Chemical Engineering and Processing 106 (2016) 33–41

Contents lists available at ScienceDirect

Chemical Engineering and Processing: Process Intensification

journal homepage: www.elsevier.com/locate/cep

Thermodynamic evaluation of a conceptual process for coal gasification coupled with chemical looping air separation

Lin Zhu[a,*], Feng Wang[a,b], Zheng Zhang[a]

[a] Key Laboratory of Gas Process Engineering, Chemistry and Chemical Engineering Institute, Southwest Petroleum University, Chengdu 610500, Sichuan Province, China
[b] China Petroleum LONGWAY Engineering Project management Co.,LTD, Langfang 065000, Hebei Province, China

ARTICLE INFO

Article history:
Received 4 May 2015
Received in revised form 28 March 2016
Accepted 1 May 2016
Available online 4 May 2016

Keywords:
Coal gasification
Chemical looping air separation
Aspen plus
Thermodynamic analysis

ABSTRACT

For the pressing need to reduce oxygen production cost of coal gasification and to utilize carbon dioxde in an effective approach, this present work, from the view of thermodynamic perspective, presents a novel technique, i.e. coal gasification integrated with chemical looping air separation (CLAS). CLAS is expected to offset 1–3% of the energy penalty associated with the world-wide oxygen production technology, i.e., cryogenic air separation unit (CASU). The thermodynamic performances, as the conversion of metal oxides (Mn_2O_3/Mn_3O_4) of redox reactions, gasification gaseous compositions and cold gas efficiency (CGE) were the main subject of focus. The four major factors, air flow rate, reduction temperature, oxygen-to-coal ratio (O/C) and steam-to-coal ratio (S/C) appear to have significant impact on the performance of the whole process, therefore comprehensive investigations related to these variations have been reached. In parallel the exergy destruction analysis has also been involved.

1. Introduction

Coal, characterized as a less costly and widely distributed fossil energy, plays an irreplaceable role in the world's energy portfolio. In China, coal reserves, accounting for 70% of the primary energy structure, will continuously last its major role in the near-coming middle age. Electricity generation is mostly derived from coal combustion [1], unfortunately several environmental issues have been appeared in the art-of-craft coal-to-electricity plants, such as cause of acid rain, ozone depletion, and the anthropogenic greenhouse effect [2]. In response to mitigation of improper effects from coal combustion, advanced Integrated Gasification Combined Cycle (IGCC) with CO_2 capture and sequestration (CCS) [3–5], and Pressurized Fluidized Bed Combustion (PFBC) [6,7] have been demonstrated as a promising approach for future coal utilization.

Between both candidates, coal gasification is posing as an attractive option in that it can effectively convert coal into syngas, followed by the sequenced electricity generation or the synthesis of useful chemical products, e.g., methanol [8].

In coal gasification process, the air- separation oxygen, as a gasification agent, is introduced to gasifier. However, air separation means increasing energy penalty for the whole electricity production process, in terms of oxygen production, cryogenic air separation unit (CASU), advanced ion-transport membrane (ITM), and nano-structured molecular sieves (NMS) [9–12], have become a gap between the wide spreading IGCC utilization and the present coal combustion technology due to their expensive separation cost as well as high energy penalty. For example, the most widely used CASU for oxygen production brings high energy penalty, leading to rise approximately 3–4% energy penalty in the oxy-fuel operations [13], in parallel resulting in revenue losses and the increasing coal utilization per unit of electricity generated, also assumed 10–40% of the thermal in the net oxygen-fuel power plant can be exploited for oxygen production with a specific power consumption of about 0.4 kWh/m^3 of O_2 [14–16]. Although the ITM and NMS can reduce around 10–25% of the capital costs in comparison to that of CASU, both technologies still remain expensive, causing the membrane, fabrication, installation, maintenance and integration issues [14–16].

Chemical looping air separation (CLAS) emerges as a novel concept of offering 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 the conventional oxygen production technologies [13,17–19]. The concept of CLAS was

* Corresponding author at: Xinndu Avenue 8#, Xindu District, Chengdu City, Sichuan 610500, China.
E-mail address: Zhulinswpi65@gmail.com (L. Zhu).

http://dx.doi.org/10.1016/j.cep.2016.05.002
0255-2701/

Nomenclature

Abbreviations
CASU Cryogenic air separation unit
CCS CO_2 capture and sequestratio
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

Greek Symbols
α_{ox} Oxidation conversion of metal oxides
α_{red} Reduction conversion of metal oxides
CGE Cold gas efficiency
Ψ Molar ratio between inert components and circulation rate of Mn_2O_3

originally proposed by Moghtaderi and his group at the University of Newcastle, Australia in 2010 [20,21]. They concluded the specific power for the CLAS (approximately 0.045 kWh/m^3 of oxygen) was much lower than the power required in the conventional CASU [13,22–25]. Later in 2013, Moghtaderi et al. developed several process options for integration of chemical looping air separation (ICLAS) at the purpose of extending the application of CLAS in the oxy-fuel-type power plants at a lower operating cost [13].

