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Facile Preparation and Charaterizations of Silica decorated Graphene nanocomposite

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ABSTRACT

An interesting method for preparing a composite nanomaterial consisting of silica (SiO_2) decorated onto the surface of graphene nanosheets is presented. Graphene/silica nanocomposite was prepared by the hydrolysis of tetraethyl orthosilicate (TEOS) in the presence of Graphene oxide (GO) obtained by a modified Hummer's method. The structure, morphology, pore diameter, pore volume, and surface area of the graphene/ SiO_2 nano composite were obtained via XRD, Raman spectroscopy, SEM, TEM, BET and BJH method. The BET analysis suggested that the silica decorated graphene have the higher surface area than bare silica and graphene. The XRD spectra indicated the coexistence of silica and graphene in the composite particles. The morphological analysis suggested that the presence of SiO_2 nanoparticles with similar sizes attached on the surface of graphene successfully. Due to its high surface area, high porosity, and other fascinating properties, it is an interesting material for in diverse energy applications such as solar cell, lithium-ion battery, electronic devices and supercapacitors.

Keywords: Graphene, TEOS, modified Hummer's method, SiO_2, TEM.

I. INTRODUCTION

In recent years, graphene-based nanomaterial has been drawn a tremendous attention in all the areas of researchers in material science and technology [1]. Graphene nanosheets are one of the advanced carbonaceous materials, which has single or few layers graphite planes [2]. It has high surface area, superior conductivity, high porosity, and other fascinating properties [3]. It has attracted significant interest as one of the most interesting and potential materials in recent years due to the diverse applications in many areas, such as solar cell, lithium-ion battery, electronic devices, and supercapacitors [4]. Furthermore, Graphene sheets show remarkable improvements in chemical and physical properties when incorporated in composite materials. Compared to the other composites, graphene-based composites have been widely studied due to their large surface area, in addition to their rather simply tuneable their remarkable properties[5]. Literature shows that the decoration of the graphene nanosheets with organic or inorganic materials can bring superior properties compared with the individual component materials [6-8]. In recent years, the nanocomposite of 2D graphene combined with zero-dimensional metal and metal oxide nanoparticles such as Ag [9], Fe [10], Pd [11] and Pt [12], ZnO [13] and CuO [14] etc. show significant scientific interest in the energy sector. Because of its intrinsically charming properties, some researchers have been attracted various composite nanostructures of silica and graphene [15-17]. Silica nanoparticles possess many special properties, such as long-term stability, high surface-to-volume ratio, easy chemical modification, size-dependent properties etc. [18-20]

In this paper, we proposed a method for preparing a composite material consisting of silica nanoparticles decorated onto the surface of graphene nanosheets is presented. The surface area, structure, and morphology of as-prepared Graphene nanosheets were studied using a range of techniques such as X-ray diffraction (XRD), Raman spectroscopy, scanning electron microscopy (SEM), transmission electron microscopy (TEM), X-ray

diffraction (XRD), nitrogen adsorption/desorption isotherms using the BET and BJH method.

II. METHODS AND MATERIAL

Graphite flakes, sulfuric acid (H_2SO_4), sodium nitrate ($NaNO_3$), potassium permanganate ($KMnO_4$), Hydrogen peroxide (H_2O_2), ethanol, tetraethyl orthosilicate (TEOS), ammonium hydroxide was used in the process. All the chemicals are received in its purest form.

2.1 Preparation of rGO/silica nanocomposite

Graphene oxide was synthesized using a modified Hummer's method based on the reference [21]. Silica nanoparticles were prepared through a process based on the Stöber method, TEOS in ethanol medium in the presence of ammonium hydroxide [22].

For the preparation of graphene/silica nanocomposite, firstly, TEOS (1M) was added into 20 ml ethanol while stirring. At the same time, the suspension was diluted to 50 ml with deionized water, ethanol and ammonium hydroxide (3M). The TEOS suspension was then added into the GO suspension in drops. After stirring continuously for 4 h, the mixture was left to age for 24 h at room temperature. The prepared sample was centrifuged with deionized water and ethanol for several times kept in an oven at 80°c for 2 hours. Finally, collect graphene/silica nanocomposite after the calcination in a hot plate.

III. RESULTS AND DISCUSSION

3.1 BET analysis

Total pore volume, mean pore diameter, and most pore size distribution of prepared samples are summarized in Table 1. The specific surface area was calculated to be 289.76 m^2 /g, 431.39 m^2 /g, and 465.51 m^2 /g for graphene, silica and graphene/silica samples, respectively, indicating that the incorporation of silica can lead to a higher specific area in comparison with pure samples.

