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GHGT-12

## A novel strategy to produce ultrapure hydrogen from coal with pre-combustion carbon capture

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### Abstract

Integrated Gasification Combined Cycles (IGCCs) are one of the emerging clean coal technologies which paves the way for producing power from coal with a higher net power efficiency than conventional PC-fired boiler power plants. It is also advantageous that in an IGCC power plant a carbon capture unit can be applied to a stream having a very high CO<sub>2</sub> partial pressure upstream of gas combustion that would not be available in case of a PC-fired boiler power plant, leading to less energy penalty involved in the carbon capture. In this study it is aimed to design a cogeneration process where a Hydrogen Pressure Swing Adsorption (H<sub>2</sub> PSA) unit is retrofitted to an IGCC power plant with pre-combustion capture for producing ultrapure hydrogen (99.99+ vol%). The ultrapure hydrogen is commonly utilised as feedstock for deep desulphurisation and hydrocracking units at refineries as well as H<sub>2</sub> fuel cells. It is found that, at the same H<sub>2</sub> purity of 99.99+%, the hydrogen recovery could be improved up to 93% with the increasing number of columns. Improving the H<sub>2</sub> recovery at the H<sub>2</sub> PSA to its maximum can contribute to reducing the power consumption for compressing the H<sub>2</sub> PSA tail gas by minimizing the yield of the H<sub>2</sub> PSA tail gas by-product. Furthermore, it is demonstrated that the H<sub>2</sub> PSA can also be designed to achieve 90% H<sub>2</sub> recovery even when a portion of the tail gas is recycled to the shift reactors in order to improve the overall advanced IGCC performance by increasing the H<sub>2</sub> yield and by reducing the auxiliary power consumption at carbon capture unit.

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*Keywords:* IGCC; Ultrapure hydrogen; Cogeneration; Pressure swing adsorption; Process simulation

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## 1. Introduction

Strong dependency on crude oil and natural gas and their associated soaring price and supply chain risk increase the need for efficient utilization of fossil fuel energy sources of being exhausted. The rising concentration in the atmosphere of various pollutants known as greenhouse gases (GHG) is identified as one of the key factors contributing to global warming effect. However, it is agreed that the fossil fuels will retain a major position in supplying heat and power in the near future before alternative technologies using renewable sources become mature enough to be substituted for conventional fossil fuels. In this respect, CCSU (Carbon Capture, Storage, and Utilisation) research has been stimulated as a pre-emptive way of sustaining fossil-fuel based economic growth without devastating the environment. Among various industrial CO<sub>2</sub> emitters, it is anticipated that the first commercial CCSU plant will be deployed to one of the fossil fuels fed power plants that currently account for approximately 30% of the global anthropogenic CO<sub>2</sub> emission [1].

The UK has set in its 2008 Climate Change Act a target to reduce its GHG emission up to 80% of 1990 levels in 2050, which will lead to allowed emissions of 150 MtCO<sub>2</sub>e per year. This target can be met only if all the industries including refining and petrochemical plants, cement plants, iron and steel manufacture as well as power stations are decarbonised. As regards refining and petrochemical industries, the Committee on Climate Change (CCC) estimated that the abatement of CO<sub>2</sub> emission would be possible partly by improving the energy efficiency of refining and petrochemical industries [2]. In addition it also proposed further reduction in CO<sub>2</sub> emission would be available by deploying carbon capture units on H<sub>2</sub> plants and replacing combustion fuels with carbon-neutral biomass. In particular, most of refining complexes need to increase their hydrogen production capacities to cope with the increased H<sub>2</sub> demand for their hydrotreating desulphurisation process that removes mainly sulphur and other impurities and hydrocracking units, which is to upgrade low-grade heavy residues to more valuable diesel and lube base oil. Given imminent carbon emission regulation, however, it is doubtful that conventional Steam Methane Reforming (SMR) process would be still the best option to produce ultrapure hydrogen.

In this study, it is aimed to design an advanced Integrated Gasification Combined Cycle (IGCC) to produce power and ultrapure hydrogen simultaneously from the coal where CO<sub>2</sub> is captured inherently for producing the ultrapure H<sub>2</sub>. It has been reported that coal is reserved most abundantly and is distributed most evenly over the globe among the fossil fuels [3]. IGCC plants can be potentially operated with other carbonaceous feedstock such as biomass, low quality petroleum residues and MSW.

