Applied Physics A (2020) 126:519
https://doi.org/10.1007/s00339-020-03710-3

Applied Physics A
Materials Science & Processing

Optical properties of graphene oxide thin film reduced by low-cost diode laser

Mahmoud Gomaa[1] · **Gamal Abdel Fattah**[1]

Received: 28 March 2020 / Accepted: 5 June 2020 / Published online: 13 June 2020

Abstract
A non-toxic, fast, low-cost, single step and highly efficient reduction method is proposed in this research for the development of high-quality multilayer graphene film using laser diode. Graphene oxide film is irradiated with a diode laser (808 nm, 6 W) for different exposure times (1, 1.5, 2, 2.5, and 3 min.) to reduce it. Surface damage was observed on the graphene oxide (GO) film for exposure time greater than 3 min and at power 6 W. Different measurement techniques (Raman spectroscopy, Uv–visible absorption, photoluminescence and FTIR) are used to study the optical properties of rGO films. After laser irradiation, the Raman spectra of rGO films showed peaks for the D, G, and 2D bands centered at 1335, 1581, and 2710 cm^{-1}, respectively. A small increase in the (I_D/I_G) ratio was observed and showed that greater reduction occurred after the graphene oxide film exposure time of 3 min. The as-prepared film of graphene oxide that showed absorption peak at 235 nm was red-shifted to 270 nm after 3 min of laser diode irradiation for reduction. The PL measurements were performed, where the sample was excited by Ar-ion laser (488 nm) and the PL signal was amplified using lock in techniques. The photoluminescence emission spectrum was measured at different positions on the graphene oxide film reduced for 3 min to show the effect of the energy spatial distribution of the laser diode beam on the rGO film. The maximum PL peaks gradually blue shifted and the peak intensities decreased as the measurements go toward the center of the reduced film. The FT-IR spectrum revealed that there are no hydroxyl, carboxyl, and epoxy groups and new peaks were found assigned to functional groups such as CH_2, C–H by reducing the graphene oxide with laser diode for 3 min.

Keywords Graphene oxide · Diode laser · Reduced graphene oxide · The photoluminescence of reduced graphene oxide · The Raman spectra of reduced graphene oxide

1 Introduction

The simple term graphene oxide (GO) was used to refer to single-atom carbon layers (called graphene) with functional oxygen groups bonded to both sides and flake area, usually defined by atomic C/O ratios of less than 3 and generally closer to 2 [1]. The addition of the oxygen groups results in double bond breakage that binds the atoms of carbon together, and the material loses its electrical conductivity [2]. The existence of hydroxyl and epoxy groups at the edges of the basal planes of the graphene oxide and carbonyl and carboxyl groups allows the flake as a whole to become polar and therefore to disperse in water or any other polar solvent [3]. Graphene oxide is considered a promising material for graphene processing and does not have a completely planar shape due to the presence of oxygen atoms on the top and bottom of the carbon sheet. Graphene oxide consisting of carbon sp^2 and sp^3 atoms, carbon sp^2 atoms are bound to the neighboring carbon atoms, while the sp^3carbon atoms are bound to the functional oxygen group [4, 5]. The sp^2/sp^3 ratio will vary the emission of photoluminescence from the visible to the near infrared range, transforming graphene oxide from an insulator into a semiconductor, and tuning the graphene oxide band gap [6]. Graphene oxide preparation involves two main steps; first, Graphite powder is oxidized to form graphite oxide, which can be dispersed in water or any other polar solvent, and then graphite oxide is exfoliated by sonication to form monolayer, bilayer or GO sheet suspensions in different solvents [7]. The popular method for graphene oxide preparation is the Hummers (oxidation with $KMnO_4$ and $NaNO_3$ in concentrated H_2SO_4) which involves

✉ Mahmoud Gomaa
mdgomaa@niles.edu.eg

[1] National Institute of Laser Enhanced Sciences, Cairo University, Giza 12613, Egypt

