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Investigation on the hydrogen production by methanol steam reforming with engine exhaust heat recovery strategy

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ABSTRACT

In this study, a reformer with an engine exhaust heat exchanger was employed to produce hydrogen. The purpose of this study is to investigate a methanol reformer with engine exhaust heat recovery to produce hydrogen with steam reforming of methanol (SRM). Steam reforming is an endothermic reaction requiring additional energy for reaction. Thus, engine exhaust heat was recovered as an energy source for the reforming to generate hydrogen. The experiments were initiated from basic engine firstly to measure the exhaust temperature and emissions; hence, the engine exhaust heat flow rates were calculated under various operating conditions. After the basic experiments, a reformer was installed in the engine exhaust system of a 1.4-L four-stroke spark-ignition engine. Additionally, the components of the reformat gas were measured to identify the correlation between hydrogen flow rate and exhaust heat exchange rate under various operating conditions.

The results show that when the engine was running between 2000 and 3000 rpm and throttle opening set at 20%, the exhaust temperature and heat flow were enough for SRM from the basic engine tests. In the reforming processes, the reformer heat recovery rate increased with engine speed. However, increasing exhaust mass flow rate resulting in poor heat exchange rate at high engine speed. Furthermore, methanol conversion efficiency increased with reformer heat exchange rate. When the S/C ratio was set to 1.2, and the methanol supply rate was fixed at 15.8 g/min, methanol conversion efficiency approached 93% and the hydrogen production was stably close to 75%. The molar rate of hydrogen was approximately 1.34 mol/min. In addition, hydrogen production per unit exhaust heat was 1.6 mol/MJ.

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Introduction

Technological advancements have led to a considerable increase in energy consumption. All over the world, fossil fuels

remain the dominant energy resource, yet they have attracted worldwide attention because of issues regarding environmental protection. Many countries have initiated measures to control greenhouse gas (GHG) emissions, among which carbon dioxide (CO₂) and methane (CH₄) are the most severe.

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Therefore, many countries have actively invested in developing two primary categories of energy: clean energy, which refers to pollution-free energy that generates zero carbon emissions, and alternative energy, which refers to reducing the hydrocarbon content of fuels in order to reduce carbon emissions. Chiriac et al. stated that hydrogen combustion emits no carbon; thus, it would be used as an attractive alternative fuel. When 2% of the energy percentage of diesel is replaced with hydrogen, CO₂ emissions can be reduced by approximately 2.5% [1].

In the past decade, continuing growth in the steel industry has been accompanied by high levels of energy consumption. Slag in furnaces after steelmaking can reach temperatures between 1450 °C and 1550 °C [2], that is, it remains much waste heat. During the production of cement, the primary heat loss results from flue gas emissions, cooling clinkers, and kilns [3]. In addition, there are remarkably huge amounts of pollution and waste heat from the worldwide vehicle exhaust systems. Hossain reported that approximately 38% of the supplied fuel energy in diesel engine is emitted through the exhaust emissions [4,5]. Shudo et al. used an exhaust gas heat recovery strategy in a homogenous charge compression ignition (HCCI) engine for hydrogen production by dimethyl ether and methanol reforming. The results showed that hydrogen addition could avoid oxidation of dimethyl ether (DME) at low temperature and control the ignition timing of the mixture in cylinder. Moreover, the methanol reforming by HCCI waste heat recovery could achieve a high overall system efficiency [6]. Bueno used a heat recovery reformer to reform glycerol into H₂-rich gases, hydrogen, and methane. The best reaction temperature for producing hydrogen is 700 °C [7]. Thus, alternative vehicle power sources should be developed to reduce the use of fossil fuels and carbon emissions. To achieve this goal, hydrogen energy plays an essential role. Water is the product of hydrogen under exothermic oxidation reaction, which is clean and does not create environmental pollution. The advantages of hydrogen energy include abundance, high heating value, excellent burning performance, extensive applications, environmental friendliness, and high potential for economic benefits [8–10]. Hydrogen can be obtained through various means such as hydrocarbon fuel reforming and water electrolysis. The popular hydrocarbon fuels for hydrogen production can be divided into two categories, which are methane (gas) and alcohol (liquid). Where, methanol possesses many advantages such as easiness to obtain, convenient transportation and storage, high energy density, and high hydrogen–carbon ratio.

