

Effect of rGO Doping Concentration on Optical Parameters of rGO-PVP Nanocomposite Toward Optoelectronics Applications

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ABSTRACT:

The important features of reduced graphene oxide (rGO), including good electrical conductivity, large surface area, and mechanical strength, have attracted widespread attention in many fields. These features elevate rGO to an important position in optical applications and energy technologies. In this study, a composite material was constructed by mixing rGO with polyvinylpyrrolidone (PVP), where PVP is a soluble, biocompatible, and flexible polymer. The nanocomposite with various concentrations of rGO was produced and its optical capabilities were investigated. The optical properties of the prepared samples including refractive index, optical conductivity, extinction coefficient, optical dielectric constant, and loss tangent were improved with increasing rGO concentrations. Moreover, with increasing rGO concentration, the optical energy gap of the synthesized composites decreased, indicating increased electronic transitions, and the PVP-G5 sample showed the lowest energy gap value, indicating better electronic transfer. This work opens the way for the development of effective, cost-effective, and environmentally friendly materials for various devices.

KEYWORDS: Nanocomposite, Reduced graphene oxide, Polyvinylpyrrolidone, Optical properties, Energy bandgap.

تأثير تركيز تطعيم صفائح أكسيد الجرافين المختزل النانوية على الخصائص البصرية لمركب rGO-PVP للتطبيقات البصرية الإلكترونية

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مستخلص:

لقد جذبت الميزات المهمة لأكسيد الجرافين المختزل (rGO)، بما في ذلك التوصيل الكهربائي الجيد والمساحة السطحية الكبيرة، والقوة الميكانيكية، اهتماماً واسع النطاق في مجالات عديدة. تعمل هذه الميزات على رفع مستوى rGO ليحتل مكانة مهمة في التطبيقات البصرية وتقنيات الطاقة. في هذه الدراسة، تم إنشاء مادة مركبة عن طريق خلط rGO مع بولي فينيل بيروليدون (PVP)، حيث يعد PVP بوليمراً قابلاً للذوبان ومتوافقاً حيوياً ومرناً. تم إنتاج المركب النانوي بتركيز متعددة من rGO وفحص قدراتها البصرية. تم تحسين الخصائص البصرية للعينات المحضرة بما في ذلك معامل الانكسار والتوصيل البصري ومعامل الخمود وثابت العزل البصري بسبب زيادة تركيزات rGO. علاوة على ذلك، فإنه بزيادة تركيز rGO، تقل فجوة الطاقة البصرية للمركبات المصنعة، مما يشير إلى زيادة الانتقالات الإلكترونية حيث أظهرت العينة PVP-G5 أقل قيمة في فجوة الطاقة، مما يشير إلى نقل إلكتروني أفضل. ويفتح هذا العمل الطريق لتطوير مواد فعالة، وجيدة من حيث التكلفة، وصديقة للبيئة لمختلف الأجهزة.

الكلمات الدالة: المركبات النانوية، أكسيد الجرافين المختزل، بولي فينيل بيروليدون، الخصائص البصرية، فجوة الطاقة.

1. INTRODUCTION

The amazing features of reduced graphene oxide (rGO) such as decent electrical conductivity, massive surface area, and mechanical strength [1], attracted more attention in a variety of fields. These characteristics make rGO an enviable material in electronics, energy storage, and sensors [2, 3]. The reduction procedure itself, which removes oxygen groups from the graphene oxide structure, improves the electrical characteristics of GO, making it a strong and flexible component for complex materials [2, 4].

Polyvinylpyrrolidone (PVP) is a widely utilized polymer due to their perfect solubility in water and many kinds of organic solvents, as well as its biocompatibility and ability to create stable films [5]. PVP is widely involved in food processing, medicines, and utilized in numerous chemical processes as a stabilizing factor [6]. The flexibility and ability to create hydrogen bonds with other molecules make PVP a good candidate for developing complex and low-cost materials with better characteristics [7]. PVP is a polymer that is made from raw materials resulting from

oil and gas. Oil equipment is often exposed to corrosion due to its presence in an acidic and salty environment. The role of PVP as a coating to prevent corrosion. Many studies were conducted that PVP-based composites can utilized as a coating for oil transformation pipe (carbon steel), , optoelectronic devices, and it has proven successful in preventing corrosion [8-10].

The combination of rGO and PVP develops unique qualities of both parts [11], rGO-doped PVP nanocomposites enhance several physical aspects e.g. the optical properties, electrical conductivity, mechanical strength, and thermal stability [12]. These nanocomposite materials expose advanced characteristics and can participate in many technological applications since rGO and PVP mixture may interact to improve light absorption, photoluminescence, and other optical properties [13, 14]. rGO-doped PVP nanocomposite has the ability to extend the light absorption better than PVP alone, due to the presence of free electrons in rGO that absorb light across a wide range of wavelengths, especially in the ultraviolet range. These adjustable properties may enhance and enlarge their applica-

tions in solar cells, photosensors, optical fibers, and optoelectronic devices that require a greater amount of light absorption [12, 13].

