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Al/PANI-MWNT/Au-Plastic Schottky Diode Solar Cells

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Abstract

Al/PANI-MWNT/Au-Plastic Schottky diode solar cells were fabricated by the electrochemical polymerization technique to make polyaniline films on the top of gold nanoparticles film. The aluminum contact was deposited by thermal evaporation. The electro-optical characteristics of these devices produced at the different polymerization time were compared. Here, we achieved the highest ever reported open-circuit voltage of 0.8 V with the electrochemical polymerization technique. The polymerization of polyaniline films was thought to be a major factor in the enhanced performance. The effects of varying the polyaniline thickness on the device performance were investigated.

Introduction

Conjugated polymers exhibit conducting or semiconducting properties. Semiconducting polymers are now attracting considerable attention as promising materials for the development of optoelectronic devices such as light emitting diodes, photovoltaic cells, and nonlinear optical systems. The development of plastic electronics into a well-established technology is a goal currently pursued by many research groups worldwide (Peumans et al. 2003, Hill et al. 2000). The success of plastic electronics depends critically on significant improvements in devices based on organic semiconductors (Forrest 2004, Yang et al. 2005). Organic semiconductors like poly (3-hexylthiophene) (P3HT), polypyrrole, and polythiophene are finding more and more applications in many optoelectronic devices including light-emitting diodes (Singh et al. 2005, Singh et al. 2006), and solar cells (He et al. 2006).

The metal/organic semiconductor Schottky junction as an alternate to the metal/inorganic semiconductor junction has been developed (Kwong et al. 2003, Rajaputra et al. 2007), which has opened the new possibility of replacing conventional inorganic devices by organic ones (Takada et al. 2002, Su et al. 2007). Among conducting polymers, Polyaniline (PANI) has received greater attention due to its advantages over other conducting polymers. The simplicity of its preparation from cheap materials, superior stability to air oxidation, controllable electrical conductivity by doping, and reversible electrochromism (Kobayashi et al. 1984) make it very useful in preparing lightweight batteries (Oyama et al. 1995), electrochromic devices (Yang et al. 1994), sensors (Shinohara et al. 1988) and electroluminescent devices (Gustafsson et al. 1992). PANI-based solar cells are of interest because of their potential as flexible, lightweight and inexpensive devices. High open-circuit voltages have been obtained in Schottky diode solar cells manufactured by electrochemical polymerization (Singh et al. 2009). However, short-circuit current densities (Isc) in these cells are not as good as in most other organic semiconductor cells (OSC). The major reason for low Isc in the organic semiconductor cells is the small exciton diffusion length of a few nm.

In this paper, we investigated a new type of Schottky diode solar cells based on electrochemical polymerization of PANI films, which exhibits the highest open-circuit voltage of 0.8 V to date. It was found that the addition of multi-wall carbon nanotubes (MWNTs) into PANI would form PANI-MWNT composite, which can lead to significant improvement in photovoltaic conversion efficiency.

Methods

Aniline monomer was distilled under reduced pressure before use. Aniline monomer was mixed to a final concentration of 0.1 M in 1 M sulfuric acid with total volume equal to 40 ml. The polyaniline was synthesized by galvanostatic step method at a constant voltage of 2 V. The working electrode was plastic film with a thin 30 nm Au layer with surface area of 1.5 cm^2. A paper clip was used as a counter electrode. The amount of the electrodeposited polyaniline was estimated by weighing the working electrode before and after the electrodeposition. Figure 1 displays the

schematic diagram of an electrochemical cell. The gold film on the plastic substrate was deposited by thermal evaporation. The Au/plastic substrates were cleaned by sonication in acetone and 2-propanol and then radio frequency plasma to remove the oil spots. Au/plastic substrates were used as working electrodes for depositing thin film of polyaniline for 60 seconds under constant voltage of 2 volt, and then washed by DI water and finally dried by blowing nitrogen gas. To obtained different coating thicknesses, we used different polymerization time: 60, 120, 180, and 240 s. A 100-nm-thick aluminum layer of area 0.09 cm^2 was thermally deposited as the electrode. To make Al/PANI-MWNT/Au-Plastic Schottky devices, the purified MWNTs were dispersed in dimethyl-formamide (DMF) at 0.2 mg/ml and airbrushed onto Au/plastic slides with a surface area of 1.5 cm^2. The schematic diagrams of these two types of solar cells are shown in Figure 2. The film was characterized by optical absorption spectroscopy, photoluminescence, and scanning electron microscopy.

