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# A Novel Technique based on Coal Gasification Integrated with Chemical Looping Air Separation

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#### Abstract

To reduce the oxygen production cost in the coal gasification process and to utilize the CO<sub>2</sub>, this study presents a novel technique based on coal gasification integrated with chemical looping air separation (CLAS) by thermodynamic methods. CLAS offers an advantage over the other mature technologies in that it can significantly reduce its capital cost with about less than 40-60% operating costs of conventional oxygen production technologies, while recycling CO<sub>2</sub> separated from gasification gas by CCS could not only promote oxygen production (oxygen uncoupling) of CLAS, but also enhance the coal gasification process by boudouard reaction. The thermodynamic performances, as the conversion of metal oxides ( $Mn_2O_3/Mn_3O_4$ ) during oxidation and reduction process, gasification gaseous compositions and cold gas efficiency ( $\eta$ ) were the main subject of focus. The effects of three key variables, as air flow rate, reduction temperature, and recycling CO<sub>2</sub> ratio ( $\beta$ ) were investigated.

Highlights: 1. The chemical looping air separation is providing oxygen and steam as gasification agents to coal gasification process.

2. CO<sub>2</sub> is getting full used by acting as inert component and also as coal gasification agent.

Keywords: Coal gasification; Chemical looping air separation; Simulation

#### Introduction

Coal, as an inexpensive and easily obtained fossil energy, plays a significant role in the world's energy portfolio. The present coal combustion technique is widely adopted for generating electricity in China [1], whereas, this utilization of coal resource brings some environmental problems, such as acid rain, ozone depletion, and the greenhouse effect [2]. Therefore, several advanced pathways for the clean and effective utilization of coal have been presented including: Integrated Gasification Combined Cycle (IGCC) with CO<sub>2</sub> capture and sequestration (CCS) [3-5] and Pressurized Fluidized Bed Combustion (PFBC) [6,7]. Between these, coal gasification is posing as an attractive option for coal utilization in that it can convert coal into syngas, and following generate electricity or synthesize other chemical products, like methanol, synthetic natural gas and petrol etc [8].

In the coal gasification process, oxygen is sent to gasifier as a gasification agent, which in turn, adds high oxygen production cost by using air separation technologies. The present existing air separation technologies mainly include: cryogenic air separation unit (CASU), advanced ion-transport membrane (ITM), and nano-structured molecular sieves (NMS) system which have been described elsewhere [9-12]. Especially, CASU is the most widely used technique for oxygen production with highly energy intensive, which leads to arising approximate 3-4% energy penalty in the oxy-fuel operations [13], resulting in revenue losses and the increasing of coal utilization per unit of electricity generated. Additionally, it should be noted that though the ITM and NMS system could reduce around 10-25% of the capital costs in comparison to that of CASU, these systems still remain expensive, causing the fabrication, installation, maintenance and integration issues [14-16].

As a promising and novel alternative air separation approach, chemical looping air separation (CLAS) offers an advantage over the other mature technologies in that it can significantly reduce its capital cost with about less than 40-60% operating costs of conventional oxygen production technologies [13,17,18]. In 2010, CLAS was firstly

presented by Moghtaderi at the University of Newcastle, Australia [19]. Moghtaderi and his group pointed out that the specific power for the CLAS (approximately 0.045kWh/ m<sup>3</sup> of oxygen) is much lower than conventional CASU system (approximately 0.4 kWh/ m<sup>3</sup> of oxygen) [13,20-22] In another related work in 2013, Shah et al. built up several process options for integration chemical looping air separation (ICLAS) to extend the application of CLAS by integration of oxy-fueltype power plants, aiming at lowering the operating costs of oxy-fuel operations [13].