a large quantity of reagents and a long reaction time [8–10], but in our previous work, a new method for the synthesis of graphene oxide by microwave plasma chemical vapor deposition on quartz substrates was established [11]. One of the aims of synthesizing graphene oxide is the large-scale processing of graphene-like carbon materials through a reduction process, and reduction is a method that transforms sp^3 carbon to sp^2 carbon [12]. Graphene oxide is not conductive and is very sensitive to higher temperatures. These properties can be restored by removing the groups formed which contain oxygen and this reduced form of graphene oxide (GO) is called reduced graphene oxide (rGO). The elimination of oxygen-containing functionalities and simultaneous transfer of basal plane carbon atoms from sp^3 to sp^2 hybridization, restores the system's aromaticity to some degree. The reduction produces a major change in the properties and microstructure of graphene oxide, so that some obvious changes can be seen directly indicating the reduction effect of the various reduction processes [13–15]. These obvious enhancements include optical properties, electrical conductivity and the atomic ratio C/O. Optical observation is a way of tracking the changes in graphene oxide before and after reduction. The reduced graphene oxide film has a metallic luster, due to the reduction process enhancing the electrical conductivity, increasing the charge carrier concentration and mobility, so increasing the reflection of incident light [16–18]. In the configuration of graphene oxide, chemical functional groups bound to the plane and structural defects in the plane, both affect the electrical conductivity of graphene oxide, while functional groups bound to the edge have a smaller effect on the conductivity. The reduction of graphene oxide can be achieved by two factors: removing functional groups on the plane, such as epoxy and hydroxyl groups, whereas other groups on the sides, such as carboxyl, carbonyl and ester groups, have less effect on the conductivity of an rGO sheet and on the treatment of structural defects [19]. There are two outcomes that should be considered during the removal of functional groups, the oxygen-containing groups should be removed and the areas should be restored to a long-range structure after removal, so that carrier transport pathways within the rGO layer are available. Thermal deoxygenation, chemical deoxygenation, or a combination of thermal annealing and chemical reactions cause loss of functional groups [19]. Thermal deoxygenation of GO can be induced by increasing the temperature. In de-hydroxylation, hydroxyl groups leave the graphene sheet directly, creating an OH radical and a graphene radical that does not allow a lattice defect to form in the plane [20, 21]. There is no exact Tc for the epoxy groups, but the hydroxyl groups can be completely eliminated by vacuum above 650 °C, while the epoxy groups are retained [22]. The carbonyl groups are more stable and can be eliminated above 1730 °C while gradually reducing the carboxyl groups by 100–150 °C. There are many methods to reduce graphene oxide, such as thermal annealing [microwave, photo-irradiation] and chemical reduction [solvothermal reduction, multistep reduction, electrochemical reduction and photochemical reduction] [23]. Laser irradiation has been adopted for GO reduction due to its unique advantages including reliability, ease, low cost and flexible design. Various low wavelength lasers, such as KrF excimer laser, pulsed UV laser, Nd: YAG laser, picosecond laser and femtosecond laser pulse, have been used to reduce graphene oxide by photochemical removal of the oxygen functional group [24, 25]. These lasers are equipped with a high cost instrumentation which cannot achieve high efficiency and a complex reduction. The 808 nm wavelength diode laser was considered to be one of the cheapest laser technologies, easy, flexible design and the absorption of irradiation transforms electromagnetic energy into thermal energy [18]. The optical absorption at 808 nm is very low from the absorption spectrum, so a modulated value of laser intensity is converted to heat. Thus, the graphene oxide film can easily be heated to a high temperature, depending on the laser power.

The thermal annealing (photo-irradiation) will be used in this work to reduce graphene oxide (rGO) films by light irradiation from a laser diode (LIMO 50 W with a wavelength of 808 nm), to be discussed in detail in the next section. Laser irradiation give us an efficient way to process reduced graphene oxide in the shortest possible time and with environmental protection. Also, experimental photoluminescence measurements is performed using Ar^+ Laser with an excitation wavelength of 488 nm, to investigate the effect of spatial energy distribution of unfocused laser beam and the degree of reduction on reduced graphene oxide film at different positions, as discussed below in more details. The optical properties of reduced graphene oxide film have been studied using different techniques such as Raman spectroscopy, Uv–visible absorption, photoluminescence (PL) and FTIR.

2 Experimental setup

In this study, different samples of graphene oxide films deposited on quartz substrates to stand the laser temperature were illuminated by a laser beam of a power (6 W, 808 nm) and a beam area of 1cmx1 cm for (1, 1.5, 2, 2.5 and 3 min). The laser beam port was 30 cm away from the sample. The illumination setup is shown in Fig. 1. The laser beam was directed toward the graphene oxide film without using any lens to achieve a large area of irradiation. The irradiated area absorbed the laser energy and the energy was rapidly converted to local heat, and GO was reduced. Surface damage was observed on the GO film for exposure time greater than 3 min and at a power 6 W.

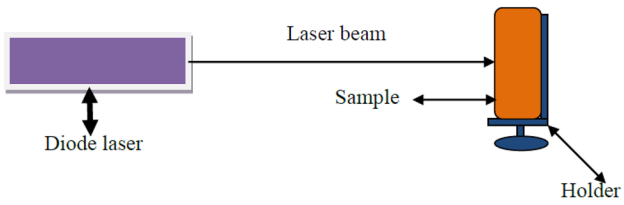

Fig. 1 Experimental setup of the reduction of graphene oxide thin film by diode laser

The graphene oxide film deposited by microwave plasma chemical vapor deposition, [11], is shown in Fig. 2a. The photograph, Fig. 2b, shows the reduction effect of laser exposure time of 3 min on the GO film. At 6 W and after 3 min of exposure time a dark black area was seen at the center of the produced spot, indicating rGO layer formation as shown in Fig. 2b. Different techniques are used to characterize the reduced samples, as discussed in more detail below.

2.1 Characterization techniques

Graphene oxide (GO) samples are reduced by irradiating the samples by a diode laser beam with a wavelength of 808 nm and a power of 6 W for different exposure times. Reduced graphene oxide films were characterized by a variety of techniques to obtain information on the reduction effect. The results of the experimental measurements of the reduced sample structural and optical properties are described in detail in the sections below. In this study, we characterized the reduced films at different exposure times (1, 1.5, 2, 2.5 and 3 min) by Raman spectroscopy alone, indicating the maximum reduction occurring at 3 min exposure time. Other characterizations, such as UV–Vis absorption, photoluminescence, and FT-IR analysis, are only measured for film irradiated by diode laser beam for 3 min of exposure time.

