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# Enhancement in performance of polycarbazole-graphene nanocomposite Schottky diode

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We report formation of polycarbazole (PCz)–graphene nanocomposite over indium tin oxide (ITO) coated glass substrate using electrochemical technique for fabrication of high performance Schottky diodes. The synthesized nanocomposite is characterized before fabrication of devices for confirmation of uniform distribution of graphene nanosheets in the polymer matrix. Pure PCz and PCz-graphene nanocomposites based Schottky diodes are fabricated of configuration Al/PCz/ITO and Al/PCz-graphene nanocomposite/ITO, respectively. The current density–voltage (J-V) characteristics and diode performance parameters (such as the ideality factor, barrier height, and reverse saturation current density) are compared under ambient condition. Al/PCz-graphene nanocomposite/ITO device exhibits better ideality factor in comparison to the device formed using pure PCz. It is also observed that the Al/PCz-graphene nanocomposite/ITO device shows large forward current density and low turn on voltage in comparison to Al/PCz/ITO device.  [http://dx.doi.org/10.1063/1.4860952]

## I. INTRODUCTION

Tunable functionality, flexibility, low cast, easy processibility and synthesis of conducting polymer, mainly conjugated polymer,[1–3] over inorganic semiconductors are attraction for researchers for fabrications of electronic device.[4–7] These features of organic conductors have opened a new possibility of replacing conventional expensive inorganic devices. Organic material based field effect transistors (OFET), solar cell, light emitting diode (OLED), Schottky diode, sensor etc are the most important electronic devices for real life applications. These devices have already been developed by researchers in the recent years mainly using solution proccessible conducting polymers or organic conductors.[8–12]

The most studied conducting polymers for above mentioned devices are polyaniline, polypyrrole, polyindole and polythiophenes.[13–16] However, polycarbazole (PCz) [Figure 1(A) and 1(B)] is a class of polymeric material which has recently drawn much more attention due to its application in development of thermal and environmentally stable devices.[17–19] In addition to its advantages as low cast, easy processible and easy synthesis in comparison to other conducting polymer.[5,19–21] However, the main concern of PCz is same as other conducting polymers viz. poor conductivity, low mobility, poor stability and mechanical properties in comparison to inorganic semiconductor materials, which restricts their future applications. To overcome mecahanical properties and stability problem, researchers have used insulating materials.[22] The use of such material which enhances the performance of poor semiconducting polymer devices as well as stability and mechanical property just by presence of smaller amount without changing its property. This concept motivated the researchers to use composite material which consist of a high conducting material as filler.[23–25] There

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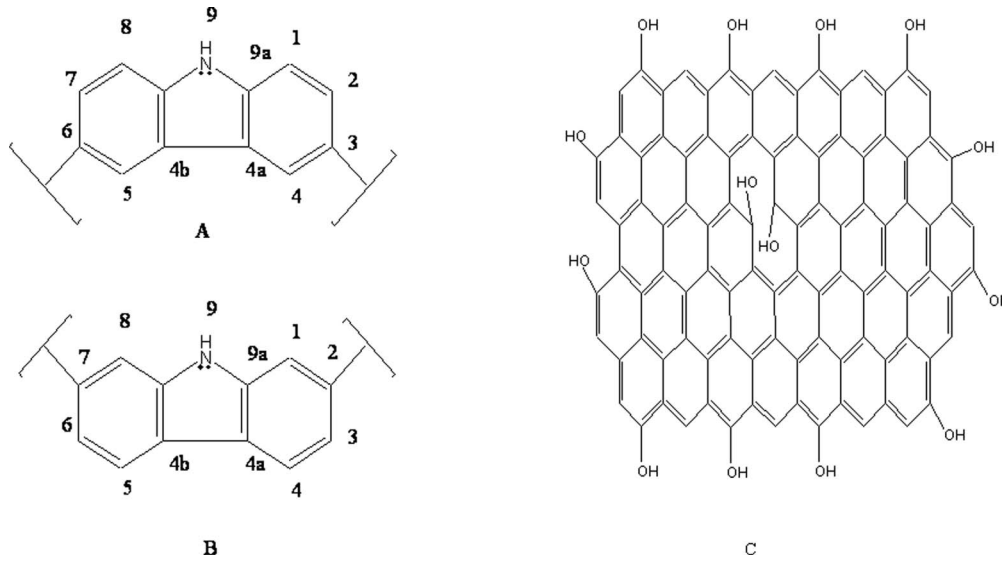


