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F4-TCNQ Concentration Dependent Capacitance-Voltage (C-V) and Conductivity-Voltage (G/w-V) Characteristics of the Au/P3HT:F4-TCNQ/N-Si (MPS) Schottky Barrier Diodes

Aslıhan Çimen[1], Hüseyin Muzaffer Şağban[1,*], Tuğba Özdemir[1], Özge Tüzün Özmen[1,2]
[1]*Department of Physics, Faculty of Arts and Science, Düzce University, 81620 Düzce,Turkey*
[2]*DÜTEM, Düzce University, 81620 Düzce, Turkey*
Corresponding Author: Hüseyin Muzaffer Şağban

Abstract :*In this work, the electrical properties of Au/P3HT:F4-TCNQ/n-Si metal-polymer-semiconductor (MPS) Schottky barrier diodes (SBDs) with the F4-TCNQ concentration values of 0.5% and 2%were investigated by capacitance-voltage (C-V) and conductance-voltage (G/w-V) measurements. The series resistance (R_s) and interface states (N_{ss}) were calculated from C-V and G/w-V measurements as a function of F4-TCNQ concentration. The preferable electrical results, such as lower capacitance, lower series resistance, higher shunt resistance and lower N_{ss}, were obtained for 2% F4-TCNQ concentration used diode. For this reason, the deeperC-V and G/w-V analysis were performed only for 2% F4-TCNQ concentration used diode. By using C and G/w measurements, diffusion potential (V_D), doping concentration of donors (N_D), depletion layer width (W_D), barrier height (ϕ_B), Fermi energy level (E_F), maximum electric field (E_m) and Schottky barrier lowering ($\Delta\phi_B$) were obtained for 2% F4-TCNQ concentration used Au/P3HT:F4-TCNQ/n-Si SBD. The results show that the series resistance and interface states decreased with increasing the frequency. Finally, it can be concluded that the concentration of F4-TCNQ has a great effect on the electrical properties of the Au/P3HT:F4-TCNQ/n-Si SBDs. Additionally,the values of R_s and N_{ss} directly affected the C-V and the G/w-V profiles of Au/P3HT:F4-TCNQ/n-Si SBDs.*

Keywords*F4-TCNQ concentration, Frequ*ency and voltage dependence, C-V and G/w-V analysis, Density of *interface states, Series resistance*

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I. Introduction

In the last few decades, the polymeric organic materials have been a subject of intensive research [1]. The main reasons of being the center of attention of organic polymers in semiconductor technology are including low-temperature processing, low cost, easy fabrication techniques and large area processing which allows the variety of large application fields and opportunity to produce of high performance devices. By considering these advantages, many devices can be fabricated with organic polymers such as organic field effect transistors (OFETs) [2], organic thin film transistors (OTFTs) [3], Schottky barrier diodes (SBDs) [4], organic light emitting diodes (OLEDs) [5] and organic photovoltaics (OPVs) [6]. SBDs are the most widely used diodes in electronic devices because of their faster response time and low forward voltage drop compared to other diodes. In these devices, organic polymer materials widely used as interfacial layer between metal and semiconductor (MS). The insertion of interfacial organic layer between MS can create new structure which referred as metal-polymer-semiconductor (MPS). In MPS structures, polymeric interfacial layer and the interface states are directly related with the main electrical and dielectric parameters of SBDs [7].

F4-TCNQ is one of the effective p-type dopant material due to the strong electron affinity of a polymer-additive systems [8].P3HT is a regioregular organic polymer material and have prevalent crystal structure with good conductivity [9].The molecules of F4-TCNQ and P3HT blend form the charge-transfer complexes by F4-TCNQ molecules accepting electrons from the P3HT molecules [10]. The charge-transfer complexes provide the strong electrostatic forces between P3HT and F4-TCNQ organics by accelerating molecular orientation [11]. Consequently, the conductivity of polymers can be enhanced by adding the F4-TCNQ dopant molecules into the polymer blend, which may improve the electrical properties of MPS SBDs as discussed in previous work [12].

The electrical parameters, which are interface states (N_{ss}),series resistance (R_s), barrier height (ϕ_{BO} and ϕ_B),diffusion potential (V_D), doping concentration of donors (N_D), depletion layer width (W_D),are important to determine the performance of MPS structures. Especially, N_{ss} and R_s are the crucial parameters on the device performance since the applied voltage is shared between R_s, depletion layer and interface layer[13]. These parameters can be deduced from capacitance-voltage (C-V) and conductivity-voltage (G/w-V) measurements and have significant importance in order to produce reliable devices with good qualities.

