Application of Aspen HYSYS for Predicting the Effects of Impurities on Thermodynamic Performance of Glycerol Autothermal Reforming for Hydrogen Production

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ABSTRACT

Hydrogen production from glycerol via autothermal reforming (ATR) has been widely investigated. However, little is known about the influence of impurities in glycerol on thermodynamic performance of the process. This study focused on the effects of impurities in glycerol on hydrogen productivity, energetic and exergetic efficiencies. The model of the entire process was simulated under thermoneutral condition in Aspen HYSYS using pure glycerol (PG) and crude glycerol (CG) as feeds. The two cases were optimized for maximum hydrogen production. From the optimized results, the hydrogen production per mole of the feed was 4.937 and 6.160 for the case of PG and CG, respectively. The thermal and exergetic efficiency of PG as feed were computed as 79.51% and 57.04% while that of CG were obtained to be 77.7 and 54.08%, respectively. The exergy destroyed to produce 1 mole of H$_2$ was found to be 133.5kJ and 157.3kJ for the case of PG and CG, respectively. It could be concluded that the presence of other constituents in CG contributed to increase in hydrogen productivity by increasing the energy demand of the plant but due to increase in both energy and exergy input, they decrease both the thermal and exergetic efficiencies.

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INTRODUCTION

The continuous release of greenhouse gases into the atmosphere and sudden change in climate conditions causing detrimental effect on our environment accounted for growing research in finding energy source that can replace fossil fuels. It has been recognized in the past that hydrogen as an energy carrier has great potential to replace fossil fuel in the future. It was stated in Kapdan and Kargi [1] that a clean energy carrier yields 2.75tons more energy than hydrocarbon fuel. The feedstock for hydrogen production can either be fossil fuel source (coal and natural gas) or alternative source (solar, nuclear and biomass). However, environmental friendly hydrogen production especially from bio-renewable feedstocks is required as mitigating technology. There is dramatic increase in crude glycerol production due to rapid expansion of biodiesel industry. Purified glycerol is used in cosmetics, food and pharmaceutical industries but high purification cost and availability of excess crude glycerol in the market made biodiesel producers to seek for alternative method for its utilization. One of the successive routes considered is the use of crude glycerol for hydrogen production. Biodiesel and glycerol are produced when vegetable oil and fats with alcohol in the presence of homogenous catalyst such as sodium or potassium hydroxide undergo transesterification and hydrolysis [2]. CG contains methanol, free fatty acid, water, glycerides, soap and fatty acids methyl esters as impurities [3].

Various techniques such as steam reforming, partial oxidation, dry reforming, aqueous phase reforming and ATR have been considered for hydrogen production [4–8]. In autothermal reforming, the required energy for the necessary reactions to take place is supplied by exothermic oxidation reaction. Therefore, the reforming technology considered for simulation in this study was autothermal reforming due to its energy efficiency and process stability. For these reasons, many works have been centered on improving hydrogen productivity in ATR of glycerol.

A thermodynamic analysis of autothermal reforming considering crude glycerol as a mixture glycerol and methanol using HYSYS was carried out by Authayanun et al. [9]. The result they obtained showed better performance when the ratio of glycerol to methanol increases. Ortiz et al. [10] conducted a thermodynamic analysis of glycerol autothermal...
reforming within a pressure range of 200-300 atm for pure and pre-treated glycerol using Gibbs free energy minimization approach. Under thermoneutral conditions, the optimum operating parameters were 0.407, 800°C and 99 for oxygen-glycerol, temperature and steam-glycerol, respectively. Abdulgani [11] in his research carried out production of renewable hydrogen from synthetics crude glycerol through ATR process. The best operating conditions were found experimentally in a packed bed reactor to be a reforming temperature of 575°C, steam-glycerol of 2.6, and oxygen-glycerol of 0.125. Many studies have been devoted to thermodynamic analysis of glycerol ATR process using Gibbs minimization technique [12–14]. Jimmy et al. [14] worked on thermodynamic modeling of hydrogen production from autothermal reforming of synthetic crude glycerol using Gibbs free energy minimization method. The optimum operating conditions under thermoneutral condition was obtained at 3.6, 0.75 and 927K for steam-crude glycerol ratio, oxygen-crude glycerol ratio and adiabatic temperature, respectively. Da Silva et al. [15] focused their study on optimization of the operating conditions of autothermal reforming of glycerol for hydrogen production for fuel cell using HYSYS as a process simulator. The simulation result showed that an optimum air to feed and steam to feed ratios of 5.5 and 3.5, respectively produced 34.7% of hydrogen.

