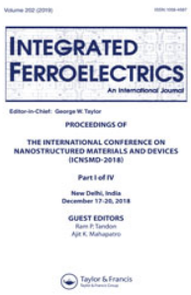

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# Electrical Properties of Self Sustained Layer of Graphene Oxide and Polyvinylpyriodine Composite

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**ABSTRACT**

A paper like self-sustained layer composed of graphene oxide (GO) and polyvinylpyrrlolidone (PVP) has been prepared by following the simple one pot chemical route. Further characterisation of the resulting layer pursued using various optical and analytical tools confirm the formation of GO-PVP composite. The appearance of C-N and C = O peaks in Fourier transform infra-red (FTIR) spectra, and increase in $I_D/I_G$ ratio from 0.92 to 0.96 in Raman spectra, after PVP addition to the GO, indicate the attachment of PVP with GO and formation of GO-PVP composite. GO-PVP paper strip of size 1cm$^2$ is silver pasted on ITO substrate and top aluminium (Al) contact is deposited using thermal deposition through shadow masking for fabrication of Al/GO-PVP/ITO device structure. The current-voltage characteristics shows decrease in electronic current through GO-PVP thin film compared to GO thin film and is attributed to the reduction in electrical conductance with inclusion of insulating polymer PVP.

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**KEYWORDS**
Graphene oxide (GO); polyvinylpyridone (PVP); Fourier transform infra-red (FTIR)

## 1. Introduction

Graphene oxide (GO) has attracted much attention due to their exceptional technological application and it consists of oxygen containing functional groups due to which it is hydrophilic in nature. This gives easily dispersion of GO in water and other organic solvents and thin films of GO can be prepared easily by spin coating, drop casting etc. The oxygen containing functional groups of GO serves as sites for chemical modification and functionalization of GO and chemical composition of GO can be chemically, thermally and electrochemically engineered [1, 2]. GO can be produced from oxidation of graphite by using Hummer's method and due to its tunability in chemical composition it has attracted application in electronics [3] and sensors and biosensors [4]. The nanocomposite of polymer and hetrojunction materials has gained interest to realise memory effect [5–9]. The use of nanocomposite of polymers for sustainable development and environmental preservation has become important because of their low cost, easy processibility, low weight, high quality surface and easy fabrication of thin and thick films [4–8].

Polyvinylpyrlolidone (PVP) has attracted more interest due to its chemical stability, excellent optical, mechanical, and electrical properties, easy process and excellent

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transparency, non-toxicity and its solubility in polar solvents. PVP a synthetic polymer that has well defined structure with an N–vinylpyrrlolidone monomer connected to long chain and can be used as copolymer in other polymer to form new nanocomposite films [5, 6]. The polymer based nanocomposite of graphene and GO are quite interesting due to its improvement in combined properties of nanocomposite.

This paper presents the optical and electrical properties of GO-PVP complex prepared by chemical route. Fourier transform infra-red (FTIR) confirms the successful formation of GO-PVP complex and scanning electron microscope (SEM) indicates the regular and transparent flakes. Increase in $I_D/I_G$ ratio in Raman spectra and rapid loss of masses in GO-PVP compared to GO observed in thermo gravimetric analyser (TGA) and decrease in electronic current of Al/GO-PVP/ITO as compared to Al/GO/ITO device structure is observed in current –voltage (I-V) characteristics.

## 2. Experimental Details

### 2.1. Synthesis of GO-PVP

GO is synthesised by using modified Hummers method [10, 11]. PVP powder (10 mg) purchased from sigma Aldrich, USA, was dispersed in 150 ml of deionised (DI) water and sonicated for 30 min. GO solution was prepared by adding 30 mg of GO in 10 ml of DI water. Both of these solutions were mixed and sonicated for next 30 min to get uniform dispersion. This homogeneous solution was filtered by milipore filter paper (Whatman grade 1), followed by washing and drying in petri-disc at room temperature. The schematic for GO-PVP composite is shown in Figure 1. A self-sustained paper like film was formed and was pasted on ITO substrate (GO-PVP/ITO) using silver pasted and top Al contacts are prepared by thermal deposition technique through shadow masking to form Al/GO-PVP/ITO device structure for further electrical measurements.

