

Research Article

Photocatalytic Activity in Phenol Removal of Water from Graphite and Graphene Oxides: Effect of Degassing and Chemical Oxidation in the Synthesis Process

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Developing new materials or modifying the existing ones is an amply field studied in the world of research. Due to its outstanding physical and chemical properties, graphene is attractive material for new applications. The methodologies for obtaining graphene are diverse and have changed over time. Graphene oxide is a versatile form of graphene, due to the presence of oxygenated functional groups. Chemical oxidation of graphite and exfoliation by ultrasonic waves is one of the preferred methods to obtain graphene oxide; chemical oxidation time and the degassing effect in the ultrasonic bath are parameters that play an important role in the features and properties of graphene oxide. Thus, in this study, the conventional times used for the oxidation of graphite and degassing in an ultrasonic bath to obtain graphene oxide were modified. The structural changes in the carbon materials were evaluated based on their photocatalytic activity in the removal of an organic pollutant in water (removing up to 38% of phenol). The band gaps of the graphitic materials were obtained by UV-vis obtaining a value range of 1.5–4.7 eV and the structure and morphology of the carbon materials were characterized by infrared and Raman spectroscopies and transmission electron microscopy, respectively.

1. Introduction

Graphite oxide (GO) is a graphite derivative which contains covalently attached oxygen-containing groups on its layers. The oxygen groups are often hydroxyl, epoxy, and carboxyl, which are embedded within the carbon layers and are generated in the course of GO synthesis by strong chemical oxidation [1, 2]. As GO contains a high concentration of exposed oxygen-containing functional groups, it is considered to be a multifunctional organic network which can be applied in a wide range of chemical transformations, most commonly with the C-OH [3, 4], OH [5, 6], and epoxy groups [7–9].

Nowadays, there are three major methods used to synthesize GO, that is, the Brodie, Staudenmaier, and Hummers

methods [10, 11]. The level of oxidation can be varied on the basis of the method, the reaction conditions, and the precursor graphite used. One of these methods, the Hummers method, is generally considered the best [12, 13], and the majority of researchers have followed it. Also, in 2008, a modification of the Hummers method improved the oxidation degree and thus the method increases its utility [14].

Graphene oxide (GEO) can be obtained from GO under conditions of thermic or ultrasonic exfoliation, which leads to individual graphene oxide sheets. The formation of oxygenated functional groups in GO makes them easier to exfoliate into monolayers of GEO by stirring or mild sonication. To obtain fewer sheets, the sonication method is used and

commonly takes place in an ultrasonic bath operated at 40 kHz [15, 16] or 50 kHz [17, 18].

It is already well known that, chemically, GO and GEO have similar structures. Both possess stacked structures with chemical functionality on their basal planes and at their edges. The difference between them is the number of stacked layers. Both materials GO [19, 20] and GEO [21] have photocatalytic activity, shown by water splitting and hydrogen production, and can reduce resazurin into resorufin [22]. Also, both carbon structures can be doped with different materials [23, 24] or atoms [25] to improve their performance as photocatalysts.

Additionally, reduced graphene oxide (RGO) modified with different compounds has been successful as a photocatalytic material in the removal of heavy metals [26, 27] and organic compounds [28]. However, the photocatalytic activity of unmodified GO and GEO has not been studied in the removal of organic compounds in water. Within the new materials to remove phenol in water are Nano ZnO [29], CeO₂/MgAl layered double hydroxides [30], and TiO₂-rGO nanocomposites [31]; in these researches, materials get removed among 50% and 90% of the phenol concentration, considering different concentrations in each study; these new materials are efficient in the removal of phenol by heterogeneous photocatalysis but long time of reaction is required and in some cases the addition of oxygen/air to accelerate the photocatalytic process; also, it is needed to work in the electron-hole recombination. The advantage to exploit the possibilities to use GO and GEO as photocatalyst is that oxygen groups in the surface that could contribute to these parameters can be subtracted generating an interesting field, related with doped and nondoped graphitic materials in photocatalysis. Thus, it is important to verify the photocatalytic activity of both of these carbon unmodified materials for the removal of organic compounds in water, considering the different changes that carbon structures undergo during the synthesis process and the effects on their structures.

In the synthesis of GO and GEO, the sonication process and the oxidation time are key factors; however, only a few studies have investigated modifications to these parameters to obtain the optimal conditions for a good chemical functionalization [32–34]. Oxidation time changes the number of functional groups and carbon hybridized sp² in both materials and the chemical effects of ultrasound derived from acoustic cavitation are relevant in this kind of process. The effects of cavitation are the basis of sonochemistry and are extremely effective in liquid degassing [35, 36]. Thus, in this study, changes in the carbon structure (GO and GEO) produced by degassing and oxidation time were analyzed. Additionally, the photocatalytic activity was demonstrated by the elimination of phenol in water. Transmission electron microscopy was used to examine the topography of the materials, FT-IR was applied to identify the functional groups in the oxidized samples, Raman spectroscopy was utilized to identify the hybridization of carbon samples and UV-vis spectroscopy, and Tauc's method was used to obtain the band gap of the graphene oxide materials.

