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AC electrical conductivity and dielectric properties of Al/NphAOEMA/PEDOT-PSS/ITO structure at different temperatures

R. ŞAHINGÖZ*, H. KANBUR ÇAVUŞ, Ş. ALTINDAL[a]
Department of Physics, Faculty of Arts and Sciences, Bozok University, Yozgat, Turkey
[a]*Department of Physics, Faculty of Arts and Sciences, Gazi University, Ankara, Turkey*

Dielectric properties and ac electrical conductivity (σ_{ac}) of Al/NphAOEMA/PEDOT-PSS/ITO structure were studied in the temperature range of 120-400 K. Experimental results show that dielectric constant (ε'), dielectric loss (ε''), dielectric loss tangent $(\tan\delta)$ and σ_{ac} are strongly both temperature and bias voltage dependent. The values of ε' are found to increase with increasing temperature while the values of ε'' decrease with increasing temperature for each bias voltage. Such behaviour of dielectric properties is dependent on the amount of interfacial polarization with temperature. In addition, we obtained the values of activation energy (E_a) from the Arrhenius plots for four different bias voltages. The high E_a that is observed in the low bias voltage may be due to the sum of the energies required for the generation of the charge carriers and their motion into vacancies.

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1. Introduction

Organic semiconductors have attracted considerable interest in the recent years with the discovery of organic conducting polymers which can be used as active components in electronic devices. These materials have a great potential advantage due to easy processing in low cost manufacturing and large area electronic and optoelectronic devices applications [1-5]. There are a lot of polymers that can be used for the fabrication of microelectronic devices due to their unique electrical, optical and dielectric properties but among them methacrylic copolymers have gained great importance in various industrial fields. Because copolymers containing the naphthyl group with a methacrylic monomers have good thermal stability [5,6].

The performance and reliability of these devices depend on various parameters such as the process of fabrication, the barrier height between metal and polymer, the density of surface states at interface, device temperature and voltage, and the stability of the polymer used. The existence of a polymer (NphAOEMA) between Al/PEDOT-PSS can have a strong influence on the dielectric properties and ac electrical conductivity of structure. There are currently a vast number of reports of experimental studies on metal/polymer structure, but very limited experimental information is available on their temperature dependent dielectric characteristics in the literature [7-9].

In this article, dielectric properties and the ac electrical conductivity of Al/NphAOEMA/ PEDOT-PSS/ITO structure have been studied by C–V and G/w–V measurements technique in the temperature range of 120–400 K at 500 kHz. To determine the dielectric constants and the ac electrical conductivity Al/NphAOEMA/ PEDOT-PSS/ITO structure, the admittance technique was used [10,11].

2. Experimental procedure

A naphthylamino group containing methacrylate based on monomer 2-(2-naphtylamino)-2-oxo-ethyl methacrylate (NphAOEMA) was synthesized by reacting 2-chloro-N-2-naphtylacetamide (NphA) with sodium methacrylate in acetonitrile. (NphA) was prepared by reacting 1-naphthylamine dissolved in benzene with chloroacetylchloride. The free-radical-initiated polymerization of (NphAOEMA) was carried out in dimethylsulphoxide (DMSO) solution at 65^{o} using 2.2'-azobisisobutyronitrile (AIBN) as an initiator. The poly (NphAOEMA) having an amorphous structure was already characterized and described elsewhere [5]. Al/NphAOEMA/PEDOT-PSS/ITO was prepared by spin-coating PEDOT:PSS on the ITO substrate, followed by the poly (NphAOEMA). The thickness of poly (NphAOEMA) was in the range of 70 nm, using a pin-speed of 5000 rpm for 30s. 200 μm aluminium as cathode was evaporated on top of the [poly (NphAOEMA)] layer by using an evaporator form West Technology Systems. Details to the structure of Al/NphAOEMA/PEDOT-PSS/ITO heterojunction structures and details of fabricated procedures have been given in our previous study [5, 6].

3. Results and discussions

The temperature dependence of the real part of complex permittivity ε', and the imaginary part of complex permittivity ε'', tan δ and σ_{ac} have been studied as well. The complex permittivity can be defined in the following complex form [12,13]

$$\varepsilon^* = \varepsilon^{'} - j\varepsilon^{''} \qquad (1)$$

where j is the imaginary root of -1. The complex permittivity formula has been used to describe the electrical and dielectric properties. In the ε^* formulation, in case of admittance measurements (C-V and G/*w*-V), the following relation equations

$$\varepsilon^* = \frac{Y^*}{j\omega C_o} = \frac{C}{C_o} - j\frac{G}{\omega C_o} \qquad (2)$$