Regardless of choice of a process producing synthetic gas, it is well-known that a pressure swing adsorption (PSA) is the only economically feasible, commercialised separation process to produce ultrapure hydrogen (99.99+ vol.%) from a synthetic gas. Various works have been published on hydrogen purification using H<sub>2</sub> PSA but they have been mainly based on feed gases originated from SMR reactors and COGs [4-9]. Therefore, it is required to redesign the H<sub>2</sub> PSA based on the condition of feed gas originated from coal gasifiers.

In this study, a H<sub>2</sub> PSA has been simulated using an in-house dynamic simulator while an exemplary IGCC plant is simulated using Honeywell UniSim Design R400. The in-house H<sub>2</sub> PSA simulator was successfully imported into Honeywell UniSim environment and integrated with the IGCC simulation that enables us to carry out various process configuration studies. The aim of this work is to design a cogeneration process capable to produce ultrapure hydrogen (100 H<sub>2</sub> MMSCFD) and power achieving 90% carbon capture.

## 2. Integrated gasification combined cycle (IGCC) with pre-combustion carbon capture simulation

Firstly a process simulation for a conventional IGCC power plant integrated with a pre-combustion carbon capture has been constructed using Honeywell UniSim R400 based on DOE report [10] in order to estimate the net power generation. Subsequently the IGCC power plant simulation is modified to an advanced IGCC plant to produce power and ultrapure hydrogen simultaneously (Figure 1). In the advanced IGCC plant, the  $H_2$ -rich fuel gas is split into two streams and one of two is fed to a  $H_2$  PSA for producing ultrapure hydrogen. The split ratio is determined so as to generate the ultrapure hydrogen at the flowrate of 100 MMSCFD. The  $H_2$  PSA tail gas needs to be compressed up to the pressure of the  $H_2$  fuel gas for feeding it to a high pressure gas cycle. This strategy of utilising the  $H_2$  PSA tail gas can augment the power generation at the combined cycle. Therefore, the net power generation from the advanced IGCC should take into account additional power consumption at the  $H_2$  PSA tail gas compressors and power generation at the combined cycle relating to the  $H_2$  PSA tail gas in addition to the  $H_2$  fuel gas.

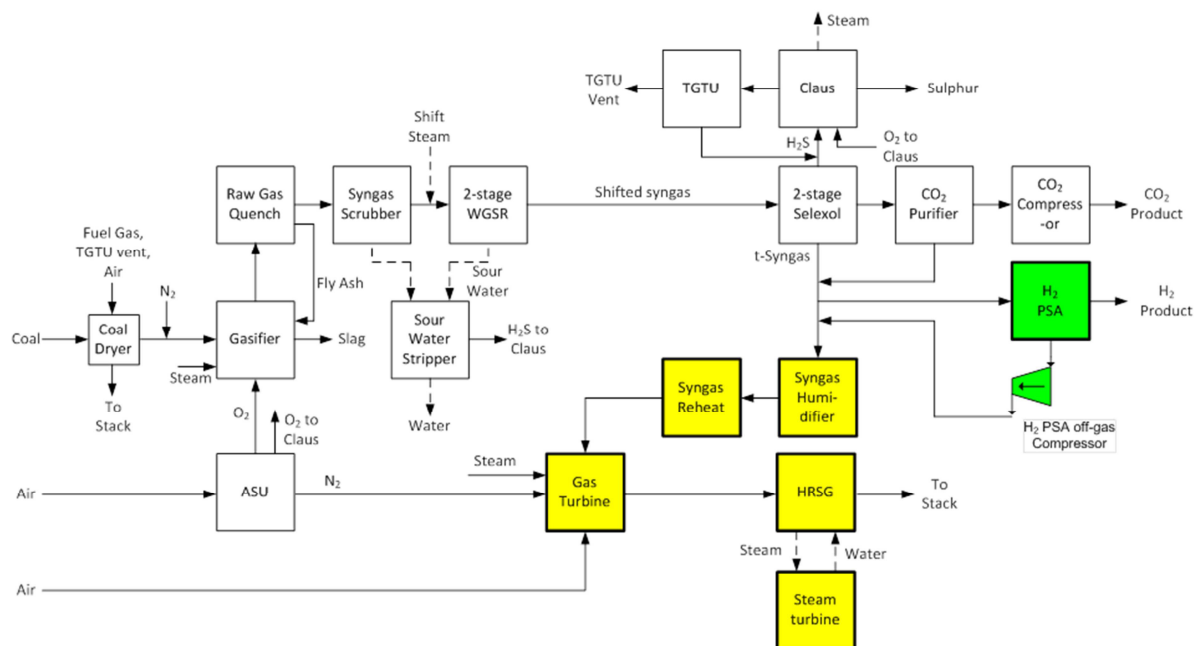


Fig. 1. Block flow diagram of an advanced IGCC plant to produce power and ultrapure hydrogen.