In recent decades, the use of both energy and exergy analysis for assessing the performance of a system has attracted the interest of the researchers. Using various assessment tools, Hajjaji et al. [16] studied the process simulation of production of hydrogen via autothermal reforming of PG. They obtained 78.7 and 67.8% as the optimum thermal and exergetic efficiencies, respectively. They also recommended S/G=5.5, O/G=0.96, T=900K as the optimum conditions for the entire process of hydrogen production. However, most of the works carried out in this field is concentrated on evaluation of thermodynamic performance of pure glycerol ATR reactor and/or catalysis in the system, but energetic and exergetic assessment of the entire process of hydrogen production from crude glycerol through ATR is very scarce in literature.

This research work is aimed to determine the effect of impurities in crude glycerol on thermal efficiency, exergetic efficiency and hydrogen productivity of glycerol autothermal reforming process. This was achieved by modeling and thermodynamic analysis of an optimized system of hydrogen production from both PG and CG through autothermal reforming.

**MATERIAL AND METHODS**

In this study, hydrogen production via autothermal reforming using both PG and CG as feeds was investigated. Aspen Hysys was used to design and simulate the entire hydrogen production process. Peng-Robinson Struyk-Vera (PRSV) equation of state was used to determine the physical and transport properties of the streams in the model. The simulation results were then optimized before hydrogen productivity, energetic and exergetic performances of the system were determined. The effect of the impurities was determined by comparing the simulation results and thermodynamic performance of the reforming processes using the two feeds.

**Model development and process simulation**

A simplified flow diagram for hydrogen production from glycerol via ATR process is shown in Figure 1. The Gibbs’ reactor considered as autothermal reformer was used to determine the equilibrium composition of the gas products. The possible reactions in the autothermal reactor are as follow:

- **Steam reforming**
  \[ \text{C}_3\text{H}_5\text{O}_3 + 3\text{H}_2\text{O} \rightarrow 7\text{H}_2 + 3\text{CO}_2 \quad \Delta H_{298K} = 127.67 \text{ kJ/mol} \]  

- **Partial oxidation**
  \[ \text{C}_3\text{H}_5\text{O}_3 + 1.5\text{O}_2 \rightarrow 4\text{H}_2 + 3\text{CO}_2 \quad \Delta H_{298K} = -603.5 \text{ kJ/mol} \]  

- **Water gas shift**
  \[ \text{CO} + 2\text{H}_2 \rightarrow \text{H}_2 + \text{CO}_2 \quad \Delta H_{298K} = -41.8 \text{ kJ/mol} \]  

**Glycerol reforming**

- \[ \text{C}_3\text{H}_6\text{O}_3 + \text{H}_2 + 0.5\text{O}_2 \rightarrow 2\text{CO}_2 + \text{CO} + 5\text{H}_2 \quad \Delta H_{298K} = -78.3 \text{ kJ/mol} \]  

**Methanol reforming**

- \[ \text{CH}_3\text{OH} + 0.5\text{H}_2\text{O} + 0.125\text{O}_2 \leftrightarrow 0.5\text{CO}_2 + 0.5\text{CO} + 2.25\text{H}_2 \quad \Delta H_{298K} = 9.6 \text{ kJ/mol} \]  

**Preferential oxidation**

- \[ \text{CO} + 0.5\text{O}_2 \rightarrow \text{CO}_2 \quad \Delta H_{298K} = -570\text{kJ/mol} \]  