In summary, we aimed to enhance the optical properties of PVP by adding different concentrations of rGO nanosheets. The increasing rGO concentrations in rGO-doped PVP nanocomposites can be attributed to improve optical absorption, energy bandgap, and many optical parameters. The understanding of the principles beneath the optical characteristics of rGO-doped PVP nanocomposites will support to design of more efficient and cost-effective materials for usage in a variety of optoelectronics devices. Lastly, this strategy broadens the uses of such kinds of composites and offers new paths for material science study including anti-corrosion coating and oil recovery applications [15].

2. Experimental and Methods

An exfoliation method was utilized to synthesize the rGO nanosheets under optimal conditions, more details were described in our prior study [16, 17]. To manufacture the PVP and rGO-PVP nanocomposite with diverse rGO con-

centrations, powder PVP and powders PVP and rGO were diffused in (100 ml) of deionized water and ultrasonicated for (60 min.) to ensure uniform diffusion of rGO in the PVP matrix. The resultant mixture was combined as PVP-G1 (12.5% rGO, 87.5 % PVP), PVP-G2 (25 % rGO, 75 % PVP), PVP-G3 (50 % rGO, 50 % PVP), PVP-G4 (75 % rGO, 25%PVP), and PVP-G5 (87.5 % rGO, 12.5 % PVP). Using a drop-casting method, 0.2 μ l of each above combination was put onto a clean quartz slide and dried in an oven at (40 °C) for (1 hour). Scanning electron microscopy (SEM) was used on the generated electrodes to evaluate their morphology with INSPECT F 50-FEA SEM. The optical characteristics of the nanocomposite films were examined using UV-vis spectroscopy on a T70/T80 Series spectrometer, with data taken in the wavelength range of (300-800 nm). Raman spectroscopy, using a type inViaTM (Renishaw, UK), helped detect the vibrational modes of rGO and PVP inside the nanocomposite films, revealing insights into their interactions. Furthermore, Fourier Transform Infrared (FTIR) spectroscopy was used using a Biotech Engineering Management

spectrometer to identify the functional groups in the nanocomposite films and validate the formation of bonds between rGO and PVP.

3. Results and Discussion

Fig. 1 demonstrates the SEM images of the prepared samples. Fig. 1a shows thin and transparent layers of rGO randomly overlapping with each other. The surface appears wrinkled and uneven,

indicating the presence of many defects and cracks in the structure of the material. In Fig. 1b, the layers of rGO are shown to be more cohesive and interconnected and the surface appears smoother and flatter compared to the image in Fig. 1a, indicating an addition of PVP has supported to cover some defects and cracks. In Fig. 1c, the image reveals fine details of the structure of rGO-doped PVP at the nanoscale.

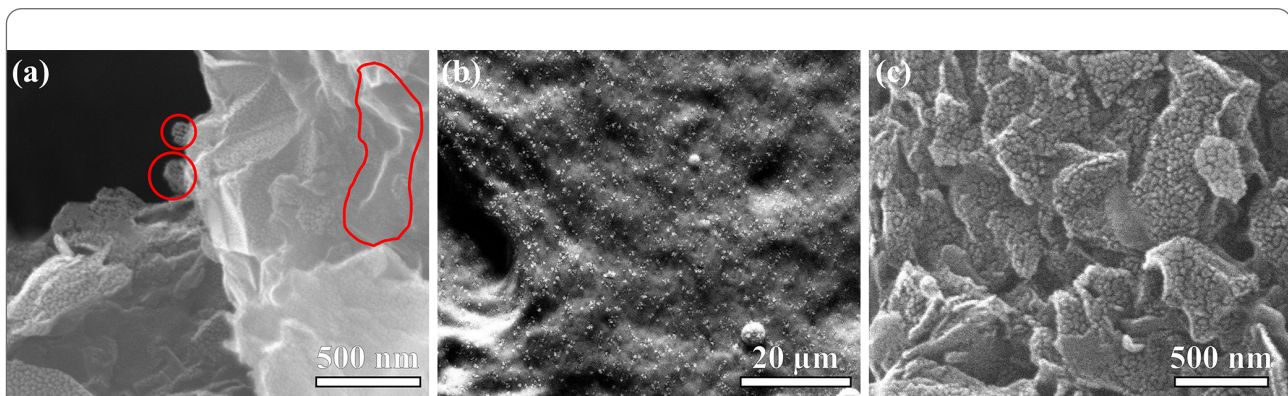


Fig. 1. SEM photograph of (a) rGO nanosheet (red colored area), (b) rGO-doped PVP nanocomposite, and (c) rGO-doped PVP nanocomposite at the nanoscale, the magnifications of the SEM images are 120,000x, 4,000x, and 60,000x, respectively.