Currently, common reforming methods for hydrogen production include partial oxidation reforming (POX), steam reforming (SR), oxidation steam reforming (OSR), autothermal reforming (ATR), coal gasification, plasma reformer, and dry reforming (DR) [11–16]. Worldwide, a considerable number of reforming experiments have been conducted for hydrogen production and introducing hydrogen into engines. Horng et al. used a methanol reformer and set multiple parameters to perform a cold-start experiment for fast hydrogen production. With a heating power of 240 W, heating temperature of 100 °C, methanol supply rate at 25 cc/min, and an airflow rate of 70 L/min, the cold start was activated in minimal time; specifically, stable hydrogen production required only 3 min

from a cold start. Hydrogen production began at a catalyst outlet temperature of 100 °C, becoming stable at approximately 380 °C [17]. Horng et al. examined the transient response of mode shifting between POX and ATR by considering heating temperature, methanol supply rate, steady-mode shifting temperature, and oxygen-to-carbon (O₂/C) and steam-to-carbon (S/C) molar ratios to evaluate the effect of reformer fast start and increased hydrogen production. When POX was used, the hydrogen concentration and hydrogen flow rate were 40% and 20.5 L/min, respectively; when the mode was shifted to ATR, the corresponding values were 49.12% and 23 L/min [18]. Moreover, Kamarudin et al. designed a 5-kW mobile proton exchange membrane fuel cell and tested it using ATR, water gas shift (WGS), and preferential oxidation (PrOX) systems. The experimental results revealed an ATR hydrogen concentration of 73% and CO concentration of 2% when the O₂/C and S/C ratios were 0.25 and 1.3, respectively. Subsequently, introducing WGS into the system decreased the CO to be smaller than 2000 ppm; further introducing PrOX reduced the CO to smaller than 100 ppm [19]. Yoon et al. compared copper-based catalysts and noble metal catalysts in SR and ATR systems under different O₂/CH₃OH ratios. Multiple start-up and shut-down cycles were applied for testing, and when noble metal catalysts were used in the ATR system, the conversion efficiency of supplied methanol approached 100% [20]. Mohammadi et al. reported that hydrogen has low ignition energy requirement, a wide flammability range, and high burning velocity, which make it a crucial fuel for engines. However, pre-mixing hydrogen externally with intake air causes backfire and engine knocking, particularly at high loads. The experimental results showed that direct injection of hydrogen during the late-compression stroke can prevent backfiring and achieve high thermal efficiency and output power. Further optimization of injection timing under high loads can reduce NO_x emissions [21]. Akansu et al. introduced various proportions of hydrogen and methane mixtures into engine to evaluate the relevant exhaust emissions and engine performance. The results indicated that introducing hydrogen and methane increased the engine combustion efficiency and reduced exhaust emissions [22]. Horng et al. introduced plasma-reformed H₂-rich gases into motorcycle engine to investigate the vehicle performance at a constant speed and transient driving conditions. At a low speed, the energy-conservation improvement rate reached 12.2%, whereas the improvement at a high speed was negligible. Regarding exhaust emissions, the CO and HC emissions of the motorcycle engine after introducing H₂-rich gases were similar to those generated by the original engine; however, the NO_x emissions exhibited a 56.8% improvement rate [23].

Referencing the aforementioned experimental methods, the present study investigated conducting steam reforming of methanol (SRM) reactions to produce H₂-rich syngas. However, SR reactions are endothermic reactions that require a supply of external energy. Exhaust heat recovery was utilized as the SRM energy source. Thus, an additional energy supply was unnecessary and hence energy consumption for reforming can be reduced. In addition, Lu utilized diesel engine exhaust heat in examining the hydrogen production characteristics of SRM. Engine exhaust heat was transferred to the reformer via a heat exchanger installed with fins. The

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