New Materials, Compounds and Applications
Vol.6, No.1, 2022, pp.27-36

ANALYSIS OF INTERFACE TRAPS OF Au/$C_{25}H_{25}BF_2N_2O$/n-Si SCHOTTKY DIODES BY HILL-COLEMAN TECHNIQUE

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Abstract. Abstract. In this work, the $C_{25}H_{25}BF_2N_2O$ organic thin film was grown on n-Si wafer by the spin coating method. Au/$C_{25}H_{25}BF_2N_2O$/n-Si Schottky diodes were prepared, and their series resistance (R_s) and interface trap (N_{it}) properties were introduced by capacitance-conductance-voltage ($C-G-V$) in the frequency range from 30 kHz–1 MHz at room temperature. Voltage dependent profile of R_s was calculated from C and G characteristics according to Nicollian and Goetzberger technique. The series resistance impact was removed from the measured capacitance and conductance values to determine the correct capacitance and conductance values of the diode. The $G_c - V$ profile has a peak for the measured frequencies between 0 V and 0.2 V due to the interface traps (N_{it}) at n-Si/$C_{25}H_{25}BF_2N_2O$ interface. The magnitude of peak increases with increasing frequency and shift towards positive voltage. By using these peak values, the frequency dependent variation of the interface traps was determined with the help of the Hill-Coleman technique. It was observed that the interface traps are decreased with increasing the frequency.

Keywords: *$C_{25}H_{25}BF_2N_2O$, Schottky diode, series resistance, interface traps.*

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Received: *2 November 2021;* ***Accepted:*** *23 Janury 2022;* ***Published:*** *19 April 2022.*

1. Introduction

Organic thin films are very important materials due to their use in light emitting diode, solar cells, and Schottky devices (Akın *et al.*, 2019; Aruna & Joseph, 2017; Eymur *et al.*; Yu *et al.*, 2019; Zhang *et al.*, 2019). As reported in many works conducted in recent years, the interface traps at the metal-semiconductor interface can be modified by using a suitable organic layer between the semiconductor and the metal. Investigation of the interfacial properties of Schottky type diodes with organic semiconductors is so important for their further improve in performance.

To increase the electronic performance of devices based on Si–based metal-semiconductor structure with organic interfacial layer, it is essential to study N_{it} between metal and semiconductor and R_S of the structure. There are many techniques to obtain the values of N_{it} and R_S from admittance measurements (Eymur & Tuğluoğlu, 2020; Goetzberger & Sze, 1969; Hill & Coleman, 1980; Nicollian, 2003; Sze, 2007). These techniques are known as Nicollian-Goetzberger and Hill-Coleman techniques, respectively (Goetzberger & Sze, 1969; Hill & Coleman, 1980; Nicollian, 2003). According to Nicollian and Goetzberger technique, the capacitance and conductance measurements based on voltage under reverse and forward biases give major information about series resistance and interface traps. While the effect of R_s is more effective at strong accumulation area at high frequencies, the effect of N_{it} is effective in the depletion region at low frequencies.

In recent years, researchers have focused on improving the electrical properties, reliability, quality, and performance of Schottky diodes with different interfacial organic layers. Eymur et al. prepared Au/CuPc/n-Si Schottky diodes and they reported the electrical characteristics of the photodiodes by current-voltage ($I-V$) and capacitance-voltage ($C-V$) measurements (Eymur *et al.*, 2020). İmer et al. prepared Au/Sunset Yellow/n-Si Schottky diodes and they introduced the illumination impact on the electrical characteristics of the Schottky diode (Imer *et al.*, 2019). Akın et al. fabricated Al/Coronene/n-Si structure for optoelectronic applications. Photovoltaic parameters of the fabricated diode have been determined at various illumination intensities (Akın, Yüksel *et al.*, 2019). Meftah et al. have investigated electrical behavior of Al/P3HT/p-Si Schottky diode using $C-V$ and $I-V$ characteristics (Meftah *et al.*, 2020).