2. Material and Methods

2.1. Synthesis of Graphite and Graphene Oxides. Graphite oxide (GO) was synthesized from graphite by an established method [14, 37]; however, the reaction time was modified to determine the influence of this parameter on the oxidation process and therefore on the structure of GO and then on the band gap results and photocatalytic activity. The reactants used were graphite (Electron Microscopy Sciences, number 70230), sulfuric acid (H₂SO₄, Baker, 98%), potassium permanganate (KMnO₄, Merck), hydrogen peroxide (H₂O₂, Baker, 30%), and distilled water (H₂O).

H₂SO₄ (46 mL) was added into the reaction flask maintained at 0°C (±2°C) (ice bath), and then graphite (2 g) and KMnO₄ (6 g) were added slowly. After an increase in temperature to 35°C (±2°C), the mixture was stirred by a magnetic stirring bar and mixed for three different time periods: 2, 4, and 6 h. Later, the excess water was incorporated into the mixture and H₂O₂ (10 mL) was added until there was no gas production.

Filtration was performed with distilled water in a glass filter, and the obtained brown GO was dried in an oven (Barnstead Thermolyne, Model 3478) at 65°C for 12 h.

Then, a solution containing 100 mg of dried GO in 10 mL of H₂O was prepared. This solution was sonicated for 3 h at room temperature with the aid of an ultrasonic bath (Branson 1510R-MTH) in two different degassing units (55 and 65). The modification in degassing could contribute to changing the amount of functional groups in the graphene oxide obtained and in the sheets numbers, thereby promoting changes in their physical and structural features of graphene oxide sheets (GEO).

2.2. Graphene Oxide Samples. The designation of the graphite oxide samples (GO) is GO-2, GO-4, and GO-6, in which the number after the acronym means the time in hours of GO oxidation reaction. For the graphene oxide samples, the designation is GEO-2-55, GEO-2-65, GEO-4-55, GEO-4-65, GEO-6-55, and GEO-6-65. The first number also indicates the oxidation time in hours from GO, and the second number (55 and 65) indicates the degassing in the ultrasonic bath.

2.3. Photocatalytic Test. Photocatalytic and photolysis experiments were conducted in a batch photoreactor. The small-scale photoreactor system consisted of a Pyrex glass flask with a capacity of 250 mL. The glass flask reactor was filled with 100 mL of an aqueous solution containing phenol (100 ppm). The pH of the solution was neutral. The temperature of the reactor was maintained at room temperature and the suspension was magnetically stirred. The experiments were performed with 2 h of UV light irradiation, following the methodology for phenol determination (ASTM-D1783-0). The percentage values of phenol removal were determined by taking aliquots at the beginning and end of the reaction time. The solution was irradiated with a UV-vis lamp at 254 nm and 10 and 20 mg of graphite oxide and graphene oxide were used in the photocatalytic test. The absorption of the samples was analyzed using a UV-vis spectrophotometer (Spectronic Genesys 2PC) at 510 nm. Oxides of graphene

and graphene were removed by filtration. Also, in the same reactor under similar conditions but without UV light, the absorption test was performed for GEO-2-55 and GEO-2-65. Each experiment was performed in at least triplicate under identical conditions. After each reaction, the resulting solution was poured into a fine filter and the filtered solution graphitic material phenol is separated and sheltered in a container. The carbon nanomaterials used also could be reutilized if desorption process is realized, research related with the number of reuses of this carbon nanomaterials, could be done in the future to meet the functionality as reusable photocatalyst.

2.4. Characterization. Fourier transform infrared spectroscopy (FT-IR) studies were performed using a Bruker-Vector 33 with a scanning range of $4000\text{--}500\text{ cm}^{-1}$ with resolution of 4 cm^{-1} . The test samples were pressed into KBr tablets and all samples were prepared with the same ratio of KBr-graphitic materials. Micro-Raman (Dilor, Lab Ram) measurements were carried out at 488 nm incident laser light with a spectral resolution of 1 cm^{-1} . The physical morphology of graphene oxide sheets was analyzed by transmission electron microscopy (TEM), using a JEOL-1010 microscope operated to 80 kV . UV-vis spectroscopy (Spectronic Genesys 2PC with wavelength of $200\text{ to }1000\text{ nm}$) was used to assess the chromophores present in the graphitic materials and to determine the band gap, the samples were prepared in aqueous solution, and a quartz cell of 3 mL was used for analysis.

