



The study of the dependence of dielectric properties, electric modulus, and ac conductivity on the frequency and voltage in the Au/(CdTe:PVA)/n-Si (MPS) structures

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ABSTRACT

The dielectric properties, electric modulus, loss tangent ($\tan\delta$), and ac conductivity of the fabricated Au/(CdTe:PVA)/n-Si (MPS) structures were investigated in wide-range frequency. By using the capacitance/conductance–voltage–frequency (C/G–V–f) measurements, the real/imaginary components of complex dielectric constant (ϵ' , ϵ'')/electrical modulus (M' , M''), loss tangent ($\tan\delta$), and σ_{ac} were calculated in wide-range frequency (2 kHz–0.7 MHz) and voltage (± 2.5 V), and all results show that these parameters are a strong function of voltage and frequency. The observed decreases of ϵ' and ϵ'' with increasing frequency were attributed to the existence of N_{ss} , interlayer, and dipole polarizations. The value of ϵ' was calculated as 128.38 (at 2 kHz and 2.5 V), which is quite higher than the maximum value of traditional SiO₂. $\ln(\sigma_{ac})$ – $\ln(f)$ plot for 0.7 V shows three linear parts, indicating three different conduction mechanisms at low, intermediate, and high frequencies. These results show that the fabricated MPS structures can be successfully used instead of traditional metal–insulator–semiconductor as ultra-capacitors, which can use more electronic charges or energy.

1 Introduction

When an insulator forms between a metal and a semiconductor, the metal–semiconductor (MS) structure or Schottky diode (SD) turns into a metal–insulator–semiconductor (MIS)-type SD or structure [1–6]; moreover, this interfacial layer is easily polarized under an external electric or magnetic field. Although conventional SiO₂ and SnO₂ are very stable and grow

easily onto semiconductors' surfaces, they cause high leakage current and cannot completely pacify unsaturated bonds or N_{ss} . These N_{ss} are usually located between the semiconductor and the interlayer, and their energies correspond to the forbidden bandgap energy (E_g) [5–11]. Depending on their occupancy, these states may act as recombination centers, releasing or capturing electrons under external influence. In general, C–G/ ω –V curves deviate from the ideal

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situation due to N_{ss} and interfacial-dipole polarization mechanisms. These situations appear particularly noticeable at low and medium frequencies in the depletion region. Because, at low and medium frequencies, as long as the period ($T = 1/2\pi f$) values exceed the lifetimes (τ) of the N_{ss} , the chargers in these traps can effectively keep up the external ac signal, allowing dipoles ample time to rotate in response to the electric field direction [10–14]. Therefore, especially at low and medium frequencies, there is a significant increase in both C and G values due to interfacial states and polarization [12–15]. However, as the frequency increases, the contribution decreases and becomes negligible to both C and G values at high frequencies.

However, instead of classical and low dielectric insulating materials, using some nanocomposite, a pure metal and metal oxide-doped organic interface layer with high dielectric properties can quite increase the performance of these devices by passivating these surface states [13, 16–19]. Pure polymers usually have low conductivity and dielectric values. However, adding small amounts of metal or metal oxide materials such as Co, Zn, Gr, ZnO, MnO, and CdTe can increase these values. Among polymers, some of the most used as interlayer between metal and semiconductor interface are usually polyvinyl alcohol (PVA) and pyrrolidone (PVP), which can be easily soluble in alcohol or water, and they also have similar functional groups [20–25]. In the other words, the basic technical and scientific problems of MS structures with/without an interlayer is to improve their performance and decrease cost production by using especially high-dielectric interfacial layer at M/S interface grown by simple and easily techniques. Especially, CdTe has moderate forbidden bandgap when compared to some insulators such as SnO_2 and TiO_2 . Cadmium telluride (CdTe) is a member of the II–VI group and is commonly used in photonic applications because of its high absorption coefficient in the visible range (VR) and moderate bandgap energy (1.45 eV), huge absorption coefficient ($> 104/\text{cm}$) near the band edge stability, high mobility ($1040 \text{ cm}^2/\text{V s}$), and low cost [26, 27]. The integration of semiconductor nanoparticles with polymers is a considerably desirable approach. Improving the performance of electronic and optic devices is a crucial issue, and using new materials is key to achieving this goal [28, 29]. PVA is a semicrystalline polymer suitable

due to its high thermo-stability, chemical resistance, dynamic strength, solubility in water and alcohol, and controllable electric and dielectric properties when doped [30].

