# A short review on various applications of carbon nanotubes and graphene nanosheets in different industries

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# Abstract

Carbon based nanomaterials including carbon nanotubes and graphene nanosheets have attracted the attention of researchers due to their outstanding extraordinary properties. In this article, we have presented the researches on the application of carbon nanotubes and graphene sheets in Nanotechnology and Advanced Materials at Materials and Energy Research Center. We have used different modification strategies based on its application for improving the obtained properties in various fields including ammonia gas sensors, supercapacitors, fuel cells, drug delivery systems, and corrosion resistant coatings in order to achieve superior properties in each field.

**Keywords:** Carbon nanostructures; Carbon nanotubes; Graphene; Surface modification.

# 1. Introduction

Carbon based nanomaterials including carbon nanotubes and graphene sheets have great potential for revolutionizing many aspects of our life [1]. The carbon nanotube as 1D and graphene as 2D allotropes of carbon nanostructures have unique properties in terms of electrical and thermal conductivity, mechanical performance and optical features [2]. They have been used in various fields due to their outstanding inherent properties. Therefore, in this article, we will introduce various applications of these nanomaterials by our research team in Department of Nanotechnology and Advanced Materials at Materials and Energy Research Center.

## 2. Carbon nanotubes

Carbon nanotube (CNT) is hexagonal network of carbon atoms rolled up into a seamless, hollow cylinder. CNT is one of the most investigated materials before invention of graphene. CNTs with intrinsic properties including high surface area, cylindrical structure, and high aspect ratio of length to diameter have attracted the attention of researchers. CNTs can be divided into two categories of single walled carbon nanotubes (SWCNTs) and multiwalled carbon nanotubes (MWCNTs). SWCNTs are made of a single graphene sheet wrapped around to form a cylinder and MWCNTs consist of two or more graphene sheets coaxially arranged around a central hollow core with van der Waals forces between adjacent layers [3]. Herein, we have presented different applications of CNTs discovered with our research team.

application in nano-electronic devices and semiconducting SWCNTs can be a promising candidate for gas sensors due to significant electronic changes by gas adsorption. In this regard, F. Goudarzi et al. [4] considered the gas sensing properties of SWCNTs. They deposited SWCNTs doped with lithium on interdigitated electrode by using electrophoresis method and the deposited SWCNTs doped with lithium were heat treated at 350 °C for 5 h. The electrical response of the hybrid sensor was measured upon exposure to 500 ppm NH<sub>3</sub> at different operating temperatures between 25 °C to 280 °C. The electrical response and sensitivity of gas sensor are calculated based on equation 1 and 2, respectively:

Sensor response = 
$$I_g - I_0$$
 (1)

Sensor selectivity = 
$$\left[\left(\lg - \log\right) / \log\right] \times 100$$
 (2)

where  $I_0$  and  $I_g$  are measured current of gas sensors upon exposure to air and ammonia, respectively. The variation of electrical response and sensitivity of gas sensor as a function of immersion time is shown in Figure 1. The results of Figure 1 (a) showed that the gas response increased by increasing the temperature and the sensor recovered rapidly by removing ammonia gas. Further, Figure 1 (b) indicated that the sensitivity of gas sensor increased with temperature due to improve of interaction between gas and sensor; however, the thermal desorption of ammonia molecules at higher temperatures, hindered the gas adsorption process and gas-sensor interaction. Therefore, the maximum gas response with high sensitivity of 34% was obtained at 150 °C [4].





## 2.1. Ammonia gas sensors

Carbon nanotubes have great potential for



Figure 1. (b) sensor response of SWCNTs/Li sensor to 500 ppm  $NH_3$  at 25, 100, 150 and 280°C [4].

# 2.2. Supercapacitors

Supercapacitors can be used in various fields including power source of camera flash, lasers, pulsed light generators, backup power source for computer memory. In this regard, Nikzad et al. [5] developed CNT/polyaniline nanocomposite via in situ polymerization of aniline. They considered the effect of pure CNT and HNO<sub>3</sub>/H<sub>2</sub>SO<sub>4</sub> acid treated CNT on electrochemical properties of the prepared supercapacitors. The cyclic voltametric curves indicated that acid treated CNT is much more electroactive than neat CNT; which can be assigned to developed functional groups on CNT. The results of galvanostatic curves in the applied current of 1 mA during 20 successive cycles are shown in Figure 2. Since the discharge time has straight relation with specific capacitance, the specific capacitance of nanocomposites is much bigger than pure acid treated CNT. The electrochemical results revealed

that the conductivity of the obtained nanocomposites increased and the supercapacitor behavior of polyaniline improves by adding CNT. Meanwhile, composites containing acid treated CNT showed higher conductivity compared to pure CNT [5].

