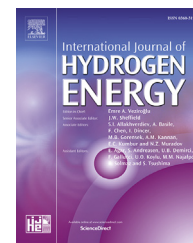


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Pt nanoparticles immobilized in mesoporous silica-coated magnetic nanocapsules: A non-leaching catalyst for hydrogen generation from hydrolysis of ammonia borane

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ABSTRACT

In this work, novel hollow mesoporous silica-coated magnetic nanocapsule that immobilizing Pt nanoparticles (NPs) in its core ($\text{Fe}_3\text{O}_4@\text{SiO}_2@\text{Pt@mSiO}_2$) was successfully synthesized. The obtained catalyst was characterized by TEM, XRD, BET, VSM, ICP and N_2 adsorption-desorption. The $\text{Fe}_3\text{O}_4@\text{SiO}_2@\text{Pt@mSiO}_2$ catalyst was used in the catalytic hydrogen generation from hydrolysis of ammonia borane and showed superior catalytic activity. Moreover, the $\text{Fe}_3\text{O}_4@\text{SiO}_2@\text{Pt@mSiO}_2$ catalyst can be easily recovered by magnetic separation attributed to the supermagnetic Fe_3O_4 NPs imbedded in the inner core. It is worth mentioning that, due to the Pt NPs were immobilized in the core of the nanocapsules and covered with mesoporous silica shell, Pt NPs leaching was efficiently prevented during the catalytic process. Therefore, this work provided a helpful platform for the preparation of noble metal based nanostructured catalysts with excellent activity, accessibility, facile recovery and almost no noble metal leaching.

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Introduction

In recent years, noble metal nanoparticles (NMNPs) based catalysts have been drawn much research attention in various catalytic applications due to their superior catalytic activities [1–3]. However, small NMNPs always suffer from the aggregation because of their high surface energy, this phenomenon subsequently reduces their catalytic efficiency [4–7].

Therefore, various supporting materials were designed to make the NMNPs highly dispersed. For example, various materials including KCC-1 [8,9], mesoporous silica [10,11], mesoporous carbon [12,13], N-doped mesoporous carbon [14,15], graphene materials [16,17] and so on, have been already employed to fabricate NMNPs based catalysts. Although NMNPs can be highly dispersed on these support materials and show excellent catalytic activity, the leaching of the NMNPs always takes place during the catalytic process

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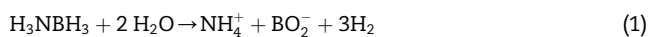
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because the NMNPs were mainly supported on the outer surface of the catalyst support.

Immobilization of the NMNPs in the framework of the support materials is one of the effective methods to prevent the NMNPs leaching. In this respect, some interesting works have been reported [18,19]. Valerie Meille and coworkers reported a Pt immobilized silica for 1-octene hydrosilylation with almost no Pt leaching [20]. Sayari et al. reported the preparation of MCM-41 silica supported monodispersed Pd NPs for the Suzuki coupling reaction with virtually no leaching [21]. Al-Abed et al. presented the synthesis of reactive nano-Fe/Pd bimetallic system-impregnated activated carbon for the simultaneous adsorption and dechlorination of 2-chlorobiphenyl [22]. Imbedding the NMNPs in the core of the mesoporous yolk-shell materials is another effective way to prevent the NMNPs leaching. In this strategy, the NMNPs were restricted in the hollow core, and the reactants can enter the hollow core through the mesoporous channels and react on the surface of the NMNPs. Such catalysts as Pd/Fe₃O₄@ γ -AlOOH [23], YS- γ -Fe₂O₃@G-GS [24], Fe₃O₄@SiO₂@PMO [25], and Bi₂S₃-PPy [26] have been fabricated and detailedly studied. Furthermore, for purpose of making the catalysts easily recovered, magnetic catalyst carrier is also important for a NMNPs based catalyst attributed to its facile recovery process that just using an external magnetic field as compared with centrifugation and filtration [27]. Therefore, in order to prevent the leaching of the NMNPs in the catalytic process and preserve the noble metal resource, developing new functional catalyst support materials and catalyst preparation method combining the above mentioned strategies are necessary.