The schematic of the CLAS concept is illustrated in Figure 1. CLAS concept is able to separate oxygen in air through the cyclic oxidation/ reduction reaction of metal oxides between two interconnected reactors, namely oxidation reactor (OR) and reduction reactor (RR). In RR, the metal oxides are reduced, where oxygen decoupling occurs in the presence of steam or  $CO_2$ , which is/are used to control the partial pressure of oxygen in RR. The reduction reaction (oxygen uncoupling) is shown as below:

$$\operatorname{Me}_{x}\operatorname{O}_{v} \to \operatorname{Me}_{x}\operatorname{O}_{v-2} + \operatorname{O}_{2} \tag{1}$$

The reduced metal oxides are transported back to oxidation reactor (OR), and fresh air is fed to OR, so that the oxides carriers can be regenerated to a higher oxidation state. The oxidation reaction (oxygen coupling) is:

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$$Me_x O_{v-2} + O_2 \to Me_x O_v$$
<sup>(2)</sup>

In theory, the thermal energy required in the CLAS process from R1 and that released from R2 will be the same. Due to the lower energy demands, the CLAS process is quite efficient.

On the basis of the CLAS concept and considering the product gas (i.e.  $O_2$ +Steam /CO<sub>2</sub>) of CLAS can be the gasification agents for coal gasification process, the coal gasification integrated with chemical looping air separation process is firstly proposed in this article. It should be noted that the CO<sub>2</sub> in gasification gas should be separated by CCS technique, and recycles back to RR, controlling the partial pressure of oxygen in RR and acting as a gasification agent to convert CO<sub>2</sub> into CO.

In this paper, the authors focus on the advantages of this integrated system. The CLAS process product gas could be the coal gasification agents and the  $CO_2$  separated from gasification gas could be an inert gas to disturb the equilibrium partial pressure of oxygen in RR. This paper firstly illustrated this novel process, subsequently, the Aspen Plus software was conducted to develop this novel process based on thermodynamic method, then some key variables that influence this integrated system have been analyzed to investigate the optimal operational conditions.

# **Process Configuration and Simulation**

## Thermodynamic analysis of CLAS reactions

In the previous research, four metal oxides (i.e.  $Mn_2O_3/Mn_3O_4$ ,  $MnO_3/Mn_3O_3$ , CuO/ Cu<sub>2</sub>O and CoO/Co<sub>3</sub>O<sub>4</sub>) were found to be the

J Fundam Renewable Energy Appl ISSN: 2090-4541 JFRA, an open access journal most suitable options for CLAS process [21]. In this work, we choose  $Mn_2O_3/Mn_3O_4$  as metal oxides due to its lower operating temperature (627-900°C) and faster reaction kinetics [21].

The reaction that occurs in Reduction Reactor (RR) is:

 $6Mn_2O_3 \rightarrow 4Mn_3O_4 + O_2 \quad \Delta H \ge 0$  oxygen uncoupling (3)

Correspondent reaction that occurs in Oxidation Rector (OR) is:

$$4Mn_{3}O_{4}+O_{2} \rightarrow 6Mn_{2}O_{3} \quad \Delta H < 0 \text{ oxygen coupling}$$
(4)

Steam /CO<sub>2</sub> is introduced to RR to reduce partial pressure of oxygen in RR, and to further illustrate the thermodynamic reduction properties of  $Mn_2O_3/Mn_3O_4$  oxide carriers, Figure 2 gives a specific example (i.e.  $Mn_2O_3$  molar flow rate is 100kmol/h) of the relationship between molar flow rate of inert components, reduction temperature and oxygen yield rate by using the Gibbs free energy minimization method. The molar ratio between inert components and  $Mn_2O_3$  is defined as

$$\psi = \frac{n(Steam / CO_2)}{n(Mn_2O_3)} \tag{5}$$

Where  $n(Steam / CO_2)$  and  $n(Mn_2O_3)$  are the molar flow rate of inert components and  $Mn_2O_3$  (here  $n(Mn_2O_3n) = 100$  kmol/h), respectively.

As shown in Figure 2, at a specified  $\Psi$ , oxygen production (oxygen uncoupling) increases with an increase in reduction temperature due to endothermic, similarly, due to the reducing of partial pressure of  $O_2$ , the oxygen production increases with  $\Psi$  when the reduction temperature is constant. In another word, the required reduction temperature could be decreased with increasing of Steam /CO<sub>2</sub> molar flow rate at a specified oxygen yield rate. For example, at  $\Psi$ =0.3, lower reduction temperature (about 800°C) is required to produce about 11 kmol/h of oxygen compares to higher temperature is required (about 860°C) to produce same amount of oxygen at  $\Psi$ =0.1. Inspired by performances of this thermodynamic properties, inert components are introduced to reduce the reduction temperature, at the same time; those inert components (Steam and CO<sub>2</sub>) could also be the gasification agents to promote the coal gasification process.

