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Synergistic catalytic effect of SrTiO₃ and Ni on the hydrogen storage properties of MgH₂



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ABSTRACT

Study on the synergistic catalytic effect of the SrTiO₃ and Ni on the improvement of the hydrogen storage properties of the MgH₂ system has been carried out. The composites have been prepared using ball milling method and comparisons on the hydrogen storage properties of the MgH₂ – Ni and MgH₂ – SrTiO₃ composites have been presented. The MgH₂ – 10 wt% SrTiO₃ – 5 wt% Ni composite is found to has a decomposition temperature of 260 °C with a total decomposition capacity of 6 wt% of hydrogen. The composite is able to absorb 6.1 wt% of hydrogen in 1.3 min (320 °C, 27 atm of hydrogen). At 150 °C, the composite is able to absorb 2.9 wt% of hydrogen in 10 min under the pressure of 27 atm of hydrogen. The composite has successfully released 6.1 wt% of hydrogen in 13.1 min with a total dehydrogenation of 6.6 wt% of hydrogen (320 °C). The apparent activation energy, E_a , for decomposition of SrTiO₃-doped MgH₂ reduced from 109.0 kJ/mol to 98.6 kJ/mol after the addition of 5 wt% Ni. The formation of Mg₂Ni and Mg₂NiH₄ as the active species help to boost the performance of the hydrogen storage properties of the MgH₂ system. Observation of the scanning electron microscopy images suggested the catalytic role of the SrTiO₃ additive is based on the modification of composite microstructure.

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Introduction

Rapid growth on the global energy demands has risen at an alarming rate. U.S Energy Information Administration (EIA) has projected an increment of 28% in world energy consumption between 2015 and 2040 [1]. In this context, utilization of energy from renewable energy sources like sun, wind and hydro has been recognized as one of the excellent approaches to tackle the issue. As an indicator, REN21 reported that about 19.3% of share on the total final energy consumption in 2015 came from renewable energy sources [2].

Energy storage is important to improve the performance of renewable energy power systems due to their intermittent power sources and independent fluctuation from the demand. Storage improves the performance of the systems by reducing the mismatch between supply and demand. It also enhances the reliability and the efficiency of the systems that improve the cost-effectiveness of the power systems [3] and as a key component in energy conservation.

Hydrogen has come into view as a benign energy storage due to its high energy content that exceeds petroleum by a factor of three [4]. Furthermore, the waste product of hydrogen combustion is water vapour, which is environmentally friendly. However, this technology is in its commercial infancy except for the compressed hydrogen storage. However, the compressed hydrogen storage requires high working pressure up to 70 MPa to achieve 4.8 wt% of hydrogen

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storage capacity [5]. Meanwhile, for the liquid hydrogen storage, the limitation on this method is due to low-temperature requirement (-253 °C) that will cause high energy lost (up to 30% during filling) as well as a short period of storage due to boiling [6]. As for the solid state hydrogen storage, it has received tremendous attention due to its high storage capacity and high volumetric density [7–9]. For instance, MgH₂, one of the lightweight hydrides is able to store up to 7.6 wt% of hydrogen with a volumetric density of 110 g/L [10–12]. However, these superiorities are nullified by its sluggish hydrogen desorption/absorption kinetics and high thermal stability that limit its commercialization [13,14].

Numerous studies were conducted to overcome the disadvantages of the metal hydrides as the hydrogen storage material. Many researchers have certified the beneficial effect of the catalyst doping on the hydrogen storage properties of the metal hydrides. In this context, transition materials like Cu, Co, Fe, Nb, Ni, Ti and V are commonly studied [10,15].

As one of the commonly studied catalysts, Ni has been corroborated as one of the promising catalysts for the MgH₂ [16-18]. For instance, Cova et al. [17] asserted that Ni was able to enhance the hydrogen sorption kinetics of the MgH₂ system. They reported that the MgH₂ - Ni composite was able to absorb about 5 wt% of hydrogen in 25 s at 275 °C and similarly to release about 5 wt% of hydrogen in 8 min under similar operating temperature. Furthermore, Zaluska et al. [19] studied the catalytic effect of 35 wt% of Mg₂Ni on the improvement of hydrogen storage properties of the MgH₂ system. Interestingly, about 5 wt% of hydrogen was able to be absorbed and desorbed by the composite at 220 °C. Meanwhile, Shahi et al. [20] reported that MgH₂ with 5 wt% of Ni started to decompose at 340 °C and was able to absorb 5.0 wt% of hydrogen at 270 °C in 15 min. As for Mao et al. [21], they showed that the addition of Ni nanoparticles into 2LiBH4+MgH2 destabilized system was able to improve the dehydrogenation kinetics at 350 °C and 400 °C. In addition, the inclusion of Ni had improved the rehydrogenation capacity; from 4.4 wt% to 5.3 wt% of hydrogen at 400 °C.