Fig. 2 represents the Raman test of rGO, PVP, and PVP-G samples. Raman spectroscopy is a valuable method for studying carbon-based materials' structures. In Raman spectroscopy, Carbon-based materials exhibit two primary vibrational modes (D and G). The D-band represents the level of

order/disorder caused by a breathing k-point phonon with A_{1g} symmetry. The G-band, corresponding to the E_{2g} phonon of Sp^2 hybrid carbon atoms, indicates the stacking structure [11]. The peaks at 1327 and 1573 cm^{-1} identify the D- and G- bands for RGO, respectively [18]. The Raman spectrum of PVP-G

film exposes a broad peak at around 1570 cm^{-1} , signifying the existence of a carbonyl ($\text{C}=\text{O}$) bond in the polymer structure. Meanwhile, the vibrations of C-N bonds present a peak at 1349 cm^{-1} [19]. In rGO, the D-band equals 1327 cm^{-1} and the G-band is 1573 cm^{-1} , the intensity ratio ($I_D/I_G = 0.67$). However, with PVA-G5 nanocomposite, the relative ($I_D/I_G = 0.82$) is higher than that of RGO, the enlargement in the ratio is ascribed to the development of new hydrogen bonds between rGO and PVP molecules [20]. The full width at half maximum (FWHM) of D- and G-bands in the rGO film are 97.2 cm^{-1} and 52.5 cm^{-1} , respectively.

However, in PVA-G5, these peaks' FWHM values increased to 102 cm^{-1} and 57.5 cm^{-1} , respectively. This broadening suggests a major rise in defect density inside the rGO structure, most likely produced by the addition of vacancies, distortions, and oxygen-containing functional groups. The Tuinstra-Koenig relationship was used to determine the mean size of neighboring carbons in the rGO and PVA-G5 nanocomposite [21, 22].

$$L_A = (2.4 \times 10^{-10}) \times (I_G/I_D) \times \lambda^4 \quad (1)$$

When comparing the pure rGO to

PVA-G5 samples, the L_A values showed a shifting trend from 22.76 nm to 22.30 nm , where λ is the laser wavelength and I_D and I_G are the Raman intensities at D and G peaks, respectively.

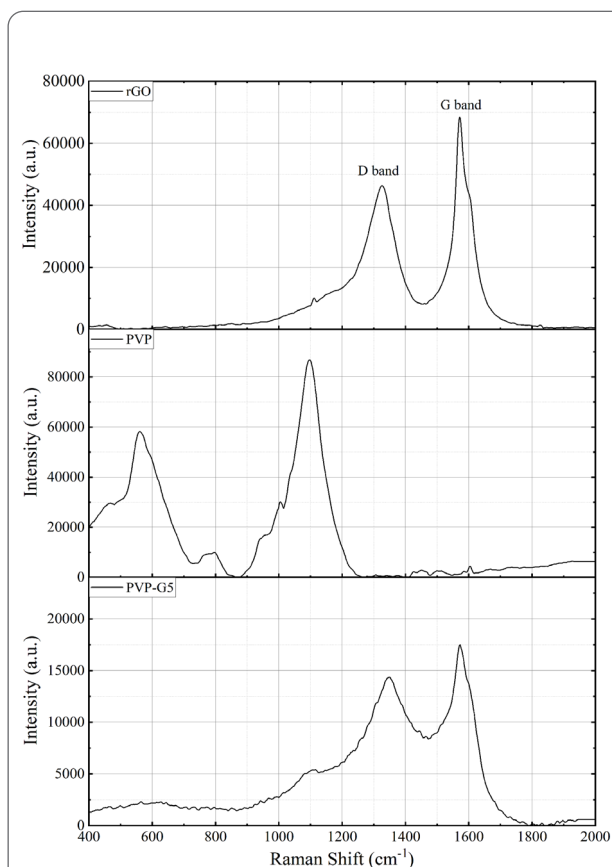


Fig.2 . Raman spectrum of rGO, PVP, and rGO-doped PVP, the excitation laser wavelengths are 633 nm, 532nm, and 532 nm, respectively.

Fig. 3 shows FTIR spectra for rGO, PVP, and PVP-G5 samples. A broad absorption peak observed between

3000 and 3500 cm^{-1} attributed to O-H symmetrical stretching. The wide peak seen at roughly 3430 cm^{-1} in pure PVP, corresponding to the O-H stretching vibration of the hydroxyl groups, moves to a lower wavenumber (around 3340 cm^{-1}) in the PVP-G5. This shift implies the establishment of hydrogen bonds between the hydroxyl groups of PVP and the oxygen-containing functional groups on the rGO surface. The sharp peak at roughly 1648 cm^{-1} in pure PVP, is caused by the C=O stretching vibration of the carbonyl group in the pyrrolidone ring [23].

