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1-D dynamic modelling and simulation of chemical looping combustion process

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A comprehensive simulation tool for the investigation of chemical looping combustion (CLC) process consisting of two interconnected fluidized bed reactors has been constructed. Dynamic fluidized bed hot-loop model is implemented into the Matlab/Simulink environment. Semi-empirical correlations are used for the calculation of reaction kinetics, heat transfer and flow dynamics. The main outputs of the model are the solid circulation rate, the conversion of the carrier and the gas composition at the reactor exit, the axial profiles of temperature and gas concentrations, and distribution of solids in the reactor. Various case studies with different boundary conditions, input parameters and reactor geometries show realistic process behaviour and stable operation.

Keywords: chemical looping combustion; CO_2 capture; dynamic modelling; process simulation; reactor system model

1. Introduction

Chemical looping combustion (CLC) has been introduced as promising combustion process with inherent separation of CO_2, initially by Richter & Knoche [1]. During the recent years, CLC has gained increasing amount of scientific interest, and it is said to have the potential for delivering the most efficient and economic technology in case of carbon capture and storage (CCS). A great number of papers considering different areas of CLC research has been listed and discussed by Hossain & de Lasa [2].

In a CLC system, the process shown in Fig. 1 is split into two interconnected fluidized bed reactors: an air reactor (AR) and a fuel reactor (FR) where two consecutive gas-solid reactions forming a chemical loop occur. A solid oxygen carrier (OC) is used to transfer the oxygen from the air to the fuel. The oxygen carrier is looping between the AR, where it is oxidized by the air, and the FR, where it is reduced by the fuel.

Depending upon the used metal oxide, the reduction reaction is often endothermic ($\Delta H_{red} > 0$), while the oxidation reaction is highly exothermic ($\Delta H_{oxd} < 0$). The total amount of released heat ΔH_c is the same as for normal combustion.

In CLC, the combustion air is not mixed with the fuel, and the CO_2 does not become diluted by the nitrogen as in the conventional combustion process. The water vapor can be condensed, and close to pure CO_2 is then obtained with minor losses of energy. In order to obtain high performance in CLC, an intimate contact between the oxygen carrier and gas phase species is important. Lyngfelt et al. [3] introduced a circulating system composed of two connected fluidized beds: high-velocity riser as an AR and low-velocity bubbling bed as a FR. Since the good gas-solids contact in the FR is crucial for avoiding possible bybass of fuel through the bubbling bed, Ruy et al. [4] and Kolbitsch et al. [5] proposed a circulating fluidized bed FR.

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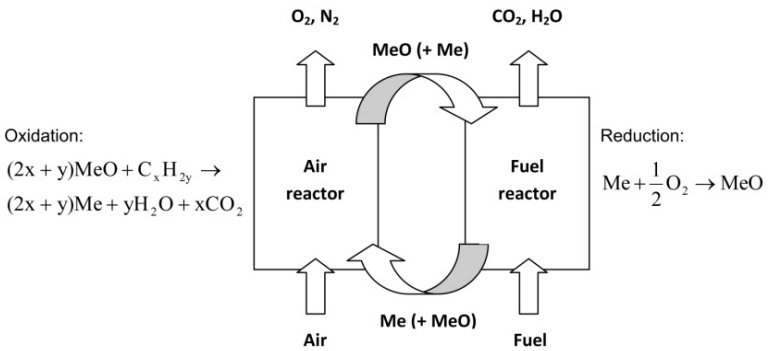

Fig. 1. The schematic picture of the CLC process.

Different models have been introduced in the literature for describing the operation of fluidized beds reactors. A wide-range review and comparison of circulating fluidized bed combustor (CFBC) models is provided by Basu [6]. Chen & Xiaolong [7], Muir et al. [8] and Park & Basu [9] demonstrated a dynamic modelling approach to predict the transient behaviour of a CFB combustor.

2. Description of the model

Studied CLC system uses CH_4 as fuel and consists of two interconnected fluidized beds, corresponding to the air and fuel reactors, separator and solids return system. Different process modules are horizontally divided into a number of 1-D elements, and for all the elements, the time dependent balance equations for mass and energy are written. Semi-empirical correlations are used for the calculation of reaction kinetics, heat transfer coefficients and flow dynamics. Each element is considered as an ideally mixed control volume. Gas and solid phases are calculated separately but the same average temperature is used for both phases. The process modules are connected together in Matlab/Simulink environment and steady state conditions with different input values are solved using ordinary differential equation solver**.**

The gas phase in the AR consists of four gas components, namely O_2, N_2, CO_2 and H_2O. In the FR, the gaseous fuel CH_4 exists at the gas phase as an additional gas component. For each gas component at the element, the mass fraction is solved using the general time dependent mass balance including reaction-based source terms for each component. The kinetic sub-model considers OC reactions in both air and fuel reactors. The reaction rate in the AR is defined for the formation of MeO (metal oxide) and the amount of O_2 is reduced by the reaction. In the FR, the reaction rate is defined for the oxidation of CH_4, and the amounts of CO_2 and H_2O are increased by the reaction, while the amount of the CH_4 is reduced. Due to the highly exothermic oxidation reaction, AR needs to be externally cooled. FR is kept adiabatic as the reduction reaction there is endothermic.

