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## Article

# Dynamic simulation and control system for chemical looping combustion

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## Dynamic simulation and control system for chemical looping combustion

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#### Abstract

Chemical looping combustion (CLC) is a technique for separating carbon dioxide from flue gas. It consists of two main processes: a fuel reactor and an air reactor. In the fuel reactor, fuel reacts with solid metal oxide particles to produce heat,  $CO_2$  and  $H_2O$  vapor which later can be easily separated by the condensation. The reacted solid metal is then sent to the air reactor for regenerating via an oxidation reaction with air before being returned to the fuel reactor. In this study, these reactors in the CLC system are integrated as a circulating fluidized bed system and a dynamic model has been developed for the system. The fuel reactor (or downer) operates in bubbling fluidization regime where the air reactor (or riser) operates in fast fluidization regime. Using the developed dynamic model, the transient responses of various control variables such as the temperature of two fluidized bed reactors, pressure and the liquid level of a condenser have been controlled by manipulating variables such as heat transfer rates of the reactors, gas outlet flow rate and liquid outlet flow rate of the condenser. A proportional–integral–derivative (PID) controller was tuned for controlling purpose. The integral of the absolute value of error (IAE) is used to evaluate the control system performance.

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Keywords: Chemical looping combustion; Fluidized bed reactors; Aspen Dynamics; Dynamic simulation

#### 1. Introduction

Process simulation of a CLC has been carried out with a variety of numerical methods and by several researcher. It can be divided into steady state and dynamic simulation. Steady state simulations typically find one solution for each set of operation conditions. For dynamic or transient simulations, operation parameters and, hence, process

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conditions can vary over time. These results show how the processes respond to external changes which provide more understanding to the process behavior. Aspen Plus software is commonly used by industries to simulate a variety of chemical and petrochemical processes. The software provides operational blocks to implement most of process units. A simulated plant is constructed by connecting relevant operational blocks through mass and energy streams. Then certain process condition and parameters are specified, and Aspen Plus will solve for unspecified condition or parameters. However, the major disadvantage of this kind of software is that it only provides basic unit operations but it does not provide a complex unit such as fluidized bed reactors. For this reason, several efforts were carried out in the past to mimic the real behavior of fluidized bed reactors [1]. A way to solve this issue has been proposed by Liu et al. [2], Jafari et al. [3] and Sotudeh-Gharebaagh et al. [4] who employed a number of basic blocks, e.g. ideal reactors usually continuous stirred-tank reactors (CSTRs) and plug flow reactors (PFRs), combined in a fashion that could simulate the real hydrodynamics and/or kinetics inside the reactor. Steady state simulations of a fluidized bed for the CLC have been developed by Porrazzo et al. [5]. The model utilized a series of CSTR and PFR models to mimic the behaviors of fast fluidized bed or bubbling beds, respectively.

In macro level approach, dynamic models needed for the CLC with methane were developed in MAT-LAB/Simulink by Peltola et al. [6]. Later, solid fuel combustion with CLOU were implemented [7]. The models were principally be able to capture dynamic effects. Haus et al. [8] modeled the entire CLC process by using dynamic simulation of solid processes (Dyssol). The basis of this software is a sequential modular. All of these unit operations were calculated in a way to allow for dynamic changes of the involved solids.

The aim of the present work is to develop a dynamic model and the control system to demonstrate the interdependency of interconnected two fluidized bed reactors, which are linked via a cyclone, operated as a chemical looping system. The control system was designed and tested their performances with the possibility of changing set points of the controllers and disturbances.

#### 2. Methodology

In this study, Aspen Plus was used to generate a flowsheet for a CLC system. Pure methane was used as representative of natural gas feedstock and NiO/Ni was chosen as an oxygen carrier due to its high oxygen carrier capacity, high melting point and high heat of reaction, compared to other carriers. Typically, both fuel and air reactors can work at high reaction temperature. The reduction reaction with methane was selected because of the availability of kinetic data from the literature [9]. It is useful for implementing and validating the proposed model. The reactor model. previously discussed [5], was applied after scaling up to 100 MW by changing its volume and keeping the same hydrodynamic conditions as shown in Tables 1 and 2.