### 2.1. Gasifier and Syngas Cooler

The Shell gasifier, fed by the Illinois No.6 bituminous dry coal transported by nitrogen, 95% oxygen with nitrogen and argon balanced, and 400°C steam, was simulated with conversion reactor where the conversion rate of each reaction was adjusted to match the mass balance as reported in the reference [11]. The Shell gasifier operates at 1,424°C and 4.2 MPa producing some HP steam in the steam jacket with the carbon conversion rate set as 99.5%. An Elevated Pressure (EP) ASU produces 95% oxygen from air for its use in the gasifier and the Claus sulphur plant. The hot syngas from the gasifier is subsequently quenched by water to have the syngas contain steam. After the water quench, the syngas is cooled to generate the HP and IP steams in series and then flows to the syngas scrubber where water-soluble impurities are removed.

### 2.2. Water Gas Shift Reactors (WGSR)

Additional steam is added to this stream before being fed to the two shift reactors in series operating at the high

and low temperatures, respectively. Given the operating conditions of the two shift reactors (Gibbs reactor), the CO conversion rate was estimated to be 95.7% in total that is consistent with the reference study. It should be noted that the Shell gasifier generates a syngas having around 0.5 of H<sub>2</sub>/CO ratio compared to around 1.0 in the GEE gasifier, so the Shell IGCC would require greater amount of CO to be converted to CO<sub>2</sub> in order to achieve 90% carbon capture. The higher load in shift reaction requires more steam consumption in shift reaction, obviously leading to higher energy penalty in the Shell IGCC when it is integrated with carbon capture unit [11].

### 2.3. Acid Gas Removal (AGR) Unit

The syngas stream from the shift reactors is fed to a dual-stage Selexol unit to recover CO<sub>2</sub> and H<sub>2</sub>S separately from the syngas. This commercialised acid gas removal (AGR) process has two absorbers where the lean solvent from the steam stripper is fed to a CO<sub>2</sub> absorber and then part of the CO<sub>2</sub>-laden solvent subsequently captures more reactive H<sub>2</sub>S in a H<sub>2</sub>S absorber. While the H<sub>2</sub>S rich solvent is regenerated by the steam stripper, pure CO<sub>2</sub> comes off the CO<sub>2</sub>-laden solvent by successive two flash drums at the different pressures. The treated syngas from the AGR unit becomes saturated with water in a fuel gas saturation column and then is fed to combustion chamber. But in case of advanced IGCC plant the treated syngas is split into two streams: one stream flows to the gas turbine and the other flows to a H<sub>2</sub> PSA. The treated syngas is composed of 88.75% H<sub>2</sub>, 2.12% CO<sub>2</sub>, 2.66% CO, 5.44% N<sub>2</sub>, 1.03% Ar at 34 bar in this study. The initial simulations based on the values of the H<sub>2</sub>S and CO<sub>2</sub> solubilities in Selexol included in the UniSim Design database led to inconsistent results. Gas-liquid equilibria have been improved modifying the Henry constants obtained by regressing experimental data found in literature [12,13].

### 2.4. Combined Cycle

Two identical advanced F class gas turbines, each having 232 MWe of net power generation, were taken into account where the combustion chamber is simulated with an adiabatic Gibbs reactor. The exhaust flue gas, exiting the gas turbine at 602°C, enters the Heat Recovery Steam Generator (HRSG) to recover the large quantity of thermal energy that it contains. The steam cycle was designed with 12.4MPa/538°C/538°C of which the HP and IP temperatures are slightly lower than those in non-capture case due to the lowered heating value of syngas. The flue gas finally exits the HRSG at 132 °C and is sent to the atmosphere.