where Y^*, C and G are the measured admittance, capacitance and conductance of the dielectric respectively and ω is angular frequency (ω=2πf) of the applied electric field [13]. The dielectric constant (ε'), at various temperatures was calculated by using the measured capacitance values at the strong accumulation region via the formula [7,8]

$$\varepsilon^{'} = \frac{C}{C_o} \qquad (3)$$

where C_o is capacitance of an empty capacitor. $C_o = \varepsilon_o(A/d)$; where A is the rectifier contact area in cm^{-2}, d is the polymer thickness and ε_o is the permittivity of free space charge (ε_o = 8.85x10^{-14} F/cm). The dielectric loss (ε''), at various temperatures was calculated by using the measured conductance values from the below equation;

$$\varepsilon^{''} = \frac{G}{\omega C_o} \qquad (4)$$

The loss tangent (tanδ) can be expressed as follows [8,12,13],

$$\tan\delta = \frac{\varepsilon^{''}}{\varepsilon^{'}} \qquad (5)$$

The ac electrical conductivity (σ_{ac}) of the dielectric materials can be calculated from the dielectric loses according to the relation [7-14],

$$\sigma_{ac} = \omega C \tan\delta (d/A) = \varepsilon^{''}\omega\varepsilon_o \qquad (6)$$

where ω is the angular frequency. The dielectric properties of Al/NphAOEMA/PEDOT-PSS/ITO structure are measured with the HP 4192A LF impedance analyzer. The instrument directly provides the values of capacitance and conductance. As can be seen from Fig. 1a, the value of ε' increases with increasing temperature due to the temperature dependent of capacitance. However, it can be seen from Fig. 1b, the value of ε'' decreases with the increase of temperature proportional to the values of conductance. Fig. 1c shows the temperature dependency of tan δ which is proportional to the values of ε' and ε''. As can be seen from Fig. 1c, the values of tanδ increase with decreasing temperature and give a minimum about zero bias. Sample (interfacial Maxwell-Wagner polarization) [14,15] and at the interface between the sample and the electrodes (space charge polarization) [16].

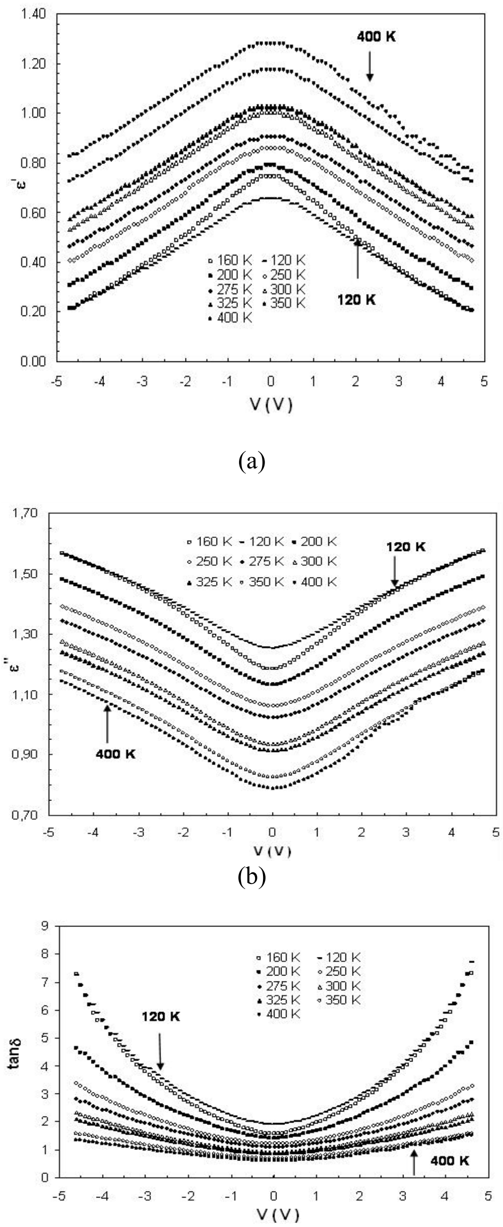

Fig. 1. The variation of the ε', ε'' and tanδ vs applied voltage for Al/NphAOEMA/PEDOT-PSS/ITO structure at different temperatures.

It can be seen from Fig. 2, the values ac electrical conductivity of the Al/NphAOEMA/PEDOT-PSS/ITO structure, in generally, decrease with increasing temperature and also give a minimum about zero bias which is the same with the conductance behaviour [6].

