Superiority of Re-circulating Fluidized Bed Reactor Over Existing Reactor Arrangements for Chemical Looping Combustion—A Review

Sachin Tomar, Nitin Lokachari and Raman Sharma

Abstract Chemical looping combustion (CLC) is an oxy-combustion CO_2 capture technique. In CLC, the fossil fuels combustion is carried out in two parts using metal oxides. First, the metal oxides are oxidized with air. Second, the metal oxides are reduced using the fuel. To fulfill this criterion of combustion for CLC, some researchers have used interconnected reactors arrangements and some have used single reactor. In interconnected reactors arrangement, metal oxides oxidation is carried out in one reactor and the reduction in other. However, in single reactor arrangement, the oxidation and reduction of metal oxide particles are carried out in cycles. The existing reactors arrangements suffer from the problems of high bed particle attrition, short residence time of the bed material/metal oxides in the air reactor (fast fluidized bed), and generation of fine particles, transfer of bed material from one reactor to another, complex loop-seal operation, heat losses during the transfer of bed material from one reactor to another and cluster formation. Re-circulating fluidized Bed (RCFB) reactor is proposed, here, to overcome some of the problems associated with the existing reactor arrangements. RCFB reactor has a concentric pipe arrangement. The inner pipe is known as riser and the outer pipe is known as downcomer.

Introduction

Scientists are working on a way to capture and sequester carbon dioxide to deal with global warming. Carbon Dioxide (CO_2) allows sunlight to reach the Earth and also prevents some of the sun's heat from radiating back into space, thus warming the planet (Metz et al. 2005). Yes, carbon dioxide and the greenhouse effect are necessary for Earth to survive. However, human inventions like power plants and transportation vehicles, which burn fossil fuels, release extra CO_2 into the air

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thereby storing more amount of heat in the Earth's atmosphere. Carbon dioxide emissions alone increased by 80% in the past decade. Many researchers believe that the process of carbon capture and storage can help us to get this number down. Currently, there are many techniques readily available for carbon capture and storage (Haag 2007; Abad et al. 2007).

Chemical looping combustion (CLC) is a new technology that seems to work effectively for CO_2 capture. Carbon capture involves trapping the carbon dioxide at its emission source, transporting it to a remote location (usually deep underground) and isolating it (Mattisson et al. 2007). Initially, this process was proposed to increase the efficiency of thermal power plants; however, later on, it was found to have the inherent advantage of CO_2 capture avoiding costly equipment and energy consumption.

Chemical Looping Combustion (CLC)

CLC is an oxy-combustion CO_2 capture technique, which divides the process of combustion into two steps (Richter and Knocke 1983; Ishida et al. 1987). The first step takes place in an air reactor, where the oxidation of metal oxide particles using the compressed atmospheric air takes place (Eq. 1) (Lyngfelt et al. 2001; Leion et al. 2008; Mattisson et al. 2007) thereby stripping nitrogen from the air. The main exhaust gases from the air reactor are nitrogen and un-absorbed oxygen. After completion of this process, the oxidized metal particles or the oxygen carriers enter the fuel reactor, where the combustion takes place (Eq. 2) (Leion et al. 2008; Mattisson et al. 2007). Fuel gases are used to fluidize bed material in the fuel reactor which in the presence of oxygen completes the reduction cycle, thereby the combustion process (Mattisson et al. 2001). The main exhaust gases from the fuel reactor or the reduction cycle are CO_2 and H_2O. Once the water vapors are condensed, CO_2 can be separated easily (Leion et al. 2007; Mattisson et al. 2007).

$$\text{Air reactor (Oxidation cycle)}: \mathrm{Me} + 1/2\,\mathrm{O_2} \rightarrow \mathrm{MeO} \quad (1)$$

$$\text{Fuel reactor (Reduction cycle)}: (2n+m)\mathrm{MeO} + \mathrm{C}_n\mathrm{H}_{2m} \\ \rightarrow (2n+m)\mathrm{Me} + m\,\mathrm{H_2O} + n\,\mathrm{CO_2} \quad (2)$$

Commonly used metal oxides are Fe, Ni, Mn, and Cu (Jin et al. 1998; Brereton and Grace 1993; Xu et al. 2009).

CLC Requirements

- Metal oxide particles (MeO) and fuel gas contact should be maximum.
- High residence time of the MeO particles in the fuel reactor.
- High solid circulation rate between the two reactors, i.e., air and fuel reactors.
- Uniform reactor temperature.

