



Pd nanoparticles with tunable diameter deposited on carbon nanotubes with enhanced hydrogen storage capacity



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ABSTRACT

In situ synthesis of Pd nanoparticles supported on multiwalled CNT (carbon nanotubes) is reported. The size of the nanoparticles can be easily tuned via application of different experimental conditions. The hydrogen storage properties of Pd supported on CNT at room temperature were examined in the pressure range of 0–50 bar. Carbon material with palladium particles of 3 nm diameter exhibits the highest hydrogen capacity at low moderate pressure than the raw materials and CNT with larger particles size of 17 and 9 nm, respectively. We also propose the mechanism of hydrogen storage in the studied samples.

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1. Introduction

The discovery of carbon nanotubes and carbon nanotubes based materials has inspired scientists for a range of potential applications. Nanotubes have attracted extensive attention for their intriguing and potentially useful structural, electrical, and mechanical properties, have unique atomic structure, very high aspect ratio [1–6]. Composites reinforced by carbon nanotubes have improved electrical conductivity and electrostatic charging behaviour, optical emitting devices, and in lightweight, high strength composites [7].

Hydrogen storage is one of the key issues for the realization of fuel-cell powered vehicles using hydrogen as the energy carrier. The advantages of hydrogen as energy sources lie in the fact that its byproduct is water, and it can be easily regenerated. Owing to the lack of a suitable storage system satisfying a combination of both volume and weight limitations, the use of hydrogen energy technology has been restricted from automobile application [8]. Hydrogen storage remains one of the main challenges in the implementation of a hydrogen-based energy economy. Although several different approaches are being pursued, sorption onto a porous high-surface-area material is a one contender. There is a great interest in finding porous solid materials that can store hydrogen for use in fuel cell vehicles [9]. Carbon-based materials,

due to their low cost and weight, have been considered as suitable adsorption substrates for the reversible storage of hydrogen [10–14]. Hydrogen storage in these materials is still at a research level and is not yet mature enough for industrial application [15–20]. Ideally, these materials would adsorb large amounts of hydrogen gas reproducibly at room temperature and moderate pressure. Among these hydrogen storage materials, nanostructured and porous carbon materials, including carbon nanotubes, graphite nanofibers, activated carbon, and graphite, have received considerable research interest due to their lightweight, high surface areas, and relative chemical stabilities [21]. Many studies have revealed that hydrogen storage capacity is enhanced by added metals e.g. (Ag, B, Ca, Fe, K, Li, Ni, Pd, Pt, Ru, Ti, TiO₂) [22] to carbon structures.

Recently, it was reported that hydrogen storage at room temperature can be improved by a phenomenon known as spillover effect initiated by metal deposited on the adsorbate surface [23]. A mechanism involves the dissociation of H₂ on the metal particles, atomic H diffusion from the particles to the sorbent, and chemisorption of atomic hydrogen on the material's winding sites. A very important attribute of this mechanism is that it is operative at ambient temperature as opposed to storage of H₂ by physisorption, which requires very low temperature [24].

In our study we present in situ deposition of palladium particles on the surface of carbon nanotubes. This method allows loading of well dispersed Pd nanoparticles with controlled diameter on CNT surface. The carbon nanotubes with Pd nanoparticles are shown to have much higher hydrogen adsorption than the pure carbon nanotubes. It is observed that the amount and the size of Pd

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nanoparticles supported on CNT are the crucial parameters determining the hydrogen storage capacity. Different stirring time resulted in the synthesis of different diameter of palladium nanoparticle and different diameter distribution. We believe that this method will pave the new way for the future storage and transport of H_2 and application of H_2 energy.

1.1. Experimental section

1.1.1. Synthesis of CNT with palladium particle

In a typical experiment, the surface treatment process is as follows. Multiwalled CNT (carbon nanotubes) (50.0 mg, from SHEENZEN NANOPORT) were weighed and placed in a 50 mL glass flask with 10 mL of HNO_3 (69%) and 30 mL of H_2SO_4 (96.2%) and

reflux for 24 h in 110 °C. In the typical synthesis of carbon nanotubes functionalized by Pd nanoparticles. 50 mg of CNT and 50 mg palladium acetate were dispersed in 150 ml of acetone and sonicated for 2 h. After sonication process, the mixture was stirred for 24, 48, 72 h respectively. Finally, each sample was dried in air at 100 °C for 24 h.

