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AC electrical behavior of nanostructure thin film ClAlPc sandwich devices with aluminum electrodes

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The AC electrical behavior of sandwich devices manufactured totally in vacuum using thermally evaporated nanostructure thin films of Chloroaluminium Phthalocyanine with aluminum electrodes (Al/ClAlPc/Al) is examined over the range of frequency 10^2 to 10^5 Hz and the temperature range 308 to 408 K. Capacitance increase with increasing temperature and decrease with increasing frequency and dissipation factor ($\tan\delta$) decrease with increasing frequency and has a pronounced maximum with increasing temperature at a given constant frequency. At low frequency the capacitance and the dissipation factor are more temperature dependent than frequency; such behavior has been shown to be in good agreement with the model of Simmons et al. In this research work the over the range of frequency < 10^3 Hz the band theory and over the range of frequency > 10^3 Hz the hopping mechanism is applicable in explaining the conduction process in ClAlPc thin films with aluminum electrodes.

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1. Introduction

Organic and polymeric electronic devices have enjoyed increasing interest because of their potential low-cost applications, easy processing, great opportunity to modify their chemical structures and good compatibility with a variety of substrates. Out of them Phthalocyanines (Pcs) plays an important role as low gap semiconductor materials [1]. These materials represent a large family of heterocyclic conjugated molecules with high chemical stability. The study of these compounds is very essential to understand the behavior of their electronic physical properties under various conditions: such as changes in temperature, pressure, frequency, ambient gases, etc [2]. DC electrical properties of various metal Phthalocyanine thin film devices have been extensively studied in recent years [3-4], but relatively little work has been done on AC electrical behavior. Sandwich devices can be used for AC measurements of capacitance as well. AC conductivity and dielectric measurements of metal Phthalocyanines was the subject of several investigations in last decades [5-6]. DC measurements of dark current provide information on conduction process in the devices. For example, it is possible to determine whether the process is electrode limited or bulk limited [7] by varying the type of electrode metal, applied electric field strength and operating temperature. AC measurements however, yield information which can be used to determine the intrinsic conduction process within the film that can be described by the hopping model, variable range hopping (VRH) or band theory, under particular operating condition [8]. The nature of the electrode material is of great importance in determining the type of conductivity observed, so in the recent years, several works have performed with several different electrodes [9]. In present work, we studied AC conductivity, capacitance, dissipation factor (tanδ) of Chloroaluminium Phthalocyanine (ClAlPc) sandwich devices with aluminum electrodes, Such measurement have been made in order to probe the electrical properties ClAlPc thin film sandwich structures having two Al electrodes and the nature of the Al / ClAlPc contact. In present work it can be seen the conduction mechanism dependent on the range of frequency is the band theory and hopping mechanism and behavior in agreement with Simmons et al model so the schottky contact of Al/ClAlPc in this condition. There are no reports on the electrical properties of simple ClAlPc sandwich structures in which both electrodes are aluminum.

2. Experimental

Al / ClAlPc / Al thin film sandwich devices fabricated by sequential thermal evaporation under a vacuum of approximately 10^{-5} mbar onto pre-cleaned glass substrates. The ClAlPc evaporation rate used was 0.8 nm per second and the thickness was monitored during deposition using a quartz crystal monitor, the thickness of Phthalocyanine films were 130-140 nm. The evaporation rate of electrodes (aluminum) was 0.5 nm per second and the thickness of the resulting Al films was 30-40 nm. A molybdenum boat was used for evaporating the Phthalocyanine layers. The Al electrode was evaporated using a tungsten boat. The electrical contacts were equipped with thin copper wires and mechanically applied to the metal electrodes using silver paint then electrical measurements were made with the film in the dark condition by MT4080A LCR multi-frequency meter. AC dependence of capacitance and dissipation factor on frequency and temperature in the range of 10^3 - 10^5 Hz and $\boldsymbol{T}$ = 303 - 413 K respectively were made. AC conductivity has been evaluated from dielectric data in accordance with the equation (1).

$$\sigma_{ac} = \omega \frac{Cd}{A} \tan\delta \tag{1}$$

Where ω the angular frequency, ω = 2πf, $\tan\delta$ = Dissipation factor, ***d*** is the thickness of sample (m), ***C*** the capacitance (F) and ***A*** the area of cross section of the sample (m^2) [10].

We can see in Fig. 1 and 2 the size of nanostructure particles in aluminum and Chloroaluminium Phthalocyanine thin films evaporated are 53 nm and 78 nm respectively.