The CO clean up section was represented in the process flow diagram by three shift reactors modeled by Gibbs reactors and preferential oxidation reactor (COPOX) modeled by conversion reactor. The three shift reactors and their temperature ranges are as follow: High Temperature Shift reactor (HTSR, 300-450°C), Moderate Temperature Shift reactor (MTSR, 220-270°C) and Low Temperature Shift reactor (LTSR, 180-250°C). The reaction taking place in the shift reactors is shown in Equation (7) while that of COPOX is given in Equations (14) and (15). In COPOX, the stream exiting the LTSR is mixed with air and CO is selectively
oxide CO₂ in the presence of noble metal [17]. However, small amount of hydrogen oxidizes to water [18]. In this work, the molar concentration of CO lower than the maximum allowable limit (10ppm) for fuel cell application was considered by varying the air molar flow rate with 2% hydrogen conversion. Heat exchangers are installed between the reactors because of the exothermic nature of the reaction. The purification section is solely based on pressure swing adsorption modeled as component splitter. The hydrogen rich gases exiting CO clean section was separated into hydrogen stream with 99.98% percentage purity and unused gases in the purification section. The process was designed by taking the specified temperature range of the reactors into consideration. The process model and the analysis were carried out on the following assumptions

- All the components possess adiabatic boundaries.
- Kinetic and potential exergy are overlooked for all system components.
- The reference temperature and pressure are 25°C and 1atm.
- Air is considered as an ideal gas with molar concentration of 21% Oxygen and 79% Nitrogen.
- The feeds conditions are at standard temperature and pressure.
- Formation of coke is overlooked due to negligible concentration.
- S/CG and A/CG of 3.5 and 5.5 are considered, respectively.
- The use of catalyst is not considered.
- The outlet stream temperature of the reactors is considered as reactor’s temperature.

The compositions of crude glycerol depend on the raw materials and processes used in biodiesel plant. The input values for the simulation of the base case are presented in Table 1. The crude glycerol composition is based on synthetic crude glycerol composition reported in literature [11].

| TABLE I. Feed compositions for the base case (CG) |
|-----------------|----------------|----------|----------|
| CG constituents | Glycerol | Methanol | Soap | Oleic acid |
| Composition (weight %) | 0.51 | 0.13 | 0.32 | 0.04 |

**Process optimization**

The whole process was optimized using the optimizer tool in Aspen HYSYS for maximum hydrogen production at a constant flow rate of both PG and CG. The optimization problem is represented mathematically as given in Equations (16) – (29).

\[
F(x) = \max(M_{H_2}^{out})
\]

The problem was subject to the following constraints

\[
300 < \text{stream 3} < 450 \text{ °C}
\]

\[
220 < \text{stream 5} < 270 \text{ °C}
\]

\[
180 < \text{stream 7} < 450 \text{ °C}
\]

\[
\max(M_{H_2}^{out}) < 10 \text{ ppm}
\]

The temperature constraint illustrated in Equations (17) – (19) were considered in order to fulfill temperature constraints of the reactors. The component molar concentration of CO was assumed not to be greater than 10ppm for the purpose of fuel cell applications. The optimizer algorithm used for this case was sequential quadratic programming because of its ability to handle equality and inequality constraints. Also, the region where the optimal solution of this case was searched for is defined on each manipulated variable as given in Equations (21) – (23).

\[
300 < \text{Air} < 700 \text{ kmol/hr}
\]

\[
200 < \text{H}_2\text{O} < 500 \text{ kmol/hr}
\]

\[
10 < \text{Air2} < 80 \text{ kmolpp/hr}
\]

**Thermodynamic analysis**

The system performance based on first law of thermodynamics can be estimated by its thermal efficiency. It is expressed as the ratio of energy output to energy input as shown in Equation (24).

\[
\eta_{\text{thermal}} = \frac{M_i \times LHV_i + W_{\text{comp1}} + W_{\text{comp2}} + W_{\text{pump}}}{(M_i \times LHV_i)}
\]

where, \(M_i\) and \(LHV_i\) are the molar flow and the lower heating
value of feed “i” respectively, \( M_{H_2} \) and \( LHV_{H_2} \) are the molar flow and the lower heating value of hydrogen; \( W_{\text{comp}1} \), mechanical work of the first compressor; \( W_{\text{comp}2} \), mechanical work of the second compressor; \( W_{\text{pump}} \), mechanical work for feed pumping.

