



International Scientific Conference “Environmental and Climate Technologies”, CONECT 2017,
10–12 May 2017, Riga, Latvia

Implementation of hydrogen plasma activation of Mg powder in two steps hydrogenation

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Abstract

Development of technologically and economically feasible solutions for hydrogen storage stimulates progress in hydrogen economy. High gravimetric and volumetric capacities of magnesium hydride makes it promising material capable to accelerate implementation of hydrogen-based technologies in our daily life. However, widely discussed limitations of sorption kinetics and thermodynamic properties must be managed in MgH_2 . This work investigates two steps hydrogenation when process of hydrogen absorption is followed after hydrogen plasma activation. Such technique initiates creation of new channels for enhanced hydrogen sorption. Moreover, synthesis of negligible amount of hydride acts as positive factor for further hydrogenation.

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Peer review statement - Peer-review under responsibility of the scientific committee of the International Scientific Conference “Environmental and Climate Technologies”.

Keywords: hydrogenation; MgH_2 ; hydrogen plasma; magnetron sputtering

1. Introduction

Growth of hydrogen economy depends on development of economically and technologically viable hydrogen production, storage, transportation and energy generation. Safety and efficiency of hydrogen storage system are of importance for deeper technological integration into the global market [1–3].

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As cryogenic liquid and pressurized gas require additional safety assurance, hydrogen storage in solid state materials (e.g. metal hydrides) becomes an advantageous solution for safer hydrogen storage in mobile and stationary systems [1, 3, 4].

Magnesium hydride (MgH_2) is considered as promising material for hydrogen storage possessing relatively high gravimetric capacity (7.6–7.66 wt%) and volumetric density (110 g H/l) [3, 5, 6]. Additionally, it is worth mentioning that prevalence of magnesium is reasonably large making this light metal inexpensive and marketable, e.g. there is 2.76 wt% of Mg in the earth crust [6] and only iron, oxygen and silicon are more abundant in the Earth [8].

On the contrary, successful adoption of MgH_2 for hydrogen storage into everyday life will be possible when limitations of hydrogenation/dehydrogenation kinetics and thermodynamic properties are enhanced.

MgO / $\text{Mg}(\text{OH})_2$ layer formation on the surface of Mg powder hinders hydrogen absorption. Moreover, if rapid growth of magnesium hydride is ensured further diffusion of H slows down significantly. In this case H_2 diffusion coefficient in MgH_2 decreases to $1.5 \times 10^{-16} \text{ m}^2/\text{s}$ compared to $4 \times 10^{-13} \text{ m}^2/\text{s}$ of Mg [8, 9]. Dehydrogenation efficiency is restricted due to previously mentioned sluggish hydrogen diffusion in hydride, high temperature and energy required to break Mg-H bond and cause nucleation of Mg and formation of H_2 on the Mg surface. In thermodynamic point of view, strong ionic bond between magnesium and hydrogen leads to absorption and desorption processes only at high temperatures. Because of stability of magnesium hydride, hydrogen is released at 300 °C and 1 bar while industry requires temperature reduction to 60–120 °C [3, 8, 9].

In order to overcome this challenge, ball milling, alloys of metals or metal oxides, catalysts and thin film hydrides are used [10]. Generally, production of MgH_2 powder is based on milling under hydrogen atmosphere and use of catalytic powder. Despite of positive impact of catalysts, such as Pd, introduction of new material may have some negative aspects for the hydride as well, e.g. chemical poisoning, leaching or corrosion. Furthermore, removal of contaminants caused by Pd may require extra investments despite of the fact that Pd itself is an expensive material [10–12].

Current research aims to implement hydrogenation of Mg powder (315–630 μm) in two steps: activation under hydrogen plasma (1) and hydrogenation at high temperature and pressure (2). Consequently, an effective hydrogenation excluding, for example, mechanical milling or catalysts is sought. Comparative analysis between hydrogenated samples of pure and activated Mg is done.

2. Experimental techniques

Two steps hydrogenation of Mg powder (315–630 μm , 99 %) was carried out in this work. First of all, powder activation was applied under hydrogen plasma. Mg powder was placed on the holder mounted in front of a magnetron sputtering system equipped with Mg target (99.99 %). Powder activation started when hydrogen plasma was generated. Bombardment of plasma consisting energetic particles (ions, atoms, molecules, etc.) could induce several processes, i.e. structural surface alterations and formation of thin films of MgH_2 .

Table 1. Experimental parameters of two steps hydrogenation.

Stage	Characteristic	Parameter
Plasma activation	Gas	Hydrogen 5.0
	Working pressure	16 Pa
	Power	240 W
	Time of treatment	3 hours
	Distance between powder and magnetron	4 cm
	Power supply	Pulsed DC
Hydrogenation	Gas	Hydrogen 5.0
	Pressure	100 bar
	Temperature	300 °C
	Time	14 days

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