In the field of energy chemistry, developing new strategy for hydrogen release and storage is a burgeoning research direction; because the hydrogen is regarded as a promising energy carrier owing to its high energy capacity and environmental friendliness [28,29]. Comparing with the physical approaches, including adsorption, compression and liquefaction, the chemical H₂ storage materials are safe and easy to be transported and stored. Ammonia borane (NH₃BH₃, AB) is currently been regarded as a promising solid chemical hydrogen storage materials due to its high hydrogen content (19.6 wt%), nontoxicity, and high stability in solid state and solution [30–32]. Hydrolysis of AB can release 3 equivalent H₂ per mole of AB in the presence of suitable catalysts even at ambient temperature according to the following Eq. (1).



NMNPs, especially Pt NPs based catalysts have been investigated for the hydrolysis of AB these years [33–35]. However, due to the scarcity resources and the high price of the Pt metal, its application in catalytic application field has been greatly restricted. On the other hand, Pt NPs have tendency to aggregate to larger particles on the catalyst support that subsequently reduces the catalytic activity. Therefore, in the view of Pt saving, preventing the Pt leaching and maintaining long time catalytic efficiency, designing functional supporting materials to stabilize Pt and making the catalysts easy to be recycled are crucial.

Based on the above considerations, in this study, hollow mesoporous silica-coated magnetic nanocapsules were fabricated, and Pt NPs were immobilized in its core. The obtained Fe₃O₄@SiO₂@Pt@mSiO₂ catalyst showed a distinct activity for the hydrogen evolution from the hydrolysis of AB. The catalyst could be effortlessly recovered by an external magnet and reused. Due to the protective mesoporous silica shell, Pt leaching was scarcely observed in the catalytic process after running several times. Therefore, this study should provide a useful strategy for preventing noble metal leaching during the catalytic process.

Experimental section

Materials

Ferric acetylacetonate (Fe(acac)₃), triethylene glycol, sodium borohydride (NaBH₄), triethanolamine (TEA), cetyltrimethyl ammonium bromide (CTAB), hexadecyltrimethylammonium chloride (CTAC), tetraethyl orthosilicate (TEOS), chloroplatinic acid hexahydrate (H₂PtCl₆·6H₂O) and ammonia borane (AB) were purchased from Tianjin Heowns Biochem LLC. Ethanol, ammonium hydroxide (NH₃·H₂O) and cyclohexane were provided by Hanlon Bower Pharmaceutical Chemical Ltd Co. The carbon nanotubes (CNTs) were purchased from Shenzhen Nanotech Port Ltd. Co. Deionized water (18.2 M Ω) was used in the whole experimental courses. All of the chemicals were analytical grade and used without any modification.

Synthesis of CNTs@Fe₃O₄ nanotubes

The preparation of CNTs@Fe₃O₄ nanotubes was carried out according to a published method with a slight modification [36]. Firstly, 100 mg of CNTs and 400 mg of Fe(acac)₃ were added to 60 mL of triethylene glycol, followed by ultrasonication for 90 min. Then, the viscous liquid mixture was heated to 278 °C and refluxed for 30 min. Finally, after spontaneous cooling to room temperature, the obtained composites were magnetically separated by a magnet, washed with ethanol several times and dried at 50 °C in a vacuum oven.

Synthesis of silica-coated magnetic CNTs (CNTs@Fe₃O₄@SiO₂)

The CNTs@Fe₃O₄@SiO₂ nanotubes were successfully synthesized by stöber sol-gel deposition approach [37]. Typically, 100 mg of CNTs@Fe₃O₄ and 700 mg of CTAB were dispersed into 160 mL of deionized water and the mixture was sonicated for 30 min. Then, the above mixture was added to 220 mL of anhydrous ethanol and sonicated for 15 min continuously to form a stable dispersion. This was followed by the immediate addition of 1.2 mL of NH₃·H₂O to the as-prepared CNTs@Fe₃O₄ dispersion. Next, TEOS solution (0.4 mL TEOS in 20 mL ethanol) was added dropwise to the mixture under ultrasonication, and the reaction mixture was magnetically stirred for another 12 h. Finally, the obtained solution was suction filtrated and washed with ethanol and water several times. The process resulted in the formation of a uniform and thick layer of silica on every individual CNTs@Fe₃O₄.

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