### 2.2. Characterisation Technique

The paper like materials is characterised using meteorological and optical tools. This includes the structural and morphological study using scanning electron microscopy (SEM, ZEISS instrument), optical study using the Raman (RENISHAW, Invia microscope), and Fourier Transform infra-red (FTIR, Perkin Elmer FTIR spectrometer). In addition, the chemical stability and chemical interaction of organic polymer PVP with GO are analysed through thermo

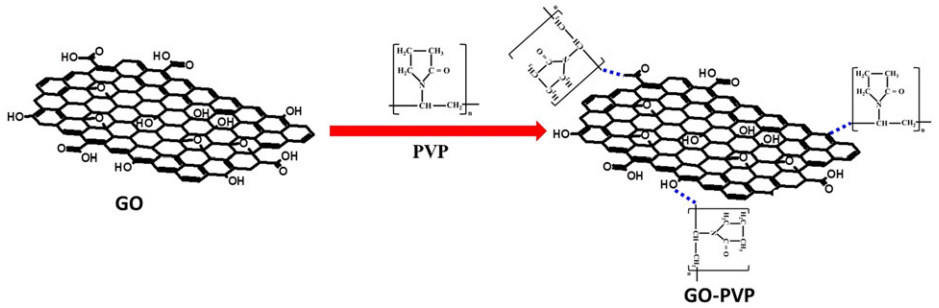


Figure 1. Schematic diagram of GO and GO-PVP complexes.

gravimetric analysis (TGA) measurements. The electronic properties are measured by recording current-voltage characteristics using Keithley-4200 semiconductor analyser.

## 3. Spectroscopic Properties of GO-PVP

### 3.1. FTIR Spectra

FTIR studies of solutions containing GO sheets, PVP molecules, and GO-PVP complexes with DI water as solvent, are recorded and shown in Figure 2. The appearances of peaks at 3323 $cm^{-1}$ corresponds to O-H stretching vibration and at 1375 $cm^{-1}$ due to O-H deformation due to C-OH groups of GO sheets. The peak at 1265 and 1065 $cm^{-1}$ is attributed to the stretching vibrations of C-O (stretching vibration of C-OH) and C-O (stretching vibration of epoxide), and peak at 1603 $cm^{-1}$ represents the C = O (carboxyl or carbonyl vibrations) of GO [10–13]. The PVP shows peaks appear at 2918 and 1474 $cm^{-1}$, corresponding to the C-H bending vibration in aliphatic compound and C-H stretching in methylene group, respectively, and broad peak at 3496 $cm^{-1}$ may attributed to the water that PVP absorbed. The absorption band found at 1652, and 1282 $cm^{-1}$ in PVP are associated with stretching vibration of the carbonyl group (C = O), and pyrrolidone structure (C-N), respectively [13]. The C-O and C-OH peaks are diminished after PVP attachment. The GO-PVP composite shows appearance of C-N and C = O peaks, which indicate the attachment of PVP with GO.

### 3.2. Raman Spectra

Raman spectra of GO and GO-PVP composites show characteristics peaks of GO at 1200 and 1500 $cm^{-1}$ corresponding to D and G bands (Figure 3), respectively. The intensity

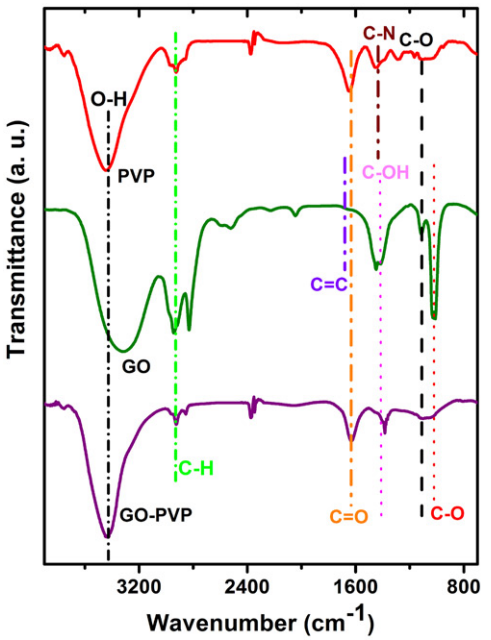


Figure 2. FTIR spectra of GO, PVP, and GO-PVP nanocomposites.