The exergy transfer associated with flow was calculated by adding the physical and chemical exergy of a particular stream.

The physical exergy, \( EX_{ph} \) of a stream is calculated by Equation (25).

\[
EX_{ph} = M[(H - H_o) - T_o(S - S_o)]
\]

where, \( M \) is the molar flow rate of a stream; \( H \) and \( S \), the specific enthalpy and entropy calculated at the stream condition; \( H_o \) and \( S_o \) are the specific enthalpy and entropy calculated at the atmospheric temperature\( (T_o) \) and pressure \( (P_o) \).

The chemical exergy, \( EX_{ch} \) of a substance not present in the environment is obtained by using Equation (26).

\[
EX_{ch} = -DG + \sum_n nEX_{ch} - \sum_R nEX_{ch}
\]

where, \( DG \) is the change in standard Gibbs’s free energy of the reaction; \( n \), number of mole of species taking part in the reaction; \( P \), product side of the reaction and \( R \), the reactant side of the product.

The chemical exergy \( EX_{ch} \) of a stream containing more than one component k of mole fraction X can be determined by Equation (27).

\[
EX_{ch} = \sum_k \{X_k \cdot EX_{ch} + RT_o \sum_k X_k ln(X_k)\}
\]

The exergy associated with heat transfer, \( EX_q \) and work transfer, \( EX_w \) is estimated by Equations (28) and (29), respectively.

\[
EX_q = (1 - T_o/T) Q
\]

\[
EX_w = \sum_n nEX_{ch}
\]

The internal exergy destruction, \( I \) within the control volume is calculated by using Equation (30).

\[
I = \sum EX_{in} - \sum EX_{out}
\]

\[
\sum EX_{in} = EX_{GLY} + EX_{WATER} + EX_{AIR} + EX_{W_{comp1}} + EX_{W_{comp2}} + W_{pump}
\]

\[
\sum EX_{out} = EX_{HYDRO} + EX_{unused}
\]

\[
EX_{unused} = EX_{WASTE} + I
\]

where, \( Q \) is the quantity of heat transfer; \( R \), gas constant; \( EX_{HYDRO} \), exergy flow of hydrogen rich stream; \( EX_{unused} \), exergy that leave the system; \( EX_{WASTE} \), exergy associated with waste stream; \( \sum EX_{in} \), total exergy flow into the system; \( \sum EX_{out} \), total exergy flowing out of the system; \( EX_{GLY} \), exergy associated with glycerol flow; \( EX_{WATER} \), flow exergy of water; \( EX_{AIR} \), flow exergy of air; \( EX_{W_{comp1}} \), \( EX_{W_{comp2}} \) and \( W_{pump} \) are the exergy flow associated with mechanical work for compressor 1, compressor 2 and feed pump respectively.

The exergetic efficiency of the overall system was evaluated by Equation (33).

\[
EX_{eff} = EX_{H_2}/\sum EX_{in}
\]

RESULTS AND DISCUSSION

Simulation results

The components molar flow rate of effluents from reformer and PSA using PG and CG as feeds are presented in Table 2. The hydrogen production was increased by 40% via shift and partial oxidation reactions installed for CO clean-up process. It is well established in the table that the CO concentration was reduced below the specified limit. Furthermore, the CO clean-up section was able to reduce hydrogen molar flow rate by 32.04% with 447.270 kg mole/h of it being produced by the reformer in the case of CG. It is well established in the table that more methane was generated in the process plant when CG was used as feed, this might be as result of the presence of alcohol and fatty acids in the feed. With these, the model developed is in conformity with the anticipated results.

Process optimization

From the results obtained, the base case possesses good hydrogen productivity relatively to values reported by previous researchers. However, the base case system still has potential for improvement in hydrogen production. The optimized results are presented in Table 3. The hydrogen production increase of 3.16% was accomplished by reducing the flow rate of the water, air to the reformer and air to the COPOX when PG was considered as feed. In the case of CG, the optimization results showed an increment of 3.62% at the detriment of increase in both air and water flow rate to the reformer.

