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## Enhancement of dilute bio-ethanol steam reforming for a proton exchange membrane fuel cell system by using methane as co-reactant: Performance and life cycle assessment



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#### ABSTRACT

The fuel processor and a proton exchange membrane fuel cell (PEMFC) integrated process fueled by cassava based bio-ethanol and methane as co-reactant is theoretically investigated and compared with that run by dehydrated bio-ethanol in this work. The methane is added to bio-ethanol reformer as co-reactant to reduce dilution effect of crude bio-ethanol and adjust very high steam to carbon ratio of this system. The hydrogen fraction increases with the reformer temperature and methane to bio-ethanol ratio until reaching a maximum point. In addition, the optimal operating conditions of mixed bio-ethanol and methane reformer and dehydrated bio-ethanol reformer, which achieve the highest reformer efficiency, are presented. The results show that superior fuel processor efficiency, fuel cell efficiency and system efficiency are obtained when the mixed bio-ethanol and methane is used to generate hydrogen. The mixed bio-ethanol and methane reforming integrated with PEMFC system has the lower environmental impact, compared to the dehydrated bio-ethanol reforming integrated with PEMFC system.

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#### Introduction

The increasing of energy demand and limitation of petroleum resource stimulate the research about alternative energy and clean technology. Fuel cell technology is much cleaner than the other conventional power generation systems because it directly converts fuel to electricity through an electrochemical process and produces only water and heat as by-products when hydrogen is used as fuel. Most of hydrogen is produced from natural gas through stream reforming process due to the high performance and economic of this process [1]. Because of increased power demand and environmental concerns, the development of a new sustainable feedstock for

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hydrogen production is necessary [2]. Considering environmental and availability aspect, the biomass and bio-based material are considered the attractive raw material for hydrogen production because of its renewability [3–5].

Bio-ethanol is a renewable resource which can be produced from several biomass sources including energy plants, waste materials from agro-industrial wastes and forestry residue materials. The bio-ethanol production process consists of four main steps; (i) the pretreatment step to reduce material size, (ii) the hydrolysis step to hydrolyze cellulose into sugar, (iii) the anaerobic fermentation step to convert sugar into ethanol and (iv) the bio-ethanol purification step to purify dilute bioethanol to pure ethanol. Bio-ethanol is also considered a good candidate for hydrogen production because of its renewability, low toxicity and high hydrogen content. However, the ethanol concentration in a bio-ethanol solution from fermentation step is generally low about 8.0-12.0 wt.% [6]. The steam to ethanol ratio in bio-ethanol stream has higher several magnitude of the stoichiometric ratio of ethanol steam reforming (steam to ethanol ratio = 3) when the bio-ethanol is directly used for hydrogen production via steam reforming process. This results in low hydrogen concentration due to dilution effect and high energy consumption for preheating reactant.

However, the use of expensive distillation units to produce water-free ethanol requires a large amount of energy [7]. This results in high capital and operating costs for pure ethanol production. Due to the water dilution in the bio-ethanol reforming process, the use of co-reactant with bio-ethanol to perform the steam reforming by using the excess water in the dilute bioethanol is considered in this work. Recently, the study of proton exchange membrane fuel cells (PEMFCs) fueled by hydrogen from ethanol steam reforming process to produce electricity has been studied by many researchers. Lima et al. [8] investigated ethanol steam reforming in presence of ceria catalysts. The main mechanism of ethanol steam reforming was proposed. The techno-economic study of ethanol autothermal reforming integrated with PEMFC was analyzed by Lopes et al. [9]. They reported that cost of electricity generation from ethanol is lower than the photovoltaic panel integrated with battery and photovoltaic panel integrated with a water electrolyzer and a PEMFC. However, there is a lack of investigation on the dilution effect and the addition of co-reactant of hydrogen production from bio-ethanol steam reforming on system performance and efficiency.

Currently, most fuel cell investigation and development aims to enhance its performance and reduce the cost. However, the pollution released during the power generation process via the hydrogen production process and the fuel cell is an important issue. Life cycle assessment (LCA) is an effective tool to study the environmental impact throughout the entire life cycle of a process [10]. Investigations of hydrogen production systems via the LCA methodology have been performed by many researchers. Hajjaji [11] investigated the environmental impact assessment of hydrogen production from poultry tallow steam reforming. The main cause of the environmental problems is from the energy production process. The environmental improvements should be focused on the minimization of this energy requirement and transport distance. The life cycle assessment of hydrogen production from renewable and fossil feedstocks was also studied by Hajjaji et al. [12]. They concluded that methane reforming has the highest global warming and abiotic depletion potential impacts, whereas bio-methane reforming systems show the lowest impact of all of the considered system. Kalinci et al. [13] studied the LCA of hydrogen production from two different biomass gasification reactor systems. However, studies of the environmental impact of fuel cell systems are still limited.

In this work, the mixed bio-ethanol and methane reforming integrated with a proton exchange membrane fuel cell (PEMFC) are theoretically investigated and compared with an integrated dehydrated bio-ethanol reforming and PEMFC system. Simulation studies were performed using Aspen Plus simulator software. The effect of operating parameters such as temperature and feed ratio on the hydrogen and CO fractions as well as the reformer efficiency were studied to determine the optimal conditions. In addition, the life cycle assessment of two scenarios of power generation from cassava based bio-ethanol was investigated. The two considered scenarios are based on dehydrated bio-ethanol reforming integrated with a PEMFC system (DE-PEMFC system) and mixed bio-ethanol and methane reforming integrated with a PEMFC system (BM-PEMFC system).

#### Methodology

#### **Process simulation**

In this work, the PEMFC integrated with bio-ethanol fuel processor system is studied in Aspen Plus simulator and demonstrated in Fig. 1. The fuel processing section of the integrated system consists of a reformer, low-temperature water gas shift reactor (LT-WGSR), high-temperature water gas shift reactor (HT-WGSR) and first preferential oxidation (PROX I) and second preferential oxidation (PROX II) as CO removal units. Two fuels are considered, dehydrated bio-ethanol and a mixture of bio-ethanol and methane.

#### Fuel processing process

The direct minimization of Gibbs free energy is applied to calculate the equilibrium compositions of the reformate gases obtained from the steam reforming of the dehydrated bioethanol and the mixture of bio-ethanol and methane. This method is solved by representing the reformer with the Rgibbs reactor module in the Aspen Plus simulator. The equation of state used in the calculation is based on the Soave-Redlich-Kwong equation. The gaseous components that appear in both steam reforming products are C<sub>2</sub>H<sub>5</sub>OH, CH<sub>4</sub>, H<sub>2</sub>, CO,  $CO_2$ ,  $O_2$  and  $H_2O$ , and the possible reactions are shown in Eqs. (1)-(5). The hydrogen yield of ethanol steam reforming from thermodynamic analysis is compared with experimental with various commercial catalysts [14] as shown in Fig. 2. The ethanol reformer is operated at pressure of 1.2 bar, steam to ethanol ratio of 6 and temperature of 300-700 °C. It is observed that the hydrogen yield obtained from the Aspen Plus simulator and the experimental data are in agreement.

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