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Efficient Vapor Sensors Using Foils of Dispersed Nitrogen-Doped and Pure Carbon Multiwalled Nanotubes

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Abstract.- We compare the performance of nitrogen-doped (CNx) and pure carbon multi-walled carbon nanotubes (MWNTs) in the fabrication of vapor sensors. The sensors were constructed by dispersing the nanotube materials in methanol so as to form millimeter-long foils (nanotube paper), consisting of compact arrays of crisscrossing nanotubes. The devices were characterized by electrical resistance measurements and SEM studies. For CNx-based sensors, low concentrations of gases such an acetone, ethanol, and chloroform were efficiently detected within 0.1-0.3 seconds via a physisorption mechanism. This process is explained in terms of a weak interaction of the vapor molecules with the pyridinic sites (N bonded to two carbon atoms) present in the doped tubes. First-principles density functional calculations confirmed that the gaseous molecules are able to interact with N-doped carbon nanotubes, via a physisorption mechanism, in which pyridine sites play a crucial role.

One of the most important applications of nanomaterials is related to environmental aspects. In this context, different nano-materials have been used to detect toxic gases and solvents [1]. Previous studies have focused in the detection of CO₂, CO, SO₂, O₂, O₃, H₂, Ar, NH₃ and H₂O [2] because of the high risks to human beings.

In particular, the surface of CNx nanotubes is chemically active without using acids treatments, due to the presence of nitrogen atoms embedded in carbon tubule (note that nitrogen has an extra electron when compared to carbon). Previous studies on sensor-based in CNx multi-walled carbon nanotubes (CNx-MWNTs) were carried out by Villalpando-Páez et al [3]. Different from this study, these authors fabricated films of *as produced aligned carbon nanotubes*. The electrical resistance response of their devices demonstrated a relatively good efficiency for detecting different gases.

CNx-MWNTs were synthesised by chemical vapour deposition (CVD) using the experimental set-up published elsewhere. Ferrocene ($Fe(C_5H_5)_2$) was dissolved in benzylamine ($PhCH_2NH_2$) (97.5 2.5 %). This solution was pyrolyzed during 20 minutes using to two-stage furnace system at 800 $^{\circ}C$ with an Ar flow of 2.5 L/min. The nanotubes were collected from the soot deposited on the surface of the quartz tube in the region located inside the furnace.

Scanning electron microscopy (SEM) image of the synthesized sample of CNx-MWNT is shown in figure 1a, at higher magnifications, the sample exhibited efficient nanotube dispersion. Figure 1b depicts different shape or geometry of the nanotube foils obtained by cutting them with a scalpel. The flexibility of adopting different morphologies represents an additional advantage with respect to sensors fabricated with pellets of aligned carbon nanotubes; difficult to manipulate due to their brittleness. The fabricated sensors are shown in figure 1c. Here, the sensor consists of a membrane (foil) made of dispersed carbon nanotubes. Finally, we plotted the normalized resistance which is defined by $R_N = (R-R_0)/R_0$, in which R represents the electrical resistance obtained in the measurement; R_0 is the mean value of the resistance. In this work, five cycles have been chosen allowing 80 s for the solvent vapor (ethanol, in this case) to enter in the reaction chamber, and 100 s for purging the system with pure Ar. In order to understand the adsorption processes of the nanotubes with the gaseous molecules, we carried out electronic structure calculations (using density functional theory) with armchair (5,5) nanotubes containing pyridine-like nitrogen atoms in the presence of ethanol, acetone and chloroform molecules.



References

Figure 1: Caption of figure 1.

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