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Synthesis of graphene with noble metals nanoparticles on its surface

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Abstract

Graphene sheets can be considered as a promising a support material for the catalytic nanoparticles. Here, we present a new approach for the preparation of graphene with noble metals nanoparticle on its surface using supercritical propanol - 2 as a reducing agent for nanocomposites graphene oxide- Pd or Pt nanoparticles. The prepared nanocomposites were characterized by X-ray diffraction analyses (XRD) and transmission electronic microscope (TEM). XRD reveals the face-centered cubic structure of Pd and Pt in the nanocomposites, TEM images show the good spatial distribution of metal nanoparticles on layered graphene sheets.

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Keywords: graphene, nanoparticles

1. Introduction

Graphene stands to be a promising 2D-carbon lattice in the field of material research due to its excellent chemical and physical properties [1-3]. Large surface area, unique thermal and chemical stability, excellent mechanical strength, and superior electrical conductivity make graphene a potential component in various fields such as solar cells [4], storage device [5], sensors [6], energy conversion [7] and so on. In recent years, graphene has been prepared by a variety of techniques, such as mechanical exfoliation of graphite [8], chemical vapor deposition [9] or chemical reduction of graphene oxide (GO) [10]. In comparison to the other methods, chemical reduction of GO is the most efficient approach to prepare graphene due to its low cost and bulk quantity production, as GO can be synthesized

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from inexpensive graphite powders [11, 12]. Graphene oxide has hydroxyl, epoxide, carbonyl and carboxyl groups at the edges and in the plane which make them hydrophilic and allow to work with aqueous solutions [13].

Carbon-supported noble metals have been widely investigated as catalysts, and the results demonstrate that the control of the "catalyst – support" nanostructures is a factor of key importance [14, 15]. The use of a two-dimensional graphene or graphene oxide supports has been proven to be quite effective in enhancing catalytic activity compared to conventional carbon supports, such as carbon black [16]. The favorable behavior of the graphene supports may be attributed to a number of advantages, such as a high dispersion of noble metals catalyst resulting from an enhanced interaction between functionalized graphene or graphene oxide surface and the noble metals nanoparticles and a large surface area of the graphene sheets support [17, 18]. Also abundant functional groups on the surfaces of GO can be used as anchoring sites for metal nanoparticles, it is possible to use them as a support to produce graphene - nanoparticle hybrid nanostructures [19]. However, the chosen reducing agent should be safe, inexpensive and available, and the process of reduction of graphene oxide to graphene should be easily scaled. This work reports the original two-step chemical synthesis and characterization of nanocomposites based on graphene with Pd and Pt nanoparticles on its surface which were obtained by supercritical propanol-2.

2. Experimental section

2.1. Synthesis of graphene - noble metals nanocomposites

GO was synthesized by the oxidation of graphite powder using P_2O_5, $(NH_4)_2S_2O_8$, H_2SO_4, and $KMnO_4$ according to the Hummers' method, where the micrographic powder was mixed with a strong oxidizing agent, filtered, washed, and dried. A portion of graphite oxide powder was dispersed in water by sonication for 1 h, forming stable graphene oxide colloid. Then precursor of corresponding metal (water solutions of H_2PdCl_4 or H_2PtCl_6) was added to graphene oxide dispersion with magnetic stirring. The reaction mixture was stirred for 30 min at room temperature before addition of the reducing agent. 1mL of 0.1M freshly prepared solution of NaBH4 was added slowly to the reaction mixture of precursor–GO suspension under vigorous stirring. The color of the reaction mixture turns at first into dark brown and then to grey depending on the concentration of the H_2PdCl_4 or H_2PtCl_6. The reaction mixture was stirred for another 2 h for the complete reduction at room temperature. The reduction product was separated by filtration and washed with large amounts of water several times to remove residual ions and then dried at 80ºC in a vacuum oven for 6 h.

For the reduction of graphene oxide with nanoparticles of noble metals located on its surface to graphene we used supercritical propanol-2 (SCI) as a reducing agent. A 100mg of graphene oxide with Pd or Pt nanoparticles was dispersed in 5 ml of propanol-2 by sonication for 30 minutes; the dispersion was put in the steel autoclave and maintained in the oven at ~ 300°C for 24 hours.

2.2. Characterization

The content of palladium and platinum nanoparticles on supports was determined by X-ray fluorescence analysis on a Zeiss Jena VRA-30 spectrometer. Powder the X-ray diffraction (XRD) analyses were performed at 25º on a Bruker D8 Advance diffractometer with Cu Kα radiation (λ = 1.54056 E). The diffraction data was recorded for 2θ angles up to 80°.Morphology analyses of samples were carried out on JEOL JEM-1011 transmission electron microscope (TEM).

3. Results and Discussion

The original method for synthesis of nanocomposite of noble metals nanoparticles on the graphene surface (NP/graphene) has been developed in this research. To the contrast to graphene oxide, graphene has only a monolayer of carbon atoms in the sp2- hybridized state and has no functional oxygen groups which could act as nucleation sites for the nanoparticles formation. That is why, it was necessary to develop an original method of immobilization of nanoparticles of Pd and Pt on the graphene surface through the formation of an intermediate nanocomposite of metal nanoparticles on the graphene oxide surface (NP/GO).