

# Utilizing Chemical Looping Combustion instead of Fired-Furnace in a Steam Methane Reforming for Enhancement of Hydrogen Production in a Multi Tubular Reactor

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## Abstract

A novel thermally coupled reactor containing steam methane reforming in the endothermic side and chemical looping combustion as an exothermic side has been investigated in this study. In this innovative configuration, huge fired furnace of conventional steam reforming process is substituted by chemical looping combustion in a recuperative coupled reactor. This reactor has three concentric tubes where the steam methane reforming is supposed to occur in the middle tube and the inner and outer tubes are considered to be air and fuel reactors of chemical looping combustion, respectively. Copper is selected as solid oxygen carrier in the chemical looping combustion process. Both oxidation and reduction of Cu in the air and fuel reactor are exothermic and used as heat sources for endothermic steam methane reforming. A steady state heterogeneous model of fixed bed for steam reformer and a moving bed for chemical looping combustion reactor predict the performance of this new configuration. The counter-current mode is investigated and simulation results are compared with corresponding predictions of the conventional steam reformer. The results prove that synthesis gas production is increased in thermally coupled reactor in comparison with conventional steam reformer.

**Keywords:** Hydrogen production; Steam reforming of methane; Thermally coupled reactor; Chemical looping combustion; Cu- based oxygen carrier.

## I. Introduction

Nowadays, there has been an enormous increase in energy demands due to fast growing of industrial development and population of the world. The shortage of fossil fuels as a main energy sources has brought energy crisis. In order to eliminate fossil fuels dependence, comprehensive research has been carried out on searching alternatives energy resources. Hydrogen is an excellent candidate due to its high potential energy.

### 1.1. Hydrogen

Hydrogen, the most common element in the earth, has been considered as an attractive energy carrier to support energy consumption. Hydrogen is an environmentally friendly, efficient, safe and sustainable energy source (Lokurlu et al., 2003; Heinzl et al., 2002). The products of hydrogen combustion are water and tiny amount of  $\text{NO}_x$  which can be reduced by proper methods. The use of hydrogen in energy sector enhances the security of energy supply and improves economic competitiveness (Muller-Langer et al., 2007). Except the nuclear fuels, hydrogen has the highest calorific value among fuels. A fuel contains higher proportion of hydrogen provides more energy (Sun et al., 2012). Therefore, pure hydrogen would be the leading fuel which can satisfy the increasing demand asked by many processes such as: methanol, electricity, ammonia, aniline, oil refining, fuel cell, vehicle engines, power plants, etc (F.Brown, 2001; Itoh et al., 2008). In fact, hydrogen is a secondary energy produced from traditional processes like: natural gas reforming, gasification of coal, electrolyses water, biomass gasification, catalytic steam reforming of natural gas, dehydrogenation of cyclic hydrocarbons, etc (Sun et al., 2012). It must be noted that hydrogen production must be low in  $\text{CO}_2$  emissions and other pollutants. Among all mentioned technologies, catalytic steam reforming of natural gas is widely used for hydrogen production, 80-85% of the world's total hydrogen production is provided by this method (Simpson and Lutz, 2007).

### 1.2. Steam reforming

Steam reforming technology is the most commercial method for synthesis gas ( $\text{CO}$ ,  $\text{H}_2$ ) production, the hydrogen cost is less than hydrogen produced by using renewable energy sources or from gasification of solid fossil fuel (Rostrup-Nielsen, 1993; Tugnoli et al., 2008). Steam reforming may involve several catalytic steps: desulfurization of the fuel, steam reforming of methane, a water gas shift reactor and purification of hydrogen using PSA unit. This method is suitable for light hydrocarbons such as natural gas (mainly  $\text{CH}_4$ ), naphtha, liquefied petroleum gas (Ryden and Lyngfelt, 2006). Three main catalytic reactions involved in steam reformer reactor are steam reforming of methane (SRM) and the water-gas shift reaction.

Conventional steam reformer consists of vertical tubes packed with Ni-based catalyst located inside huge furnace. The required heat for endothermic reforming reaction is provided by direct combustion of fuel in the furnace. Therefore, the reformer tubes are under very high thermal stress (F.Brown, 2001). In order to solve this issue, the recuperative coupled reactor has been suggested.



### 1.3. Coupled reactors

Recently, Coupling exothermic and endothermic reactions are more interested in order to improve the thermal efficiency of process and consequently enhance the production. This type of reactors aims to use energy released by exothermic reaction for proceeding endothermic reaction. In general, the coupled reactors exist into three main classes: direct coupling, recuperative coupling and finally regenerative coupling. At present, recuperative coupling has attracted the most attention of many researchers (Song et al. 2003). Hunter and McGuire (1980) were pioneers in

coupling endothermic and exothermic reactions without direct heat transfer. A review on the process intensification for methane steam reforming in a thermally coupled membrane separation technology was studied by Bhat and Sadhukhan (2009). Patel and Sunol (2007) suggested a thermally coupled membrane reactor that is composed of three channels for methane steam reforming. A numerical model for natural gas steam reforming and coupling with a furnace was developed by Ventura and Azevedo (2010). In an interesting idea, Ryden and Lyngfelt (2006) studied steam methane reforming coupled with chemical looping combustion reactor in order to enhance  $H_2$  production with  $CO_2$  capture. In their suggested configuration, reformer tubes are located inside the fuel reactor of chemical looping combustion. A disadvantage of their innovative configuration is erosion of reformer tubes due to harsh environment of the fuel reactor. This issue can be solved by using thermally coupled multitubular reactor which is the main goal of this study.

#### 1.4. Chemical looping combustion

Chemical looping combustion (CLC) is a high-quality candidate which has a good potential to become an efficient technique for separating  $CO_2$ . It is easy to produce clean energy from fossil fuel by using this novel method which separates  $CO_2$  inherently (Villa et al., 2003; Hossain and de Lasa, 2008; Zhang et al., 2009). In chemical looping combustion, a gaseous fuel like natural gas or synthesis gas is burnt with oxygen carrier which is usually a metal oxide and used to transfer oxygen from the combustion air to the fuel. Therefore fuel and combustion air never mixed and combustion products ( $CO_2$ ,  $H_2O$ ) do not become diluted with  $N_2$ ; thus pure  $CO_2$  is obtained after condensation of water. The process consists of two separate reactors (a fuel and an air reactor) and solid oxygen carrier transports oxygen between them, see Fig. 1 (Anheden and Svedberg, 1998; Ishida et al., 1987

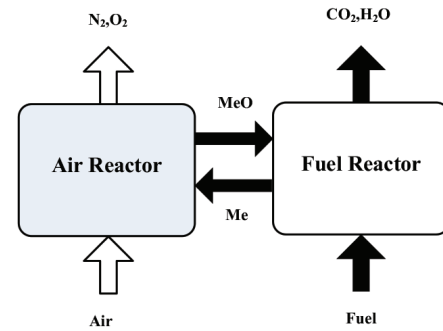
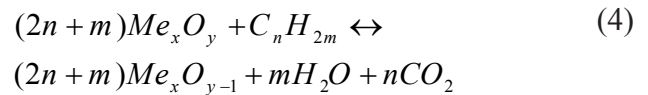
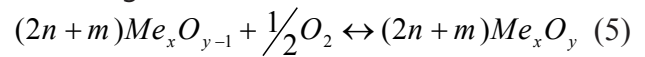