FIG. 1. Structural formula of Polycarbazole (A) Polymerization at 3, 6 position and (B) 2,7 positions. (C) Structural formula of functionalized graphene sheet.

are mainly three nano carbon materials are available viz. fullerene (zero dimension) carbon nanotube (CNT) (one dimensional) and graphene (two dimensional) which have higher conductivity in comparison to not only PCz but also the other conductor polymers. Among these nanomaterials graphene (Figure 1(B)) has attracted tremendous interest as a novel electronic material with various potential applications. Graphene, single to a few atomic planes of graphite, have unique electronic, optical and mechanical properties, including the exceptionally high carrier mobility at room temperature.[26,27] However, interaction of nanofillers with polymer matrixes and uniform distribution of nanofillers are major issues in development of reproducible and stable devices.

In recent years, Schottky diodes using conducting polymer nanocomposites with nano fillers as graphene, carbon nanotube, nanoclay and metal nanoparticles have been developed mainly using chemically synthesized polymers.[28–32] These hybrid structures have emerged as excellent candidates for the fabrication of a variety of electronic and optoelectronics devices.[33,34] However, uniformity of composites and processibility are the major challenges. It has also been suggested that uniformly distributed conducting nanofiller in the polymer acts as nanometric heat sinks, preventing the build-up of large thermal effects and thus reducing material and device degradation.

In view of above we explored electrochemical synthesis of conducting polymer-nanofiller composite and chosen PCz-graphene system as it is not developed and characterized yet to the best of our knowledge. Our technique not only produces uniform composite but also gives an easy method for direct deposition of compact composite film over the substrates for fabrication of devices. We directly deposited active semiconducting composite layer by electrochemical method for fabrication of Schottky diode with a configuration of Al/PCz-graphene nanocomposite/Indium tin oxide coated glass (ITO). PCz-graphene devices showed excellent rectifying behavior and lower turn on voltage in comparison to pure PCz. The composite layer is characterized by SEM, UV-vis and Fourier Transform-Infrared (FT-IR) studies before fabrication of Schottky diode.

## II. EXPERIMENTAL PROCEDURE

The synthesis of functionalized graphene by using methanol were described somewhere else.[35] Methanol derive functionalized graphene has been taken as filler nanomaterial in PCz matix. 0.1 mg of graphene was dispersed in 2 ml dichloromethane ($CH_2Cl_2$) and sonicated for 15 minute for making good suspension. In order to get uniform and stable suspension (at least for 30 minutes) we removed the precipitated graphene and used suspended graphene solution only for further

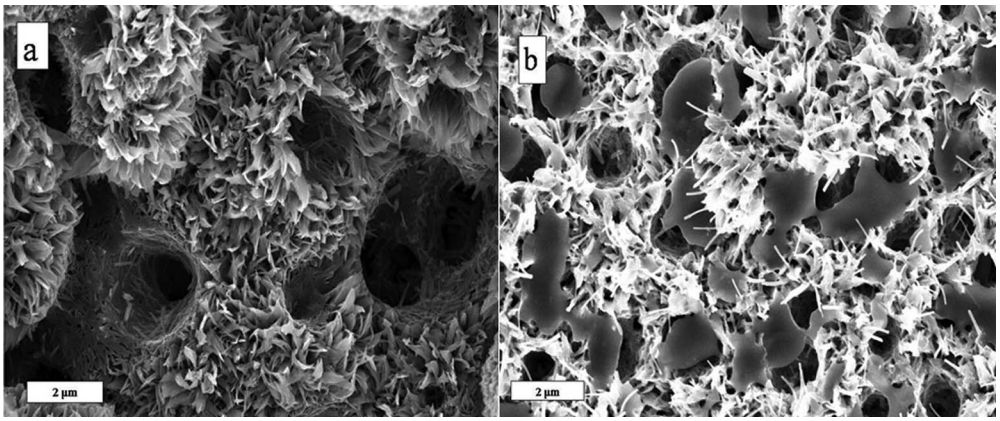


FIG. 2. Scanning electron microscopy of electropolymerized (a) PCz and (b) PCz/graphene nanocomposite film over ITO substrate.