In this work, firstly, the effects of F4-TCNQ concentration on the electrical characterization of the Au/P3HT:F4-TCNQ/n-Si MPS SBD's were investigated. For this aim, the Au/P3HT:F4-TCNQ/n-Si MPS SBD's with the F4-TCNQ concentration value of 0.5% and 2%were fabricated, and then, the values ofN_{ss} and R_s were analyzed to determine the concentration effects investigated by using C-V and G/w-V measurements. The results show that increasing the F4-TCNQ concentration improves the Au/P3HT:F4-TCNQ/n-Si diode quality by increasing shunt resistance and reducing capacitance, series resistance and N_{ss}. By considering this conclusion, the deeper C-V and G/w-V analysis that give the electrical parameters such as diffusion potential (V_D), Fermi energy level (E_F), depletion layer width (W_D), barrier height (ϕ_B), density of donor atoms (N_D), maximum electrical field (E_m), Schottky barrier lowering ($\Delta\phi_B$) were studied only for 2% F4-TCNQ used Au/P3HT:F4-TCNQ/n-Si MPS SBD.

II. Experimental

P3HT (poly(3-hexylthiophene)) and F4-TCNQ ((2,3,5,6-Tetrafluoro-2,5-cyclohexadiene-1,4-diylidene) dimalononitrile, 7,7,8,8-Tetracyano-2,3,5,6-tetrafluoroquinodimethane) organic materials were purchased from Sigma-Aldrich Ltd. The mixtures of P3HT:F4-TCNQ with 0.5%and 2%F4-TCNQ concentrations were prepared by dissolving in 1-2 dichlorobenzene with a concentration of 10g/L at 60^oC and then the solution stirred 3 hours at room temperature.

The Au/P3HT:F4-TCNQ/n-Si MPS SBDs were fabricated on n-type single silicon wafers with <100> orientation, 4.8Ω-cm resistivity and 350±25 μm thickness. For the fabrication process, n-type Si wafers were cleaned chemically by RCA cleaning procedure with respectively H_2O-H_2O_2-NH_4OH (5:1:1), H_2O-HF (50:1) and H_2O-H_2O_2-HCl (6:1:1) solutions at 80^oC. After cleaning steps, silver with a thickness of ~2500Å was evaporated onto whole back side of Si wafers by using thermal evaporation method. For low resistivity ohmic contact, Si wafers annealed for 30min at 450^oC in N_2 atmosphere. Ohmic contact formation was followed by the cleaning of n-type Si wafers in 50% HF solution to prepare the front side of Si wafer for polymer coating. Immediately, P3HT:F4-TCNQ solutions were spin-coated on the front surface of Si wafers with 1500rpm/s acceleration for 30s to form ~100nm thick organic film. After that, the samples were dried on the hot plate for 15min at 150^oC.

Finally, high purity (99.999%) circular-shaped Au rectifying contacts with 2500Å thickness and 1mm diameter were deposited onto the P3HT:F4-TCNQ surface of the wafers through a shadow mask. Fig.1 shows the Au/P3HT:F4-TCNQ/n-Si diode structure. Afterwards, the capacitance-voltage (C-V) and conductance-voltage (G/w-V) measurements were performed by using HP 4192 A LF impedance analyzer (10kHz-2MHz) at room temperature. The C-V and G/w-V measurements were carried out with computer through IEEE-488 AC/DC converter card.

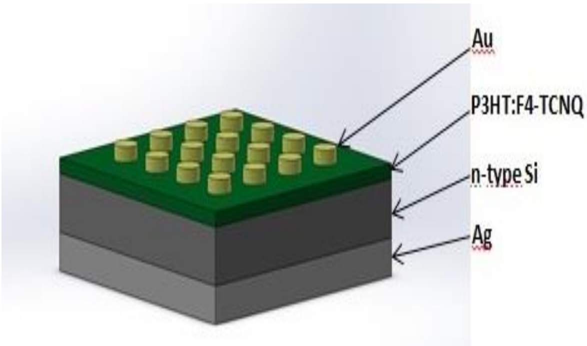

Figure 1. Au/P3HT:F4-TCNQ/n-Si diode structure.

