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CO₂-Oxy underground coal gasification integrated proton exchange membrane fuel cell operating in a chemical looping mode of reforming

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ABSTRACT

O₂/CO₂ based underground coal gasification (UCG) is a promising clean coal technology for the exploitation of deep underground coal resources with a minimal difficulty of the operation. However, the generated syngas of the underground is enriched with carbon monoxide and it should be converted into a clean gas on the above ground for the end use. In the present study, a clean mode of power generation is proposed using the CO enriched UCG syngas. Firstly, the UCG gas is reformed into pure hydrogen using chemical looping reforming (CLR) method. Secondly, the pure hydrogen obtained from CLR process is used suitably to generate electric power in a proton exchange membrane fuel cell (PEMFC) system. Hence, carbon energy is converted into hydrogen energy through the CLR technique and this clean gas is efficiently transformed into electrical energy using the PEMFC system. A net efficiency for converting CO enriched gas into H₂ through conventional system using water gas shifting reactors is evaluated and compared with the CLR system. An energy analysis of O₂/CO₂ based UCG integrated with CLR-PEMFC shows 43.6% net efficiency with carbon capture and storage (CCS).

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Introduction

Depletion of fossil fuel demands to improve the efficiency of the existing technologies as well recovering the unutilized energy resources. Deep coal resources can be utilized using an unconventional technology known as underground coal gasification (UCG). As nearly 40% of coal reserves are found at a depth higher than 300 m, UCG is the appropriate technology for the recovery of these resources effectively. The UCG is an inherent clean technology as it avoids several conventional difficulties of coal mining and processing [1]. In addition, the production of NO_x and SO_x during the UCG operation is

limited. This is due to the in-situ conditions prevailing on fuel enriched combustion of UCG. Therefore, the formation of these pollutant gases is minimized.

The use of gasifying medium for the UCG plays a vital role in deciding the syngas composition. Air/H₂O gasifying medium leads to N₂ rich syngas. H₂O/O₂ gasifying medium produces a hydrogen rich syngas whereas CO₂/O₂ gasifying medium generates a CO enriched product gas. These gases can be post processed for the clean recovery of the energy utilization. It is proposed that CO₂ gasification of UCG may be economical and easier in operation compared to steam gasification of UCG. However, the generated syngas is not a clean source of energy as it is enriched with carbon energy (in the

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form of CO). It would be appropriate if the downstream processing of UCG syngas could be performed in the context of carbon capture and storage (CCS) in order to avoid global warming. Therefore, the present study aims at the conversion of CO₂/O₂ based UCG syngas into a clean source of energy in the context of CCS.

Research on hydrogen generation is essential for clean environment. A lot of research is ongoing for the production of clean fuel gas. Several literature are available on hydrogen generation through advanced technologies. Ouyang et al. [2–5] research group works extensively on assessment of hydrogen generation using metal hydrides. They estimated the energy efficiency of various metal hydrides. They estimated the energy efficiency of the hydrolysis and regeneration of sodium metaborate is 49.91% [2]. The energy efficiency of the MgH₂, H–Mg₃La and H–La₂Mg₁₇ hydrolysis cycles were estimated as 45.3%, 40.1% and 41.1%, respectively [3]. They prepared a new lanthanum based alloy as electrode for Ni/MH batteries and it shows high discharge capacity with cyclic property [4]. They further studied the effect of ammonium chloride on hydrogen generation in hydrolysis of MgH₂ [5]. One of the recent emerging technologies for the hydrogen generation is chemical looping reforming with inherent carbon capture. Fan [6] explained about the novel applications of chemical looping techniques. He discussed about the suitability of metal oxides for fuel, thermodynamic phase equilibrium of metal oxides and chemical looping techniques for solid and gaseous fuels. Cormos [7] evaluated the chemical looping system for hydrogen production and power generation using iron based metal oxides in the context of CCS. The paper evaluates the plant concepts and critical gasifier design factors to increase the efficiency of the power plant system. Chen et al. [8] carried out experimental investigation on the reactivity of iron based metal oxides with Al₂O₃ or TiO₂ inert supports for chemical looping hydrogen production. Their study shows the better reactivity of Fe₂O₃ as the oxygen carrier and it is found that there is no deterioration of metal/metal oxide structural property after multi-cycle experiments. Chiesa et al. [9] analyzed chemical looping techniques for hydrogen generation using natural gas as fuel. They proposed three reactors for CLR using iron as the metal oxides. The study estimated hydrogen efficiency and electrical efficiency of gas turbine and steam turbine systems for power production. Cleeton et al. [10] estimated the hydrogen production efficiency of chemical looping combustion system integrated steam based coal gasification with Fe₂O₃ as oxygen carrier. They found 48.4% and 58.3% of exergetic efficiency for hydrogen production at 1 and 10 atm, respectively. Ryden et al. [11] carried out chemical looping reforming for hydrogen production in a fluidized bed reactor using Fe₃O₄ metal oxides supported on MgAl₂O₄ inert materials. They found that the reactivity of metal oxide particles is high with CO and syngas fuels. A decrease in specific surface area and particle size and an increase in the bulk density of the metal/metal oxide particles are observed after the chemical looping treatment. Kathe et al. [12] carried out ASPEN simulations on the chemical looping techniques for hydrogen production with iron based oxygen carriers using methane as fuel. They found an increase of 6% higher thermal efficiency of chemical looping techniques than the conventional reforming process. Chan