In our previous work [15], the voltage and frequency profiles of D_{it} , τ , and R_s were obtained with various methods. Some of the main electrical parameters were also calculated from the reverse bias C^{-2} vs V plot for each frequency. The N_{ss} and their lifetimes were also obtained from the parallel conductance method, which is known as the most accurate and reliable one. Therefore, in this study, the real and imaginary components of the complex dielectric constant ($\epsilon^* = \epsilon' - j\epsilon''$), electrical modulus ($M^* = 1/\epsilon^* = M' + jM''$), and impedance ($Z^* = Z' - jZ''$) of the Au/(CdTe:PVA)/n-Si MPS structure, loss tangent ($\tan\delta$), and the Nyquist plots (M'' vs M') have been investigated in detail over the frequency range of 2 kHz to 0.7 MHz at ± 2.5 V. All of these parameters were found to have strong functions of frequency and applied voltage. The measured value of ϵ' was found to be 128.38 (at 2.5 V), which is 33.78 times bigger than the value of traditional SiO_2 , even at 2 kHz. The double logarithmic plot of σ_{ac} vs $\ln(f)$ shows three linear components corresponding to low, intermediate, and high frequencies with different slopes. This observation suggests the existence of two different charge transport mechanisms in the sample. The experimental results indicate that the (CdTe:PVA) interlayer may serve as a viable alternative to traditional insulators for ultra-capacitors, as it exhibits a high value of ϵ' even at 2 kHz, allowing for the storage of more electronic charges/energy.

2 Experimental procedure

The Au/(CdTe:PVA)/n-Si (MPS) structure was fabricated onto n-Si. First, the n-Si substrate was cleaned in the ultrasonic bath using a mixture of chemical acid and deionized water. It was then rinsed in deionized water and dried with N_2 gas. Secondly, the whole back of the n-Si substrate was coated by very pure Al with 150-nm thickness and then annealed at 500°C for 5 min to form the ohmic contact. The CdTe-PVA was performed by dispersing 10 mg CdTe nanostructure in 5 ml of 0.05% PVA solution. The solution was then applied to the front of an n-Si substrate using the spin coater method.

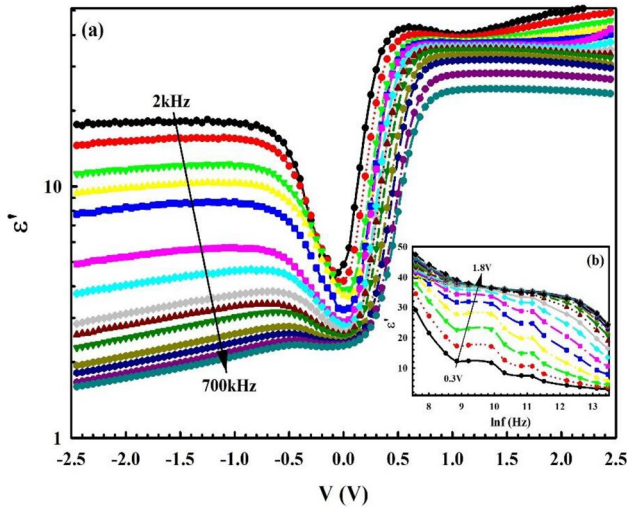


Fig. 1 **a** The ϵ' vs V , **b** the inset ϵ' vs $\ln(f)$ plots of the MPS structure

Finally, pure Au rectifier contacts with a thickness of 150 nm were thermally grown onto (CdTe-PVA) interlayer. The C - V and G/ω - V measurements were performed utilizing an HP4192-A impedance meter at room temperature (RT). For more detailed information on the chemical cleaning of the surface of p-Si wafers, fabrication procedures, measurement systems, and structural analysis, refer to our previous work [15].