experiments. In the stable suspension, 60 mM carbazole and 0.1 M tetrabutyl ammonium perchlorate (TBAP) (both purchased from Aldrich, Germany) were added and mixed properly. The PCz-graphene composite films were electrochemically polymerized under ambient condition by same method as polycarbazole was formed in our previous paper.[34] The electropolymerized samples were dip washed in $CH_2Cl_2$ to remove the carbazole monomers and oligomers from the surface of film before further characterizations. The thickness of polymer and composites film was of the order of $850 \pm 30$ nm as estimated from atomic force microscope AFM measurement (NT MTD, Russia). The synthesized samples were characterized by UV-vis spectroscopy using Perkin Elmer Lamda-25 model, Germany. The structural analysis was done by using Fourier Transform-Infrared (FT-IR) spectroscopy by using Nicolet Thermo Scientific 6700, Germany. The formation of nanocomposite was confirmed from Scanning electron microscope (SEM) model GEMINI Zeiss (SUPER TM 40) micrographs.The sandwich cells of Al/PCz-graphene composite/ITO and Al/PCz/ITO were fabricated by thermal vacuum evaporation (model: HIND HIVAC model 12A4D) method by depositing 1 mm diameter circular dot (area .785 $mm^2$). The thickness of metals on top of the polymer/composite film was pre-adjusted to 60 nm in each case. Current density-voltage (J-V) measurements of devices Al/PCz-graphene composite/ITO and ITO/PCz/Al structure were carried out with source meter of Keithley (model 2612A) at room temperature (27 °C) in air under dark condition.

## III. RESULTS AND DISCUSSIONS

The morphology of electrochemically synthesized sample of pure polymer (PCz) and its nanocomposite (PCz-graphene) film over ITO were studied using SEM and images are shown in Fig. 2. There are clear visible differences in morphology of pure PCz and its nanocomposite (PCz-graphene) films. SEM images confirm the uniform distribution of graphene sheets in the flower globular like morphology of PCz matrixes. In addition, it was also observed that the cavities (spaces) between the globules of PCz were completely filled with the graphene sheets and a dense film was formed in case of composites. UV-vis spectra of pure PCz and PCz-graphene nanocomposite films were shown in Figure 3. Typical absorption in pure PCz film was observed at 336 nm, 377 nm and 656 nm. Each monomer in PCz consists of $\pi$ orbital of nitrogen hetero atoms, which have already consisted a non bonding orbital. For such a monomer molecule, lowest energy transition must be n-$\pi$* while generation of defect during electropolymerization over polymer chains produce polaron which generates a site for lowest energy transitions.[36,37] The spectrum of electropolymerized film showed the absorption peaks at 336 nm ($\pi$-$\pi$*) and at 377 nm (n-$\pi$*). The 656 nm peak can be assigned as polaronic excitation peak which lie between lowest absorption sides. The absorption spectra of PCz/Graphene nanocomposite film show the different nature in comparison to pure polymer. Only n to $\pi$* transition was present in composite film while $\pi$-$\pi$* transition was