Existing Reactor Configuration for CLC

The most common interconnected fluidized bed configurations for CLC is shown in Fig. 1 in which air reactor is operated at high velocity and the fuel reactor is operated at low velocity. In this configuration, cyclone separator is used to retain the bed material in the system, which leave air reactor and diverted them to fuel reactor. The position of the fuel reactor is slightly elevated so that the bed material can easily flow from the fuel reactor to the air reactor under the influence of the gravity. Bed material circulation and flow of gases are governed by the loop-seals between the air reactor and the fuel reactor and between the cyclone separator and fuel reactor. Bed material circulates between the air reactor and the fuel reactor with the help of high velocity in the air reactor(Kolbitsch et al. 2009; Shen et al. 2009;

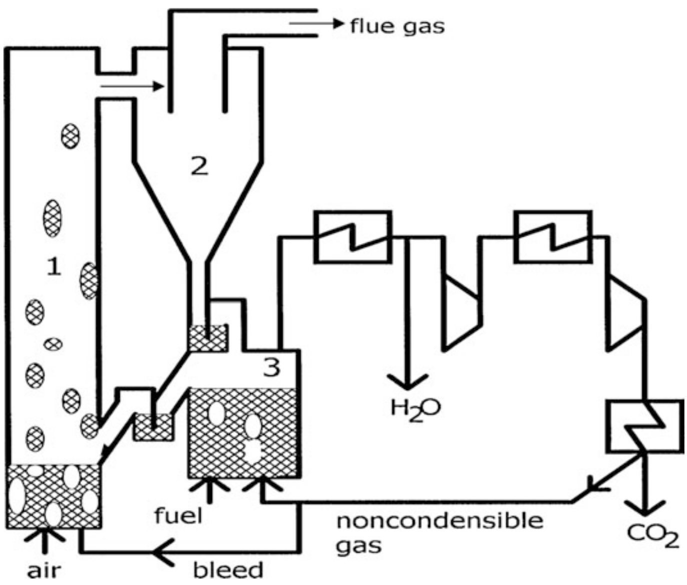


Fig. 1 Schematic diagram of interconnected reactor configuration for the CLC (Lyngfelt et al. 2001)

Basu and Cheng 2000; Johansson et al. 2003; Hossain and de Lasa 2008; Adanez et al. 2012).

Issues Related to the Existing Interconnected Reactors Arrangement

The main issues related to the existing interconnected fluidized bed reactors arrangements for chemical looping combustion (CLC), where air reactor is operated in a fast fluidization regime and the fuel reactor in bubbling fluidization regime are discussed here. The application of CLC principle for any of the fluidized bed systems mainly depends on the design of the fluidization column and the oxygen carrying particles (Merrow 1985). In order to overcome these problems, many modifications in the existing designs and many more new designs have been proposed. This paper mainly focuses on the following issues/drawbacks:

- Bed particle attrition
- Loop-seal operation
- Heat losses
- Cluster formation
- Fluidization gas bypassing
- Short residence time

Bed Particle Attrition

While moving in the reactor, the bed particles face attrition which is a mechanical stress due to inter-particle collision and bed-to-wall impacts. In the designing of the fluidized bed reactor, attrition is one of the main factors which are to be considered (Reppenhagen and Werther 2000). Due to the attrition, generation of fines take place which are finally passing through the dust recovery system resulting in the change in the properties of the bed material, particle size distribution gets too coarse, loss of valuable material, and wear and tear of the reactor (Bemrose and Bridgwater 1987). There are three main attrition sources in an interconnected reactor arrangement, namely, cyclone separator sections, the grid jet, and the bubbling bed as shown in Fig. 2. The cyclone separator attrition is higher than the jet and bubble-induced attrition at higher velocities. Equation 3 shows the overall loss rate as the sum of the individual sources responsible for the generation of fines.

$$\dot{m}_{\mathrm{loss,tot}} = \dot{m}_{\mathrm{loss},j} + \dot{m}_{\mathrm{loss},b} + \dot{m}_{\mathrm{loss},c} \tag{3}$$

