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INVESTIGATION OF ELECTRICAL PROPERTIES OF Al/PAN/p-Si MPS DEVICES FABRICATED BY SPIN COATING AND SCREEN-PRINTING TECHNIQUES

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Abstract

In this work, the polyacrylonitrile polymer was coated on top of p-Si by two different techniques, and the electrical characterization of the fabricated devices was held. Screen-printing and spin coating techniques were used to fabricate the metal/polymer/semiconductor devices, and the devices were finished with Ohmic contact of aluminum on the Si side, and with silver on the polymer side by using evaporation via chemical vapor deposition. The dark current - voltage and frequency dependent capacitance - voltage measurements of the fabricated Metal Polymer Semiconductor structure have been performed. Current and voltage measurements were carried out in the dark and in the voltage range of -2.0 V to +2.0 V. Capacitance voltage measurements were carried out in the dark, in the voltage range of -4.0 V to +4.0 V, and in the frequency range of 20 kHz to 1 MHz. The results of electrical characterization have been discussed in view of rectification of devices, interface states, interface dipoles, conduction of carriers, polarization mechanism, and relaxation process.
Keywords: Polymer, Polyacrylonitrile, MPS, Interface, Polarization mechanism, Relaxation

DÖNDÜREREK KAPLAMA VE SERİGRAFİ BASKI YÖNTEMLERİ İLE ÜRETİLEN Al/PAN/p-Si MPY AYGITININ ELEKTRİKSEL ÖZELLİKLERİNİN ARAŞTIRILMASI

Özet

Bu çalışmada poliakrilonitril polimeri p-Si üzerine iki farklı teknikle kaplanmış ve üretilen aygıtların elektriksel karakterizasyonu akım voltaj, frekans bağımlı kapasitans – voltaj ölçümleri aracılığı ile yapılmıştır. Metal/Polimer/Yarı iletken aygıtları üretmek için serigrafi baskı ve döndürerek kaplama teknikleri kullanıldı ve her iki aygıt da kimyasal buhar biriktirme tekniği kullanılarak buharlaştırma yoluyla p-Si katmanından alüminyum ile polimer katmanından gümüş ile Ohmik kontak yapısıyla tamamlandı. Üretilen Metal Polimer Yarı iletken aygıtların karanlık akım - gerilim ve frekansa bağlı kapasitans - gerilim ölçümleri yapılmıştır. Akım - Gerilim ölçümleri karanlık ortamda ve -2,0 V - +2,0 V aralığında yapılmıştır. Kapasitans - Voltaj ölçümleri de yine karanlık ortamda -4,0 V - +4,0 V voltaj aralığında ve 20 kHz - 1 MHz frekans aralığında gerçekleştirilmiştir. Ölçümlerin sonuçları analiz edilerek aygıtların elektriksel karakterizasyonu yapılmıştır. Akım - voltaj ölçümlerinin analizi ile aygıtların doğrultma faktörleri; frekans bağımlı kapasitans - voltaj ölçümlerinin analizleri ile ise arayüz durumları, arayüz dipolleri, taşıyıcıların iletimi, kutuplanma mekanizması ve durulma süreci açısından tartışılmıştır.
Anahtar Kelimeler: Polimer, Poliakrilonitril, MPS, Arayüz, Polarizasyon mekanizması, Durulma