ratio ($I_D/I_G$) is calculated by dividing the intensity of $I_D$ peak to $I_G$ peak and are observed to be 0.92 and 0.96 for GO and GO-PVP composites, respectively. Here, the increase in $I_D/I_G$ ratio after PVP attachment is attributed to the generation of more defect sites with removal of C-O and C-OH functional groups (as FTIR supports) in the GO sheets.

### 3.3. SEM of GO-PVP Complex

The SEM image in Figure 4 indicates the presence of flake like structures in both GO (Figure 4a) and GO-PVP (Figure 4b) complexes. SEM of GO powder represents flakes

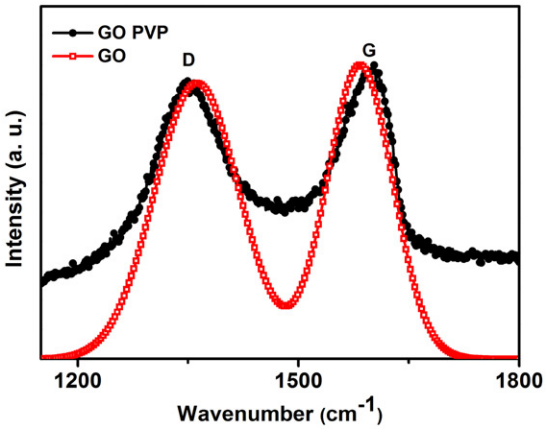


**Figure 3.** Raman spectra of GO (open circle) and GO-PVP (filled circle) composites.

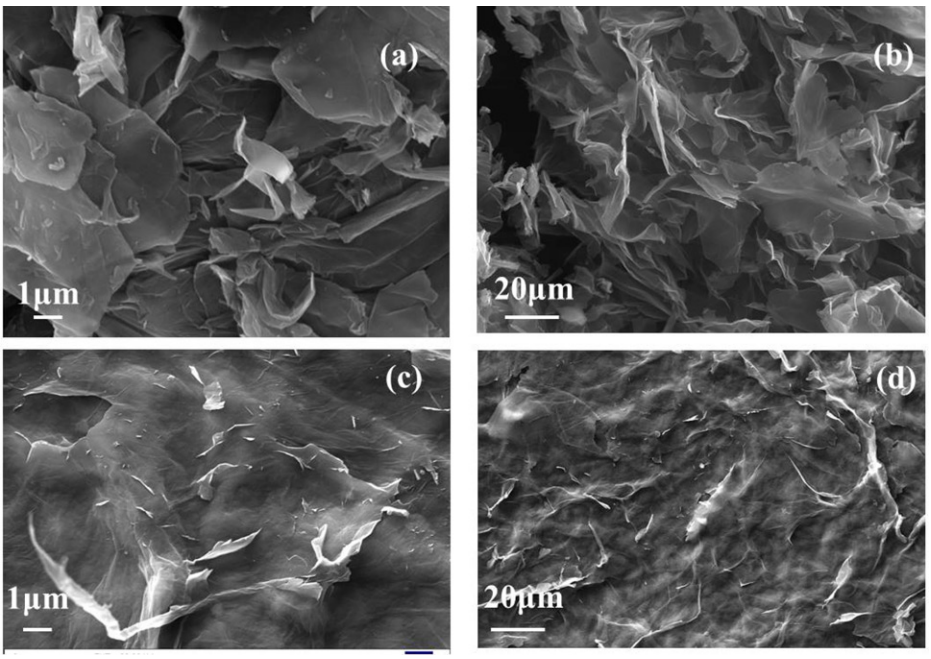


**Figure 4.** SEM images of (a, b) GO and (c, d) GO-PVP composites in 1 $\mu$m and 20 $\mu$m scale.

of sizes 5-20 µm with appearance of wrinkle like patterns. In contrast, the layer of GO-PVP complex (Figure 4c and d) shows arrangement of GO flakes with densely packed and settled in relatively planar arrangement of GO and helps distributing the flakes randomly and uniformly in the entire volume.