## 3. Design of H<sub>2</sub> PSA at advanced IGCC

The hydrogen PSA unit produces ultrapure hydrogen (99.99+ vol%) at 34 bar while it discharges the tail gas at 1 bar. Since the H<sub>2</sub> PSA tail gas contains a considerable amount of hydrogen and CO, it should be utilised for power generation by feeding it to a combined cycle. To do this, the H<sub>2</sub> PSA tail gas needs to be recompressed before it being sent to a high pressure combustion chamber. Given the fact that the tail gas compression involves significant power consumption, it is essential to increase the hydrogen recovery at the H<sub>2</sub> PSA close to its maximum in order to minimise the required H<sub>2</sub> PSA feed flowrate and the tail gas compression work at the same time. Once the mass balance around the H<sub>2</sub> PSA is determined based on the required product flowrate and the maximum H<sub>2</sub> recovery achievable, it is possible to estimate net power generation taking into account the tail gas compression work.

In our previous work [14], an in-depth study on design of H<sub>2</sub> PSA integrated with an IGCC power plant was carried out to maximize H<sub>2</sub> recovery at the H<sub>2</sub> PSA unit. The study demonstrated that the H<sub>2</sub> recovery could be increased by increasing the complexity of the PSA step configuration that enables a PSA cycle to have a lower feed flow to one column for adsorption and to accommodate more pressure equalization steps. In the study, the column dimensions were kept constant in all the PSA configurations so the total cycle time increases considerably and the bed productivity decreases with increasing number of columns from four to twelve. This is because more than one column undergo simultaneously the adsorption step so the total flowrate is reduced to half or one third. In this work the PSA simulations were performed at constant total cycle time of 1200 s regardless of the various configurations. As a result, the bed productivity can be almost kept constant. Note that with the increasing number of columns, the volume of one column should be reduced at the constant total cycle time but the ratio of column length to diameter is kept constant at 20.

Table 1. Effect of the one-column residence time during adsorption step on hydrogen purity, recovery and productivity at different H<sub>2</sub> PSA configurations.

One-Column Residence time [m <sup>3</sup> ·s/mol]	Column length [m]*	Column diameter [m]*	H <sub>2</sub> purity [%]	H <sub>2</sub> recovery [%]	H <sub>2</sub> productivity [mol <sub>H<sub>2</sub></sub> /kg <sub>ads</sub> /day]
Four-Column PSA with two-stage pressure equalisation					
0.100	0.467	0.0233	99.979	78.25	218.39
0.1225	0.500	0.0250	99.993	75.28	171.51
0.150	0.535	0.0267	99.996	71.85	133.69
0.175	0.563	0.0281	99.998	68.42	109.12
Six-Column PSA with two-stage pressure equalisation					
0.100	0.371	0.0185	99.976	86.38	321.44
0.1225	0.400	0.0200	99.993	83.43	253.44
0.150	0.424	0.0212	99.997	80.32	199.26
0.175	0.447	0.0223	99.999	75.93	161.46
Nine-Column PSA with three-stage pressure equalisation					
0.100	0.324	0.0162	99.971	92.79	345.12
0.1225	0.346	0.0173	99.992	91.48	277.78
0.150	0.371	0.0185	99.995	89.71	222.56
0.175	0.390	0.0195	99.998	86.96	184.39
Twelve-Column PSA with four-stage pressure equalisation					
0.100	0.324	0.0162	99.983	93.41	260.57
0.110	0.334	0.0167	99.992	92.94	235.92
0.1225	0.346	0.0173	99.994	92.52	210.71
0.150	0.371	0.0185	99.997	90.48	168.35

\*Note that the column dimensions were estimated based on the feed flowrate scaled down with a factor of 10<sup>-6</sup>.

It has been explored at which one-column residence time during the adsorption step ( $\tau = V/F$ ) the H<sub>2</sub> PSA can achieve the target H<sub>2</sub> purity of 99.99+ vol%. Table 1 shows the effect of residence time on the performance of each H<sub>2</sub> PSA configuration with respect to hydrogen purity, recovery and productivity (see [14] for step configuration of each H<sub>2</sub> PSA configuration). It is expected that with increasing residence time the H<sub>2</sub> purity can be improved as a sacrifice of H<sub>2</sub> recovery and productivity. In the four-, six- and nine-column systems the targeted H<sub>2</sub> purity is obtained with a H<sub>2</sub> PSA run having a column size to give a residence time in the vicinity of 0.1225 m<sup>3</sup>·s/mol. This is because as the feed flowrate to one column during adsorption step is reduced from total feed flowrate at the four-column PSA to the one third at the nine-column PSA through the one half at the six-column PSA, the required volume of one adsorption column is reduced almost at the same ratio. However, the residence time is notably reduced to 0.110 m<sup>3</sup>·s/mol from nine-column PSA to twelve-column PSA since the feed flowrate to one column during adsorption step does not change but the adsorption step time decreases from the one third to the one fourth of cycle time, i.e. the total amount of feed to be treated by one column during adsorption step is reduced.

