Fabrication, illumination dependent electrical and photovoltaic properties of Au/BOD-Pyr/n-Si/In Schottky diode

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ABSTRACT

4,4-Difluoro-4-bora-3a,4a-diaza-s-indacene (BODIPY)-based BOD-Pyr compound was synthesized according to the literature and HOMO and LUMO energies of the BOD-Pyr were calculated by DFT/B3LYP/6-311G(d,p) method using Gaussian 09W. Au/BOD-Pyr/n-Si/In Schottky diode were fabricated using thermal evaporation and spin coating technique. The electronic and photovoltaic properties of Au/BOD-Pyr/n-Si/In photodiode have been investigated by current-voltage (I-V) measurements at dark and under various illumination intensities. The calculated ideality factor and barrier height of the diode in dark were found to be 2.84 and 0.75 eV, respectively. These parameters were also obtained under 100 mW/cm\textsuperscript{2} illumination level as 1.55 and 0.87 eV, respectively. The values of open-circuit voltage and short circuit current density were obtained as 0.26 V and 0.56 mA/cm\textsuperscript{2} under the illumination level of 100 mW/cm\textsuperscript{2}. These all findings suggest that Au/BOD-Pyr/n-Si/In device can be used as photodiode in optoelectronic applications.

Key words: Organic semiconductor, Schottky diode, Photodiode, BODIPY

1. Introduction

Due to their technological advantages in optoelectronic applications, the electrical properties of metal-semiconductor (MS) and metal-interface layer-semiconductor (MIS) type Schottky barrier diodes (SBDs) have been studied in detail in recent years [1, 2]. It is well known that organic compounds used as an interfacial layer for MS contacts have allow us to modify electrical parameters of diodes such as an ideality factor ($n$) and a barrier
height \( f \) [3, 4]. Many studies have showed that the organic thin layer on the inorganic semiconductor substrate
could affect the performance of these diodes owing to a change of the density of interface states, saturation current and resistance of diode [4-13].

Recently, organic dyes as a semiconducting compound have become an exciting area for their use in molecular optoelectronic devices. The most important advantages of organic dyes are their tunable electronic and easy processing features such as high optical and thermal stability, the compatibility with flexible substrates, low cost and easy production in large area applications (3). Especially, the chemical tunability of organic dyes makes them more promising candidates for modifying charge-transport mechanism and band gap and properties. The implementation of organic dyes as semiconductor has been widely used in the fabrication of several optoelectronic devices such as Schottky diodes, photodiodes, organic light-emitting diodes, and solar cells [6, 7, 9, 14, 15].

4,4-Difluoro-4-bora-3a,4a-diaza-s-indacene (BODIPY)-based dyes have gained a great deal of attention in last decades in very diverse applications due to their good favorable chemical, photonic and electronic properties, including easy synthetic functionalization, high photochemical stability, high molar extinction coefficients and high fluorescence quantum yield. These advantages make BODIPY an excellent candidate for use in electronic and optoelectronic devices [16-18]. Nevertheless, compared to considerable interest in outstanding properties of BODIPY dyes, there are only a few studies have made to characterize the electronic parameters of different Schottky diodes which were fabricated by forming BODIPY organic layer on ohmic contact. For example, Kilicoglu and Ocak measured the ideality factor, barrier height and interface state density of the Phenyl-BODIPY/n-Si structure by current-voltage and capacitance-voltage-frequency techniques [19]. Ozcan et al. fabricated the Al/Subphthalocyanine-BODIPY dyads/p-Si/Al diodes and photoelectrical properties of these diodes was investigated at solar light illuminations [15]. Therefore, further electrical and optical characterizations to investigate π- extended BODIPY compounds are highly desired to show their full potential in different optoelectronic applications. In this paper, we fabricated a novel In/n-Si/ BOD-Pyr/Au diode and studied its electrical and photoelectrical characteristics. These properties were explored by illumination-dependent current-voltage measurements.