Cite

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1. Introduction

Recently, hybrid structures gained attention; electronics have been developing by Investigations on such that Metal Polymer Semiconductor (MPS) devices. MPS devices serve good properties of enhanced performances, flexibility, and easy processing. Being ideal for Schottky diodes because of their optical and electrical features, organic (i.e. polymeric) materials play a role as an interface layer in metal/semiconductor devices (MS) [1-7]. Schottky diodes have beneficial electrical properties of lower voltage drop, lower forward pre-voltage and high-speed switching, lower space occupation in the integrated circuits, easy construction, and compatibility, so they are preferably used in the industry [8-10]. Such beneficial properties make the polymers favorable material to investigate. Polymers have been started to be used as interface layer in between metal and semiconductor in recent decades and a new type of device MPS emerged [11-14]. Polymers have key role in designing the future materials. The polymer materials can solve degradation problem in Schottky devices. Therefore, stability gets improved resulting the electrical properties to be improved under atmospheric conditions, and for mass production [15-22]. And also, polymer synthesis and fabrication using polymers are easy, quick and cost effective regarding conventional Schottky diodes using closed chambers and high vacuum, those are slow to operate, extreme and expensive. In order to benefit all the features served, such MPS devices have been reported wide variety of applications of solar cells, sensory devices, light emitting devices, photodetectors, and energy harvesters for their advantages due to rectifying properties, improved performances, and easy processing and modifications. There have been many polymers exhibiting dielectric properties, such as polyvinylidene fluoride (PVDF), poly(3,4-ethylenedioxythiophene) (PEDOT), polypyrrole (PPy), trimethylolpropane triacrylate (TMPTA), and polyacrylonitrile (PAN), etc. [23]. PAN is composed of acrylonitrile of dipolar structure. The dipole structure might give the PAN potential dielectric properties [24]. The PAN has been used as piezoelectric, dielectric, supercapacitor, capacitor, membrane in variety of applications . The PAN has been also on demand material serving some features, including easy deposition, durability, and conductivity, etc.

Here, MPS devices have been prepared by using polyacrylonitrile (PAN) on top of p-Si. The devices were fabricated by two different fabrication techniques of spin coating and screen-printing after threatening the p-Si substrate. Both techniques are easy to process and serve flexibility in atmospheric conditions [19, 20, 25]. PAN spin coating was studied but PAN screen-printing has never been studied. Nevertheless, the electrical properties have never been investigated such as the dielectric function, the interface states and the polarization of interface states in such device. The main difference was the thickness of the films regarding the techniques, the thickness was clearly observable by naked eye. After the devices were fabricated, the electrical properties were comparatively investigated by using the current-voltage measurements and the admittance spectroscopy. The rectification ratio RR, the series resistance R_s, the capacitance C, and the dielectric function are the electrical parameters that point out electrical properties of the interface states and the polarization of these states. Then, the results revealed the p-Si/PAN MPS device to be able to be an appropriate candidate for electronic use.

2. Experimental

2.1. Materials and Preparation

A boron doped Silicon (Si) wafer of 3-inch diameter and (100) orientation was used as a semiconductor substrate. After cutting the Si wafer with 2×2 cm^2, the cleaning procedure has been employed by RCA procedure: The Si wafers were put into a water solution of NH_4OH and HCl and boiled in order to remove the impurities which could be formed on the surface of the Si wafer. Then, they were rinsed and dried with a manual blowing pump. As all the procedure was in the air so inevitably the Si wafer would have a couple of nm SiO_2 layer on top and then the surfaces were rinsed with H_2O:HF solution by 1:1 volume ratio to have a Si wafer free of oxide layer.

2.2. Fabrication

After the preparation, the aluminum ohmic contact was evaporated on the bottom of the Si wafer under the pressure of 10^{-7} mBar by Physical Vapor Deposition (PVD). The PAN precursors were prepared in two different concentrations: one for spin coating and the other is for screen-printing. PAN was dissolved in DMF for the both precursors. The precursor for spin coating was prepared in a PAN to solution weight ratio of 2:100 and the screen-printing precursor was in 9:100 weight ratio. Firstly, 50 µL of PAN precursor spin coated on top of cleaned Si wafer for 60 second with 2000 rpm. Secondly, a 1 mL drop of PAN precursor was swiped at once on p-Si substrate.

2.3. Electrical Measurements

The MPS device was finished by evaporating silver (Ag) on top of the PAN coated samples. The I-V and C-V measurements were performed by using Keithley 4200 semiconductor system under dark and at room temperature conditions. The voltage interval is in between -4 V and +4 V and the frequency of the electric signal is applied from low frequency 20 kHz up to high frequency 1.0 MHz.