III. Results And Discussion

Surface morphologies of MPS SBDs with different F4-TCNQ concentration were investigated by Quanta FEG Scanning Electron Microscope (SEM). The operating distance and operating voltage of SEM were set to 4.8mm and 10kV, respectively. To decrease the resistivity during SEM measurements, P3HT:F4-TCNQ surfaces of the diodes were coated by the gold layer prior to load the samples into SEM chamber.F4-TCNQ concentration dependent SEM images of the P3HT:F4-TCNQ surfaces were shown in Fig. 2(a) and 2(b) with the scale of 5 μm under the 10kV acceleration voltage. In these figures, the grey areas are the P3HT polymer layer [14] and bright clusters are the F4-TCNQ dopant about 200nm diameter [15]. As shown in Fig. 2(a), only one F4-TCNQ portion was detected by using 0.5% F4-TCNQ in P3HT:F4-TCNQ organic blend for the scale of

5 μm in the SEM image. However in Fig. 2(b), 3 portions of the F4-TCNQ were detected for the 2% F4-TCNQ concentration used MPS diode by the same SEM image scale. The difference of the SEM images for 0.5% and 2% F4-TCNQ concentration used MPS diodes is compatible with the literature [15,16]. Additionally, P3HT:F4-TCNQ/n-Si structure with 2% F4-TCNQ concentration was monitored as cross-sectional by SEM to measure the thickness of P3HT:F4-TCNQ polymer layer as shown in Fig. 3.By using the cross-sectional SEM image, the thickness of P3HT:F4-TCNQ polymer layer was found about 100nm.

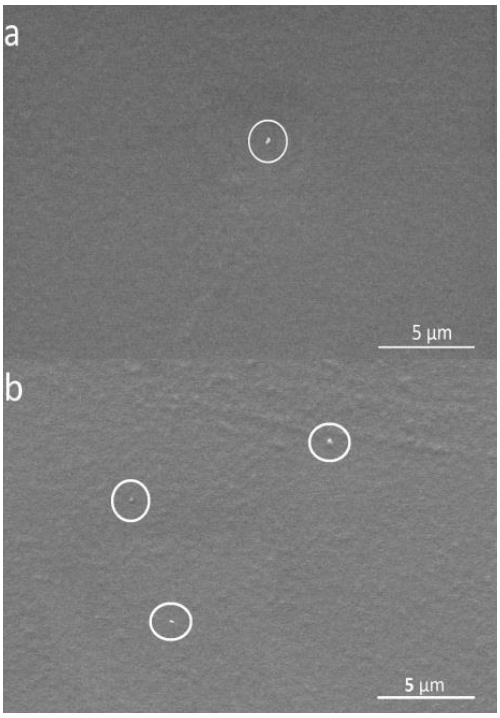

Figure 2.SEM images of (a) 0.5% F4-TCNQ and (b) 2% F4-TCNQ concentration used P3HT:F4-TCNQ surfaces.

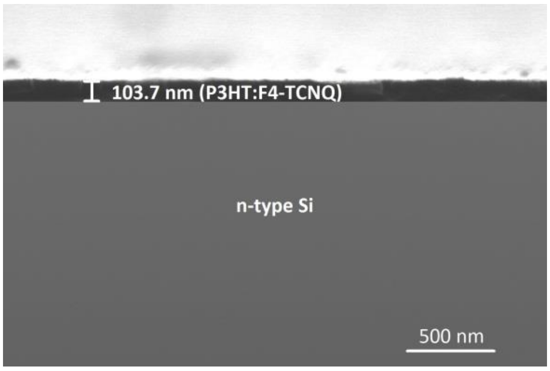

Figure 3. Cross-sectional SEM image of P3HT:F4-TCNQ/n-Si structure with 2% F4-TCNQ concentration.

The behaviors of C, N_{ss} and R_s characteristics as a dependent on F4-TCNQ concentration (0.5% and 2%) for the Au/P3HT:F4-TCNQ/n-Si SBDs were investigated at room temperature at 1MHzfrequencyand in the voltage range between -10V and +10V.The comparative curves of C-V analysis are given in Fig. 4 for the 0.5% and 2% F4-TCNQ used Au/P3HT:F4-TCNQ/n-Si MPS SBDs. As shown in figure, F4-TCNQ concentration had a great effect on the C-V profile of Au/P3HT:F4-TCNQ/n-Si MPS SBDs through the forward bias. The capacitance value decreased with the increasing of F4-TCNQ concentration in the Au/P3HT:F4-TCNQ/n-Si SBD's. This can be due to the enhancement in the passivation of the interface states between semiconductor and organic polymer in the MPS structure by increasing the F4-TCNQ dopant molecules, i.e. F4-TCNQ concentration. By increasing the F4-TCNQ concentration in the P3HT:F4-TCNQ organic blend, more trap states may be disappeared by resulting the reduction in the number of charges that were contributed the conductivity. Therefore, these disappeared charges might give rise to lower capacitance value for 2% F4-TCNQ concentration used MPS SBD.

