An easy and innovative one-step in situ synthesis strategy of silica nanoparticles decorated by graphene oxide particles through covalent linkages

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Abstract

This research work presents the development of an easy synthesis method for functionalized and covered graphene oxide (GO) nanosheets with silica nanoparticles through one-step in situ sol-gel approach by using two different silane molecules: a tetraethylorthosilane (TEOS) grafted with N(\textbeta-aminooethyl)-\textgamma-aminopropyltrimethoxysilane (Z6020) and a TEOS with \textgamma-aminopropyltriethoxysilane (A1100). Both synthetized GO/SiO\textsubscript{2}-Z6020 and GO/SiO\textsubscript{2}-A1100 nanoparticles were characterized and compared in terms of exfoliation level, GO chemical partial reduction and self-condensation efficiency of silica nanoparticles over carbon planes. This way, microstructural and morphological analysis were performed using UV-visible, Fourier Transform InfraRed and Raman spectroscopies, Wide Angle x-rays Scattering, scanning electron microscopy, confocal optical microscopy and thermogravimetric analysis. Laser granulometry and zeta potential measurements on colloidal solutions in water allowed the determination of the particles size distribution and dispersion quality of these nanohybrid materials.

1. Introduction

The discovery of carbon nanomaterials, especially graphite and graphene and their derivatives, has triggered particular attention in the recent years. Their unique and various structures, which consist in stacking of two-dimensional honeycomb lattice of carbon atoms (graphene nanosheets), give them peculiar properties such as: high specific surface area, in plane flexibility, low density, extraordinary high mechanical, thermal and electrical behaviors \cite{1,2}. Many researches have reported the use of graphene as a single-layer structure in many industrial fields, especially in the polymer nanocomposites science, chemical reaction catalysis, water purification, electronics, electrochemistry, electromagnetics, biomedical and energy storage industries \cite{3–7}. However, their cost-effective synthesis processes, their potential involved structural defects and their problems of exfoliation and distribution at the time of the elaboration of polymer based composites limit their applications \cite{8–10}. With the aim to overcome these difficulties, the functionalization of graphene oxide nanosheets by inorganic nanoparticles is a promising way that enables to avoid the aggregation phenomenon and to reinforce the surface of graphene \cite{11–14}.

Besides, the exceptional potential of the surface modification of graphene oxide (GO) is always one of its main advantages due to the improvement of their physical and chemical properties compared to conventionally carbon structure \cite{15,16}. It is clear that graphene oxide has numerous suitable reactive sites, including hydroxyl and epoxide groups on basal planes and carboxyl groups on the edges, which make it stable and dispersable in polar solvents and potentially can be covered covalently with a great number of chemical reagents \cite{16,17}. 
The molecular structure of organosilanes and their potential chemical reactivity with various substrates have already been studied and have shown great potential [18, 19]. When the silane is hydrolyzed, the alkoxy groups react and produce hydroxyl groups allowing siloxane linkages and condensation reactions of silane [20]. However, to reach an optimal achievement of this kind of surface functionalization, an homogeneous dispersion of particles in water is needed [21–23]. Chemical surface modification of graphene oxide sheets using silane based coupling agents is then an effective way to enhance the interfacial interaction of such GO particles and significantly improve their exfoliation ability [24, 25]. Given the good characteristics and properties of SiO$_2$ nanoparticles: high surface area, high thermal stability, great chemical, mechanical, optical and electrical properties, silica nanoparticles are a suitable candidate to develop new advanced synthesis protocols to produce nanohybrids materials combining both characteristics of GO and silicates [26, 27].

In order to investigate the effect of organosilanes on the modification of the surface of graphene oxide nanosheets and as a strategy to control the self-condensation reactions of SiO$_2$ nanoparticles, the present work aims to establish a one-step in situ synthesis by a sol-gel technique that allows the self-condensation of silica nanoparticles on the surface of GO controlling the conversion and the chemical deposition. Two main approaches have been adopted. The first one consists in the use of the tetraethyl orthosilicate (TEOS) as a source of silica nanoparticles and the use of N-(b-aminomethyl)-caminopropyltrimethoxysilane (Z-6020) to control the self-condensation step and bring chemical coupling ability. The second approach consists in the use of TEOS with γ-aminopropyltriethoxysilane (A1100). Furthermore, to better evaluate these two routes of synthesis, the functionalized GO/SiO$_2$-Z6020 (called GSZ), GO/SiO$_2$-A1100 (called GSA) and graphene oxide (GO) were characterized and compared.