BODIPY (4,4-difluoro-4-bora-3a, 4a-diaza-s-indacene) is known as one of the popular dyes that are frequently used in the literature. Due to its properties such as high absorption coefficient, fluorescent quantum efficiency, p-conjugate structure and long excited state, BODIPY-based organic semiconductor structures are highly suitable for various applications such as dye-sensitized solar cells, fluorescent molecular probes and optoelectronics (Kaplan *et al.*, 2021; Poddar & Misra, 2020; Sahin *et al.*, 2021; Sevgili *et al.*, 2021). For instance, Kılıcoglu and Ocak studied the ideality factor and barrier height of the Phenyl-BODIPY/n-Si diode by current-voltage measurements (Kılıçoğlu & Ocak, 2011). Ozcan et al. fabricated the Al/Subphthalocyanine-BODIPY dyads/p-Si/Al diodes and electrical properties of these diodes was studied at various illumination intensities (Özcan *et al.*, 2018). The synthesis of BOD-Z-EN ($C_{25}H_{25}BF_2N_2O$) compound to fabricate Au/BOD-Z-EN/n-Si diode was previously reported by us (Tezcan *et al.*, 2021). We investigated electrical parameters of Au/BOD-Z-EN/n-Si/In diode such as ideality factor, barrier height and series resistance by current-voltage measurements at dark and under illumination conditions.

As far as we know, there are no capacitance, conductance, interface trap and series resistance profile results on Au/$C_{25}H_{25}BF_2N_2O$/n-Si Schottky diode yet. The aim of this work is to explore the changes in R_s and N_{it} of these structures with voltage and frequency at room temperature, respectively. The series resistance and interface trap properties of Au/$C_{25}H_{25}BF_2N_2O$/n-Si Schottky diode was studied for the first time from $C-V$ and $G-V$ characteristics in the frequency range from 30 kHz to 1 MHz. We selected the methods introduced by Hill-Coleman and Nicollian-Goetzberger, which is considered to be the most suitable method for determining the N_{it} and R_s, respectively (Goetzberger & Sze, 1969; Hill & Coleman, 1980; Nicollian, 2003).

2. Experimental

$C_{25}H_{25}BF_2N_2O$ compound was synthesized according to published procedure (Tezcan, Eymur *et al.*, 2021; Ucuncu *et al.*, 2016). Au/$C_{25}H_{25}BF_2N_2O$/n-Si Schottky structure was fabricated on n type Si crystal with 20 Ω cm resistivity, 500 μm thickness, (100) orientation, and grown by CZ (Czochralski) technique. The silicon crystal was first washed in trichloroethylene, methanol and acetone, respectively. As a second process, the crystal was cleaned using the RCA process (Akın, Yüksel *et al.*, 2019). In the third process, indium metal (99.999%) in a 150 nm thick was deposited in the thermal evaporation system at 10^{-5} Torr on the non-shiny side of the crystal. The crystal is placed on a tungsten plate (with the aluminum coated surface facing up) in the thermal evaporation system. After a period of voltage applied to the plate, the indium-coated surface deposited into the silicon and ohmic contact was formed.

The solution was prepared using 5 mg of $C_{25}H_{25}BF_2N_2O$ and 1 ml of chloroform. The prepared solution was deposited on the shiny surface of the crystal by operating the Laurell spin coating system at 500 rpm for 30 seconds and then at 1200 rpm for 60 seconds. Then, rectifying (Schottky) contacts were created using the mask with 2 (two) mm diameter holes. The crystal layer (with the metal uncoated surface down) was placed on the mask and placed in the sample holders of the thermal evaporation system. When the pressure reached 1 x 10^{-5} Torr in the evaporation system, 150 nm thick gold (Au, 99.99% purity) metal was deposited and a rectifying contact was formed. As a result, Au/$C_{25}H_{25}BF_2N_2O$/n-Si/In Schottky diode structure was produced. HP Agilent E4990A (20 Hz to 10 MHz) Impedance Analyzer for capacitance-voltage $(C - V)$ and conductivity-voltage $(G - V)$ measurements of the fabricated Au/$C_{25}H_{25}BF_2N_2O$/n-Si/In Schottky diodes in the dark, at room temperature and in the frequency range of 30 kHz-1 MHz was used.