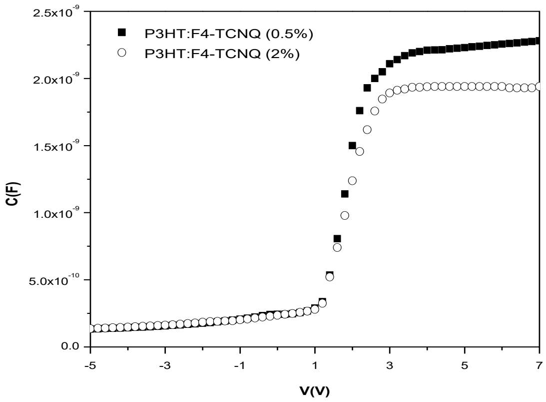

Figure 4. The F4-TCNQ concentration dependent capacitance-voltage characteristic for Au/P3HT:F4-TCNQ/n-Si MPS SBDs at 1MHz.

The electrical characteristics of the SBDs are affected by diode resistance (R_i) which includes series resistance (R_s) and shunt resistance (R_{sh}) and is important to determine the performance of the SBDs. R_i can be calculated by using the following equation [17]:

$$R_i = \frac{G_m}{G_m^2+(wC_m)^2} \quad (1)$$

where G_m and C_m are the conductance and capacitance values and w is the angular frequency. In ideal diodes, the value of R_s should be low (~0) whereas R_{sh} is quite high. The values of R_s and R_{sh} describe the SBD resistance (R_i) in forward bias and reverse bias, respectively. The R_s and R_{sh} values can be determined by using the R_i vs V plot which is shown in Fig. 5for 0.5% and 2% F4-TCNQ used Au/ P3HT:F4-TCNQ/n-Si (MPS) SBDs at 1MHz frequency as a function of voltage.

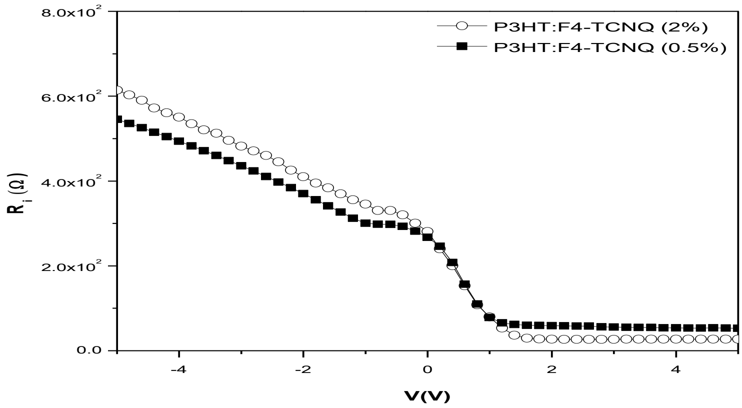

Figure 5. The frequency dependent R_i-V characteristic comparison of 0.5% and 2% F4-TCNQ concentration used Au/P3HT:F4-TCNQ/n-Si SBDs at 1MHz.

By using this figure, R_s value of 2% F4-TCNQ concentration used MPS SBD was found 25.51Ω while 0.5% F4-TCNQ concentration used MPS SBD has the R_s value of 47.59Ω. Moreover, the shunt resistance values of 0.5% and 2% F4-TCNQ concentration used MPS SBDs are 545.38Ω and 613.95Ω at -5V, respectively. These results show that the F4-TCNQ concentration enhancement in the Au/P3HT:F4-TCNQ/n-Si MPS SBD gives rise to more ideal diode with the lower R_s and higher R_{sh}. Finally, it can be concluded that the increasing of F4-TCNQ concentration in the Au/P3HT:F4-TCNQ/n-Si SBD causes the lower capacitance and lower series resistance, which may reduce the N_{ss} values of SBDs [18].

In this work, Hill-Coleman method [19] was used to calculate the N_{ss} values for the both F4-TCNQ concentrations used MPS SBDs. From this method N_{ss} values obtained by following relation:

$$N_{ss} = \frac{2}{qA}\frac{\left(\frac{G_m}{w}\right)_{max}}{[(G_m/w)_{max}C_{ox}]^2+(1-\frac{C_m}{C_{ox}})^2} \quad (2)$$

where C_m and $(G/w)_m$ are the measured capacitance and conductance, respectively, w is the angular frequency (= 2πf), A is the diode area and C_{ox} is the capacitance of polymer layer.