The peak at 1648 cm^{-1} shifts to a lower wavenumber in the combination. This little change implies a probable interaction between the carbonyl group and the rGO surface. Peaks in PVP at 1272 cm^{-1} and 2935 cm^{-1} correspond to C-N stretching and C-H stretching vibrations, respectively [24]. The results indicate the strength of the interaction between the O-H group in PVP and the functional groups in rGO

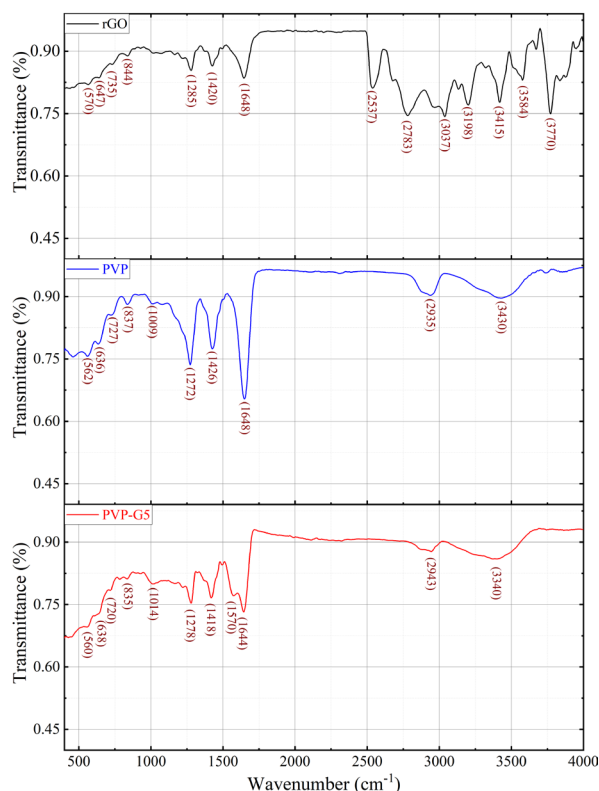


Fig. 3. FTIR spectrum of rGO, PVP, and rGO-doped PVP.

UV-vis spectroscopy was utilized to examine the optical characteristics of rGO, PVP, and rGO-doped PVP nanocomposite across the wavelength range of 300 to 800 nm. Fig. 4a depicts the absorption spectra of rGO, pure PVP, and five rGO-PVP samples with various rGO concentrations. At Ultraviolet absorption, the absorption is mostly due to $\pi-\pi^*$ electronic transitions inside the

molecules, these transitions take place between (C=C) groups and the carbon-yl groups (C=O) present in the samples [25]. PVP has the lowest absorption, rGO has the highest absorption, and rGO-doped PVP samples fall between them. The intensity of absorption increases with increasing concentration of rGO in PVP and the absorption increases due to the strength of the interaction between rGO and PVP.

Fig. 4b shows that PVP has the highest permeability and rGO the least transmission, and samples PVP-G1-PVP-G5 are located between them, where the transmission decreases with increasing rGO concentration in PVP. The absorption of the samples PVP-G1, PVP-G2, PVP-G3, PVP-G4, and PVP-G5 moved towards the highest wavelength (red shift) compared to rGO and PVP. The reason for this shift is the rGO concentration increases in the PVP matrix, where the sample concentration is directly proportional to its absorption at a specific wavelength according to the Lambert-Beer law [26]. This proves the occurrence of fusion between PVP and rGO as a result of the formation of new bonds. Consequently, there is a difference in the amount of

absorption for each sample depending on its concentration.

Figures 4c and 4d show the extinction coefficient and the refractive index, respectively. The extinction coefficient is the extinction that occurs for the electromagnetic wave inside the material, and it is the amount that the material's electrons absorb from the energy of the incident photons. On this basis, its value is determined by the interactions of the electromagnetic wave with the medium. It was observed that PVP has the lowest extinction coefficient and the highest extinction coefficient for rGO, and PVP-G1-PVP-G5 is located between them, where the extinction coefficient values rise with increasing concentration of rGO in PVP, meaning the absorption process increases with increasing concentration. More precisely, the higher the extinction coefficient, the greater the amount of light lost from the beam. The extinction coefficient serves to understand the properties of different materials and how they interact with light. The extinction coefficient can be calculated from the following equation [27]:

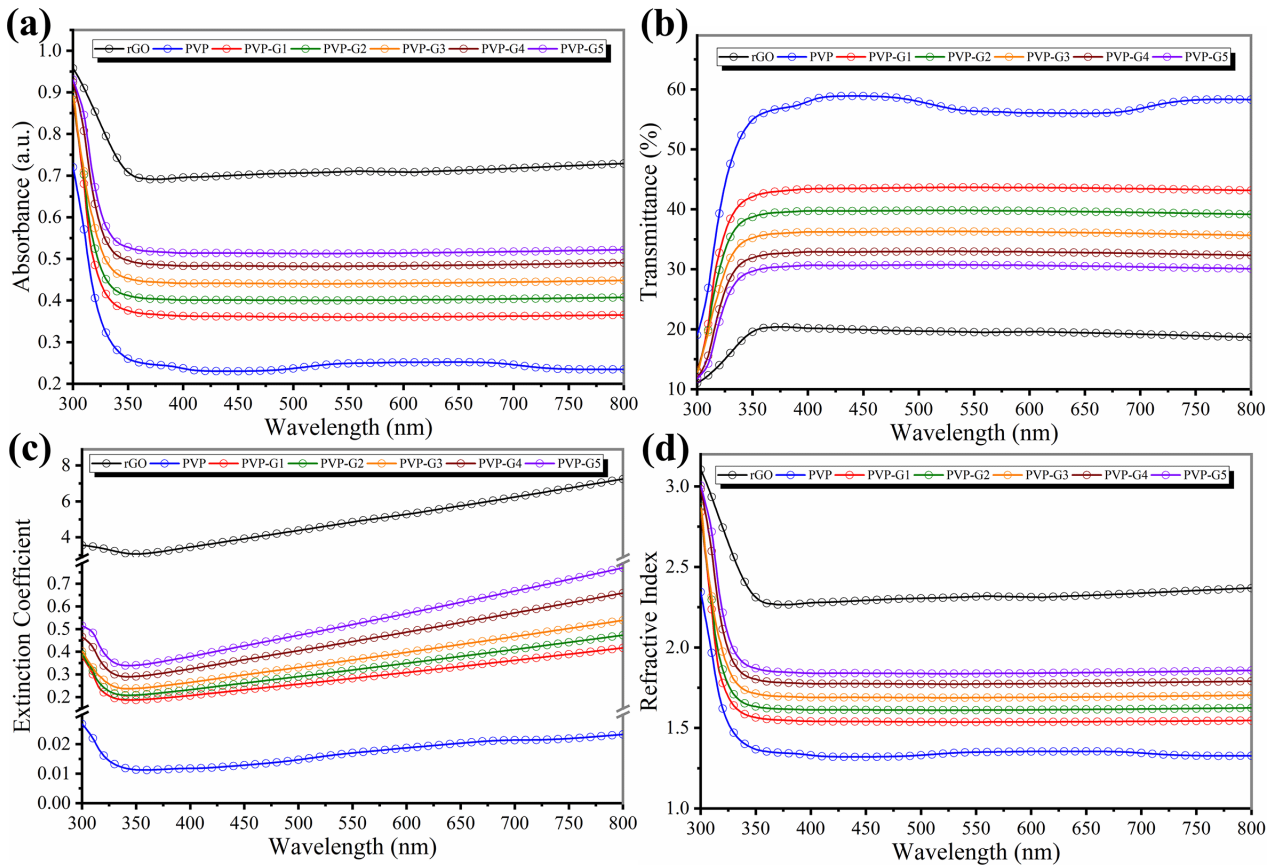


Fig. 4. Absorbance (a) and transmittance (b) spectrum for rGO, PVP, and PVP-G1-PVP-G5 nanocomposite samples, the extinction coefficient (c), and (d) the refractive index.

hence α is the absorption coefficient and λ is the wavelength.

$$k = \alpha \lambda / 4\pi \quad (2)$$

Fig. 4d shows the diagrammatic plots of the refractive index of PVP and PVP-G1-PVP-G5 as functions of wavelength, where it was observed that PVP has the lowest refractive index, while

rGO has the highest refractive index. As for the samples from PVP-G1-PVP-G5, they fall between them, as the refractive index increases with increasing rGO concentration. The refractive index of pure rGO, pure PVP, and rGO-doped PVP (PVP-G1-PVP-G5) was computed using the equation [28]:

$$n = (1 + R)/(1 - R) + \sqrt{4R/(1 - R)^2 - k^2} \quad (3)$$

hence, R is the reflectance.

Fig. 5 depicts the optical band gap (E_g) of PVP-G composites as determined by the Tauc plot method. There is a definite pattern of lowering E_g as rGO concentration increases. The calculated direct band gaps for the PVP-G samples vary from 3.82 to 3.51 eV. This decrease in E_g is attributable to the creation of new energy levels within the bandgap as a result of defects and chemical interactions between PVP and rGO [29]. These defect states enable improved electronic transmission, resulting in greater electrical conductivity in the composite material. The association between E_g and rGO concentration is inverse. The $\Delta E_g/E_g$ value decreases as rGO concentration increases at a decay constant of 2.77 ± 0.13 . This indicates that progressively introducing rGO into the PVP minimizes the energy gap. This shift in the energy gap is likely owing to changes in the material's electrical band structure caused by the presence of rGO. For a more precise view, by increasing the rGO concentration the $\Delta E_g/E_g$ variation presents a growth feature due to the neglecting of the PVP impacts on the rGO-doped PVP nanocomposites [27, 30].