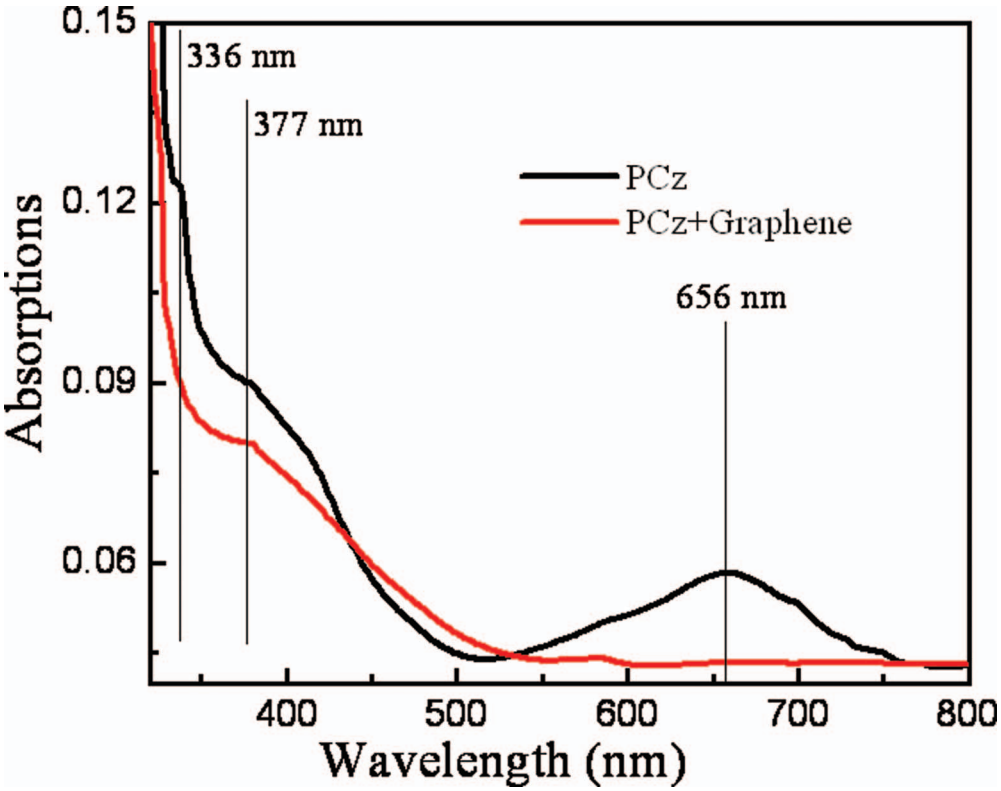


FIG. 3. UV-vis spectra of electropolymerized pure PCz and PCz/Graphene nanocomposite film.

shifted to higher energy side with reduced intensity of polaronic peak (656 nm), which revealed the interactions between graphene and PCz.

Figure 4 discloses the structural characterization by FTIR spectra of pure graphene, pure PCz electropolymerized film, and PCz/graphene nanocomposite electropolymerized film, respectively over ITO substrate. It has already been reported in literatures that composite is a class of material in which all phases present in their own form with presence of interaction and do not form any new phase. The interaction between different phases can be seen by appearance or disappearance of new peaks, disappearance of older one and shift of already existing peaks, respectively. FTIR spectrums were recorded for methanol derived pure graphene, pure PCz as well as PCz/graphene composite film. The peaks at 1600 $cm^{-1}$, 2920 $cm^{-1}$, and 3400 $cm^{-1}$ were observed to predict the interaction between the polymer and graphene in nanocomposite film. The presence of OH peak in graphene might be due to partial oxidation of graphene during synthesis of graphene. PCz shows the presence of broad absorptions peak just below the OH one. This peak might be due to the presence of N-H stretching absorption of heterocyclic rings in PCz. The composite shows the absorptions intensity and peaks between the graphene and PCz with lower wave number shift. 1500 $cm^{-1}$, and 2920 $cm^{-1}$ peaks also confirm the presence of interaction among the polymer and graphene in composite.

The electronic properties of the devices Al/PCz/ITO and Al/PCz-graphene nanocomposite/ITO are investigated by current density–voltage (J–V) measurements at room temperature (27 $^0$C) in air under dark conditions. The cross sectional view of fabricated device ITO/PCz/Al Schottky junction is shown in the schematic diagram (inset of Fig. 5(a)). The J–V characteristic of devices Al/PCz/ITO and Al/PCz-graphene nanocomposite/ITO show good quality rectifying behavior as shown in Fig. 5(a). The schematic structure of the device is shown in inset of Fig. 5(a). The junction properties of polymer semiconductors are depend upon the interfacial properties of two electrodes used for diode fabrications and the difference of their respective work function. For fabrications of diode which can be used as rectifier, one need to choose electrodes in such a way that one electrode makes ohmic contact with semiconducting layer and other electrode makes non ohmic contact. According