3 Results and discussion

The frequency-dependent admittance technique, which includes a set of capacitance/conductance (C/G - V) measurements, is generally utilized to calculate the real/imaginary components of complex dielectric constant ($\epsilon^* = \epsilon' - j\epsilon''$). With this, the

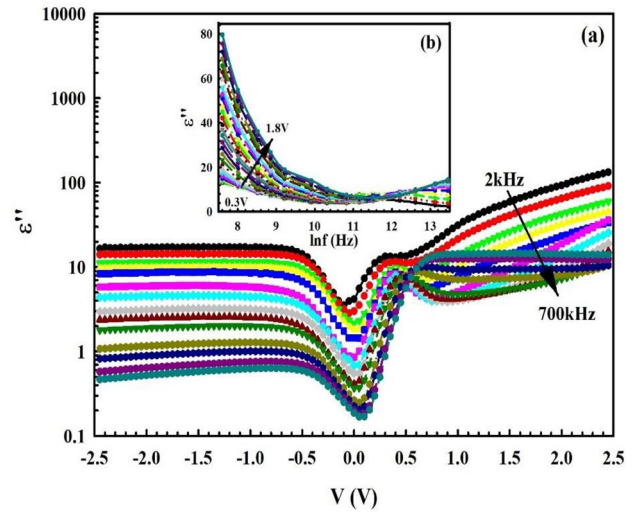


Fig. 2 **a** The ϵ'' vs V and **b** the inset ϵ'' vs $\ln(f)$ plots of the MPS structure

obtained dielectric data can be employed to analyze the complex electrical modulus ($M^* = M' + jM''$), loss tangent ($\tan\delta$), and electrical conductivity (σ). The measurements were performed in a wide voltage and frequency range to ensure accuracy and reliability. Therefore, in this study, C and G versus V measurements were carried out with a frequency range of 2–700 kHz and a voltage range of ± 2.5 V at RT. After that, the ϵ' , ϵ'' , and $\tan\delta$ ($= \epsilon''/\epsilon'$) values were extracted from the measured C and G using the following relations [12, 16, 24, 26, 31–33]. The real part of the dielectric constant is represented with ϵ' , and it is dimensionless and a measure of the capability of a material to store electronic charges. Under an electric field, the dielectric film can be easily polarized, resulting in energy loss due to internal friction. In the literature, this energy loss is called dielectric loss (ϵ''). $\tan\delta$ is another method to measure the energy loss as heat under an electric field.

$$\epsilon^*(\omega) = \epsilon'(\omega) - j\epsilon'' = \left(\frac{C_m d_i}{A \epsilon_0} \right) - j \left(\frac{G_m d_i}{A \epsilon_0} \right) = C_m / C_0 - j(G_m / \omega C_0) \quad (1)$$

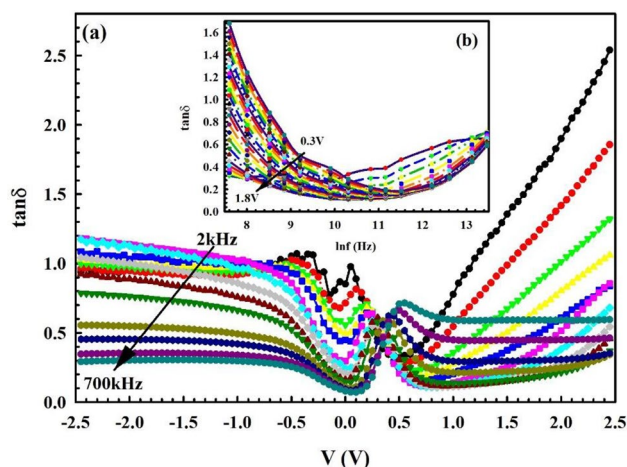


Fig. 3 **a** The $\tan\delta$ vs V and **b** the inset $\tan\delta$ vs $\ln(f)$ plots of the MPS structure

In Eq. (1), $\left(\frac{\epsilon_{0A}}{d_i}\right)$ is the C value of the empty capacitor (C_0), ϵ_0 (8.85×10^{-12} F/cm) is the dielectric constant of free space, d_i is the interlayer thickness, and ω is the angular frequency ($2\pi f$). C_m and G_m/ω are the measured C and G values, respectively. The voltage-dependent profiles of $\epsilon' - V$, $\epsilon'' - V$, and $\tan\delta - V$ plots for different frequencies, as well as frequency-dependent profiles for various positive biases, are given in Figs. 1, 2, and 3, respectively. Figures 1 and 2 show that the curves for both $\epsilon' - V$ and $\epsilon'' - V$ have three distinct regions corresponding to voltage ranges of approximately $-2.5/0.0$ V (inversion), $0.0/0.7$ V (depletion), and $0.7/2.5$ V (accumulation). The values of both ϵ' and ϵ'' increase as the frequency decreases for the entire bias voltage range. The charges at traps lead to an excess polarization at low frequencies, depending on their lifetimes (τ). However, this contribution disappears at sufficiently high frequencies because the period ($T = 1/\omega$) becomes lower than τ [33, 34]. In the other words, surface states/traps are effective especially in the depletion regions. At low and intermediate frequencies, these effects can be observed. The external ac signal depends on their relaxation/lifetimes (t), leading to an increase both in the real and imaginary parts of the complex dielectric constant. But at sufficiently high frequencies, the value of period ($T = 1/2\pi f$) becomes lower than t and hence both surface states do not have enough time to follow ac signal. As a result, dipoles can only rotate around their own axis, leading to a decrease in the contribution of surface states and dipole polarization with increasing frequency.