2. Chemical elaboration process

2.1. Materials

Natural graphite powder with particle size < 20 μm was purchased from Sigma-Aldrich. Chemical reagents used for the synthesis and functionalization of graphene oxide (GO): sulfuric acid (H$_2$SO$_4$, 98%), sodium nitrate (NaNO$_3$, 99%), hydrogen peroxide (H$_2$O$_2$, 30%), chlorhydric acid (HCl, 37%), ethanol (C$_2$H$_5$OH, 98%) and sodium hydroxide (NaOH). Potassium permanganate (KMnO$_4$, 99.5%) was purchased from Fisher and used as received. Tetraethyl orthosilicate (TEOS, > 98%) was also purchased from Sigma-Aldrich. γ-aminopropyltriethoxysilane (A-1100, > 99%) and N-(b-aminomethyl)-γ-aminopropyltrimethoxysilane (Z-6020, > 98%) were generously donated by Witco Corp (USA).

2.2. Synthesis of graphene oxide nanosheets

GO nanosheets were prepared from natural graphite powder according to a modified Hummers method [28]. The details of the synthesis were developed in our previous publication [26] and could be resumed as follow. First, 2.0 g of graphite and 1.5 g of NaNO$_3$ were added into 50 ml of concentrated H$_2$SO$_4$ and stirred for 3 h. Subsequently, 6 g of KMnO$_4$ were gradually added to the above solution while keeping the temperature close to 20 °C. The mixture was then stirred at the ambient temperature for 24 h. After that, 100 ml of water was slowly added. Finally, 250 ml of water and 10 ml of H$_2$O$_2$ (30% solution) were added to the above mixture. After that, the product was washed, first with an aqueous solution of hydrochloric acid (HCl, 2 M) and then with distilled water until the pH of the suspension became neutral. The graphite oxide was dispersed in deionized water and exfoliated to yield GO nanosheets. Finally, the product was dried in a vacuum oven at 100 °C for 24 h.

2.3. Synthesis of nanohybrid materials

The graphene oxide nanosheets functionalized and filled with silica nanoparticles (SiO$_2$) were fabricated by the developed in situ sol–gel process, which allows the self-condensation, the self-deposition of silica nanoparticles and the grafting of organosilane on the surface of GO at the same time. Figure 1 represents the chemical reaction that occurred during the synthesis of the nanohybrid materials using both A-1100 and Z-6020 reagents.

For this purpose, 70 ml of a water–alcohol–TEOS solution (50:15:5, v/v/v) was mixed at room temperature for hydrolysis for 3 h. Then, 30 mg of GO sheets were dispersed in water (1 mg ml$^{-1}$) through sonication for 30 min and then added to the above mixture. Thereafter 1 ml of γ-aminopropyltriethoxysilane (A-1100) was added to the solution. Then, the mixture was kept under agitation at room temperature for 24 h and at pH = 4 adjusted by using the acetic acid concentration. Eventually, the silica nanoparticles deposited on the surface of GO nanosheets after 24 h of the sol-gel reaction via increasing the pH up to 9 using a solution (5 wt%) of sodium hydroxide. Then, the mixture was kept for 1 h at 60 °C for condensation reactions.

In the second method, 1 ml of N-(b-aminomethyl)-c-aminopropyltrimethoxysilane (Z-6020) was slowly added to the mixture of water–alcohol–TEOS solution (80:15:5, v/v/v) and GO (30 mg), drop-by-drop, and allowed the mixture to react for hydrolysis reaction at room temperature during 2 h. In order to initiate the self-
condensation reaction, the hydrolyzed Z-6020 (HZ-6020) molecules increase the pH of the mixture up to 10. At the end of 24 h, the gel containing GO and SiO₂ was stirred for 1 h at 60 °C. After that, the mixture was centrifuged and washed at least 5 times with deionized water, then with ethanol. Finally, the prepared particles were placed in a vacuum oven at 100 °C for 24 h before further use. The schematic of the synthesis procedure of particles is displayed in figure 2.

3. Products characterization methods

- Fourier transformed infrared spectroscopy (FT-IR) was performed using a JASCO-IR spectrometer. The spectra were collected in the wavenumber range from 4000 to 400 cm⁻¹ at a resolution of 4 cm⁻¹.

- UV-vis absorption spectra of the synthesized products have been recorded in deionized water (0.5 mg ml⁻¹) by Shimadzu UV-1800 UV-vis spectrophotometer. The optical absorption was measured within the wavelength range from 200 to 800 nm.