Figure 1. A schematic view of chemical looping combustion

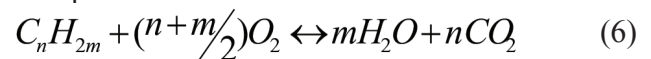
$CO_2$  and  $H_2O$  exit from the fuel reactor while the outlet gas from the air reactor consists of  $N_2$  and unused  $O_2$ . The oxygen carrier is reduced with fuel which occurs in the fuel reactor according to:



Where  $Me_xO_y$  is the common abbreviation for metal oxide in chemical looping combustion and  $Me_xO_{y-1}$  stands for the reduced oxide which is then circulated to the air reactor and oxidized according to:



Reaction (5) is always exothermic while the amount of energy released or required in reaction (4) depended on choice of oxygen carrier. The net energy in the reactor is equal to the amount of heat released from normal combustion. That is apparent because summation of reaction (4) and (5) yields reaction (6); the conventional complete combustion of fuel.



The pressure and the temperature of air and fuel reactors are  $10^5$ Pa and  $800-1200^\circ C$ , respectively. Up to now, researchers suggest various designs for chemical looping combustion including: moving bed, fluidized bed, packed bed and dense membrane reactor but circulating fluidized beds is common to use (Nalbandian et al., 2011; Fan et al., 2008; Son and Kim, 2006; Noorman et al., 2007). Richter and Knoche, (1983) proposed the principle of CLC process to increase the thermal efficiency in fossil fuel fired power plants for the first

time. Some years later, Lyngfelt and Leckner (1999), successfully ran a 10kW CLC prototype at Chalmers University of Technology. Choice of oxygen carriers is one of the critical steps in CLC and numbers of studies have been done on this course. An appropriate oxygen carrier has the following properties:

- 1) Have suitable rate of reaction both reduction and oxidation.
- 2) Be thermodynamically suitable to convert fully the fuel to  $\text{CO}_2$  and  $\text{H}_2\text{O}$ .
- 3) Have good strength so that resistance to attrition and breakage.
- 4) Does not tend to agglomeration.
- 5) Be cheap and healthy.
- 6) Have high melting point.
- 7) Have low tendency to become deactivate with carbon and sulphur.

Based on above properties, some metal oxides like Ni, Cu, Fe, Mn could be probable oxygen carriers (Cho, 2005; Johansson, 2007; Adanez et al., 2004). In this work, Cu is used as solid particle. An advantage of chemical looping combustion compared with ordinary combustion would be the inherent capture of  $\text{CO}_2$  from the rest of the flue gas without spending energy. (Ryden et al., 2009). Also, since indirect combustion in chemical looping combustion does not involve high temperature flame, the formation of  $\text{NO}_x$  is avoided.

### 1.5 Literature review

Many efforts have been done for improvement of steam methane reforming. Arab Aboosadi et al. (2011a) have considered a novel integrated thermally coupled configuration for methane steam reforming. In their simulated reactor, hydrogenation of nitrobenzene to aniline in the exothermic side is used as a heat source for endothermic reaction of steam methane reforming. The exothermic reaction takes place in the shell side and endothermic reaction occurs in the tube side. Moreover, Arab Aboosadi et al. (2011b) simulated and optimized tri-reformer (TRM) reactor for producing synthesis gas using differential evolution (DE) method. In TRM process, steam reforming,  $\text{CO}_2$  reforming and partial oxidation of methane occurred in a single reactor. Finally, methane steam reforming and hydrogenation of nitrobenzene in hydrogen

perm-selective membrane thermally coupled reactor has been optimized using differential evolution (DE) method by Rahimpour et al. (2012). Recently, Rahimpour et al. (2013) have simulated methane steam reforming technology coupled with fluidized bed chemical looping combustion using Fe- based as oxygen carrier. From previous studies, it is found that there is no modeling information available in the literature about using chemical looping combustion as a heat source for steam methane reforming in order to increase  $\text{H}_2$  production. Therefore, it was decided to first study on this system.

### 1.6. Objectives

The main goal of this study is enhancement of hydrogen production theoretically in a thermally coupled steam reformer (TCSR). The endothermic and exothermic reactions are chosen the steam methane reforming and indirect combustion of methane in a chemical looping combustion process, respectively. The motivation is to combine the energy efficient concept of coupling exothermic and endothermic reactions and enhancement of hydrogen production. A steady state 1-D heterogeneous model of the thermally coupled multitubular reactor is used to estimate the performance of the proposed reactor. Ultimately, the simulation results of the TCSR were compared with the ones in conventional steam methane reforming.

## 2. Process description

### 2.1. Conventional steam reformer

Fig. 2 represents the schematic diagram of a conventional Lurgi-steam methane reformer (CR) to produce syngas for methanol synthesis process. This reactor has vertical tubes which are located inside a huge fired furnace. Natural gas is mixed with steam and entered to the steam reformer tubes as feed. Vertical tubes is packed with Ni- based catalyst, the generated heat related to natural gas combustion in burners of furnace transfers to reformer tubes (Methanol documents of Lurgi in Assaluyeh-Iran.). Table 1 shows the specification of reactor and operating conditions of the CR

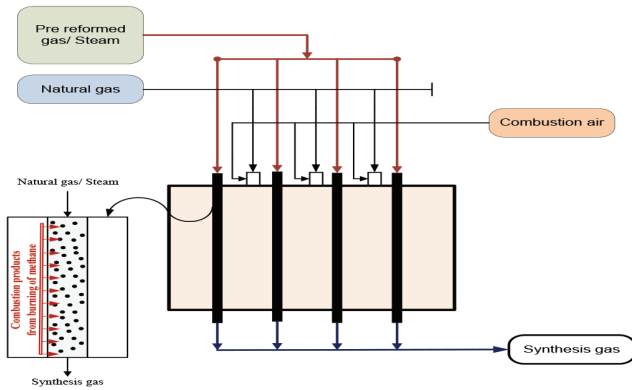


Figure 2. A schematic diagram of conventional steam methane reforming

Table 1: The specification of reactor and operating conditions

Parameter	Value	Unit
Feed composition		mol%
CO <sub>2</sub>	1.72	[-]
CO	0.02	[-]
H <sub>2</sub>	5.89	[-]
CH <sub>4</sub>	32.59	[-]
N <sub>2</sub>	1.52	[-]
H <sub>2</sub> O	58.26	[-]
Inlet temperature	793	°K
Inlet pressure	40	bar a
Total feed gas flow	9129.6	Kmol/h
Number of tubes (in 4 rows)	184	[-]
Inside diameter	125	mm
Heated length	12	m
Catalyst volume filled in (total)	27.8	m <sup>3</sup>
Design pressure	41	bar g
Design temperature	1063	°K
Catalyst properties		
Catalyst shape	HOLE 10 rings	
Particle size	19 × 16	mm
Void fraction	0.4	