The $\tan\delta - V$ curves exhibit distinct peaks for each frequency (see Fig. 3). The observed peak can be attributed to the reorganization and restructuring of electronic charges localized between the interfacial thin film and semiconductor under an electric field [29]. Figures 1, 2, and 3 show that the ϵ' , ϵ'' , and $\tan\delta$ values of the (CdTe:PVA) interfacial thin film are strongly dependent on f and V in both depletion–accumulation regimes. Higher values of ϵ' and ϵ'' are due to the thin interfacial layer, surface-dipole polarization, and interface traps [33, 34]. When the dipoles have sufficient time to keep up with the change in the direction of the electric field depending on the frequency ($T = 1/2\pi f$), increases in ϵ' and ϵ'' values occur at these frequencies [26, 29, 32, 34]. However, charges localized at traps

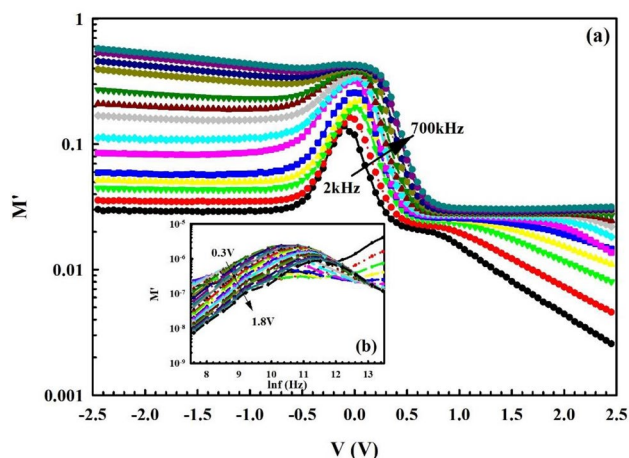


Fig. 4 **a** The M' vs V , **b** the inset M' vs $\ln(f)$ plots of the MPS structure

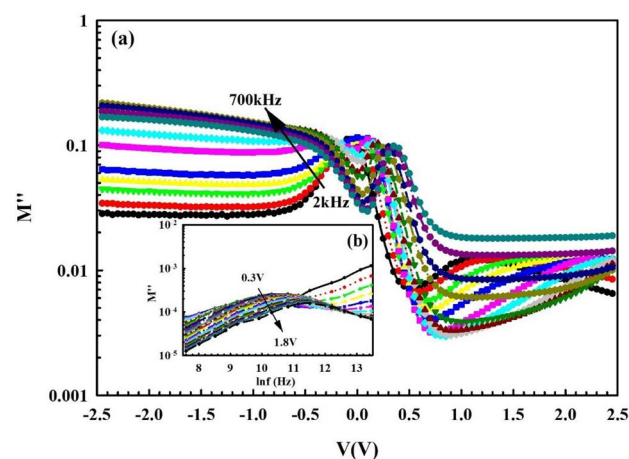


Fig. 5 **a** The M'' vs V , **b** the inset M'' vs $\ln(f)$ plots of the MPS structure

with an adequately long relaxation time ($\tau \gg T$) to allow migration between the traps cannot contribute to ε' and ε'' at higher frequencies.

The electric modulus value typically increases because of the relaxation of the electric field in interfacial thin films/layers, as the electric displacement remains almost constant. The complex formalism of the electric modulus can be analyzed employing the following expression:

$$M^*(f) = \left(\frac{\varepsilon'}{\varepsilon'^2 + \varepsilon''^2} \right) + j \left(\frac{\varepsilon''}{\varepsilon'^2 + \varepsilon''^2} \right) \quad (2)$$

The obtained experimental M' and M'' values from Eq. 2 are given in Figs. 4 and 5 for various frequencies and applied voltage, respectively. These plots are necessary to see the polarization and interface traps/states' effects on the dielectric and electric modulus characteristics for various applied dc bias voltages.