2. Experimental Details

BOD-Pyr was prepared according to published literature procedures [20]. The synthetic route of Bod-Pyr is shown in Scheme 1. First, compound 3 was synthesized from the reaction of benzoyl chloride (1) and 2,4-dimethylpyrrole (2) in the presence of Et3N and BF3·Et2O. Then, a Vilsmeier Haack’s formylation reaction of compound 3 gave compound 4. Lastly, Wittig olefination of compound 4 by using ylide 5 gave entirely the E-isomer of the BOD-Pyr. The structure of BOD-Pyr was confirmed by 1H, and 13C NMR spectroscopy [20].
The ground state optimize molecular geometry at the gas phase of the isolated compound was obtained with the DFT/B3LYP/6-311G(d,p) computational level within the Gaussian 09W program [21]. By using this optimized molecular structure, the GaussView5 [22] graphical interface software was used to form the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) simulations. The graphical presentation of HOMO, LUMO orbitals and orbital energy levels are shown in Figure 1. As seen in Fig. 1, the energy difference between HOMO and LUMO levels ($\Delta E_g$) is 2.73. According to the literature, it is known that the range for $\Delta E_g$ is 0.5 to 3.5 eV for semiconductors [23]. The calculated $\Delta E_g$ value (2.73 eV) of BOD-Pyr indicates that BOD-Pyr can have a semiconducting behavior.

**Fig. 1** 3D frontier molecular orbital schemes for BOD-Pyr
The Au/BOD-Pyr/n-Si/In device was constructed by using a n type Si (100) wafer which has 20 Ω-cm resistivity and 500 μm thickness. Then, the solution was stirred for 1h at ambient temperature. Ultrasonic bath was used to clean n-Si wafer by using acetone, methanol and deionized water. Then, the impurities and the native oxide layer on the surfaces were removed by using a HF:H2O (1:10) solution. Ohmic contact was prepared on the back of the n-Si wafer by thermal evaporating of indium metal, and then the wafer was annealed at 350°C for 30 sec in N2 atmospheres. Then, 10 mg of BOD-Pyr compound was dissolved in 1 ml chloroform. Thin films of BOD-Pyr were made by spin coating at 1200 rpm and 30s. Finally, the front Au contact was formed on the BOD-Pyr thin film by thermal evaporating as a metallic contact and Au/BOD-Pyr/n-Si/In device design completed as given in Fig. 2.

Fig. 2 (a) Schematic diagram of the Au/BOD-Pyr/n-Si/In device; (b) Schematic energy level diagrams for Au/BOD-Pyr/n-Si/In device

3. Results and discussion
The forward and reverse bias I–V characteristics of the Au/BOD-Pyr/n-Si/In device were examined to obtain the junction parameters. The semi logarithmic I-V data for diode were taken in the dark at room temperature and under various illumination intensity (20-100 mW/cm²). As clearly seen in Fig. 3, the Au/BOD-Pyr/n-Si/In diode shows good rectification behavior in the dark. It was calculated that the rectification ratio of the structure at ± 2V was 100. Thus, thermionic emission theory can be used to obtain diode characteristics [2, 24]. According to the theory, the relation between the current and voltage for the device is described as [2, 24]:

\[
I = I_0 \left[ \exp \left( \frac{qV}{nkT} \right) - 1 \right]
\]

where q is the electronic charge, n is the ideality factor, k is the Boltzmann’s constant, T is the temperature, \( I_0 \) is the reverse saturation current and defined as:
\[ I_0 = A_0 A^* T_2 \exp(-q\beta k T) \]
Here, $A$ is the diode contact area, $A^*$ is the effective Richardson constant for n-Si ($A^* = 112 \text{ AK}^{-2} \text{ cm}^{-2}$) and $f$
is the effective barrier height which is determined by the following equation:
\[ \mathcal{Q} = \frac{kT}{q} \ln \left( \frac{AA'\gamma^2}{I_0} \right) \]
The ideality factor value can be calculated from the slope of the linear region of the forward bias \( I-V \) characteristics by the following relation:

\[
 n = \frac{q}{kT} \frac{dV}{d(\ln I)}
\]  

(4)

As can be seen in Fig. 3, the current value in reverse bias increases with increasing light intensity and this shows that Au/BOD-Pyr/n-Si/In structure is highly light sensitive and the diode exhibits photovoltaic behavior. The values of barrier height and ideality factor of Schottky diode for dark were found to be 0.75 eV and 2.84, respectively. Under illumination with light intensity 100 mW/cm\(^2\), \( \Phi_b \) and \( n \) values were also obtained as 0.87 eV and 1.55, respectively. The attained \( n \) values are greater than unity which may be attributed to factors such as presence of interfacial layer, shunt and series resistances and leakage currents [25-27].