Heat load on (tube(100% design case	68,730	Kcal/m <sup>2</sup> h
Reformer duty (100% (design case	248.2	GJ/h
<b>Shell side</b>		
<b>Combustion air</b>		
Temperature	603	°K
Pressure	1	bar
Flow rate	114,313	sm <sup>3</sup> /h
<b>(Feed gas/fuel</b>		
Temperature	307	°K
Pressure	3	bar
Flow rate	29,608	sm <sup>3</sup> /h

## 2.2. Thermally coupled steam reformer (TCSR)

A conceptual schematic diagram of TCSR is shown in Fig .3. Chemical looping combustion of methane is used as exothermic reaction instead of normal combustion of natural gas in conventional steam reformer. This reactor is consisted of three concentric tubes, the inner tube is used as air reactor of CLC and the outer tube is assumed to be fuel reactor. Endothermic reaction of steam methane reforming takes place on the Ni-based catalyst counter currently in the middle tube. Air fed to the air reactor from the bottom of the reactor and natural gas combined with steam is fed to steam reformer from top of the middle reactor. A small part of steam reformer product stream recycled and combined with CH<sub>4</sub> to use as feed for fuel reactor. Cu as an oxygen carrier enters into air reactor of CLC from top of the reactor, moves down ward and after oxidation exits in the form of CuO from bottom of reactor. Then, copper oxide circulates in a loop and after regeneration transfers to fuel reactor and reduced with CH<sub>4</sub>. The specific properties and operational conditions of TCSR are tabulated in Table 2. Table 1 (excluding data in the shell side) is also used for endothermic side of thermally coupled steam reformer.

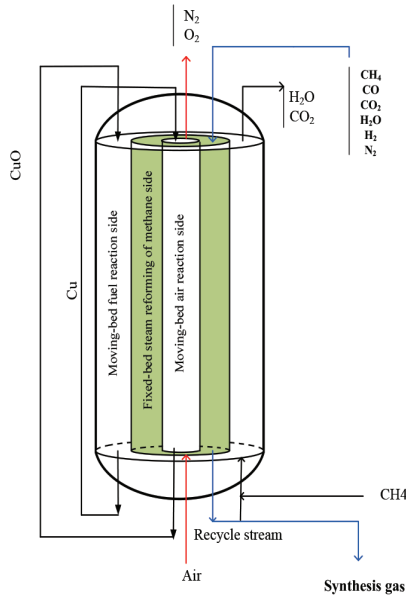


Figure 3. A conceptual schematic diagram of thermally coupled steam reformer

Table 2. The specific properties and operational conditions of TCSR

Parameter	Value
Inlet temperature of endothermic side (*K)	793
Inlet pressure of air reactor (kPa)	101.325
Inlet pressure of fuel reactor (kPa)	101.325
Inner tube or air reactor diameter (m)	0.1
Middle tube or steam reformer diameter (m)	0.1601
Outer tube or fuel reactor diameter (m)	0.1887
Length of the reactor (m)	12
Feed composition	
<b>Air reactor</b> (composition mol %)	
O <sub>2</sub>	20.94
N <sub>2</sub>	78.08
Ar	0.98
<b>Oxygen carrier properties</b> (CuO/Al <sub>2</sub> O <sub>3</sub> )	
Particle size (mm)*	0.1-0.3

Table 2. Continued...

*Porosity	0.57
*(Apparent density (kg m <sup>-3</sup>	1800
*(Molar density of CuO (molm <sup>-3</sup>	80402
*(Molar density of Cu (molm <sup>-3</sup>	140252
(Flow rate(kg/s	0.01
Obtained from (García-Labiano * (et al., 2004	

### 3. Reaction scheme and kinetics

#### 3.1. Steam methane reforming (endothermic side)

Three main reaction reactions occurred in steam reformer are steam reforming of methane and the water-gas shift reaction (Equations 1–3). The reaction rates of CR reactions proposed by Xu and Froment are as follows (Xu and Froment, 1989; Xu et al., 2002):

$$R_1 = \frac{k_1}{P_{H_2}^{2.5}} (P_{CH_4} P_{H_2O} - \frac{P_{H_2}^3 P_O}{K_I}) \times \frac{1}{\phi^2} \quad (7)$$

$$R_2 = \frac{k_2}{P_{H_2}^{3.5}} (P_{CH_4} P_{H_2O}^2 - \frac{P_{H_2}^4 P_{O_2}}{K_I}) \times \frac{1}{\phi^2} \quad (8)$$

$$R_3 = \frac{k_3}{P_{H_2}} (P_O P_{H_2O} - \frac{P_{H_2} P_O}{K_{III}}) \times \frac{1}{\phi^2} \quad (9)$$

$$\phi = 1 + K_O P_O + K_{H_2} P_{H_2} + K_{CH_4} P_{CH_4} + K_{H_2O} \frac{P_{H_2O}}{P_{H_2}} \quad (10)$$

The reaction equilibrium constants and Arrhenius kinetic parameters are listed in Table 3. Table 4 shows the Van't Hoff parameters for species adsorption

#### 3.2. Chemical looping combustion (exothermic sides)

In general, metal oxides are used as oxygen carrier in chemical looping combustion. The focus of the literature is on Ni, Cu, Fe and Mn that each one has specific advantage. In this work, Cu is selected as oxygen carrier because reduction of Cu is exothermic while for most other materials, it is endothermic.

Additionally, it is environmentally benign and reactive (Jerndal et al., 2006). The reduction and

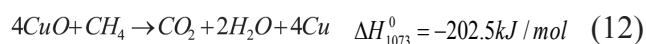
Table 3. The reaction equilibrium constants and Arrhenius kinetic parameters for steam reforming reactions

Reaction, j	Equilibrium constant	$K_{0j}$ $\left(\frac{\text{mol}}{\text{kgcats}}\right)$	$E_j$ $\left(\frac{\text{J}}{\text{mol}}\right)$
1	$K_I = \exp\left(\frac{-26830}{T_s} + 30.114\right) (\text{bar}^{-2})$	$1.17 \times 10^{15} (\text{bar}^{-0.5})$	240100
2	$K_{II} = K_I \cdot K_{III} (\text{bar}^{-2})$	$2.83 \times 10^{14} (\text{bar}^{-0.5})$	243900
3	$K_{III} = \exp\left(\frac{4400}{T_s} - 4.036\right)$	$5.43 \times 10^5 (\text{bar}^{-1})$	67130

Table 4. The Van't Hoff parameters for species adsorption

Components	$K_{0i} (\text{bar}^{-1})$	$\left(\frac{\text{J}}{\text{mol}}\right) \Delta H_i$
CH <sub>4</sub>	$10^{-4} \times 6.65$	-38280
CO	$10^{-5} \times 8.23$	-70650
H <sub>2</sub>	$10^{-9} \times 6.12$	-82900
H <sub>2</sub> O	$10^5 \times 1.77$	88680
$K_{III} = \exp\left(\frac{4400}{T_s} - 4.036\right)$		

oxidation reaction of Cu in fuel and air reactor are as follows:



One of the main drawbacks of this material is its low melting point (1085°C). This makes necessary to perform at temperatures lower than its melting point in order to avoid agglomeration and loss of activity (Noorman et al. 2010). In order

to describe the kinetics of metal oxide reduction and oxidation, many efforts have been done. In this way, two general types of models can be found in the literature for chemical looping combustion: one is nucleation growth model referred to as Avrami Erofeev models and the other is shrinking core models which are common method (Levenspiel, 1998; Koga and Harrison, 1984). The following equations described the unreacted shrinking core model for gas- solid reactions:

$$\frac{t}{\tau_j} = X_{s,j} \quad \frac{dX_{s,j}}{dt} = \frac{1}{\tau_j} \quad \tau_j = \frac{\rho_{m,i} L_i}{b_j k_j C_g^n} \quad (13)$$

Where  $\rho_{m,i}$  is the molar density of reactive material in the oxygen carrier,  $L_i$  is the layer thickness of reacting solid,  $b_j$  is the stoichiometric factor in reaction  $j$  ( $j$ =oxidation, reduction),  $k_j$  is the chemical reaction rate constant in reaction  $j$ . The rate of oxidation and reduction can be calculated by following equation:

$$-r_j = \frac{\rho_{m,i}}{b_j} \frac{dX_{s,j}}{dt} \quad (14)$$

In this study, the kinetics data for the reduction and oxidation of  $\text{CuO}/\text{Al}_2\text{O}_3$  with methane and air were used (García-Labiano et al., 2004). Table 5 shows these kinetics parameters for both oxidation and reduction reactions

Table 5. The kinetics parameters of reactions in exothermic side for Cu based oxygen carrier

Kinetic parameters	Fuel reactor	Air reactor
$n$	0.4	1
$E \left( \frac{\text{kJ}}{\text{mol}} \right)$	$60 \pm 3$	$15 \pm 2$
$K_0 (\text{mol}^{-n} \text{m}^{3n-2} \text{s}^{-1})$	$4.5 \times 10^{-4}$	$4.7 \times 10^{-6}$

## 4. Mathematical model

Fig. 4 shows a schematic diagram for counter-current mode of the heat exchanger reactor configuration. One-dimensional homogenous model including a set of coupled mass and energy balances are taken into consideration. The hypotheses considered in the model of both exothermic and endothermic sides are summarized as follows:

- Steady state condition.
- Gas phase is considered as an ideal.
- Plug flow pattern is assumed.

- Bed porosity in axial and radial direction is constant.
- Heat loss is neglected.
- Axial diffusion of mass and heat are ignored.

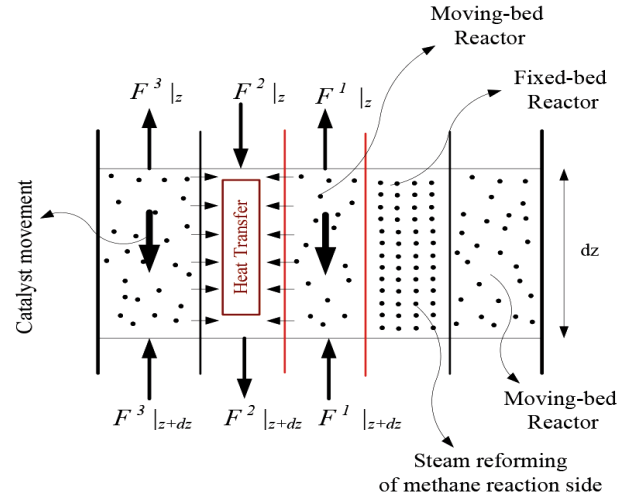


Figure 4. A schematic diagram of counter-current mode for thermally coupled steam reformer (TCSR).

### 4.1. Packed bed steam methane reformer tube (endothermic side)

Based on the above assumptions and considering the element with length  $\Delta z$  along axial direction, the differential equations describing mole and energy transfers are obtained.

The mass and energy balances for fluid and solid phases are listed in table 6.

Where  $T_s$  is temperature of steam reformer side and  $\eta$  is effectiveness factor defined as the ratio of the reaction rate observed on the real rate of reaction.

### 4.2. Moving bed chemical looping combustion (exothermic side)

There are a few investigations in the modeling of chemical looping combustion reactors. Abad et al. (2010) studied modeling of chemical looping combustion of methane using Cu-based oxygen carrier. Modeling of a 120kw chemical looping combustion reactor using Ni based oxygen carrier was introduced by Kolbitch et al. (2009). Kang et al. (2010) investigated the modeling of a counter-current moving bed for fuel and steam reactors in three reactor



Table 6. Mass and energy balance and boundary conditions for solid and fluid phases in packed bed steam methane reformer tube (endothermic side).

Parameter	Value
Mass and energy balance for solid phase	$a_v c k_i^g (y_i^g - y_i^s) + r_i \rho_b = 0$ (15)
	$-\frac{c_p^g}{A} \frac{d(F T^g)}{dz} + a_v h_f (T_s^s - T_s^g) - \frac{\pi D_i}{A} U_f (T_s - T_f) - \frac{\pi D_i}{A} U_a (T_s - T_a) = 0$ (16)
	$a_v h_f (T_s^g - T_s^s) + \rho_b \sum_{i=1}^N r_i (\Delta H_{fi}) = 0$ (17)
Mass and energy balance for fluid phase	$-\frac{1}{A_a} \frac{d(F_i y_i^g)}{dz} + a_v c k_i^g (y_i^s - y_i^g) = 0$ (18)
Boundary condition	$P_j^g = P_{j0}^g, y_{i,j}^g = y_{i0,j}^g, T_j^g = T_{j0}^g, z = 0,$ (19)

chemical looping (TRCL). Their results show that the heat transfer from oxygen carrier to gas phase is fast so that the temperature of gas and solid phases become equal along the reactor. Kang et al., 2012 suggested that a counter-current moving bed is expected to obtain high CO<sub>2</sub> selectivity in comparison with fluidized bed reactor. To obtain mass and energy balances in fuel and air reactor, an element with length  $\Delta z$  was considered. Table 7 shows the equations of mass and energy balances for fluid and solid phases for both air and fuel reactor.

Where  $C_p^s$  (J/Kg.K) is the heat capacity of the solids,  $F^s$  (kg/s) is the solid loading.

### 4.3 Auxiliary correlations

In order to complete the mentioned simulation equations, auxiliary correlations containing physical properties of components, mass and heat transfer coefficients should be added, see table 8.

## 5. Numerical solution

A governing equations of this model consists of a set of differential algebraic equations

including mass balances for all sides with given boundary conditions incorporate the reaction rates as well as basic assumptions. In order to solve the set of equations, back-ward finite difference approximation was used. The reactor length is then divided into 100 separate sections and the Gauss-Newton method in MATLAB programming environment was used to solve the non-linear algebraic equations in each section. All parameters of chemical looping combustion reactors like inlet fuel reactor compositions and flow rates are determined by using trial and error method.

