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Hydrogen generation from aluminum hydride for wearable polymer electrolyte membrane fuel cells

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ABSTRACT

Aluminum hydride (AlH₃) has been identified as a promising H₂ storage material for fuel cell systems and offers a significant weight savings over conventional Li-ion batteries, due to its high H₂ storage capacity and simple balance of plant requirements for H₂ generation. This work reports on the development and characterization of a novel, wearable AlH₃ based PEM fuel cell system. System characterization revealed an unexpectedly low energy density value, 25% lower than anticipated, (436 Wh kg⁻¹ vs. 582 Wh kg⁻¹ for 1440 Wh) due in part to a previously unpublished phenomenon of incomplete α -AlH₃ thermolysis. Based on literature reports, complete thermolysis was expected to occur at <180 °C, however this work reports on conditions where the full H₂ yield cannot be obtained despite high temperature (>250 °C) exposure. This work provides an experimental characterization of this phenomenon and quantitatively describes it by developing a new model in the framework of the Avrami–Erofeev phase transformation model, which can be utilized for the optimum design of high energy density AlH₃ cartridges.

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Introduction

Power and energy are critical to assure communications and situational awareness capabilities for soldiers. To effectively deploy these capabilities on the soldier and to avoid increased risks of musculoskeletal injuries [1,2], higher density power and energy sources are required. As a potential solution, the authors together with industry partners, are investigating Aluminum hydride based wearable fuel cells [3,4].

Aluminum hydride (AlH₃, Alane) is a covalently bonded, binary hydride which can release up to 10.1 wt. % H₂ resulting in a very high H₂ capacity at 0.149 kg H₂ L⁻¹. While seven different polymorphs of AlH₃ (α , α' , β , γ , δ , ϵ , and ζ) have been identified, this work utilized a metastable α -AlH₃ polymorph

[5], which is the most thermally and hydrolytically stable form [6].

A previously developed AlH₃ PEM based fuel cell system is shown in [Supplementary Figure S1a](#) and the cartridge in [Supplementary Figure S1b](#). This system is able to provide a high energy density of 582 Wh kg⁻¹ for a 1440 Wh load, resulting in significant weight savings vs. the conventional 150 Wh kg⁻¹ Li-ion wearable battery. Nevertheless, based on soldier evaluation for a wearable application, this fuel cell system required: decreased system thickness (20 mm); and a flexible form factor. This feedback resulted in the development and evaluation of a novel wearable AlH₃ fuel cell system as described in [Sections Fuel Cell System and Fuel Cell System-Power and Energy Performance Measurements](#). As part of evaluating the system's α -AlH₃ cartridge performance,

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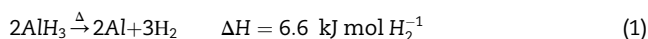
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the α -AlH₃ dehydrogenation kinetics were investigated resulting in the observation of a previously unpublished phenomenon of incomplete α -AlH₃ dehydrogenation and the development of a new α -AlH₃ dehydrogenation model.

As part of understanding the fuel cell system's cartridge performance, a brief review of α -AlH₃ dehydrogenation characterization and modeling is presented below, additional details can be found in a review by Graetz et al. [5] and Maehlen et al. [7].

Review of α -AlH₃ dehydrogenation

The metastable α -AlH₃ polymorph decomposes to produce up to 10.1 wt. % H₂ and Al metal in a single step endothermic reaction as shown in Eq. (1).



As reported [5–7], depending on the heating rate and the stabilization method, full decomposition ranges from 130 to 180 °C. Stabilization is done by the formation of an Al₂O₃ surface layer that prevents an α -AlH₃ gas interface and subsequent H₂ release as observed by *insitu* XPS [8] and *insitu* TEM [9].

Isothermal decomposition follows a sigmoidal shape, with an induction period followed by an acceleratory period and then a decay period. The induction and acceleratory periods correspond to Al nucleation and growth, respectively [5], while the decay period corresponds to hindered phase transformation. Nucleation is initiated at surface defects resulting in phase transformation and volume change, further disrupting the inhibiting Al₂O₃ surface layer and accelerating the dehydrogenation process [8]. This Al₂O₃ layer can also be disrupted by mechanically alloying dopants that provide a passage for H₂ transfer from AlH₃ to the gas phase [10].

