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Hydrogen-rich syngas production of urea blended with biobutanol by a thermodynamic analysis

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ABSTRACT

Both biobutanol and urea are the environment-friendly hydrogen carrier. This study is to compare hydrogen production between steam reforming of biobutanol and autothermal reforming of biobutanol feed using pure steam and vaporization of aqueous urea (VAU) by a thermodynamic analysis. Hydrogen-rich syngas production, carbon formation, thermal neutral temperature (TNT), and hydrogen production cost are analyzed in both steam reforming and autothermal reforming. The results show that hydrogen-rich syngas production with the use of VAU is higher than that with pure steam not only in steam reforming but also in autothermal reforming. When the VAU/butanol molar ratio is 8, and the O_2 /butanol molar ratio equals 3, the reforming efficiency reaches up to 81.42%. At the same condition, the hydrogen production cost is lower than that without blending urea. Therefore, using VAU to replace pure steam in biobutanol reforming leads to benefits of increasing the hydrogen-rich syngas yield and lowering cost.

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Introduction

Because carbon dioxide emissions are increasing rapidly, climate change and instability in the crude oil supply cause prices to continue to grow. More and more countries have put an emphasis on the development of biofuels in order to obtain alternative sources of energy to replace gasoline and diesel fuel. Biodiesel and hydrogen-treatment vegetable oil (HVO) are alternatives for fossil diesel and bio-alcohols such as bio-ethanol or biobutanol have been developed to reduce the demand for gasoline. Both bioethanol and biobutanol are made from biomass through the fermentation process. The most popular and highest production bio-alcohol is bioethanol, production of which was 117.7 million m^3 in 2016 globally [1]. On the other hand, production and application of biobutanol have been of great importance in the last few decades.

Biobutanol is the same as bioethanol in which it is produced via a fermentation process using microorganisms from biomass such as corn, wheat crops, and rice straw [2,3]. The formula for biobutanol is C_4H_9OH , and it has similar characteristics with butanol, which is produced from petroleum. Because biobutanol has longer carbon chain than ethanol, its chemical and physical properties are closer to gasoline than ethanol. Although its heating value ranges from 10% to 20% that is lower than that of gasoline, it is higher than that of other alternative fuels such as bioethanol. Moreover, it is a potential biofuel because it is more stable than bioethanol. Therefore, biobutanol is mainly used to blend with gasoline, and the blend ratio of Vol 12.5% has been put into the fuel standard of the United States government. Dernette J et al. [4] conducted a gasoline engine experimental study using biobutanol blended with gasoline, and their results showed that biobutanol could reduce emissions from gasoline engine

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vehicles. There were also some studies conducted to assess engine emissions using biobutanol blended with diesel, and their results also revealed that using biobutanol blended with diesel decreased soot and carbon monoxide emissions [5,6].

While bioethanol and biobutanol are used as alternative fuels in internal combustion engines, emissions and efficiency are still of great concerns because of the requirement to protect the environment [7]. Up to this point, the hydrogen energy fuel cell is known as the best technology to improve and reduce emissions [8,9]. Therefore, many countries all over the world have conducted the development and application of stationary and on-board fuel cell power generation. On-board fuel cell power generation has a higher technique threshold than a stationary system because of constraints related to volume and vibration problems. Moreover, the problems including hydrogen storage, transportation, supply infrastructure, and high costs are still severe limitations. Thus, the best way to produce hydrogen is considered to be a reforming process. Fuel reforming is widely applied to industry and fuel cell systems. For on-board applications, liquid phase fuels such as gasoline, diesel, and biofuel all can serve as reforming fuels, and the on-board fuel supply system can be used without any modifications. For example, Nissan Motor Company [10] announced that they had developed an electric vehicle with a SOFC running on bioethanol to provide electric for the extended range. The solid oxide fuel cells (SOFCs) need to be operated under high temperature condition and can be fed pure hydrogen as well as hydrogen-rich syngas (hydrogen and carbon monoxide) [11], which reduces the hydrogen purification device and decreases the application limitations. On the other hand, the energy density of a liquid fuel is higher than that of a gas phase fuel, including natural gas (methane), petroleum gas (propane) and ammonia. It is also easier to transport and store. Because of the carbon dioxide reduction demands, a couple of researchers have focused primarily on hydrogen production for reforming technologies using bio-fuels [12,13]. Biobutanol is the same as the butanol as a good hydrogen carrier because it comprises 13.5 wt% hydrogen. Therefore, butanol reforming has increasingly been the object of study in recent years. Nahar et al. [14] performed a thermodynamic analysis of steam reforming of butanol using the Gibbs free energy minimization method. Their results showed that the best operating parameters occurred when H_2O/C_4H_9OH molar ratios ranged from 9 to 12, and the highest syngas yield could be gained. The H_2 reached 81.27%, and CO was 65.48%. Horng et al. [15] also analyzed hydrogen-rich syngas production of the autothermal reforming of butanol using the Gibbs free energy minimization method. Their results revealed that when the H_2O/C_4H_9OH molar ratio equaled 3, and the O_2/C_4H_9OH molar ratio was 1.5, the H_2 yield was 59.3%, and the CO yield was 39.5%. Wang et al. [16] investigated the hydrogen production of dry reforming of butanol using a thermodynamic analysis, and they found that the highest H_2 concentration reached 57.34%–57.87% when $CO_2/butanol$ molar ratios ranged from 3.5 to 4 at reaction temperatures ranging between 1150 K and 1200 K. Huang et al. [17] conducted partial oxidation reforming of butanol using the Gibbs free energy minimization method, and their results showed that when the O_2/C_4H_9OH molar ratios ranged from 1.5 to 2.0, the highest H_2 production was gained. Harjua et al.