## 6. Results and Discussion

Model validation was carried out by comparing the simulated results of steam reforming side of TCSR with the observed experimental data of the industrial tubular fixed bed steam reformer reactor which is shown in Table 9. According to this table, this simulation results is in good agreement with experimental data. The simulation results of the reactor in the exothermic side are not compared with any reference case .

Table 7. Mass and energy balance and boundary conditions for solid and fluid phases in moving bed chemical looping combustion

Parameter	Value
Mass and energy balance for solid phase	$-\frac{1}{A} \frac{d(F^s y_j^s)}{dz} + \rho_b r_j b = 0$ (20)
	$-\frac{cp^s}{A} \frac{d(F_i^s T_j^s)}{dz} + a_v h_f (T_j^s - T_j^g) + \rho \sum br_j (-\Delta H_{Rxn}) = 0$ (21)
Mass and energy balance for fluid phase	$\frac{1}{A} \frac{d(F^g y_j^g)}{dz} + \rho_b r_j = 0$ (22)
	$\frac{cp^g}{A} \frac{d(F_i^g T_j^g)}{dz} + a_v h_f (T_j^s - T_j^g) + \frac{\pi D_i}{A} U (T_s^g - T_j^g) = 0$ (23)
Boundary condition	$z = 0; y_i^g = y_{iout}^g, y_{i0}^s = 0, T^g = T_{iout}^g$ (24)

Table 8. Auxiliary correlations.

Parameter	Equation	Number
component heat capacity	$C_p = a + bT + cT^2 + dT^3$	(25)
mixture heat capacity	$C_{p,mix} = \sum_{i=1}^n y_i \times C_{p,i}$	(26)
viscosity	$\mu = \frac{C_1 T^{C_2}}{1 + \frac{C_3}{T} + \frac{C_4}{T^2}}$	(27)
overall heat transfer coefficient	$\frac{1}{U} = \frac{1}{h_i} + \frac{A_i \ln(D_o/D_i)}{2\pi K_w} + \frac{A_i}{A_o} \frac{1}{h_o}$	(28)
heat transfer coefficient between gas phase and reactor wall	$\frac{h}{C_p \rho \mu} \left( \frac{C_p \mu}{K} \right)^{2/3} = \frac{0.458 \left( \frac{\rho u d_p}{\mu} \right)^{-0.407}}{\epsilon_B}$	(29)
Pressure drop	$\frac{dP}{dz} = 150 \frac{(1-\epsilon)^2 \mu u_g}{\epsilon^3 d_p^2} + 1.75 \frac{(1-\epsilon) u_g^2 \rho}{\epsilon^3 d_p}$	(30)

Table 9. Comparison between model prediction and plant data.

Parameter	Plant	CR
Temperature (°K)	710	720.5
Composition (mol %)	Plant	CR
CO <sub>2</sub>	5.71	5.70
CO	3.15	3.18
H <sub>2</sub>	31.39	31.45
CH <sub>4</sub>	20.41	20.37
N <sub>2</sub>	1.29	1.30
H <sub>2</sub> O	38.05	38.01
Methane conversion (%)	26.5	26.6

In this section, predicted mole fraction, pressure drop and temperature profiles in the counter current coupled reactor are presented. The results are shown in the following figures. One definition is introduced to examine the methane conversion through the reactor length:

$$\text{Methane conversion} = 100 \times \frac{F_{\text{CH}_4, \text{in}} - F_{\text{CH}_4, \text{out}}}{F_{\text{CH}_4, \text{in}}} \quad (31)$$

### 6.1.1. Thermal behavior

Fig. 5(a) shows the axial temperature profiles in conventional steam reformer reactor (CR) as well as the endothermic side of thermally coupled steam reformer (TCSR). As seen, the temperature profile of CR is linear while it has a curvy profile in the TCSR. Although the initial temperature of both configurations is the same, the temperature profile of TCSR is higher than that of CR as a result of distribution of solid oxygen carriers in the exothermic side which improves overall heat transfer coefficient. Since the rates of endothermic reactions are increased as temperature increases, hydrogen production is improved in TCSR. In order to make a driving force for heat transfer from exothermic side

to endothermic side, the temperature of endothermic side must always lower than that of exothermic side. Fig. 5(b) represents the temperature profile of exothermic side of fuel reactor. In general, the highest temperature in thermally coupled reactors is related to exothermic side where heat generation is occurring. Part of generated heat from air and fuel reactors is used for driving endothermic reaction and the rest is used to heat the reaction mixtures in three sides of TCSR. Although steam reforming is an endothermic process, its temperature increased along the reactor; it happens because the generated heat in exothermic side is so higher than consumed heat in endothermic side. The most heat transfer is occurring at the beginning of the reactor because of high temperature difference between endothermic side and exothermic sides. It is understood from Fig. 5(b) that the variation of temperature of fuel reactor is 70 K and the outlet temperature is 1160 K; fortunately, this temperature is lower than melting point of copper so that no agglomeration and loss of activity occur in the chemical looping combustion.

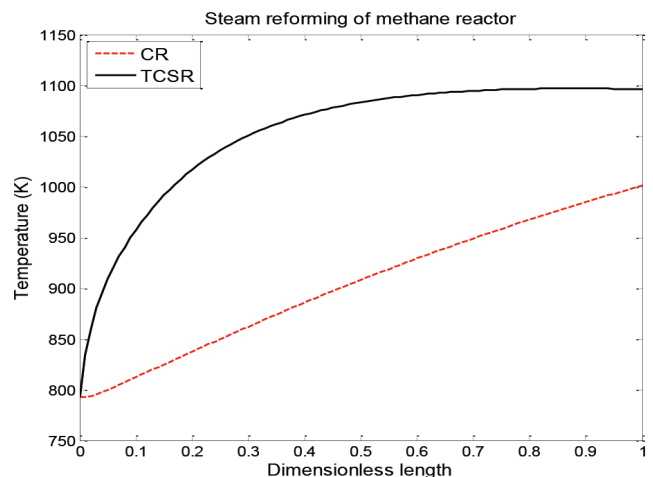


Figure 5(a). Variation of temperature for endothermic side of TCSR and CR.

### 6.1.2. Molar behavior

The total molar flow rate of endothermic side in CR and TCSR are compared in Fig. 6. The total molar flow rate of TCSR is higher than that of CR. The temperature profile of endothermic

side and total molar flow rate have the same trend, because these two parameters are proportional to each other (see fig 5(a)). As total molar flow rate increases, the residence time and conversion decreases.