- Wide Angles x-rays Scattering diffractograms in reflection mode of the synthesized products were performed at room temperature using a Phillips XPERT Pro diffractometer with a CuKα radiation source and x-rays wavelength (λ = 0.154 nm) using a 40 mA current and a 45 kV voltage generator. The samples were analyzed at 2θ scanning range from 5° to 80° with a step interval of 0.016°.

- The Raman spectra were recorded at room temperature from 1000 to 2000 cm⁻¹ on a LabRAM-HR Evolution (Horiba) spectrometer using an excitation wavelength of 532 nm and exposure time of 10 s.

- Thermal analysis (TGA-DTG) were carried out using a TA–SDT Q600 thermal analyzer at a heating rate of 10 °C min⁻¹ from 25 to 700 °C under a nitrogen atmosphere.
Scanning electron microscopy (SEM) was employed to observe the surface morphology of synthesized products. The SEM micrographs were recorded on a Quanta 200 commercialized by FEI. The operating parameters were an accelerating voltage of 15 kV and a high vacuum of...

Confocal optical images were obtained using a Leica DCM8 microscope. Samples were prepared by depositing a clear solution of GO, GSA and GSZ (in water) on a clean microscope slide substrate followed by vacuum drying for 24 h.

The particle size distribution was estimated by using laser diffraction particle size analyzer type LA-690 in an aqueous solution.

The magnitude of surface charge and dispersion stability of colloidal suspensions were analyzed by using a nanoparticle analyzer type Nano Partica SZ-100 in an aqueous medium solution at room temperature.

4. Results and discussion

4.1. UV-Visible spectra analysis

UV–vis absorption spectra of GO nanosheets, GO/SiO$_2$–A1100 and GO/SiO$_2$–Z6020 particles were recorded to identify the characteristic features of GO and to determine the grafting density of silica nanoparticles on the carbon surfaces [29]. The UV–vis absorption behaviors of the different aqueous suspensions are shown in figure 3. The absorption spectra of GO nanosheets consists in a sharp band at 230 nm and a shoulder at 310 nm which are related to the $\pi-\pi^*$ bond vibration mode of the graphitic structure (C=C) and the n-\(^*$) bond vibration mode of the functional groups such as –COOH, –OH, C–O–C, respectively. This last indicates both the presence of hydroxyl, carboxylic and epoxide groups either on the edges or on the basal plane of GO and their degree of oxidation [30, 31]. In comparison, it can be seen in the UV-visible spectrum of particles that the GO/SiO$_2$–Z6020 can be identified through the observation of two characteristic absorption bands at 229 nm and 275 nm, which are attributed to the absorption of aromatic rings and to the deposition of silica nanoparticles on the surface of GO, respectively.

Furthermore, the shoulder observed at ~310 nm disappeared, which is mean that the oxygen functionalities groups existing on the surface and on the edges of GO nanosheets have been reacted to form covalent linkages with silica nanoparticles and/or covalent functionalization of silane Z-62020 molecules.

In the case of GO/SiO$_2$–A1100 particles, it can be enlightened an intensive absorption peak at 229 nm and a weaker another at 275 nm, which show the low recovery degree of SiO$_2$ nanoparticles on the surface of GO compared to the GO/SiO$_2$–Z6020 [29, 32].
4.2. FT-IR spectrophotometry analysis

The chemical structures, the formation of functional groups and the presence of silica nanoparticles on the basal plane of GO nanosheets were investigated by FT-IR analysis.

