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Ammonium phosphate as promised hydrogen storage material

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ABSTRACT

Most of the reported hydrogen storage materials are either expensive or based on synthetically complicated compounds. Ammonium phosphates are cheap and available product; it is being used as fertilizer. These compounds could be utilized as hydrogen storage materials. Stoichiometrically, mono-, di- and tri-ammonium phosphate can lead to evolve 3.875, 7.867 and 11.903 hydrogen equivalent, respectively. Using Pt/C (20 wt %) leads to produce 17.28, 11.35 and 10.15% from the equivalent hydrogen in the mono-, di- and tri-ammonium phosphate, respectively. Analyzing the obtained gases by GC confirms evolution of pure hydrogen. Moreover, ion chromatography detects and ions in the liquid which indicates hydrolysis of the ammonium phosphate. Ni/C and Co/C NPs can be exploited as catalysts; Ni/C containing 20 wt% metal reveals comparable results to Pt/C. Overall, this study opens promising avenue to develop new catalysts to enhance the hydrolysis of ammonium phosphate to produce hydrogen. Comparing to the reported hydrogen storage materials, ammonium phosphates possess comparable hydrogen content 97.28, 118.3 and 115.72 kg/m³ for mono-, di- and tri-ammonium phosphate, respectively which is very close to the MgH₂ (115.5 kg/m³) and not so far from ammonia borane (148.2 kg/m³). However, in contrast to these compounds, hydrogen can be released from the introduced storage materials by hydrolysis only using the proper catalyst. Furthermore, the hydrolysis process results in producing different acids; nitrous, nitric and phosphoric acid which should be considered in the reactor construction precautions.

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Introduction

Hydrogen has a high energy density by weight between 120 and 142 MJ/kg, compared to 44 MJ/kg for petroleum. Storage of hydrogen considers the bottleneck of wide spreading of hydrogen energy. Hydrogen can be stored in a high-pressure storage tank and cryogenic liquid storage tanks. The previous routes for hydrogen storage facilities are complicated because of its low boiling point ($-252.87\text{ }^{\circ}\text{C}$) and low density in the gaseous state (0.08988 g/L) at 1 atm. High-pressure storage of hydrogen gas is limited by the weight of the storage canisters and the potential for developing leaks. Liquid hydrogen requires the addition of a refrigeration unit to maintain a cryogenic state [1] thus adding weight and energy costs, and a resultant 40% loss in energy content [2]. The energy density of hydrogen can be improved by storing hydrogen in a liquid state. However, the issues with liquid hydrogen tanks are hydrogen boil-off, the energy required for hydrogen liquefaction, volume, weight, and tank cost. The energy requirement for hydrogen liquefaction is high; typically, 30% of the heating value of hydrogen is required for liquefaction. Moreover, storage of hydrogen in liquid or gaseous form poses important safety problems for on-board transport applications. Recently, researchers are doing high efforts to find solid materials being capable to carry hydrogen and then reversible. Some of the hydrogen storage material commandments are the capacity of the hydrogen storage material should be $\sim 6.5\text{ wt. \%}$ and 65 g/L hydrogen available, the decomposition temperature to be between 60 and $120\text{ }^{\circ}\text{C}$, for realization the economic and environmentally being considered the cost of storage and keep low toxicity. Chemical storage materials with high hydrogen contents are highly promising as hydrogen sources for fuel cells. Among them, boron- or nitrogen-based compounds, such as $\text{LiNH}_2\text{--LiH}$, Li/NaBH_4 and N_2H_4 , etc., have attracted much attention [3,4].

