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Review Article

Destabilization of lithium hydride by the substitution of group 14 elements: A review

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ARTICLE INFO

Article history:

Received 1 September 2015

Received in revised form

4 January 2016

Accepted 18 February 2016

Keywords:

Hydrogen storage

Mechanical alloying

Thermodynamics

Light element hydrides

ABSTRACT

Lithium metal has been considered as an important candidate in energy sector due to its unique properties of hydrogen binding and electrochemical merits. Lithium hydride has high hydrogen capacity (12.7 mass %), however, it requires 900 °C for 0.1 MPa desorption pressure due to the presence of strong ionic bond which generates many thermodynamic constraints. These thermodynamic constraints can be removed using a third element that forms a new compound with Li upon dehydrogenation of LiH. This new compound should have more stability than elemental Li. Group 14 elements of periodic table may have the ability to fulfil this criterion. The paper presents a comprehensive review based on our recent studies on hydrogen storage properties of a Li-M-H system.

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E-mail address: tichi@hiroshima-u.ac.jp (T. Ichikawa).<http://dx.doi.org/10.1016/j.ijhydene.2016.02.069>

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Introduction

Energy has always been a back-bone for all the developments, modern civilization and economic growth worldwide. That is why, most of the biggest problem on this planet is somehow dependent on so called “Energy crisis”. Energy crisis is nothing but termed as more demand to supply ratio and the shortage of fossil fuels. The seriousness of this crisis is understood by the experts, policy makers and researchers globally and many of them are actively engaged to replace the current fossil fuel infrastructure with a new infrastructure based on alternative sources of energy e.g. solar energy, geothermal energy, ocean energy. Hydrogen energy is one of the leading contender which fulfils required characteristics to replace the current energy infrastructure i.e. plenty of availability and its environmental compatibility. Hydrogen is generally considered as secondary energy, referred as energy carrier, can be used to store and deliver the energy as per the requirement. It has three times higher energy content than gasoline and other similar fuels gravimetrically, but its volumetric energy content is low. The characteristics of this unique gas form a bottleneck to store it efficiently so that it can be used onboard in the transportation sector especially for light motor vehicles due to their small size and limited space for fuel storage. Currently hydrogen is being stored as a compressed gas that requires high pressure of 70 MPa or cryogenic liquid that used extremely low temperature of 20 K. Numerous studies have shown that solid state storage provide much safer and viable option than the other two mentioned above, provided it fulfil the storage goals as documented by U.S. department of energy (DOE) [1], according to which 7.5 wt% or 70 kg/m³ densities are required. Light element hydrides e.g. MgH₂ and LiH meet the above requirements. MgH₂ containing 7.6 wt% H₂ requires 300 °C as a working temperature i.e. more stable than required.

Lot of work has been devoted in literature to destabilize MgH₂ [2,3]. The condition is rather poor with LiH even if it has higher content of hydrogen i.e. 12.7 wt%. It requires 900 °C for 0.1 MPa desorption pressure due to high enthalpy of dehydrogenation of 181.2 kJ/mol H₂. The presence of strong ionic bond is responsible for the high stability of LiH [4,5]. A strong activation barrier is formed for atomic motion due to high directionality of the above mentioned ionic bonds, which in result create many thermodynamic constraints [6] and lower the kinetics of sorption reaction. This is the reason LiH could not be considered a practical storage media for long time and limited to be used as the model system for study of basic properties and simulation purposes to design more complex systems [7,8]. Reilly et al. were the first who proposed a thermodynamic alteration by forming alloys with Mg in dehydrogenated/hydrogenated state [9,10] with a well-known example of destabilized system Mg₂NiH₄ [11]. Using the same concept, Vajo et al. tried to destabilize LiH using a third element Si which is capable to form a new compound with lithium upon dehydrogenation [6,12]. This can be explained through Fig. 1. Suppose some hydride say MH desorbs hydrogen to reach its dehydrogenated state M with enthalpy x without any destabilization effect; x value would be much higher. If an intermediate state of MA_{*n*} is introduced which is

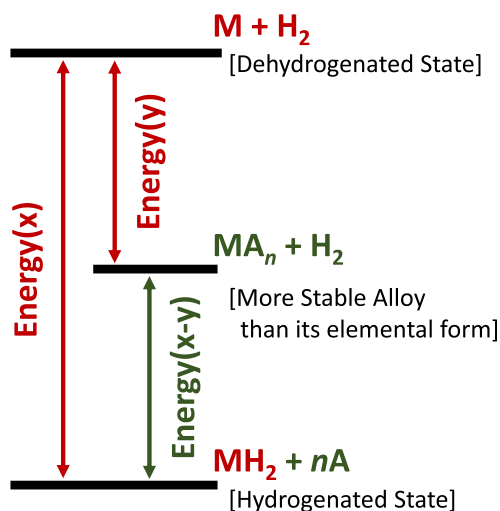


Fig. 1 – Schematic of destabilization process of a hydride MH using third element A.

more stable in comparison to pure M with the energy of formation say y , the sorption reaction will be cycled between MA_{*n*} + H₂ and MH + n A with a less enthalpy which would be equal to the difference $x-y$. This in turn will destabilize MH system effectively. Thus the reduction in stability of MH is directly dependent on the relative stability of M-A system. On the basis of electrochemical data, it is found that Group 14 elements require positive potential for the lithiation [13–16], which in turn suggests the more stable nature of Li-M alloy (M = Group 14 elements i.e. C, Si, Ge, Sn) in comparison to pure Li. This motivated us to study the destabilization effect of LiH using Group 14 elements. In this paper we are reviewing our recent results on the destabilization of LiH using C, Si, Ge, Sn.

Li-C-H system

Carbon is the first element of Group 14 and has been considered as important material for energy storage in the form of hydrogen storage [17–21] and battery electrode [22–24]. Particularly in hydrogen storage, the C-H system has high hydrogen content of more than 4 wt% by forming nano structure, but at the same time it requires more than 700 °C to squeeze out all the hydrogen due to very strong covalent bonds between C and H [25]. Although it looks inappropriate to discuss the C-H system at a first sight in a paper devoted to destabilization of LiH. But surprisingly and interestingly, Li-H and C-H covalent bonds are found to be destabilized [26–28] when reached to a closed contact during intercalation by the milling process. This intend us to place the C-H system as unavoidable part of discussion while making efforts to destabilize LiH using carbon. However we will keep our main focus on the destabilization of C-H covalent bonds.

In order to study Li-C-H system, we started with different graphite materials prepared by 3 h milling using 3 different forms of carbon as starting material: (i) as received graphite (G); (ii) 8 h pre-milled graphite under 1 MPa H₂ (HG) and (iii) 8 h pre-milled graphite under 1 MPa Ar (AG). Graphite is highly

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