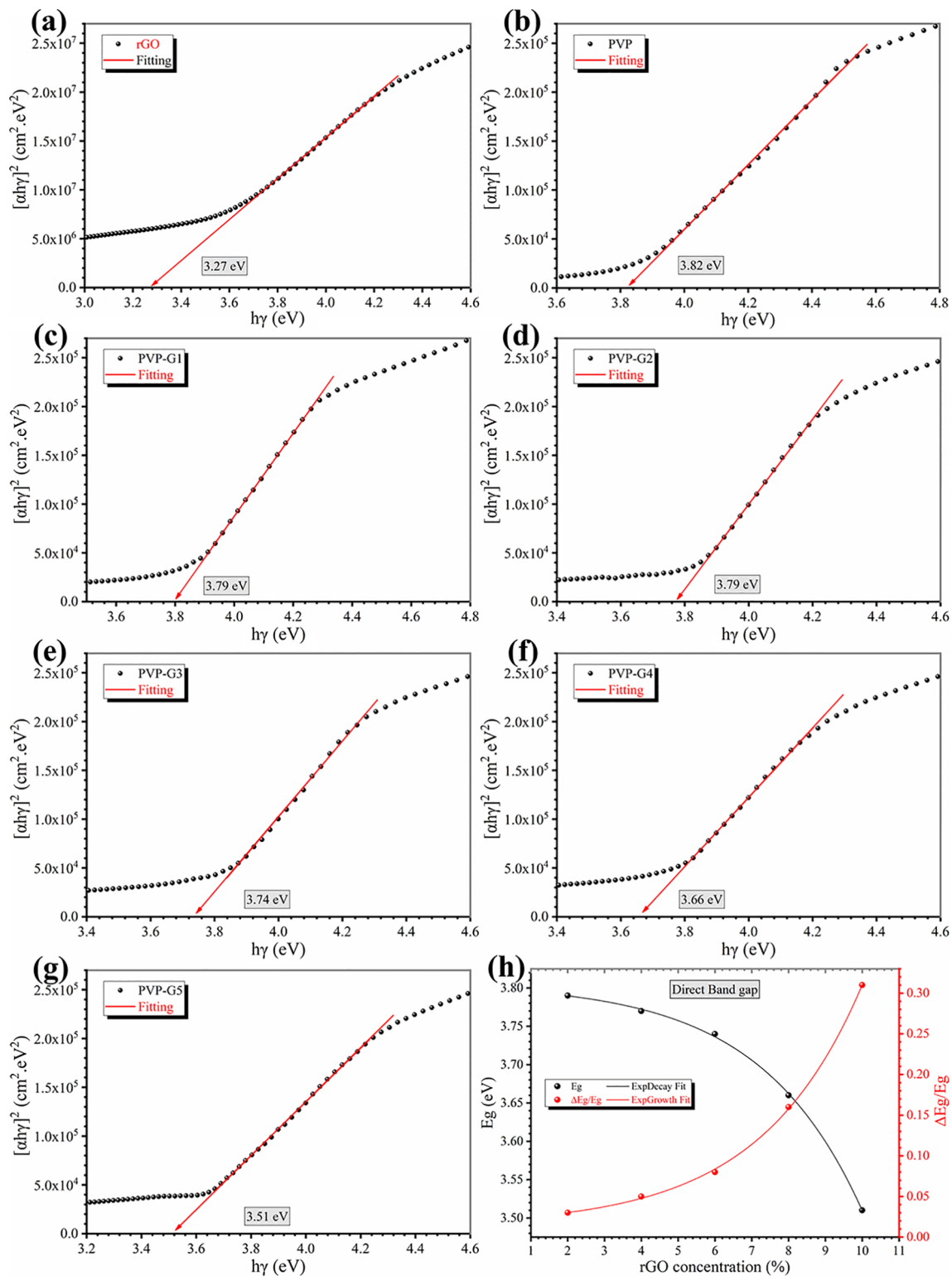


Fig. 5. Tauc plots of direct optical bandgap estimation, (a) rGO nanosheets, (b) PVP polymer, (c) PVP-G1 sample, (d) PVP-G2 sample, (e) PVP-G3 sample, (f) PVP-G4 sample, and (g) PVP-G5 sample; (h) linear relationship of E_g vs. rGO concentrations and plots of $\Delta E_g/E_g$ vs. rGO concentrations for the experimental results.

Figures 6a and 6b show the real (ϵ_r) and imaginary (ϵ_i) components of the dielectric constant for rGO, PVP, and rGO-PVP composites (PVP-G1 to PVP-G5) as a function of wavelength. It was revealed that the rGO-PVP samples (PVP-G1 to PVP-G5) exhibit low dielectric constants at short wavelengths (300-400 nm), moderate dielectric constants at medium wavelengths (300-800 nm), and increasing dielectric

constants at long wavelengths. These findings show that rGO-PVP samples can be used in applications requiring varying dielectric constants at various wavelengths.

