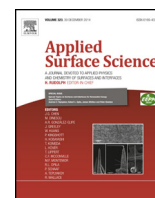




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Effect of pyrolysis temperature on the properties of carbon/nickel nanocomposites prepared by sol–gel method

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ABSTRACT

Carbon–nickel nanocomposites (C/Ni) were prepared by sol–gel method after the incorporation of nickel oxide (NiO) nanoparticles in organic matrix based on pyrogallol–formaldehyde (PF). The nanocomposites heated under inert atmosphere have been characterized by various techniques such as X-ray Diffraction (XRD), Scanning Electron Microscope (SEM), Transmission Electron Microscopy (TEM), and electrical analysis. The XRD spectra exhibited the presence of NiO or metallic Ni phase in amorphous carbon matrix at low pyrolysis temperature, while at 1000 °C the graphite structure line was observed. The TEM images indicate the presence of multiwall carbon nanotubes (MWNT) around Ni nanoparticles for the sample treated at high pyrolysis temperature. The AC conductance shows that our nanocomposites have two behaviors: semiconductor and metal, depending on the pyrolysis temperature. The voltage–current $V(I)$ characteristics of the compound show two different regions: an Ohmic region at low current and a negative differential resistance (NDR) region at higher current. This switching phenomenal behavior has been explained by an electrothermal model.

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1. Introduction

Carbon–nickel (C/Ni) nanocomposite materials received great attention because of their application in a variety of processes, especially the growth of carbon nanowires and nanotubes [1–3], oxidation of alcohols in methanol and ethanol fuel cells [4,5], and hydrogenation of various organic compounds [6,7]. C/Ni nanocomposites could be attractive for hydrothermal gasification of organic compounds [8,9], application in electrochemical devices as sensors [10], and electrodes [11], or hydrogen storage [12,13].

In our work, these nanocomposites were prepared by the sol–gel method followed by a heat treatment at different pyrolysis temperatures. During carbonization, which takes place over the range 500–1200 °C, the physical properties undergo strong modifications [14]. This evolution is experimentally related to a loss of heteroatoms (mainly hydrogen atoms) during the pyrolysis. The process of carbonization appears as a solid-phase nucleation. In fact, the loss of hydrogen promotes the production of bi-dimensional microlayers through a genuine polymerization [15]. The resulting crystal nuclei allow the development of crystallites in the case of graphitizable carbons.

Recently, the negatrons have been subject to several researches. They are widely used for the manufacture of electronic devices [16–18]. In fact, the negatrons have the negative value of a basic differential parameter such as; resistance, inductance, and capacitance. We can have the electrical switching phenomenon in negatrons. This phenomenon has from the beginning, attracted considerable interest [19]. It is one of the numerous interesting effects arising in strong electric field [20]. The phenomenon has been observed in a great number of crystalline, amorphous, and liquid semiconductors [21–23]. Melanin was shown to process a high conductivity state and negative differential resistance NDR [24]. A synthesis of one-dimensional organic molecules has demonstrated NDR using a molecular sized junction at low temperature and room temperature [25,26]. Logic operations have been demonstrated using NDR devices made with organic materials [27,28].

In this paper, we present the different properties of organic–inorganic nanocomposites PF/Ni. Multiwall carbon nanotubes were observed around the nickel particles at high pyrolysis temperature. The dc conductivity exhibited the presence of conduction percolation phenomenon, and the dominance of 3D-GVRH conduction model in the studied materials. The AC conductance shows that the nanocomposites have two behaviors which depend on the pyrolysis temperature: semiconductor and metallic. The $V(I)$ characteristic indicates the dominance of

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the electrothermal model with the appearance of NDR at room temperature in the sample treated at 600 °C.

2. Experimental

2.1. Sample preparation

NiO nanoparticles were first prepared by sol–gel process using the nickel (II) chloride ($\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$) precursor in a mixture of methanol solution. After 15 min, under magnetic stirring at room temperature, the solution was then placed in an autoclave and dried in supercritical condition of ethyl alcohol. The obtained aerogel was annealed at 500 °C in air for 2 h.

The synthesis of our samples has been accomplished in three steps. In the first one, organic wet gels were prepared by mixing formaldehyde (F) with dissolved pyrogallol (P) in water (W) solution, using picric acid as a catalyst, and nickel oxide (NiO) as an incorporated element with 5% of mass ratio. In the second step, the obtained wet gels were dried in humid atmosphere at 50 °C for two weeks. To obtain a structured xerogel, the sample was transferred in an incubator and dried at 150 °C at a heating rate of 10 °C/day. The drying temperature was then maintained for two days. In the present study, the thermal treatment was carried out in a tubular furnace under nitrogen atmosphere between 600 °C (PF/Ni-600) and 1000 °C (PF/Ni-1000). Each sample was heated up to the desired pyrolysis temperature with a heating rate of 5 °C/min. A constant selected pyrolysis temperature was then maintained for 2 h. After that, the sample was cooled naturally. The samples for electrical measurements were prepared by sculpting monoliths on a parallelepipedic shape ($10 \times 5 \times 2$) mm³ and silver paint on two parallel faces. The latter was used to assure Ohmic contacts.