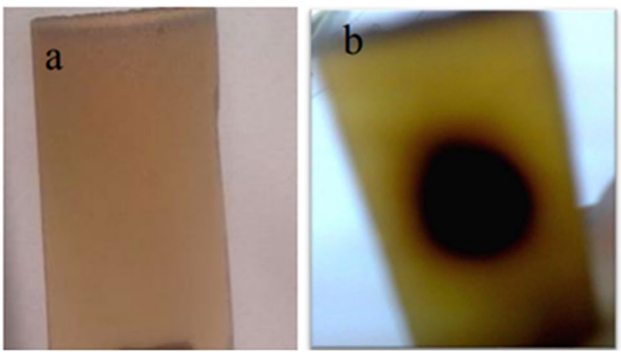

Fig. 2 **a** GO film photo before laser irradiation and **b** a photo of reduced film after 3 min laser exposure time (6w, 808 nm)

3 Results and discussion

3.1 Raman spectroscopy

The Raman shifts range from 500 to 4500 cm^{-1} in Figs. 3 and 4 show the Raman spectra measured on graphene oxide films before and after laser irradiation for different exposure times (1, 1.5, 2, 2.5 and 3 min). The Raman spectrum of graphene oxide films displayed only D and G bands before the laser reduction as shown in Fig. 3, and the G band intensity was higher than that of the D band. After laser irradiation the Raman spectra of GO films showed the D, G, and 2D bands centered at 1335, 1581, and 2710 cm^{-1}, respectively, as shown in Fig. 4. The bands D and G reflected the material defects and the vibration of the carbon sp^2 atoms in the plane. The stacking order was expressed in 2D band.

The measured Raman spectra at different reduction times of all rGO films showed that the presence of the 2D peak corresponded to graphene formation due to the laser-induced reduction reaction as shown in Fig. 4. The relative intensity ratio of D/G (I_D/I_G) was commonly used to measure material defects in graphene and the relative intensity of 2D/G (I_{2D}/I_G) was used to calculate the number of layers of graphenes [26]. The average (I_D/I_G) ratio of GO films was 0.6. The (I_D/I_G) ratios for rGO films were 0.81, 0.86, 0.87, 0.89, and 0.9 while the laser irradiation exposure time were 1, 1.5, 2, 2.5, and 3 min, respectively. The Raman spectra showed that the maximum reduction occur after 3 min of exposure time. The I_D/I_G ratios of rGO films was greater than those of GO films because some material defects were created by the laser irradiation. The increase in strength ratio (I_D/I_G) with the increase in reduction time suggests that new graphitic domains were being created and that the number of sp^2

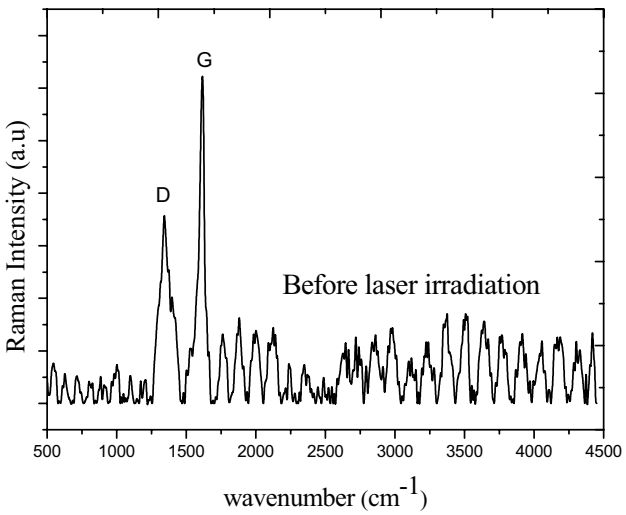

Fig. 3 Raman spectrum measured for GO film before laser irradiation

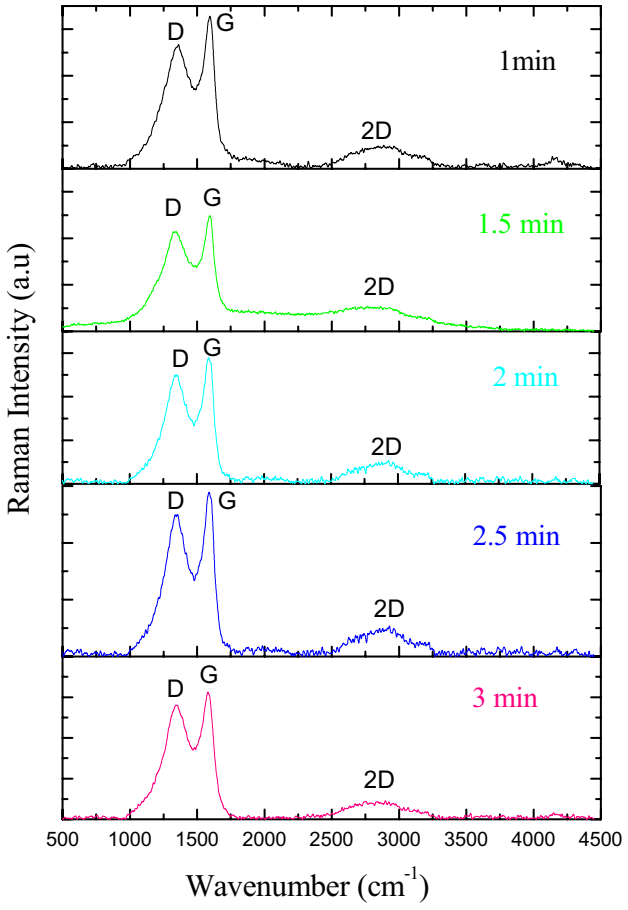

Fig. 4 Raman spectra measured of rGO films at different reduction times (1, 1.5, 2, 2.5 and 3 min)