#### 3. Graphene

Graphene is the thinnest material with one-atom-thick which consists from sheets of sp<sup>2</sup>-bonded carbon atoms closely packed in a honeycomb crystal lattice. Graphene is the basic structure of graphite, carbon nanotubes and fullerene. Graphene shows unique properties such as high surface area (2630 m<sup>2</sup>/g), high aspect ratio, low density, ultra-high mechanical strength Young modulus of ~ 1 TPa), tunable electronic band gap, high thermal conductivity (~ 5000 W/mK) and fast charge carrier mobility (~200,000 cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup>) [3, 6].



Figure 2. Galvanostatic curves of (a) CNT, (b) polyaniline/CNT composite, (c) acid treated CNT, and (d) polyaniline/acid treated CNT composite [5].

Our research team has used graphene and its derivatives such as graphene oxide (GO) for different

applications which are discussed below.

#### 3.1. Drug delivery systems

Graphene and its derivatives show good biocompatibility and they can be applied in biomedical applications such as drug delivery, MRI, hyperthermia, bio-sensors and anti-bacterial materials.

Farazi et al. decorated the surface of graphene oxide with magnetic ( $Fe_3O_4$ ) nanoparticles via coprecipitation method and then, they synthesized  $Fe_3O_4/GO$ -gelatin nanocomposite to obtain a stable suspension. They used  $Fe_3O_4$  nanoparticles due to their biocompatibility, low toxicity and acceptable magnetic properties. Also, gelatin was selected because it is a water-soluble biopolymer with advantages such as low-cost, non-toxicity and biodegradability. Further, they studied the potential application of the prepared nanocomposite as a

drug nano-carrier. Doxorubicin hydrochloride (DOX) is anti-breast cancer drug which could be loaded on the surface of different nano-carriers via  $\pi$ - $\pi$  stacking for application in drug delivery systems. Therefore, Farazi et al. [7] loaded the DOX on the synthesized Fe<sub>2</sub>O<sub>4</sub>/GO-gelatin nanocomposite and studied the effect of pH and temperature on releasing the drug. The results (Figure 3) showed that the amount of released DOX increased by increasing temperature from 25 °C to body temperature. The DOX cumulative release in 144 h was 8.84% and 20.98% at 25 °C and 37 °C, respectively. The DOX cumulative release increased from 20.98% to 41.98% by decreasing the pH from 7.4 to 5.4. Accordingly, the DOX drug is better released from the synthesized nanocomposite at body temperature and in acidic pH which mimics tumor environment [7].



Figure 3. The in-vitro release profile of DOX from Fe3O4/GO-DOX [7].

## 3.1. Methanol oxidation in fuel cells

Proton exchange membrane fuel cells, with outstanding advantages such as low operation temperature and high voltage output, are an appropriate candidate for solving the world's energy power. Methanol oxidation is a promising alternative for next generation energy conversion systems. The main approach in direct methanol fuel cells is to oxidize methanol completely to carbon dioxide. However, commercialization of direct methanol fuel cell is challenging because of its low kinetics [8-10]. Various approaches have been considered for developing the reaction kinetics and to reduce the metal loading on the surface.

In this regard, Asghari et al. [11] prepared reduced graphene oxide and nitrogen doped graphene based platinum catalysts and compared the results with commercial Pt/C catalyst. Graphene with unique chemical and physical properties such as high specific surface area, ultra-thin thickness, electrical conductivity, structural flexibility and high stability can be used as support for fuel cell electrocatalysts. The XRD results for reduced graphene oxide (Pt/G) and nitrogen doped graphene (Pt/NG) based platinum catalysts are presented in Figure 4. According