![Experimental forward and reverse I-V characteristics of Au/BOD-Pyr/n-Si/In diode](image)

**Fig. 3** Experimental forward and reverse \( I-V \) characteristics of Au/BOD-Pyr/n-Si/In diode

The light intensity dependent \( \Phi_b \) and \( n \) values for Au/BOD-Pyr/n-Si/In diode between 20 and 100 mW/cm\(^2\) are listed in Table 1. The \( n \) and \( \Phi_b \) values have changed from 2.27 and 0.81 eV for 20 mW/cm\(^2\) to 1.55 and 0.87 eV for 100 mW/cm\(^2\), respectively. As seen, the barrier height values improve with increase in applied illumination intensity while the ideality factor values decrease with an applied illumination intensity. This change in \( \Phi_b \) and \( n \) values, which depends on the light intensity, is an expected situation for Schottky barrier diodes [25, 26].
Table 1 Illumination intensity dependent values of $n$, $I_0$ and $\int$ obtained from $I-V$ measurements
| Illumination intensity (mW/cm²) | $n$ | $I_0$ (A) | $h$ (eV) |
|-------------------------------|-----|-----------|-----------|


One of the important parameters that allows us to understand the electrical properties of the diode is series resistance. By using Cheung-Cheung method [28], as introduced by the Eqs. (5) and (6), it is possible to extract series resistance \( R_s \) value from the forward biased diode for high voltages.

\[
H(I) = V - n \left( \frac{kT}{q} \right) \ln \left( \frac{I}{A_{AA} T^2} \right) \tag{5}
\]

\[
H(I) = n \phi_b + IR_s \tag{6}
\]

The \( H(I) \) versus \( I \) function plots for various illumination intensities are given in Fig. 4. As expected, all \( H(I) \) vs. \( I \) curve have a good linear behavior. Thus, the values of series resistance of diode were calculated from the slope of \( H(I) \) vs. \( I \) curve. As seen, the values of \( R_s \) were decreased strongly with increase in illumination intensity which is due to generation of free charge carriers by incident light absorption. For instance, \( R_s \) was calculated as 3.53 k\( \Omega \) for dark, whereas it decreased up to 1.74 k\( \Omega \) by illumination under 100 mW/cm\(^2\).

![Fig. 4 Illumination dependence of \( H(I) \) versus \( I \) for In/BOD-Pyr/n-Si/Au diode](image-url)
The interface state density \( N_{ss} \) in Schottky diodes performs a key role in calculating ideality factor and barrier height. The values of ideality factor \( n \) against \( N_{ss} \) that varies with voltage are introduced by Card and Rhoderick, which is expressed as following relation [29]

\[
N_{ss}(V) = \frac{1}{q} [\varepsilon_i (n(V) - 1) - \varepsilon_s] \frac{1}{W_D}
\]

where \( \varepsilon_s \) and \( \varepsilon_i \) are dielectric constant of semiconductor and interface, respectively. The interface layer thickness of organic layer is \( \delta_i \) and depletion region width is \( W_D \). In n type semiconductor, the relation of conductivity band limit energy of semiconductor \( (E_c) \) with the energy of interface state density \( (E_{ss}) \) is expressed by [29]
\[ E_c - E_{ss} = q_e - V \] (8)
where $\mathcal{E}$ is the effective barrier height and $q$ is the electron charge. The variation of interface state density against
energy distribution of the Au/BOD-Pyr/n-Si/In Schottky diode at dark and under different illumination intensity is introduced in Fig. 5. As a result of the calculations made according to the plot of Fig. 5, \( N_{ss} \) values of Au/BOD-Pyr/n-Si/In Schottky diode are between \( 4.25 \times 10^{11} \text{eV}^{-1}\text{cm}^{-2} \) for \( E_c - 0.55 \text{ eV} \) and \( 9.38 \times 10^{10} \text{eV}^{-1}\text{cm}^{-2} \) for \( E_c - 0.70 \text{ eV} \) in the dark and also it ranges from \( 2.36 \times 10^{11} \text{eV}^{-1}\text{cm}^{-2} \) for \( E_c - 0.53 \text{ eV} \) to \( 2.81 \times 10^{10} \text{eV}^{-1}\text{cm}^{-2} \) for \( E_c - 0.70 \text{ eV} \) under 100 mW/cm\(^2\) illumination intensity. As observed in Fig. 5, the \( N_{ss} \) values decrease exponentially from the valence band of the semiconductor towards the middle of the forbidden energy band. Considering these calculations, it is observed that the increase in the lighting intensity causes a decrease in the interface states. Similar results were obtained by Ersöz et al. [30]. The \( N_{ss} \) values of the Au/PPY/n-Si diode fabricated by Ersöz et al. are between \( 1.7 \times 10^{13} \text{eV}^{-1}\text{cm}^{-2} \) and \( 6.0 \times 10^{12} \text{eV}^{-1}\text{cm}^{-2} \) for dark and range from \( 1.5 \times 10^{13} \text{eV}^{-1}\text{cm}^{-2} \) to \( 4.2 \times 10^{12} \text{eV}^{-1}\text{cm}^{-2} \) for 100 mW/cm\(^2\). The \( N_{ss} \) values calculated by Ersöz et al. are considerably higher than the values obtained for our sample.