3. Results and Discussion

3.1. FT-IR Spectroscopy. Figure 1 shows the FT-IR spectra of GEO and GO; the band at 1730 cm^{-1} is attributed to carboxyl groups $\nu(\text{C}=\text{O})$. The peak around 1619 cm^{-1} corresponds to the stretching band $\nu(\text{C}=\text{C})$. The peaks at $1020\text{--}1090\text{ cm}^{-1}$ are attributed to the C-O-C, C-O, and C-OH bands, giving evidence for the presence of oxygen-containing groups, caused by the chemical oxidation reaction [8, 14, 37]. All spectra show these characteristic bands reported for graphite and graphene oxides. The band corresponding to $\nu(\text{C}=\text{O})$ at 1730 cm^{-1} shows a decrement in the spectra related with GEO obtained at 65 degassing units. In the same spectra appear the band at 1690 cm^{-1} and also a clear increment in the band around 1410 cm^{-1} ; these two signals indicate that more degassing units produce carboxylate groups [38]. The signals related to $\nu(\text{C}=\text{O})$ and C-O-C, C-O, and C-OH are more intense in the GO-4 and GO-2 spectra than in the GO-6 spectrum, indicating that longer oxidation does not necessarily produce a larger amount of functional groups in GO. In the same figure, the bands related to the oxidation process (C=O, C-O-C, C-O, and C-OH) show clear differences with greater intensity in the spectra of GEO when 55 units of degassing were used (GEO-2-55, GEO-4-55, and GEO-6-55) in comparison with the spectra of the samples treated with 65 units of degassing (GEO-2-65, GEO-4-65, and GEO-6-65). In addition, the spectra of GEO exposed to 65 units of degassing show the peak around 1645 cm^{-1} attributed to $\nu(\text{C}=\text{O})$ related to ketones, and also the bands at 1690 cm^{-1} have been related

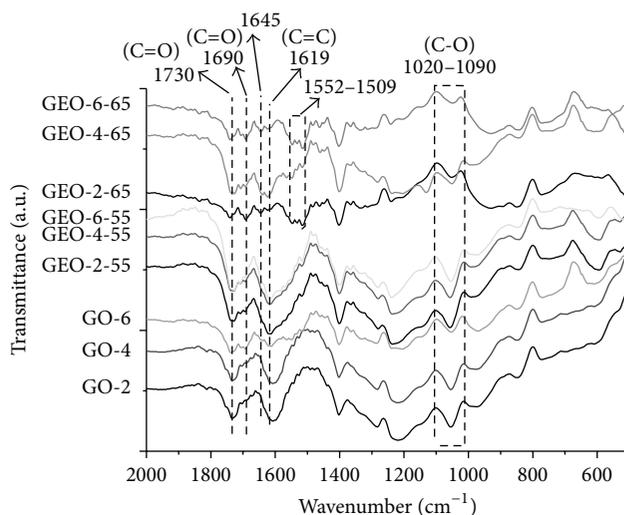


FIGURE 1: FT-IR spectra of GO-2, GO-4, GO-6, GEO-2-55, GEO-4-55, GEO-6-55, GEO-2-65, GEO-4-65, and GEO-6-65.

with quinone groups [2, 38]. These signals are not presented in the spectra of GEO exposed to 55 units of degassing; this fact, together with the bands only presented in the spectra of the samples treated to 65 units of degassing, located around 1552 cm^{-1} to 1509 cm^{-1} and assigned to unoxidized graphitic domains [39], corroborates the notable changes in the GEO samples due to the degassing action of the ultrasonic bath. This effect can be attributed to the cavitation effect which generates increased outgassing; this effect is stronger with 65 units than with 55 units of degasification, leading to the elimination of oxygenated functional groups present in the graphite oxide.

3.2. Raman Spectroscopy. Raman spectroscopy is a widely used tool for the characterization of carbon products, especially considering conjugated and double carbon-carbon bonds. The Raman spectra of GO are shown in Figure 2(a) and for GEO in Figure 2(b). The graphitic materials show different peaks; the D band located around 1356 cm^{-1} is due to the out-of-plane breathing mode of sp^2 atoms and is active in the presence of defects, whereas 2D (around 2680 cm^{-1}) is the second order of this vibrational mode [5, 6]. The tangential G mode at 1593 cm^{-1} corresponds to the first-order scattering of the E_{2g} mode in-phase vibration of the graphite lattice. In the same way, the intensity and broadness of the 2D band in graphene is a function of the number of layers. This band is a second-order process related to a phonon near the K point in graphene, activated by double resonance processes, which are responsible for its dispersive nature and cause a strong dependence on any perturbation to the electronic and/or phonon structure of graphene. Thus, the 2D band is very important for characterizing specific sp^2 nanocarbons [6, 7, 14, 37]. The ratio of the intensities of the D and G bands (ID/IG) is often used as a means of determining the number of layers in a graphene sample and its overall stacking behavior; high D/G ratios indicate a high degree of exfoliation