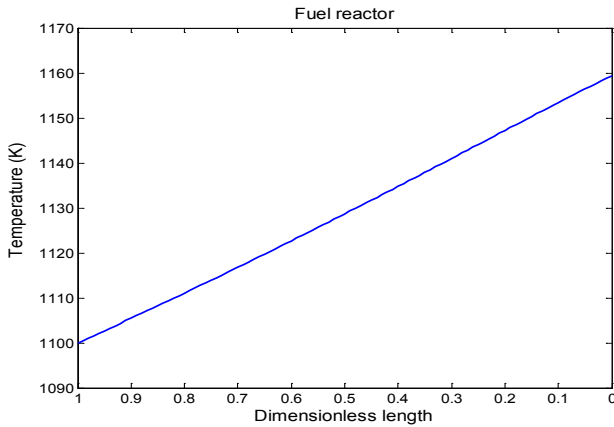


Figure 5(b). Variation of temperature for exothermic side of fuel reactor

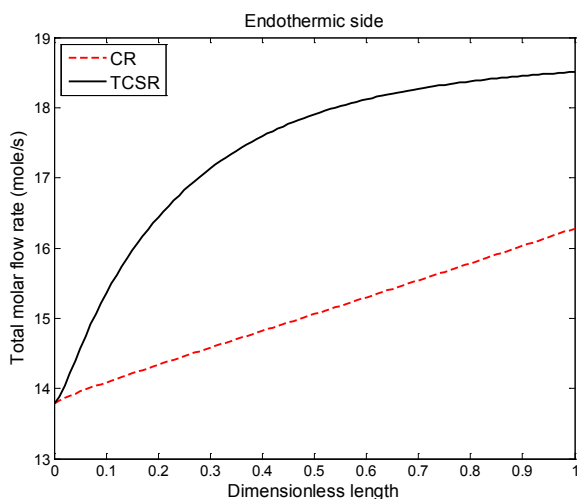


Figure 6. Comparison of total molar flow rate between endothermic sides of TCSR and CR.

Fig.7 (a)-(d) presents the comparison of mole fraction of components in endothermic side of TCSR and CR. Fig.7 (a) and (b) illustrate the mole fraction of hydrogen and carbon monoxide (CO) along the reactor at steady state. Hydrogen and CO are the most desirable products of steam reforming and fortunately their mole fractions increase in the coupled configuration in comparison with CR, because thermal effect of coupled reactor provides a good condition for heat transfer and consequently more

production. The  $\text{CH}_4$  mole fraction is depicted in Fig. 7 (c). As can be seen, the consumption rate of  $\text{CH}_4$  as a main reactant in steam methane reforming process increases in coupled configuration. According to this figure,  $\text{CH}_4$  conversion reaches to 52.6 % in TCSR while 26% methane conversion occurs in CR. The difference between  $\text{CH}_4$  mole fraction profiles in CR and TCSR is owing to the temperature increase in endothermic side which causes the increase in reaction rate and  $\text{CH}_4$  consumption.  $\text{CO}_2$  mole fraction as an undesired product in TCSR is compared with the one in CR in Fig.7 (d). At the reactor entrance, the  $\text{CO}_2$  mole fraction in TCSR is higher than the one in the CR configuration but at the end of reactor it becomes lower.

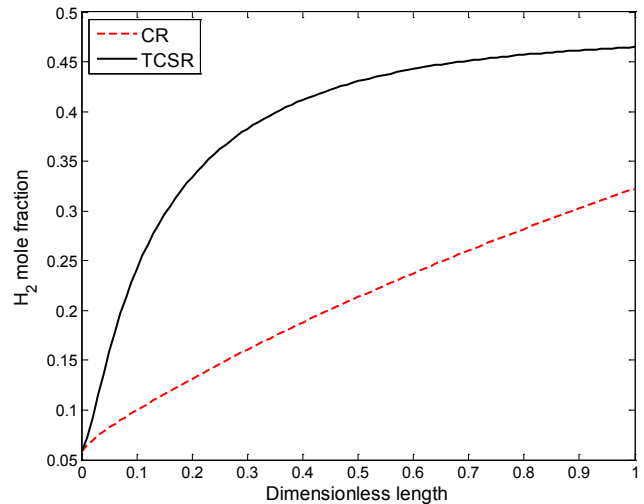


Figure 7(a). Comparison of  $\text{H}_2$  mole fraction along the reactor axis between endothermic side of TCSR and CR.

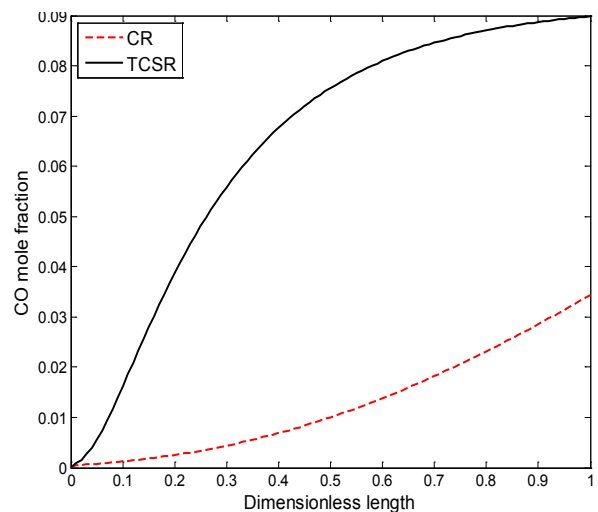


Figure 7(b). Comparison of CO mole fraction along the reactor axis between endothermic side of TCSR and CR.

Fig. 8 (a)-(c) demonstrates the mole fractions of components in exothermic side of fuel reactor. As the reaction scheme in fuel reactor shows, the mole fraction of  $\text{CH}_4$ , as reactant, decreases linearly along the fuel reactor; it is shown in Fig. 8 (a).  $\text{H}_2\text{O}$  and  $\text{CO}_2$  are the main products of reduction reaction in fuel reactor and their mole fraction increases (see Fig 8(b) and (c)) and it can be said that all component behaviors are normal

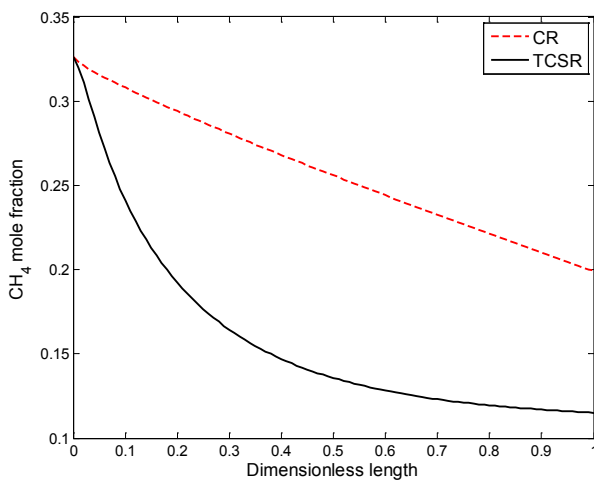


Figure 7(c). Comparison of  $\text{CH}_4$  mole fraction along the reactor axis between endothermic side of TCSR and CR.

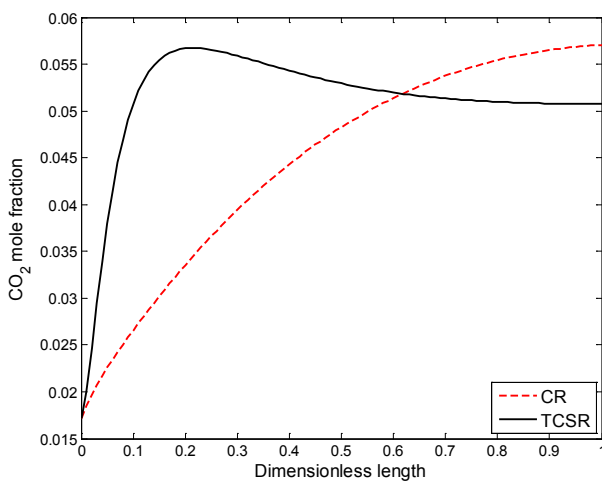


Figure 7(d). Comparison of  $\text{CO}_2$  mole fraction along the reactor axis between endothermic side of TCSR and CR.