3. Results

An insulator layer formed in between metal and the semiconductor plays important role for a Schottky barrier diode. The insulator layer could either be formed naturally/unintentionally or one can put it intentionally there. So, the density of states/traps are created for the conduction of the electron or hole, and the current voltage characteristics are of some significant changes. Thermionic emission is the best to explain the conduction mechanism in MPS device [26-28]. The current is expressed in the thermionic emission mechanism for the majority carrier flowing across the junction barrier namely Schottky barrier as;

$$I = I_o e^{q(V-IR_s)/nkT}\left(1 - e^{q(V-IR_s)/nkT}\right) \tag{1}$$

$$I_o = AA^*T^2 e^{-q\Phi_{Bo}/kT} \tag{2}$$

where, V is the forward-bias voltage, I_o is the reverse saturation current, q is the charge of the electron, R_S is the series resistance, k is the Boltzmann constant, T is the absolute temperature, A is the contact area, A* is the effective Richardson constant for p-Si with a value of 32 A cm^{-2} K^{-2}, Φ_{Bo} is the barrier height. [26, 27];

As measured at room temperature (300 K), the MPS device Al/PAN/p-Si exhibits quite clearly Schottky barrier diode behavior. The I-V plot is given in Figure 1. As the rectifying property of a diode is characterized by rectification ratio, the rectification ratio is calculated through the Equation 3 [29, 30], and plotted in Figure 2. Some numerical values for the rectification ratio have

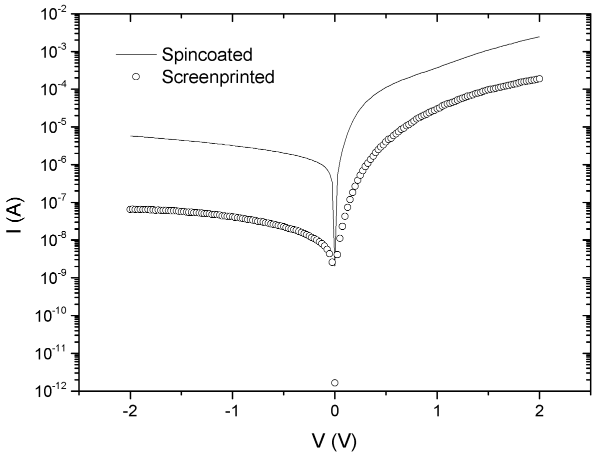


Figure 1. The Current vs. Voltage graph of measurements of Al/PAN/p-Si spin coated and screen-printed devices in semi-log I-V plot.

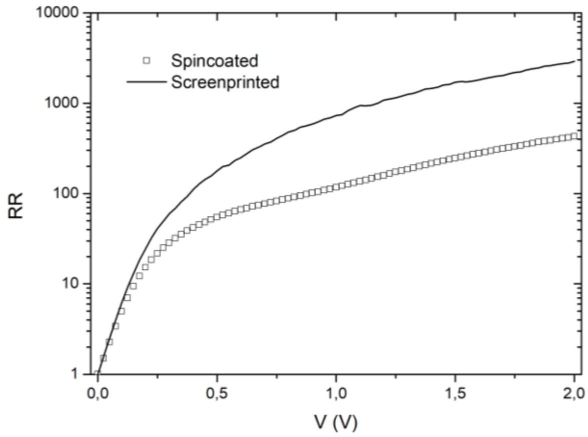


Figure 2. The graph of Rectification Ratio vs. Voltage of Al/PAN/p-Si spin coated and screen-printed devices in a semi-log I-V plot.

been tabulated in Table 1. The screen-printed device has higher RR values indicating it as a better MPS device in comparison with spin coated one. Numerically, RR of the screen-printed device is greater than that of the spin coated device by factors of 3.27, 6.24, and 6.79 at symmetrical bias voltages of 0.5 V, 1.0 V, and 2.0 V, respectively.