to Figure 4, the characteristic peak of graphene oxide can be seen at  $2\theta = 12^{\circ}$ . In the case of G/ Pt and NG/Pt, different facets of Platinum nanocrystals can be observed in XRD patterns beside the diffraction peaks at 27 to 29 degrees attributed to carbon support. The synthesized catalysts were evaluated for methanol and CO electro-oxidation. Figure 5 shows the cyclic voltammetry results of the synthesized catalysts. The results proved that the nitrogen doped graphene based platinum catalysts have higher electrochemical catalytic activity. Indeed, the Pt/NG catalysts showed higher comprehended peak current and higher ratio of forward peak to backward peak (I,/I,); announcing that Pt/NG sample is more stable toward intermediates of electrooxidation of methanol (mostly CO) and therefore, higher stability can be achieved versus CO-poisoning of Pt nanoparticles. Moreover, although the onsets of three catalysts were almost the same, the Pt/G and Pt/NG showed faster pace to the forwarding peaks due to their higher current densities. The enhanced electrochemical performance of the synthesized catalyst is due to nitrogen functional groups on the surface of graphene sheets which can alter the defects and electronic structure of the support.





Figure 4. XRD patterns of GO, Pt/G and Pt/NG [12].

**Figure 4.** Methanol oxidation reaction in a three electrode halfcell measurements under Ar atmosphere at room temperature, 0.5 sulfuric acid and 2M methanol, normalized by CO-stripping surface area [11].

Moreover, Aghabarari et al. [12] synthesized N-doped graphene based Platinum nanocatalysts at different temperatures using melamine as nitrogen source and compared the results with commercial 40 wt.% platinum on carbon catalyst. As Figure 6 shows, proper distribution of nitrogen on the synthesized samples is observed by elemental mapping. Further, they characterized the electrochemically active surface area of catalysts by electrochemical methods. Figure 7 shows the CO stripping and the subsequent voltammograms in acidic media. The results present that the Pt/NG-400 and Commercial Pt/C catalysts behave approximately similar with each other in onset or peak potential. However, onset of Pt/NG-600 and Pt/NG-800 catalysts happens very faster than that of Pt/C (almost in 0.5 V in more negative potentials). In acidic media, all catalysts develop a main CO oxidation peak with some differences. For example, Pt/C presents only a symmetric peak at 0.61 V, while the other catalysts exhibit an asymmetrical peak with

several humps. These small anodic contributions are linked to CO oxidation on catalytic sites of diverse nature. Catalytic sites of diverse activity justify the absence of only one symmetric main peak in conjunction with a CO diffusion impediment, which can be occasioned by adsorbed sulfate on special sites (e.g. edge) or by some geometric factor (e.g. particle agglomeration) that impedes the free CO diffusion towards the most catalytic sites. Additionally, an electronic charge transfer by the catalyst support may produce higher CO binding energy and consequently slower CO diffusion. Consequently, multiple anodic peaks associated with CO oxidation on different sites are observed during the CO stripping voltammogram [12]. Besides, methanol oxidation cyclic voltammetry results indicate that the synthesized N-doped graphene based catalysts have higher activity for methanol oxidation over commercial catalyst for fuel cell applications.



**Figure 6.** FE-SEM images of (a, b) GO and (c) NG sample synthesized at 800 °C with its nitrogen elemental mapping [12].



**Figure 7.** CO-Stripping curves of (a) commercial Pt/C catalyst, (b) Pt/NG-400, (c) Pt/NG-600 and (d) Pt/NG-800 in acidic media. Recorded in 0.5M H2SO4, Ead= 0.1 V, Scan rate= 20 mVs-1 [12].

# Potential (V vs. Ag/AgCl)

3.2. Oxygen reduction reactions in fuel cells

The oxygen reduction reaction is the primary electrochemical reaction occurring at the cathode of polymer electrolyte fuel cells. Precious metals such as Pt with inherent properties such as high activity and durability in acidic and alkaline electrolytes have been used as catalyst to overcome the disadvantage of slow kinetics of oxygen reduction reactions [13].