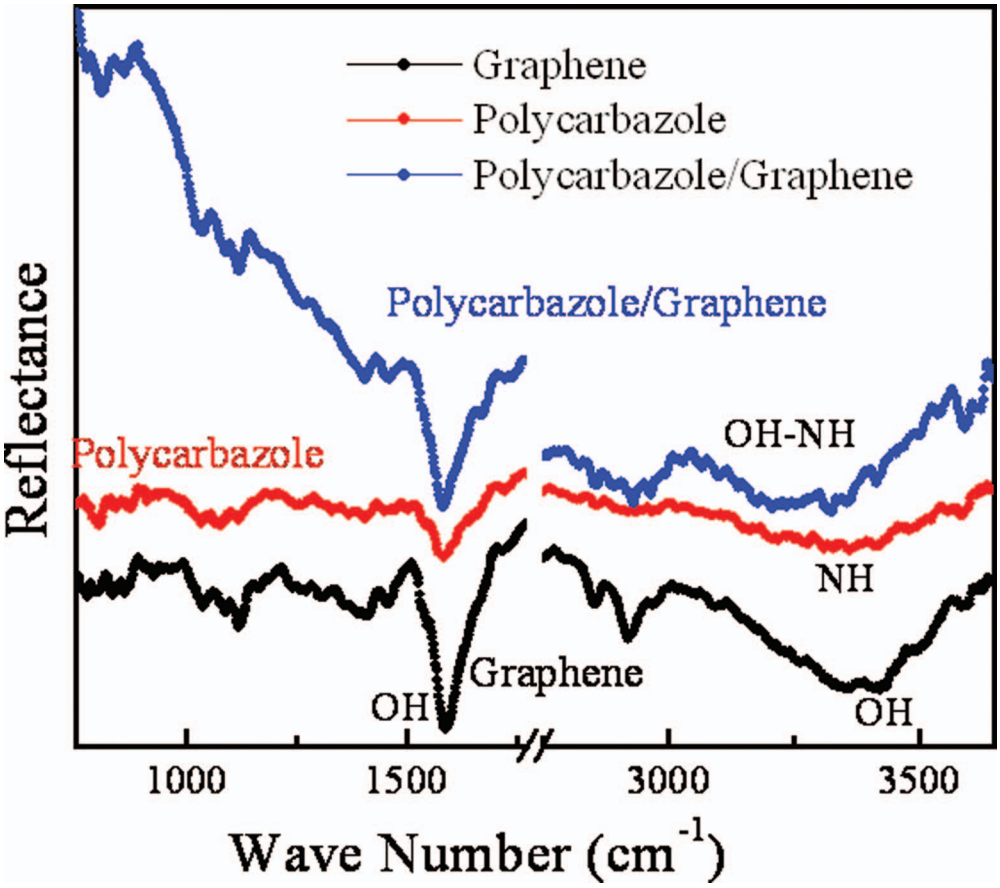


FIG. 4. FTIR spectra of methanol derived graphene, polycarbazole (PCz) and polycarbazole (PCz)/graphene nanocomposite, respectively.

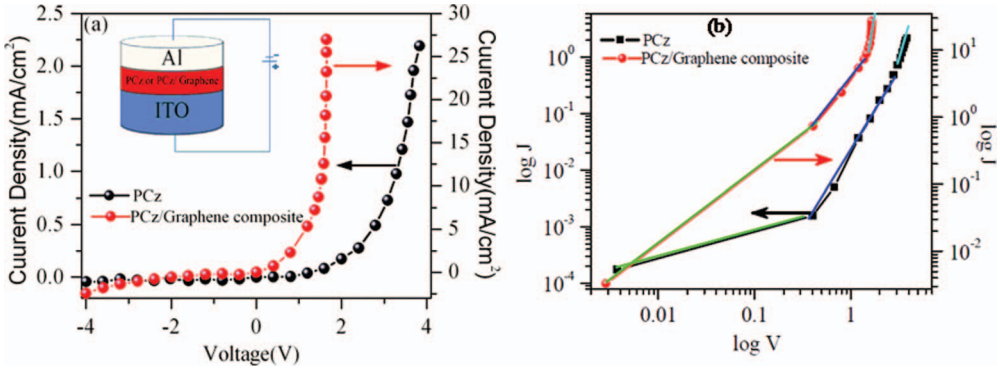


FIG. 5. (a) J-V characteristics of PCz and PCz/graphene composite. Inset shows the scheme of fabrication of device and measurement. (b) Log J vs Log V plot for PCz and PCz/graphene composite film.

