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Performance analysis of a water–gas shift membrane reactor for integrated coal gasification combined cycle plant

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ABSTRACT

In integrated gasification combined cycle (IGCC) systems, the water–gas shift reaction, which promotes the conversion of CO present in syngas mixtures into hydrogen, is an important step for hydrogen production. Application of the water–gas shift membrane reactor (WGSMR) in IGCC systems is an attractive option for CO_2 capture compared with conventional methods because of smaller heat loss in gas purification and high CO conversion by selectively removing hydrogen from the reaction zone through the membrane. In this study, we proposed and evaluated commercial-scale WGSMR models combined with IGCC using reported laboratory-scale experimental data to optimize their operational parameters. Various models were developed using the *Aspen Plus*® Ver. 8.6 process simulator to investigate the impacts of hydrogen separation, pressure loss, and the flow direction between the sweep gas on the permeate side and syngas on the retentate side on the WGSMR performance with respect to CO conversion, H_2 yield, and reactor temperature. The membrane reactor model gave approximately 20% higher CO conversion than a reactor model without H_2 separation and approximately 4% lower CO conversion than a membrane reactor model with a pressure drop. A counter-current membrane reactor model gave approximately 2% higher CO conversion than a co-current model; the H_2 yield on the permeate side was 9.3% higher in the counter-current model by separation of H_2 through the membrane. A sensitivity analysis indicated that a high flow rate and low pressure of sweep gas are advantageous for H_2 recovery, and high catalyst loading and high syngas inlet temperature are preferable for higher CO conversion.

1. Introduction

Coal is the largest energy source for electricity generation worldwide; however, the burning of coal is one of the major contributors to climate change. Crises of fossil fuel depletion and environmental degradation mean that efficient utilization of energy and technology improvements have become important agendas [1]. Coal gasification is an important technology for the efficient production of hydrogen and electricity [2–4]. In recent years, the integrated coal gasification combined-cycle (IGCC) process, which produces H_2 and CO in a coal gasifier and generates electricity by combusting the produced H_2 in gas turbines and fuel cells, is one of the successful technologies to replace conventional coal-fired power plants [5–7]. In particular, IGCC has greater advantages when low-rank coals and biomass are used as energy sources [8–10] and has scope for further development [11–16]; however, owing to the strong demand for reduction of CO_2 emissions from power generation plants, carbon capture and storage (CCS) has become a required technology in recent years [17].

The water–gas shift membrane reactor (WGSMR), which continuously removes H_2 from the products of the water–gas shift (WGS) reaction, is a promising means of syngas-to-hydrogen conversion with favorable H_2 production and/or CCS capabilities [18,19]. The WGS reaction is represented by Eq. (1) [20]:

$$CO + H_2O \rightleftharpoons CO_2 + H_2 \qquad \Delta H_{298K} = -41.1\ \text{kJ/mol} \tag{1}$$

WGSMR is well suited to IGCC applications owing to its simple operating process [21–24]. In WGSMR, syngas (including sulfur and steam) flows directly into the reactor and the WGS reaction takes place on a catalyst that has sulfur tolerance around 300–400 °C. The H_2 and CO_2 produced are simultaneously separated by a membrane. The high pressure and temperature of the syngas are advantageous to producing hydrogen gas and transporting it across the membrane [25]. The driving forces to separate H_2 in a WGSMR can be further enhanced if a sweep gas is used. Iulianelli et al. [26] analyzed recent advances in WGSMR, especially those that can operate at reaction temperatures (370–400 °C) with high hydrogen selectivity. Extensive laboratory-scale experimental studies were conducted. Augustine et al. [27] investigated a palladium alloy (Pd-alloy)-based WGSMR. They observed that such

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Nomenclature

Abbreviations

ASU	air separation unit
EOS	equation of state
IGCC	integrated gasification combined cycle
LHV	lower heating value
PFR	plug-flow reactor
S:C	steam:carbon ratio
TMOS	tetramethoxysilane
WGS	water–gas shift
WGSMR	water–gas shift membrane reactor

