



Life cycle greenhouse gas assessment of hydrogen production via chemical looping combustion thermally coupled steam reforming

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ABSTRACT

Chemical looping combustion thermally coupled steam reforming (CLC-SR) emerges as a new alternative to achieving hydrogen production simultaneously with inherent CO₂ capture. To evaluate the environmental performances of any competing technology, it is of necessity to consider total emissions in association with the technology over its entire lifetime. In this study, a life cycle greenhouse gas emission (LCE) assessment of the CLC-SR is conducted, together with a comparison with the conventional natural gas steam reforming (SR). The energy efficiency of 75.2% for achieving 97.0% inherent CO₂ capture is obtained in the CLC-SR, with the LCEs of 3009 g CO₂ eq./kg H₂, approximately accounting for one third in those of conventional natural gas SR. The main LCE contributors in the CLC-SR are plant operation and natural gas losses from the natural gas production and transport. To further meet the emission reduction potential, several key parameters are varied to illustrate their influence on LCE performances of the CLC-SR, including reformer operation temperature, different types of oxygen carriers (OCs) and the lifetime of OCs. The results of this investigation demonstrate the CLC-SR is a promising alternative to conventional SR for cleaner hydrogen production from a LCE perspective.

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1. Introduction

Growing concerns about the shortage of fossil fuels and climate change have motivated researches for new energy sources and sustainable energy systems (Zhu and Fan, 2015). Hydrogen is generally considered as an ideal alternative to fossil fuels due to its ability to reduce anthropogenic emissions of greenhouse gases (GHGs) (e.g. CO₂) and its high efficiency when fed into fuel cells (McIntosh and Gorte, 2004).

Yet, hydrogen is a secondary source of energy, generated from various primary sources including fossil fuels (natural gas, coal and oil) and renewable energy sources (e.g. biomass, solar, wind and nuclear) (Fan and Zhu, 2015). The environmental performances of H₂-production processes are highly dependent on the form of primary sources (fossil fuels or renewable energy sources) and the employed conversion processes (Serrano et al., 2012). Although dozens of demonstration projects of hydrogen production from renewable electricity have been realized in the last two decades,

some technical and economic barriers have to be solved before these projects can be commercially successful (Götz et al., 2016; Gahleitner, 2013). It is predicted hydrogen productions will continue to be highly relied on fossil fuels in the coming decades (Hosseini and Wahid, 2016). Specifically, natural gas steam reforming (SR) is the dominant route for industrial hydrogen production, which is responsible for about 50% of the hydrogen production worldwide (Sharma et al., 2017). The main reactions are given in Eqs. (1)–(3) (Zhu et al., 2015b).

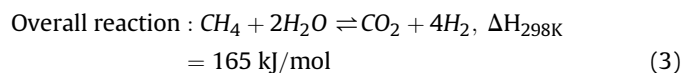
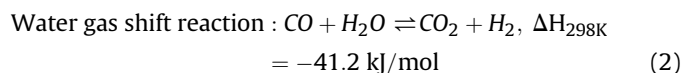
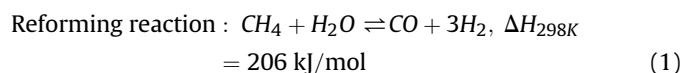


Fig. 1(a) presents a schematic diagram of the conventional

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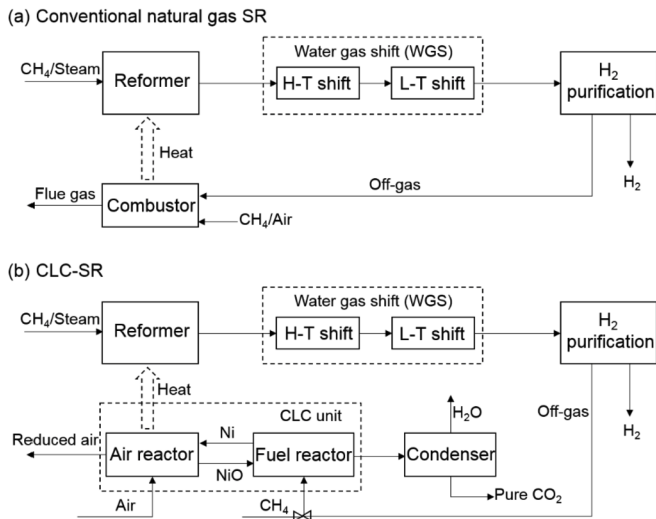


Fig. 1. Schematic diagram of conventional natural gas SR and CLC-SR process.

natural gas SR process. Since the reforming reaction is highly endothermic, a huge amount of supplemental energy is required by combustion of additional natural gas (if necessary) or the off-gas from the H₂ purification unit. The reforming and combustion processes result in high CO₂ emissions, accounting to be approximate 11 kg CO₂ eq./kg H₂ (11,888 g CO₂ eq./kg H₂ in (Spath and Mann, 2000); 10,640 g CO₂ eq./kg H₂ in (Dufour et al., 2009); 11,893 g CO₂ eq./kg H₂ in (Cetinkaya et al., 2012); 10,560 g CO₂ eq./kg H₂ in (Susmozas et al., 2013)).