In this regard, Aghabarari et al. [14] revealed a significant improvement for oxygen reduction reaction by anchoring Pt nanoparticles (PtNPs) onto a new hybrid chitosan derivativecarbon black support. In the approach used, the chitosan derivative was prepared by a Michael reaction with methyl acrylate followed by an amidation reaction with diethylentriamine to chitosan-(N-(2-(2-aminoethylamino)ethyl) obtain propanamide) (CSD). Then, the chitosan derivative obtained was coated on the carbon black (Vulcan XC 72R) with different mass ratios of CSD/carbon (CSD.C) by two different methods to obtain 5 wt% and 10 wt% CSD.C (method 1), and 13 wt% CSD.C (method 2). Method 1 was based on mixing the carbon black dispersed in isopropanol with the appropriate amount of chitosan derivative in 2 wt% acetic acid aqueous solution over 2 h, after which the solvent was removed in a rotary evaporator. Method 2 was used to maximized the amount of biopolymer incorporated by mixing the chitosan derivative in 2 wt% acetic acid aqueous solution with carbon black (mass ratio CSD/carbon = 5/1), dispersing with ultrasonic treatment for 30 min, stirring the mixture at room temperature for 24 h and then separating the obtained CSD.C by centrifugation. Finally, Pt nanoparticles (30 wt%) were anchored onto the hybrid CSD.C by impregnation of an appropriate aqueous solution containing chloroplatinic acid, followed by addition of a NaBH, solution to reduce the metal at room temperature. According to Figure 8, the halfwave potentials of the Pt.5.CSD, Pt.10.CSD, Pt.CSD. M2 and Pt/C (JM) are 0.86, 0.85, 0.84 and 0.85 V, respectively. The onset potential for the oxygen reduction reactions in the chitosan-based catalysts is approximately 20 mV lower than commercial catalyst and does not shift suggesting that the nature of the active site has not changed. Therefore, the results exhibit that the chitosan-based catalysts posses a better performance than the commercial catalyst. Based on the ring current generated from the oxidation of  $H_2O_2$ , we can see that both the Pt.CSD. M2 and commercial catalyst produced a lower level of  $H_2O_2$  than the Pt.5.CSD and Pt.10.CSD catalysts.



**Figure 8.** Polarization curves on (A) a glassy carbon rotating ringdisk electrode and (B) H2O2 yield of Pt/C (JM) (1), Pt.5.CSD (2), Pt.10.CSD (3) and Pt.CSD.M2 (4).

In order to reduce the costs and realize sustainable development, Aghabarari et al. [13-15] developed bio-nanocomposites of chitosan with graphene and heteropolyacid for oxygen reduction reactions in alkaline media. They proposed a facile and nonprecious metal catalyst approach and synthesized nitrogen doped carbon by using melamine and chitosan as low-cost nitrogen source. Further, they prepared hybrid N-based composites by adding heteropolyacid H<sub>3</sub>PMo<sub>12</sub>O<sub>40</sub> (HPMo) and different transition metals (Fe, Co, Cu). They investigated the electro-catalytic oxygen reduction reaction activities of the composites by using three electrodes set up utilizing cyclic voltammetry and rotating disc electrode in alkaline environment. According to Figure 9, the oxygen reduction reaction activity presents the following order: Fe/HPMo/MeCS > Co/HPMo/MeCS > Cu/HPMo/MeCS. Composites of Fe exhibited the best performance while Cu-composites showed the lower activity. Indeed, nitrogen-doped carbon with transition metals such as Fe or Co improves catalytic activity for oxygen reduction reaction in alkaline electrolyte. Besides, the presence of the heteropolyacid HPMo can improve the activity of chitosan composites.



**Figure 9.** Oxygen reduction reaction of composites recorded in O2 saturated 0.1 M NaOH solution with a rotation speed of 1600 rpm and potential scan rate of 0.01 Vs-1 [13].

#### 3.4. Corrosion resistant nanocomposite coatings

Polymer coatings are widely used in various industries for enhancing the corrosion resistance of metallic structures. However, these organic coatings could not provide long term corrosion protection due to presence of cracks and defects in the coatings beside their hydrolytic degradation upon exposure to corrosive environment. In order to enhance the life time of polymer coatings, carbon based nanomaterials with outstanding inherent properties have attracted a great deal of attention. In this regard, Pourhashem et al. [16] considered the effect of graphene oxide on corrosion resistant properties of solvent based epoxy coatings, revealing that epoxy coatings containing graphene oxide show significantly higher corrosion resistance compared to pure epoxy samples. They showed that by adding graphene oxide directly to polymer with lower viscosity (hardener or resin), the graphene oxide nanosheets can be dispersed

uniformly in coating matrix. Besides, the wt.% of nanofiller has great effect on dispersion quality and therefore, on corrosion protection performance of coatings. In this study, best corrosion protection was obtained by adding 0.1 wt.% graphene oxide to low viscosity hardener, followed mixing by epoxy resin.

