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Hydrogen production from steam reforming using an indirect heating method

Hyunjin Ji*, Junghun Lee, Eunyeong Choi, Ilsung Seo

Agency for Defense Development, Yuseong P.O. Box 35-44, Daejeon, Republic of Korea

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ABSTRACT

In this work, a methanol steam reforming (MSR) reactor was operated using an indirect heating method. A thermal circuit was constructed between the MSR reactor and the electrical heater to supply the heat required for the endothermic reaction, and deionized water was used as the heat transfer medium (HTM). The MSR reactors featured a shell-and-tube type design to operate at high pressures. A Cu/Zn catalyst was installed on the tube side, and HTM was supplied to the shell side. To improve the heat transfer performance, the heat transfer area between the shell and tube was increased from 598 to 1117 cm². Because the MSR reactor had a sufficient heat exchange area corresponding to the catalytic reaction rate, the heat exchange area had little effect on methanol conversion. However, the heat exchange area had a greater effect on the performance because the operating temperature of reactor was lower. Under the same operating temperature conditions, the MSR reactor operated under the indirect heating method showed relatively higher methanol conversion than the MSR reactor operated in an electric furnace because of the effective heat transfer by the latent heat of saturated steam. The MSR reactor based on the indirect heating method was continuously operated at 250 °C for 72 h to verify characteristic start-up and operation. The results showed that the MSR reactor could be operated at a constant temperature; however, low methanol conversion at low operating temperatures led to slow catalyst degradation. In addition, the MSR reactor required more than 2 h for initial start-up and for restart after emergency shutdown because the HTM needed to be evaporated and pressurized to the target pressure.

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Introduction

Conventional diesel-electric submarines use a diesel generator to charge batteries when floating above sea level, and the batteries are discharged to drive the propulsion motor underwater. However, in contrast to a nuclear submarine, which can remain indefinitely submerged, diesel-electric submarines have limited activity while submerged because of their low battery capacity. To overcome these limitations and increase the endurance time in underwater air-independent

propulsion (AIP), methods that do not require the presence of air have been introduced. Fuel cells are considered the most advanced AIP method available for submarines [1–4].

Fuel cells are energy conversion devices that directly convert chemical energy into electrical energy. Fuel cells are considered next-generation power systems that can replace internal combustion engines and solve air pollution problems because of their high efficiency, lack of noise and vibration, and eco-friendliness. For this reason, various applications in the defense field have recently been reported. In particular, the operating capability of unmanned underwater vehicles

* Corresponding author.

E-mail address: grgmachine@add.re.kr (H. Ji).

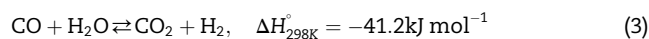
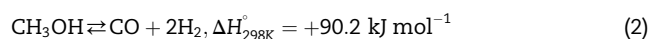
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(UUVs) and submarines has been dramatically improved by the installation of fuel cell systems because of their low noise levels and high specific energy density [5–7]. Despite the excellent performance of fuel cells, limited H₂ storage remains an unsolved problem [8,9].

In many modern naval submarines, e.g., those in Germany, the Republic of Korea and Greece, numerous metal hydride cylinders have been installed to supply H₂ to a polymer electrolyte membrane (PEM) fuel cell module [5]. The H₂ storage method using metal hydrides advantageously provides high H₂ storage density per unit volume and excellent storage safety [3,10,11]. However, metal hydride cylinders require high-purity H₂ and long charging times [12]. These factors must be improved in terms of logistic support. Various methods have been studied to overcome these drawbacks. Biert et al. [13] reviewed various fuels, such as liquid hydrogen (LH₂), diesel, liquefied natural gas (LNG), and dimethyl ether (DME), applicable to marine fuel cell systems. Han et al. [14,15] studied a diesel autothermal reformer with hydrogen peroxide for low-oxygen environments. Nikiforov et al. [16] studied hydrogen production based on the hydrolysis of aluminum onboard a submarine. Hahn et al. [17] dissolved commercial magnesium AZ31 in seawater for hydrogen production. Luo et al. [18] and Oh et al. [19] studied the performance and characteristics of direct sodium borohydride/hydrogen peroxide (NaBH₄/H₂O₂) fuel cells for potential application in AIP for underwater vehicles. Nevertheless, reforming technology remains one of the most practical methods to apply to underwater vehicles when considering maturity and difficulty [20,21]. Metal hydrides increase the number of cylinders in direct proportion to the demand for hydrogen, but a fuel processor directly produces hydrogen using a high energy-density hydrocarbon fuel; thus, increasing the fuel tank size will result in more hydrogen. Therefore, fuel processors are suitable for large underwater vehicles. Additionally, compared with metal hydrides, fuel is easier to acquire, store, transport, and handle and can be quickly replenished [22].

