

Electrical Properties of InSe/PMItz Nanocomposite Thin Films

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An outperforming inorganic semiconductor InSe thin films were growth on glass/ITO substrate by electrochemical deposition method and one group annealed. Organic semiconductor PMItz was successfully growth on two separate films by physical vapor deposition (PVD) method. For electrical analyses, heterojunctions were contacted with indium and their I-V characteristics were investigated under varying light intensities. From I-V measurements, it is calculated that ideality factors n were between 7.53-3.94 and 1.45-1.42, fill factors were between 0.25 and 0.26 and serial resistance R_s values were $\approx 10^8 \Omega$. It is observed from overall data that first time growth inorganic-organic heterojunctions herein exhibited photodiode and photovoltaic behaviours.

Keywords: InSe; Organic semiconductor; hybrid heterojunction; photovoltaic cell; photodiode.

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1. Introduction

Outstanding optoelectronic properties of indium selenides (In_xSe_y) that can constitute varied elemental compositions still make them peculiar over the decades for the development of multivarious and functional electro-photonics [1-6]. Since their photodetective characteristics that outperform classical 2D semiconductors are promise for advanced data processing, storage and optical imaging technologies [7-12]. Their various and compatible heterojunctions with other III-VI semiconductors through Van der Waals epitaxy facilitate thermal and electrostatic tunability to achieve desired optical and electrical properties as well as to increase the diversity of the devices [13-17]. Dangling covalent bonds at their contact surfaces as their inherent property gives opportunity for easy heterointerface processing by reducing trap cites and ignoring lattice mismatch [18-20]. On the other hand, nonflexible film

structure, relatively high production cost and paucity in variety of inorganic semiconductors are their major drawback for commercialization whereas small molecule-based organic semiconductors are more advantageous and have started to be more preferable in terms of their high abundance, relatively low fabrication costs, tunability of optoelectronic properties by chemical modification and more flexible film structure which also enables the design of biocompatible devices [21-25]. Therefore, studies based on organic semiconducting heterojunctions attract an increasing attention for last few decades. In connection with this, Phenanthrimidazoles as a popular class of π -conjugated organic semiconductors exhibit excellent photophysical and optoelectronic behaviours and are especially utilized for the fabrication of organic electroluminescent devices for a few decades [26-32]. But optoelectronics where phenanthroimidazoles were deposited on inorganic substrates are relatively rare. Also, organic-inorganic heterostructures where In_xSe_y were used as substrate were

not encountered in the literature so far to our knowledge. Moreover, heterojunction of organic and inorganic layers increases the variety of fabricated devices and eliminates the aforementioned drawbacks of inorganic semiconductors to some extent. In our previous study, we fabricated an ITO supported photodiode thin film of a newly synthesized phenanthroimidazole derivative (PMItz) which displays strong negative differential resistance (NDR) property both in the reverse and forward bias conditions [33]. As our ongoing research we herein deposited PMItz on In_xSe_y substrate by physical vapor deposition (PVD) method to fabricate a new organic-inorganic hybrid heterojunction. One of separately prepared two films was also annealed. Thus, it is aimed to take advantage of prominent electrical features of both In_xSe_y and PMItz. The obtained photodiode and photovoltaic behaviours of the first time fabricated InSe and PMItz based hybrid heterojunctions were observed.

2. Experimental

Autolab PGSTAT128N model electrochemical impedance spectroscopy instrument was used for electrochemical deposition. A Vaksis PVD-MT/2M2T thermal evaporation thin film deposition system was used for thermal deposition.

2.1. Construction of Glass/ITO/InSe thin films

In_xSe_y was grown on a glass/ITO substrate having specific resistance of lower than 20Ω by electrochemical deposition method. The substrate was cleaned with deionized water, acetone, propanol and dried with nitrogen before deposition.