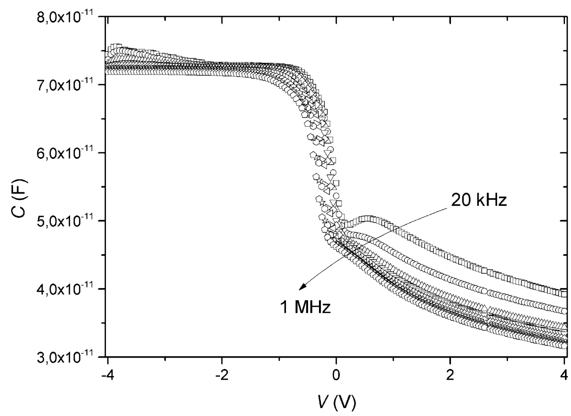
$$RR = \frac{I_{fwd}}{I_{rew}}; at\ any\ voltage\ V \quad (3)$$

Table 1. Some numerical values of rectification ratio of the Al/PAN/p-Si spin coated and screen-printed devices.

Device	RR		
	for ± 0.5 V	for ±1.0 V	for ±2.0 V
spin coated	54.5	116.8	427.1
screen-printed	178.2	728.9	2897.9

For electronic applications, it is important to reveal the dielectric properties of the MPS devices. Modelling is the best method to reveal dielectric properties. In modelling such a device/a diode, the device is characterized by admittance spectroscopy method [30, 31]. The capacitance through the device is of three regions as accumulation, depletion, and inversion: these regions cover the interface and the semiconductor.

The capacitance (C-V) and the conductance (G/ω-V) measurements of the MPS device Ag/PAN/p-Si were performed and are a function of voltage in a wide range frequency such that between 20 kHz and 1 MHz. The C-V and G/ω-V plots have been given in Figure 3, and Figure 4 and the plots for the both devices are compatible with that of p-type semiconductor device reflecting accumulation, depletion, and inversion regions, clearly. For spin coated device, in both the accumulation and the inversion region, the capacitance and conductance decreased as the frequency increased. Biasing with ac signal, that the charge carriers are objected follow the changing E-field results some change in the capacitance due to the change in the dielectric function [32-35]. The lower frequency the easier following the ac signal contributes some more capacitance; or vice versa at higher frequencies i.e. $f \geq 100$ kHz. There have been seen peaks located from -0.1 to -0.3 V bias with increasing frequency 20 kHz to 1 MHz. Such peaks are generally attributed to the presence of the interface states localized



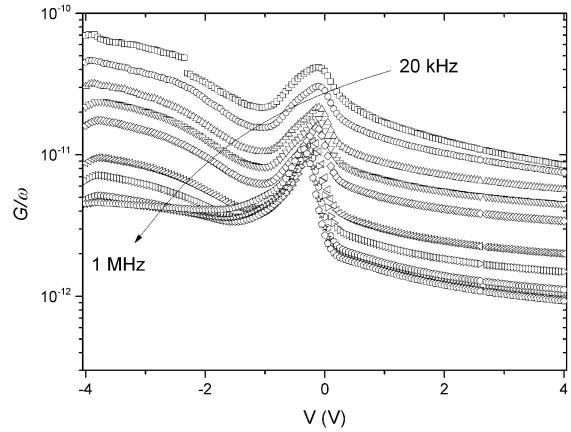


Figure 3. C-V and G/ω-V plot of Al/PAN/p-Si spin coated device.