Figures 4 and 5 show that the M' and M'' versus voltage graphs peak at around zero bias voltage when $\omega\tau = 1$, indicating maximum dielectric loss. These peaks demonstrate the transition in charge carriers' mobility from short to long ranges. Additionally, the peak positions shift to positive voltage values as the frequency increases. The main reason for this peak position shift is due to the reordering and restructures of electronic charges and dipoles under the dc electric field [35–37]. The observed essential changes in the real/imaginary parts of M^* with both frequency and voltage are the result of

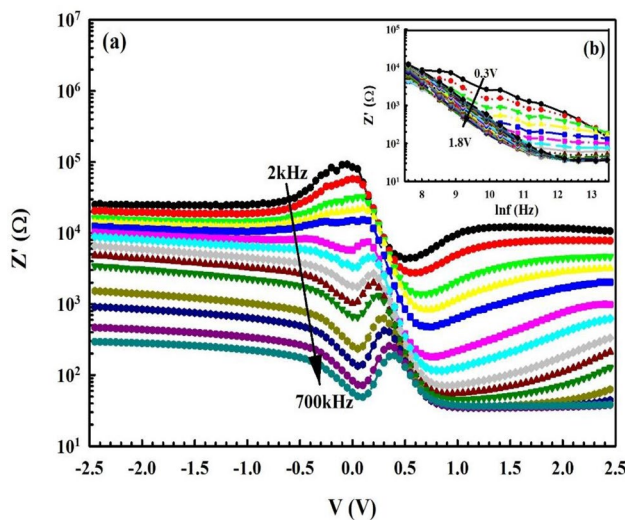


Fig. 6 **a** The Z' vs V , **b** the inset Z' vs $\ln(f)$ plots of the MPS structure

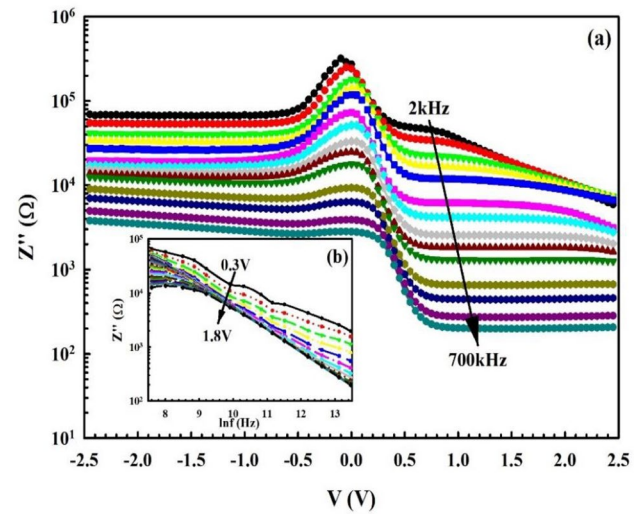


Fig. 7 **a** The Z'' vs V , **b** the inset Z'' vs $\ln(f)$ plots of the MPS structure

localized charges and dipoles in the interfacial thin film, and such a special distribution is usually called Maxwell–Wagner polarization (MWP), which occurs at low and moderate frequencies. The decrease in both M' and M'' values as frequency increases is demonstrated in Figs. 4 and 5. This is a result of the short-range mobility of electronic charges. It can be readily concluded that the observed change in the real and imaginary parts of ε^* and M^* with frequency is due to relaxation phenomena and N_{ss} effects.

The dielectric relaxation behavior can also be explained using complex impedance spectroscopy

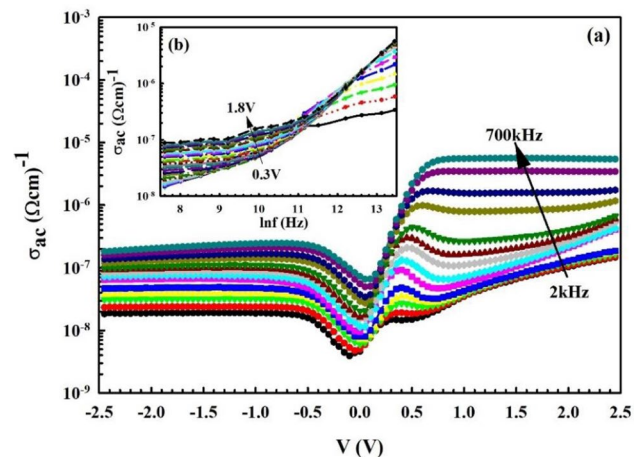


Fig. 8 **a** The σ_{ac} vs V and **b** the inset σ_{ac} vs $\ln(f)$ plots of the MPS structure