### 3.4. TGA of GO-PVP

The TGA of GO and GO-PVP (Figure 5) records major weight loss at 100 °C. The GO powder shows, initially, a 10% weight loss below 100 °C due to removal of hydroxyl groups. The 40% weight loss during increase in temperature from 100 to 150 °C is due to removal of oxygen containing functional groups. The 10% weight loss occurs from 150 to 200 °C [11, 12]. After 200 °C, the weight loss is very slow. GO-PVP nanocomposite shows ~8% weight loss below 100 °C due to the removal of adsorbed water. The 40% weight loss during 150 to 200 °C should be ascribed as the removal of oxygen containing functional groups. The trends of weight loss curve for GO-PVP is shifted towards higher temperature and very fast compared to the GO, indicates more loss of masses.

## 4. I-V Characteristics of GO and GO-PVP Based Thin Film Devices

Figure 6 represents the I-V characteristics of Al/GO/ITO and Al/GO-PVP/ITO vertical device structures, as schematised in inset (e). Here, both device structures show linear behaviour at low bias during $0 \rightarrow +8V$ and $0 \rightarrow -8V$. The ln(I) versus ln(V) plot of GO (shown in inset (a, b) of Figure 6) and GO-PVP (shown in inset (c, d) of Figure 6) complexes indicate a slope of 1 [11–13]. The current levels through GO-PVP and GO thin films are observed to be $5 \times 10^{-8}$ A and $8 \times 10^{-2}$ A, respectively at low bias. The increase in resistance values from $1 \times 10^{3}$ to $2 \times 10^{9}$ Ω after mixing of PVP in GO is attributed to the reduction in overall electrical conductance in presence of insulating polymer PVP.

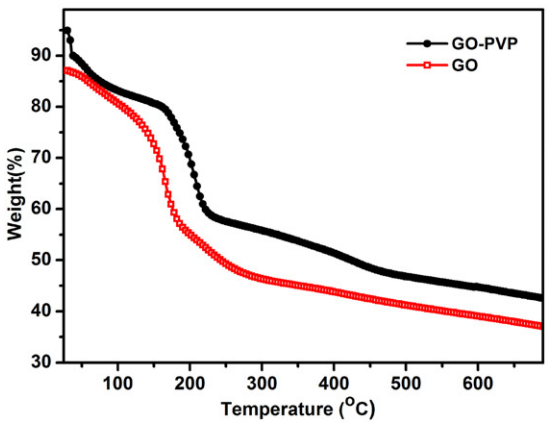


**Figure 5.** TGA of GO (open square) and GO-PVP (dark circle) composites.

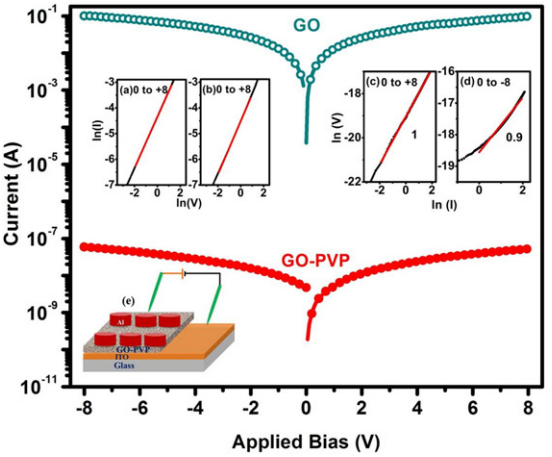


**Figure 6.** I-V characteristics of GO (open circle) and GO-PVP (dark circle) composite thin films based Al/GO/ITO and Al/GO-PVP/ITO device structures. (a, b) ln(I) and ln(V) plot of GO, (c, d) ln(I) and ln(V) plot of GO-PVP, and (e) schematic diagram of GO-PVP thin films.

