ARTICLE IN PRESS

Applied Energy xxx (2015) xxx-xxx



Contents lists available at ScienceDirect

Applied Energy



journal homepage: www.elsevier.com/locate/apenergy

Compact PEM fuel cell system combined with all-in-one hydrogen generator using chemical hydride as a hydrogen source $\stackrel{\circ}{\sim}$

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HIGHLIGHTS

• Compact fuel cell system was developed for a portable power generator.

• Novel concept using an all-in-one reactor for hydrogen generation was proposed.

• Catalytic reactor, hydrogen chamber and separator were combined in a volume.

• The system can be used to drive fuel cell-powered unmanned autonomous systems.

ARTICLE INFO

Article history: Received 17 November 2014 Received in revised form 12 February 2015 Accepted 18 March 2015 Available online xxxx

Keywords: Fuel cell Chemical hydride Hydrogen generator High energy density Portable power generator

ABSTRACT

Compact fuel cell system was developed for a portable power generator. The power generator features a polymer electrolyte membrane fuel cell (PEMFC) using a chemical hydride as a hydrogen source. The hydrogen generator extracted hydrogen using a catalytic hydrolysis from a sodium borohydride alkaline solution. A novel concept using an all-in-one reactor was proposed in which a catalyst, hydrogen chamber and byproduct separator were combined in a volume. In addition, the reactor as well as a pump, cooling fans, valves and controller was integrated in a single module. A 100 W PEMFC stack was connected with the hydrogen generator and was evaluated at various load conditions. It was verified that the stable hydrogen supply was achieved and the developed system can be used to drive fuel cell-powered unmanned autonomous systems.

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1. Introduction

Recently, a variety of mobile unmanned autonomous systems (UAS) are being developed. In addition, currently-developed UAS has been widely used for civilian and military applications. The mobile UAS uses secondary batteries such as a lithium–polymer battery as a main power source. However, the operating duration is seriously limited due to the low energy density of existing secondary batteries. Particularly, small unmanned aerial vehicles (UAV) for aerial reconnaissance are performing missions with the limited operational radius because the battery power can offer them an endurance of around 1 h. For the above reason, many groups have tried to use a fuel cell as an alternative power source

http://dx.doi.org/10.1016/j.apenergy.2015.03.084 0306-2619/© 2015 Elsevier Ltd. All rights reserved. because of its higher energy density than existing batteries. It is expected that the operation duration and mission range can be extended by applying the fuel cell to the UAS as a power source.

In the case of UAVs, fuel cell-powered UAVs have been widely developed in worldwide, such as Phantom Eye by Boeing [1], Hyfish by the German Air & Space Center [2], FAUCON H2 by EnergyOr Technologies [3], and Korea Institute of Science and Technology [4]. Moreover, the fuel cell system for UAVs has been studied at universities such as the Washington State University [5] in the United States, National Cheng Kung University [6] in Taiwan, Nanyang Technological University [7] in Singapore, Korea Advanced Institute of Science and Technology, and Chosun University in Korea [8,4,9]. Up to now, a polymer electrolyte membrane fuel cell (PEMFC) has been mainly adopted for the UAS but the hydrogen storage density was too low to realize the high energy density of the fuel cell system using the compressed as a hydrogen source. Basically, hydrogen has gravimetric energy density (120 MJ/kg) three times higher than gasoline (44 MJ/kg). However, the volumetric energy density (0.01 MJ/L at STP) is much lower than gasoline (32 MJ/L). Thus, the high-density hydrogen

Please cite this article in press as: Kim J, Kim T. Compact PEM fuel cell system combined with all-in-one hydrogen generator using chemical hydride as a hydrogen source. Appl Energy (2015), http://dx.doi.org/10.1016/j.apenergy.2015.03.084

^{*} This article is based on a short proceedings paper in Energy Procedia Volume 161 (2014). It has been substantially modified and extended, and has been subject to the normal peer review and revision process of the journal. This paper is included in the Special Issue of ICAE2014 edited by Prof. J Yan, Prof. DJ Lee, Prof. SK Chou, and Prof. U Desideri.

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storage is important for determining the energy density of PEMFC systems for UAS applications. In addition, the hydrogen storage method would have high storability and good mobility [10–12].

Currently-available hydrogen storage methods include compressed hydrogen, liquid and cryo-compressed hydrogen, sorbents, metal hydrides, metal-organic frameworks (MOF), and carbonbased material [13–17]. However, conventional hydrogen storage methods are still problematic; for the compressed hydrogen, the hydrogen storage density is very low and for the liquid hydrogen, the device for refrigeration is bulky and complex. Recently, it is reported that MOF and carbon-based hydrogen storages have realized the desirable hydrogen storage density but they require high manufacturing technologies [10]. As a result, these hydrogen storage methods suffer from technical difficulties in manufacture, the increased expense for the maintenance, and problems that make the system bulky and heavy. Thus, we need an alternative method for hydrogen storage to complement the above problems.