clusters was increasing. This increase also shows the elimination of functional groups, suggesting that the increased defects from GO to graphene and these observations are consistent with the References [13, 27]. The (I_{2D}/I_G) ratios of rGO films were 0.16, 0.19, 0.23, 0.26 and 0.3, while the laser irradiation times were 1, 1.5, 2, 2.5 and 3 min, respectively. In this study, rGO film's (I_{2D}/I_G) ratios were less than 1 so it was revealed that the products have multilayer graphene structures. The relation between the intensity ratio (I_D/I_G) and reduction time is shown in Fig. 5. Due to the development of new sp^2 clusters which are smaller in size relative to those present in graphene oxide before reduction, the reduction process reduced the average size of the sp^2 clusters. The intensity ratio (I_D/I_G) measures the degree of disorder in the sample and the size of the sp^2 domains average. The crystal size was determined using the following equation found by Cancado et al. [28], using the (I_D/I_G) intensity ratio.

$$L_a(\text{nm}) = (2.4 \times 10^{-10})\lambda^4 \left(I_D/I_G\right)^{-1} \quad (1)$$

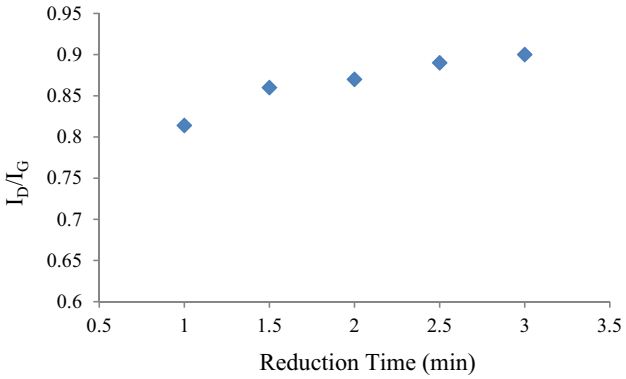

Fig. 5 Variation of the D-band to G-band intensity ratio (I_D/I_G) as a function of the reduction time

where λ is the wavelength of dispersive Raman microscope (Bruker–Senterra), which using a laser beam of 532 nm.

The relationship between average size (L_a) and reduction time is expressed in Fig. 6. Table1 displays the (I_D/I_G) and (I_{2D}/I_G) ratios of GO, rGO films, and L_a (nm) for different laser irradiation times.

3.2 UV–Vis absorption spectra

The spectra for UV–Vis absorption of graphene oxide as prepared and the reduced graphene oxide with exposure time of 3 min are shown in Fig. 7. From the figure, one can observe that graphene oxide exhibits a maximum absorption at 235 nm due to π–π^* transitions of aromatic C–C bonds. Such results indicate that GO has functional groups of oxygen. Graphene oxide film used in this work was prepared by microwave plasma chemical vapor deposition as in our previous work [11]. The absorption of 300 nm ($n \rightarrow \pi^*$ transitions) of graphene oxide indicated the degree of oxidation.

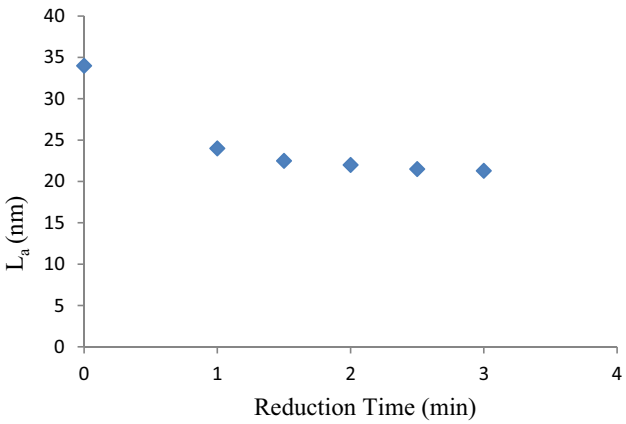

Fig. 6 Relation between the average size (L_a) and reduction time

Table 1 The I_D/I_G and I_{2D}/I_G ratios of GO and rGO films for different laser irradiation times

Film types	GO	rGO				
Laser irradiation time (min)	–	1	1.5	2	2.5	3
I_D/I_G	0.6	0.81	0.86	0.87	0.89	0.93
I_{2D}/I_G	–	0.16	0.19	0.23	0.26	0.3
L_a (nm)	32	24	23	22	21	20

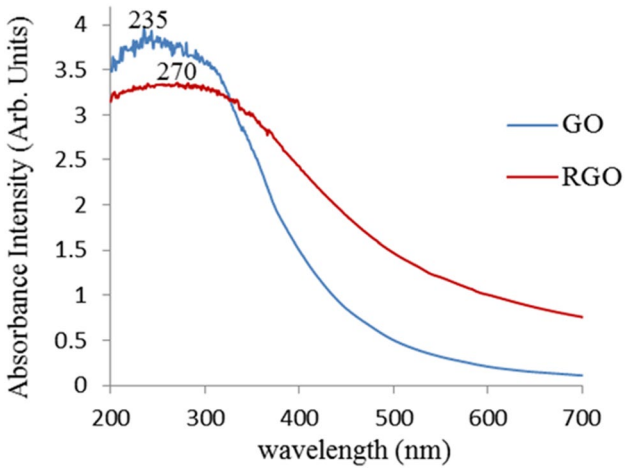

Fig. 7 UV–Vis absorption spectra of the as-prepared GO and the reduced GO for 3 min using diode laser of a power 6 W, 808 nm

