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H_2 pressure swing adsorption for high pressure syngas from an integrated gasification combined cycle with a carbon capture process

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HIGHLIGHTS

- PSA processes were developed for H_2 recovery from high-pressure IGCC syngas.
- A layered bed using two adsorbents enhanced the separation efficiency of PSA process.
- A dynamic model predicted well the PSA results using a five-component syngas.
- The four-bed PSA improved recovery by 5–6% compared to the two-bed PSA.

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ABSTRACT

The integrated gasification combined cycle (IGCC) process, possessing high efficiency and environmental advantages, produces H_2-rich syngas at high pressures (30–35 bar) after capturing CO_2. Since the syngas pressure is very high for conventional PSA processes, development of an efficient PSA process at the pressure conditions is required for H_2 production. In this study, the H_2 PSA process for IGCC syngas was developed experimentally and theoretically. Breakthrough and PSA experiments using activated carbon or activated carbon/zeolite LiX were performed at 25–35 bar by using a five-component hydrogen mixture (H_2:CO:N_2:CO_2:Ar = 88:3:6:2:1 mol%) as a simulated syngas. The overall PSA performance was evaluated in terms of the purity, recovery and productivity of H_2 product. According to the results from using single or layered beds, the two-bed PSA process produced 99.77–99.95% H_2 with 73.30–77.64% recovery experimentally. A four-layered bed PSA at 35 bar was able to produce 99.97%-purity H_2 with 79% recovery, and it contained Ar and N_2 impurities. The quality of tail gas from the PSA process could be used for the gas turbine without losing H_2 and CO. A rigorous mathematical model that included mass, energy, and momentum balances was employed to elucidate the dynamic behaviors and separation performance of the adsorption bed and PSA process.

1. Introduction

The heavy reliance of modern industries on fossil fuels is a central obstacle to improving the air quality and preventing catastrophic climate change. Carbon dioxide that is emitted from coal-based thermal power plants all over the world is cited as one of the major sources of greenhouse gas. Naturally, hydrogen is considered as one of the most important energy carriers for the future, not only as a raw chemical material but also as a clean energy source. "Hydrogen can be produced from fossil fuels in stand-alone plants with CO_2 capture, but it may be advantageous to co-produce hydrogen and electricity. The flexible co-production plants could become increasingly attractive in future when electricity grids include a large proportion of variable renewable energy generation" [1]. An integrated gasification combined cycle (IGCC), that uses a gasifier to turn coal and other carbon-based fuels into syngas (synthesis gas) [2,3], allows the overall efficiency of the plant to be increased while producing both electricity and hydrogen simultaneously [4].

In reference to the Department of Energy's Clean Coal Initiative with carbon capture and sequestration (CCS), it is expected that an IGCC plant can be developed that has near-zero carbon emissions and high efficiency [5,6] because IGCCs generate a massive amount of H_2 after the CO_2 capture process [7,8]. IGCC can become one of dominant technologies in the power industry due to its ability to produce pure H_2 and to reduce carbon emissions [9]. Furthermore, an increasing demand for hydrogen provides an alternative market for IGCC syngas [10] because IGCCs generate a massive amount of H_2 after the CO_2 capture process. In addition, it is reported that

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Nomenclature

A	cross sectional area, cm^2
B	dual site Langmuir isotherm parameter, Pa^{-1}
C	concentration, mol/cm^3
C_p	heat capacity, cal/g K
D	diameter, cm
D_z	mass axial dispersion coefficient, cm^2/s
h	heat transfer coefficient, J/cm^2 s K
K	effective axial thermal conductivity, J/cm s K
k	parameter for the dual site Langmuir model or valve sizing coefficient
L	bed length, cm
P	pressure, pa
q, q^*, $\bar{q}_i$	amount adsorbed, equilibrium amount adsorbed, and average amount adsorbed, respectively, mol/g
q_m	dual site Langmuir isotherm parameter, mol/g
r or R	radius, cm, or gas constant, J/mol K
t	time, s
T	temperature, K
T_w	wall temperature, K
T_{atm}	ambient temperature, K
u	interstitial velocity, cm/s
y	mole fraction in gas phase
z	axial position in an adsorption bed, cm

Greek letters

ε	interparticle void fraction
μ	viscosity, cm/g s
ν	superficial velocity, cm/s
ρ	density, cm^3/g
ω	LDF coefficient, s^{-1}

Subscripts

b	bed
g	gas phase
i	inner bed
j	component j
o	outer bed
p	particle
s	solid phase
w	wall
z	axial

hydrogen production from just 1% of the produced syngas from a 500 MW IGCC plant is enough to provide the fuel needs of 10,000 vehicles because 1 kg of hydrogen is the same LHV (lower heat value) as 1 US gallon of gasoline (113,700 BTU). Hydrogen produced in this manner could well be the lowest-cost hydrogen because of the economies of scale provided by the large gasifiers in the IGCC plant [11].