It is clearly demonstrated that given the targeted H<sub>2</sub> purity of 99.99+vol% the H<sub>2</sub> recovery increases from 75% to 93% with increasing number of columns as shown in Tables 1 and 2. Along with the change in the H<sub>2</sub> recovery, the required ratio of H<sub>2</sub> PSA feed to total H<sub>2</sub> fuel gas and the required tail gas compression work are also changed significantly. As the ultrapure hydrogen product rate was determined a priori, the required feed flowrate for the H<sub>2</sub> PSA decreases with the increasing H<sub>2</sub> recovery and simultaneously the tail gas compression work decreases as well (Table 2). Moreover, the lower PSA feed flowrate resulting from the higher H<sub>2</sub> recovery can contribute to augmented power generation at the combined cycle since more H<sub>2</sub> fuel gas can be sent to the gas turbine at constant coal feed rate.

Table 2. Flowrate ratios of PSA feed to total H<sub>2</sub> fuel gas and tail gas compression works among various H<sub>2</sub> PSA configurations at the condition of 99.99+vol% H<sub>2</sub> purity and total cycle time of 1200 s.

H <sub>2</sub> PSA configuration	H <sub>2</sub> recovery [%]	Ratio of H <sub>2</sub> PSA feed to total H <sub>2</sub> fuel gas [%]	Tail gas compression work [MW]
Four-Column PSA with two-stage pressure equalisation	75.28	41.85	8.08
Six-Column PSA with two-stage pressure equalisation	83.43	37.62	5.70
Nine-Column PSA with three-stage pressure equalisation	91.48	34.43	3.82
Twelve-Column PSA with four-stage pressure equalisation	92.94	33.91	3.52

#### 4. Improving the performance by recycling H<sub>2</sub> PSA tail gas to shift reactors

It has been reported that the ultrapure hydrogen yield could be improved by recycling the H<sub>2</sub> PSA tail gas to shift reactors [15] as shown in Figure 2. This is because the CO contained in the tail gas can be converted to CO<sub>2</sub> and H<sub>2</sub> in reaction with steam. This modification can also lead to reduce the energy penalty involved in carbon capture.

However, it should be noted that it is not possible to recycle the entire tail gas to the shift reactors due to a build-up of impurities in the recycle loop being proportional to the amount of tail gas recycle. It implies that it is essential to bleed a portion of the tail gas out of the recycle loop by sending it to the gas turbine as shown in Figure 2. Therefore, it is important to see if the H<sub>2</sub> PSA can also be designed to achieve as high as 90+% H<sub>2</sub> recovery even with the lowered H<sub>2</sub> mole fraction in the feed resulting from the recycle of a certain amount of the tail gas to the shift reactors.

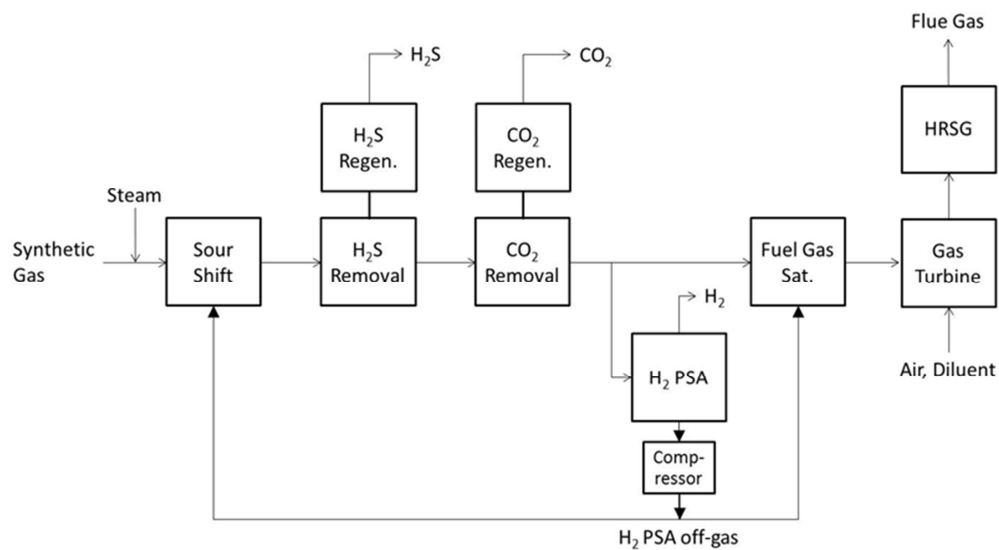


Fig. 2. Block flow diagram of an advanced IGCC plant with a recycle of H<sub>2</sub> PSA tail gas to shift reactors.