Table 1. Main parameters used for simulating the fuel reactor.

Name	Parameter	Value	Unit
Operating temperature	T <sub>FR</sub>	1350	°C
Operating pressure	P <sub>FR</sub>	10	atm
Inlet superficial gas velocity	Uo	0.36	m/s
Inlet solid mass flow rate	$F_{\rm s}$	25	kg/s
Gas volume fraction in bubble phase	$\varepsilon_b$	0.9	-
Gas volume fraction in emulsion phase	$\varepsilon_m$	0.5	-
Volume in fuel bubbling bed	V <sub>total</sub>	133	m <sup>3</sup>

Table 2. Main parame	ers used for	simulating the	he air reactor.
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Name	Parameter	Value	Unit
Operating temperature	$T_{\rm AR}$	1350	°C
Operating pressure	P <sub>AR</sub>	10	atm
Inlet superficial gas velocity	$U_{\rm o}$	7.7	m/s
Inlet air mass flow rate	Fair	22	kg/s
Inlet air temperature	T <sub>air</sub>	320	°C
Solid volume fraction in dense phase	$\varepsilon_{sd}$	0.16	_
Total volume in riser	V <sub>total</sub>	187.4	m <sup>3</sup>

 Table 3. Kinetic parameters and physical properties of solid particles.

Name	Reduction reaction	Oxidation reaction	Unit
Kinetic coefficient $k_0$	0.71	$1.8 \times 10^{-3}$	$mol^{(1-n)} m^{(3n-2)} s^{-1}$
Activation energy $E_a$	78,000	7000	$J mol^{-1}$
Order of reaction n	0.8	0.2	-

#### 2.1. Reaction conditions

The reactions involved in the analysis are the following Eqs. (1) and (2). The kinetic data for the NiO reduction and the physical properties of the solid particles was adopted from [9] as shown in Table 3. Reduction reaction

$$CH_4 + 4NiO \rightarrow CO_2 + 2H_2O + 4Ni \tag{1}$$

Oxidation reaction

$$2O_2 + 4Ni \rightarrow 4NiO$$
 (2)

The reduction and the oxidation rates in the CLC system are usually expressed in percentage of solid conversion over time ([=]% min<sup>-1</sup>). Aspen Plus does not accept a kinetic expression in % min<sup>-1</sup> unit and thus, some manipulations of the kinetic expressions are needed. The modified kinetic expression for shrinking core model was applied to spherical grain geometry for both reduction and oxidation reaction.

#### 3. Simulation setup

#### 3.1. Steady state model

For a bubbling bed fuel reactor, the bubble growth is determined by gas passing through the bed and thus part of the gas does not encounter the solid particles. Kunii and Levenspiel [10] proposed various strategies to model the actual behavior of the fluidized bed depending on the class of particles involved. According to this theory, a fluidized bed was assumed to be composed of two phases [11]: the bubble phase at low solid content, and the emulsion phase characterized by perfect mixing of solids. The two-phase model pointed out the importance of the gas mass transfer between the bubble phase and the emulsion phase. Under Aspen Plus environment, several authors proposed combinations of CSTRs and PFRs reactors to mimic the real behavior of the fluidized bed [5] The bubble phase was modeled with plug flow reactors (PFRs) and the emulsion phase was modeled with perfect mixing reactors (CSTRs) in Fig. 1. The calculator blocks (in FORTRAN) were used to calculate the mass transfer between the emulsion and the bubble phases and transfer blocks were used to verify the mass continuity in the system.