The configuration of the CLAS is similar to the chemical looping oxygen uncoupling (CLOU), which can be found elsewhere [26–30]. And the schematics of both CLAS and CLOU concepts are illustrated in Fig. 1a and b, respectively. The most significant difference between two concepts is the different application of oxygen. Both of them are able to separate oxygen in air through the cyclic redox reaction between two interconnected reactors. Oxidation of metal oxides occurs in an oxidation reactor (OR) for both concepts. In terms of CLAS concept, reduction of metal oxides takes place in reduction reactor (RR), where oxygen decoupling occurs in the presence of steam or CO_2 to reduce the partial pressure of oxygen in RR. The reduction reaction (oxygen uncoupling) is shown as below:

$$Me_xO_y \rightarrow Me_xO_{y-2} + O_2 \quad (1)$$

As shown in Fig. 1a, the oxygen generated from CLAS can be employed as an oxidation agent for any kinds of oxy-fuel plants, for comparison, oxygen derived from CLOU is directly combusted with coal (Fig. 1b). Therefore, it can be concluded CLAS provides a more flexible pathway for oxygen utilization.

In the CLAS concept, the reduced metal oxides from RR are then transported back to oxidation reactor (OR), for the purpose of reoxidation of reduced oxides, air supplies necessarily required oxygen to maintain full regeneration. The oxygen coupling step has been illustrated in Reaction (2).

$$Me_xO_{y-2} + O_2 \rightarrow Me_xO_y \quad (2)$$

As illustrated above, the concept of CLAS is relatively simple and involves the cyclic oxidation and reduction of metallic oxide particles in two separate reactors to separate oxygen from air. The simplicity of its hardware and operation significantly reduces the CLAS's capital cost [23,24,31]. Additionally, the process enjoys low operating costs because of its small energy footprint given that much of the heat for endothermic reduction of metal oxides is provided by exothermic oxidation of metals; reducing the need of large thermal energy input [13,23].

Inspired by this concept, the coal gasification integrated with chemical looping air separation process is proposed in this article. Coal is used as the raw material, gasified by oxygen produced from CLAS, to achieve high-purity hydrogen production and by-product steam produced by application of heat recovery steam generator (HRSG). The main focus of this manuscript is to investigate the potential advantages of this aforementioned integrated process from a thermodynamic perspective. Aspen Plus (v7.2) software was used to develop this novel process based on a thermodynamic method. Some key variables with significant influence on the overall plant performance were discussed in details. Also, the exergy destruction analysis of the novel process was involved.

2. Process configuration and simulation

2.1. Thermodynamic analysis of CLAS reactions

According to the previous studies from Moghtaderi and his partners [13,22], CLAS is mainly a thermo- chemical process that can be carried out at biospheric pressure. In addition, CuO/Cu_2O, CoO/Co_3O_4, MnO_2/Mn_2O_3 and Mn_2O_3/Mn_3O_4 perform well as

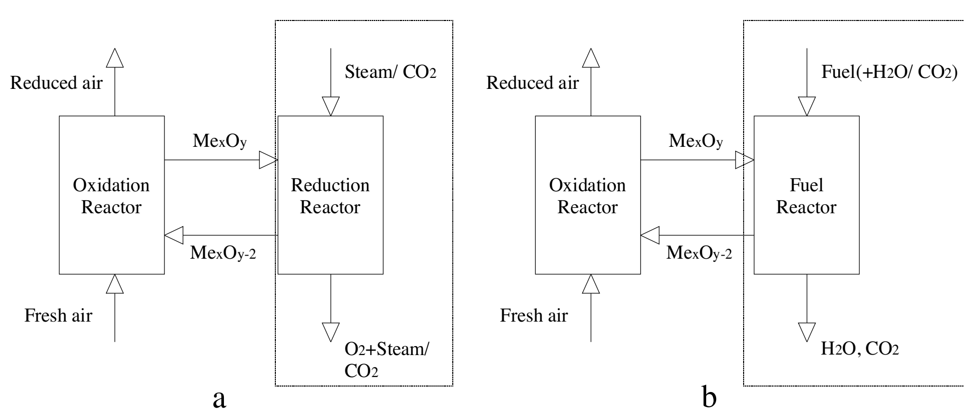

Fig. 1. Schematic of the chemical looping air separation (CLAS) concept (a) and the chemical looping oxygen uncoupling (CLOU) concept (b).