Meanwhile, positive effects on the hydrogen storage properties done by their chlorides, fluorides and oxides like NiCl₂, TiCl₃, VCl₃, NbF₅, TaF₅, TiO₂ and ZrO₂ were reported by previous researchers. Mao et al. [16] had substantiated the catalytic effects of CoCl₂ and NiCl₂ on the hydrogen storage properties of MgH₂. They reported that the addition of the catalysts had reduced the activation energy from 158.5 kJ/mol (milled MgH₂) to 121.3 kJ/mol (with CoCl₂) and 102.6 kJ/mol (with NiCl₂). Meanwhile, Malka et al. [22,23] studied the property of MgH₂ sorption kinetics with the addition of TiCl₃, VCl₃, NbF₅ and ZrF₄. They reported that the lowest desorption activation energy of the studied systems was the ZrF₄ with a value of 77 kJ/mol and followed by 83 kJ/mol for the NbF₅, 85 kJ/mol for the TiCl₃, and 97 kJ/mol for the both of TaF₅ and VCl₃. As for the non-catalyzed MgH₂, the reported value was 152 kJ/mol. On the other hand, Mao et al. [24] found that the introduction of NbF5 into LiBH4-MgH2 system was able to enhance the rehydrogenation capacity by 1.6 wt% at 400 °C. Meanwhile in other works, Mao et al. [25,26] found that the addition of TiF₃ to LiAlH₄ - MgH₂ and LiAlH₄ - MgH₂ - LiBH₄ destabilized systems had significantly reduced the initial decomposition temperature of the systems. The initial temperature was reduced from 150 $^\circ$ C to 60 $^\circ$ C for the LiAlH₄ – MgH₂ system and from 130 $^{\circ}$ C to 60 $^{\circ}$ C for the LiAlH₄ – MgH₂ – LiBH₄ system. As for the oxide catalysts, Chen et al. [27] found that a composite made of MgH₂ and nano-size of ZrO₂ with a molar ratio of 95:5 had an incredibly low absorption activation energy with a value of 13.1 kJ/mol. Thanks to this effect, the composite was able to absorb 4 wt% of hydrogen at 25 °C. Pandey et al. [28] reported that the incorporation of 5 wt% of TiO₂ nanoparticles (50 nm) was able to reduce the absorption activation energy to 52.7 kJ/mol. The composite was able to absorb 5.1 wt%. of hydrogen in 6 min at 250 °C and 50 atm. Meanwhile, recently Pukazhselvan et al. [29] found that a composite made of MgH_2 and 3 wt% of $0.5TiO_2$ had a desorption activation energy of 110.9 kJ/mol. Deduction based on the presented DSC profile shows that the composite started to decompose at 280 °C.

Non-reactive additives that provide promising catalytic effects despite the catalysts above are worth mentioning. These additives are known for its catalytic effect by acting as a lubricant and cracking agent that will introduce the pulverization effect for smaller metal hydride particle sizes [10,30]. In this regard, Sulaiman and Ismail [31] corroborated that the inclusion of 5 wt% of carbon nanotubes (CNTs) to the MgH₂ -10 wt% K_2NiF_6 composite was able to reduce the onset decomposition temperature to 245 °C. Luo et al. [32], on the other hand, found that the hydrogen absorption of the MgH₂ – 5 wt% NbF₅ composite was increased by 0.5 and 0.8 wt% at 200 °C and 100 °C with the addition of 5 wt% of single-wall CNTs. Meanwhile, Huang and Chuang [33] reported that the addition of 5 wt%. of single-wall CNTs to the activated composite of $MgH_2 - 5$ wt% ZrO_2 demonstrated a remarkable result. The composite was able to absorb 4.0 wt% of hydrogen at 25 °C.

Ceramics like silicon carbide (SiC) and titanium carbide (TiC) are also known as promising additives for the MgH₂ [34–37]. Given the hardness of the materials (8–9 Mohs) [38,39], many studies have corroborated the favourable catalytic effects of the SiC and TiC in improving the performance of the hydrogen storage properties of MgH₂. Referring to Ranjbar et. Al [35], they found that the SiC was able to reduce the decomposition temperature by 20 °C as well as promoting the hydrogen absorption capacity by 0.7 wt% at 350 °C. In another study [40], they found that the incorporation of 10 wt % of Ni to $MgH_2 - 5$ wt% SiC was able to reduce the decomposition temperature by 80 °C as well as increasing the hydrogen desorption capacity to 4.7 wt% at 300 °C under 0.1 atm. Imamura et al. [41] reported that 10 mol% and 75 mol % of SiC were able to reduce the decomposition temperature of the MgH₂ – SiC composite to 207 $^{\circ}$ C and 164 $^{\circ}$ C, respectively. Meanwhile, Tian and Shang [37] claimed that a composite made of $MgH_2 - 2$ mol% of TiC that was cryo milled for 8 h and ball milled for another 60 h had a decomposition temperature of 164 °C. In addition, Fan et al. [36] reported that the incorporation of 10 wt% of TiC to MgH2 had caused 95 °C of decomposition temperature reduction. The decomposition activation energy was calculated to be 144.6 kJ/mol, which is 46.7 kJ/mol lower than the milled MgH₂. As for El-Eskandarany et. al [42], they reported that 10 wt% of ZrNi₅ powder which is inert and doped with MgH₂ was able to absorb 3.1 wt% of hydrogen in 179.8 min at 50 °C. Recently Yahya and Ismail [43]

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