Further, Pourhashem et al. [17] used different silane coupling agents such as 3-aminopropyl triethoxysilane (APTES) and 3-glysidyloxypropyl trimethoxysilane (GPTMS) with amine and epoxy endgroups, respectively, for functionalizing the surface of graphene oxide. According to electrochemical impedance spectroscopy results in Figure 10, that both epoxy/APTES-GO and epoxy/GPTMS-GO nanocomposite coatings (containing 0.1 wt.% nanofiller) showed superior corrosion resistance than epoxy/GO and pure epoxy coating which is due to the positive effect of silane agent on enhancing the interfacial interaction between modified GO sheets with polymer matrix. Their results revealed that the adhesion strength of epoxy coatings to metallic substrates and the water contact angle on epoxy coatings increases by incorporating silane modified GO nanosheets. The improved properties are due to functionalization of GO sheets with silane agents which prevents GO agglomeration in coating matrix, leading to increase of diffusion path for corrosive agents into the coating. Also, the presence of Si-O-Si and Si-O-C bonds within polymer matrice is another parameter for improving the corrosion resistance and bonding strength of nanocomposite coatings. Meanwhile, since the silane functionalized GO sheets are directly added to polyamide hardener with low viscosity, epoxy coatings containing APTES modified GO sheets showed higher corrosion resistance than epoxy/GPTMS-GO nanocomposite coatings; this behavior could be attributed to chemical compatibility between the amine end groups of APTES with polyamide hardener.



**Figure 10.** Bode plots derived from electrochemical impedance spectroscopy for pure epoxy, epoxy/GO, epoxy/APTES-GO and epoxy/GPTMS-GO nanocomposite coatings after 14 days of immersion in 3.5 wt.% NaCl solution.

Since epoxy coatings loaded with 0.1 wt.% APTES-GO nanosheets showed higher corrosion resistant than epoxy/GO and epoxy/GPTMS-GO nanocomposite coatings, Pourhashem et al. [18] considered the effect of APTES-GO wt.% (0.05, 0.1, 0.3, and 0.5 wt.%) on corrosion protection performance of epoxy coatings. As presented in Figure 11, epoxy coatings loaded with 0.1 wt.% APTES-GO nanosheets showed the highest corrosion resistance and by increasing the amount of nanofiller in coating matrix, the agglomeration of nanosheets happens; leading to development of pores in the coating and decrease of corrosive agents diffusion path in the coating.



**Figure 11.** Bode plots for epoxy coatings containing different loadings of APTES-GO (0, 0.05, 0.1, 0.3, and 0.5 wt.%) after 7 days of immersion in 3.5 wt.% NaCl solution.

Moreover, Pourhashem et al. [19] investigated the effect of GO nanosheets decorated by  $SiO_2$ nanoparticles on corrosion protection performance of epoxy coatings. In this research,  $SiO_2$  nanoparticles were synthesized in situ by hydrolysis of tetraethyl orthosilicate on GO sheets. The electrochemical results (Figure 12) demonstrate that  $SiO_2$ -GO nanohybrid is promising nanofiller for increasing the corrosion resistance of epoxy coatings. The synthesized nanohybrid can uniformly be dispersed in polymer matrix and increase the barrier performance of epoxy coatings. The presence of active silane groups on the surface of  $SiO_2$ -GO nanosheets provides high compact polymer coatings with limited defects and pores.



**Figure 12.** The bode plots of pure epoxy, epoxy/GO and epoxy/ SiO2-GO coatings after 28 days of immersion in 3.5 wt.% NaCl solution.

#### Conclusion

Carbon nanotubes and graphene nanosheets with significant electrical, chemical, physical, mechanical and thermal properties are candidate for developing the properties of materials. By selecting appropriate treatment methods, these carbon nanomaterials can be used in various fields. Our studies have shown than carbon nanotubes are an excellent option for ammonia gas sensors and supercapacitors. Moreover, the potential application of graphene and its derivatives in drug delivery systems, fuel cells, and anti-corrosion coatings have been demonstrated. This review article would pave the way for other researchers and outlines the great potentials of these carbon nanomaterials.

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