to the theory of Schottky barrier, the work function of the metal must be smaller than that of the p-type semiconductor for the formation of a rectifying barrier at the interface. As reported in literature, the work function of ITO is comparable with polymer while Al forms rectifying contact due to lower work function with polymer.[38] We choose these materials for formation of rectifying contact. The rectifying behavior of the devices Al/PCz/ITO and Al/PCz-graphene nanocomposite/ITO were

TABLE I.

| Devices | $J_0$(A/cm$^2$) | $\phi_B$ (eV) | $\eta$ |
|---|---|---|---|
| ITO/PCz/Al | $8.43 \times 10^{-8}$ | 0.83 | 4.3 |
| ITO/PCz-graphene/Al | $3.29 \times 10^{-6}$ | 0.71 | 2.8 |

analyzed by assuming the standard thermionic emission theory. According to this theory, the *J–V* relationship (also called Shockley Diode equation for diode) is expressed as

$$J = J_0 \left[ \exp\left(\frac{qV}{\eta kT}\right) - 1 \right]$$

where $\eta$ is ideality factor and $J_0$ is the saturation current density in absence of external bias, q is electronic charge, V is the applied voltage, T is temperature in Kelvin and k is the Boltzmann constant. $J_0$ is related to the Schottky barrier height, $\phi_B$ as

$$J_0 = AT^2 \exp\left(\frac{-q\phi_B}{kT}\right)$$

Where $A^*$ is the effective Richardson constant. The value of $A^*$ is calculated and found to be 1.2 $A$ cm$^{-2}$ K$^{-2}$ by using effective mass of charge carrier in PCz thin film[5] and $\phi_B$ is the barrier height. Based on the above equations, we calculated the saturation current density $J_{0,}$ barrier height $\phi_B$ and ideality factor $\eta$ of devices Al/PCz/ITO and Al/PCz-graphene nanocomposite/ITO and given in Table I. The ideality factor of device Al/PCz-graphene nanocomposite/ITO is much lower than device Al/PCz/ITO. The better ideality factor of composite is may be due to the presence of graphene in polymer matrix enhances the charge transport across the interface. However the ideality factor of both devices is greater than one. The obtained ideality factor value for Al/PCz/ITO is almost two times larger than previously reported value by our group for same polymer.[5] This discrepancy generated due to the increase in the thickness of film. The thickness of device used for electrical characterizations is two times more in comparison to previously reported Al/PCz/ITO. Ideality factors from 1.0 to 2.0 are normally attributed to the carrier drift diffusion process and the Sah–Noyce–Shockley generation recombination process.[39] The exceeding value from 2.0 originates from the trap-assistedtunneling,[40–43] carrier leakage[46] and barrier inhomogeneity.[5,47] The ideal diode equation assumed that all the recombination occurs via band to band in the bulk areas from the device (i.e. not in the junction). Using this assumption the derivation produces the ideal diode equation and the ideality factor, $\eta$, is equal to one. However, recombination does occur in other ways and in other areas of the conducting polymer device like recombination via traps and in the junction area of device. The number of recombination sites increase with increasing the thickness of bulk region of device. This type of additional recombination's produce ideality factors higher than one that deviate from the ideal diode.[33] The current density obtained for Al/PCz/ITO configurations is higher in comparison to previously reported paper[5] by our group. This discrepancy comes due to difference in active area of the devices.[44] Previously we had made device of active area $4 \times 4$ mm$^2$,[5] however this time we have made device area of 0.785 mm$^2$.

In the forward region current increases exponentially while in reverse region current does not follow the trend of forward region. Asymmetric and non-linear J–V curves in forward region show that these devices exhibit rectification behavior. Saxena and Santhanam[45] have proposed the conduction mechanism in non linear J-V characteristics. The conduction mechanism in non-linear J–V characteristics of the polymer/metal junction could be explain on the basis of space-charge limited conduction (SCLC), Poole–Frenkel emission or thermoionic emission.[46,48,49] A system will follow the SCLC mechanism if the plot of log J vs. log V is linear. The log–log plot of J–V characteristics of devices Al/PCz/ITO and Al/PCz-graphene nanocomposite/ITO have three different linear regions (green, red and cyan). The first region (green region) exhibits an ohmic behavior at lower voltage. In the second region (red region) charge transport is mainly governed by SCLC conduction mechanism. In the third region (at higher voltages) conduction mechanism

is governed by SCLC with an exponential distribution of traps in the band gap of the organic material.

