



Portable ammonia-borane-based H₂ power-pack for unmanned aerial vehicles



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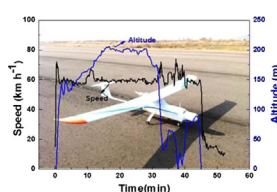
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HIGHLIGHTS

- A portable power-pack fueled by ammonia borane (AB) was newly designed to power an unmanned aerial vehicle (UAV).
- The power-pack was demonstrated to drive a UAV for 57 min with fast load-following ability and rapid response time.
- *In situ* monitoring system was introduced to determine the filter capacity of hydrogen purification equipment.

GRAPHICAL ABSTRACT



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ABSTRACT

An advanced ammonia borane (AB)-based H₂ power-pack is designed to continually drive an unmanned aerial vehicle (UAV) for 57 min using a 200-W_e polymer electrolyte membrane fuel cell (PEMFC). In a flight test with the UAV platform integrated with the developed power-pack, pure hydrogen with an average flow rate of 3.8 L(H₂) min^{−1} is generated by autothermal H₂-release from AB with tetraethylene glycol dimethylether (T4EGDE) as a promoter. During take-off, a hybridized power management system (PMS) consisting of the fuel cell and an auxiliary lithium-ion battery supplies 500 W_e at full power simultaneously, while the fuel cell alone provides 150–200 W_e and further recharges the auxiliary battery upon cruising. Gaseous byproducts identified by *in situ* Fourier transform infrared (FT-IR) spectroscopy during AB dehydrogenation are sequestered using a mixed absorbent in an H₂ purification system. In addition, a real-time monitoring system is employed to determine the remaining filter capacity of the purifier at a ground control system for rapidly responding unpredictable circumstances during flight. Separate experiments are conducted to screen potential materials and methods for enhancing filter capacity in the current H₂ refining system. A prospective reactor concept for long-term fuel cell applications is proposed based on the results.

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1. Introduction

Ammonia borane (NH₃BH₃, AB) is a promising hydrogen storage material that has been extensively studied as a potential fuel to replace oil for transportation applications in the last decade due to

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its reasonable stability under ambient conditions, high hydrogen storage capacity (19.6 wt%), and potential regenerability. The AB fuel can release hydrogen on demand via hydrolysis [1], alcoholysis [2], or thermolysis [3]. The thermally induced dehydrogenation of AB at <250 °C proved capable of enabling the generation of sufficient quantities of hydrogen with high H₂ storage density (material-based, >13 wt%). The hydrogen production via AB thermolysis is thus of particular interest for long-term applications.

For this method to be useful for desired purposes, the sluggish kinetics for AB dehydrogenation need to be improved. In this context, a number of approaches for enhancing the slow rate of AB dehydrogenation have been proposed. For example, transition-metal-based catalysts including Rh [4], Ir [5], Ru [6], Ni [7], and Fe [8] were utilized to accelerate the rates of H₂ release. Various types of chemical promoters such as proton sponge [9], ionic liquids [10,11], Lewis and Brønsted acids [12], mesoporous silica [3], diammoniate of diborane (DADB) [13], and activated boron nitride [14] were also demonstrated as efficient additives for promoting the liberation of H₂ from AB. For a continuous reactor, a liquid AB-based fuel blended with an ionic liquid (e.g., 1-butyl-3-methyl-imidazolium chloride, bmimCl) was employed for continuous hydrogen production from AB under mild conditions [10,11], which could potentially satisfy the 2015 Department of Energy (DOE) target of 5.5 wt% (system-based) for transportation applications [15]. The ionic liquids were further suggested to play a dual role as a transporting medium of AB and as a promoting additive for AB dehydrogenation [11]. Based on experimental and density functional theory (DFT) methods, a series of polyetheral promoters were suggested to enhance the reactivity of B–H bonds at AB through intermolecular hydrogen bonding interaction, which facilitated the formation of DADB, a reactive intermediate, to promote AB dehydrogenation [11,16]. Another strategy recently proposed by Varma and coworkers [17] involved a non-catalytic hydrothermal method with efficient heat management, which could produce a large quantity of hydrogen from AB with a considerably high H₂ storage capacity of ca. 14 wt% (materials-based).