3. Results and Discussion

Fig. 1 depicts the values of capacitance (C) depend on voltage (V) of the Au/$C_{25}H_{25}BF_2N_2O$/n-Si Schottky diode in the voltage range from -4 V to 4 V and between 30 kHz and 1 MHz frequency. It is observed from Fig 1. that the curves have the inversion, depletion and accumulation areas for all frequency. The curves show a peak and the peak position is shifted towards zero voltage with increasing frequency. This shifting is referred to the existence of interface traps following A.C. signal at low frequencies (Alialy *et al.*, 2014; Bilkan *et al.*, 2015; Werner *et al.*, 1988).

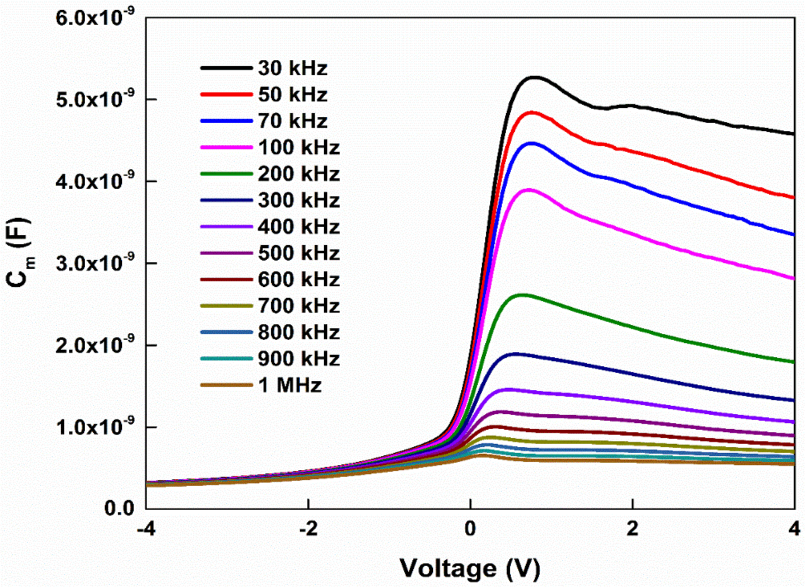

Figure 1. Variation of the measured capacitance (C_m) with voltage (V) for Au/$C_{25}H_{25}BF_2N_2O$/n-Si Schottky diode at various frequencies

Furthermore, the values of peak and capacitance have increased with decreasing frequency. Therefore, the increase in charge density increases capacitance value and correct capacitance creates from capacitance of the space charge region and interface layer.

The measured conductance-voltage ($G_m - V$) curves for Au/$C_{25}H_{25}BF_2N_2O$/n-Si Schottky diode are illustrated in Fig. 2. It can be seen from Fig. 2 that all conductance curves show three areas as in capacitance curves. Furthermore, the curves show a peak and the peak position is shifted towards zero voltage with increasing frequency. Moreover, as seen in Fig. 2, the values of conductance has influenced by the charges at interface traps and as a result of this, the value of G increased with increasing both voltage and frequency at the forward and reverse bias voltage region.

As a result, capacitance values increase as voltage increases and decrease as frequency increases. On the other hand, conductivity values increase as both voltage and frequency increase. However, from Fig. 1 and 2, C and G values are highly frequency dependent, and it can be seen that the effect of frequency is more especially in the accumulation and depletion areas.