According to figure 4, there is no absorption band observed in the spectrum of pristine graphite, indicating the chemically inert graphitic structure of bulk graphite [33]. After oxidation of graphite (figure 4(b)), new vibrational bands appear at (2983, 2851 cm\(^{-1}\)), (1735, 1361 cm\(^{-1}\)), (1216, 1034 cm\(^{-1}\)) and a new band in the range 3700–3000 cm\(^{-1}\), which confirms the presence of –CH\(_2\), C=O, C–O–C,–OH and –COOH functional groups, respectively. These changes in the chemical structure of graphite indicate that oxidation well occurred. Another characteristic band is observed for GO at 1625 cm\(^{-1}\) attributed to the absorption band corresponding to the C=\(\equiv\)C skeletal vibration bond in the unoxidized graphitic domain [34–38]. Then, the organic–inorganic hybrid structures based on GO and SiO\(_2\) are created and an intensive band is obviously detected at 1090 cm\(^{-1}\) in FT-IR spectra of GSZ which corresponds to Si–O–Si bending vibration and indicates the significant self-condensation reaction of TEOS precursors forming silica nanoparticles on the GO surface [39]. Furthermore, additional vibrational bands appear at 676, 785, 3735, 1538 and 1455 cm\(^{-1}\), which can be attributed to the stretching vibrational mode of the new Si–O–C, Si–OH, NH\(_2\), N–H and C–N groups, respectively. These new signals reveal the silane (Z6020) that is now fixed onto the GO surface and can play the role of a coupling agent [40, 41]. FT-IR spectra of GSA confirms the surface structure modification and the presence of functional
groups. The presence of bands at 3700–3000 cm⁻¹ and 1361 cm⁻¹ demonstrate the partial reduction of GO and the low tendency of silane to react with GO surface. These results put in evidence that the Z6020 silane highly tends to react with the functional groups of GO nanosheets through self-condensation reactions whereas the A1100 silane considerably intensifies the deposition of TEOS as nano-silica particles on the GO surface [1, 42].

4.3. Wide Angles x-rays scattering analysis

1D WAXS acquisitions in reflection mode were performed to study the crystalline phase structure, the stacking order and the morphology of flake graphite, GO nanosheets, GSA and GSZ particles. Using Bragg’s and Debye–Scherrer equations, the layer spacing and the average crystallite size can be calculated, respectively:

\[ d_{hkl} = \frac{\lambda}{2\sin(\theta)} \]  
\[ D = \frac{K\lambda}{\beta \cos(\theta)} \]  

Where \( \lambda \) is the x-rays incident wavelength, \( \beta \) the full width at half maximum intensity (FWHM) of the diffraction peak, \( \theta \) the Bragg’s diffraction angle, \( d_{hkl} \) the interreticular distance between each \( (hkl) \) crystalline planes and \( D \) the crystallite particle size.

The inset figure in figure 5 presents the WAXS diffractograms of pristine graphite powder since the characteristic diffraction peak at 2\( \theta \) = 26.5° of the (002) crystalline planes is clearly observable. The interlayer spacing of 0.34 nm is then found. The crystallite size is about 15 nm. Similar observations were reported in the literature [43]. In the case of the exfoliated GO, an important displacement of the previous discussed 2\( \theta \) angle to lower values can be observed (2\( \theta \) = 10.5°). This observation demonstrates the good exfoliation of the GO nanofilms and the formation of oxygen functional groups on the surface of GO with an interlayer spacing of about 0.84 nm and the crystallite size is 8.6 nm [44]. The WAXS diffractogram (figure 5) of GSZ particles shows a diffraction peak at 13.6° and a broad halo of diffusion centered at 22.4°, which are mainly due to the GO and amorphous structure of the nano-silica, respectively. It reveals the in situ decoration of silica nanoparticles on the surface of the GO nanosheets [1, 45]. Concerning the GSA organosilane modification, the WAXS results displayed intense diffraction peaks around 22.4° and 26.5° which are assigned to the crystal plane of the nano-silica (d_{100}) and the plane (d_{002}) of the well-ordered graphene sheets, respectively. This result indicates that the use of A100 silane and manual options for pH adjustment are disrupted the self-reaction and changed the amorphous crystalline structure of SiO₂ which contributes to heightening the crystalline size of the synthetized nanohybrid particles [46, 47].

The crystalline size of the synthesized products was summarized in table 1.

4.4. Raman spectroscopy

Raman spectroscopy was performed to investigate the amount of defects inside the graphite based structures and to further confirm or not the silica nanoparticles coating on the GO surface. Raman spectra of GO, GSA and GSZ are depicted in figure 6.
The Raman spectrum of GO sheets illustrates the presence of the two characteristic bands of graphite based materials, thereby a typical D and G bands which are assigned to the sp² ordered crystalline graphite-like structures at 1354 cm⁻¹ and a disordered sp³ carbon structure at 1583 cm⁻¹, respectively [48, 49]. The shifting of the G bands to higher wavenumbers (1605 cm⁻¹) was very remarkable in the case of GSA and GSZ particles indicate the better exfoliation of GO sheets and formation of silica nanoparticles on the GO surface [50, 51].

In addition, it’s clear that the difference in intensity and broad of band between GSA and GSZ particles. Therefore, the increase of the D and G band in GSZ indicates that the silica nanoparticles were covalently attached on the surfaces of GO, indirectly proving the self-condensation mechanism of silica nanoparticles on GO without significant defects of carbon lattice [52]. The D and G bands of the GSZ Raman spectrum refer to the good exfoliation after the successful in situ sol-gel deposition of the nano-silica particles on the layered structure of the GO.