Symbols

A_0	contact area in each membrane module [m^2]
C_p	heat capacity of gas [J/(K mol)]
d_p	particle diameter of catalyst [m]
F	flow rate of gas [kmol/h]
$F_{CO,in}$	flow rate of CO in syngas inlet [kmol/h]
$F_{CO,out}$	flow rate of CO in syngas outlet [kmol/h]
$F_{H2,perm}$	flow rate of H_2 on permeate side [kmol/h]
$F_{H2,ret}$	flow rate of H_2 on retentate side [kmol/h]
$F_{R''N''A}$	hydrogen flow rate in stream R″N″A to N^{th} module [kmol/h]
ΔH_{298K}	enthalpy of reaction [kJ/mol]
h_h	heat transfer coefficient of hot side [W/(m^2 K)]
h_l	heat transfer coefficient of cold side [W/(m^2 K)]
J_{H2}	hydrogen permeation rate [kmol/h]
$J_{i''N''}$	permeation flow of component *i* in module ″*N*″ in reactor [kmol/h]
K	equilibrium constant [–]
k_j	thermal conductivity of layer *j* (stainless tube and insulator) [W/(m K)]
L	reactor length [m]
N	N^{th} module [–]
Δp	pressure drop [Pa]
$P_{e,i}$	hydrogen permeability in component *i* [mol m/(m^2 s Pa)]
p_i	partial pressure of component *i* [Pa]
$p_{i,perm}$	partial pressure of component *i* on permeate side [Pa]
$p_{i,ret}$	partial pressure of component *i* on retentate side [Pa]
Q	heat loss from the reactor [W]
R'	gas constant [J/(mol K)]
$R_{''N''}$	reaction rate for hydrogen product [kmol/h]
r_1	radius of inner side in reactor [m]
r_j	radius of layer *j* (stainless tube and insulator) [m]
T	reactor temperature [K]
T_h	temperature of hot side in reactor [K]
T_l	temperature of cold side in reactor [K]
U	overall heat-transfer coefficient [W/(m^2 K)]
u	superficial gas velocity [m/s]

Greek alphabets

$\alpha_{A/B}$	separation coefficient between A and B [–]
γ	thickness of membrane [m]
ε	voidage of catalyst bed [–]
μ	viscosity of fluid [Pa s]
ρ_f	density of fluid [kg/m^3]

membranes are highly hydrogen-selective, which makes them one of the most popular membrane technologies for replacing the water–gas shift reactor. Basile et al. [28] studied the performance of a WGSMR in the temperature range of 320 to 400 °C and reaction pressures up to 0.17 MPa. They concluded that optimal CO conversion could be achieved with a high degree of H_2 removal from the system. Mendes et al. [29] investigated the behavior of a WGSMR with respect to H_2 recovery in the low-temperature range (200 to 300 °C). They found that higher CO conversion was achieved at lower temperatures owing to the equilibrium advantage. Different membrane materials for WGSMR have also been examined [27,30–33]. The performance and economic attractiveness of metallic and non-metallic membrane reactors used for H_2 production from coal plants were investigated by Dolan et al. [34,35]. Recently, proton ceramic membrane reactors have been proposed and applied in simultaneous steam methane reforming and WGS, coal steam gasification and WGS [36–38].

Some researchers investigated the WGSMR with an IGCC system [39–43]. Al-Zareer et al. [44] carried out modelling and performance analysis of an IGCC system with a WGSMR for hydrogen production. They investigated the effects of mass flow rate of oxygen, steam, and coal type. They concluded that the parameters that maximize the hydrogen production rate differ from those that maximize overall net energy efficiency of the IGCC system. Lotric et al. [45] simulated a WGSMR for an IGCC plant with CO_2 capture. They found that greater than 70% of CO conversion to H_2 was achieved in the first 40% of reactor length and the hydrogen yield was dominated by the permeation rate of hydrogen. Franz et al. [46] evaluated the pre-combustion CO_2 capture in IGCC power plants and reported that efficiency losses in the WGSMR can be reduced as low as 5.8% for a CO_2 degree of separation of 90%. Maas and Scherer [47] investigated the pre-combustion CO_2 capture with ceramic membranes in a lignite-fired IGCC plant. Their results showed that the net energy efficiency of 41.97% in the IGCC system with a WGSMR and a CO_2 separation of 97.6% could be achieved. Bracht et al. [48] investigated a WGSMR for CO_2 removal in IGCC systems, finding that the net thermal efficiency of the IGCC system was 42.8% (lower heating value, LHV basis) with a CO_2 recovery of 80%.

Numerous studies of WGSMR operations have been carried out, most of which focused on laboratory-scale experiments, computational fluid dynamics (CFD) [20], and economic analyses [46,47]. The hydrodynamics are important for analyzing detailed heat and mass transport in a WGSMR [20]; however, CFD capability is limited by massive computational time and cost. The application of CFD to IGCC has therefore been limited to CO_2 capturing units and reactors and has not yet been conducted to analysis of the entire process of IGCC with a WGSMR in a commercial scale. Nevertheless, integration of WGSMR into the commercial IGCC process and its detailed analysis are indispensable for its practical use. Although a process simulator is beneficial for such whole-process calculation, its application to this technology has been quite limited.

In the present study, we attempted to model, analyze, and optimize the performance of a WGSMR in commercial-scale IGCC applications using a process simulator (*Aspen Plus*® V8.6) for the first time. Four models were created to analyze the influences of (i) pressure loss, (ii) direction of flow (i.e., co-current and counter-current), and (iii) operating parameters (i.e., sweep gas flow rate, sweep gas pressure, catalyst loading, and syngas inlet temperature) on CO conversions and H_2 yields. In the IGCC process, integration of a WGSMR depends on the membrane materials (polymeric, ceramic, or metallic); thus, non-metallic ceramic membranes (made from tetramethoxysilane (TOMS) [49]) were assumed to be used because they are inexpensive compared with metallic membranes and can operate around 300–400 °C.

2. Methodology and models

The main input data were temperature, pressure, molar flow rate,