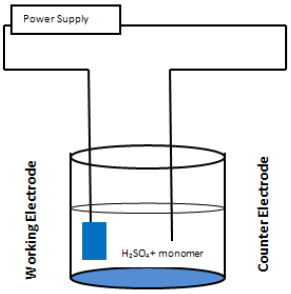

Figure 1. The diagram for electrochemical cell.

Figure 2. Schematic diagram of Al/PANI/AuPlastic.

Results and Discussion

Morphology of PANI and PANI-MWNT composite

Figure 3 shows the surface topography of polyaniline thin film using the scanning electron microscopy. The PANI thin film exhibits a nano-network structure that helps increase mechanical and electrical properties for the heterojunction Schottky diode solar cell. However, when the aniline monomer was electrochemically polymerized onto the MWNT-coated Au/Plastic, the network structure disappeared in the PANI-MWNT composite. This means that the polymer filled the vacancy space in the MWNT network, generating an improved uniform surface.

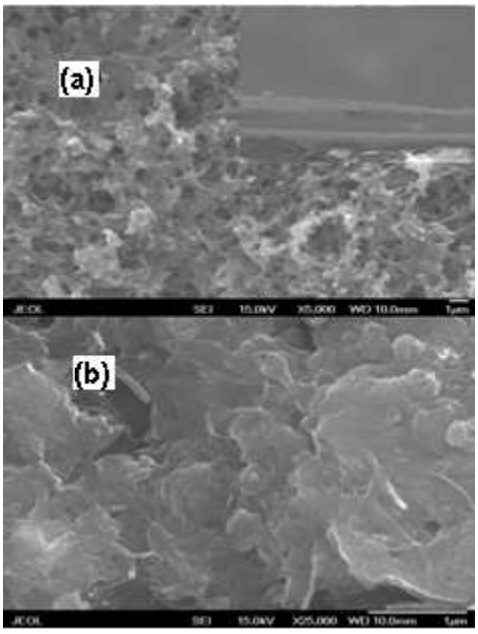

Figure 3. SEM images of polyaniline deposited onto gold foil (a) and PANI-MWNT composite (b). The inset in (a) displays the cross section view of PANI/Au.

Optical absorption spectroscopy

The optical absorption spectroscopic measurements were conducted on PANI nanofibers using UV-1700 spectrometer by dispersing equal concentration of the fibers in double distilled water. Fig 4 shows the UV-Vis-NIR absorption spectra of polyaniline/gold sheet for different polymerization time. The polyaniline matrix from the reduced state to the partially oxidized conducting form shows four absorption bands. The spectra for the reduced form of polyaniline reveals a strong absorption band gap and the absorption peaks change with the deposition time. The PANI nanofibers, which are synthesized in the emeraldine salt form, exhibit three prominent peaks. The absorption peak at around 320nm is attributed to the π-π^* band transition. There are four peaks: the 430nm, a 550nm, and two at 700nm and 900nm that merge as polymerization time decreases. The peak at a wavelength of around 430nm is attributed to the

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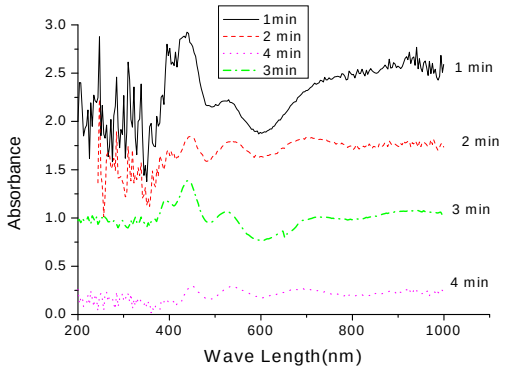

Figure 4. Absorption spectra of the polyaniline/gold sheet for different polymerization times.