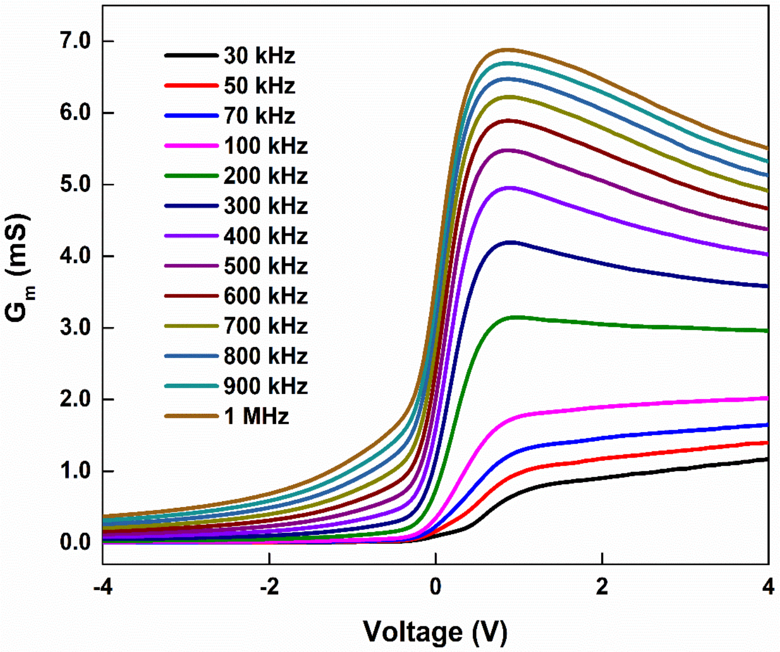

Figure 2. Variation of the measured conductance (G_m) with voltage (V) for Au/$C_{25}H_{25}BF_2N_2O$/n-Si Schottky diode at various frequencies

In the inversion area (at sufficiently small negative voltages), there is no significant change in C and G values and the voltage has almost no effect on the C and G values. However, the highest value of the capacitance values shifts to negative voltages as the frequency decreases. The same situation occurs as the frequency increases in conductivity values. The capacitance and conductance values depend on various factors such as the thickness and formation of the interfacial layer, the density of the interfacial states, and the series resistance (Castagne & Vapaille, 1971; Nicollian, 2003). The effect of interface state density can be eliminated when the $C - V$ and $G - V$ curves are measured at sufficiently high frequency, because charges in the interface states cannot follow an ac signal (Castagne & Vapaille, 1971; Sharma & Tripathi, 2016; Zeyrek *et al.*, 2013). In this case, the interface states are in equilibrium with the semiconductor (Sharma & Tripathi, 2016; Tugluoglu, 2007; Tugluoglu *et al.*, 2013).

Similar behavior has been reported in the literature for organic/inorganic Schottky contacts. Tuğluoğlu et al. prepared an Au/perylene-monoimide/n-Si Schottky diode and took $C-V$ and $G-V$ measurements at different frequencies (Tugluoglu, Yuksel *et al.*, 2013). They observed that the C and G values for the sample they prepared depend on both voltage and frequency, and the changes occurred especially in the depletion and accumulation areas. Tozlu and Mutlu investigated the effect of polymer interface on interface states with the help of frequency dependent capacitance and conductance measurements in Au/PVP:PMF/p-Si Schottky diodes (Tozlu & Mutlu, 2016). They emphasized that the conductance values for the Au/PVP:PMF/p-Si Schottky diode change with both voltage and frequency, which is due to the interface states in the energy band gap.

The series resistance (R_s) of the Au/$C_{25}H_{25}BF_2N_2O$/n-Si/In Schottky diode is derived from the capacitance and conductivity measurements measured in the condensation region using the Nicollian and Brews method (Nicollian, 2003). R_s is an important factor that causes deviations in the ideal behavior of the measured capacitance (C_m) and conductivity (G_m) properties of the diode. The series resistance of Schottky diode is given as (Kavasoglu *et al.*, 2009; Nicollian, 2003; Tugluoglu, 2007) (a,b,c):

$$R_s = \frac{G_m}{G_m^2+\omega^2 C_m^2} \ , \qquad (1)$$

where C_m and G_m are the capacitance and conductance measured in the accumulation area, respectively.

Fig. 3. depicts the voltage dependence of R_s at different frequencies for the Au/$C_{25}H_{25}BF_2N_2O$/n-Si/ Schottky diode. A peak value was observed at all frequencies. R_s values decreased as the frequency increased. The R_s values of the diode are calculated in the depletion region (0.5 V). The calculated R_s values were obtained as 313 Ω and 112 Ω for 30 kHz and 1 MHz, respectively.