1.1.2. Characterization

Transmission electron microscopy (TEM) characterization of the samples were performed on a Tecnai F30 electron microscope operated at an acceleration voltage of 200 kV. Hydrogen adsorption capacity was measured using a Sievert-type volumetric apparatus (IMI Hiden Analytical Ltd). The TGA (thermogravimetric analysis) measurements were carried out in a DTA-Q600 SDT instrument TA

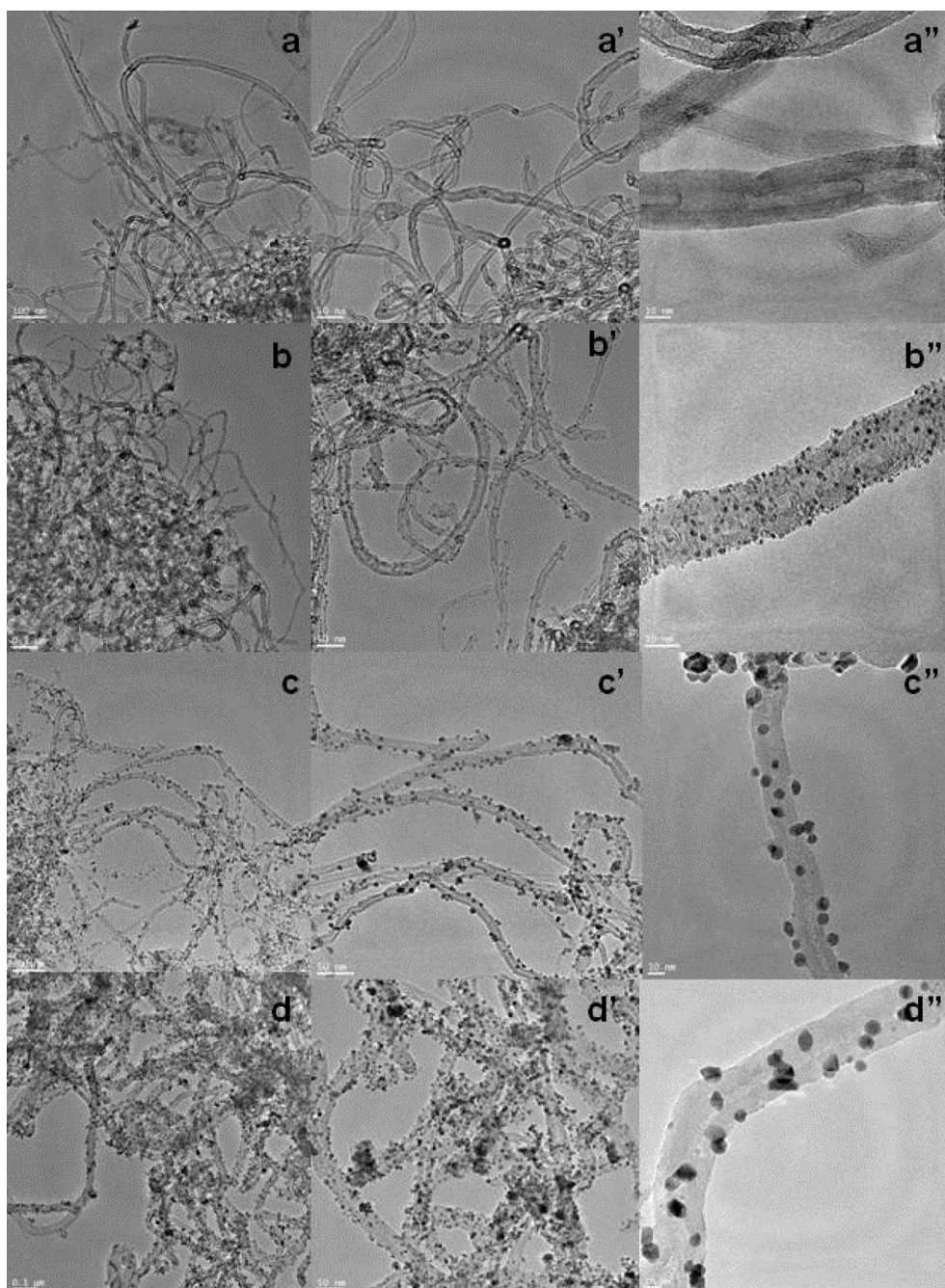


Fig. 1. TEM images of pristine carbon nanotubes (a, a', a'') CNT with Pd nanoparticles Pd@CNT₂₄ h (b, b', b''), Pd@CNT₄₈ h (c, c', c''), Pd@CNT₇₂ h (d, d', d'').

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