2.2. Characterization techniques

The (XRD) patterns of nanocomposites were obtained by a Bruker D5005 diffractometer, using Co K α radiation ($\lambda = 1.78901 \text{ \AA}$). The synthesized products were characterized using a JEOL JSM-5310 scanning electron microscope (SEM) and a JEOL-100C transmission electron microscope (TEM). Electrical measurements in a temperature range of 80–300 K were carried out under vacuum using a liquid nitrogen cryostat. Voltage–current measurements were performed using a computer-controlled setup comprising an Agilent 34401A multimeter and an Keithley 220 current source. The AC impedance measurements were obtained over a wide frequency range from 40 Hz to 100 MHz using an Agilent 4294A impedance analyzer. An alternating signal with voltage amplitude of 50 mV was employed to measure conductance G in a parallel mode.

3. Results and discussions

Fig. 1 shows the X-ray diffraction for different nanocomposites PF/Ni. From these diffractograms patterns, three peaks appear around 43°, 51°, and 74°, which are identified respectively to lattice planes (202), (122), and (223), corresponding to the structure of nickel oxide for the xerogel PF/Ni-150. After heat treatment under an inert atmosphere for two hours at 650 and 1000 °C, we note the presence of two peaks of metallic nickel around 52° and 61°, which are the reductions of nickel oxide. They are identified, respectively, to plans (200), and (220). A line corresponding to the structure of graphite observed around 30° for the sample treated at 1000 °C is attributed to lattice plane (020).

The average grain size G can be calculated using the Scherrer equation [29]:

$$G = \frac{0.9\lambda}{B \cos \theta_B} \quad (1)$$

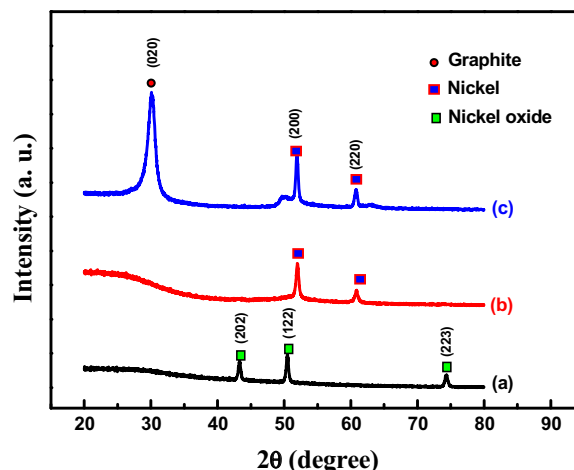


Fig. 1. XRD patterns of different PF/Ni nanocomposites.

where λ is the X-ray wavelength, θ_B is the maximum of the Bragg diffraction peak (in radians) and B is the line width at half maximum. After a correction for the instrumental broadening, the average value of the crystallites was found of about 35 nm for nickel oxide, 25 nm for metallic nickel, and 10 nm for the graphite.

The SEM images for the studied PF/Ni nanocomposites are illustrated in Fig. 2. According to these images, the increase of the pyrolysis temperature contributed to the agglomeration of the microparticles.

TEM images performed on the NiO nanoparticles and the PF/Ni-650 samples are shown in Fig. 3. We notice in Fig. 3a that the average size of NiO nanoparticles is about 35 nm. The TEM image of PF/Ni-650 (Fig. 3b) indicates that material was composed by metallic nickel nanoparticles dispersed in amorphous carbon matrix. It is worthwhile to notice that nickel oxide was reduced to metallic nickel which leads to the decreasing of the nanoparticles size.

For the nanocomposites treated at high pyrolysis temperature (700 and 1000 °C), the TEM images in Fig. 4 show the appearance of carbon nanotubes around the nanoparticles of metallic nickel. Obtaining these carbon nanotubes has been explained by several models, such as carbon filament growth model proposed by Baker [30], itself has adapted the model of silicon filaments proposed by Wagner and Ellis [31]. Note that these models are also the basis for the germination and growth mechanisms at high temperatures. The Ni nanoparticles were like a germination center where on the surface of these nanoparticles, the amorphous carbon is exploded to the carbon nanotubes. The nanotube growth comes from the difference between the surface tension of the nanoparticles of Ni, which is very high due to its nanoscale size, and the low surface energy of graphite [32]; we note that this growth mechanism is similar to that of high-temperature processes based on the minimization of the surface energy of the carbon/nanoparticle system.

Fig. 5 shows the TEM images of carbon nanotubes observed in the nanocomposite PF/Ni-1000. It is the multiwalled carbon nanotubes (MWNTs) with the inter-plane distance of 0.34 nm. They are equivalent approximately to the distance between the graphene layers in a graphite crystal [33].

The physics and mechanics of bimaterial interface adhesion have been developed to a level comparable to that for cohesion in homogeneous, isotropic solids [34–36]. Strong adhesion is generally caused by capillary, electrostatic, van der Waals forces, and other kinds of chemical forces [37,38]. In our study, since the characteristic dimension of carbon nanotubes is down to nanometers, there is a strong adhesion between carbon nanotubes and the Ni nanoparticles, due to van der Waals forces [38].

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