## 5. Conclusion

Self-sustained layers of GO-PVP composite were prepared by following chemical route. FTIR spectra indicates the appearance of C-N and C = O in GO-PVP composite, suggesting chemical attachment of PVP with GO layer. SEM imaging shows arrangement of densely packed GO-PVP compared to GO flakes. Raman spectra indicate increase in $I_D/I_G$ ratio after PVP attachments to the carbon backbone of GO that generates more defect sites. The decrease in values of current in I-V characteristics through Al/GO-PVP/ITO compared to Al/GO/ITO device structures is atributed to the inclusion of insulating polymer PVP with GO layer. Owing to the good bio-compatibility and non-toxic behaviour of PVP, the currently developed GO-PVP complexes could be further tested for translating their interesting behaviours into bio-sensing applications and further fabrication of medical devices for detection of bio-entities

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## References

1. S. Stankovich *et al.*, Synthesis of graphene-based nanosheets via chemical reduction of exfoliated graphite oxide, *Carbon*, **45**, 1558 (2007). DOI: 10.1016/j.carbon.2007.02.034.
2. R. K. Singh, R. Kumar, and D. P. Singh, Graphene oxide: Strategies for synthesis, reduction and frontier applications, *RSC Adv.* **6** (69), 64993 (2016). DOI: 10.1039/C6RA07626B.
3. P. Saini *et al.*, Probing the switching mechanism in annealed thin films of graphene oxide, *ACS Appl. Mater. Interfaces* **10** (7), 6521 (2018). DOI: 10.1021/acsami.7b09447.

4. S. K. Bhardwaj *et al.*, Biosensing test-bed using electrochemically deposited reduced graphene oxide, *ACS Appl. Mater. Interfaces* **8** (37), 24350 (2016). DOI: 10.1021/acsami.6b04562.
5. G. M. Moazzami, and F. Sharif, Enhancement of dispersion and bonding of graphene-polymer through wet transfer of functionalized graphene oxide, *Express Polym. Lett.* **6**, 1017 (2012). DOI: 10.3144/expresspolymlett.2012.107.
6. J. Liu *et al.*, Preparation of $MoS_2$ –polyvinylpyrrolidone nanocomposites for flexible nonvolatile rewritable memory devices with reduced graphene oxide electrodes, *Small* **8**, 3517 (2012).
7. Y. Zhang *et al.*, Interactions of graphene and graphene oxide with proteins and peptides, *Nanotechnol. Rev.* **2**, 27 (2013).
8. S. S. Nanda, G. C. Papaefthymiou, and D. K. Yi, Functionalization of graphene oxide and its biomedical applications, *Crit. Rev. Solid State Mater. Sci.* **40** (5), 291 (2015). DOI: 10.1080/10408436.2014.1002604.
9. B. Paulchamy, G. Arthi, and B. D. Lignesh, A simple approach to stepwise synthesis of graphene oxide nanomaterial, *J. Nanomed Nanotechnol.* **6**, 253 (2015).
10. M. Singh *et al.*, Tuneable physicochemical properties of thermally annealed graphene oxide powder and thin films. *J. Nanosci. Nanotechnol.* **18** (3), 1763 (2018). DOI: 10.1166/jnn.2018.14319.
11. P. Saini *et al.*, Spectroscopic and electronic properties of polyallylamine functionalized graphene oxide films, *Vacuum* **154**, 110 (2018). DOI: 10.1016/j.vacuum.2018.04.056.
12. P. Saini *et al.*, Functionalization of polyallylamine with graphene oxide surface, *Appl. Mater. Proc.* **2** (3), 209 (2017). DOI: 10.5185/amp.2016/3015.
13. P. Saini *et al.*, Temperature dependent electronic properties in thin films of graphene oxide. *Integrated Ferroelectric* **184** (1), 210 (2017). DOI: 10.1080/10584587.2017.1368665.