#### **Process description**

The diagram of chemical looping air separation integrated with coal gasification process is shown in Figure 3. This process mainly consists



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of three parts, namely CLAS, coal gasifier and gas cleaning processes, where syngas (steam,  $CO_2$  and  $O_2$ ) generating, coal gasification and acid gas removal can take place at correspondingly suitable operating parameters.

The CLAS process could separate oxygen in air by recirculation of metal oxides (here  $Mn_2O_3/Mn_3O_4$ ) between a set of two interconnected reactors through a loop seal to prevent gas leakage between two reactors, where reduction and oxidation of metal oxides take place. To promote oxygen production and to effectively make full use of  $CO_2$ , steam and recycling  $CO_2$  are introduced together as inert components to reduction reactor (as illustrated in section 2.1.).

Coal is introduced to an entrained-flow gasifier along with syngas from CLAS process to produce gasification gas under certain conditions; these reactions take place in sequence: coal pyrolysis, volatile combustion and char gasification. The reactions that occur during gasification process are as below

Combustion reactions

 $C + O_2 \rightarrow CO_2 - 394 \text{kJ/mol}$  (6)

$$C + 0.5O_2 \rightarrow CO - 111 \text{kJ/mol}$$
<sup>(7)</sup>

 $0.5O_2 + H_2 \leftrightarrow H_2O - 241.8 \text{kJ/mol}$  (8)

Boudouard reaction

$$C + CO_2 \leftrightarrow 2CO + 172 \text{kJ/mol}$$
(9)

Carbon gasification

$$C + H_2O_{(g)} \leftrightarrow CO + H_2 + 131 \text{kJ/mol}$$
 (10)

$$C + 2H_2O_{(g)} \leftrightarrow CO_2 + 2H_2 + 90kJ/mol$$
 (11)

Water-gas shift(WGS)

$$\rm CO+H_2O_{(g)} \leftrightarrow \rm CO_2+H_2-42.4kJ/mol$$
 (12)

Reforming reaction

$$CH_4 + H_2O_{(g)} \leftrightarrow CO + 3H_2 + 206 kJ/mol$$
 (13)

Methanation

$$C + H_2 \leftrightarrow CH_4 - 75 \text{kJ/mol}$$
 (14)

It could notice that the  $CO_2$  gasification agent could enhance gasification process by boudouard reaction (R9) [23,24], converting  $CO_2$  into CO, meanwhile, making full use of  $CO_2$ .

The gasification gas, composed primarily of H<sub>2</sub> and CO, is then sent to cleaning process to remove the acid gas components (such as CO<sub>2</sub>, H<sub>2</sub>S, COS, etc.) by absorption method (for example, selexol). Due to the limit of this research, the heat recovery and integration units are not considered in this article. The CO<sub>2</sub> from cleaning process is then recycled back to reduction reactor, starting a new cyclic life.

## Simulation and assumption

All of the systems were developed and simulated using Aspen Plus 7.2. The initial step is to define the main components which occur during the process. In this work, the components taken into consideration for this novel technique are: C, S,  $Mn_2O_3$ ,  $Mn_3O_4$ , CO,  $CO_2$ ,  $H_2O$ ,  $N_2$ ,  $O_2$ ,  $H_2$ ,  $CH_4$ ,  $H_2S$ , COS,  $SO_2$ , ash and coal. Due to their structural complexity, coal and ash are defined as non-conventional components in Aspen Plus, meanwhile, DCOALIGT and HCOALGEN models are used to calculate the density and enthalpy of non-conventional components, respectively.

The reduction and oxidation reactors are simulated using Aspen Plus model R gibbs. It should be added that at the outlet of those two reactors, a cyclone separator represented by Sep model is used to separate solid from gases. The simulation of cyclic  $Mn_2O_3/Mn_3O_4$  solids is realized by the method of "tear stream" in Aspen Plus to improve convergence of this system.