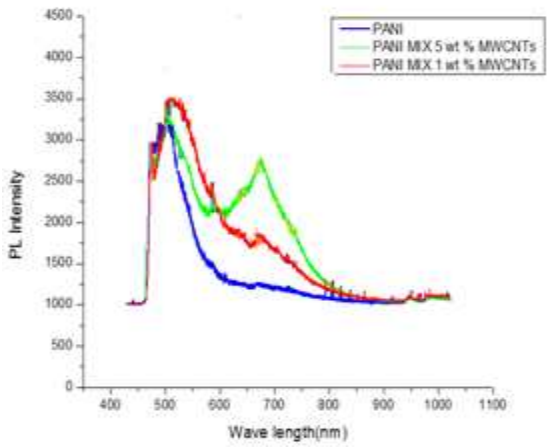

Figure 5. The photoluminescence spectra of PANI and PANI-MWNT composites.

transitions between band-polaron band and the one at 900 nm is due to the polaron band-π* band transitions. The UV-Visible spectra indicate that there is a single broad polaronic band deep in the band gap of the PANI nanofiber. It has been proposed that the presence of coulombic interactions, dielectric screening and local disorder in the polyaniline lattice act to stabilize the delocalized polaron state. The UV-results indicate the formation of a polaronic band in between the band gap of the polymer upon doping. It is observed that the peak attributed to the π-polaron band transition at 430 nm intensifies with the increase in dopant concentration. This is due to the fact that with the increase in dopant concentration, the density of the localized defect states increases in the polaronic band. This leads to an increase in the density of charge carriers and as such the direct current conductivity of the polyaniline nanofibers increases with the increase in dopant concentration.

Photoluminescence

The Photoluminescence (PL) spectra of the PANI nanofibers were taken with an exitonic wavelength of 228 nm, using a Perkin Elmer Ls-55 fluorescence spectrometer. Figure 5 shows the PL spectra of the PANI nanofibers. All the curves show a common peak at around 580 nm. The UV-Visible spectra in the Figure 5 indicate a single broad polaronic band at around 450 nm, which is due to transitions from the polaronic band to the π-band (HOMO) and therefore the PL peak at 450 nm intensifies. The increase in intensity of the PL peaks can also be attributed to the reduction of diameter of nanofibers.

The I-V characteristics of gold/PANI Schottky diode solar cells

Dark I-V characteristics of the gold/PANI Schottky diode heterodiode shown in Figure 6 exhibit a rectifying behavior with a rectification of 1.39 at 0.72 V (rectification stands for the ratio of the forward to reverse current at a certain bias voltage). The rectifying behavior indicates the formation of a diode between gold thin film and PANI p-type film. The reverse current of this diode illustrates a gradual increase. The forward current shows an exponential behavior of the form:

$$\mathbf{I = I_o \exp\left(\frac{qV}{nkT}\right)} \qquad \mathbf{(1)}$$

Where I_o is the saturation current, V is the bias voltage, q/kT is the thermal energy and n is the ideality factor.

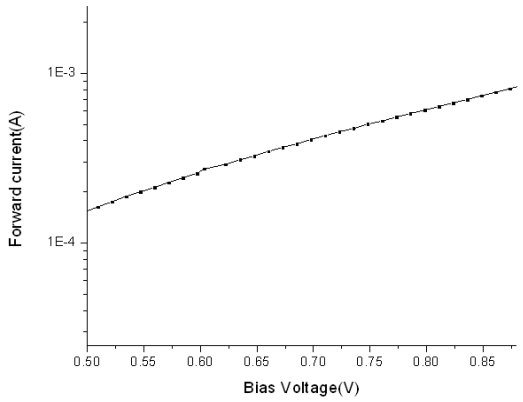

Figure 6. The dark I-V characteristic curve of gold thin film /PANI Shotcky diode.

The dark semi-log I-V curve of the gold/PANI Schottky diode device (Figure 6) exhibits a linear behavior in the bias region of 0.05-0.1691 V. The empirical equation of this diode is:

$$I = 1.7 \times 10^{-4} \exp(8.87V / n) \qquad \textbf{(2)}$$

The equation shows an extrapolated saturation current of 1.7×10^{-4} A, while the ideality factor that is calculated from this equation is 5.4. The high value of *n* suggests that the carrier transport of this device is dominated by more than one mechanism.

The J-V characteristics of the gold thin film/PANI thin film solar cell exhibits considerable photovoltaic performance as depicted in Figure 7, with short circuit current density (I_{SC}), open circuit voltage (V_{OC}) and fill-factor (FF) for different polymerization time in the Table 1. The highest conversion efficiency of the cell is 0.25% for 3 min polymerization time, indicating a significantly considerable performance.

