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Metal hydride material requirements for automotive hydrogen storage systems

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ABSTRACT

The United States Department of Energy (DOE) has published a progression of technical targets to be satisfied by on-board rechargeable hydrogen storage systems in light-duty vehicles. By combining simplified storage system and vehicle models with interpolated data from metal hydride databases, we obtain material-level requirements for metal hydrides that can be assembled into systems that satisfy the DOE targets for 2017. We assume minimal balance-of-plant components for systems with and without a hydrogen combustion loop for supplemental heating. Tank weight and volume are driven by the stringent requirements for refueling time. The resulting requirements suggest that, at least for this specific application, no current on-board rechargeable metal hydride satisfies these requirements.

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1. Introduction

Energy-efficient cars emitting zero greenhouse gases: this ultimate goal makes fuel cell vehicles running on hydrogen a very attractive concept. Furthermore, light-duty vehicles represent a large fraction of the commercial vehicle market, so it is important to focus on this class of vehicles. Due to the special volume and weight constraints for this sector of the market, as well as the expectations generated from the performance of currently existing vehicles, hydrogen-powered vehicles must satisfy certain performance requirements if they are to compete with light-duty vehicles based on other technologies.

Starting with performance demands on the vehicle, the United States Department of Energy (DOE) has produced a series of performance targets for the hydrogen storage system itself [1]. The targets are divided into phases, to guide the pace of technology improvement by research and development teams. The three phases correspond to 2010, 2017, and Ultimate Full Fleet targets.¹ The ultimate targets are designed to describe vehicles that would be competitive with other light-duty vehicles in the market.

Even though the DOE hydrogen storage targets have been modified over recent years, the basis for the targets remains the same: to develop a vehicle operating on hydrogen whose performance is not compromised when compared to today's

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¹ The phases used to be 2010, 2015, and Ultimate Full Fleet. The 2015 phase has been changed to 2017.

current vehicles. Currently, there are over 20 targets listed by DOE that deal with the characteristics of the on-board hydrogen storage system as it relates to vehicle performance, energy efficiency, safety, cost as well as the system's overall size and weight. While all the targets are important, some, such as volumetric and gravimetric density and cost, are among the more difficult targets to meet for many hydrogen-based systems.

In this paper we use simplified storage system and vehicle models, combined with minimal balance-of-plant components, to obtain the material-level requirements for metal hydrides to satisfy the 2017 system-level targets. Because we are dealing with hypothetical materials, some additional assumptions are needed in order to complete the analysis, and we turn to metal hydride databases to anchor these assumptions. Sec. 3 details the approach and Sec. 4 makes explicit some additional assumptions. In Sec. 5 these assumptions come together to result in material requirements.

2. Hydrogen storage materials and baseline metal hydride systems

Because hydrogen is a lightweight gas at normal conditions, compressing it to high pressures (350 and 700 bar) and liquefying it at extremely low temperatures (20 K) have become common storage methods. While both of these “physical” storage methods are being applied to on-board hydrogen vehicles today, neither can meet all of the DOE targets and many scientists and researchers are actively pursuing other options.

One of these options that has and continues to be evaluated is storage of hydrogen on solid materials. This can include adsorption on high-surface materials by low energy molecular bonding (physisorption) or by absorption into materials with stronger chemical bonding (chemisorption). Like physical storage methods, solid storage methods also have deficiencies for automotive applications. Physisorption systems, while not requiring liquid hydrogen temperatures, still require cryogenic temperatures near to that of liquid nitrogen (77 K) to store enough hydrogen for today's vehicle requirements. Chemisorption systems can be divided into two material classes: reversible and non-reversible systems. For automobile applications, the non-reversible systems are systems based on materials that cannot be readily recharged with gaseous hydrogen at a fueling station. These systems, that we often refer to as chemical hydride systems, require off-board recharging where more complex processing conditions can be carried out to successfully rehydrogenate the material [2,3]. Another class of materials, reversible materials, typically can be recharged with hydrogen under conditions typically found at some of today's gaseous or liquid refueling hydrogen stations. These systems are often referred to as metal hydride systems. Table 1 shows some of the candidate adsorbent, chemical hydride and metal hydride materials that are currently being evaluated for DOE by the Hydrogen Storage Engineering Center of Excellence (HSECoE) [4].

Table 1 – Hydrogen storage materials considered by the HSECoE, currently and in the past.

	Tier 1 Developed materials	Tier 2 Developing materials	Materials no longer considered
Adsorbents	AX-21 ^a MOF 5 ^b	Pt/AC-IRMOF 8 ^c PEEK ^d	MOF 177 ^b
Chemical Hydrides	NH ₃ BH ₃ (l) NH ₃ BH ₃ (s) AlH ₃	LiAlH ₄	
Metal Hydrides	NaAlH ₄ TiCr(Mn)H ₂	Mg(NH ₂) ₂ + MgH ₂ + 2LiH	MgH ₂ 2LiNH ₂ + MgH ₂ Mg ₂ NiH ₄

a Activated carbon.
b Metal organic framework.
c Isoreticular metal organic framework.
d Polyether ether ketone.

2.1. Metal hydride materials

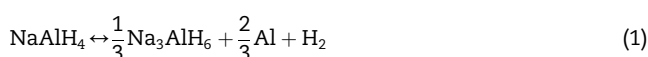
Over the past 30 years several hydrogen storage systems based on reversible metal hydrides have been evaluated for vehicle applications [5]. Most of these have involved either pure metals (like Mg) or, more commonly, intermetallic alloys (like LaNi₅, TiCrMn, and FeTi) as metal hydrides. While hydrogen storage systems based on these metal hydride materials were often able to reversibly store and deliver hydrogen suitably for several industrial vehicle applications [6–8], most of these systems were considered as being much too heavy for today's commercial vehicle market.

In the late 1990s some hydride materials, like NaAlH₄, which were widely considered to be non-reversible, were shown to be reversible under reasonable operating conditions with the addition of certain additives [9]. Since then, several demonstration projects [10–14] have evaluated NaAlH₄ as a possible reversible metal hydride for vehicle applications. While improvements in the overall weight of a storage system appears possible using higher capacity sodium alanate material, the slower absorption and release rates for this material, coupled with higher heats of reaction or enthalpy, has resulted in little overall improvement of these systems for vehicle applications.

2.2. Baseline systems

Two metal hydride systems have received most attention as potential on-board hydrogen storage systems for light-duty vehicles. The first system is based on sodium alanate (NaAlH₄) while the second system is based on a high-pressure metal hydride (Ti_{1.1}CrMn). These two metal hydrides, because of significantly different operating conditions and kinetics of hydrogen absorption reactions, require substantially different system configurations.

Hydrogen absorption/desorption in the sodium alanate system is governed by the following reactions:



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