25 mM InCl_3 (sigma aldrich %99,999) for In source, 25 mM $\text{H}_2\text{O}_3\text{Se}$ (sigma aldrich %98) for Se source, 250 mM LiCl (sigma aldrich %99) and 350 mM KCl (sigma aldrich %99) as buffer solutions were used. For one-step electrochemical deposition, electrolyte volume was set to 60 ml using 15 ml from each solution. The pH of the electrolyte was adjusted to 1,5 using 9,45 M HCl solution and applied voltage set to $-0,95 \text{ V}$. Platin plate as counter electrode with the dimensions of $1,0 \times 1,5 \text{ cm}$, Ag/AgCl reference electrode, and glass/ITO as working electrode with the surface area of 1 cm^2 were used. The reaction was performed at room temperature during 60 min. and in nitrogen atmosphere with 0.1 bar. A set from obtained films was annealed at 70 cmHg and $105 \text{ }^\circ\text{C}$ temperature during 60 min.

2.2. Fabrication of Glass/ITO/InSe/PMItz and Glass/ITO/InSe(annealed)/PMItz heterojunctions

PMItz as organic layer in powder form was grown on Glass/ITO/InSe and Glass/ITO/InSe(annealed) by using physical vapor deposition (PVD) method. The temperature

and pressure of the chamber was set to $26 \text{ }^\circ\text{C}$ and 10^{-6} torr respectively using Mo pot.

2.3. Fabrication of In/InSe/PMItz/In and In/InSe(annealed)/In heterojunctions

In metal contacts were coated on InSe, InSe(annealed) and PMItz layers using the PVD method using a mask.

3. Results

3.1. Electrical Analyses

Fluctuations or changes in currents within heterojunctions by applied external potential are pivotal for electronic applications. Sometimes variations in measurements are possible by changing external parameters such as high-low temperature [34], exposition of heterojunction to incident light with different frequencies [35] or as in our study, varying of light intensity in constant frequency. Electrical analyses were performed in dark and 420 W/m^2 light intensity. The fabricated heterojunctions were contacted with indium by PVD as illustrated in Figure 1.

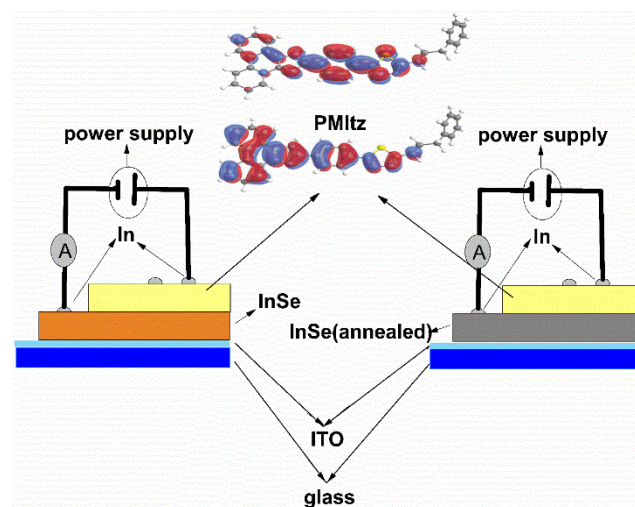


Figure 1. Schematic representation of In/InSe/PMItz/In(left) and In/InSe/PMItz(annealed)/In(right) devices.

In Figure 2a and 2b, $\ln(I)$ versus V graphs of In/InSe/PMItz/In and In/InSe/PMItz(annealed)/In devices in $\pm 5 \text{ V}$ range and under forward and reverse bias conditions at dark and illumination environment. The asymmetry of the plots in right and left parts is clearly seen. Unsaturated condition is observed in reverse bias, which originates from image force lowering of barrier height between PMItz layer and In contact [36-42]. This condition is more stable in In/InSe/PMItz(annealed)/In device. It is seen that the electrical conductivity of both heterojunctions increases in forward bias under light.