Because methanol (MeOH) can be converted into hydrogen at low temperatures (200–300 °C), has a relatively high H/C ratio, and enables a simple layout of the fuel processor [23], in this study, MeOH was selected as the fuel instead of other hydrocarbons. In the overall MeOH steam reforming (MSR) reaction (Eq. (1)), two side reactions are commonly considered: MeOH decomposition (Eq. (2)) and the water-gas shift reaction (Eq. (3)) [24]:



As shown in Eq. (1), MSR is an endothermic process; thus, a continuous supply of heat is required to maintain a steady process temperature. From a systems viewpoint, the most common method is to obtain heat from a combustion gas generated by the burner. The burner uses the remaining reformate gas after the electrochemical reaction at the fuel cell stack and additional MeOH supplied to the burner [25–30]. This method is a direct heating method because heat is produced (burner) and consumed (reactor) in nearby locations.

Therefore, the direct heating method can simplify the MSR reactor structure. However, a temperature gradient can occur in the reactor [31,32] because the combustion gas is in the gas phase during heat transfer and has a low overall heat transfer coefficient. Furthermore, the heterogeneous temperature distribution inside the reactor is exacerbated with an increasing reactor size. To improve heat transfer, researchers installed a latent-heat phase change material (PCM) [33,34] around the reactor. Maruoka et al. [35] investigated a new MSR reactor with machined copper (latent heat = 13.3 kJ mol⁻¹) as the PCM, dual pipes and both covers made of chromium. The results revealed that the PCM temperature was constant despite intermittent heating and that hydrogen was continuously generated. However, when the reactor size was increased, uniform heating of the entire area of the PCM surrounding the reactor with the burner became difficult. Dusterwald et al. [36] and Wiese et al. [37] supplied heat to the MSR reactor using latent heat from saturated steam as a heat transfer medium (HTM). However, because the longitudinal length of the single-tube-type reactor was 1 m, available space was limited. Their studies also focused on the reactor performance rather than on the reactor design using saturated steam as the HTM.

In this study, a MSR reactor using indirect heating was designed as a shell-and-tube type that can operate at high pressures and improve space efficiency because multiple tubes can be installed in parallel. A thermal circuit was constructed between the MSR reactor and an electrical heater to supply the heat required for the endothermic reaction. Given the operating temperature and size of the MSR reactor, deionized water was selected as the HTM. Because saturated steam has fluidity and latent heat at the target temperature, it can supply heat efficiently to the reactor. Using these experimental setups, the effect of the heat transfer rate between the shell and tube was investigated. We also compared the reforming performance between the direct heating method using an electric furnace and the indirect heating method with HTM. Finally, the MSR reactor with a thermal circuit was operated for 72 h to monitor performance changes.

Experimental setup

Fig. 1 shows a schematic of the experimental setup for high-pressure MSR. It mainly consists of three parts: a MSR reactor, a thermal circuit and a gas analyzer. Briefly, the MSR reactor was designed as a shell-and-tube type according to the American Society of Mechanical Engineers Section (AMSE) VIII pressure vessels [38] because both the catalyst zone (tube side: design pressure = 60 bar(g)) and the HTM zone (shell side: design pressure = 100 bar(g)) were exposed to high-pressure environments. The reaction zone was several stainless-steel tubes equipped with app. 232 g of Cu/Zn catalysts (RP-60, BASF Co., Germany). The catalyst was activated by reducing copper oxide to copper at 250 °C at 4 vol% H₂ (Ar balance). The shell side of the MSR reactor was a region in which the HTM flowed such that the heat of the HTM was effectively transferred to the tube side. The reactor had a multi-hole shell side with a diameter greater than the tube so that the HTM could flow parallel to the longitudinal direction of the tube, as shown in Fig. 2. To compare the performance of the MSR

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