The solid phase (OC) is divided into MeO and Me (metal) by having the conversion ratio based on the mass fractions of the solid components. The time dependent conversion ratio in the element includes the source term from the OC reaction. To describe the hydrodynamics of fast fluidized bed, the vertical density profile of solids in the reactor is modelled by using an empirical correlation provided by Johnsson & Leckner [10]. In the current modelling approach, the reactors are divided into the core and wall layer regions. In the core region, the solids are moving upward, and downward in the wall layer. The wall layer thickness and density are estimated based on the reactor dimensions and fluidizing conditions. The mass flow entering into the wall layer is defined for each element, based on a given velocity parameter. Mixing of solids between the core and wall layer regions is modelled using a certain back flow ratio, which defines the mass flow from the wall layer back to the core.

In order to solve the time dependent temperatures of the elements, the energy equation of gas-solid suspension is written. The equation includes the convective heat flows of solids and gas mixture, energy dispersion due to mixing of solids by turbulence, energy source from chemical reactions and heat transfer to the internal heat exchangers. Convective heat flows are divided into the gas and solid phases and considered separately. The dispersion of energy decreases the temperature gradients between the elements, and thus, the axial temperature profile is equalized. The heat transfer rate into the internal heat exchangers can be calculated by using an empirical correlation for the total heat transfer coefficient, including convection and radiation, by Dutta & Basu [11].

3. Results and conclusions

The presented 1-D model is a useful tool for evaluating the performance of the chemical looping combustion. To study the model capabilities, simulation results of a CLC process, where methane was used as a fuel and NiO as oxygen carrier, were conducted. Methane conversion in a bubbling bed FR and conversion degrees of OC at the FR inlet and outlet, as a function of temperature are shown in Fig. 2 (a). As expected, the methane conversion increases with the temperature. In order to guarantee adequate methane conversion, the solid circulation rate of oxygen carrier should be well above theoretical limit value which leads to values $\Delta X = (X_{FR,in}\text{-}X_{FR,out}) << 1$. Modelled profiles for the oxygen volume fraction and reaction rate for the oxidation in the circulating fluidized bed AR for the same case are shown in Fig. 2 (b) and (c), respectively. In the current model, the kinetic parameters for the oxygen carrier reactivity are evaluated based on thermogravimetric data [12]. In order to validate the model, the material specific parameters for the carrier reactivity should be adjusted based on experiments in more realistic fluidized bed conditions.

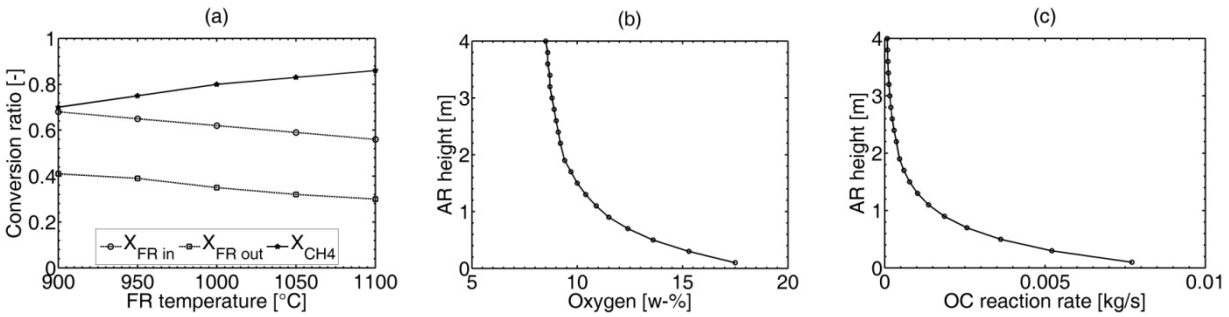

Fig. 2. (a) Methane conversion and conversion degrees at the FR as a function of temperature. **(b)** Mass fraction of oxygen and **(c)** reaction rate of oxidation as a function of height in the AR at T = 1050 °C. Simulations have been done with fuel power of 100 kW and global air-to-fuel ratio 1.1.

Reactions and fluid dynamics in a system of two interconnected fluidized bed reactors makes a complex system with many controlling parameters. After the validation of pilot scale dynamical model, the model offers a possibility to study the required material fluxes and needed control systems for the steady operation of a CLC plant also in larger scale.

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4. References

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