![Fig. 5](image)

**Fig. 5** Illumination intensity dependent \( N_{ss} \) against \( E_c - E_{ss} \) plots for the Au/BOD-Pyr/n-Si Schottky diode.

The thickness of the BOD-Pyr interface layer fabricated between the semiconductor and the rectifier contact has been found using the following equation

\[
C_{org} = \frac{\varepsilon_0 A}{d}
\]  

(9)

In the equation (9), \( A \) is the effective contact area, \( d \) is the thickness of the organic interface layer, \( \varepsilon_0 \) is the permittivity constant of the space and \( \varepsilon_i \) is the permittivity constant of the interfacial layer. Interfacial organic layer capacitance value \( (C_{org}) \) was computed to be \( 2.41 \times 10^{-10} \text{ F} \) from the capacitance value in the accumulation region of 1 MHz C-V measurement in Fig. 6. The thickness of the organic interface layer was computed as \( 3.46 \times 10^{-5} \text{ cm (346 nm)} \) from Eq. (9).
The light current density-voltage ($J$-$V$) curves of the Au/BOD-Pyr/n-Si/In are shown in Fig. 7. To understand the photovoltaic performance of diode at different illumination intensities, the basic photovoltaic parameters include open circuit current voltage ($V_{OC}$) and short circuit current ($J_{SC}$) of the diode were analyzed. The $V_{OC}$ value is extracted from the point where the $I$-$V$ curve intersects the voltage axis at this region. The value of $J_{SC}$ is also obtained from the point where the $I$-$V$ curve intersects the current axis at same region. The all obtained values are listed in Table 2. As can be seen, $V_{OC}$ and $J_{SC}$ values have increased while the light intensity increases. This behavior can be considered to be due to an increase in the number of charge carriers by the illumination.
Fig. 7 Current density-voltage (J-V) plots of Au/BOD-Pyr/n-Si/In diode under various illumination intensity.

Besides, the values of maximum current ($I_m$), maximum voltage ($V_m$) and fill factor ($FF$) were calculated from Fig. 7 and Eq. (10). The fill factor ($FF$) is described as follows:

$$FF = \frac{V_mI_m}{V_{oc}I_{sc}}$$  \hspace{1cm} (10)

| Power (mW/cm²) | $V_{oc}$ (V) | $I_{sc}$ (mA/cm²) | $V_m$ (V) | $I_m$ (mA) | FF  |
|----------------|-------------|------------------|-----------|-----------|-----|
| 20             | 0.23        | 0.26             | 0.10      | 4.23 x 10⁻⁷ | 22.45 |
| 40             | 0.25        | 0.38             | 0.11      | 6.29 x 10⁻⁷ | 20.84 |
| 60             | 0.25        | 0.46             | 0.11      | 7.50 x 10⁻⁷ | 20.86 |
| 100            | 0.26        | 0.56             | 0.11      | 9.12 x 10⁻⁷ | 20.10 |

Various conduction mechanisms could be cause for non-ideal forward I-V characteristic of Au/BOD-Pyr/n-Si/In Schottky diode. The forward bias I-V measurements of Schottky diode for dark are offered in log-log dimension as showed in Fig. 8. As seen in Fig. 8, the characteristic under dark of the sample composed of four linear different areas. The first and second zones are ohmic i.e., the current changes linearly with voltage ($V^{1.1}$ and $V^{1.4}$). These values indicate that the ohmic conduction mechanism is dominant in these zones. The current in third zone depends on voltage as $V^{3.82}$. This shows that the third region is identified trap charge limited current (TCLC) mechanism. The current in fourth zone depends on voltage as $V^{2.02}$. This shows that the fourth zone is identified space charge limited current (SCLC) mechanism. Hence, the lower traps are active in this zone and the injected free carriers reason the SCLC conduction mechanism [31-34]. Furthermore, the SCLC conduction technique is generally utilized to identify the characterization of organic Schottky diodes [32, 34, 35].
The photosensitivity ($S$) of the Au/BOD-Pyr/n-Si Schottky diode can be obtained by the following equation [36, 37]

$$S(\%) = \left( \frac{I_{\text{photo}} - I_{\text{dark}}}{I_{\text{dark}}} \right) \times 100$$  \hspace{1cm} (11)

where $I_{\text{photo}}$ and $I_{\text{dark}}$ are the photo and dark current, respectively.