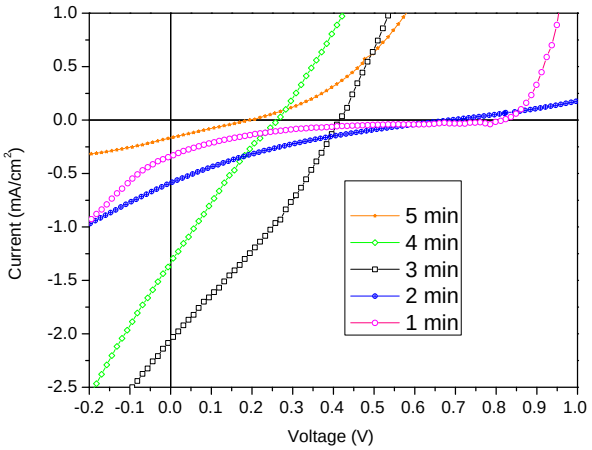

Figure 7. I-V characteristics gold thin film/PANI at different polymerization time.

Table 1. The photovoltaic performances of the gold thin film /PANI with different polymerization time.

Polymerization time (min)	I_{sc} (mA/cm^2)	V_{oc} (Volt)	FF	Efficiency (%)
1min	0.035	0.82	0.10	2.6×10^{-2}
2 min	0.060	0.69	0.16	6.7×10^{-2}
3 min	0.204	0.41	0.29	2.5×10^{-1}
4 min	0.136	0.26	0.23	7.9×10^{-2}
5 min	0.017	0.19	0.25	0.8×10^{-2}

The Al/PANI-MWNT/Au-Plastic Schottky devices

The electrochemical polymerization method can aid the formation of very good omic contact. The optimum condition to obtain high photovoltaic conversion efficiency for polyaniline on the gold foil is at a polymerization time of 1 min.

As seen in Figure 8, the PANI-MWNT composite can alter the photovoltaic performance as compared to those made of PANI itself. For instance, when the MWNT concentration is 5%, the Voc is 0.45V and FF improves more significantly from 0.1 to 0.51, and the total conversion efficiency increases from 0.03% to 0.31%. Adding MWNT material can tune the performance of Schottky solar cells. The synthesis by in situ polymerization processes leads to effective site-selective interactions between the quinoid ring of the PANI and the MWNTs, and the formation of a genuine PANI-MWNT composite. This facilitates charge transfer processes between the two components and results in enhanced electronic properties (Maser et al. 2003). According to other studies, (Su et al. 2007) the electrical conductivity of a dedoped PANI-MWNT composite with a 16.3 wt % concentration of MWNTs reached 3.0×10^{-3} S/cm, which was 6 orders of magnitude higher than that of dedoped PANI nanorods. The coexisting composites of PANI nanorods and MWNTs coated with PANI had high electrochemical activity and good cyclic stability.

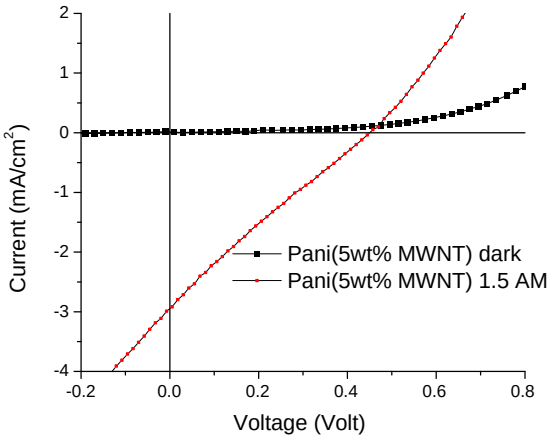

Figure 8. I-V characteristics of Al/PANI-MWNT/Au-Plastic Schottky devices.

Conclusions

In brief, we fabricated plastic/gold/PANI/Al Schottky diode solar cells and Al/PANI-MWNT/Au-Plastic devices by electrochemical polymerization of

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PANI on the evaporated gold on the plastic foil. The solar cell exhibited highest Voc values of 0.8V after 1 min electrochemical polymerization of PANI. With proper concentration of MWNT in the PANI-MWNT composite, the plastic/gold/PANI-MWNT/Al solar cell can reach efficiency above 0.31%.

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