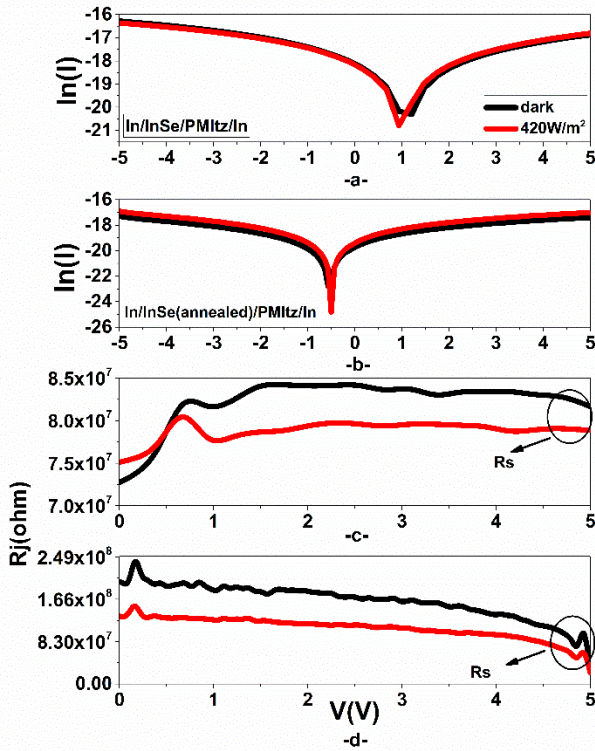


Figure 2. a) $\ln(I)$ versus V , c) R_j versus V plots of In/InSe/PMItz/In device and, b) $\ln(I)$ versus V d) R_j versus V plots of In/InSe(annealed)/PMItz/In device in dark and $420\text{W}/\text{m}^2$ light intensity condition.

In Figure 2c and 2d, R_j (junction resistance, $R_j = dV/dI$) versus V plots are given. R_j values of In/InSe/PMItz/In device changes between $8.5 \times 10^7 \Omega$ and $7.2 \times 10^7 \Omega$ while In/InSe/PMItz(annealed)/In device, R_j values are between $2.65 \times 10^8 \Omega$ and $1.1 \times 10^8 \Omega$. R_j values of both heterojunction decreased with the light. R_s serial resistance can be also estimated from the above plot, where low R_s value indicates high current flow across the junction [43]. In heterojunctions under light, photons of higher energy than forbidden energy band gaps (E_g) generate new electron-hole pairs through depletion region. These pairs which make stress in the structure splits by strong internal electric field induced in particle boundaries. Electrons flow quickly through PMItz layer while holes are slower owing to trap levels. Accordingly, photoconductivity increased while R_j values decreased [36, 44].

Current-voltage relations of diodes (MIS, MPS, MOS, MS) are determined according to thermionic emission theory and diode parameters can be estimated by different methods [45-49], Norde method [50] and Cheung methods [51] are one of them. In our study, n and R_s values were calculated from the following equation which is the first one used in Cheung & Cheung method.

$$\frac{dV}{d\ln(I)} = IR_s + \frac{nkT}{q} \quad (9)$$

The slope of $\frac{dV}{d\ln(I)}$ versus I plot gives R_s values while n ideality factor is calculated from the y-intercept using other constants together [51].

The plot of $\frac{dV}{d\ln(I)}$ versus I , were given in Figure 3a-b for In/InSe/PMItz/In and In/InSe(annealed)/PMItz/In devices respectively. n values calculated from these curves are between 7.53 - 3.94 and 1.45 - 1.42 for In/InSe/PMItz/In and In/InSe(annealed)/PMItz/In devices respectively. Although n values decreased with the light intensity, they are extremely far from unity, ideal diode value, especially for In/InSe/PMItz/In device. This arise from the existence interface states within PMItz layer, high density N_{ss} values and barrier inhomogeneities [52]. Also the effects of image force effect, recombination and expansion of depletion region are possible.