The air reactor, or the riser, was operated in the fast fluidization regime, where its superficial gas velocity,  $U_o$ , was higher than the terminal velocity of solid particles,  $U_t$ . In this regime, gas and solid were assumed to be perfectly mixed and two regions were identified in the bed: a dense phase at the bottom of the bed, characterized by high average solid concentration, and a lean phase, above the dense phase, characterized by an exponential decrease of the solid volume fraction along the height of the bed. The implementation of the riser employed four CSTRs in series one CSTR was used to simulate the dense phase and the three CSTRs were used to simulate the lean phase as shown in Fig. 1, at different values of the solid volume fraction and thus different kinetics rates.

#### 3.2. Dynamic model

Aspen Plus provides a library of models that can be used to develop bubbling bed models. Thus steady-state design and optimization studies of the processes with bubbling bed model can be evaluated. Typically, these models can be exported and evaluated their dynamic responses in Aspen Dynamics. However, in our case, this cannot be done due to the developed model containing calculator blocks and transfer blocks which are inactive in Aspen Dynamics. Therefore, this paper presents a simple approximate method for achieving the objective of having



Fig. 1. Process flow sheet of the CLC model in Aspen Plus.



Fig. 2. Process flow sheet of the CLC model in Aspen Dynamics.

fluidized bed reactors model that can be exported and evaluated in Aspen Dynamics. The basic idea is to simplify the responses of the bubbling phase and emulsion phase with lumped reactor models having the same dynamic responses. In this case, the CSTRs were employed as shown in Fig. 2. Their dynamic properties were determined by system properties such as equipment sizes, stream flow rates.

Fig. 2 shows the interconnected fluidized bed reactors linked by a cyclone and its control structure. The system was controlled by several control loops. Temperature controller 1 (TC1) and Temperature controller 2 (TC2) were employed for controlling the reactor temperatures by using constant duty method. This is the simplest method of the heat transfer option, but it is unrealistic since it does not consider the dynamic response taking place to transfer the heat between units. Therefore, time constant blocks were implemented to represent response time of the heat units. Time constant 1 and 2 (Time\_1 & Time\_2) of 15 min and 20 min were implemented for each heat unit, respectively. The level controller 1 (LC1) and pressure controller 1 (PC1) were implemented for the separator.

#### 4. Results and discussion

#### 4.1. Comparative results: lumped model vs. bubbling bed model

The two reactor models (lumped model and bubbling bed model) were tested under the same operating condition, superficial gas velocity, mass flow rate and volume of reactors. The bubbling bed model was simplified by using a



Fig. 3. Conversion both gas and solid between two models by using kinetic data from Abad et al.



Fig. 4. Conversion both gas and solid between two models by changing kinetic data from Abad et al.

lumped model. The comparison at steady state condition in Fig. 3 showed that the simplified model gives higher conversion in terms of gas/solid conversion than the bubbling bed model since the by-pass gas through the bubble phase was neglected in the lumped model. To obtain the proper responses, the activation energy from Abad et al. were adjusted and compared the result with bubbling bed model. The differences of gas conversion between the two models were less than 10% as shown in Fig. 4.

#### 4.2. Relative gain array (RGA)

To develop proper control loops and select the most promising control structures, the pair of manipulated and controlled variables were evaluated using RGA analysis [12]. The open-loop gain matrix for each scenario was obtained from steady-state conditions. Table 4 lists the controlled, manipulated variables and their selected variables for the present analysis.

		1			
Controlled variables	Parameter	Set point	Manipulated variables	Parameter	Nominal steady state value
Temperature of fuel reactor	$T_{\rm FR}$	1350 °C	Heat transfer rate of fuel reactor	$Q_{\rm FR}$	-15.12 MW
Temperature of air reactor	$T_{\rm AR}$	1350 °C	Heat transfer rate of air reactor	$Q_{\mathrm{AR}}$	-12.89 MW
Level liquid of condenser	$H_{\text{SEP}}$	0.385 m	Outlet liquid flow rate of condenser	$F_{ m L}$	12,824.70 kg/h
Pressure of condenser	P <sub>SEP</sub>	10.13 bar	Outlet gas flow rate of condenser	$F_{\rm G}$	390.15 kmol/h

Table 4. List of controlled variables and manipulated variables.