The values of N_{ss} calculated by using Eq. (2) are given in Fig. 6.As shown in this figure, the N_{ss} values of 2% F4-TCNQ concentration used diode are changing from 8.16×10^{12}cm^2/eV to 6.26×10^{11}cm^2/eV between 10kHz and 2MHz while 0.5% F4-TCNQ concentration used diode has the N_{ss} values from 9.46×10^{12}cm^2/eV to

6.90×10^{11}cm^2/eV for the same frequency range. Finally, it can be deduced that increasing the F4-TCNQ concentration in the Au/P3HT:F4-TCNQ/n-Si SBDs reduces the N_{ss} values. This decreasing in the N_{ss} values by the enhancement of the F4-TCNQ concentration in organic interface layer can be explained by the barrier height lowering, increasing the mobility of charges in MPS structure and trap passivation between semiconductor and organic polymer. Additionally, these results show that N_{ss} values strongly dependent on frequency as shown in Fig. 6.

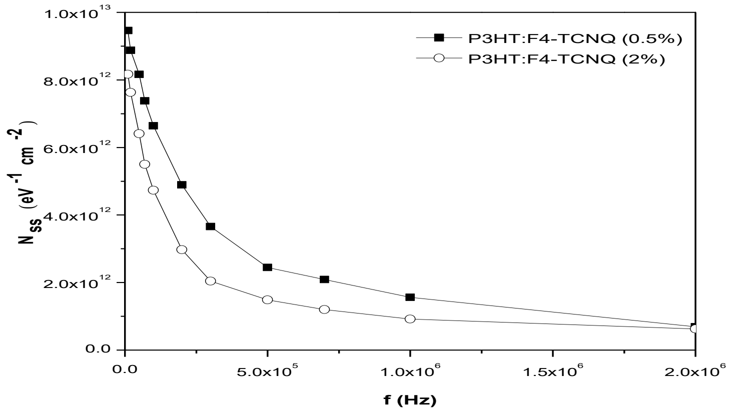

Figure 6. The frequency dependent N_{ss} characteristic for 2% and 0.5% F4-TCNQ concentration used Au/P3HT:F4-TCNQ/n-Si MPS SBDs.

The results obtained so far including the first experimental observations of the different F4-TCNQconcentrations effect on C-V, R_i and N_{ss} analysis show that increasing the F4-TCNQ concentration improves the quality of the Au/P3HT:F4-TCNQ/n-Si SBD. Therefore, the preferable results have been obtained for 2% F4-TCNQ concentration used Au/P3HT:F4-TCNQ/n-Si MPS SBD. For this reason, the following investigations were performed only for 2% F4-TCNQ concentration used SBD which has the higher F4-TCNQ concentration.

The frequency dependent (10kHz-2MHz)C-V and G/w-V characteristics of the Au/P3HT:F4-TCNQ/n-Si SBD were measured at room temperature and are given in Fig. 7 and Fig. 8, respectively. As shown in Fig. 7 and Fig. 8, C and G/w parameters are the function of the frequency and the voltage. Lowering the frequency enhances the capacitance and conductance values, especially at low voltage range (~0V-2V) which is called as accumulation region. The ac signal followed by the charges in interfacial polymer layer gives rise to the frequency dependent capacitance and conductance dispersions. However, when the frequency is increased to the high frequency limits (>500kHz), ac signal cannot be followed easily by the charges. Therefore, capacitance values tend to decrease at high frequencies (>500kHz). Moreover, the C-V and G/w-V plots exhibit the peaks in positive voltage range. These peaks are gradually disappeared as frequency increased from 500kHz to 2MHz. The possible reasons of these peaks in C-V and G/w-V characteristics may be due to the presence of the deep states in the band gap, series resistance and charges in the interfacial polymer layer [20,21].

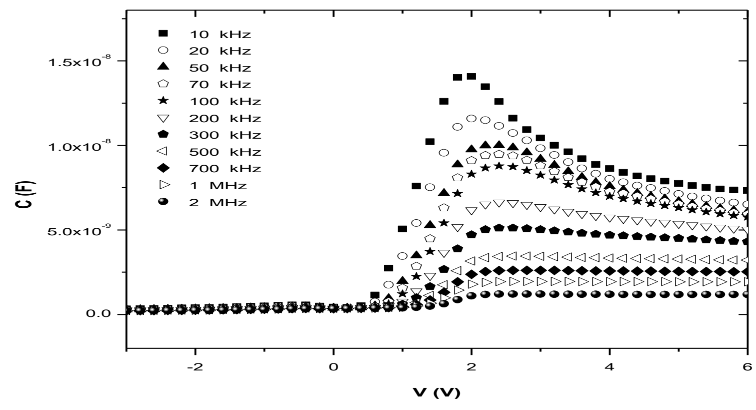

Figure 7. The frequency dependent capacitance-voltage characteristic of 2% F4-TCNQ used Au/P3HT:F4-TCNQ/n-Si SBD.