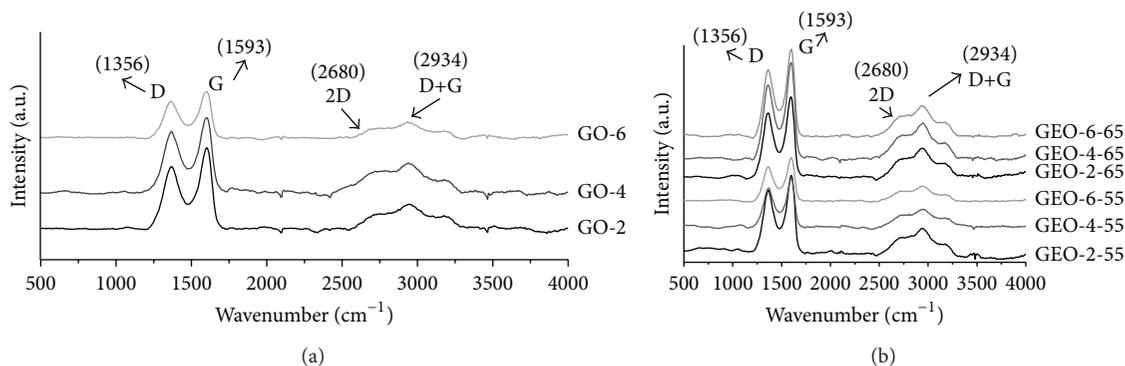


FIGURE 2: Raman spectra of (a) GO-2, GO-4, and GO-6 and (b) GEO-2-55, GEO-4-55, GEO-6-55, GEO-2-65, GEO-4-65, and GEO-6-65.

disorder. Changes in the Raman spectra occur due to changes in the number of layers or due to doping and reflect the evolution of the electronic structure and electron-phonon interactions [40]. Consequently, changes in the 2D band in graphene oxide are related to considerable defects in the graphene sheet, produced by oxidation [41]. In the Raman spectra (Figure 2(a)) of GO-2, GO-4, and GO-6, it can be seen that the D, G, 2D, and D+G bands (at 1356 cm^{-1} , 1593 cm^{-1} , 2680 cm^{-1} , and 2934 cm^{-1} , resp.) are very similar in terms of intensity and width for GO-2 and GO-4 but dissimilar in comparison with the GO-6 spectrum, where the three bands decrease in intensity. Also in the Raman spectra all samples show the D+G band which is a combination band related with the concentration of defects due to the present oxygenated groups [42, 43].

The ID/IG ratio (Table 1) was lower in GO-6 than in GO-4. Thus, in 6 h of reaction to oxidize graphite, apparently more oxygen-containing functional groups were not generated. This also agrees with the bands analyzed by infrared spectroscopy. Table 1 includes all the values for the ID/IG ratios. Conversely, the ratio obtained for ID/IG decreased for the GEO samples obtained with 65 degassing units, in comparison with GEO treated at 55 units; thus, it can be concluded that sheets with less oxygenated groups were obtained with 65 units in comparison with sheets obtained at 55 units, due to degassing effect.

3.3. Transmission Electron Microscopy. In Figure 3(a), the sheets obtained for GEO-2-55 are shown. The characteristic roughness of graphene sheets is observed, including transparency and uniformity in the morphology. Figure 3(b) shows sheets of GEO-4-55, which are less corrugated than GEO-2-55, but similar in transparency although more stacked. Figure 3(c) shows GEO-6-55 materials, and the sheets tend to be in shades of gray, depending on the number of overlapping layers of carbon; it is important to mention that the formation of a layer or multilayer is not a factor controlled by chemical method. Thus, Raman and FT-IR characterizations show a clear evidence that graphite and graphene oxides are obtained, and also, the morphology obtained by TEM is considered acceptable and useful to this research. For the GEO sheets treated at 65 degassing units,

TABLE 1: ID/IG and IG/ID ratio values in the Raman spectra.

Sample	ID/IG
GO-2	0.67
GO-4	0.71
GO-6	0.68
GEO-2-55	0.75
GEO-4-55	0.74
GEO-6-55	0.72
GEO-2-65	0.7
GEO-4-65	0.65
GEO-6-65	0.66

less corrugated surface is observed in GEO-4-65 and GEO-6-65 (Figures 3(e) and 3(f)) than in the samples obtained with degassing at 55 units, but the irregular shape of the sheets is similar to that obtained for GEO treated at 55 units. Figure 3(d) shows the micrograph obtained from sheets of GEO-2-65, which are very similar to GEO-2-55 sheets in transparency and roughness and unlike the sheets of GEO-4-65 (Figure 3(e)) have more transparency, indicating less carbon layers. Finally in the sheets of GEO-6-65 (Figure 3(f)), clear folds at the ends of these layers are found; although the transparencies as well as shape are very similar to the other sheets, these sheets are stacked. It is important to mention that, in a previous work reported, the average value of the thickness of this kind of GEO was 0.56 nm obtained by atomic force microscopy (AFM). According to the thickness, GEO should not exceed 3 or 4 overlapping leaves [37]. And in the case of graphite oxide (GO), it is known that the thickness value is in the order of microns.