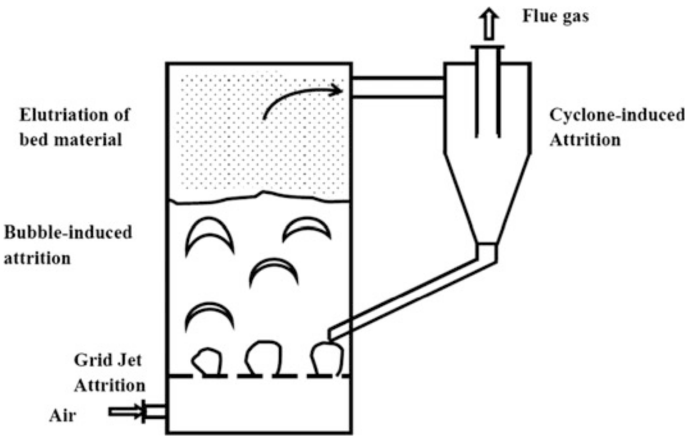


Fig. 2 Regions of bed particle attrition in an interconnected reactor configuration for CLC (Werther and Reppenhagen 1999)

where $\dot{m}_{\text{loss},j}$, $\dot{m}_{\text{loss},b}$ and $\dot{m}_{\text{loss},c}$ are the masses of elutriable fines that are produced per unit time by attrition in the jetting region, the bubbling bed, and the cyclone, respectively.

Loop-Seal Operation

Mechanical and nonmechanical valves are used for the transferring of the bed particles from one reactor to another reactor in the interconnected fluidized bed reactor system. Mechanical valves include rotary, screw, butterfly, and slide valves. These valves do not work properly under high temperature and pressure due to sealing and mechanical problem. At high temperature and pressure nonmechanical valves like loop-seal, *L*-, *J*-, *V*-valves (Knowlton and Hirsan 1978; Leung et al. 1987) are commonly employed to transfer bed particles from one reactor to another reactor by aeration as shown in Fig. 3. Loop-seal requires proper pressure balance around them, otherwise, seal failure may occur (Basu and Cheng 2000). Seal failure results in the gas bypassing which affects the overall efficiency of the CLC system. At a given gas velocity and solid circulation rate, the pressure drop across the loop-seal increases linearly with increasing solid inventory in the bed.

Heat Losses

In an interconnected fluidized bed reactor arrangement, there are many sites for the heat losses which include cyclone separator, loop-seal, transport line, standpipe, and reactor walls (Fig. 4) (Grace et al. 1987).

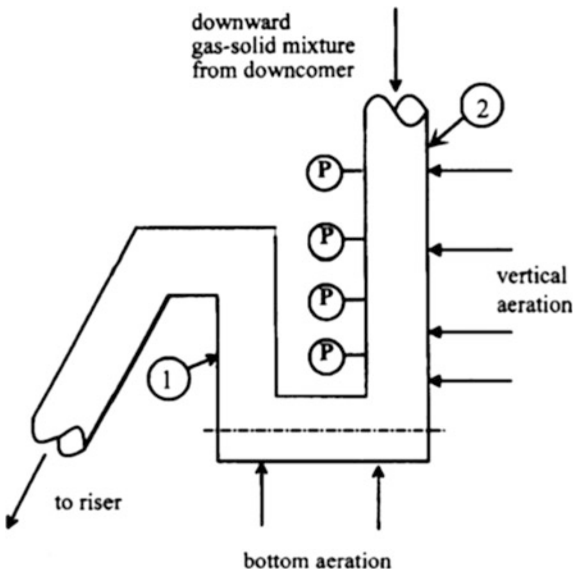


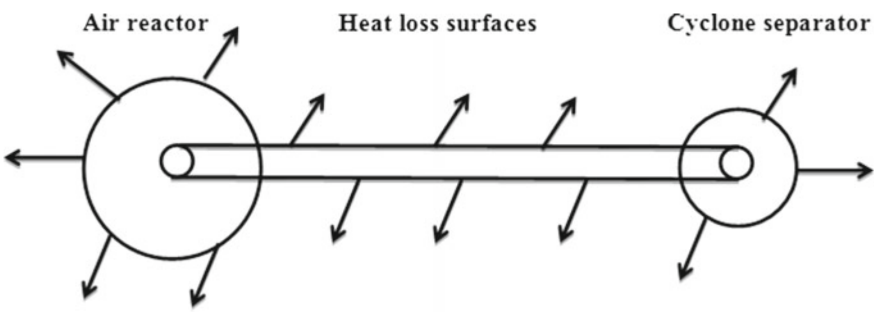
Fig. 3 Weir section (*1*) and vertical section of a loop-seal in an interconnected reactor configuration (*2*) (Kim et al. 1999)