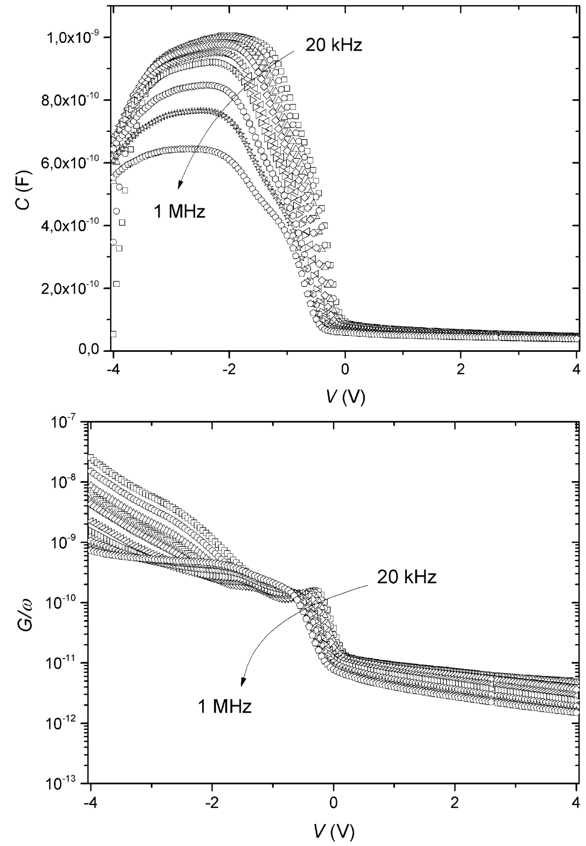


Figure 4. C-V and G/ω-V plot of Al/PAN/p-Si screen-printed device.

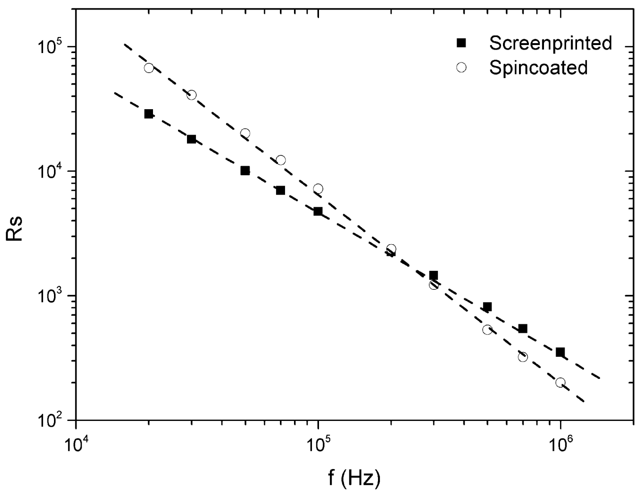


Figure 5. The series resistance, R_S vs frequency, f plot of Al/PAN/p-Si screen printed and spin coated devices.

in between the semiconductor and the polymer [36]. That the device has a decreasing trend in capacitance and conductance with increasing voltage and frequency is clear in the Figure 3, and indicates and supports the idea that of interface states.

For the screen-printed device, the capacitance and the conductance characteristics are given in the Figure 4. In the accumulation region, the device had the capacitance values which were closer values in the low frequency range (f < 100 kHz) whereas the capacitance decreases rapidly at high frequencies. Such behavior could be attributed to the deep impurity states of the semiconductor interface states between the semiconductor and the polymer, and the series resistance [37-40]. The capacitance from those of the states diminishes since the states cannot follow the E-field as the frequency increases [33,34].

The series resistance may cause deterioration on the impedance – admittance data. The series resistance at higher frequencies gets important role because of non-ideal behavior of the semiconductor devices. It is well-known that R_s is resulted from doping in the semiconductor, the resistance from ohmic and/or rectifying contacts, the impurities between the back contact and the semiconductor. In a list of methods to determine the series resistance [41], the method put forward by Nicollian and Brews [39] has been selected:

$$R_s = \frac{G}{G^2 + (\omega G)^2} \quad (4)$$

where C and G are the measured *capacitance* and *conductance*, and ω is the angular frequency. In Figure 5, R_s vs f characteristics have been plotted. The behavior that the series resistance decreases exponentially with the increasing frequency could be attributed to the carrier response at those frequencies in the interface states.