Coal, due to its structural complexity and composition diversity, cannot be handed directly by Aspen Plus. It's recommended that coal should be broken down into its constituent reactants, which is realized by a yield reactor, where the coal is characterized by the proximate and ultimate analysis and decomposed into its corresponding constituents [25-29], of which the behaviors can further be simulated by Rgibbs model in Aspen Plus. In this work, Shenhua coal [30] is chosen as feedstock, of which the proximate and ultimate analyses are listed in Table 1.

To limit the length of this research, we don't take heat recovery and integration units into account. The gasification gas is cooled to the suitable operating temperature for removal of the acid gas components, followed to separate acid components by Sep model in Aspen Plus.

The PR-BM method uses the Peng Robinson cubic equation of state with Boston-Mathias alpha function for all thermodynamics properties, and is recommend for gas processing application, consequently, chosen as global method.

Based on the process configuration described above, some assumptions have considered, and summarized as follows:

- The heat and pressure losses in all process have been ignored, and heat recovery and integration units are out scope of this paper.
- (2) The simulation is in a steady state, and the residence time is long enough to obtain both chemical and phase equilibria.
- (3) Coal pyrolysis is considered to happen immediately when coal is fed into the gasifier.
- (4) Ash is regarded as inert component, which doesn't react with any gasification products.

It should be noted that CLAS process is still a laboratory concept and the data published in this article using equilibrium method is simply for presenting the advantages of this novel technique, giving a quick impression of this integrated technique in comparison to the previous CLAS or coal gasification process. The main input parameters of this novel technique are listed in Table 2.

	Proximate analysis					Ultimate analysis					
	Moisture	Volatiles	Fixed carbon	Ash	С	Н	0	N		S	
wt%	6.01	35.10	54.13	4.76	69.57	4.30	13.81	1.0	3	0.52	27.1

Table 1: Proximate and ultimate analysis of shenhua coal (wet basis).

	Value/Range			
Reduction temperature	810-860°C			
Oxidation reactor pressure	100kPa			
Reduction reactor pressure	100kPa			
Gasifier pressure	4000kPa			
Mn <sub>2</sub> O <sub>3</sub> / Mn <sub>3</sub> O <sub>4</sub> flow rate	100kmol			
Coal feed flow rate	800kg/h			
Air flow rate	100-250 kmol/h			

Table 2: Main input parameters of this novel technique.

#### **Results and Discussion**

The conversion of metal oxides is used to describe the reaction thermodynamic performances, and defined as below

#### Oxidation

$$\alpha_{ox} = \frac{m - m_{red}}{m_{ox} - m_{red}} \tag{15}$$

Reduction

$$\alpha_{red} = \frac{m_{ox} - m}{m_{ox} - m_{red}}$$
<sup>(6)</sup>

where  $\alpha_{ox}$  and  $\alpha_{red}$  are the metal oxides conversion during oxidation and reduction, respectively, while  $m_{ox}$ ,  $m_{red}$  and m represent the metal oxides weight at fully oxidized state, fully reduced state, and the weight at certain operational conditions, respectively.

The coal gasification efficiency is expressed using cold gas efficiency, which is defined as

$$\eta = \frac{Q_{LHV,gas}}{Q_{LHV,coal}} \tag{17}$$

where  $Q_{LHV,gas}$  and  $Q_{LHV,coal}$  are the lower heating value of gasification gas and coal, respectively, and the lower heating value of gasification gas could be calculated by eqs 18 and 19.

$$Q_{LHV,gas} = \sum_{i} LHV_{i} x_{i} \sum_{i} n_{i} \quad i = H_{2}, \text{ CO } CH_{4}$$
(18)

$$Q_{LHV,\text{coal}} = LHV_{\text{coal}} m_{\text{coal}}$$
(19)

where *LHV* represents the lower heating value, and  $x_i$  and  $n_i$  are the molar fractions and molar flow rate of gasification gas components i, respectively.  $m_{\text{coal}}$  represents the mass flow rate of coal.