Thus, the morphology of GEO is apparently not modified in general as a factor of degassing units, and only the roughness and stacking were affected by oxidation time. So, the samples treated for 2 h independently of degassing units are the most rough (Figures 3(a) and 3(d)); 4 h of oxidation time produces smoother sheets (Figures 3(b) and 3(e)). However, if a longer oxidation time was used (6 h), the surface is not modified in relation to the samples treated for 4 h, but more stacking is found in these sheets (Figures 3(c) and 3(f)).

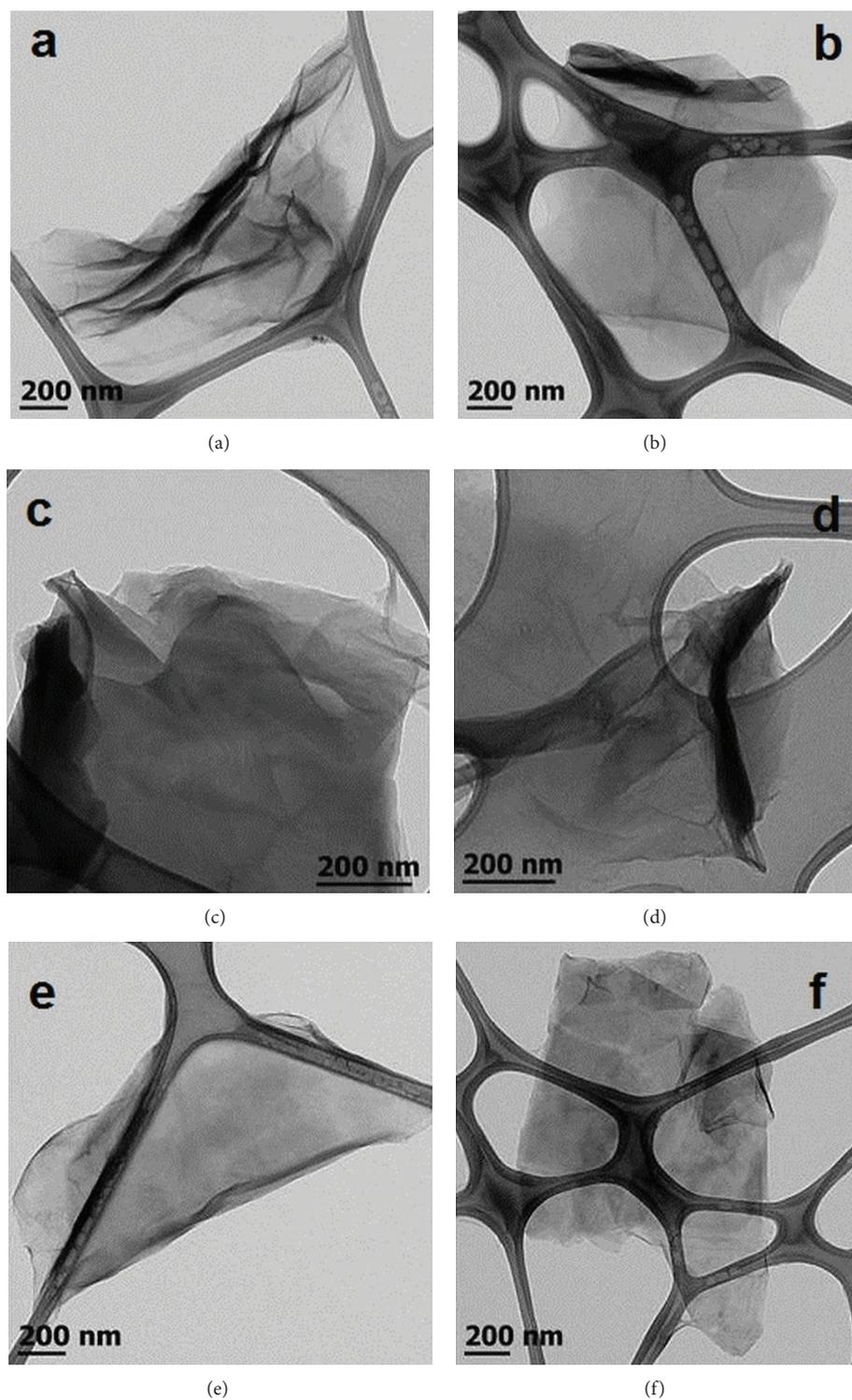


FIGURE 3: TEM micrographs of (a) GEO-2-55, (b) GEO-4-55, (c) GEO-6-55, (d) GEO-2-65, (e) GEO-4-65, and (f) GEO-6-65.