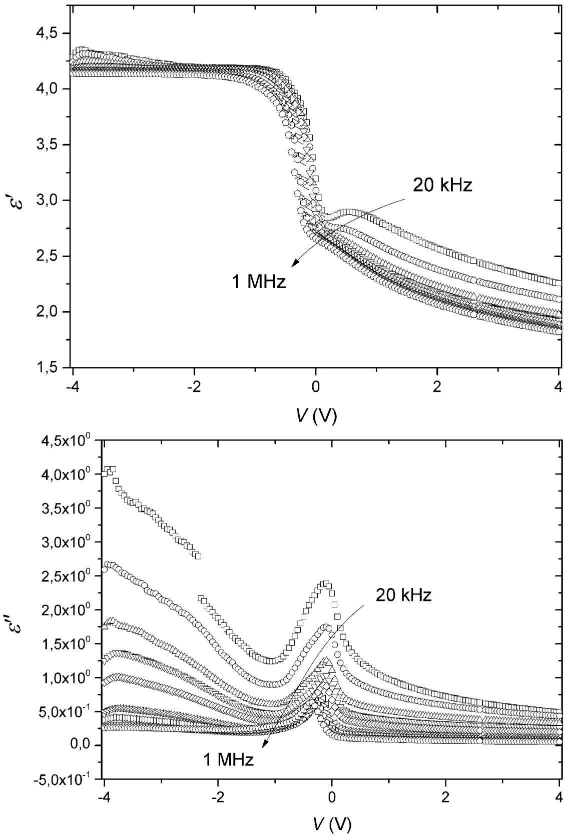


Figure 6. The dielectric measurements of Al/PAN/p-Si spin coated device.

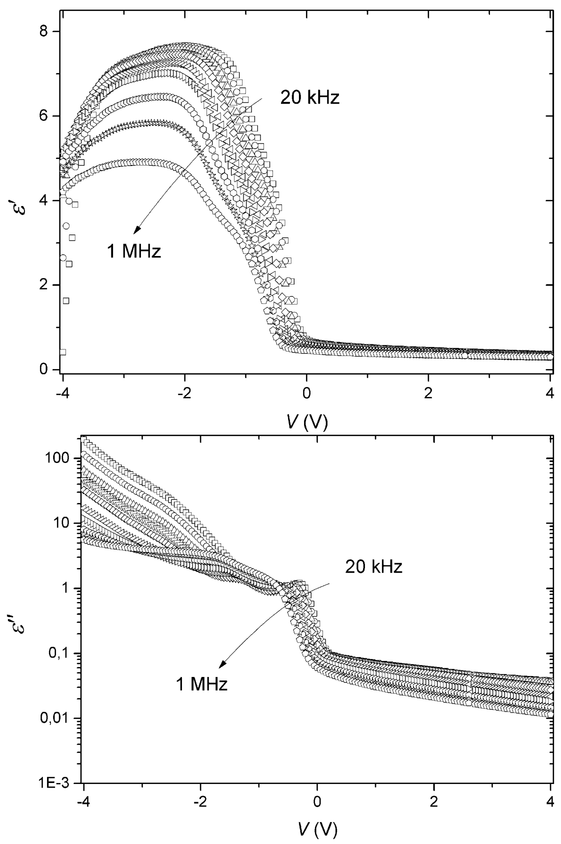


Figure 7. The dielectric measurements of Al/PAN/p-Si screen-printed device.

The dielectric properties of the polymer interface material make the device have practical applications in electronic devices, so it needs to be revealed. The dielectric function/permittivity, ε is composed of reel and imaginary parts that are the dielectric constant ε' and the dielectric loss ε'', respectively, and is/are given by the equations:

$$\varepsilon_r = \varepsilon' - i\varepsilon''; \; \varepsilon' = \frac{Cd}{A\varepsilon_o}, \varepsilon'' = \frac{Gd}{\omega A\varepsilon_o} \quad (5)$$

$$\tan\delta = \frac{\varepsilon'}{\varepsilon''} \quad (6)$$

$$M = \frac{1}{\varepsilon_r} = M' + iM'';$$
$$M' = \frac{\varepsilon'}{\varepsilon'^2 + \varepsilon''^2}, M' = \frac{\varepsilon''}{\varepsilon'^2 + \varepsilon''^2} \quad (7)$$

where C is the capacitance of free space, ω is the angular frequency, t is the thickness of the sample, A is the cross-sectional area, and $tan\delta$ is the tangent loss. Then, one can drive the equation for the electric modulus, M by utilizing the dielectric function. The dielectric constant ε' and the dielectric loss ε'' measurements and $tan\delta$ have been plotted in Figure 6, Figure 7, and in Figure 8 for both the devices. ε', ε'' and $tan\delta$ decrease as the frequency increases for the both MPS devices. Why the higher frequency the lesser dielectric constant and loss have been observed could be attributed to the dipoles located interface being unable to follow the applied electric field

