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Fuel reactor model validation: Assessment of the key parameters affecting the chemical-looping combustion of coal

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ABSTRACT

The success of a Chemical Looping Combustion (CLC) system for coal combustion is greatly affected by the performance of the fuel reactor. When coal is gasified in situ in the fuel reactor, several parameters affect the coal conversion, and hence the capture and combustion efficiencies. In this paper, a mathematical model for the fuel reactor is validated against experimental results obtained in a 100 kW_{th} CLC unit when reactor temperature, solids circulation flow rate or solids inventory are varied. This is the first time that a mathematical model for Chemical Looping Combustion of coal with in situ gasification (*i*G-CLC) has been validated against experimental results obtained in a continuously operated unit. The validated model can be used to evaluate the relevance of operating conditions on process efficiency. Model simulations showed that the reactor temperature, the solids circulation flow rate and the solids inventory were the most relevant operating conditions affecting the oxygen demand. However, high values of the solids circulation flow rate must be prevented because they cause a decrease in the CO_2 capture. The high values of CO_2 capture efficiency obtained were due to the highly efficient carbon stripper. The validated model is a helpful tool in designing the fuel reactor to optimize the CLC process. A CO_2 capture efficiency of $\eta_{CC} = 98.5\%$ and a total oxygen demand of $\Omega_T = 9.6\%$ is predicted, operating at 1000 °C and 1500 kg/MW_{th} in the fuel reactor.

1. Introduction

In recent years, increasing attention has been paid to the application of Chemical-Looping Combustion (CLC) for coal combustion with CO_2 capture (Adanez et al., 2012). The CLC process is based on the transfer of oxygen from air to the fuel by means of a solid oxygen carrier which avoids direct contact between fuel and air. The oxygen carrier is composed of a metal oxide, Me_xO_y, and often uses an inert material acting as support. The CO_2 capture is inherent in this process. Fig. 1 shows a general scheme of the CLC system using coal as fuel.

A CLC system consists mainly of two reactors, namely air and fuel reactors, with the oxygen carrier circulating between the two. In the in situ Gasification Chemical Looping Combustion concept (*i*G-CLC), coal is fed to the fuel reactor, where the in situ gasification of coal takes place, generating volatile matter and gasification products through reactions (1)–(3). Reducing gases evolving during gasification are oxidized by reactions (4)–(6) with the oxygen carrier, Me_xO_y. The water–gas shift (WGS) – reaction (7) – can also be significant to the process as CO, H_2, CO_2 and H_2O are present in gases. The reduced oxygen carrier, Me_xO_{y-1}, is transferred to the air reactor where it is regenerated with air – reaction (8) – to be later transferred to the fuel reactor and start a new cycle. In *i*G-CLC, the CO_2 capture efficiency can be reduced if char particles are by-passed to the air reactor where they will be burnt (9), releasing some CO_2 with the depleted air.

$$\text{Coal}\,(s) \rightarrow H_2O\,(g) + \text{Volatile matter}\,(g) + \text{Char}\,(s) \tag{1}$$

$$\text{Char}\,(s) + H_2O\,(g) \rightarrow H_2\,(g) + CO\,(g) + \text{ash}\,(s) \tag{2}$$

$$\text{Char}\,(s) + CO_2\,(g) \rightarrow 2CO\,(g) + \text{ash}\,(s) \tag{3}$$

$$CH_4 + 4Me_xO_y \rightarrow CO_2 + 2H_2O + 4Me_xO_{y-1} \tag{4}$$

$$CO + Me_xO_y \rightarrow CO_2 + Me_xO_{y-1} \tag{5}$$

$$H_2 + Me_xO_y \rightarrow H_2O + Me_xO_{y-1} \tag{6}$$

$$H_2O + CO \leftrightarrow H_2 + CO_2 \tag{7}$$

$$Me_xO_{y-1} + ½O_2 \rightarrow Me_xO_y \tag{8}$$

$$\text{Char}\,(s) + O_2 \rightarrow CO_2 + \text{ash}\,(s) \tag{9}$$

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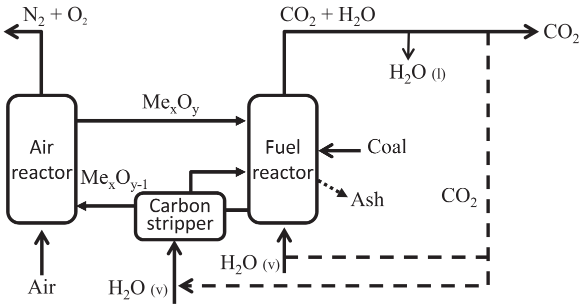

Fig. 1. Reactor scheme of the *i*G-CLC process for solid fuel.

A fundamental part of the reliability of a CLC system with coal is based on the behaviour of the fuel reactor. This will determine the loss of unburnt gas in the exit gas stream and the amount of char exiting from the fuel reactor. The separation of char from oxygen carrier particles in a carbon stripper and their recirculation to the fuel reactor has been proposed in order to reduce the carbon flow entering the air reactor (Cao and Pan, 2006).