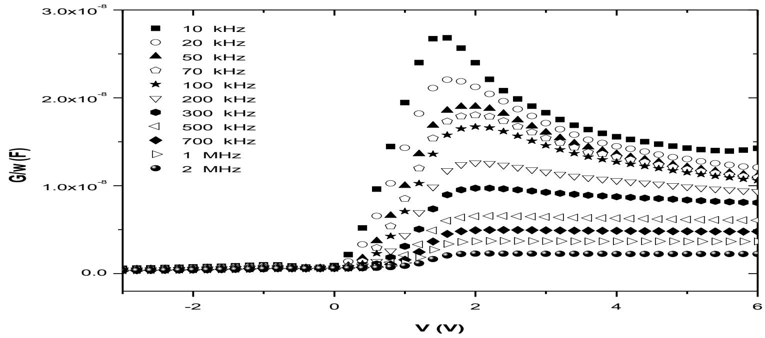

Figure 8. The frequency dependent conductance-voltage characteristic of 2% F4-TCNQ used Au/P3HT:F4-TCNQ/n-Si SBD.

As depicted in Fig. 9, R_i values give the quite distinct peaks in small voltage range(-1V – 1V), which are strongly dependent on frequency. However, these peaks decrease by increasing the frequency. The behavior of $R_i$can be attributed to the localized density of the interface states between semiconductor and polymer interfacial layer [22-24].Moreover, R_s values are almost constant at high frequency region where R_s values are independent from the effects of N_{ss}.

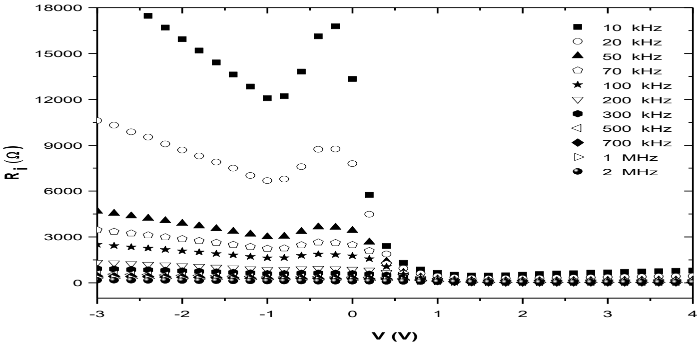

Figure 9. The characteristics of diode resistance (R_i) for various frequencies of the Au/P3HT:F4-TCNQ/n-Si Schottky barrier diode at room temperature.

For detailed C-V analysis of 2% F4-TCNQ concentration used Au/P3HT:F4-TCNQ/n-Si MPS SBDs, C^{-2}-V was obtained by using following relation [24-27]:

$$C^{-2} = \frac{2(V_R+V_o)}{q\varepsilon_s N_D A^2} \tag{3}$$

In Eq. (3), $V_R$is the reverse bias voltage, V_0 is the built in voltage at zero bias, q is the electronic charge, ε_s is the dielectric constant, N_D is the concentration of donor atoms and A is the area of the diode. Fig. 10 shows the C^{-2}-V plots of Au/P3HT:F4-TCNQ/n-Si MPS structure. By using this figure, Fermi energy level (E_F), depletion layer width (W_D) and doping concentration (N_D) were calculated for whole frequency values by following equations [27]:

$$V_o = V_D - \frac{kT}{q} \tag{4}$$

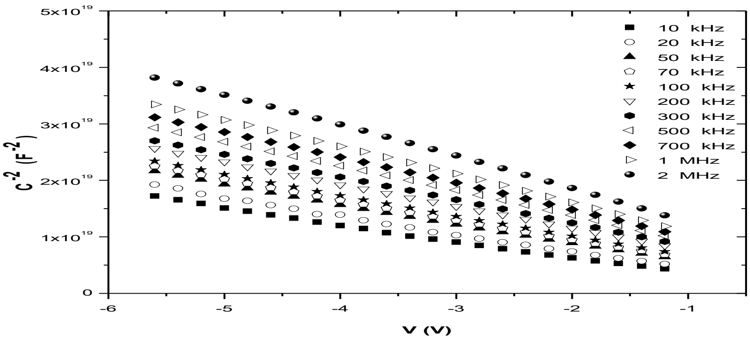

Figure 10. The C^{-2}-V plots of the Au/P3HT:F4-TCNQ/n-Si Schottky barrier diode at room temperature as a function of voltage

where$V_D$is the diffusion potential of zero bias. E_F was calculated by [26]