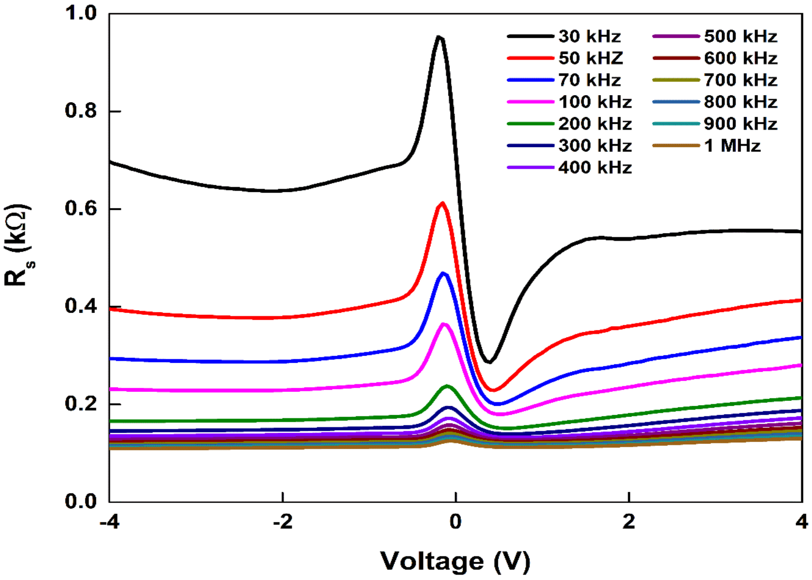

Figure 3. Variation of the calculated series resistance (R_s) with voltage (V) for Au/$C_{25}H_{25}BF_2N_2O$/n-Si Schottky diode at various frequencies

The series resistance, which decreases with increasing frequency, is attributed to the escape of trapped charge carriers with sufficient energy at high frequency from the trap levels between the insulator and semiconductor interface (Kavasoglu *et al.*, 2009; Nicollian, 2003; Tugluoglu, 2007). As can be seen in Fig. 3, the series resistance gives a high peak at low frequency values. The interface traps are dense at voltages where a peak is present. If the variation of the series resistance with the bias voltage gives a peak, this behavior indicates a special distribution of interface traps (Kavasoglu *et al.*, 2009; Nicollian, 2003; Tugluoglu, 2007).

The calculated series resistance values are used to obtain the correct values of the capacitance and conductance. The values of corrected capacitance (C_c) and corrected conductance (G_c) were determined by the following relations (Kavasoglu *et al.*, 2009; Nicollian, 2003; Tugluoglu, 2007) (h):

$$a = G_m - (G_m^2 + \omega^2 C_m^2) R_s \tag{2}$$

$$C_c = \frac{(G_m^2+\omega^2 C_m^2) C_m}{a^2+\omega^2 C_m^2} \tag{3}$$

$$G_c = \frac{(G_m^2+\omega^2 C_m^2) a}{a^2+\omega^2 C_m^2} \tag{4}$$

Fig. 4 and 5 depict the $C_c - V$ and $G_c - V$ curves between 30 kHz and 1 MHz, respectively. As given in Fig. 4 and 5, the values of G_c are increased, while the C_c values are decreased with increasing frequency. The peaks that occur in the consumption region in the conductivity curves are due to the concentration of the interface states in the forbidden energy range in a special region (Kavasoglu *et al.*, 2009; Nicollian, 2003; Tugluoglu, 2007).

This behavior in both $C_c - V$ and $G_c - V$ curves shows that the R_s effect is very important and the measured $C_m - V$ and $G_m - V$ curves must be corrected by considering the effect of this R_s value. If the R_s effect is not eliminated, the reliability and accuracy of the obtained parameters will decrease.

There are many methods in the literature to determine the interface state density in Schottky structures (Haddara & Elsayed, 1988; Hill & Coleman, 1980). The distribution profile of the interface traps was obtained using the Hill-Coleman method at different frequencies. According to this method, if the corrected conductance values in the depletion region of the Schottky structure give a peak, these peaks indicate the existence of interfacial traps occurring at the organic/inorganic interface. After subtracting the series resistance effect from the conductance values measured for the Au/$C_{25}H_{25}BF_2N_2O$/n-Si Schottky diode, the corrected conductance values showed peaks at all frequencies in the depletion region. According to Hill-Coleman, the interface trap (N_{it}) values can be calculated from the corrected capacitance and conductivity values from the following equation.