The current density of PCz-graphene nanocomposite is much higher (up to ten time of magnitude) than pure PCz as shown in Fig. 5(a). Now a question arises, why composite shows the high current density in comparison to pure PCz while both the films are deposited for same time for same thickness. This can be explained on the basis of Mott's law of variable-range hopping for variety of disordered materials. There are three type of conduction are possible in the composite materials. First one is hopping through polymer chains, second one is hopping between the polymer chains and conducting fillers and third one is conduction between conducting fillers (for high percentage of fillers) through polymer chains. It is unlikely that the disordered polymeric material in the composite plays a significant part in this large increase in conductivity because conductivity of these polymer chains is probably much less than that of the graphene nanosheets. It means graphene nanosheets play a huge role for large improvement in current. This behavior is indicative of percolative character in composite systems. Percolation theory deals with the effect of varying the number of interconnections present in a random polymer system. In this case the interconnections are the highly conductive graphene sheets. By using the percolation theory and the transport property described by Mott's law of variable-range hopping for a large variety of disordered materials and metal–semiconductor transition in which conduction occurs by hopping (i.e. phonon-assisted tunneling between electronic localized states centered at different positions) in disordered semiconductors with localized states in the band gap. Introduction of a very small amount of graphene nanosheets (as conducting filler) as high conducting material in disordered material (PCz matrix) provides a easier path (percolation path) between two electrodes through which charge can percolate.[50,51] This percolation path facilitates the conduction mechanism by reducing the variable range hopping path between two the electrodes. This phenomena cause increase in current by result of reduction in the path resistance. The J-V curve already discloses and reduction of recombination through the traps, defect etc. cause decrease in ideality factor as given in Table I. SEM studies of the PCz-graphene composites also supported uniform distribution of graphene and formation of compact film (covering cavities by fillers). Interaction between the filler (graphene sheets) and polymer is probably resulted better path and less traps for the charge carriers. The increase in the reverse saturations current density '$J_0$' and lower barrier height '$\phi_B$' in PCz-graphene nanocomposite can be explained on the basis of uniform distributions of graphene in PCz matrix. It has already been reported in the literature about good conductivity of graphene. This uniform distribution of graphene in PCz matrix causes the reduction in the work function of composite results in decrease in the barrier height with increase in the current density in comparison to pure PCz. Forment *et al.* has reported about the reduction in barrier height distributions for smooth surfaces.[47] The incorporations of graphene in PCz matrix causes the enhancements in uniformity of composites film surface which reduces the barrier height of device. In another way, graphene also acts as dopant in PCz matrix and increases its conductivity. It is well know that the depletion width and barrier height of Schottky diodes depend on the doping concentrations.[52] Thus graphene in PCz matrix decreased the depletion width and barrier height of diode and increased the reverse saturations current density in device Al/PCz-graphene nanocomposite/ITO in comparison to device Al/PCz /ITO.

Our electrochemical technique for formation of composites and direct coating of substrates for fabrication of devices showed potential for electronic applications and may be explored for other polymers and fillers. The incorporation of conducting nanofiller in polymer matrix reduced the barrier height and increased the reverse saturation current, same time, decreased the diode turn on voltage with decrease in ideality factor.

## IV. CONCLUSION

We have directly synthesized polycarbazole (PCz)–graphene nanocomposite over indium tin oxide (ITO) coated glass substrates using electrochemical polymerization technique. The synthesized nanocomposite is characterized by SEM, UV Vis. and FTIR spectroscopy. The SEM images clearly showed difference in morphology of pure PCz and PCz-graphene nanocomposite. It is observed that graphene nanosheets are uniformly distributed in PCz matrix. Composite coated substrates are

used for fabrication of Schottky diodes of configuration Al/PCz-graphene nanocomposite/ITO and compared with devices formed by pure PCz. The diode performance parameters (such as the ideality factor, barrier height, and reverse saturation current density) of both the devices are calculated using J-V characteristics. We observed much better ideality factor and also very high forward current density & lower turn on voltage for device Al/PCz-graphene nanocomposite/ITO in comparison to device Al/PCz/ITO. Possible conduction mechanism and reason for better device performance is extensively discussed in paper.

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