# Effect of air flow rate

The air flow rate would have a significant effect on the oxidation of  $Mn_2O_3/Mn_3O_4$  metal oxides. Figure 4 shows the effects of air flow rate on the oxidation conversion ( $\alpha_{ox}$ ) of  $Mn_2O_3/Mn_3O_4$  oxygen carriers. With the increasing of air flow rate range from 100 to 250 kmol/h,  $\alpha_{ox}$  is getting rapidly increased due to that more oxygen is reacted with reduced metal oxides ( $Mn_3O_4$ ) to getting a higher oxidized state ( $Mn_2O_3$ ). The  $\alpha_{ox}$  reaches its maximum value of 1 at the air flow rate of around 240 k-mol/h.

#### Effects of reduction temperature

In this section, we focus on the effects of reduction temperature on the reduction conversion ( $\alpha_{red}$ ) of  $\rm Mn_2O_3/Mn_3O_4$  oxygen carriers and on the gasification performances.

The effect of reduction temperature on  $\alpha_{red}$  is shown in Figure

5. When reduction temperature in the range of 810-830 °C, the  $\alpha_{red}$  increases from 0.28 to 0.53, after that,  $\alpha_{red}$  increases at a faster speed from 0.53 to 1. Due to the endothermic of reduction reaction, the increasing temperature would promote the reduction of Mn<sub>2</sub>O<sub>3</sub> into Mn<sub>3</sub>O<sub>4</sub> for oxygen uncoupling, leading to increasing of  $\alpha_{red}$ .

Figure 6 shows the effect of reduction temperature on gasification compositions (dry basis). When the reduction temperature increases from 810 to 840°C, the CO and H<sub>2</sub> concentrations increase from 0.55 to 0.62 and from 0.13 to 0.30, respectively, while, shown as an opposite tendency, CO<sub>2</sub> concentration decreases rapidly from 0.18 to 0.07, with corresponding decreasing CH<sub>4</sub> concentration to few. After the reduction temperature of 840°C, the gaseous compositions show slight change with reduction temperature.

Similarly, Figure 7 shows the effect of reduction temperature on gasification performances. The gasification temperature rises significantly from 830 to 1220°C over the reduction temperature range from 810 to 840°C, whereas among the reduction temperature range, cold gas efficiency ( $\eta$ ) firstly increases from 0.74 to 0.82 and then decreases, reaching a maximum at reduction temperature of about 830°C.





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Figure 6: Effect of reduction temperature on gasification gaseous compositions (dry basis).





The reasons might be as follows: Due to the thermodynamic properties of  $Mn_2O_3/Mn_3O_4$  carriers, more oxygen is released with the increasing reduction temperature range from 810 to 840°C (see Figure 5), promoting the combustion reactions (R6-R8), and results in gasification temperature, as a result of which, the endothermic carbon gasification reactions (R10 and R11) are promoted towards

positive direction to produce more CO and H<sub>2</sub> Due to the endothermic boudouard reaction (R9) and reforming reaction (R10), the CH<sub>4</sub> and CO<sub>2</sub> concentrations decrease at higher gasification temperature. The sudden decrease in cold gas efficiency over the reduction temperature of 830°C may be caused by the rapid decreasing of CH<sub>4</sub>, illustrated in eqs 17 and 18 (the lower heating value of CH<sub>4</sub> is about 3 times than that of CO or H<sub>2</sub>).

## Effects of recycling CO<sub>2</sub>

The  $\text{CO}_2$  is removed from gasification gas by cleaning process, and then is recycled back to reduction reactor. The recycling ratio of  $\text{CO}_2$ has a significant influence on system efficiency, defined as below

$$\beta = \frac{n_{\rm CO_2}}{n_{\rm CO_2}^*} \tag{20}$$

where  $n_{CO_2}$  and  $n_{CO_2}$  are the molar flow rate of CO<sub>2</sub> recycling back to reduction reactor and the total molar flow rate of CO<sub>2</sub> removed from gasification gas.