Cluster Formation

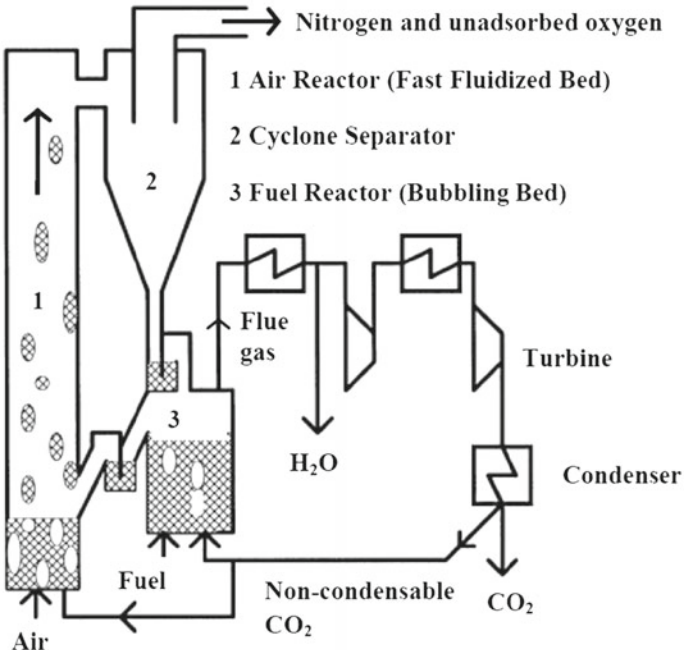
The metal oxide particles used for carrying oxygen from air reactor to the fuel reactor have a great tendency to agglomerate or cluster formation, which is not a favored phenomenon in any of the fluidized bed operations. This might result in back-mixing of the particles, which affects the overall heat transfer rate and improper chemical interactions between metal oxide particles and the gas used for fluidization (Brereton and Grace 1993; Bai et al. 1995). When operated in a fast fluidization regime, riser or the draft tube section has very few upward moving particles and the downcomer section has many downward moving clusters (Chandel and Alappat 2006).

Gas Bypassing

The 'Gas' here refers to the one that is used for fluidization, i.e., air and fuel in the air reactor and the fuel reactor respectively. Gas bypassing can be defined as the escape of gas (air) from jet to downcomer, which is actually directed into the riser and vice versa (Kolbitsch et al. 2009; Chandel and Alappat 2005). This leads to escape of unburnt fuel from the fuel reactor resulting in improper combustion (Diana et al. 2011).



(a) Top View (heat loss from air reactor, transport line, and cyclone separator)



(b) Side view

Fig. 4 Heat loss surfaces in an interconnected fluidized bed reactor

Short Residence Time

Fast fluidization regime is preferred in the air reactor, which might affect the interactions of gas and the metal oxide particles, thus leading to a lower residence time. As a result of this, oxidation of metal oxide particles is improper which ends up in decreasing the performance of the reactor (Xu et al. 2009). In order to overcome the issues related to existing fluidized bed designs/setups, several modifications, and many new designs have been proposed by the researchers and one such design is the Re-circulating fluidized bed (RCFB) reactor. The reactor's design and its working have been briefly discussed here.

Re-circulating Fluidized Bed Reactor Design for CLC

RCFB has a concentric pipe arrangement, where the inner tube is called the draft tube and the outer or annular region is known as the downcomer. The fluidizing gas is supplied to the base of the draft tube through the air jet. RCFB can be compared to that of a spouted fluidized bed with a draft tube (Alappat and Rane 2001; Yang and Keairns 1978, 1983). RCFB eliminates the requirement for the cyclone separator, instead uses the freeboard section at the top. RCFB also consists of spacer section, whose length is changeable and the bed particles reside on the perforated plate. The principle of CLC can be applied to the RCFB design, where the inner draft tube acts as an air reactor and the downcomer as the fuel reactor.

Working

The fluidizing gas (compressed air) from the air jet is sent into the draft tube section, which comes into contact with the bed material, takes it along the draft tube (Alappat and Rane 2000; Yang and Keairns 1974) and in the mean-time the particles get oxidized and enter the freeboard section, lose their energy and fall back into the downcomer section, where the fluidizing gas will be any fossil fuel, reduces the metal oxide particles. Though the process of combustion is divided into two parts, the main part remains to take place in the downcomer section i.e. fossil fuel reacting with the oxidized metal particles, producing energy. These particles again enter the bottom of the draft tube to start a new oxidation cycle. The schematic representation of RCFB is explained with the real-time cold model setup available in BITS Pilani (Fig. 5).