$$E_F = \frac{kT}{q}\ln\left(\frac{N_C}{N_D}\right) \tag{5}$$

with,

$$N_C = 4.82 \times 10^{15} T^{3/2} \left(\frac{m^*_e}{m_o}\right)^{3/2} \tag{6}$$

where N_C is the effective density of states in Si conductance band, m_e^* is the effective mass of electron, m_0 is the rest mass of the electron and kT/q is the thermal energy [28]. Additionally,ϕ_B(C-V) barrier height can be calculated from C^{-2}-V plot by following relation:

$$\phi_B(C-V) = V_D + E_F - \Delta\phi_B \tag{7}$$

where$\Delta\phi_B$ is the image force barrier lowering; and it can be calculated by [23,26]

$$\Delta\phi_B = \left[\frac{qE_m}{4\pi\varepsilon_s\varepsilon_o}\right]^{1/2} \tag{8}$$

In Eq. (8), E_m is the electrical field and calculated by following relation [27]:

$$E_m = \left[\frac{2qN_DV_D}{\varepsilon_s\varepsilon_o}\right]^{1/2} \tag{9}$$

By using these equations, the values of N_D, V_D,E_F, ϕ_B, $\Delta\phi_B$, R_s and N_{ss} in the frequency range between 10kHz and 2MHz were calculated and given in Table 1.According to Table 1, N_{ss} and R_s values decrease by increasing the frequency. The reason of this behavior is that ac signal can not be followed easily by the charges located in the

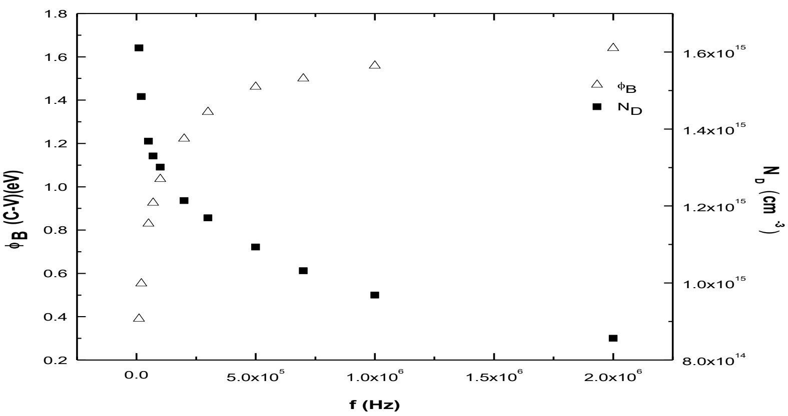

Figure 11. Theϕ_B(C-V)and N_D parameters for various frequencies for the Au/P3HT:F4-TCNQ/n-Si Schottky barrier diode

interface states. Therefore, at high frequency region, R_s values become independent from frequency and N_{ss} values decrease. Moreover, reduction in N_D values and enhancement in ϕ_B (C-V) values were obtained by increasing the frequency (Fig. 11). This expected behavior can be due to the presence of interface states and organic interfacial layer [29-30]. Fig. 12 shows the variation of frequency dependent R_s and N_{ss} values for the Au/P3HT:F4-TCNQ/n-Si SBD. As shown in this figure, values of R_s and N_{ss} tend to decrease by increasing the frequency. This same behavior dependent on the frequency shows the correlation between the R_s and the N_{ss}.

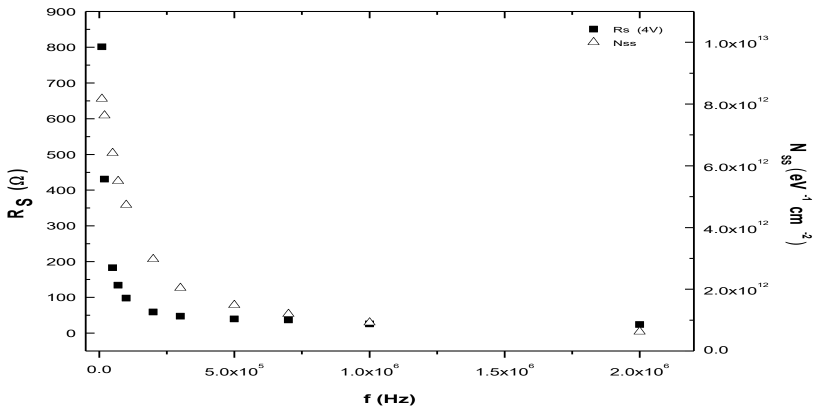

Figure 12. The variation of N_{ss} and R_s for various frequencies for the Au/P3HT:F4-TCNQ/n-Si Schottky barrier diode