Great advances have been made in the evaluation of operational conditions – e.g. reactor temperature (Berguerand and Lyngfelt, 2009; Cuadrat et al., 2011a,b; Gu et al., 2011; Song et al., 2013), solids circulation flow rate (Cuadrat et al., 2012a), fluidizing gas flow or composition (Cuadrat et al., 2012a), coal feeding rate or coal rank (Cuadrat et al., 2012b) – or the effect of oxygen carrier properties (Mendiara et al., 2013; Linderholm et al., 2012) on the CLC performance from experimental work in CLC units with coal. In all these cases, gasification of coal is an intermediate step taking place in the fuel reactor. In these studies, high CO_2 capture can be achieved by using high temperatures and/or implementing a carbon separation system between the fuel and air reactors. However, complete combustion of gases from the fuel reactor has not been achieved in existing *i*G-CLC units (Gayán et al., 2013).

Modelling and simulation of the *i*G-CLC system is an important tool for analysing the effect of various operational conditions. Thus, the main issues affecting the process have been identified. Some papers have been presented in the literature for modelling the process involved in the fuel reactor of an *i*G-CLC system with different degrees of complexity in their formulation (Cuadrat et al., 2012c; Kramp et al., 2012; Ströhle et al., 2010; Brown et al., 2010; Schöny et al., 2011; Mahalatkar et al., 2011; Abad et al., 2013; García-Labiano et al., 2013; Berguerand et al., 2011; Markström et al., 2013a). Any mathematical model should be validated against experimental results in order to be confident in its predictions. Simulation and validation of theoretical predictions are found for batch fluidized bed reactors (Brown et al., 2010; Mahalatkar et al., 2011). Results from analytical models have also been compared to results from continuously operated units in the range 0.5–100 kW (Cuadrat et al., 2012c; Berguerand et al., 2011; Markström et al., 2013a), but so far no comprehensive numerical model that has been validated against experimental results obtained from a continuously operated CLC unit with coal has been found in the literature. This validation step is required before the design, optimization, and scale-up of the process where a mathematical tool is used.

In this paper, a previously formulated (Abad et al., 2013; García-Labiano et al., 2013) mathematical model was validated against experimental results obtained in a 100 kW_{th} CLC unit erected at Chalmers University of Technology. Later, this model was used to determine the more relevant parameters influencing the performance of the CLC process. The information shown in this paper was then used to identify the operating conditions and design parameters which optimize the CO_2 capture and combustion efficiency of the process.

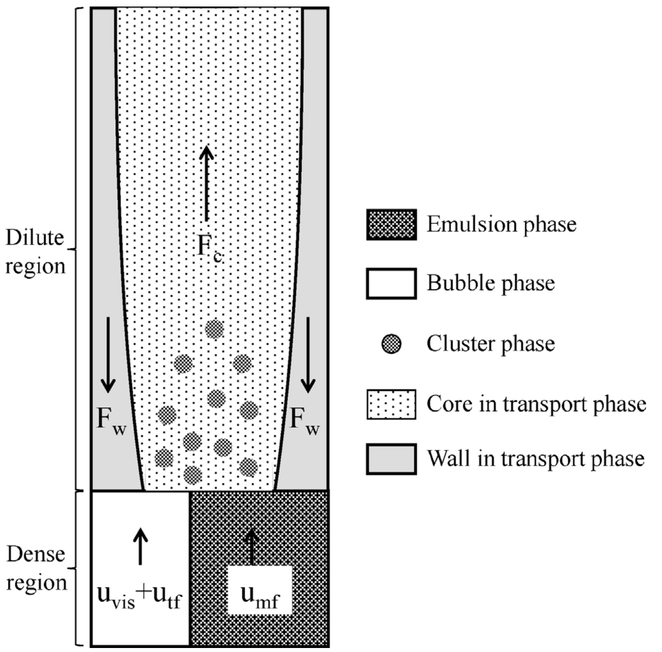

Fig. 2. Gas and solids distribution in the fuel reactor, considered as a high-velocity fluidized bed reactor.

2. Fuel reactor model

2.1. General description of the theoretical model

In a previous paper (Abad et al., 2013), a theoretical model describing the fuel reactor in the in situ Gasification Chemical-Looping Combustion process (*i*G-CLC) was presented. The model developed included the reactor fluid dynamics, coal conversion and reaction of the oxygen carrier with gases evolved from coal. More information on the model was presented in Abad et al. (2013) and García-Labiano et al. (2013)

The model developed was focused on the fuel reactor of the 1 MW_{th} CLC unit built at TU Darmstadt. Details about the 1 MW_{th} CLC unit can be found elsewhere (Orth et al., 2012). The fuel reactor was a fluidized bed working at a high-velocity regime. The fluid dynamic model considers the gas and solids flows inside the reactor and the gas–solids mixing patterns in the different regions in which it could be divided. The reactor was divided into two vertical regions with respect to axial concentration and backmixing of solids, see Fig. 2: (1) a dense region in the bottom bed with a high, roughly constant concentration of solids; and (2) a freeboard above the dense region, the dilute region, where there is a pronounced decrease in the concentration of solids as height increases.

The model can be considered 1.5 dimensional, with the main dimension in the axial direction. Gas distribution and mixing between the emulsion and bubbles in the dense region was taken into account. Thus, the gas flow in the dense region was shared between the emulsion and bubble phases, with gas mixing between the two, controlled by diffusion. Solids were in the emulsion phase, where the gas flow maintained the minimum fluidizing conditions; the remaining gas passed through bubbles, with no solids present. The dilute region had a cluster phase and a transport or dispersed phase. Both the cluster and transport phases were superimposed but with different mixing behaviour. The cluster phase had a strong solids backmix with solids in the dense region. The transport phase was characterized by a core/annulus flow structure. A net flow upward of solids passed through the core and particle backmixing occurred at the reactor walls. Also, a lateral exchange of solids