The temperature of plasma during the deposition of GO reduces the oxidation degree and the absorption of 300 nm in GO spectrum, as reported in previous studies [13–15, 21]. The 235 nm absorption peak red-shifted to 270 nm for the graphene oxide film reduced for 3 min, which can be related to the restoration of the electronic conjugation of C–C bonds within the graphene film. The red shift in maximum absorption is due to the elimination of oxygen-containing bonds from graphene oxide film and is related to an increase in the atomic volume of sp^2 hybridized carbon atoms with an increase in reduction time. The red shifting of the maximum absorption also shows the degree to which graphene oxide was reduced, indicating that the hybridized carbon atoms sp^3 were transformed into hybridized carbon atoms sp^2 and is consistent with the one reported [29].

3.3 Photoluminescence analysis

One of the most important photoluminescence properties of GO is that the oxidation degree of the graphene defines the emission wavelength. The degree of oxidation was investigated by the photoluminescence. The Photoluminescence setup consisted of Ar^+ Laser [LEXEL 95] with an excitation wavelength of 488 nm and a power of 125 mW, a 750 mm long monochromator [SPEX 750 M] with a grating (1200 gr/mm) and the output signal has been amplified using the lock-in technique (SR510).

The graphene oxide film was reduced by using laser irradiation from a laser diode (3 min, 6 W). Photoluminescent (PL) emission spectra of reduced graphene oxide film was measured for different positions on the film surface, from the center of the reduced spot on the film (position 0) to the edge of the spot (position 4) as shown in Fig. 8a. The PL emission was measured at different distances (0, 1, 2, 3 and 4 mm) to study the effect of the spatial distribution of laser beam energy on the degree of the reduction process, where each area of the sample was exposed to different laser energy (according to Gaussian distribution and due to the relatively large laser beam area) Fig. 8b, c. Depending on the distribution of laser beam energy and the degree of oxidation measured at different positions on the reduced GO film as shown in Fig. 8a. From Fig. 8, the degree of reduction increased toward the reduced spot center.

The PL emission spectra of reduced graphene oxide film with an excitation wavelength of 488 nm have blue-shifted, became narrow and the peak intensity decreased with the measurement toward the center of the spot, this can be due to the higher intensity distribution of the diode laser beam at the center, which implies a higher reduction effect. The PL peak position of as-prepared graphene oxide film at (604 nm) before reduction as reported in previous work [11] shifted to 568 nm after reduction for 3 min in this work as in Fig. 9. The maximum shift of PL peak was observed at the center of the reduced graphene oxide after reduction for 3 min. The maximum PL peaks were blue-shifted from 587 nm at the edge of the film (4 mm) to 568 nm at the center of reduced graphene oxide film (0 mm). The relationship between peak positions and different positions of reduced graphene oxide film is shown in Fig. 10.

The PL emission intensity peak of the reduced graphene oxide film has decreased from the edge to the center due to the decrease in defect density owing to the gradual removal of epoxy and hydroxy functional groups from the GO film with increased laser beam effect and reduction time [30, 31]. The relationship between the peak intensity and positions on RGO film is shown in Fig. 11. Also, the maximum half-width (FWHM) of the PL peak is lower at the center of the reduced graphene oxide film compared to (FWHM) at the edge. This can be due to the sp^3 decreases as the laser beam

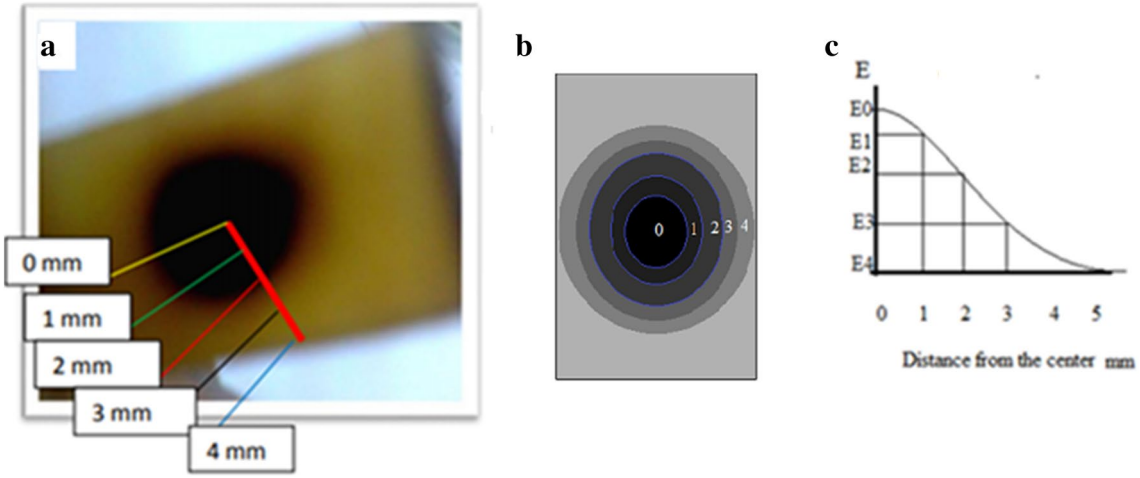

Fig. 8 **a**, **b** Positions of PL measurements on the reduced graphene oxide film (3 min, 6 W and 808 nm) started from the center (0) to the edge of the sample (4), **c** the relation between energy of laser beam and positions where 0 correspond to maximum energy E0 and 4 correspond to minimum energy E4