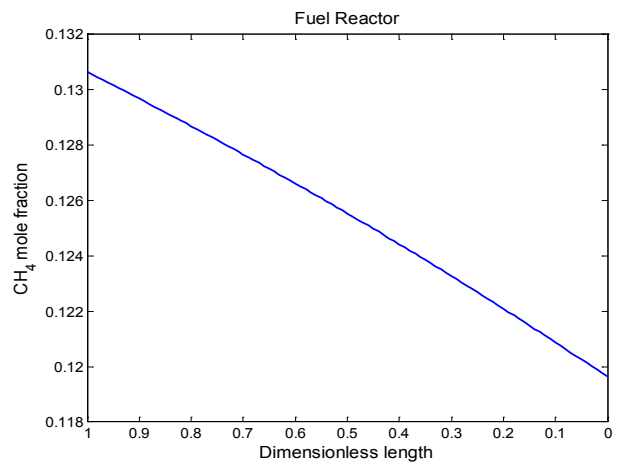


Figure 8(a). Profile of  $\text{CH}_4$  mole fraction along the reactor axis in the fuel reactor of exothermic side in TCSR

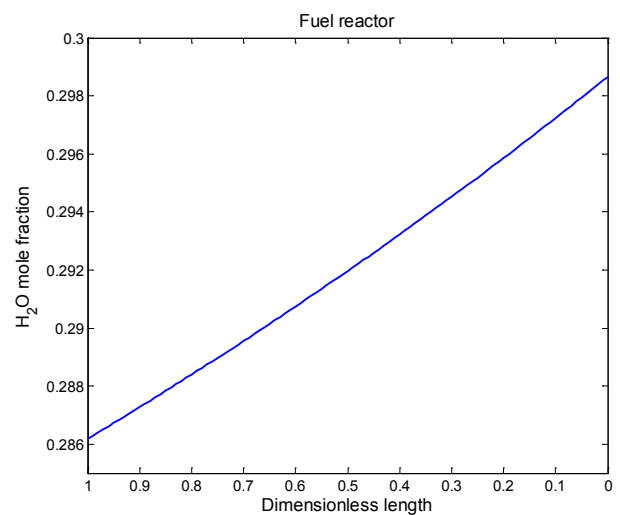


Figure 8(b). Profile of  $\text{H}_2\text{O}$  mole fraction along the reactor axis in the fuel reactor of exothermic side in TCSR

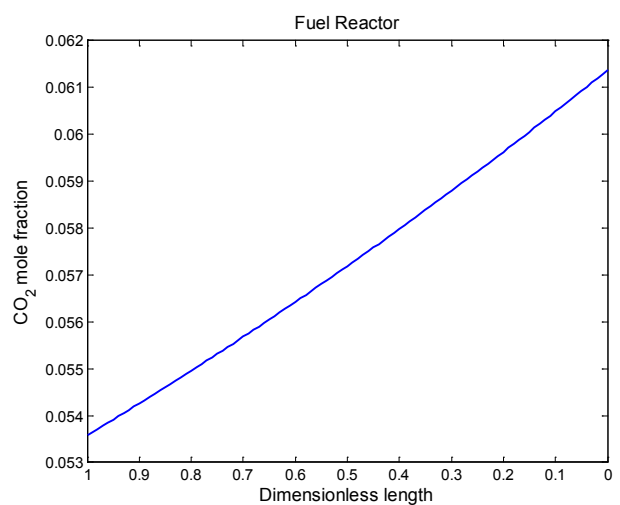


Figure 8(c). Profiles of  $\text{CO}_2$  mole fraction along the reactor axis in the fuel reactor of exothermic side in TCSR

### 6.1.3. Pressure drop

Fig. 9 shows the pressure along steam methane reforming for both conventional and thermally coupled steam reformer. The Ergun Equation (which is equation (23)) usually used to calculate pressure drop through catalytic packed bed.

Since total molar flow rate and temperature profile of thermally coupled steam reformer is higher than temperature profile of conventional steam reformer, the density of gas phase and consequently the velocity and related viscose losses of TCSR become higher. As a result, pressure drop profile through this reactor is higher than conventional steam reformer.

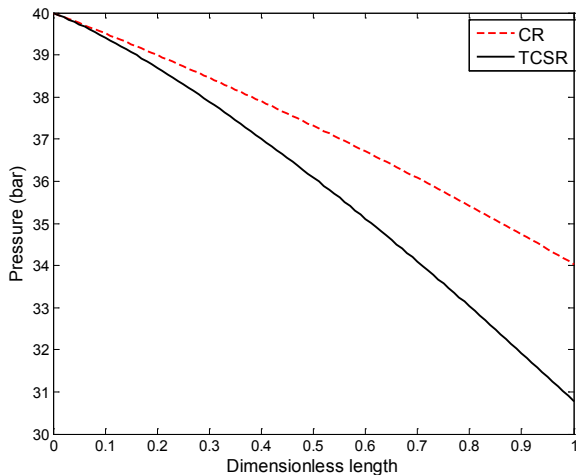


Figure 9. Pressure drop along the endothermic side CR and TCSR.

### 6.1.4. Influence of Inlet pressure of Methane Steam Reforming

In this section, the reactor performance is investigated for various inlet pressure of the endothermic stream. As it can be seen in Fig 10(a) and (b), by increasing the inlet pressure, the methane conversion and consequently hydrogen mole fraction in the methane steam reforming side decrease due to equilibrium related to steam reforming reactions. Fig 10 (c) shows how total molar flow rate changes with increasing inlet pressure of endothermic side. As seen, total molar flow rate decreases from 19.56 to 17.9  $\frac{mole}{s}$  because of their inverse relationship. Fig 10 (d) illustrates that the pressure drops along the reactor axis increases when the inlet

pressure of endothermic stream increases from 30 to 50 bars.

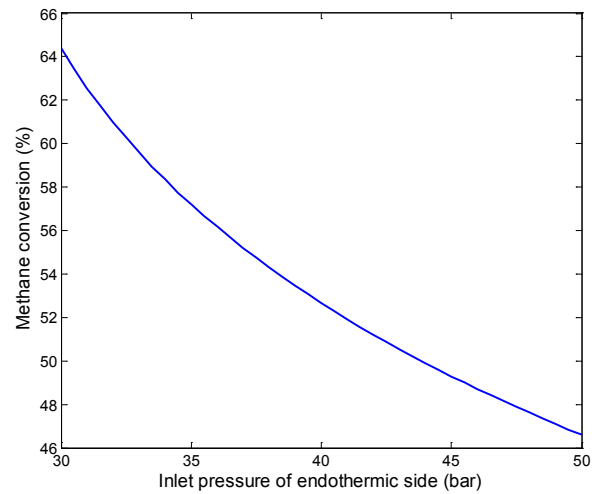


Figure 10(a). Influence of inlet pressure of endothermic stream on methane conversion along the reactor length.