Although natural gas SR is a mature industrial process, the associated high CO₂ emissions should be reduced to mitigate global warming (Zhu et al., 2016). As one possible option, the implementation of CO₂ capture and storage (CCS) techniques is quite promising, which includes pre-combustion capture, post-combustion capture and capture in oxy-combustion (Kanniche et al., 2010). But the high cost and energy penalty of the current CCS techniques keep them far from being mature in the commercial application. The energy penalty in the CO₂ capture process decreases the overall energy efficiency by 6% in SR (Tzanetis et al., 2012) and 5–20% in power plant (Davison, 2007). The up-to-date information about CCS can be found elsewhere (Araújo and de Medeiros, 2017).

Fortunately, chemical looping combustion (CLC) process emerges as a promising technology which is also called “next-generation” CCS. The CLC process is capable of obtaining inherent separation of CO₂ from other flue gas components (Zhu et al., 2015a), so no other gas separation equipment is required and no extra energy is consumed for the gas separation (Boot-Handford et al., 2014). The estimated cost of CLC is lower than other CO₂ capture options and it is even more preferred if the environmental impact is also considered (Adánez et al., 2012). In CLC, the fuel combustion is fulfilled by two sub-reactions occurring in two reactors, i.e., a fuel reactor (FR) and an air reactor (AR) (Fan et al., 2017b). A kind of metal oxide as oxygen carriers (OCs) is applied to oxidize fuel in FR and to be reoxidized in AR by fresh air, which avoids the direct contact between fuel and air. The exhaust from FR mainly contains CO₂ and water vapor. After water vapor condensation, an almost pure CO₂ stream ready for transport and storage is obtained (Wang et al., 2017). The majority of the CLC plants existing worldwide so far use the configuration composed of two interconnected fluidized-bed reactors which can achieve a good contact between gas and solids as well as the flow of solid material between FR and AR (Lyngfelt et al., 2001). More detailed process description

of CLC can be found in (Adánez et al., 2012).

Proposals on applications of CLC for H₂ production have been expanded significantly over the last ten years, e.g. chemical looping combustion thermally coupled steam reforming (CLC-SR), auto-thermal chemical-looping reforming (CLR) and chemical-looping hydrogen (CLH). The differences among these processes have been elaborated elsewhere (Adánez et al., 2012). Among them, CLC-SR is the easiest approach to being realized because of its superior character of directly integrating with on-site reformer by replacing the conventional combustion chamber into CLC combustor. This concept was first proposed by Rydén and Lyngfelt in 2006 with the aim of reducing carbon footprint for hydrogen production (Rydén and Lyngfelt, 2006). A schematic diagram of CLC-SR is presented in Fig. 1(b). The main difference with respect to conventional SR is that a CLC unit is applied instead of combustor to supply heat to the endothermic reforming reactions, as such for the mission of inherent CO₂ capture. With pioneering R&D efforts, researches concerning on this concept has been mainly extended into two branches including: (1) design of novel reactor which is capable of thermally integrating of SR with CLC, and the challenges ahead are maintaining stable circulating rate of OCs between FR and AR as well as high-efficiency heat transfer between AR and reformer (Rahimpour et al., 2012); (2) process simulation and optimization for the purpose of improving energy efficiency, exergy efficiency and heat transfer efficiency thermodynamically (Fan et al., 2016).

Even though CLC-SR is capable of producing hydrogen with inherent carbon capture, this process does not guarantee a better environmental performance from its life-time operation. Doubters are still questioning if this technology is qualifying to reduce GHGs (particularly CO₂) from the life cycle view because of the extra emissions resulting from OC manufacturing as well as its make-up assigning by reactivity losses and attritions during successive redox reactions. As such, life cycle GHG emission (LCE) assessment of CLC-SR is the main focus of this study, considering emissions from plant construction and operation to final decommissioning. Although abundant LCE assessment work have been reported associated with technologies related to hydrogen production from both fossil fuel and renewable resources, including SR (Spath and Mann, 2000), gasification (Kalinci et al., 2012), water electrolysis (Cetinkaya et al., 2012), water splitting (Ozbilen et al., 2012) and methane decomposition (Dufour et al., 2010), the study on examining the LCEs in CLC-SR is not reported to the best of our knowledge. The very limited life cycle assessment (LCA) work on the CLC process for H₂ production focuses on three-stage Fe-based chemical looping hydrogen process (Petrescu et al., 2014) and CLR for hydrogen production (Salkuyeh et al., 2017). Sensitivity analyses of key system parameters are still not well-known to examine the environmental feasibility of such process. Notably, in most carbon-capture related technologies, providing environmental benefits is somehow against the anticipation of technical feasibility, since capturing carbon leads to process efficiency losses. However in this CLC-SR process, we are intended to demonstrate the synergistic effect for simultaneously obtaining environmental benefits and technical feasibility associated with CLC. Furthermore the relationship between LCEs and the pathway of integrating CLC with SR is not deeply analyzed and remains questionable.

Inspired by this, we conduct a life cycle assessment (LCA) of CLC-SR system to count the overall carbon emissions from plant construction until final decommissioning in light of our pervious study which demonstrates the thermodynamic feasibility together with economic feasibility of this process in relative to conventional SR process (Fan et al., 2016). Herein, focuses are addressing on the environmental benefits of thermal coupling CLC with SR for hydrogen production.

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