Both the CO<sub>2</sub> and steam could be employed as inert components to reduce the partial pressure of oxygen (Figure 2), thus CO<sub>2</sub> could replace partial steam to obtain fully oxygen uncoupling. To further indicate this, Figure 8 illustrates the effect of recycling ratio of CO<sub>2</sub> ( $\beta$ ) on the required steam flow rate in the  $\beta$  range of 0-1. The required steam (100°C, 101.325kpa) decreases from 8kmol/h to 6kmol/h with the increasing  $\beta$  rang of 0-1, which further indicates that the recycling CO2 could reduce the required steam flow rate.

Figure 9 shows the effects of  $\beta$  on gasification gaseous compositions. The CO concentration increases with  $\beta$  at a moderate speed from 0.62 to 0.64, while H<sub>2</sub> concentration presents an opposite tendency, decreasing from 0.34 to 0.31. Interestingly, The CO<sub>2</sub> concentration is independent with increasing  $\beta$ , remaining almost unchanged at low value of around 0.02. As mentioned above, the increasing  $\beta$  means more CO<sub>2</sub> is introduced to gasifier. As a result of the boudouard reaction, CO<sub>2</sub> is converting to CO, which leads to the CO<sub>2</sub> concentration unchanged, whereas, the increasing CO<sub>2</sub> would inhibit the water-gas shift reaction towards the forward direction, resulting in a continuous decrease in H<sub>2</sub> content.

Figure 10 indicates that no obvious indirect effect is observed between  $\beta$  and cold gas efficiency ( $\eta$ ), but the  $\beta$  has shown little influence on gasification temperature.

With increasing  $\beta,$  the  $\eta$  is staying almost unchanged due to the little change of lower heating value of gasification gas (see epn 17 and



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18). Because the boudouard reaction is endothermic, consequently, the gasification temperature exhibits a decreasing tendency.

# Conclusions

A novel technique based on coal gasification integrated with chemical looping air separation is presented in this article. This process mainly consists of three parts, i.e., chemical looping air separation, coal gasifier, and gas cleaning process (acid gas removal). The CO<sub>2</sub> separated from gasification gas is recycled back to reduction reactor, utilization of CO<sub>2</sub> by promoting oxygen production and gasification process.

To determine the feasibility of this process, the thermodynamic performances, including the redox conversion, gasification gaseous compositions and  $\eta$  were the main subject of focus. The effects of three key variables, as air flow rate in the range of 100-250kmol/h , reduction temperature in the range of 810-860°C, and  $\beta$  in the range of 0-1were investigated. The increasing of air flow rate would promote the oxidation of  $Mn_2O_3/Mn_3O_4$  oxygen carriers, which reaches the fully oxidized state at the air flow rate of 240kmol/h. An increase in reduction temperature would promote oxygen uncoupling, increasing of CO and  $H_2$  from 0.55 to 0.62 and from 0.13 to 0.30, respectively, and decreasing of CO<sub>2</sub> and CH<sub>4</sub> in gasification gaseous compositions.

The increasing  $\beta$  could significantly reduce the required steam flow rate and promotes the CO concentration at a moderate speed from 0.62 to 0.64.

Though there is still a long way to go from commercial use, this results presented in this work can be used as an initial guess for the process, and the economic assessment, heat integration, chemical kinetics, and hydrodynamics are expected for the further studies.

# Nomenclature

## Abbreviations

CASU= cryogenic air separation unit

- $CCS = CO_2$ , capture and sequestration
- CLAR=chemical looping air separation
- CLC=chemical looping combustion

FR=fuel reactor

IGCC= Integrated Gasification Combined Cycle

ITM= advanced ion-transport membrane

 $H/M=H_2O$  introduced to FR to methane ratio

NMS= nano-structured molecular sieves

OR=Oxidation reactor

PFBC= Pressurized Fluidized Bed Combustion

RR=Reduction reactor

S/M=Steam introduced to carbonator to methane ratio

#### Variables

n=molar flow rate

m=mass flow rate

Q<sub>LHV</sub>=lower heating value

## **Greek Symbols**

 $\alpha_{ox}$  =oxidation conversion of metal oxides

 $\alpha_{red}$  =reduction conversion of metal oxides

η=cold gas efficiency

 $\beta$  = recycling ratio of CO<sub>2</sub>

 $\Psi$ = molar ratio between inert components and circulation rate of Mn<sub>2</sub>O<sub>3</sub>

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