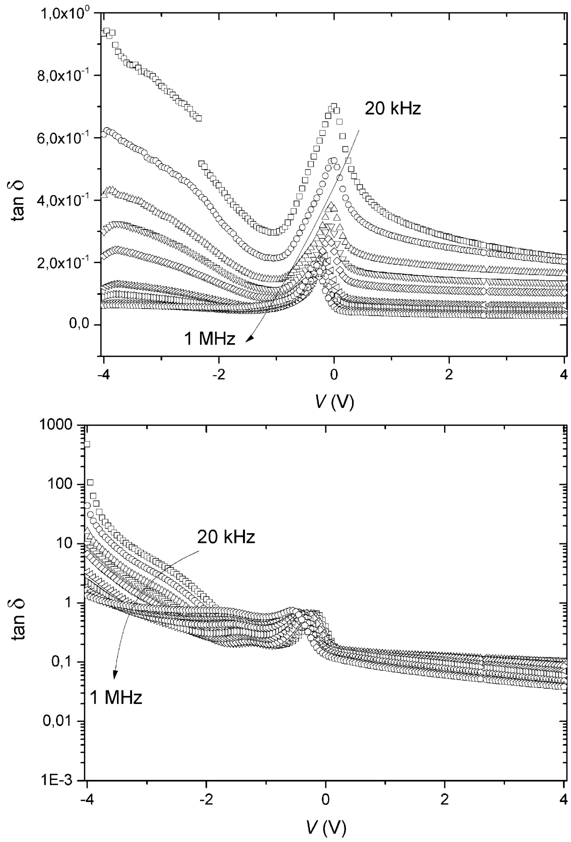


Figure 8. tanδ – V graphs of Al/PAN/p-Si spin coated (top) and screen-printed (bottom) devices.

(E-field) of bias. Because, as the frequency increases the period of the electric signal decreases; as they are inverse of each other. So, the dipole would have less time to reorder/reorient in the direction of the E-field of the applied electric signal. Then, the dielectric constant became less frequency-dependent or independent at high frequencies.

In addition to providing insight information into the electrical processes occurring in the materials at various frequencies and voltages, the study of electric modulus is a beneficial tool in investigation more about of bulk characteristics, electrical conductivity, polarization processes, and relaxation times. M' and M'' calculations made by using the Equations 5 to 7 and plotted in the Figure 9 for spin coated device and in the Figure 10 for the screen-printed device. M' and M'' displayed dependances on the applied bias voltage and the frequency. Similar behavior that of in ε', ε'' was observed in the graphs of M' and M''. Both samples' M'' data presented a peak about zero bias and the peaks showed shift to the negative bias region with increasing frequency. The zero bias corresponds to space charge region; here, the carriers were to follow electric signal at low frequencies by contributing the capacitance in contrast the increasing frequency resulted the carriers not to follow the signal and the contribution to the capacitance decreased [42-45]. In the Figure 10 of M'' graph of the screen-printed device data, which was

