 2003. Today, chemical looping operations described in literature amount to at least 9000 h in 34 pilots (Lyngfelt and Linderholm, 2016). Most of the operational experience related to chemical looping combustors was obtained using gaseous fuels and manufactured oxygen carriers, with oxygen carrier development reviewed by Lyngfelt, 2015) and Wang et al. (2015). In the case of chemical looping operation using solid fuels, low-cost materials such as ores or waste materials are often used. The reason for the increased attention enjoyed by low-cost materials is that (i) solid fuels normally contain significant quantities of ash, which is expected to

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Nomenclature

Abbreviations and symbols	Explanation [Unit(s)]
AR	air reactor
CCS	carbon capture and storage
CFB	circulating fluidized bed
CLC	chemical-looping combustion
CR	circulation riser
CS	carbon stripper
CSTR	continuous stirred tank reactor
FR	fuel reactor
FR+	fuel reactor and adjacent reactor vessels, including CR, CS LS2, LS3, LS4
LS	loop seal
PFR	plug flow reactor
RTD	residence time distribution
A, A_c	cross sectional area
d50	the mass-median diameter; one way of expressing the average particle size by mass [μm]
G_s	solids flux [kg/(s,m^2)]
m_{FR+}	solids inventory in FR+ [kg]
$\dot{m}$	circulation, or global solids flow [kg/min]
$\dot{m}_{riser}$	solids circulation in air reactor riser [kg/min]
N	number of CSTRs [–]
$(O_2)_{AR}(t)$	concentration of O_2 in AR exit gas at time t [%]
p	pressure [Pa]
r^2	coefficient of determination [-]
T_{FR}, T_{AR}	temperature in FR/AR [°C]
u_0	superficial velocity [m/s]
u_t	terminal velocity [m/s]
ρ_{bulk}	bulk density of oxygen carrier material [kg/m^3]
ρ_{exit}	Solids loading at the exit of the air reactor riser [kg/m^3]
τ	residence time [s]
τ_i	residence time in bed i [s]

lower the oxygen carrier lifetime, and (ii) hydrocarbons contained in typical gaseous fuels such as natural gas require more reactive oxygen carrier materials than the gas released from solid fuels, which contains larger amounts of reactive H_2 and CO. Several research groups have reported the design and operation of 0.5–50-kW chemical looping combustors (Bayham et al., 2013; Sozinho et al., 2012; Thon et al., 2014; Markström et al., 2013; Adánez et al., 2014; Xiao et al., 2012; Shen et al., 2009; Tong et al., 2014; Mendiara et al., 2012), and a 1-MW pilot has been built and autothermally operated using ilmenite (a natural mineral widely used as an oxygen carrier in CLC applications) as an oxygen carrier (Ohlemüller et al., 2015; Ströhle et al., 2014). More than 3000 h of operation using solid fuels has been reported in total, with 1570 h corresponding to low-cost oxygen carriers Low-cost materials include iron ores and iron-based waste materials, ilmenite, and manganese ores. Using ilmenite as oxygen carrier and bituminous coal as fuel, the expected lifetime of oxygen carrier particles was investigated in a 100 kW reactor system – described below – and found to be 700–800 h (Linderholm et al., 2014). Compared to ilmenite, manganese ore has shown higher reactivity with syngas. This was demonstrated in a study by Sundqvist et al. (2015) who investigated 11 manganese ores in a laboratory fluidized bed, and estimated the rate constant for the 8 most reactive ores to be 3–6 times higher as compared to ilmenite. Moreover, the presence of manganese ore has also been shown to give a higher rate of steam gasification of char in both lab tests (Arjmand et al., 2012) and a continuous unit (Linderholm et al., 2012).

In order to find out how particles will behave over time in full-scale applications, evaluation in larger, continuous units is a vital step. The expected lifetime of the oxygen-carrying particles may be a very important parameter when it comes to process upscaling. Lyngfelt and Leckner (2015) recently proposed a design of a 1000 MW CLC system, which is very similar to a state-of-the-art CFB boiler. The authors find that the additional cost of the CLC–CFB system, relative to conventional state-of-the art CFB technology, is around 20 €/tonne CO_2. In other words, the cost of CO_2 capture is significantly lower as compared to other capture technologies.

Satisfactory circulation of solids is a fundamental prerequisite for the design and control of any chemical looping process, with insufficient circulation causing (a) a high temperature difference between the AR and FR, i.e., low fuel reactor temperature, and (b) an insufficient amount of oxygen supplied to fuel. Although the circulation of solids in fluidized beds is difficult to measure due to high temperatures and mechanical wear caused by the moving bed material, a large number of methods have been proposed and investigated. Bhusarapu et al. (2004) listed six groups of such measurements, namely optical, radioactive, electrical, tracer, acoustical, heat/mass transfer, and mechanical. Most of these methods have been developed for cold conditions and are excellent for predicting the flow of solids in cold-flow units only, being unsuited for high-temperature systems. Furthermore, a large number of these methods are invasive, making them hard to implement in the case of continuous operation. Thus, an ideal method should allow direct on-line measurements of the solids circulation.

In CLC, the circulation of solids supplies oxygen and heat to the fuel in the fuel reactor. Using a 1.5-kW continuous chemical looping combustor, Abad et al. (2012) developed a valve (situated between the cyclone and the fuel reactor) for diverting the flow of solids, enabling circulation measurement and control. However, mechanical valves are normally impractical or very difficult to manage in larger units. Dietrich et al. (2013) investigated four approaches to achieve the circulation of solids, with two of them supposedly being suitable for hot conditions. However, both of these methods rely on deactivating fluidization in a loop seal for a certain period of time, which might lead to agglomeration, sintering, and operation discontinuation for some oxygen carrier materials. Using a Ni-based oxygen carrier and natural gas as a fuel in a 10-kW CLC unit, Linderholm et al. (2009) developed a method for measuring the solids circulation based on the temperature difference between the AR and FR, which requires autothermal operation and full or close-to-full fuel conversion, thus being unsuitable for operation employing solid fuel. Using a 150-kW CLC unit for gaseous fuels, Kolbitsch (2009) showed that the mass flow of solids through a loop seal can be estimated from the corresponding pressure drop, i.e., the pressure difference between a point at a specific height on the incoming side of the seal and that at the same height on the exit side. This loss of pressure results from frictional forces of the two-phase flow (gas-solids) in the loop seal, increasing with increasing circulation. Studying two loop seals, Kolbitsch observed a linear correlation between the mass flow of solids and the abovementioned pressure drop. At identical circulation, different pressure loss responses were observed for the two studied loop seals, which was attributed to "slight differences in fluidization".