Despite these accomplishments, the development of a continuous hydrogen generator utilizing AB fuel is still challenging due to the following concerns: (i) continuous supply of solid AB, (ii) purification of hydrogen containing gaseous byproducts, and (iii) discharging of liquid/solid spent-fuels from a reactor. Many studies have been conducted for the creation of an AB-based hydrogen generation system that ensures continuous feeding of AB as well as discharging of waste fuels. For example, DOE's Hydrogen Storage Engineering Center of Excellence (HSECoE) has been making significant efforts in relevant reactor design using the mixture of AB with either silicon oil [18] or ionic liquid [19]. Devarakonda et al. recently reported an auger reactor simulation using solid AB and a liquid-state AB fuel [20,21]. A novel reactor fueled by AB beads with a spinning wheel feeding system was also recently developed, which demonstrated capability to successively power a 200-W_e polymer electrolyte fuel cell (PEMFC). In this system, spherical solid AB beads were sequentially delivered into a semi-batch-type reactor filled with tetraethylene glycol dimethylether (T4EGDE), a liquid promoter [16,22]. The T4EGDE additive in the continuous reactor was suggested to play roles in (i) accelerating the H₂-release kinetics, (ii) suppressing the formation of scatters/impurities and foaming upon dehydrogenation, and (iii) fluidizing solid spent-fuels [23]. For practical applications, an improved reactor concept was proposed by introducing H₂ purification equipment with high filter capacity and an efficient drainage system for spent fuels.

We report here on a portable hydrogen power-pack fueled by solid AB pellets, which mainly comprises an AB-based hydrogen generator, a hybridized power management system (PMS) installed with a 200-W_e PEMFC, an auxiliary battery, and a control unit. The

performance of the developed hydrogen power-pack was then assessed by integration into an unmanned aerial vehicle (UAV) platform under collaboration with the Korea Aerospace Research Institute (KARI). To the best of our knowledge, no continuous AB-based H₂ generator has previously been applied for a practical application to date. The ability of the power-pack to continuously drive a UAV for 57 min with fast load-following ability and rapid response time was verified. The fuel cell with the AB-based H₂ production system was found to provide supplemental power to recharge the auxiliary battery during operation. Plausible strategies for enhancing the efficiency of the hydrogen generator are discussed based on additional experiments.

2. Experimental setup

2.1. Materials

The AB solids (Aviabor, 98%) were pulverized into fine powers by a commercial grinder under N₂ atmosphere, followed by pelletizing into spherical beads using a manual dry pressing process [23]. The AB beads weighed ca. 0.082 g, with a diameter of 5.7 mm, and density of ca. 0.74 g cm⁻³. Supposing that one AB bead releases 2 equiv. of H₂, the AB pellet can produce ca. 130 mL of H₂. Tetraethylene glycol dimethylether (T4EGDE, Sigma Aldrich, 99%) and triethylene glycol dimethylether (T3EGDE, Sigma Aldrich, 99%) were employed without further purification and stored under an atmosphere of N₂ at room temperature.

2.2. Procedure for hydrogen production with the developed H₂ generator

An AB-powered continuous H₂ generator with fast dehydrogenation rates up to 3.3 L(H₂) min⁻¹ and fast load-following capability was recently constructed [23]. Providing a continuous supply of the AB/T4EGDE mixtures (AB:T4EGDE = 50:50, wt%) in a liquid phase was found to be difficult due to the limited AB solubility towards T4EGDE. In addition, since the T4EGDE promoter was exuded upon pelletizing the mixtures of AB and T4EGDE, the delivery of the AB/T4EGDE mixtures in a solid state was unsuccessful. For these reasons, desired amounts of the liquid T4EGDE promoter (120 g) were pre-loaded in a semi-batch-type reactor while solid AB pellets (with a stored weight of 110 g) were supplied into the reactor by adjusting feeding rates in the range of 2.46–2.54 g min⁻¹. The AB contents (relative to AB + T4EGDE) in the reactor thus varied from 0.0 to a maximum of 48 wt% as a function of time. The AB powders were pelletized into a spherical form, and one AB bead had an average weight of 0.082 g (Fig S1), which could theoretically produce ca. 130 mL(H₂) upon releasing 2 equiv. of H₂. Continuous supply of the AB beads into a semi-batch-type reactor was achieved using a spinning-wheel-type conveyor.

2.3. Configuration of the developed fuel cell system for a UAV

The fuel cell power-pack utilizing both AB beads and T4EGDE consisted of (i) a continuous hydrogen generator, (ii) a hybridized power management system (PMS) equipped with a commercial 200-W_e PEMFC stack (AEROPAK [24]) as well as an auxiliary lithium-ion battery (Kokam), and (iii) control panels, as depicted in Fig. 1a. The developed H₂ generator was also composed of (1) a storage tank for the AB beads, (2) a conveying system, (3) a semi-batch reactor, and (4) an external H₂ purification system (Fig. 1b, red box). This H₂ production system has different features from the previous H₂ generator, in that (i) The dimensions of the fuel tank for the AB beads were expanded to increase the maximum storage from 80 g (1000 ea.) [23] to 120 g (ca. 1500 ea.) of AB, (ii) a worm-

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