	$Q_{ m FR}$	$Q_{ m AR}$	$F_{ m L}$	$F_{ m G}$
T <sub>FR</sub>	9.997E-01	8.756E-09	3.066E-04	-1.565E-07
$T_{\rm AR}$	-1.299E-09	9.999E-01	1.060E - 04	-1.498E-09
$H_{\rm SEP}$	3.064E-04	1.060E - 04	1.002E+00	-2.401E-03
$P_{\text{SEP}}$	-1.522E-10	-6.645E-11	-2.402E-03	1.002E+00

Table 5. Relative gain array (RGA).

The RGA matrix is calculated as follows Eq. (3).

$$\Lambda = K \otimes \left(K^{-1}\right)^T \tag{3}$$

The RGA matrix is shown in Table 5. According to the rules of pairing for RGA, the relative gain elements  $(\lambda_{ij})$  between the controlled variable *i* and the manipulated variable j that is closed to one will be selected. Thus, the results suggested the pairings of T<sub>FR</sub>-Q<sub>FR</sub>, T<sub>AR</sub>-Q<sub>AR</sub>, H<sub>SEP</sub>-F<sub>L</sub>, P<sub>SEP</sub>-F<sub>G</sub> due to their relative gain elements.

#### 4.3. Performance evaluation

PID controllers were implemented for each control loop. Controller's parameters, which are controller gain ( $K_c$ ) integral time ( $\tau_I$ ) and derivative time ( $\tau_D$ ), were tuned using Ziegler–Nichols closed loop method [13]. The controller parameters were determined as shown in Table 6.

Table 0. Tun	ing parameters it	a control structur	с.		
Name	CV	MV	K <sub>c</sub>	$ au_{ m I}$	$ au_{\mathrm{D}}$
TC1	$T_{\rm FR}$	$Q_{\mathrm{FR}}$	30.35	0.15	0.024
TC2	$T_{\rm AR}$	$Q_{\mathrm{AR}}$	34.43	0.15	0.024
LC1	$H_{\text{SEP}}$	$F_{ m L}$	412.89	0.075	0.012
PC1	$P_{\rm SEP}$	$F_{ m G}$	23.31	0.075	0.012

Table 6. Tuning parameters for control structure.

The performances of the controllers were evaluated by using Integral of the Absolute value of Error (IAE). It was calculated as follow Eq. (4) [14] to show integral of the absolute value of error of processes against its set point. For example, Figs. 5–8 show the responses of the control loops when temperature of fuel reactor was raised by 50 °C. The system reached the new steady state at 300 s and integral of the absolute value of error was 0.18. Other scenario was tested by changing the controller set points and the gas input flow rate as shown in Table 7.



Fig. 5. Response of temperature controller (TC1).

#### 5. Conclusion

In this study, the steady-state and dynamic models of the natural gas chemical looping combustion were developed using Aspen Plus and Aspen Dynamics, respectively. The models were used to evaluate their dynamic responses



Fig. 6. Response of temperature controller (TC2).



Fig. 7. Response of level controller (LC1).



Fig. 8. Response of pressure controller (PC1).

and the control structures were determined. The control loops were set up and the controllers were tuned and tested their performance under several transient conditions. It was found that the control system satisfactory performed with the changing of the assigned disturbances and set points.