($Z = Z' - jZ''$). This is described by the following relations [12, 15, 32–36].

$$Z^*(\omega) = Z' - jZ'' = \frac{1}{j\omega C_0 \epsilon^*} = \frac{\epsilon''}{\omega C_0 [\epsilon'^2 + \epsilon''^2]} - \frac{\epsilon'}{\omega C_0 [\epsilon'^2 + \epsilon''^2]} \quad (3)$$

In Eq. 3, the quantities of ϵ_0 , A , and d represent the permittivity of free space, rectifier-contact area, and the distance between rectifier and ohmic contacts, respectively. Thus, the Z' and Z'' values versus voltage curves for each frequency were extracted from Eq. 3 and are represented in Figs. 6 and 7.

The figures demonstrate distinct peaks in both Z' and Z'' values as the voltage decreases. However, the values of these peaks decrease with increasing frequency for almost every applied dc bias voltage, as shown in the inset of the figures. The ac electrical conductivity (σ_{ac}) values of the Au/(CdTe:PVA)/n-Si MPS structure can be designated with the $\tan\delta$ or ϵ'' values by utilizing the following expression [37–40].

$$\sigma_{ac}(\omega) = 2\pi f \omega C(\tan\delta) \left(\frac{d_i}{A} \right) = 2\pi f \epsilon_0 \epsilon'' \quad (4)$$

It can be clearly seen in Fig. 8 that there are two distinctive peaks in the inversion and depletion regions due to a special density distribution of surface states/traps at interlayer/semiconductor interface. On the other hand, the change in peak magnitudes and their positions with frequency are generally dependent on the lifetimes of traps and

their reordering/restructuring under external electric field. The decrease in the series resistance value with increasing frequency causes an increase in the conductivity values ($G = 1/R$).

Some researchers were also investigated both the basis electric and dielectric parameters and conduction mechanisms depend on frequency, temperature, and applied bias voltage in MS-type structures with an interfacial layer in recent years and they reported similar results on the surface states, series resistance, interfacial layer, and barrier inhomogeneities effects on the performance of these structures [41–45]. The inset of Fig. 8a shows that the ac value remains constant at lower frequencies and increases as the frequency increases. This is due to the decrease in the R_s of the MPS structure and the electronic charges at traps. These two different frequency ranges correspond to DC and AC conductivity, as represented in Eq. 3.

$$\sigma_{ac} = \sigma_{dc} + A\omega^s \quad (5)$$

In Eq. 5, A is a constant, and s represents the degree of applied frequency. The slope (s) values were calculated from Fig. 9. This provides information about the conduction mechanism, with expected values between 0 and 1. To achieve this, double logarithmic σ_{ac} vs f plots were obtained for three different dc bias voltages, as shown in Fig. 9.

Figure 9 shows the $\ln(\sigma_{ac})$ vs f plots for three different voltages, each with three linear parts having different intercept points within the measured frequency range. This enables direct calculation of the

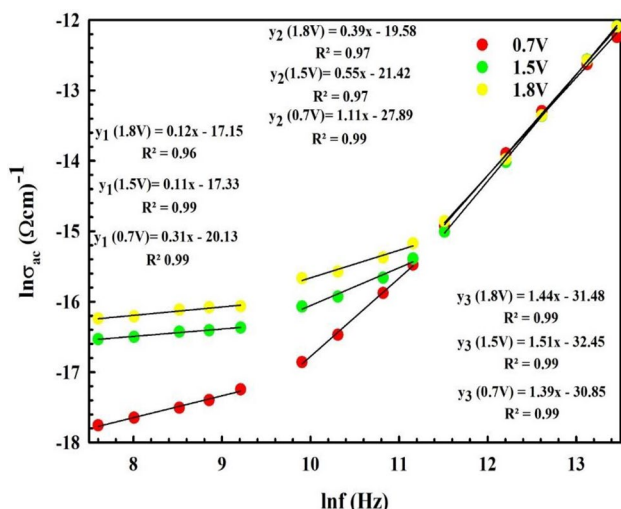


Fig. 9 The double logarithmic σ_{ac} vs f plots of the sample for various applied bias voltages