Recently, chemical hydrides have attracted a great interest as an alternative of hydrogen storage for UAS applications. In the present study, a fuel cell system using the hydrogen storage based on the chemical hydride was developed for portable applications. There are many chemical hydrides available including NaBH₄, ZnBH₄, CaBH₄, LiAlH₄, NaBH(OCH₃)₃ and so on. Chemical hydrides are stored stably as it is at the atmospheric condition, while hydrogen can be extracted readily by chemical decomposition when needed. The chemical hydride has advantageous to portable fuel cells because of its high hydrogen capacity and possibility of hydrogen storage/supply at the atmospheric condition. Particularly, chemical hydrides can be stored and carried in the state of an alkali solution, and hydrogen can be extracted from the solution as well as directly in the solid state [18]. The chemical hydrides mainly consist of alanate hydrides, alkali metal hydrides, and metal hydro-borates [19]. Among them, sodium borohydride (NaBH₄) metal that is a kind of hydro-borates has been widely studied as the chemical hydridebased hydrogen storage because it is cheaper and the energy density per unit volume/mass is higher than other types of chemical hydrides [20–24]. In addition, the NaBH₄ has gravimetric and volumetric energy densities of 25.6 MJ/kg and 6.6 MJ/L at STP, respectively. The gravimetric energy density is comparable with gasoline and the volumetric energy density is much higher than hydrogen (0.01 MJ/L at STP). Thus, the NaBH₄ was selected as a hydrogen source for the PEMFC system in the present study.

Hydrogen can be generated by the hydrolysis reaction of NaBH₄. The hydrolysis reaction should be accelerated and stabilized to supply hydrogen stably and continuously. There are two ways that have been widely used; one is to use a catalyst and the other is to control pH value by acids [21]. When acids are used, the hydrogen generation is reliable but the hydrogen generation rate was not controllable without control mechanisms and the acid is difficult to handle and store. Considering the aspect of safety, the catalytic hydrolysis of NaBH₄ was selected for portable applications. Up to now, noble catalysts such as Ru, Rh, and Pt, and non-noble catalysts such as Ni, Co, and Cu have been studied for the catalytic hydrolysis of NaBH₄. In the present study, Co–B/Ni foam was used as a catalyst because of its relatively low cost and high performance [24–26].

Kim et al. [8,4,9] reported that the fuel cell-powered small unmanned aircraft was developed, where the fuel cell system featured a 100 W PEMFC combined with a hydrogen generator using a catalytic hydrolysis of NaBH₄ solution. However, there were still problem in the catalytic hydrolysis. First, the catalyst can be seriously deactivated by the deposition of sodium metaborate (NaBO₂) that is a byproduct of the hydrolysis of NaBH₄ [27–29]. In addition, the hydrogen generation rate was controlled by varying the feeding rate of NaBH₄ solution. This control strategy was not suitable to provide the rapid response to the dynamic change of the hydrogen supply for the fuel cell of UAVs. The most serious problem is that the device for generating pure hydrogen from NaBH₄ should equip the catalytic reactor, hydrogen chamber, and NaBO₂ separator, which are very complex and makes the system bulky and heavy [30,29]. Seo et al. [9] developed ammoniaborane-based hydrogen fuel cell system for UAVs because ammonia-borane has higher hydrogen storage density (19.8 wt.%) compared to the NaBH₄ (10.8 wt.%). However, ammonia gas, which seriously deactivates the electrode catalyst of the fuel cell, was contained in product gases so that a bulky hydrogen purification system was required. Ultimately, the energy density of the system (95 W h/kg) was lower than those with NaBH₄-based system (200–400 W h/kg) [8,4,9].

In the present study, the all-in-one reactor was designed in which the catalytic reactor, hydrogen chamber, and NaBO₂ separator were combined in a single space. In the present study, the all-in-one reactor was designed in which the catalytic reactor, hydrogen chamber, and NaBO₂ separator were combined in a single space. By taking this design, the response characteristic for the dynamic hydrogen supply was considerably improved and the borate disposal was easier to prevent the catalyst from being deactivated by the NaBO₂ deposition. In addition, all of balance-of-plant (BOP) such as a pump, valve, cooling fan, and controller were integrated to develop the complete hydrogen generator. The developed hydrogen generator was connected to a 100 W PEMFC stack and the performance was evaluated according to the electronic load.

2. Experiments

2.1. Fuel cell system design

The developed compact PEMFC system using the hydrogen generator based on the NaBH₄ chemical hydride is shown in Fig. 1. The system consists of three parts; the PEMFC stack, hydrogen generator, and NaBH₄ fuel tank. First, 100 W lightweight PEMFC stack was used. Particularly, bipolar plates were manufactured by metal forming so that the weight and volume of the stack could be reduced considerably. Next, the hydrogen generator has an all-in-one reactor in which the NaBH₄ hydrolysis takes place and the generated hydrogen is pressurized temporally. The exploded view of the all-in-one reactor is shown in Fig. 2. The reactor consists of three parts; the catalytic reactor, byproduct separator, and hydrogen chamber. The Co-B/Ni foam catalyst was packed in the catalytic reactor as shown in Fig. 3. The Co–B is an active material for the NaBH₄ hydrolysis and the Ni foam is the support of Co–B. The catalyst was prepared by electroless plating. The foam catalyst was very easy to be shaped and the replacement was very simple.

The NaBH₄ solution was supplied into the catalytic reactor by the pump and then the hydrolysis reaction took place on the catalyst, immediately generating hydrogen. The generated hydrogen was stored temporally in the hydrogen chamber maintaining the pressure of 5 bar. Hydrogen was supplied to the fuel cell after the pressure was adjusted to 1.6 bar through the pressure regulator. The NaBO₂ was collected in the byproduct separator and discharged through the valve into the outside according to the amount of NaBH₄ supplied by the pump. The cooling fins were attached on the reactor to cool the heat of reaction of the NaBH₄ hydrolysis that is exothermic. Cooling fans were installed on the case of the fuel cell system to maintain the constant temperature in the reactor. The NaBH₄ solution was stored in the fuel tank that is easy to be detachable from the system, which facilitated the safe storage and porterage.

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