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Scenario	Integral of Absolute value of Error (IAE)					
	TC1	TC2	LC1	PC1	Total	
+50 °C T <sub>FR</sub>	1.8E-01	3.3E-04	4.9E-06	6.0E-03	1.8E-01	
$-15$ °C $T_{\rm FR}$	3.5E-02	1.5E-04	6.6E-06	1.6E-02	5.1E-02	
+50% F <sub>fuel</sub>	5.6E-05	0.0E + 00	1.6E-07	7.4E-05	1.3E-04	
$-50\%$ $F_{\text{fuel}}$	5.8E-04	6.7E-04	7.1E-06	1.3E-02	1.4E-02	
$+50$ °C $T_{AR}$	2.8E-04	2.1E-01	5.2E-07	2.7E-04	2.1E-01	
$-10$ °C $T_{AR}$	1.5E-04	2.0E-02	1.3E-07	4.5E-05	2.0E - 02	
+50% Fair	0.0E + 00	0.0E + 00	3.1E-08	0.0E + 00	3.1E-08	
-50% Fair	0.0E + 00	2.6E - 04	2.5E-07	1.3E-04	3.9E-04	
+20% H <sub>SEP</sub>	0.0E + 00	0.0E + 00	7.6E-04	1.5E-05	7.8E-04	
-20% H <sub>SEP</sub>	0.0E + 00	0.0E + 00	7.6E-04	1.4E-05	7.7E-04	
$+2$ bar $P_{\text{SEP}}$	0.0E + 00	0.0E + 00	1.5E-06	1.9E-03	1.9E-03	
$-2$ bar $P_{\text{SEP}}$	0.0E + 00	0.0E + 00	2.0E-06	2.6E-03	2.6E-03	

 Table 7. Summary of the performance evaluation.

#### References

- [1] Porrazzo R, White G, Ocone R. Fuel reactor modelling for chemical looping combustion: From micro-scale to macro-scale. Fuel 2016;175:87–98.
- [2] Liu B, Yang X, Song W, Lin W. Process simulation development of coal combustion in a circulating fluidized bed combustor based on Aspen Plus. Energy Fuels 2011;25(4):1721–30.
- [3] Jafari R, Sotudeh-Gharebagh R, Mostoufi N. Modular simulation of fluidized bed reactors. 2004, p. 123-9.
- [4] Sotudeh-Gharebaagh R, Legros R, Chaouki J, Paris J. Simulation of circulating fluidized bed reactors using ASPEN PLUS. Fuel 1998;77(4):327–37.
- [5] Porrazzo R, White G, Ocone R. Aspen plus simulations of fluidised beds for chemical looping combustion. Fuel 2014;136:46-56.
- [6] Peltola P, Ritvanen J, Tynjälä T, Pröll T, Hyppänen T. One-dimensional modelling of chemical looping combustion in dual fluidized bed reactor system. Int J Greenh Gas Control 2013;16:72–82.
- [7] Peltola P, Ritvanen J, Tynjälä T, Hyppänen T. Fuel reactor modelling in chemical looping with oxygen uncoupling process. Fuel 2015;147:184–94.
- [8] Haus J, Hartge E-U, Heinrich S, Werther J. Dynamic flowsheet simulation for chemical looping combustion of methane. Int J Greenh Gas Control 2018;72:26–37.
- [9] Abad A, Adánez J, García-Labiano F, de Diego LF, Gayán P, Celaya J. Mapping of the range of operational conditions for Cu-, Fe-, and Ni-based oxygen carriers in chemical-looping combustion. Chem Eng Sci 2007;62(1):533–49.
- [10] Kunii D, Levenspiel O. Fluidized reactor models. 1. For bubbling beds of fine, intermediate, and large particles. 2. For the lean phase: freeboard and fast fluidization. Ind Eng Chem Res 1990;29(7):1226–34.
- [11] Davidson J, Harrison D. Fluidized particles. Cambridge, New York: Cambridge University Press; 1963.
- [12] Bristol EH. On a new measure of interaction for multivariable process control. IEEE Trans Automat Control 1966;11(1):133-4.
- [13] Ziegler JG, Nichols NB. Trans ASME 1942;759-68.
- [14] Seborg DE, Mellichamp DA, Edgar TF, Doyle FJ. Process dynamics and control. John Wiley Sons; 2010.