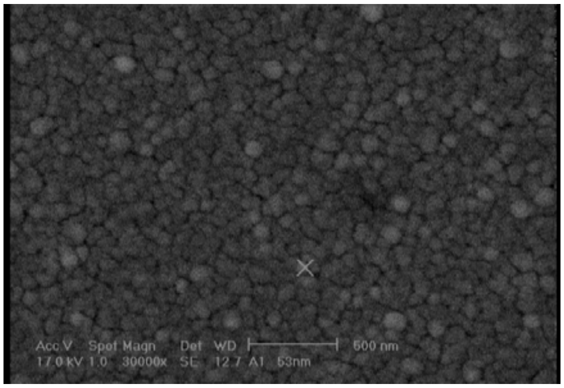

Fig. 1. SEM of aluminum evaporated nanostructure thin film.

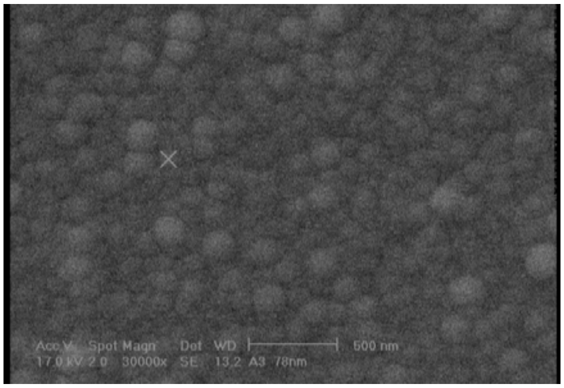

Fig. 2. SEM of ClAlPc evaporated nanostructure thin film.

3. Results and discussion

3.1 Capacitance and dissipation factor

In the current report we are investigating the dependence of capacitance and dissipation factor on frequency and temperature for Al / ClAlPc / Al structure. The frequency dependence of capacitance for the devices over the range of frequency 10^5 to 10^2 Hz was measured at temperatures 308, 318, 333, 353, 373, 393 and 408 K as show in Fig. 3. Fig. 4 shows the temperature dependence of capacitance for the device over the range of temperature 308 to 408 K at frequencies 10^2, 10^3, 10^4 and 10^5 Hz .The capacitance is shown to be strongly frequency dependant at relatively high temperatures and at low frequencies but, became less so at low temperatures and high frequencies. Increasing capacitance with increasing temperature was interoperated as arising from an increase in the number of free carriers with increasing temperature [11].

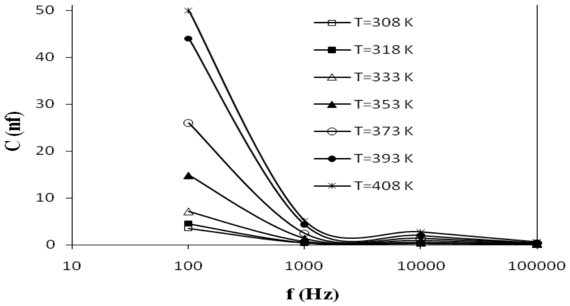

Fig. 3. Dependence of capacitance on frequency at different temperatures.

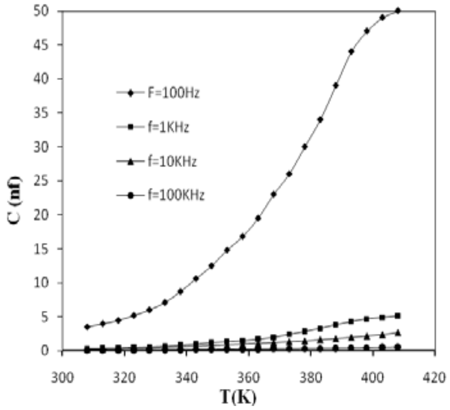

Fig. 4. Dependence of capacitance on temperature at different frequencies.

Fig. 5 shows the variation of the dissipation factor with frequency at different temperatures. It can be seen that in ClAlPc thin films dissipation factor decrease with increasing frequency and has a pronounced maximum with increasing temperature at each curve of given constant frequency. It is obvious that the temperature of the maximum point of curves increase with increasing frequency.

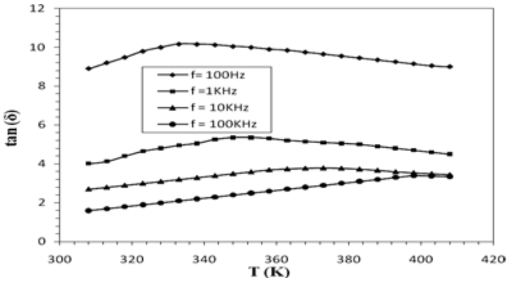

Fig. 5. Dependence of dissipation factor on temperature at different frequencies.

If we take into account of the existence of two schottky barriers set up due to the electrodes and also the capacitance due to the bulk of ClAlPc, then such behavior can be explained in terms of the AC equivalent-circuit model proposed by Simmons et al [12] as shown in Fig. 6.