3.4. UV-Vis Spectroscopy. The UV-vis spectra (Figure 4) exhibit two characteristic absorption bands for graphite oxide (GO) and graphene oxide (GEO), with a maximum at 228 nm, corresponding to the $\pi-\pi^*$ transitions of aromatic C=C and C-C bonds; the shoulder at ~ 300 nm is ascribed to

the $n-\pi^*$ transitions of C=O bonds [27, 35, 36]. These bands are presented specifically with the incidence of ultraviolet light in graphitic materials and in the GO and GEO samples of this research are clearly defined. Thus, the values of the band gap are calculated based on these spectra.

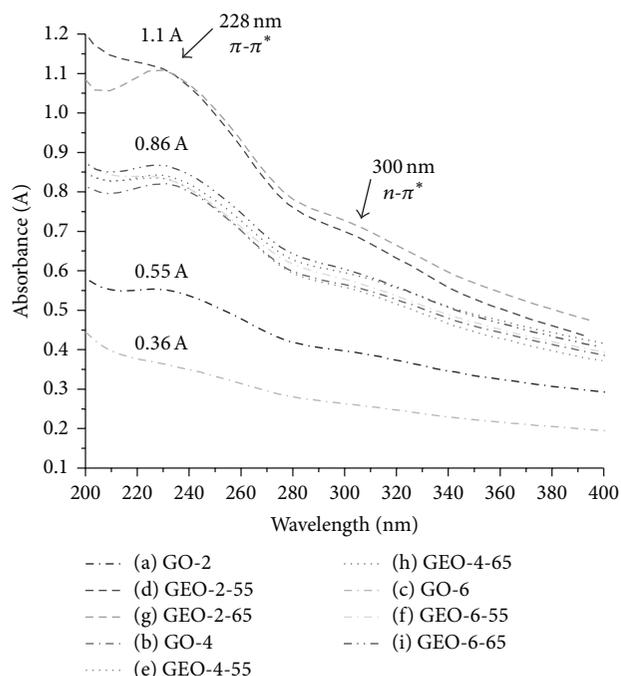


FIGURE 4: UV-vis spectra of (a) GO-2, (b) GO-4, (c) GO-6, (d) GEO-2-55, (e) GEO-4-55, (f) GEO-6-55, (g) GEO-2-65, (h) GEO-4-65, and (i) GEO-6-65.

Band gap values were determined from Tauc's plot of $(\alpha h\nu)_2$ versus photon energy ($h\nu$), using the UV-vis spectra wherein the absorption coefficient varies as a function of frequency, according to $\alpha h\nu \propto (h\nu - E_g)^n$, where $h\nu$ is the photon energy and E_g is the band gap; $n = 2$ for indirect transitions (I) and $n = 1/2$ for direct transitions (D) [2, 27, 37], as shown in Table 2. Also in Figure 5 is shown a graph for the value obtained in GEO-2-55 specifically. The indirect band gap values correspond to small values, because the absorption band is not specified, but there are areas with lower activity and thus the chart detects even the slightest intersection giving a small band gap value, indicating that certain spaces in the graphitic materials have less separation of the valence and conduction bands.

3.5. Photocatalysis Test. To determine the photocatalytic effect of GEO-2-55, adsorption and photolysis tests were performed (see the results in Table 3). The aim was to confirm that this material was not effective as an absorbent material, because the percentage of phenol removal was almost nil, indicating that it was not able to absorb phenol molecules. However, we observed that 7% of phenol is removed from water by photolysis. Photolysis is a technique that is not used alone, and although the encapsulated air in the reactor and incident UV light generate oxidizing radicals able to degrade phenol in water, these are not able to achieve a high degradation percentage [44, 45].

Table 4 shows a summary of photocatalytic probe using different samples of GO and GEO. With 200 mg/L of graphitic material in the photocatalytic process, the increase in

TABLE 2: Band gap values (E_g), indirect (I) and direct (D), obtained through spectra of UV-vis for all samples of GO and GEO.

Sample	E_g (I) eV	E_g (D) eV
GO2	2.80	4.70
GO4	1.94	4.90
GO6	1.90	3.96
GEO-2-55	1.87	4.04
GEO-4-55	1.89	4.1
GEO-6-55	1.89	4.07
GEO-2-65	1.5	4.0
GEO-4-65	2.05	4.6
GEO-6-65	2.1	4.0

TABLE 3: Removal percentage of phenol in water by absorption test using GEO-2-55 and GEO-2-65 and in the photolysis test.