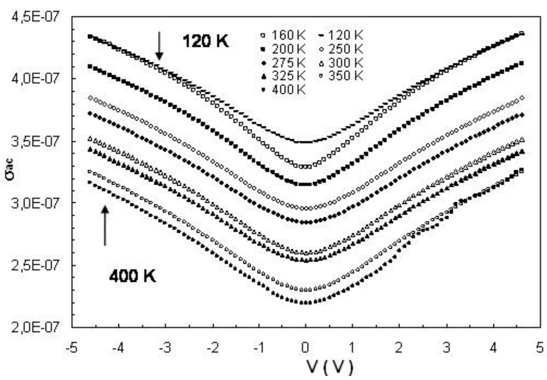

Fig. 2. The variation of ac electrical conductivity (σ_{ac}) vs applied voltage for Al/NphAOEMA/PEDOT-PSS/ITO structure at different temperatures.

The variation of the ε', ε'' and tan δ for Al/NphAOEMA/PEDOT-PSS/ITO structure with temperature at various bias voltages are shown in Fig. 3(a), (b) and (c), respectively. It is clear that the values of the ε', ε'' and tan δ are sensitive to bias voltage at relatively low temperatures. In other words the high values of ε', ε'' and tan δ interestingly be observed at low bias voltages and temperatures. As can be seen from Fig. 3 (a) values of ε' increases with increasing temperature for each bias voltage. On the other hand, the values of ε'' (Fig. 3(b)) decreases with increasing temperature. This increase in ε' towards the high temperature region may be due to interfacial space charge formation, which would be effective at high temperatures and hence the capacitance or ε' being to increase [14]. Also it may be attributed to free charge build-up at interfaces within the bulk of the

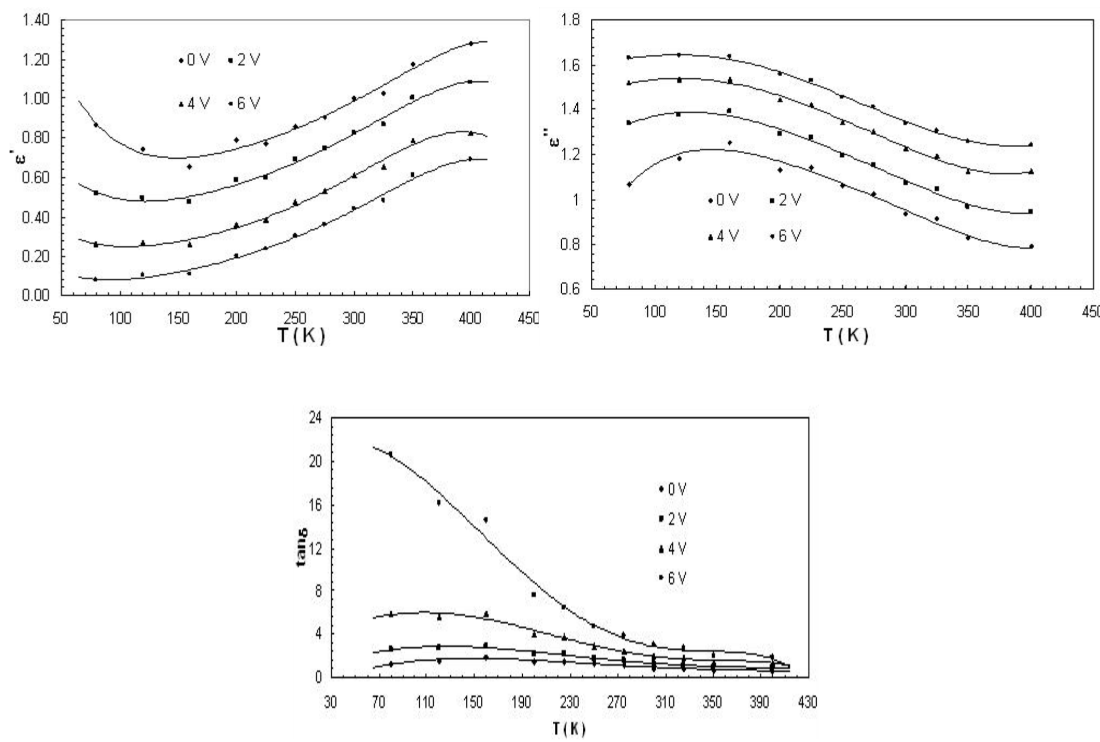

Fig. 3. Dependence of ε', ε'' and tanδ of Al/NphAOEMA/PEDOT-PSS/ITO structure on temperature and voltage.