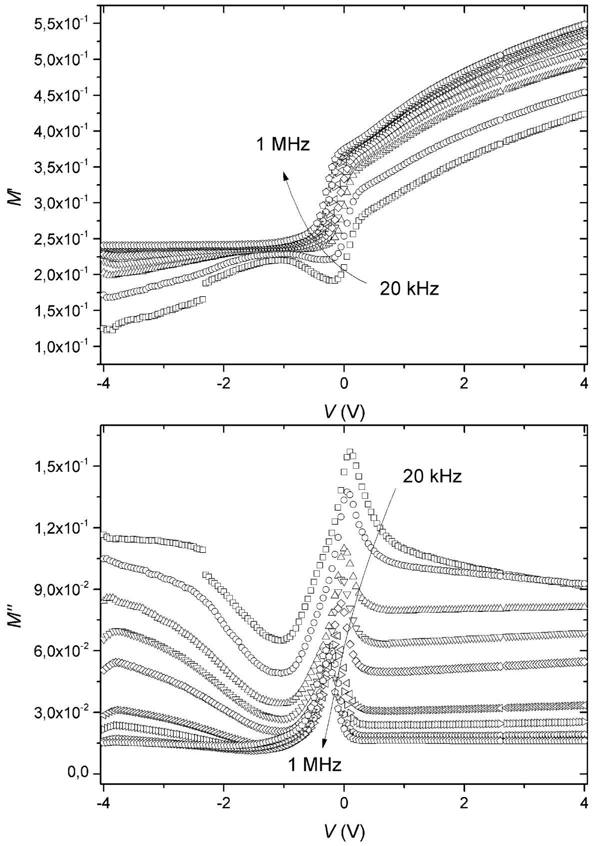


Figure 9. M' and M'' – V graphs of Al/PAN/p-Si spin coated device.

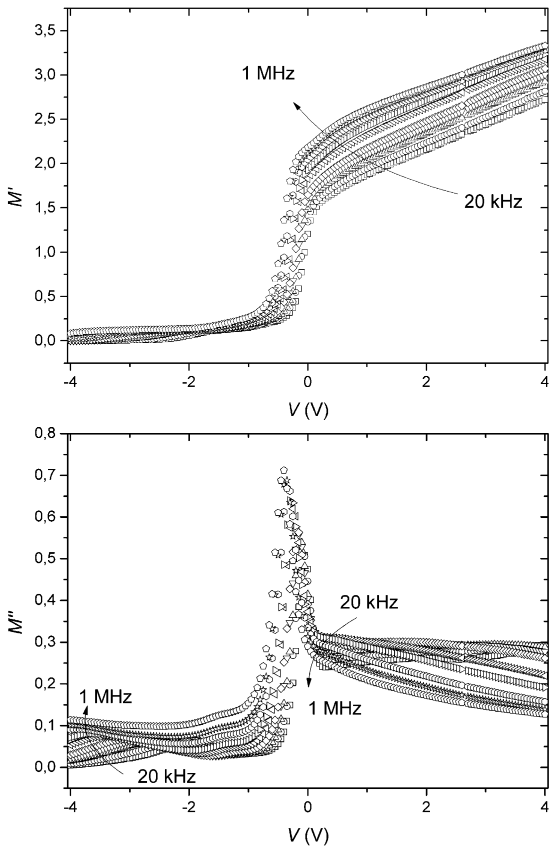


Figure 10. M' and M'' – V graphs of Al/PAN/p-Si screen-printed device.

different from the other device's data, an increasing change in M'' was observed with increasing frequency in the accumulation region then the change was reversed to the decreasing behavior in the inversion region. The interface states and polarization mechanism plays the important role, and regarding the data, the change in the direction of the behavior might be attributed to the change of polarization mechanism [29, 33-35, 46]. Both the value of M' and M'' of screen-printed device approached about zero at low frequencies, the main idea behind that was attributed to dielectric relaxation mechanisms [47-49].

4. Conclusion

This study focused on fabrication and electrical properties of an MPS device which has been formed by coating the polyacrylonitrile (PAN) on p-Si semiconductor. Two distinct manufacturing methods were utilized to create an MPS device: spin coating and screen printing. Rectification of the screen-printed device showed much higher value *i.e., 2897* at 2.0 V forward bias voltage. R_s for both devices decreased with increasing frequency. R_s results were attributing the interface states. Then, the dielectric analysis of the devices addressed and strongly showed the interface dipoles localized between the semiconductor and the polyacrylonitrile coating for both. Finally, the electric modulus calculations and analysis explained the mechanisms that play important role in explaining the conduction of the carriers. The polarization mechanism and the relaxation process are the two defining how the carriers follow external E-field, applied voltage. The results showed that the devices are the candidates of the electronic applications.

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