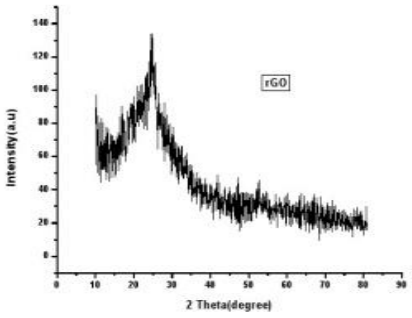
Table 1. Summarizes the BET surface area and BJH pore diameter of nanocomposite

Sample	BET surface area (m^2/g)	Pore volume (cc/g)	Pore diameter (nm)
Graphene	307.13	0.261	1.564
Silica	431.39	2.267	21.380
Graphene /silica	465.51	1.155	1.563

3.2 XRD analysis

The XRD patterns obtained from the graphene, silica nanoparticles and graphene/silica nanocomposite are shown in fig (1). From fig (1a), the XRD pattern has been obtained for synthesized graphene nanosheets. The disappearance of the peak at 10° and appearance of the peak at 24° shows that the product is completely reduced after the thermal treatment and exfoliation. [23]. The strong broad hump, between 21 and 25° (Fig 1b) indicates that the synthesized silica is amorphous, along with some crystalline structures also seen [24].

The XRD pattern has been obtained for graphene/silica nanocomposite shown in fig (1c). The diffraction peaks at 2Θ=24.4° and 53.7° can be attributed to the silica structures. The characteristic diffraction peak of graphite at 26.4° didn't appear, revealing that silica nanoparticles with the size of 42 nm deposited on the graphene surface could efficiently suppress the stacking of reduced graphene layers, thus, in this case, no graphite-like layered structure formed again.



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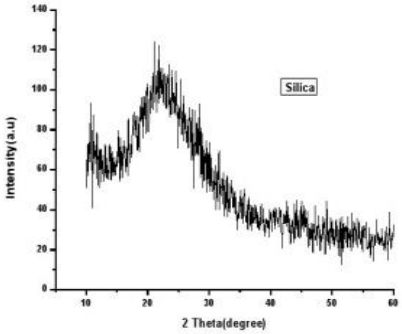

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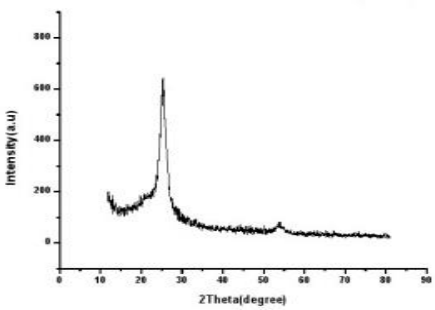

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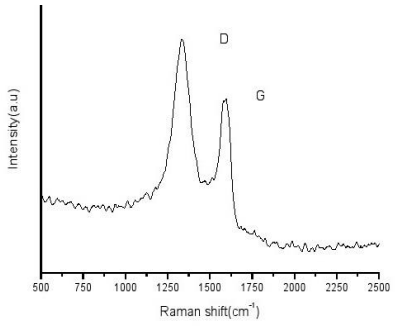
Figure 1. XRD images of (a) graphene (b) silica and (c) graphene/silica nanocomposite

3.3 Raman analysis

Fig (2a) shows the Raman spectra of graphene reduced by thermal method. The presence of D and G bands confirms the formation of graphene with lesser defects and crystalline structure. The higher intensity of D band suggested the presence of more isolated graphene domain.

The specific frequencies of the characteristic peaks at 410 cm^{-1}, 490 cm^{-1},800 cm^{-1},980 cm^{-1} confirm the formation of silica nanoparticles [fig (2b)]. The principle features located at $\nu = 490\ cm^{-1}$ is the breathing mode of 4-membered rings, usually called the D1 line.the characteristic peaks at $\nu = 800\ cm^{-1}$ is the SiO_2 network optical mode and $\nu = 980\ cm^{-1}$ is the vibration of the (OH)-group with respect to Si .these results are consistent with previous works[25].

The Raman spectra of graphene/ SiO_2 nanocomposite were presented in Fig. (3c). The existence of the D and G bands in the Raman results is evidence of the presence of carbon in the study nanomaterials. The D and G bands of the graphene/SiO_2 composite appeared at 1344 cm^{-1} and 1566 cm^{-1}, respectively. The decrease in the I_D/I_G ratio indicates an increase in the number of graphene layers or a partial reduction in the GO into graphene, which is consistent with the reports.



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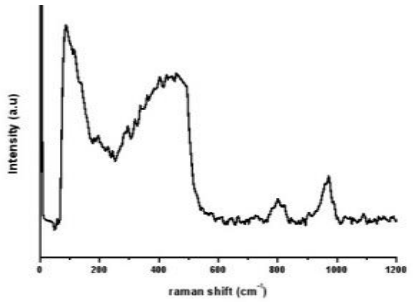

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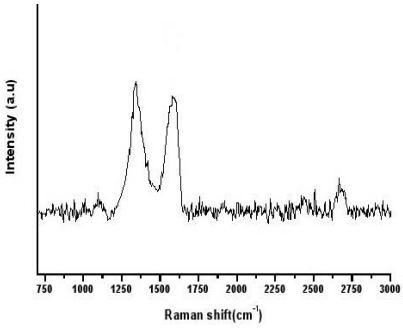
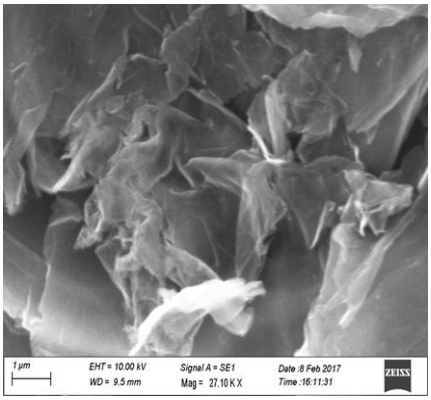


