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Mathematical modeling of a two-stage fuel reactor for chemical looping combustion with oxygen uncoupling of solid fuels

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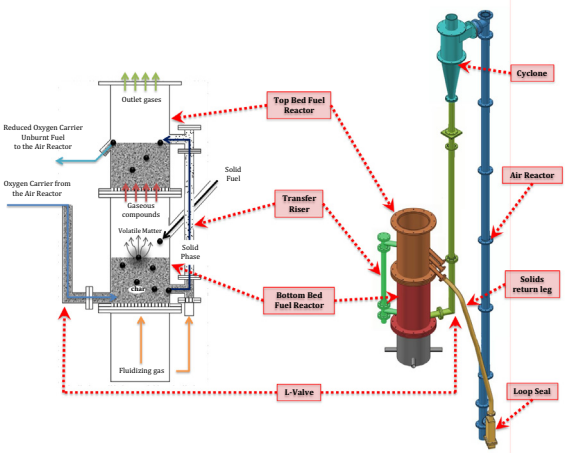
HIGHLIGHTS

- A novel two-stage fuel reactor for CLOU process of solid fuels has been proposed.
- Gas–solid co-current flow pattern was established in the fuel reactor.
- Mathematical modeling of the proposed fuel reactor has been developed.
- The performances of fuel reactor were evaluated varying operating conditions.
- The performances of different fuel reactor configuration were compared.

GRAPHICAL ABSTRACT



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ABSTRACT

The success of a Chemical Looping Combustion (CLC) process for solid fossil fuel combustion is critically affected by the performance of the oxygen carrier and by proper design and operation of the fuel reactor.

In this study, a novel configuration of the fuel reactor for chemical looping combustion with oxygen uncoupling of solid fossil fuels is proposed. The configuration is based on a two-stage reactor with the aim of overcoming the main drawbacks of the single-stage design: limited conversion, slip of unburnt volatiles, extensive elutriation of char fines. The two stages of the configuration operate in series and accomplish different tasks. The bottom bed is mainly devoted to conversion of the char, taking advantage of the full oxidative power of the oxygen carrier coming from the air reactor. The top reactor exploits the residual oxidative power of the oxygen carrier to oxidize volatile matter and gasification products as well as the unconverted char issuing from the bottom bed.

A mathematical model has been developed with the aim of assessing the performances of the two-stage fuel reactor varying operating conditions in comparison with a benchmark case consisting of a single-stage fuel reactor. Two options were considered in the benchmark, depending on whether the single stage fuel reactor is or is not equipped with a carbon stripper at the exhaust. The operation of the fuel reactor has been simulated by considering chemical looping combustion of a bituminous coal with an oxygen carrier consisting of CuO supported on zirconia.

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Nomenclature

a	char surface-to-volume ratio [m^2/m^3]
$c_{p,i}$	specific heat capacity [J/(mol K)]
d	Sauter mean particle diameter [mm]
E_C	amount of carbon fines elutriated from the fuel reactor [kg/s]
h	freeboard axial coordinate [m]
$[i]$	gas phase concentration [mol/m^3]
$\dot{m}_{char}^{in\ cs}$	inlet char flow rate from the fuel reactor to the carbon stripper [kg/s]
$\dot{n}_i$	molar flow rate [mol/s]
P_i	partial pressure [Pa]
$\dot{Q}$	thermal power [kW]
R	gas constant [J/(K mol)]
r_j	volumetric reaction rate [mol/(s m^3)]
R_j	reaction rate [mol/s]
S_{FB}	cross-section of the fuel reactor [m^2]
T	absolute temperature [K]
u	gas superficial velocity [m/s]
W_i	inventory in the fuel reactor [kg]
x_{CH_4}	methane conversion degree [–]
x_{char}	char conversion degree [–]
y_i	gaseous molar fraction [–]

Greek letters

α	stoichiometric coefficient [–]
$\Delta H_{R_j}^0$	standard enthalpy of reaction [J/mol]
η_{CC}	CO_2 capture efficiency [–]
η_{CO_2}	carbon-to-CO_2 efficiency [–]
η_{CS}	efficiency of the carbon stripper [–]
η_{CS}^{max}	maximum efficiency of the carbon stripper [–]
η_{FR}	combustion efficiency [–]
ρ_C	char density [kg/m^3]
Φ	oxygen-to-fuel ratio [–]
Ψ	kinetic parameter [–]