Fig. 9 presents the variation of photosensitivity ($S(\%)$) with the illumination intensity ($P$) of our sample at the -2 V of reverse bias. According to Fig. 9, the photosensitivity $S(\%)$ value increases with increasing the intensity of illumination. Akın et al. [36] and also Hendi [37] introduced that the reason for the increase in $S(\%)$ value is due to the photogenerated charges produced as the electric field increases.
The photoresponsivity ($R$) of the diode is the key parameter to utilize as photodiode. The photoresponsivity ($R$) is expressed according to [36, 37]

$$R = \frac{l_{\text{photo}}}{P \cdot A}$$  \hspace{1cm} (12)

where $A$ is the diode area and $P$ is the illumination intensity. The curves of variation of $R$ against $V$ of the Au/BOD-Pyr/n-Si Schottky diode with increasing illumination intensity is illustrated in Fig 10. As seen, the $R$ values increase with increasing the bias voltage for 20, 40, 60, 100 mW/cm². These $R$ values reflect that Au/BOD-Pyr/n-Si Schottky diode can be utilized to fabricate the photodiodes for active applications. These results suggest that the achievement of a n-Si/BOD-Pyr could be characterized as an inorganic/organic photodiode.
The linear variation of the current versus the intensity of illumination is the one of important parameter for a tool to be utilized as a photodiode [31, 36, 38]. The variation of the photocurrent against illumination intensity can be expressed by the following equation [38-40]

\[ I_{\text{photo}} = \gamma P^\beta \]  

(13)

where \( \gamma \) value is a constant, \( \beta \) is an exponent which is determined from the slope of the plot and \( P \) is the intensity of illumination. Fig. 11 presents the curve of \( \ln I_{\text{photo}} - \ln P \) for the data of \(-2V\) of Au/BOD-Pyr/n-Si Schottky diode. As can be seen from the curve, these curves present that the photocurrent \( I_{\text{photo}} \) increases as the intensity of illumination increases. The data on this curve present excellent linearity. The value of \( \beta \) describes whether the process of recombination is monomolecular or bimolecular [36]. The \( \beta \) value of our sample was computed as 0.77, which reflects bimolecular recombination mechanism [36, 41]. The \( \beta \) value is also attributed to the trap levels in the gap of energy band [42].
4. Conclusions

In this study, Au/BOD-Pyr/n-Si/In structure was produced by coating a BOD-Pyr thin film as an organic interlayer between metal and semiconductor. The electrical and photoelectrical properties were studied in dark and under illumination (20-100 mW/cm²) using forward and reverse bias I-V measurements. The diode has a high rectification ratio of 100 at dark and ± 2 V. Moreover, it was seen that main diode parameters were strongly dependent upon illumination intensity. The decrease in the ideality factor of the diode and an increase in the barrier height values were observed with the light intensity. The ideality factor and barrier height were found to be 2.84 and 0.75 eV in dark, and 1.55 and 0.87 eV under 100 mW/cm² illumination level, respectively. Moreover, the series resistance values obtained by Cheung’s method were found in the range of 3.53 kΩ (for dark) - 1.74 kΩ (for 100mW/cm²). The $N_{ss}$ values have been determined for $E_c$= 0.70 eV as $9.38 \times 10^{10}$ and $2.81 \times 10^{10}$ eV⁻¹ cm⁻² at dark and 100 mW/cm² illumination intensity, respectively. The open-circuit voltage and short circuit current density values were found to be as 0.26 V and 0.56 mA/cm² under 100 mW/cm² illumination level, respectively. Photocurrent ($I_{photo}$) of the sample increased with increasing the intensity of illumination that reflected bimolecular recombination mechanism. These all findings suggest that Au/BOD-Pyr/n-Si/In device can be used as photodiode in optoelectronic applications.

Declaration of Competing Interest

The authors report no declarations of interest.
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