Although other kinetic models [11] can be utilized to predict the dehydrogenation process, the Kolmogorov–Johnson–Mehl Avrami (KJMA) model, also known as the Avrami–Erofeev (AE) phase transformation model [12–14], has been widely accepted. This model has been utilized to predict α -AlH₃ long term stability [12] and storage risk analysis [14].

Experimental

The as received, stabilized α -AlH₃ powder (ATK Launch Systems Inc., [6]) was characterized by Thermal Gravimetric Analysis (TGA), Scanning Electron Microscopy (SEM) and X-ray diffraction (XRD). The α -AlH₃ is handled and stored in air at 21 °C in a climate controlled laboratory environment, with no additional precautions, since the stabilized powder shows no decomposition at 43 °C in air for at least 300 days [6]. This α -AlH₃ is also utilized in a wearable α -AlH₃ fuel cell system described below.

TGA

As received, stabilized α -AlH₃ powder samples, 10 mg, were loaded into a platinum pan for use with the TGA (Perkin Elmer

Pyris 1). The sample enclosure was pretreated with N₂ (99.999% purity) for a 2 h period to purge ambient air. After the purge, the sample was heated at 10 °C min⁻¹ in N₂ to the final desired temperature. Additional information on thermal desorption methods and measurements of α -AlH₃ are available in literature [8,14].

SEM

As received, stabilized α -AlH₃ powder samples and α -AlH₃ powder samples after TGA isothermal exposure at 135 and 155 °C, were directly observed using a SEM (Hitachi 4700). Samples were uncoated and analyzed using an accelerating voltage of 10 kV and a beam current of 10 mA.

XRD

As received, stabilized α -AlH₃ powder samples and α -AlH₃ samples after TGA isothermal exposure at 90 and 185 °C, were loaded in a X-ray powder diffractometer (Bruker D8 Advance XRD) and characterized with Cu-K α radiation at a beam intensity of 35 kV and 30 mA at 21 °C, utilizing a flat glass sample holder. Based on previous literature [14], oxidation beyond initial surface passivation starts at temperature > 377 °C, and thus no special sample holders are required.

Fuel cell system

The wearable α -AlH₃ 20 W fuel cell system developed is shown in Fig. 1a with the dimensions, while the setup is shown in Fig. 1b. This system is divided into an AlH₃ cartridge compartment, a stack with balance of plant, and a battery section. The cartridge compartment and battery section are mounted on hinges to the stack section, enabling the system to be flexible in the transverse plane. The stack utilizes an air cooled 40 W PEM stack (FCgen[®]-micro, Ballard Power Systems) with an open cathode flow field. The balance of plant includes a centrifugal cooling fan, H₂ manifold with valve to purge water and N₂ from the H₂ circuit, and power electronics. The 101 g cartridge, Supplementary Figure S2, comprised of 82 g α -AlH₃ in an Aluminum can, is inserted into a vacuum insulated chamber that feeds H₂ to the fuel cell stack via a H₂ manifold. This chamber also contains a thin film polyimide heater that increases cartridge temperature for H₂ dehydrogenation in response to the load requirements [4]. The battery section contains a battery management system to control the charge and discharge of the 40 Wh Li-ion battery pack. The battery pack provides start up, instant and bus power, along with load following and energy storage ability. The weight and volume of this prototype system is 0.91 kg and 0.74 L respectively. For normal operation the system will be placed in a wearable pouch with limited ventilation.

The system performance was measured utilizing an ARBIN 5A PBT electronic load for the following tests: 1) Constant Current, 1.3 A; 2) Constant Current in a Pouch, 1.3 A; and 3) Cycling Current in a Pouch, 0.3 A for 45 s/2.3 A for 15 s. The measured energy is the sum of the energy from the internal battery and the cartridge. The battery energy is calculated

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