Many hydrogen storage materials have been studied for safe use, efficiency, and economics of extracting hydrogen from these materials such as ammonia–borane complex [5–7], metal hydrides [8–12], graphene-based materials [13–16], and others [17,18]. Among the introduced hydrogen storage materials, ammonia borane (AB) possesses the highest hydrogen content (19 wt%, 148.2 kg/m^3). Hydrogen release from AB can be performed by three main strategies; thermolysis, ionic liquids (ILs)-assist and hydrolysis by the aid of proper catalyst [19–21]. Besides the high temperature required, the thermolysis approach is confounded by the practical disadvantages associated with delivery of a solid material which constrains utilizing it [22,23]. The last strategy depends on using efficient catalysts for hydrolysis of AB in aqueous solution. Overall, up to date, the introduced hydrogen storage materials have not showed satisfactory performance for commercial vehicular application. Recently, researchers made a great effort to synthesize new hydrogen storage materials to fulfill the previous requirements with low synthesizing cost.

Industrially, ammonium phosphate can be produced in three forms based on the ammonium content; mono ($(\text{NH}_4)\text{H}_2\text{PO}_4$), di ($(\text{NH}_4)_2\text{HPO}_4$), and tri ($(\text{NH}_4)_3\text{PO}_4$) ammonium. Specifically, the mono and di-ammonium phosphates are widely

used as fertilizer due to its good stability compared to the third formulation [24]. All of the three forms can be industrially synthesized by reaction between phosphate rock and ammonia [25] or nitric acid (old process) [26]. Stoichiometrically, the hydrogen content in the three forms is 5.22, 6.82 and 8.05 wt% for the mono, di and tri ammonium phosphate, respectively. Compared to the reported hydrogen storage materials, ammonium phosphates are very cheap, abundant and stable material. Therefore, it would be great achievement if the hydrogen could be extracted from these materials and exploiting them as hydrogen storage materials. Chemically, and especially for the tri-ammonium phosphate, dissociation of the ammonium phosphates results in producing ammonia gas. However, the reaction can be controlled to evolve hydrogen if proper effective catalyst is utilized.

It is known that, among the transition metals, nickel reveals effective catalytic activity in the dehydrogenation reactions [27,28]. Moreover, cobalt has shown good performance extraction of hydrogen from some materials [29]. The mass transfer rate of the reactants is a main parameter affecting the heterogeneous catalytic reactions. In other words, enhancing the diffusion rate of the reactants molecules to the surface of the solid catalyst distinctly enhances accelerates the whole process. Consequently, due to its adsorption capacity, carbon showed considerable performance as support for the functional catalytic materials in important applications such as fuel cells [30,31].

Platinum is amazing metal as it shows excellent catalytic activities in many reactions; but unfortunately it is rare metal and very expensive. Here, commercial Pt/C nanoparticles with two different metal contents (20 and 40 wt%) were invoked as effective catalysts to investigate the catalytic hydrolysis dissociation of the ammonium phosphate.

Experimental

Materials

Ammonium phosphates were purchased (SamChun Chemicals Company, South Korea), nickel acetate tetra-hydrate (NiAc, 98% assay Junsei Chemical Co., Ltd, Japan), cobalt acetate tetra-hydrate (CoAc, 99% assay Junsei Chemical Co., Ltd, Japan), poly(vinyl alcohol (PVA, MW = 65000, Junsei Chemical Co., Ltd, Japan) and Pt/C (fuelcells Co. USA) were utilized without any further modification. Distilled water was used in Ni/C NPs fabrication and ammonia phosphate hydrolysis. Pt-loaded carbon nanoparticles (Pt/C) is Vulcan XC-72 Carbon nanoparticles loaded by Pt metal; two formulations were utilized 20 and 40 wt% metal content. These materials have been purchased from FuelCell.com Company (USA).

Ni/C and Co/C NPs preparation

First, aqueous solutions from NiAc, CoAc and PVA (10 wt%) were prepared. The solutions were adjusted and mixed to produce final solutions having different contents from CoAc and NiAc. Typically, four NiAc/PVA solutions were prepared having NiAc content of 10, 20, 30 and 40 wt%. With the same strategy four CoAc/PVA solutions were prepared. The

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