[18] carried out steam and autothermal reforming of n-butanol using an Rh/ZrO₂ catalyst. Their experimental results depicted that the carbon conversion could reach 100% at a reaction temperature of 700 °C, and the H_2 yield was 70% of the theoretical value. Hartley et al. [19] carried out steam and autothermal reforming of butanol using a thermodynamic analysis and an experiment. Their results revealed that the thermal neutral temperature occurred at 700 °C when the $O_2/butanol$ molar ratios ranged from 2.7 to 2.8, and the highest H_2 yield was 5.56 mol for a steam/butanol molar ratio of 12 at the same reaction temperature. In part of the experiment, the H_2 yield was very close to the theoretical value with an $O_2/butanol$ molar ratio of 2.7, and an $H_2O/butanol$ molar ratio of 9 at 700 °C using an Rh/Al₂O₃ catalyst. Bimbela et al. [20] also conducted steam reforming of butanol using a Ni–Al catalyst and found that the fuel conversion could reach 99.05% when the GHSV was equal to 57000 h⁻¹ at a reaction temperature of 750 °C. Cai et al. [21] used bimetallic catalysts on the autothermal reforming of butanol, and they identified the best composition of the catalyst in their research. The catalytic partial oxidation reforming of iso-butanol was determined in an article by Chakrabarti et al. [22]. Their results showed that for the catalytic partial oxidation reforming of iso-butanol without steam, both CO and H_2 molar fractions were higher than 70%; moreover, the H_2 molar fraction was able to reach as much as 103% for the catalytic partial oxidation reforming of iso-butanol with steam.

Urea is also known as the best hydrogen carrier, and it is an organic compound composite with carbon, nitrogen, oxygen, and hydrogen. It contains 6.7 wt% hydrogen, and its formula is CO(NH₂)₂ [23]. It is expected that urea production will increase to over 200 million tons by 2018, and over 90% of urea produced is used as a fertilizer [24]. There is a small percentage of urea used in vehicle applications as a NO_x reduction reactant reacting in a selective catalytic reduction system [25]. Urea is formed as either a powder or crystal that is non-flammable, non-toxic, stable, colorless, and tasteless. It is friendly to the environment and human health. It can dissolve into water, ethanol, and glycerin. Furthermore, urea contains hydrogen content of 6.7 wt%, and its safety properties satisfies the definition of a hydrogen carrier in transportation application according to the U.S. Department of Energy [26]. Therefore, more researchers are putting an emphasis on urea reforming as well as fuel cell applications. Lan et al. [27] utilized urea as a fuel for a fuel cell and generated power from fertilizer and waste. Cinti et al. [28] also evaluated urea applied to the SOFC system, and their experimental results depicted that SOFC system efficiency could reach over 40% by feeding syngas reformed from urea. In addition, Fan et al. [29] focused on a direct urea fuel cell, which utilized hydrogen peroxide as an oxidant. Zamfirescu et al. [30] carried out on-board urea reforming to produce hydrogen-rich syngas that was then fed into an engine. They heated solid urea to become a liquid, mixed it with steam, and then fed it into the reformer. Their results displayed that syngas from urea reforming could enhance engine combustion, reduce cost by 45%, and decrease greenhouse gas emissions by 64%. Rollinson et al. [31] conducted steam reforming of urea to produce hydrogen-rich syngas. They utilized a continuously fluidized bed reactor with a Ni-based catalytic and observed that the H_2 yield was

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