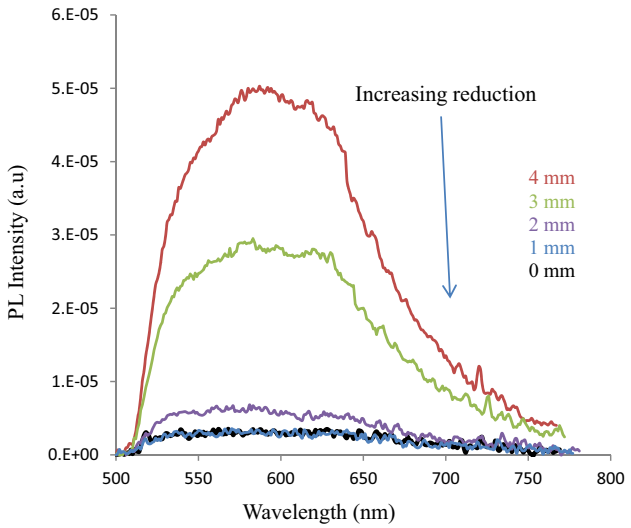

Fig. 9 PL spectra with increasing photothermal reduction on the reduced graphene oxide film surface at different positions from the center

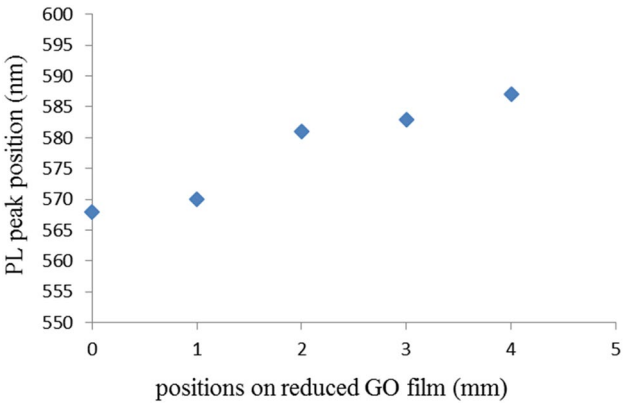

Fig. 10 Relation between the PL maximum peak positions and positions on RGO film (3 min, 6 W)

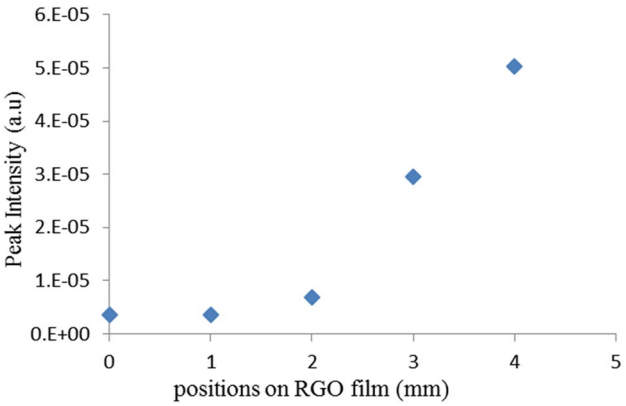

Fig. 11 Relation between peak intensity and positions on RGO film (3 min, 6 W)

effect increases and where at the center of spot was reduced but not over-heated and amorphous contents did not exist or were undetectable. FWHM's relationship to the position on RGO film is shown in Fig. 12.

3.4 FT-IR analysis

FT-IR spectral analysis was performed to confirm the chemical structure of the graphene oxide and reduced graphene oxide as shown in Fig. 13a, b. The characteristic FT-IR properties of graphene oxide deposited by microwave plasma chemical vapor deposition on quartz substrate are shown

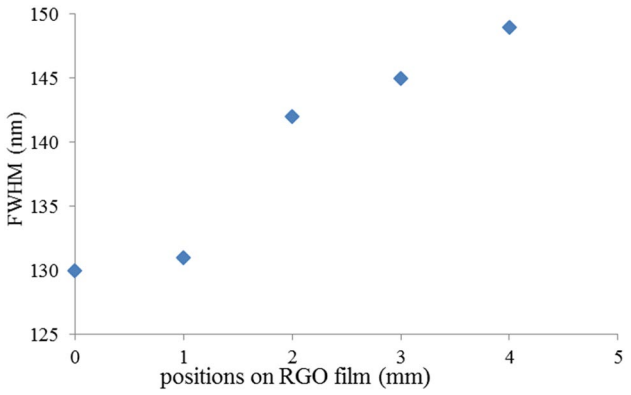

Fig. 12 Relation between FWHM and positions on RGO film (3 min, 6 W)