Subscripts or superscripts

0	standard state
AR	air reactor
D	dense zone in the fuel reactor
El	elutriated material
FB	Freeboard
FR	fuel reactor
S	oxygen carrier support
i	*i*th element or compound
in	Inlet
in-V	incoming volatile matter
j	*j*th reaction
mf	incipient fluidization
out	Outlet
R_j	moles reacted by the *j*th reaction

1. Introduction

Chemical Looping with Oxygen Uncoupling (CLOU) stems out as a very promising chemical looping combustion concept in applications to combustion of solid fossil fuels [1,2]. The CLOU concept is based on two cyclic steps, carried out in separate reactors. The first step, carried out in the Fuel Reactor, consists of the thermal reduction of the oxygen carrier, which thereby releases gaseous oxygen which becomes available for fuel oxidation. The second step, carried out in the Air Reactor, consists of the re-oxidation of the reduced oxygen carrier in air. The Fuel and Air Reactors are most typically arranged as dual interconnected fluidized bed reactors, an arrangement that enables continuous recirculation of the oxygen carrier between the two reactors, while preventing gas leakage from one reactor to the other. Accordingly, direct contact between the fuel and the atmospheric oxygen is avoided, flue gases issuing from the Fuel Reactor are mainly composed by CO_2 and H_2O, whence the latter can be easily separated by condensation.

Not any oxygen carrier is suitable for CLOU. The most promising ones are composed by an active phase consisting of metal oxides belonging to the transition group (Co, Cu, Mn). The choice of the oxygen carrier must meet several requirements: large oxygen transport capacity, high reactivity in both reduction and oxidation stages, limited cost, environmental friendliness, resistance to attrition and good fluidization properties [2].

Application of chemical looping with oxygen uncoupling to the conversion of solid fuels presents, however, some criticalities when compared with conversion of gaseous fuels. If the release of gas-phase oxygen during carrier reduction is not sufficiently fast, the performance of a single-stage fuel reactor may suffer from elutriation of unburnt carbon fines produced by attrition of fuel particles, from slip of volatile matter and gasification gaseous products generated by solid fuel and from extensive recirculation of unburnt char to the air reactor. The overall process efficiency may be significantly reduced [2,3].

Different fuel reactor configurations are currently under scrutiny to improve the performance of chemical looping combustion. First of all, the basic dual interconnected fluidized bed system made of a fast fluidization air reactor and a bubbling fluidized bed fuel reactor has been integrated with a carbon stripper. The stripper, either located at fuel reactor exhaust or inside it [4–13], separates unburnt char particles from the oxygen carrier present in the solid stream directed to the air reactor, so that the former can be recirculated to the fuel reactor. Accordingly, the amount of carbon burnt in the air reactor is reduced and the CO_2 capture efficiency is increased. Novel designs of the fuel reactor have been targeted at approaching plug flow patterns where gas and solids are directed in counter-current flow patterns in multi-stage dense fluidized beds or moving beds [14–22]. Thanks to a tailored residence time distribution (RTD) of gas and solids, these novel fuel reactor designs may improve the conversion of solid carbon and of volatiles/gasification products increasing the contact time between combustible gases and oxygen carrier.

Pröll and Hofbauer [14] proposed a gas–solid counter-current flow pattern for the fuel reactor where the gas–solid contact is increased by the presence of several and equally-spaced cross section restrictions along the fuel reactor. Dense fluidized beds are established above the restrictions generating a multi-stage configuration with RTD characteristics approaching plug flow [15,18]. Fuel feeding is located along the fuel reactor in order to increase the residence time of char particles and volatiles and to improve gas–solid contact between the combustible gases and the oxygen carrier particles.

Werther and co-workers proposed a two-stage bubbling fluidized bed fuel reactor characterized by gas–solid counter-current flow pattern and by fuel feeding at lower bubbling fluidized bed

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