Energy analysis

As shown in Table 4, the presence of impurities in CG caused 24.77% increment in hydrogen production. This occurs as a result of higher stoichiometric hydrogen content in the feed. Therefore, the higher the ratio of glycerol to methanol in CG, the higher the hydrogen productivity and vice-versa. This is

| TABLE 2. The component molar flow rate of effluents in the system |
|---------------------------------------------------------------|
| **Components (Kgmol/hr)** | **ATR** | **PSA** | **ATR** | **PSA** |
|---------------------------|---------|---------|---------|---------|
| CH₄                       | 0.365   | 0.365   | 17.527  | 17.527  |
| H₂O                       | 402.261 | 270.498 | 293.618 | 150.126 |
| CO                        | 143.957 | 0.003   | 162.922 | 0.011   |
| CO₂                       | 155.596 | 299.562 | 153.527 | 316.457 |
| H₂                        | 346.786 | 478.560 | 447.270 | 590.582 |
| N₂                        | 446.041 | 485.561 | 446.134 | 485.669 |
| GLY                       | 0.000   | 0.000   | -       | -       |
| CGLY                      | 0.000   | 0.000   | 0.000   | 0.000   |
| O₂                        | 0.000   | 0.000   | 0.000   | 0.000   |
To determine the influence of other constituents apart from glycerol present in CG on hydrogen productivity, thermal efficiency and energetic efficiency was explored under thermoneutral condition. The optimized results indicated a slight reduction of 1.73% in thermal efficiency was contributed to increase in power consumption for feed pumping and air compression. This consequently led to 5.06 increments in specific energy consumption. It is worth noting that this analysis was based on the optimized simulation results shown in Table 3. It is observed that the thermal efficiency is very close to the values reported in the literature; glycerol: 83.6%, [15] and gasoline: 82.84%, [19].

**Exergy analysis**

The exergetic efficiencies of the system were computed to be 57.04 and 54.08% for PG and CG, respectively. The exergetic efficiency of CG is about 3% less than that of PG. This might probably due to internal exergy loss as result of numerous reactions taking place during reforming process. The exergetic efficiency of PG is approximately equal to the value obtained by Hajjaji et al. [16] (57%). It is noticed that the exergetic efficiencies are less than thermal efficiencies. The inclusion of exergy destruction in the exergetic balance might responsible for the differences.

### Table 3. Results of the optimization study

| Objective Function | PG                  | CG                  |
|--------------------|---------------------|---------------------|
| M\(^{\text{ex}}\)\(_{\text{H}_2}\) (kgmole/hr) | 478.560  493.700   | 590.582  616.976   |
| Control Variables  |                     |                     |
| W/PG or W/CG       | 3.5  2.654          | 3.5  4.860          |
| A/PG or A/CG       | 5.5  4.752          | 5.5  6.331          |
| Air 2 (kgmole/hr)  | 50  33.25           | 50  36.740          |
| Reformers’ Parameters |                   |                     |
| P (kpa)            | 456  456            | 456  456            |
| TR (°C)            | 842.8  775.5        | 729  765.8          |

For the case of PG, an approximate value of 42.95% of the total exergy fed to the system is unused of which 74.51% of it disappears within the process plant as exergy destruction. With the CG as feed to the system, about 45.92% of the total exergy available for the process is not used. 77.79% of the exergy unused vanishes as internal exergy loss. It also indicated in Table 5 that 133.5kJ of exergy is associated with production of 1 mole of H\(_2\) when PG is considered as feed. In the case of CG, production of 1 mole of H\(_2\) caused 157.3kJ of internal exergy loss. These could be ascribed to exergy loss caused by mixing of streams in the reactor, chemical reactions and heat transfer with wide temperature difference. These values are comparatively higher than the values reported in the literature for SMR, 100.60kJ/mole H\(_2\) [20] and for ATR of glycerol, 98.82kJ/mole H\(_2\) [16]. This higher value might be as a result of PSA included in this model.