$$N_{it} = \frac{2}{qA} \frac{(G_{c,max}/\omega)}{\left(G_{c,max}/\omega C_{ox}\right)^2 + (1 - C_c/C_{ox})^2} \tag{5}$$

where A is the diode area, q is the electron charge and C_{ox} is the insulating layer capacitance. This layer is the $C_{25}H_{25}BF_2N_2O$ organic layer capacitance in the produced

Schottky diode, and it is obtained from the values in the accumulation region of the corrected capacitance and conductance measurements at high frequency (1 MHz) with the help of the following equation (g).

$$C_{ox} = C_c\left[1 + \frac{G_c^2}{(\omega C_c)^2}\right] = \frac{\varepsilon_{ox}\varepsilon_o}{d_{ox}}A \tag{6}$$

Here ω, ε_{ox}, ε_o, $d_{ox}$and A are angular frequency (=2πf), interface layer permittivity constant, free space permittivity constant, organic layer thickness and diode area, respectively. $C_{25}H_{25}BF_2N_2O$ organic layer thickness was determined as 320 nm from organic capacitance in 1 MHz frequency.

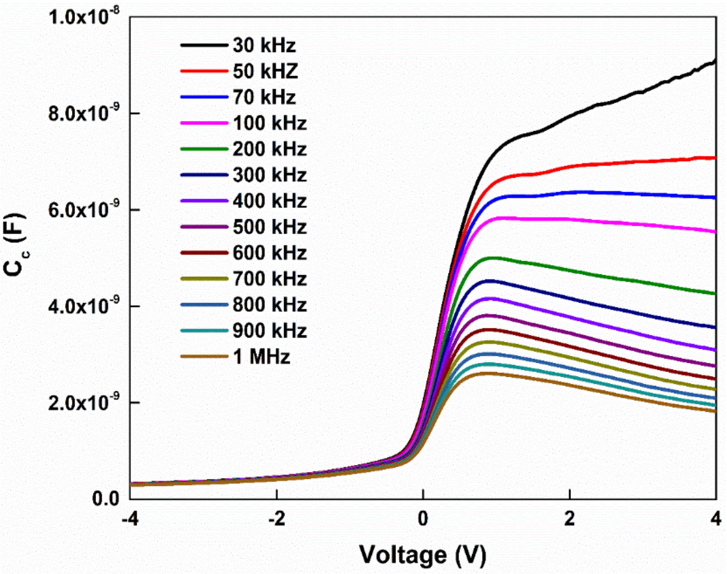

Figure 4. Variation of the corrected capacitance (C_c) with voltage (V) for Au/$C_{25}H_{25}BF_2N_2O$/n-Si Schottky diode at various frequencies

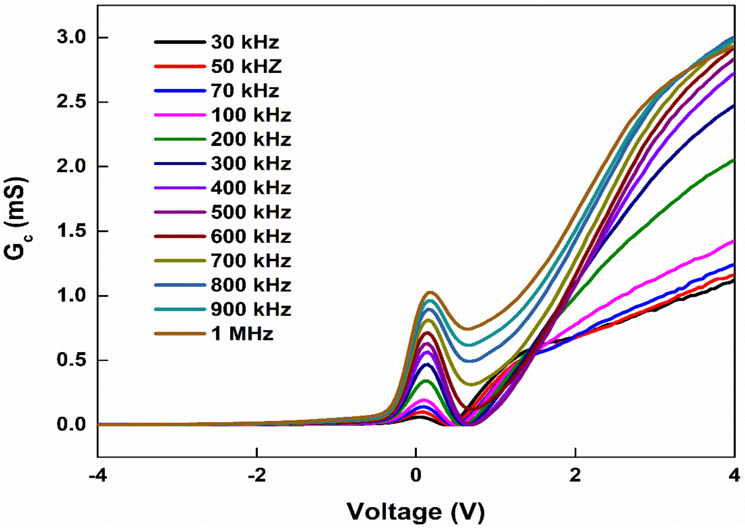

Figure 5. Variation of the corrected conductance (G_c) with voltage (V) for Au/$C_{25}H_{25}BF_2N_2O$/n-Si Schottky diode at various frequencies