How RCFB overcomes the issues related to the existing designs? A design by the name RCFB has been introduced which can possibly overcome some of the issues related to the existing reactor arrangements. The construction of this particular fluidized bed is done in such a way, keeping in mind the sole purpose, i.e., to overcome the existing problems.

- As cyclone separator is the main site of attrition at the higher velocity, so in RCFB reactor a large cross-sectional area called freeboard section is used in place of cyclone separator to retain the bed particles in the reactor which results in lesser particle attrition than the existing reactor configuration, without losing the bed particles.

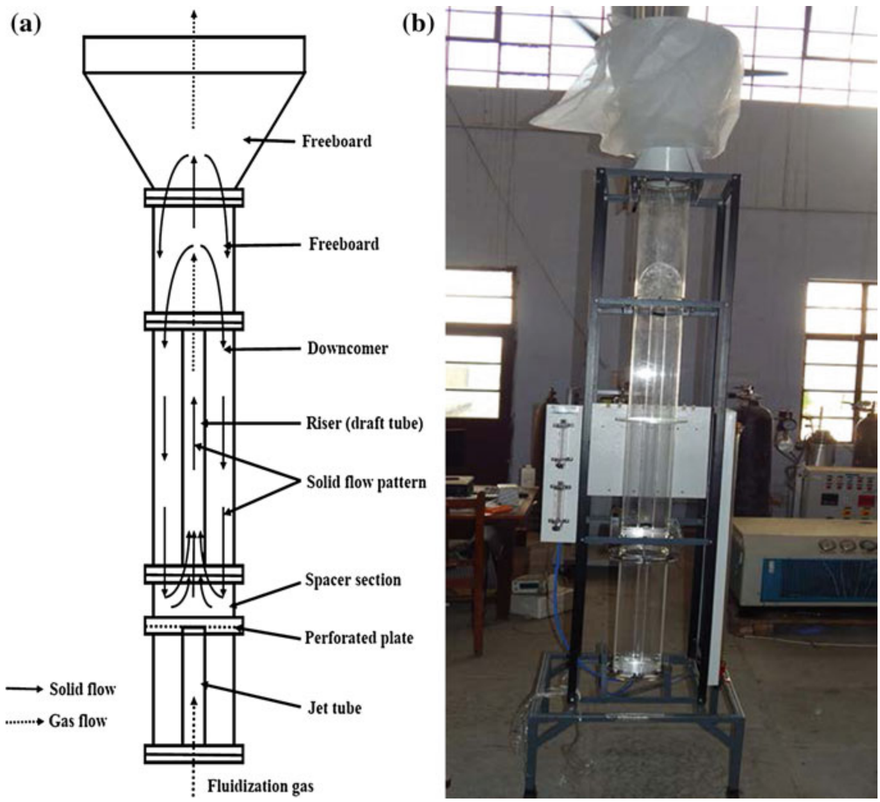


Fig. 5 **a** Schematic diagram of RCFB reactor. **b** Lab scale RCFB experimental setup in BITS Pilani

- The complex loop-seal arrangement is not needed in the RCFB reactor which prevents gas bypassing.
- In RCFB reactor configuration, cyclone separator and complex loop-seal arrangement are not used and hence less heat loss.
- The draft tube also induces high solid circulation rate and bed particles keep circulating within the reactors, which ensures no cluster formation.
- The extra travel path provided by the draft tube will ensure more contact time between the particles and the gas (Yang and Keairns 1978).
- Maintaining a large bed inventory in the spacer section and also the lesser length of the spacer section will not allow the gas to bypass.

Conclusions

Existing interconnected reactor arrangements for CLC have some issues that have been discussed in this paper. To overcome some of these issues, RCFB reactor has been proposed here. The draft tube in the RCFB ensures good solid mixing and solid circulation, longer residence time which results in maintaining uniform temperature throughout the reactor and better distribution of bed inventory and fuels. The construction of the RCFB reactor is not complex as it does not have cyclone separator and complex loop-seals, which makes it less expensive with flexible operating conditions. Further experimental studies are needed to verify the claims made.

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