### 4.5. TGA-DTG analysis

Thermal stability of graphite, GO nanosheets, GSA and GSZ hybrid nanomaterials were investigated through thermogravimetric analysis (TGA—figure 7) and differential thermal analysis (DTG—figure 8).

The TGA and DTG graphs of pristine graphite show a single mass loss at 620 °C demonstrating its high thermal stability as reported in the literature [53]. In the case of GO nanosheets, three well-defined weight losses can be identified at three different temperature regions: the first stage occurring between 0 and 130 °C is ascribed to the evaporation of the trapped residual water on GO sheets surface; the second stage observed in the range of 130 °C–320 °C corresponds to the elimination of the oxygen based functional groups existing on the surface and on the edges of GO nanosheets (i.e. hydroxyl, carboxylic and epoxide groups) vanished in the form of gases such as H₂O and CO₂ as byproducts; subsequently, the third stage at 320 °C–650 °C is assigned to the degradation of the carbon-carbon skeleton into volatile gases such as CO. The residual weight (16.02%) observed at 700 °C can clearly reveal that GO nanosheets exhibit three weight loss steps and is not stable anymore at a higher range of temperatures. Regarding the GSA and GSZ nanohybrid particles, curves exhibit only one weight loss at 700 °C of 9.43% and 7.51%, respectively, that illustrate the high heat resistant capacity and the lower hydrophilic nature of GSZ’s surface compared with the GSA’s one. Finally, the presence of Z6020 silane content that leads to the self-condensation process increases the possibility of nano-silica to form a covalent linkage of nano-silica particles on the GO surface [32].

| Sample       | Graphite | GO   | GSA | GSZ  |
|--------------|----------|------|-----|------|
| Crystallite particles size D (nm) | 15       | 8.6  | 9.1 | 8.7  |
| Interlayer distance d_(002) (nm) | 0.34     | 0.84 | 0.59| 0.65 |
4.6. Scanning electron microscopy (SEM) micrographies

The morphology of the pristine graphite, the GO sheets, and the synthesized particles (SGA and SGZ) was analyzed via scanning electron microscopy (SEM).

It can be seen from figure 9(a) that the flakes of pristine graphite show a well-ordered structure of graphene layers [51]. Figure 9(b) displays the surface morphology of GO nanosheets where the formation of thin ripples and stripes can be distinguished. Moreover, the multi-layered structure could be easily figured out at the edges [54]. Figures 9(c)–(d) illustrate the SEM micrographies of the synthesized hybrid particles. The use of the A1100 silane seems to significantly promote the dense coverage of GO by silica particles, thereby forming large agglomerated particles and makes the sheets relatively thick. However, some vacancies can be observed suggesting the non-uniform coating of silica nanoparticles and the formation of undesired agglomerates. In the opposite, the synthesized hybrid nanoparticles using the Z6020 silane show a more pronounced reduction in size of the silica particles and of the GO sheets thickness, simultaneously. These observations corroborate the uniform coverage of the nanosheets obtained via a self-condensation of in situ silica precursors on the surface of the GO nanosheets that successfully validates the engaged hybridization process [40, 55].

Figure 7. TGA weight loss curves of (a) graphite, (b) GO, (c) GSA and (d) GSZ.

Figure 8. DTG curves of (a) graphite, (b) GO, (c) GSA and (d) GSZ.
4.7. Confocal optical microscopy observations

Micrographies by confocal optical microscopy were obtained in order to measure the average thickness of GO and GO–SiO2 particles. Thus, it was expected to enlighten the effect of the SiO2 nanoparticles grafting on the GO sheets surface.

From the figure 10 results, it can be seen that GO, GSZ and GSA particles have an average thickness of about 2.50 nm, 8.00 nm and 3.50 nm, respectively. The GSA particles present an higher thickness than the two others that can be due to the agglomeration of silica into bulky dense particles on the GO surface after having been hydrolyzed for 24 h. It can also be seen that after surface modification of GO nanosheets by the Z6020 silane, the thin thickness and the surface roughness increased comparing with the A1100 silane. However, in GSZ particles the thickness is only 3.50 nm, as shown in figure 10(c), which is due to the presence of the covalent C–O bonds, implying the existence of functionalized silane Z6020 chains on the edge of GO as well as SiO2 nanoparticles on the GO surface. From the results obtained, the SiO2 nanoparticles have covered the surface of GO sheets and the height is about 1.00 nm [56, 57].