## 7. Conclusion

Coupling endothermic reaction with an appropriate exothermic reaction improves the thermal efficiency of processes. In this study, a novel thermally coupled steam reformer has been proposed for hydrogen production by using chemical looping combustion as a heat source. Chemical looping combustion is carried out in exothermic side which consists of two moving bed reactor named as air and fuel reactors and supply the necessary heat for endothermic side. Solid metal oxide transfers oxygen between these two reactors. Copper oxide is used in this work because both reduction and oxidation of copper are exothermic.

Steam methane reforming takes place in endothermic side for hydrogen production. Thermally coupled steam reformer consists of three concentric tubes; the inner, middle and outer sides are used for air reactor, steam reformer reactor and fuel reactor, respectively. One dimensional heterogeneous model is used to simulate TCSR and comparison with conventional steam reformer. Thermal and molar behaviors of all sides as well as pressure drop profiles were investigated along the reactors. The results show that an increment

about 47% occurs in hydrogen mole fraction in TCSR compared with conventional steam reformer. In addition, the methane conversion in steam reforming reaches to 52.6%. Finally, these preliminary simulation results demonstrate that substitution of huge fired-furnace of conventional steam reformer with chemical looping combustion process is highly efficient for synthesis gas production.

## Nomenclature

$A_a$	Cross section area of each tube ( $m^2$ )
$av$	specific surface area of catalyst pellet ( $m^2/m^3$ )
$b$	Stoichiometric factor for reaction in CLC
$C^g$	Gas concentration ( $mol\ m^{-3}$ )
$cp^g$	Specific heat of the gas at constant pressure ( $J\ mol^{-1}$ )
$cp^s$	Heat capacity of oxygen carrier in CLC ( $J\ kg^{-1}\ K^{-1}$ )
$Di$	Inside diameter of steam reformer (m)
$F_t$	Total flow rate per each reaction side ( $mol\ s^{-1}$ )
$K_1$	Reaction rate constant for 1 <sup>st</sup> rate equation of steam reforming ( $mol\ kg^{-1}\ s^{-1}$ )
$K_2$	Reaction rate constant for 2 <sup>nd</sup> rate equation of steam reforming ( $mol\ kg^{-1}\ s^{-1}$ )
$K_3$	Reaction rate constant for 3 <sup>rd</sup> rate equation of steam reforming ( $mol\ kg^{-1}\ s^{-1}$ )
$K_j$	Chemical reaction constant of $j$ reaction ( $j$ =oxidation, reduction) ( $mol^{1-n}\ m^{3n-2}\ s^{-1}$ )
$L_i$	Layer thickness of the reacting solid (m)
$N$	Number of components
$n$	Reaction order
$R_1$	First rate of reaction for steam reforming of $CH_4$ (reaction 4) ( $mol\ kg^{-1}\ s^{-1}$ )
$R_2$	Second rate of reaction for steam reforming of $CH_4$ (reaction 5) ( $mol\ kg^{-1}\ s^{-1}$ )
$R_3$	Water gas shift reactor (reaction 6) ( $mol\ kg^{-1}\ s^{-1}$ )
$r_j$	Rate of reaction occurs in chemical looping combustion based on gas phase ( $mol\ m^{-3}\ s^{-1}$ )

$t$	Reaction time (s)
$T_s$	Bulk gas phase temperature related to steam reformer (k)
$T_a$	Temperature of air reactor (k)
$T_f$	Temperature of fuel reactor (k)
$U_f$	Overall heat transfer coefficient between fuel and steam reforming reactors ( $W\ m^{-2}\ K^{-1}$ )
$U_a$	Overall heat transfer coefficient between air and steam reforming reactors ( $W\ m^{-2}\ K^{-1}$ )
$X_s$	Solid conversion in the chemical looping reactor

### Subscripts

$0$	Inlet condition
$a$	Air reactor
$f$	Fuel reactor
$g$	Gas phase
$i$	Chemical species
$j$	Oxidation, reduction
$s$	Solid phase

### Greek letters

$\Delta H_{fi}$	Enthalpy of formation of component ( $J\ mol^{-1}$ )
$\Delta H_{rxn}$	Enthalpy of reaction ( $J\ mol^{-1}$ )
$p_b$	Density of catalyst bed ( $kg\ m^{-3}$ )
$\rho_{mi}$	Molar density of reactive material in the oxygen carrier ( $mol\ m^{-3}$ )
$\eta$	Effectiveness factor used for the intra particle transport limitation

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## جایگزینی چرخه شیمیایی احتراق به جای کوره در فرآیند تبدیل بخار با استفاده از کاتالیست مس

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### چکیده

این مقاله، به بررسی مدل سازی راکتور کوبلینگ حرارتی دو واکنش کاتالیستی ریفرمینگ متان با بخار آب و چرخه شیمیایی احتراق جهت بهبود میزان تولید هیدروژن می پردازد. کوبلینگ حرارتی دو واکنش گرماگیر و گرماده باعث بهبود بازده حرارتی و در نتیجه افزایش میزان تولید میشود. ریفرمینگ متان با بخار آب فرآیندی گرماگیر است که گرمای آن توسط یک کوره فراهم می شود. در این حالت، لوله های ریفرم تحت تنش حرارتی بالایی قرار دارند. با جایگزینی این کوره با چرخه شیمیایی احتراق برانیم تا علاوه بر حل این مشکل تولید هیدروژن را در یک فرآیند کوبلینگ افزایش دهیم. چرخه شیمیایی احتراق نوعی احتراق غیر مستقیم است که از دو راکتور هوا و سوخت تشکیل شده است. در این فرآیند از تماس مستقیم سوخت با اکسیژن جلوگیری می شود و شرایط برای واکنش سوخت با یک اکسید فلز فراهم می شود. کوبل این دو واکنش کاتالیستی در یک راکتور سه لوله هم مرکز انجام می شود. راکتور درونی و بیرونی به ترتیب به عنوان راکتورهای هوا و سوخت چرخه شیمیایی احتراق و راکتور میانی راکتور ریفرمینگ متان منظور می شود. راکتور چرخه شیمیایی احتراق بستر متحرک است که مس در آن کاتالیست متحرک است. راکتور ریفرمینگ متان بستر ثابت است که از کاتالیست نیکل پر شده است. شبیه سازی فرآیند با استفاده از یک مدل یک بعدی هتروژن انجام شد. صحت مدل انجام شده با داده های راکتور ریفرمینگ متان پالایشگاه گاز زاگرس عسلویه بررسی شد. نتایج حاصل با موارد مشابه در راکتور معمولی ریفرمینگ متان مقایسه شد که افزایش چشمگیر میزان هیدروژن مشاهده شد.

واژگان کلیدی: تولید هیدروژن، ریفرمینگ متان با بخار آب، چرخه شیمیایی احتراق، کاتالیست نیکل و مس