Absorption for 2 h	
Absorbent	Removal percentage
GEO-2-55 (100 mg)	0%
GEO-2-55 (200 mg)	1%
GEO-2-65 (100 mg)	0%
GEO-2-65 (200 mg)	0%
Photolysis (UV-254 nm) for 2 h	
Phenol solution	Removal percentage
100 ppm	7%

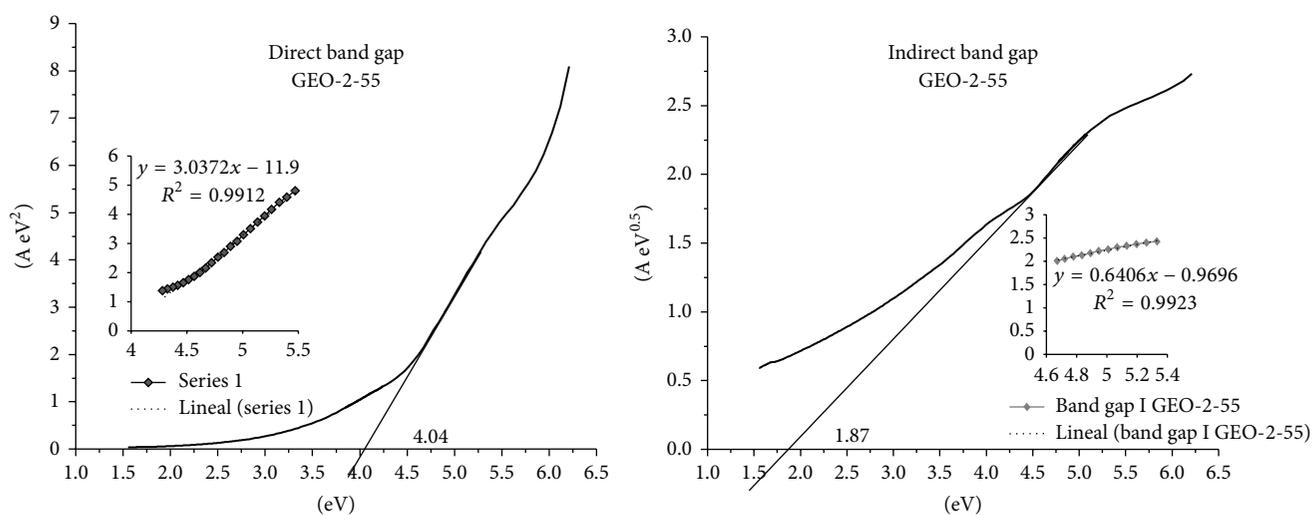
the percentage of phenol degradation in water is improved in the case of GEO-2-55, GEO-4-55, and GEO-6-55. The increase in degradation is minimal when the probes are used at 200 mg/L rather than 100 mg/L for almost all samples. When the amount of catalysts is duplicated, the increase in the degradation percentage of phenol is around 1% in the majority of probes, with the exception of GEO obtained with 55 units of degassing (GEO-2-55, GEO-4-55, and GEO-6-55).

As mentioned before, the photocatalytic activity of GO and GEO has been verified using different probes [19–28] but any for eliminating phenol in water. Thus, the results indicate that the minimal degradation of phenol obtained using GO is due to the dimensions of the graphite oxide (3D), since a greater number of graphite layers allow less interaction between oxygenated functional groups (C=O, C-O, and O-H) on the graphite surface with phenol molecules, such that the photocatalytic process is impaired.

In the case of the samples of GEO-2-65, GEO-4-65, and GEO-6-65, the low percentage of degradation is attributed to the nature of the samples, which contained fewer functional groups at the surface as they are eliminated by the degassing process in the ultrasonic bath (this is demonstrated in an earlier section). It is clear that oxygenated groups play an important role in generating oxidizing radicals (such as OH or O^{*}) in photocatalytic probes. It is assumed that the effect in removal that is generated in the photolysis is not completely reflected in photocatalysis, because graphitic material in the solution of phenol modifies the interaction of UV radiation with the solution, and then, the effect of radiation is not

TABLE 4: Removal percentage of phenol in water in the photocatalytic test using GO and GEO.

Photocatalysts (10 mg)	Removal percentage of phenol solution	Error (%)	Photocatalysts (20 mg)	Removal percentage of phenol solution	Error (%)
GO-2	5.05%	±0.1	GO-2	7.89%	±0.1
GO-4	3.89%	±0.1	GO-4	4.81%	±0.08
GO-6	6.43%	±0.05	GO-6	7.89%	±0.08
GEO-2-55	12.11%	±0.05	GEO-2-55	38.62%	±0.1
GEO-4-55	4.90%	±0.15	GEO-4-55	12.29%	±0.1
GEO-6-55	6.32%	±0.05	GEO-6-55	14.96%	±0.08
GEO-2-65	3.45%	±0.1	GEO-2-65	5.86%	±0.06
GEO-4-65	2.84%	±0.05	GEO-4-65	4.86%	±0.08
GEO-6-65	3.87%	±0.15	GEO-6-65	5.05%	±0.12

FIGURE 5: Determination of direct and indirect band gap from Tauc's plot of $(\alpha h\nu)_2$ versus photon energy $(h\nu)$, for GEO-2-55.

exactly the same, in spite of the encapsulated air, because the photocatalyst plays a prominent role.