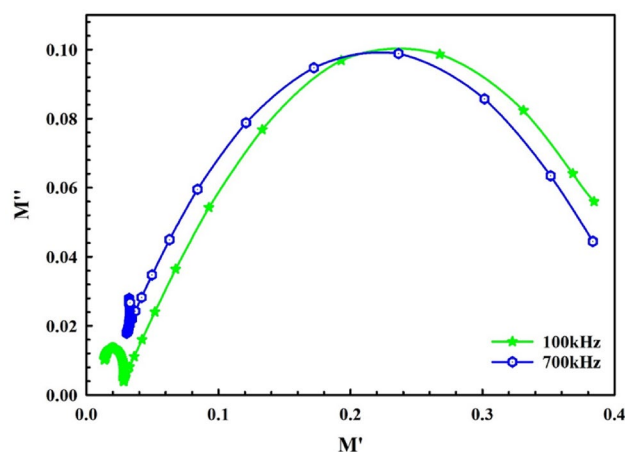


Fig. 10 The Cole/Cole curves (M'' vs M') of the MPS structure for 0.1 and 0.7 MHz

value of s for each bias voltage. The curves also suggest the presence of three distinct charge transports at low/intermediate/high frequencies. The values of s for three applied bias voltages were 0.31, 1.11, and 1.39 for 0.7 V, 0.11, 0.55, and 1.51 for 1.5 V, and 0.12, 0.39, and 1.44 for 1.8 V, respectively. These results indicate that the conduction mechanism known as the relaxation model can occur when electrons migrate from one state/trap to another [39, 40].

Using Cole–Cole curves, (M'' vs M') can provide significant information regarding the nature of the relaxation process of the interfacial layer. Therefore, the Cole–Cole (M'' vs M') curves of the MPS structure at 0.1 and 0.7 MHz are presented in Fig. 10. As demonstrated in this figure, the dielectric relaxation deviates from ideal Debye behavior. The electric modulus curves for interfacial layers or dielectric materials do not always form perfect circles or semicircular arcs; they may become asymmetric. The smaller semicircle observed in the cole-cole curve at 100 kHz disappeared at 700 kHz. The two semicircles observed at 0.1 MHz disappeared in the M'' vs M' curve, indicating an enhancement of the material's conductivity. The high-frequency arc observed in these composites cannot be attributed to interface traps between the interlayer and semiconductor.

4 Conclusion

In this present study, we were especially focused on the changes in the dielectric parameters, electrical modulus, and ac electrical conductivity with both voltage and frequency for the Au/(CdTe:PVA)/n-Si MPS structure rather than the MIS structures. Therefore, these parameters were calculated from the impedance dielectric spectroscopy, including C/G-V-f measurements. Thus, the ϵ' , ϵ'' , $\tan\delta$, M'' , M' and σ_{ac} values versus voltage and frequency were drawn between wide-range frequency (2 kHz–0.7 MHz) and voltage (± 2.5 V). The experimental results suggest that the parameters are f - and V -dependent, especially at low to moderate frequencies in the depletion zone. This dependence is caused by the existence of N_{ss} , interlayer, and dipole polarizations, which are dependent on relaxation time. The obtained higher values of ϵ' ($= 128.38$ at 2.5 V) even at 2 kHz is almost 34 times higher than the maximum value of traditional SiO₂. Additionally, the frequency-dependent AC

conductivity data reinforces this conclusion. The $\ln(\sigma_{ac})$ – $\ln(f)$ plot for three different bias voltages also exhibits three linear regimes, supporting the hypothesis of hopping conduction in the composites. The higher values of ϵ' observed for the (CdTe:PVA) interlayer suggest that it can be used as a successful alternative to traditional insulators for ultra-capacitors, electronic charge storage, or energy storage. The two semicircles observed at 0.1 MHz are absent in the M'' vs M' curve, indicating an enhancement in the material's conductivity.

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Author contributions

Çiğdem Şükriye Güçlü: Manufacturing, investigation, measurements, calculations, writing, reviewing, and editing. Şemsettin Altındal: Reviewing, editing, and supervising. Murat Ulusoy: Investigation, measurements, calculations, reviewing, and editing. Ayşegül Eroğlu Tezcan: Writing, calculations, and editing.

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Data availability

The data that support the findings of this study are available upon reasonable request from the authors.

Declarations

Conflict of interest The authors declare that they have no known competing financial interests or per-

sonal relationships that could have appeared to influence the work reported in this paper.

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