(c)

Figure 2. Raman spectra of samples

3.3 SEM analysis

Fig (3a-c) shows the typical SEM image of as-prepared graphene nanosheets, silica nanoparticles, and graphene /silica nanocomposite respectively. The SEM photograph of graphene (Fig.3a) shows the presence of randomly aggregated and thin crumpled sheets which are closely associated with each other in reduced graphene oxides [26]. Furthermore, graphene was almost transparent and had one or few layer 2D sheet morphology. It should be pointed out that spherical silica particles with distribution of closely spaced have been seen in fig 2b [27]. From Fig (2c), we can see that the samples of prepared graphene /silica the graphene sheets and silica nanoparticles are distinguishable clearly. Also seen that SiO_2 particles with similar sizes were grafted on the surface of graphene nanosheets successfully.



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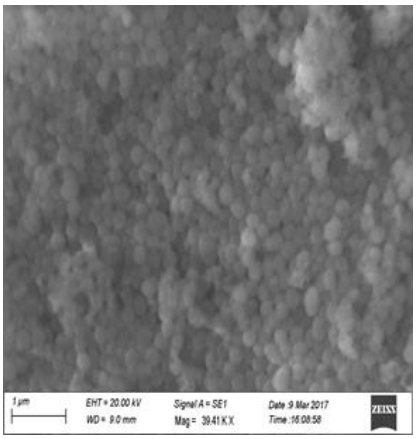

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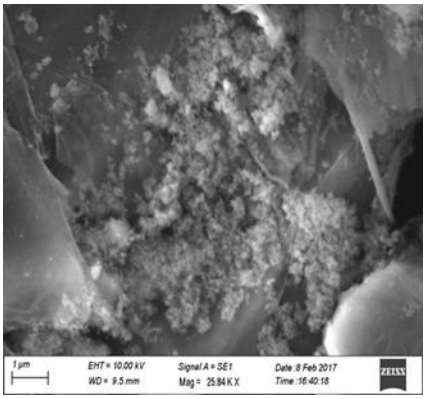

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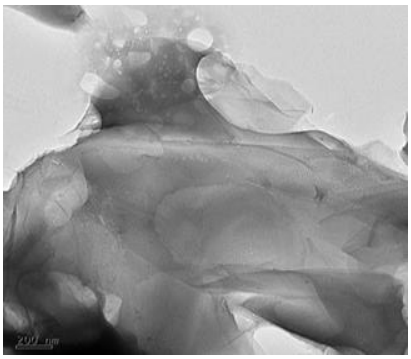
Figure 3. SEM images of (a) graphene, (b) silica nanoparticles and (c) graphene /silica nanocomposite

3.4 TEM analysis

The TEM analysis again confirms the decoration of silica on graphene surface in grapheme/silica nanocomposite. Fig (4b) reveals that the graphene nanomaterial still presents a flexible sheet-like appearance similar to that of SEM image. Silica nanoparticles with the size of about 32-48 nm were well dispersed and attached to the transparent grapheme sheets.



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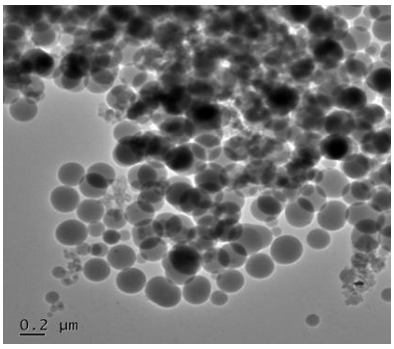

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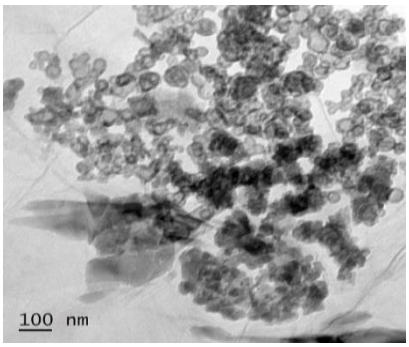

(c)

Figure 4. TEM images of (a) graphene, (b) silica nanoparticles and (c) graphene /silica nanocomposite

IV. CONCLUSION

Spherical silica particles with the distribution of closely spaced have been synthesised by the hydrolysis reaction of TEOS in ethanol containing water and ammonia. The BET analysis suggested that the silica decorated graphene have the higher surface area (465.51m2 /g) than bare silica and graphene. The XRD spectra indicated the coexistence of silica and graphene in the composite particles. All the images from TEM and SEM indicated that the silica nanoparticles successfully bonded onto the graphene nanosheets.

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