et al. [13] studied the capability of Ca₂Fe₂O₅ metal oxide particles for higher conversion of steam to hydrogen. They found that an equilibrium conversion of 75% is achieved for the production of steam to hydrogen using the modified Fe particles at 1123 K.

In our earlier studies, CO₂ based UCG was proposed and the net electrical efficiency of the conventional and advanced power generating systems was evaluated [14,15]. In continuation of the earlier studies, a chemical looping reforming (CLR) based proton exchange membrane fuel cell (PEMFC) operation is proposed for converting carbon energy enriched syngas into a clean hydrogen gas. Further, it is converted efficiently into electric power using the PEMFC system. The trace level of CO in feed gas severely affects the performance of PEM fuel cell [16]. Several researches are progressing for cleaning the feed hydrogen to the PEM fuel cell. Ersoz et al. [17] studied the reforming options for PEM fuel cell using steam reforming, partial oxidation and auto thermal reforming technologies for fossil fuels and concluded that steam reforming is more suitable process for the conversion. Apart from conventional reforming technologies, chemical looping reforming is more appropriate technology for the production of ultrapure hydrogen, which is highly suitable for PEM fuel operation. Silva et al. [18] developed an electrochemical model and performed material and energy analysis for the PEMFC system integrated biogas CLR and found that a net electrical efficiency of 45% at a low load conditions. Chen et al. [19] proposed a hybrid power plant, which integrates solid oxide fuel cell (SOFC)/gas turbine (GT) with the CLR for hydrogen generation in the context of CCS. They estimated a net power efficiency of 43.53% at a cell operating conditions of 20 bar and 900 °C. Diego et al. [20] conducted auto-thermal CLR based experiments in a circulating fluidized bed reactors using methane gas. They found the methane conversion of 98% with 2.5 mol of H₂ per mol of CH₄ using NiO supported on alumina (Al₂O₃) inerts. Luo et al. [21] investigated the performance of iron based oxygen carriers such as pure Fe₂O₃, synthetic Fe₂O₃/MgAl₂O₄ and iron ore in the reduction process using thermogravimetric analyzer (TGA) and concluded that a high reactivity of methane fuel is observed in the case of Fe₂O₃/MgAl₂O₄ oxygen carriers. Ortiz et al. [22] studied the performance of iron waste material in a CLR process using tail gas, which is obtained from pressure swing adsorption (PSA) process. They found that the oxygen carriers experienced a higher oxygen transport capacity at 800 °C. Galvita et al. [23] proposed a novel combined chemical looping process with Fe₃O₄ and CaO using CH₄ and CO₂ as feed, for energy storage and release. The above studies show the feasibility of hydrogen production through CLR techniques. Therefore, the present study deals with the production of hydrogen through CLR process using CO enriched UCG syngas, which is generated from CO₂ based underground coal gasification.

In the present study, thermodynamic analyses have been carried out for the PEMFC power production using CO enriched syngas in the context of the CCS. Mass and energy analysis have been carried out to estimate the net electrical efficiency of the proposed systems. The net thermal efficiencies of conventional and advanced technologies are evaluated and compared. Fig. 1 shows the conceptual representation of CO₂/O₂ UCG integration with PEM fuel cell

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