Table 1. The obtained N_{ss}, R_s, V_D, N_D, W_D, ϕ_B, $\Delta\phi_B$ and E_F values for various frequencies.

f (kHz)	N_{ss} ($eV^{-1}cm^{-2}$)	R_s (Ω)	V_D (eV)	N_D (cm^{-3})	W_D (cm)	ϕ_B (eV)	$\Delta\phi_B$ (eV)	E_F (eV)
10	8.16×10^{12}	8.01×10^{2}	0.16	1.61×10^{15}	3.64×10^{-5}	0.39	1.06×10^{-2}	0.227
20	7.63×10^{12}	4.31×10^{2}	0.32	1.48×10^{15}	5.34×10^{-5}	0.55	1.23×10^{-2}	0.229
50	6.40×10^{12}	1.83×10^{2}	0.60	1.37×10^{15}	7.55×10^{-5}	0.83	1.41×10^{-2}	0.231
70	5.50×10^{12}	1.34×10^{2}	0.69	1.33×10^{15}	8.26×10^{-5}	0.93	1.45×10^{-2}	0.231
100	4.73×10^{12}	9.81×10^{1}	0.80	1.30×10^{15}	8.98×10^{-5}	1.04	1.49×10^{-2}	0.232
200	2.97×10^{12}	5.88×10^{1}	0.99	1.21×10^{15}	1.03×10^{-4}	1.22	1.55×10^{-2}	0.234
300	2.04×10^{12}	4.75×10^{1}	1.11	1.17×10^{15}	1.11×10^{-4}	1.34	1.58×10^{-2}	0.235
500	1.48×10^{12}	3.96×10^{1}	1.22	1.09×10^{15}	1.21×10^{-4}	1.46	$1,59\times10^{-2}$	0.237
700	1.19×10^{12}	3.66×10^{1}	1.26	1.03×10^{15}	1.26×10^{-4}	1.50	1.58×10^{-2}	0.238
1000	9.19×10^{11}	2.55×10^{1}	1.32	9.68×10^{14}	1.33×10^{-4}	1.56	1.57×10^{-2}	0.240
2000	6.26×10^{11}	2.38×10^{1}	1.40	8.56×10^{14}	1.46×10^{-4}	1.64	1.54×10^{-2}	0.243

IV. Conclusion

The C-V and G/w-V characteristics of 0.5% and 2% F4-TCNQ concentration used Au/P3HT:F4-TCNQ/n-Si SBDs have been investigated at room temperature in the frequency range of 10kHz-2MHz.

The electrical performance of 0.5% and 2% F4-TCNQ concentration used Au/P3HT:F4-TCNQ/n-Si SBDs were compared in terms of C-V analysis in the voltage range between -10V and +10V and at the frequency of 1MHz. According the experimental results, 2% F4-TCNQ concentration used diode has the lower capacitance values than those of 0.5% F4-TCNQ concentration used diode. The reduction of capacitance values can be due to the enhancement in the passivation of trap states by increasing the F4-TCNQ dopant concentration. Moreover, increasing the F4-TCNQ concentration results the lower R_s and higher R_{sh}, which shows the more ideal diode behavior. On the other hand, frequency dependent N_{ss} values were compared for 0.5% and 2% F4-TCNQ concentration used Au/P3HT:F4-TCNQ/n-Si SBDs. The increasing of F4-TCNQ concentration reduces the N_{ss} values which can be due to the barrier height lowering, increasing the mobility of charges in MPS structure and trap passivation between semiconductor and organic polymer.

The experimental results show that increasing of F4-TCNQ concentration improves the Au/P3HT:F4-TCNQ/n-Si diode quality. Therefore, the preferable electrical results have been obtained for 2% F4-TCNQ concentration used diode. For this reason, the detailed C-V and G/w-V analysis, which consists of R_s, N_{ss}, V_D, N_D, W_D, ϕ_B, E_F, E_m and $\Delta\phi_B$ calculations, were performed only for 2% F4-TCNQ concentration used Au/P3HT:F4-TCNQ/n-Si MPS SBD in the frequency range of 10kHz-2MHz.

The capacitance and conductance values of Au/P3HT:F4-TCNQ/n-Si MPS SBD strongly dependent on the voltage and frequency regions. The dispersion of the capacitance and conductance values dependent on frequency can be attributed to the presence of R_s, interfacial layer between semiconductor and polymer and the charges in the interfacial polymer layer.

As similar to C and G/w, R_s varies by changing the voltage and frequency. The decreasing of R_s values was obtained by increasing the frequency. However, in the high frequency region (>500kHz), R_s values become independent from the frequency region where N_{ss} has no effect on the R_s.

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