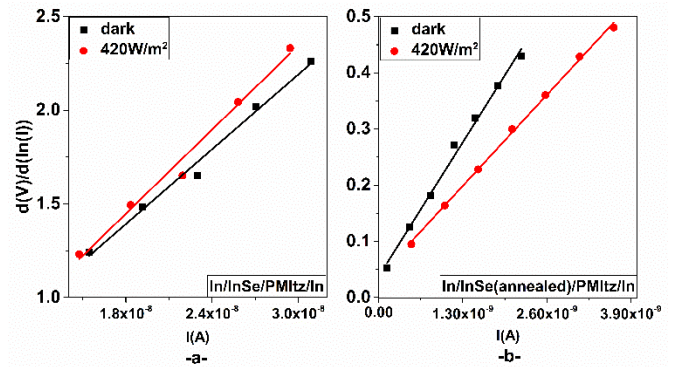


Figure 3 a) the plot of $\frac{dV}{d\ln(I)}$ versus I , of In/InSe/PMItz/In device and b) the plot of $\frac{dV}{d\ln(I)}$ versus I , of In/InSe/PMItz(annealed)/In device in dark and $420\text{W}/\text{m}^2$ light intensity condition.

R_s values of In/InSe/PMItz/In and In/InSe(annealed)/PMItz/In devices determined versus dark and light by different methods. Although there is not much change in In/InSe/PMItz/In with light, it is seen that there is a significant change in In/InSe(annealed)/PMItz/In. While R_s value from $d(V)/d(\ln(I))$ - I plot of In/InSe(annealed)/PMItz/In decreased from $1.83 \times 10^8 \Omega$ to $1.24 \times 10^8 \Omega$ from dark to light, R_s value from R_j - V plot decreased from $1.26 \times 10^8 \Omega$ to $7.38 \times 10^7 \Omega$ from dark to light. R_s values calculated with different methods are in accordance with each other and behaviours against light are similar.

The fill factors (FF) were calculated from the formula $FF = \frac{I_{mpp} \cdot V_{mpp}}{I_{sc} \cdot V_{oc}}$ [53]. Where I_{sc} is short circuit current, V_{oc} is open circuit voltage, I_{mpp} is maximum power point current and V_{mpp} is maximum power point voltage values. In Figure 4a-b solar cell parameters of In/InSe/PMItz/In and In/InSe(annealed)/PMItz/In devices were given, respectively. The solar cell parameters of the devices were estimated from

the plots. It has been observed that both heterojunctions exhibit photodiode behaviour due to the change of I_{sc} value under light. In addition, the FF value of the In/InSe(annealed)/PMItz/In device was higher than that of the In/InSe/PMItz/In device.

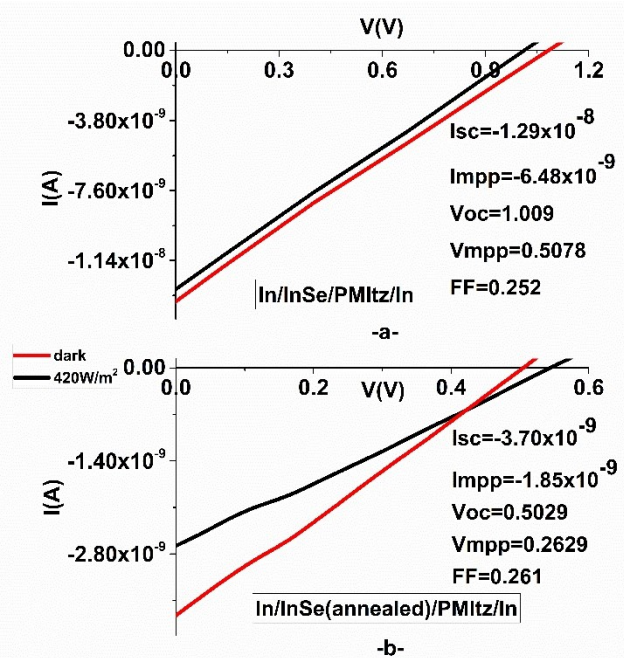


Figure 4. Solar cell parameters of a) In/InSe/PMItz/In and b) In/InSe(annealed)/PMItz/In devices.