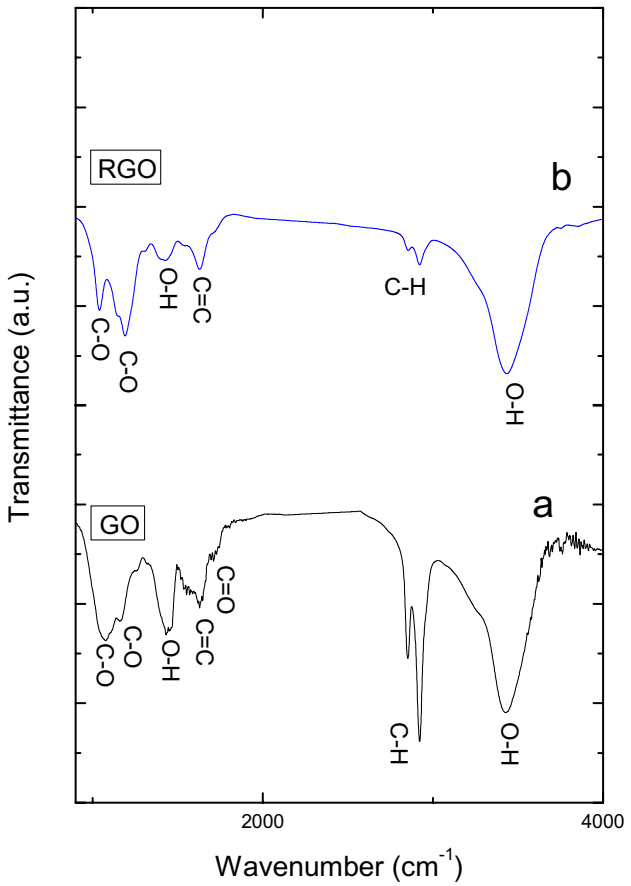

Fig. 13 **a** FT-IR spectra of graphene oxide **b** and reduced graphene oxide (3 min, 6 W)

in Fig. 13a, as discussed in [11]. The characteristic FT-IR properties of graphene oxide include the presence of different forms of oxygen functionality verified by the 1625 cm^{-1} and 1735 cm^{-1} bands corresponding to the aromatic groups C=C and C=O carbonyl/carboxyl, respectively. Bands of 1078 and 1196 cm^{-1}, corresponding to C–O in epoxy groups, strong bands of 2860 and 2926 cm^{-1}, corresponding to symmetrical and anti-symmetrical stretching vibrations of CH_2 and 3425 cm^{-1} bands corresponding to O–H as shown in Fig. 13a. In the case of reduced graphene oxide, FT-IR spectroscopy has also confirmed the reduction of the groups containing oxygen in GO by laser diode irradiation, as shown in Fig. 13b. Due to deoxygenation of the GO oxygenated functionalities, the absorption bands of groups containing oxygen and a double at 2860 and 2926 cm^{-1} decreased in intensity after the reduction of graphene oxide film by diode laser of a power 6 W for 3 min. Therefore, the stretching vibration of C=O has disappeared which suggests a reduction in graphene oxide. According to the remaining epoxy groups, the stretching vibration of C–O at 1078 and 1196 cm^{-1} becomes sharper, even after laser reduction [29].

4 Conclusion

In this research, the reduction of graphene oxide films using laser diode with a wavelength of 808 nm at room temperature and ambient air was successfully achieved. Different optical techniques characterized the r-GO films, which examined the changes in thin film surfaces and approved the efficient, fast and non-toxic method of reducing graphene oxide film by the laser. The experimental setup of photoluminescence enables us to measure the emission of photoluminescence at different positions on the reduced film of graphene oxide and was aware of the degree of oxidation or reduction and effect of laser at each position. At different reduction times, the measured Raman spectra of all rGO films revealed that the higher reduction in our work occurred at 3 min exposure time. We noticed the laser damage to film surface after 3 min. The apparent 2D presence of all rGO films peaks in the Raman spectra also corresponded to the formation of graphenes. The use of continuous wave diode laser will promote the development of multilayer graphene films, which is highly expected to welcome the processing based on diode laser by industrialists and scientists. There are several potential applications using the reduced film of graphene oxide, such as the super capacitor. For future research, by altering the laser parameters and increasing the electrical conductivity, we aim to increase the amount of reductions.

References

1. T. Szabó, A. Szeri, I. Dékány, Composite graphitic nanolayers prepared by self-assembly between finely dispersed graphite oxide and a cationic polymer. Carbon **43**, 87–94 (2005)
2. C. Zhu, S. Guo, Y. Fang, S. Dong, Reducing sugar: new functional molecules for the green synthesis of graphene nanosheets. ACS Nano **4**, 2429–2437 (2010)