The following relations were used to calculate ϵ_r and ϵ_i from the computed values of n and k [27]:

$$\epsilon_r = n^2 - k^2 \quad (4)$$

$$\epsilon_i = 2nk \quad (5)$$

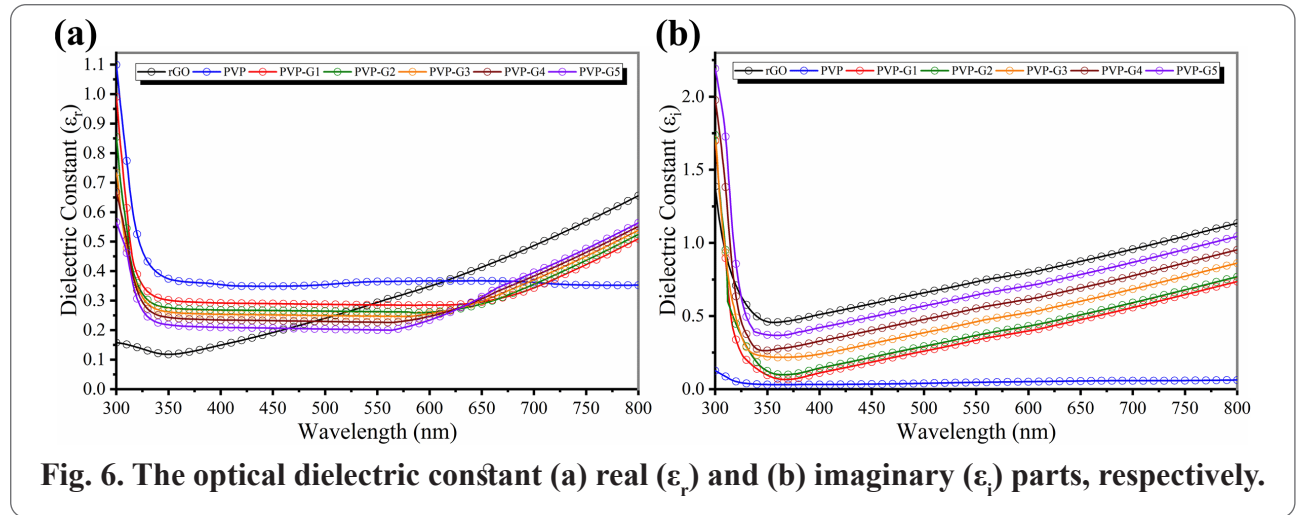


Fig. 6. The optical dielectric constant (a) real (ϵ_r) and (b) imaginary (ϵ_i) parts, respectively.

Fig. 7a demonstrates optical conductivity, the samples are identified as rGO, PVP, PVP-G1, PVP-G2, PVP-G3, PVP-G4, and PVP-G5. The optical conductivity of PVP rises dramatically as the concentration of rGO increases. At shorter wavelengths, the rise in optical conductivity is more apparent, demonstrating a high dependence on rGO concentration. At medium and longer wavelengths, the optical con-

ductivity stabilizes and exhibits a more linear behavior with minor fluctuations depending on the concentration of rGO in the PVP matrix. The optical conductivity of the produced samples was examined using the formula [27]:

$$\sigma = \frac{n\alpha c}{4\pi} \quad (6)$$

hence, σ and c are the optical conductivity and speed of light, respectively.

Fig. 7b shows the relationship between loss tangent ($\tan \delta$) and wavelength (nm) for several different samples labeled as rGO, PVP, P-G1, P-G2, P-G3, P-G4, and P-G5. At short wavelengths (300–400 nm), all samples show high values of loss tangent. rGO shows the highest loss tangent value at short wavelengths. PVP and (P-G1 to P-G5) show lower values of rGO, with a gradual increase in loss with increasing rGO concentration. At intermediate wavelengths (400–600 nm), the loss tangent begins to decrease gradually for most samples. At long wavelengths (600–800 nm), most samples show stability in loss tangent values, with some convergence between samples.

The optical loss tangent epitomizes the ability of the material to dissipate electromagnetic energy. Higher values

of the loss tangent at short wavelengths indicate greater energy dissipation, which is clearly observed in samples with higher rGO concentrations. As the wavelength increases, the loss tangent gradually decreases, which means that the samples become more efficient in reducing energy dissipation. This effect is more pronounced in samples containing higher rGO concentrations. At higher wavelengths, the loss tangent is stabilized due to a slight effect of rGO, and the differences between loss tangent values in the samples become negligible. The stability in this region could be valuable in applications preferring limited power loss within a wide range of wavelengths. The following equation was utilized to gauge the loss tangent ($\tan \delta$) [31]:

$$\tan \delta = \varepsilon_i / \varepsilon_r \quad (6)$$

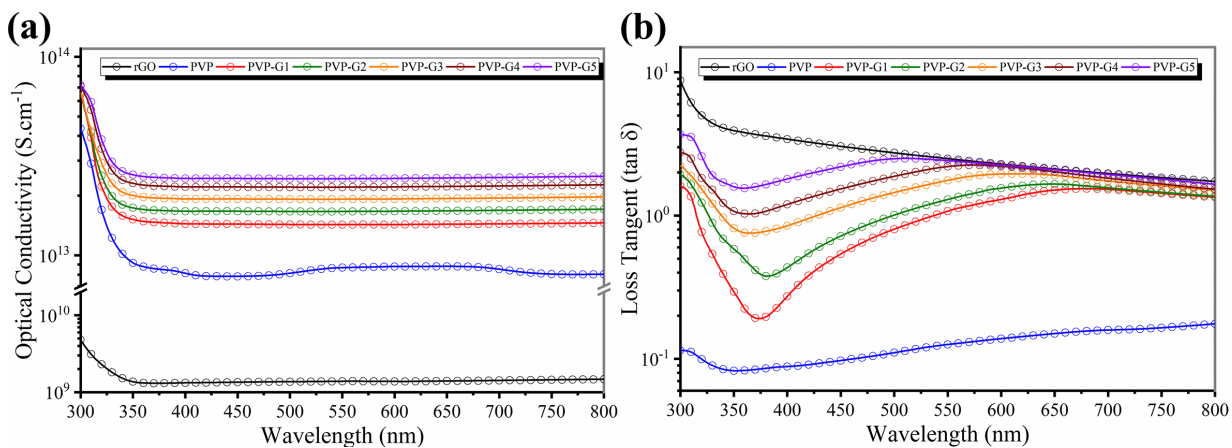


Fig. 7. (a) the optical conductivity of the prepared samples, and (b) the loss tangent as a function of wavelength.

The most important results obtained in this research are a gradual increase in absorption and a gradual decrease in transmittance of samples (PVP-G1 to PVP-G5) in the ultraviolet region due to the rGO concentration levels, as these nanocomposites showed a greater amount of absorption than the PVP and less transmittance for nanocomposites compared to PVP alone, as these two properties can be used in solar cell applications as well as in reducing the risks of ultraviolet radiation. Also, a decrease in the energy gap of the polymer was observed with an increase in the concentration of rGO, and this property can be used in solar cell and supercapacitor applications. Also, the results showed a gradual increase in the extinction coefficient of the nanocomposites with an increase in the concentration of rGO, meaning that the nanocomposites absorb most of the light passing through them. Therefore, these results enhance the performance of solar cells. Also, the refractive index of the nanocomposites was observed to increase gradually with an increase in the concentration of rGO, as this property can be used in optical fibers to control the light path. The results also

showed an increase in the optical conductivity of the nanocomposites with an increase in the concentration of rGO, in contrast to the conductivity being optically lower for rGO and PVP. This result is important in anti-corrosion coating for oil transformation pipe, and oil recovery and UV protection applications because these nanocomposites have the ability to absorb UV radiation and transfer energy well. The dielectric constant of pure PVP is lower, and when rGO is added to it, the dielectric properties of the nanocomposite can be improved, and this feature can be used in applications of supercapacitors and optoelectronic devices.

Table 1 represents the best values of optical parameters for each prepared sample, and shows that higher values of extinction coefficient are likely due to increased interactions with light, which may contribute to a reduction in the energy gap and improved electrical conductivity. Increasing the values of refractive index gradually from 1.34 to 1.83 shows that the material is becoming more able to refract light by increasing the rGO nanosheets, which is good for applications such as optical lenses. The results also showed a

decrease in the optical bandgap from 3.97 to 3.51, indicating improved electronic conductivity and increased light absorption in the materials.

Table 1. The optical calculated parameters of all the prepared samples (extinction coefficient, the refractive index, direct optical bandgap, optical dielectric constant real and imaginary, optical conductivity, and the loss tangent).

Samples	k	n	E_g	ϵ_r	ϵ_i	σ	Tan δ
PVP	0.10	1.34	3.82	0.36	0.04	8.41×10^{12}	0.07
rGO	2.98	2.26	3.27	0.10	0.45	1.22×10^9	3.84
PVP-G1	0.17	1.54	3.97	0.30	0.06	1.44×10^{13}	0.18
PVP-G2	0.21	1.62	3.97	0.27	0.11	1.69×10^{13}	0.37
PVP-G3	0.23	1.70	3.94	0.26	0.20	1.94×10^{13}	0.75
PVP-G4	0.28	1.78	3.66	0.24	0.26	2.23×10^{13}	0.98
PVP-G5	0.33	1.83	3.51	0.20	0.35	2.49×10^{13}	1.49

4. Conclusion

In this research, the rGO-doped PVP nanocomposites are manufactured and evaluated successfully, these nanocomposites could be a good candidate for next-generation optoelectronics devices. The rGO additive outstandingly improves the optical characteristics of the PVP matrix. The UV-vis, Raman, and FTIR spectroscopy investigations verified that the optical features of the rGO-doped PVP nanocomposites were significantly enhanced as a result of the rGO injection into PVP. The increasing rGO concentration formulates a narrower optical bandgap, this

indicates an improvement in electronic transmission and better interaction between the components. These findings suggest that rGO-PVP nanocomposites are an interesting potential candidate for flexible, smart, and even next-generation optoelectronics applications. The results emphasize to development of more efficient, environmental, and low-cost materials for diverse optical applications. Overall, this work offers new promises for material science and the creation of novel, high-performance nanocomposite materials can be involved in cost reduced advanced industrial technologies such as anti-cor-

rosion coating and optoelectronic devices.

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