Photocurrent ($I_{ph} = I_{ill} - I_{dark}$), photoresponsivity ($R = I_{ph}/P_{inc} \cdot A$) and photosensitivity ($S = R \cdot T/V$) are important parameters for diodes [54-60]. Where, I_{ill} is the current in illuminated condition, I_{dark} is the dark current, P_{inc} is the power of incident light (420 W/m^2) and A is contact area ($0.31 \times 10^{-4} \text{ m}^2$) of diode, T is thickness of active layer (PMItz thickness $\approx 6.12 \times 10^{-7} \text{ m}$) and V is applied bias voltage. In Figure 5, the variation of a) I_{ph} , b) R and c) S values versus different voltage values of devices under forward bias and in 420 W/m^2 light intensity is given. The I_{ph} , R and S values of the In/InSe/PMItz/In device are $9.74 \times 10^{-10} - 3.47 \times 10^{-9} \text{ A}$, $7.38 \times 10^{-8} - 2.63 \times 10^{-7} (\text{A/W})$, $4.43 \times 10^{-14} - 3.16 \times 10^{-14} \text{ S.m/W}$, respectively. The I_{ph} , R and S values of the In/InSe(annealed)/PMItz/In device are $3.75 \times 10^{-9} - 1.36 \times 10^{-8} \text{ A}$, $2.84 \times 10^{-7} - 1.03 \times 10^{-6} (\text{A/W})$, $1.70 \times 10^{-13} - 1.23 \times 10^{-13} \text{ S.m/W}$, respectively. It is seen that I_{ph} , R and S values of In/InSe(annealed)/PMItz/In device are higher than these of In/InSe/PMItz/In device at all voltage values. It is found that In/InSe(annealed)/PMItz/In device is a better diode than In/InSe/PMItz/In device. Annealing of the InSe layer improved the optical properties of the device as well as its electrical properties.

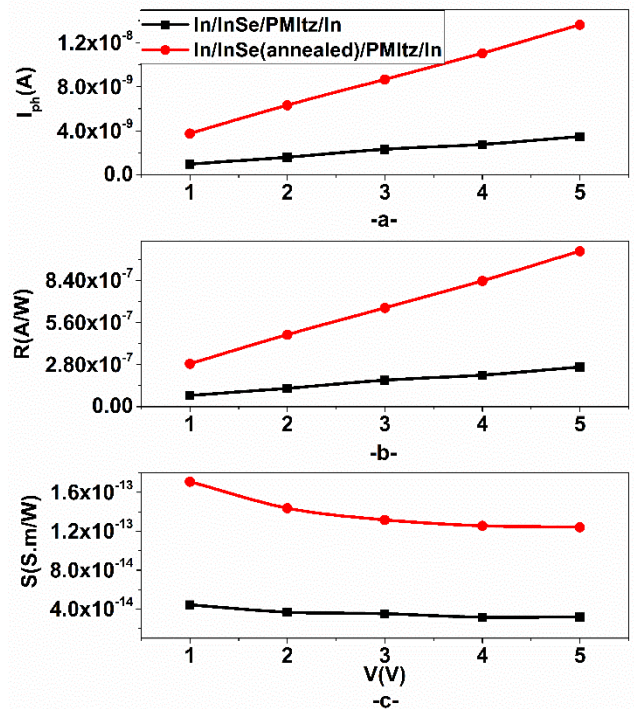


Figure 5. the variation of a) I_{ph} , b) R and c) S values versus different voltage values of devices under forward bias and in 420 W/m^2 light intensity

4. Conclusion

InSe thin films were obtained successfully on to glass/ITO substrate by electrochemical deposition method. Organic PMItz semiconductor was deposited on the obtained thin films by PVD method. Indium contacts were jointed heterojunctions and electrical parameters were investigated under dark and 420 W/m^2 light intensity. It is concluded that heterojunctions are light sensitive, power generation take places in photovoltaic region and they act as photodiode under reverse bias conditions.

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