The variation of the σ_{ac} with temperature and the Arrhenius plots for the temperature dependence of σ_{ac} for Al/NphAOEMA/PEDOT-PSS/ITO structure at various bias voltages are shown in Fig. 4(a) and (b), respectively. As can be seen from Fig. 4(a), the values of σ_{ac} decrease with increasing temperature. On the other hand, the value of σ_{ac} increases with increasing bias voltage. Such behaviour of σ_{ac} is attributed to the amount of impurities, which are located at the grain boundaries. This means that the contribution of σ_{ac} or conduction mechanism comes from the grain boundaries. We calculated the activation energy (E_a) using a well-established empirical relation [17];

$$\sigma_{ac} = \sigma_o \exp(-E_a / kT) \tag{7}$$

where σ_o is the pre-exponential factor, k the Boltzmann constant. The temperature dependent ac conductivity data obtained in the range of 120 - 400 K can be fitted to Arhenius plot Eq.7 as shown in Fig. 4(b). As shown in Fig. 4(b), there is a linear relationship between the total conductivity and the inverse absolute temperature except for two low temperatures (120 and 160 K). The values activation energy (E_a) of the structures were obtained from the slope of the Ln (σ_{ac}) vs q/kT plot (Fig. 4b), decreases with increasing bias voltage and changed from 13.55 meV (at zero bias) to the 8.76 meV (at 6 V). The high E_a that is observed in the low bias voltage may be due to the sum of the energies required for the generation of the charge carriers and their motion into vacancies [8]. It can be concluded that electrical conductivity (σ_{ac}) depends on temperature and it is reasonable to assume that a range of E_a is involved.

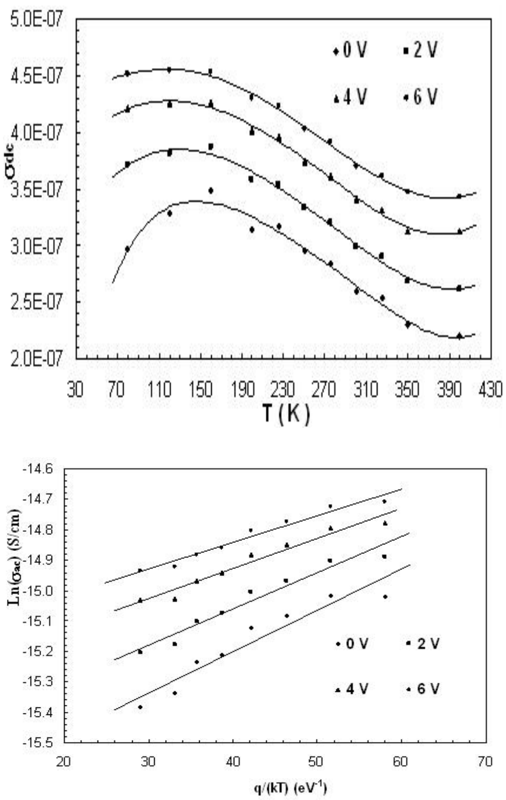

Fig. 4. Dependence of ac electrical conductivity (σ_{ac}) and Arrhenius plots of Al / NphAOEMA / PEDOT-PSS/ITO structure on temperature and voltage.

Experimental results of the Al/NphAOEMA/PEDOT-PSS/ITO structure show that the values of dielectric constant (ε'), dielectric loss (ε''), loss tangent (tan δ) and ac electrical conductivity (σ_{ac}) are strongly temperature and bias voltage dependent. While ε' increases with increasing temperature, ε'', σ_{ac} and tan δ decrease with increasing temperature. This behaviour of dielectric properties of Al/NphAOEMA/PEDOT-PSS/ITO structure may be attributed to interfacial polarization [17].

4. Conclusions

The temperature dependence of dielectric properties and ac electrical conductivity (σ_{ac}) of Al/NphAOEMA/PEDOT-PSS/ITO structure have been investigated in the temperature range of 120-400 K. Experimental data show that ε', ε'', tanδ and σ_{ac} are strongly temperature and bias voltage dependent. The value of ε' increases with increasing temperature while the value of ε'' decreases with increasing temperature for each bias voltage. The interfacial polarization depends on temperatures, and the number of interface state density at Al/Polymer interface, consequently, contributes to the improvement of dielectric properties of Al/NphAOEMA/PEDOT-PSS/ITO structure. The high value of E_a which is observed in the low bias voltage may be due to the sum of the energies required for the generation of the charge carriers and their motion into vacancies. In conclusion, we can say that the dielectric properties of Al/NphAOEMA/PEDOT-PSS/ITO structure obtained from impedance measurements are changed due to thermal restructuring and reordering at the metal-semiconductor interface under the temperature effect.

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*Corresponding author: recep.sahingoz@bozok.edu.tr