The syngas produced by an IGCC plant has a value even if it is not needed immediately for electricity generation [9]. For example, the recovered H_2 can be supplied to generate the hybrid power of an H_2 turbine (Combined Cycle Gas Turbine: CCGT) and a fuel cell (Integrated Coal Gasification Fuel Cell: IGFC) for green power generation as a next generation IGCC process. Especially, some studies suggested that IGFC with CCS configuration can be a promising way to significantly decrease the greenhouse gas emission concerns of coal-based power plants because of the most economical H_2 production [12–15]. Therefore, academic and industrial consortium of the EU demonstrated a H_2-IGCC project [16]. The solid oxide fuel cell (SOFC) and molten carbonate fuel cell (MCFC), which are expected in the IGFC process, are not poisoned by carbon monoxide and carbon dioxide. And the hydrogen recovered from syngas after reforming/gasification can either be used directly within the IGFC process [17]. In addition, it was reported that IGFC using pressurized SOFC (more than 20 bar) has higher efficiency than that of atmospheric pressure SOFC [13]. Therefore, the issue is how to effectively supply high-pressure hydrogen of good quality at a low cost from IGCC syngas to the IGFC process. A fraction of the H_2-rich decarbonized syngas is fed to a pressure swing adsorption (PSA) unit. According to the suggested plant configuration, the tail gas from the PSA unit is compressed and combined with the remaining of the decarbonized syngas and combusted in the gas turbine [1,18].

Hydrogen plants are almost invariably designed using the pressure swing adsorption (PSA) process for final hydrogen purification. The PSA process is widely viewed as a promising way to recover H_2 from various effluent gases because of its high efficiency. Impurities from different crude H_2 streams can be removed by the selective adsorption of impurities on porous adsorbents such as zeolites and activated carbons. The quality of the hydrogen produced is a major issue for fuel cell [19] and other industrial application [20]. Therefore, many PSA process concepts have been reported as the state-of-the-art method for hydrogen separation/purification [21–29]. In addition, because of challenges relating to global warming, low-quality H_2 mixtures from various sources that have been frequently burned as thermal sources have also become attractive for use in various chemical processes via an efficient H_2 PSA separation process [27,30]. Furthermore, the use of PSA processes as purifiers could reportedly produce high-purity H_2 successfully with less than 10 ppm CO for fuel cell applications [31,32]. However, most studies on H_2 PSA for the production of 99.9–99.99+% H_2 have been focused on adsorption pressures of 10–20 bar because H_2 effluent gases from the present industrial fields are emitted in this pressure range; these effluents include reforming gas [21,22], coke oven gas [23,24], water gas shift reaction gas [27], and coal gas [31–33]. As a pre-combustion CO_2 capture, a PSA study was reported for CO_2 capture from H_2/CO_2 binary mixture at a high-pressure (about 34 bar) [34]. Generally, since minor impurities of weak adsorbates played a key role in process efficiency in PSA processes for purification, the PSA results using the feed gas, similar to a real gas composition, can significantly contribute to designing the corresponding real PSA process.

In reference to the National Energy Technology Laboratory, "There are several conventional H_2 separation processes, but modern installations preferentially choose pressure swing adsorption (PSA) near the feed pressure, which is a well-proven technology offering high availability and low cost" [35]. The effluent pressure of H_2 syngas from the IGCC process is expected to be close to 30–35 bar, even after CO_2 capture. PSA processes, which can be operated at the pressure range of IGCC syngas, are required in a cost-effective manner because the operation of PSA processes in their current state through the depressurization of the IGCC syngas is not desirable with respect to the overall capital and operating costs for the whole IGCC process.

Despite the importance of the efficient PSA process for the IGCC process, it is hard to find the studies for experimental and theoretical H_2 PSA processes because of high pressure of the carbon captured syngas. Recently, only one simulation study suggested that a multi-bed PSA with at least 12 columns containing a single