The variation of the interface traps (N_{it}) with frequency of the prepared Au/$C_{25}H_{25}BF_2N_2O$/n-Si Schottky diode is illustrated in Fig. 6. N_{it} values were calculated as 3.41 x10^{12} eV^{-1} cm^{-2} and 5.48x10^{11} eV^{-1} cm^{-2} at 30 kHz and 1 MHz frequencies, respectively. These values decreased as the frequency increased. The reason for the decrease is the reduced contribution of carriers at the interface at high frequencies (Goetzberger & Sze, 1969; Haddara & Elsayed, 1988; Nicollian, 2003; Yakuphanoglu, 2010). Similar results have been reported by other researchers (Haddara & Elsayed, 1988; Kavasoglu *et al.*, 2009; Tozlu & Mutlu, 2016; Tugluoglu, 2007; Tugluoglu *et al.*, 2013; Yakuphanoglu, 2010).

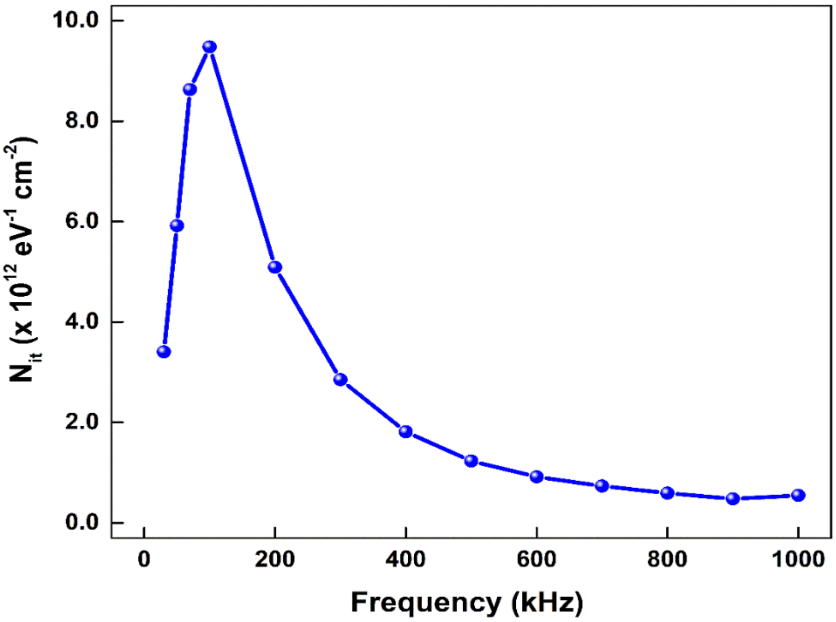

Figure 6. Variation of the interface traps (N_{it}) with frequency for Au/$C_{25}H_{25}BF_2N_2O$/n-Si Schottky diode

4. Conclusion

In this present study, we have prepared Au/$C_{25}H_{25}BF_2N_2O$/n-Si Schottky diode. The capacitance, conductance, series resistance and interface trap properties of the diode were investigated in the frequency range from 30 kHz to 1 MHz and discussed. The spin coating occurred $C_{25}H_{25}BF_2N_2O$ organic layer film was the thickness of 320 nm. The calculated R_s values in the depletion region (0.5 V) were obtained as 313 Ω and 112 Ω for 30 kHz and 1 MHz, respectively. N_{it} values were determined as 3.41 x10^{12} eV^{-1} cm^{-2} and 5.48x10^{11} eV^{-1} cm^{-2} at 30 kHz and 1 MHz frequencies, respectively. It was observed that the values of R_s and N_{it} were decreased with increasing frequency.

In summary, the scope of this study is the analysis of the interface trap from voltage–dependent capacitance and conductance measurements for Au/$C_{25}H_{25}BF_2N_2O$/n-Si Schottky diode over a wide frequency range. It was found that Au/$C_{25}H_{25}BF_2N_2O$/n-Si Schottky diode exhibited a metal-insulator-semiconductor (MIS) type behavior with low interface trap. The results demonstrated that the characteristic properties of Au/$C_{25}H_{25}BF_2N_2O$/n-Si diode can be controlled by interface traps, series resistance, and interfacial layer values.

Acknowledgment

This work was supported by The Management Unit of Scientific Research Project of Giresun University under Project No. FEN-BAP-A-150219-06.

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