New materials have been tested in the removal of phenol by means of photocatalysis, not just carbon materials, in previous section in this paper, such as Nano ZnO [29], CeO₂/MgAl layered double hydroxides [30], and GRO-TiO₂ nanocomposites [31], and these photocatalytic materials remove 80, 50, and 90% of phenol, respectively. Additionally, photocatalytic activity by graphene oxide (GEO) composite materials has been reported, for instance, TiO₂-graphene oxide [46] and, more recently, BiOI-GO (graphene oxide) composite [47] and GO/PPy (graphene oxide/polypyrrole) [48]; these materials remove around 90% of phenol. However, the application of pure graphite materials as photocatalysts materials has been investigated minimally [49, 50], generating great expectation to investigate the photocatalytic effectiveness individually in the removal of pollutants in water. Thus, the 38% of removed phenol is low compared with the percentage of other materials mentioned. However, the aim to evaluate the effectiveness of photocatalyst of GEO-2-55 opens a field to apply these materials without doping in the removal of organic pollutants and diversify its potential

as a photocatalyst material. Nevertheless, the most important aspect of this study is understanding the parameters that play an important role in carbon material preparation, specifically in this research with a focus on photocatalytic activity without other materials. Also, if the amount of catalyst material that is used in this research is considered, the removal of phenol in water achieved, that is, 38%, 12%, and 15%, using GEO-2-55, GEO-4-55, and GEO-6-55, respectively, could be significant. The removal percentage of phenol is almost proportional to the increase in the photocatalytic effect for these samples (GEO treated with 55 degassing units; see Table 4). Thus, these results open up the range of possible applications of graphene oxide as a photocatalytic material by considering the processing conditions. Also, it might assume good semiconductor capabilities for these GEO materials, which could be activated with UV light at $\lambda = 254$ nm; this positively affected the reaction rate of catalysis process and therefore promoted the degradation percentage of phenol present in water. This indicates that GEO-55, especially GEO-2-55, may be a good material for use as a photocatalyst in heterogeneous photocatalysis. Also, to complement the results, the sample that shows the best removal percentage

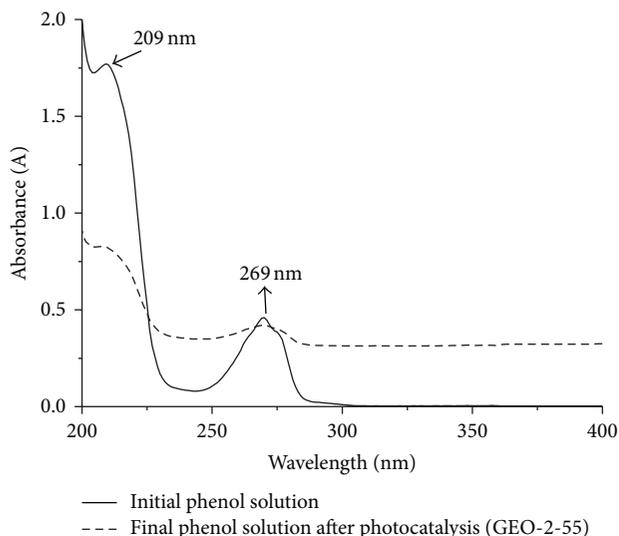


FIGURE 6: UV-vis spectra of photocatalytic test for phenol solution using GEO-2-55, in the initial and final of reaction (2 hr).

was characterized by UV-vis spectroscopy. The spectra of initial and final solution of phenol used in the photocatalytic reaction with GEO-2-55 are shown in Figure 6; the results show a clear reduction in the characteristic absorption bands of phenol after photocatalytic process performed.

4. Conclusion

Oxygen functional groups generated in graphitic sheets do not increase linearly with chemical oxidation time in graphite oxide; only some morphological changes are observed in terms of stacking and roughness with variation in this parameter. However, in obtaining graphene oxide using an ultrasonic bath with degassing, modification of the other parameter produces changes in the graphene oxide sheets. This removes oxygenated functional groups and affects the photocatalytic activity of graphene oxide. Although the band gap values are in the same range, oxygenated functional groups appear to play an important role. On the other hand, the number of layers and therefore dimensionality are also a factor affecting photocatalytic activity in this case and led to a difference between the effectiveness of graphite oxide and graphene oxide. It is found that GEO-2-55 is the graphitic nanometric material that presents the best photocatalytic activity. The combination of oxygenated functional groups, the band gap range (1.8–4 eV), and the roughness in the sheet are more efficient with the incidence of UV light at 254 nm than only photolysis or adsorption process.

The removal of 38% of the phenol in water with GEO-2-55 is a very favorable result and makes this material viable and efficient for applications in alternative advanced oxidation processes. GEO-2-55 shows photocatalytic activity with the incidence of UV light and can be considered as a photocatalytic material capable of degrading organic pollutants present in water. Results presented here could be as starting point to get chemical and physical structural

changes in this kind of carbon materials using single changes in processing conditions and then understand the principles of graphitic materials, to get better performance as photocatalyst to remove some pollutants from water or other typical applications of photocatalysis.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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