3. S.J. Park, R.S. Ruoff, Chemical methods for the production of graphene. Nat. Nanotechnol. **4**, 217–224 (2009)
4. G. Eda, Y.-Y. Lin, C. Mattevi, H. Yamaguchi, H.-A. Chen, I.-S. Chen, C.-W. Chen, M. Chhowalla, Blue photoluminescence from chemically derived graphene oxide. Adv. Mater. **22**, 505–509 (2010)
5. J.L. Chen, X.-P. Yan, K. Meng, S.-F. Wang, Graphene oxide based photoinduced charge transfer label-free near-infrared fluorescent biosensor for dopamine. Anal. Chem. **83**, 8787–8793 (2011)
6. T. Gokus, R.R. Nair, A. Bonetti, M. Böhmler, A. Lombardo, K.S. Novoselov, A.K. Geim, A.C. Ferrari, A. Hartschuh, Making graphene luminescent by oxygen plasma treatment. ACS Nano **3**, 3963–3968 (2009)
7. H. He, J. Klinowski, M. Forster, A. Lerf, A new structure model for graphite oxide. Chem. Phys. Lett. **287**, 53–56 (1980)
8. W.S. Hummers, R.E. Offeman, Preparation of graphitic oxide. J. Am. Chem. Soc. **80**(6), 1339 (1958)
9. J.Y. Lim, N.M. Mubarak, E.C. Abdullah, S. Nizamuddin, M. Khalid, Recent trends in the synthesis of graphene and graphene oxide-based nanomaterials for removal of heavy metals—A review. J. Ind. Eng. Chem. **66**, 29–44 (2018)
10. R. Giovannetti, E. Rommozzi, M. Zannotti, C.A. D'Amato, Recent advances in graphene based TiO_2 nanocomposites ($GTiO_2Ns$) for photocatalytic degradation of synthetic dyes. Catalysts **7**(10), 305 (2017)
11. M. Gomaa, G.A. Fattah, Synthesis of graphene and graphene oxide by microwave plasma chemical vapor deposition. J. Am. Sci. **12**(3), 72–80 (2016)
12. H. Zhang, J. Wang, Q. Yan, W. Zheng, C. Chen, Yu, Z, Vacuum-assisted synthesis of graphene from thermal exfoliation and reduction of graphite oxide. J. Mater. Chem. **21**, 5392–5397 (2011)
13. K. Krishnamoorthy, M. Veerapandian, R. Mohan, S.-J. Kim, Investigation of Raman and photoluminescece studies of reduced graphene oxide sheets. Appl. Phys. A. **106**, 501–506 (2012)
14. T.F. Emiru, D.W. Ayele, Controlled synthesis, characterization and reduction of graphene oxide: a convenient method for large scale production. Egypt. J. Basic Appl. Sci. **4**, 74–79 (2017)
15. S. Kumar, A. Kumar, Chemically derived luminescent graphene oxide nanosheets and its sunlight driven photocatalytic activity against methylene blue dye. Opt. Mater. **62**, 320–327 (2016)
16. S. Stankovich, D.A. Dikin, R.D. Piner, K.A. Kohlhaas, A. Kleinhammes, Y. Jia et al., Synthesis of graphene-based nanosheets via chemical reduction of exfoliated graphite oxide. Carbon **45**(7), 1558–1565 (2007)
17. J. Chen, B. Yao, C. Li, G. Shi, An improved Hummers method for eco-friendly synthesis of graphene oxide. Carbon **64**, 225–229 (2013)
18. Y.-L. Zhang, L. Guo, H. Xia et al., Photoreduction of graphene oxides: methods, properties, and applications. Adv Opt Mater. **2**, 10–28 (2014)
19. W. Gao, L.B. Alemany, L. Ci, P.M. Ajayan, New insights into the structure and reduction of graphite oxide. Nat. Chem. **1**(5), 403–408 (2009)
20. X. Gao, J. Jang, S. Nagase, Hydrazine and thermal reduction of graphene oxide: reaction mechanisms, product structures, and reaction design. J. Phys. Chem C. **114**(2), 832–842 (2009)
21. K. Krishnamoorthy, M. Veerapandian, G.-S. Kim, S.J. Kim, A one step hydrothermal approach for the improved synthesis of graphene nanosheets. Curr. Nanosci. **8**, 934–938 (2012)
22. S. Pei, J. Zhao, J. Du, W. Ren, H.-M. Cheng, Direct reduction of graphene oxide films into highly conductive and flexible graphene films by hydrohalic acids. Carbon **48**(15), 4466–4474 (2010)
23. T.V. Cuong, V.H. Pham, Q.T. Tran, S.H. Hahn, J.S. Chung, E.W. Shin, E.J. Kim, Photoluminescence and Raman studies of graphene thin films prepared by reduction of graphene oxide. Mater. Lett. **64**, 399–401 (2010)
24. D. Bhattacharjya, C.-H. Kim, J.-H. Kim, I.-K. You, J.B. In, S.-M. Le, Fast and controllable reduction of graphene oxide by low-cost CO_2 laser for supercapacitor application. Appl. Surf. Sci. **462**, 353–361 (2018)
25. C.-R. Yang, S.-F. Tseng, Y.-T. Chen, Characteristics of graphene oxide films reduced by using an atmospheric plasma system. Nanomaterials. **8**, 802 (2018)
26. Hu Maocong, Z. Yao, X. Wang, Graphene-based nanomaterials for catalysis. Ind. Eng. Chem. Res. **56**(13), 3477–3502 (2017)
27. H. Kang, A. Kulkarni, S. Stankovich, R.S. Ruoff, S. Baik, Restoring electrical conductivity of dielectrophoretically assembled graphite oxide sheets by thermal and chemical reduction techniques. Carbon **47**, 1520–1525 (2009)
28. L.G. Cancado, K. Takai, T. Enoki, M. Endo, Y.A. Kim, H. Mizusaki, A. Jorio, L.N. Coelho, R.M. Paniago, M.A. Pimenta, General equation for the determination of the crystallite size La of nanographite by Raman spectroscopy. Appl. Phys. Lett. **88**, 163106(1–3) (2006)
29. D. Li et al., Processable aqueous dispersions of graphene nanosheets. Nat. Nanotechnol. **3**, 101 (2008)
30. R. Maiti, A. Midya, C. Narayana, S.K. Ray, Tunable optical properties of graphene oxide by tailoring the oxygen functionalities using infrared irradiation. Nanotechnology. **25**, 495704 (2014)
31. Q. Mei, B. Liu, G. Han, R. Liu, M.-Y. Han, Z. Zhang, G. Oxide, From tunable structures to diverse luminescence behaviors. Adv. Sci. **6**, 1900855 (2019)