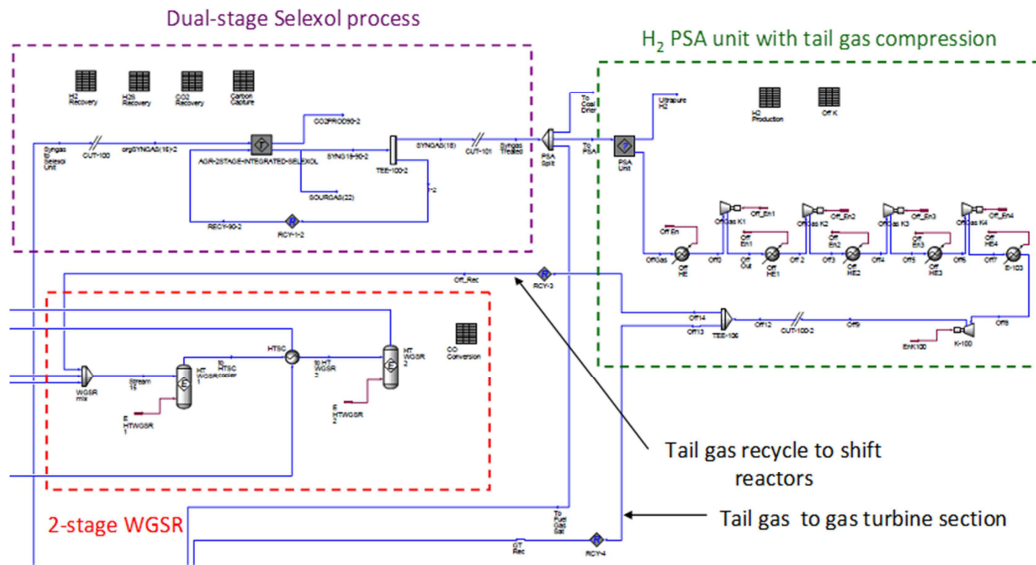


Fig. 3. UniSim PFD of an advanced IGCC plant with a recycle of H<sub>2</sub> PSA tail gas to shift reactors where the dynamic simulation (H<sub>2</sub> PSA with tail gas recycle) is integrated with Honeywell UniSim simulation (IGCC power plant simulation).

In this study, the adsorption column volume of the nine-column H<sub>2</sub> PSA with the total cycle time of 1200 s is sought to give the targeted 99.99 vol% H<sub>2</sub> purity when the H<sub>2</sub> mole fraction in feed is lowered to 0.85 due to the tail gas recycle. To do this study, it is prerequisite to construct an integrated process simulation where the in-house dynamic H<sub>2</sub> PSA simulator is imported into the UniSim IGCC simulation as shown in Figure 3. As a result of the simulation study, it is estimated that the one-column residence time should be increased to 0.140 m<sup>3</sup>·s/mol from 0.1225 m<sup>3</sup>·s/mol, i.e. the H<sub>2</sub> PSA unit should be designed with a larger column to accommodate the higher feed flowrate and to remove more impurities. Accordingly the H<sub>2</sub> recovery is lowered to around 90% from 91.5% due to the increase in the column volume.

## 5. Conclusions

A detailed simulation of an advanced IGCC plant to produce power and ultrapure hydrogen simultaneously where CO<sub>2</sub> is intrinsically captured by a pre-combustion capture has been proposed in this study. The H<sub>2</sub> PSA is designed such that its H<sub>2</sub> recovery can be increased to its maximum in order to avoid the excessive power consumption involved in tail gas compression and minimise the H<sub>2</sub> PSA feed gas flowrate.

The advanced IGCC for cogeneration can be improved by implementing the tail gas recycle to shift reactors since it can improve the H<sub>2</sub> product yield and the overall power generation and can alleviate the power consumption at pre-combustion carbon capture process.

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