### Table 5. Summary of the results from the exergy analysis of the whole process plant

| Exergy exchanged                          | PG   | CG   |
|-------------------------------------------|------|------|
| Exergy in (kJ/moleH\(_2\))               | 417.5| 440.3|
| Exergy out (kJ/moleH\(_2\))              | 283.9| 283.0|
| Exergy destruction (kJ/moleH\(_2\))      | 133.5| 157.3|
| Exergy unused (kJ/moleH\(_2\))           | 179.3| 202.2|
| Exergy efficiency (%)                    | 57.04| 54.08|

### CONCLUSION

The simulation of ATR for hydrogen production was conducted using both PG and CG as feeds. The simulation model was optimized for maximum hydrogen production. The influence of other constituents apart from glycerol present in CG on hydrogen productivity, thermal efficiency and exergetic efficiency was investigated under thermoneutral condition. The optimized results indicated more generation of methane when CG was considered. It also shows that the presence of other constituents in CG contributed to increase in hydrogen productivity by increasing the energy demand of the plant but due to increase in both energy and exergy input, they decrease both the thermal and exergetic efficiencies.

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**Persian Abstract**

چکیده

تولید هیدروژن از گلیسرول از طریق اصلاح اوتومرال (ATR) به طور کستردهای مورد بررسی قرار گرفته است. با این حال، در مورد تأثیر ناخالصی‌های موجود در گلیسرول بی‌عملاً بر عملکرد نیترودنیزی فرآیند کمی در این مطالعه بر تأثیر ناخالصی‌های گلیسرول بر بهره‌وری هیدروژن، راندمان انرژی و گرزی به عنوان خروجی از گلیسرول خالص (PG) و گلیسرول خام (CG) استفاده از ATR HYSYS Aspen به سیستم شتاب‌دهنده و اثر در هر چاپک سایر غلظت هیدروژن به ترتیب ۱۴/۱۷و۱۳/۷۹/۱۴/۰۱/۴۵/۷۸ و ۱۳/۷۱/۱۴/۰۱/۴۵/۷۸ در حالی که به ترتیب ۱۲/۷۷و۱۳/۷۱/۱۴/۰۱/۴۵/۷۸ و ۱۳/۷۱/۱۴/۰۱/۴۵/۷۸ به ترتیب ۱۲/۷۷و۱۳/۷۱/۱۴/۰۱/۴۵/۷۸ و ۱۳/۷۱/۱۴/۰۱/۴۵/۷۸ به ترتیب ۱۲/۷۷و۱۳/۷۱/۱۴/۰۱/۴۵/۷۸ و ۱۳/۷۱/۱۴/۰۱/۴۵/۷۸ به ترتیب ۱۲/۷۷و۱۳/۷۱/۱۴/۰۱/۴۵/۷۸ و ۱۳/۷۱/۱۴/۰۱/۴۵/۷۸ به ترتیب ۱۲/۷۷و۱۳/۷۱/۱۴/۰۱/۴۵/۷۸ و ۱۳/۷۱/۱۴/۰۱/۴۵/۷۸ به ترتیب ۱۲/۷۷و۱۳/۷۱/۱۴/۰۱/۴۵/۷۸ و ۱۳/۷۱/۱۴/۰۱/۴۵/۷۸ به ترتیب ۱۲/۷۷و۱۳/۷۱/۱۴/۰۱/۴۵/۷۸ و ۱۳/۷۱/۱۴/۰۱/۴۵/۷۸ به ترتیب ۱۲/۷۷و۱۳/۷۱/۱۴/۰۱/۴۵/۷۸ و ۱۳/۷۱/۱۴/۰۱/۴۵/۷۸ به ترتیب ۱۲/۷۷و۱۳/۷۱/۱۴/۰۱/۴۵/۷۸ و ۱۳/۷۱/۱۴/۰۱/۴۵/۷۸ بنا بر گرفت که صحور سایر تکنیکات در CG به فازهای هیدروژن با الکتری شیمیایی نافذان اثر انرژی گیاه می‌شود، اما با توجه به الکتری شیمیایی انرژی و اکترزی، آن‌ها هم باید حزارتی و هم از نظر مصرف کنند از کاوشانه می‌دهند.