4.8. Particles size distribution

The particle size distribution of GO, GSA and GSZ was obtained in an aqueous solution. Results are summarized in figure 11.

It can be demonstrated from figure 11(a) that GO particles exhibit an average size of about 7.9 μm. This decrease in size and size distribution range of GO might be ascribed to the graphitic structure and to the oxygen based functional groups that exist on the GO sheets surface. It has to be noticed that the water molecules interposed within the interlayer of the GO nanosheets increase stability in various polar solvents.

From figure 11(b), GSA hybrid particles show a higher diameter size distribution with an average value of about 26.8 μm. This higher range might be ascribed to the dense deposition of silica nanoparticles on the surface of GO sheets and the more important concentration of A1100 silane groups on the surface of nano-silica leads to an increment in the size distribution of the individual particles and as a consequence reach a bulky hybrid structure.
Meanwhile, it can be demonstrated from figure 11(c) that GSZ exhibit an average size distribution value of 19.9 μm. In addition, the hybridization of GO nanosheets with TEOS and Z6020 silane allow to obtain a uniform deposition of nanoparticles on the surface and facilitate the partial reduction of GO particles resulting in the disappearance of the H-bonds interaction [58–60].

4.9. Zeta-potential measurements
The quality of exfoliation and dispersion of nanoparticles can be characterized by various indicators such as zeta potential, solution pH, specific surface area, electrical and thermal conductivity and particle size distribution [61]. The zeta potential of the synthesized products was performed in an aqueous solution to optimize the dispersion stability of the colloidal suspensions. As seen in figure 12, the zeta potential of GO sheets is about −32.7 mV, which confirms that the surface charge is negative [62]. The negative zeta potential in water indicates
that GO sheets form stable aqueous colloids due to high hydrophilicity and electrostatic repulsion with the ionized functional groups, such as carboxylic (COOH) acid groups.

A negative zeta potential value is achieved for GSZ (−49.4 mV) as shown in figure 12(c). The higher negative value reveals the improved stability of the GSZ nanohybrid particles in aqueous solution. Moreover, a relatively lower negative value of the synthesized GSA (−38.8 mV) imply a moderate stability in aqueous medium because of the presence of positive functional groups, such as protonated amine (−NH₃⁺) groups as well as the

Figure 11. Size distribution analysis of (a) GO, (b) GSA and (c) GSZ.
disappearance of the oxygen based functional groups from the GO sheets surface. In addition, the result reflects the poor dispersion stability behavior of GSA in aqueous solution. This investigation demonstrates that the zeta potential of synthesized GSZ particles is more negative than those of GSA ones that illustrates the influence of the modification process [63–65].

The zeta potentials of GSA and GSZ particles were then more negative than those of GO sheets. This is probably due to the formation of several electronegative groups in terms of siloxane bonds (Si–O–Si) and silanol (Si–OH) on the surface of GO sheets covered with the silica nanoparticles.

5. Conclusions

In conclusion, we have successfully synthesized GO nanosheets covered by silica nanoparticles through a one-step in situ sol-gel process. This way, we propose an easy and innovative method for taking the potential advantages of silane molecules by covalent bonding with GO sheets surface via self-condensation. Two different silane molecules (A1100 and Z6020) were studied and allowed to determine their impact on the synthesis parameters and on the final functionalization of particles. The various characterization and measurement techniques employed allowed to investigate the efficiency of this novel synthesis process and to validate the expected characteristics of the final synthesized particles. It is confirmed that the disordered structure, the SiO₂ nanoparticles deposition on GO nanosheets and the presence of the covalent grafting with silane molecules have well been obtained through this one-step chemical procedure. The results on GO sheets showed that an excellent oxidation degree of the pristine graphite powder was obtained via a modified Hummers method. All the results proved that the adopted synthesis method and the use of Z6020 silane produced a more homogeneous GO/SiO₂
nanohybrid particle due to the establishment of covalent bonds and self-condensation which lead to a higher morphological and thermal properties then GSA particles.

We believe that the proposed synthesis strategy is an alternative approach for taking the advantages of both GO nanosheets and SiO2 nanospheres and could be an appropriate method for large scale and industrial production. This kind of nanoparticles should be a great candidate to develop polymer based nanocomposites coatings with high-anticorrosion and barrier properties.

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