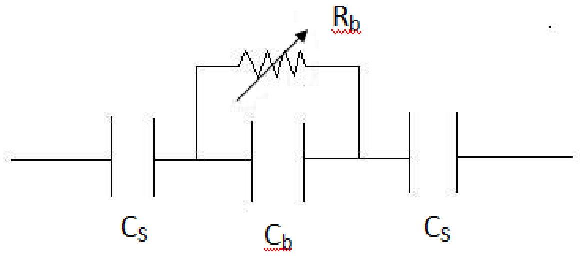

Fig. 6. Equivalent circuit model of Simmons et al.

In this proposed model, each capacitor system is assumed to be comprised of two schottky barrier capacitance, C_s in series with the bulk capacitance, C_b which is itself in parallel with the temperature dependent resistance, $R_b = R_0 \exp\frac{\Delta E}{KT}$ where ΔE is the activation energy. The capacitance of total system is given by [13] as

$$C = \frac{C_s}{2}\left[\frac{1+\omega^2 R_b^2 C_b\left(\frac{C_s}{2}+C_b\right)}{1+\omega^2 R_b^2\left(\frac{C_s}{2}+C_b\right)^2}\right] \tag{2}$$

So at low temperature and high frequency equation (2) becomes

$$C = \frac{C_b C_s}{C_s + 2C_b} \tag{3}$$

Similarly at high temperature and low frequency it is independent of the film thickness and corresponds to the capacitance of the schottky barriers [14] and it reduces to

$$C = \frac{1}{2} C_s \tag{4}$$

Using equation (4) together with measured value of C = 6 nf at high temperature and low frequency (308 K and 10^3 Hz), a schottky capacitance value obtains $C_s = 12$ nf. Similarly in equation (3) by assuming the same value of C_s together with measured capacitance C = 0.5 nf at low temperature and high frequency (408 K and 10^5 Hz), a bulk capacitance of $C_b = 0.54$ nf is obtained. This value is consistent with the measured value of capacitance at low temperature and high frequency that indicates that the total capacitance is governed by the bulk capacitance at low temperature and high frequency.

Also this model predicts a pronounced maximum for dissipation factor with increasing temperature at given constant frequency [12] as we can see in our results.

3.2 Conductivity

Fig. 7 shows the frequency dependence of the AC conductivity of the devices at different constant temperatures. It is obvious that the conductivity is strongly frequency dependent. Over the range of frequency < 10^3 Hz the conductivity decreases with increasing frequencies at constant temperatures. Over the range of frequency > 10^3 Hz the conductivity increases with increasing frequencies at constant temperatures. Generally, decrease of conductivity with increasing frequency is associated with band-type condition process, while increase with a hopping-type condition mechanism. Therefore, over the range of frequency < 10^3 Hz the band theory and over the range of frequency > 10^3 Hz the hopping mechanism is applicable in explaining the conduction process in ClAlPc thin films with aluminum electrodes.

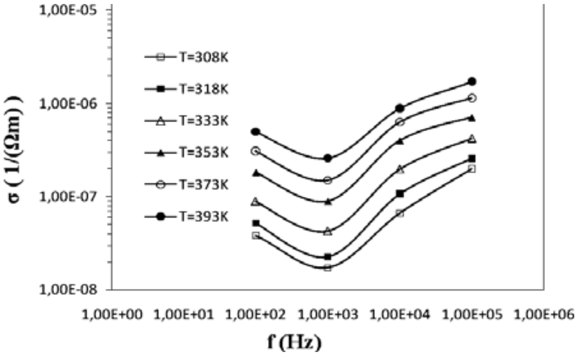

Fig. 7. Dependence of AC conductivity (σ_{AC}) on frequency at different constant temperatures for Al / ClAlPc / Al devices.

Over the range of frequency > 10^3 Hz the conductivity obeys the power law [15]:

$$\sigma_{AC} = A\omega^s \tag{5}$$

Where A is a constant independent on temperature, ω the angular frequency, ω = 2πf and s is the frequency exponent. The exponent s generally has values in the range $0 < s \leq 1$ and it has been found to decrease with increasing temperature. Hassan and Gould [16] have shown that the AC conductivity for CuPc is proportional to ω^s. In particular, for law frequency and low temperature region the exponent s decreases linearly with increasing temperature. This behavior is consistent with the correlated barrier hopping (CBH) model for carriers over the potential barrier separating two centers in a random distribution [17]. In this model the temperature dependence of the frequency exponent s can be expressed as

$$s = 1 - \frac{6KT}{W_m} \tag{6}$$

Where W_m is the effective potential barrier at infinite intersite separation ($R = \infty$), which for the case of two-electron transition, is given as

$$W_m = \frac{2e^2}{\pi \varepsilon \varepsilon_0 R} + W \tag{7}$$

Where $\varepsilon\varepsilon_0$ the permittivity of the semiconductor, R is is the separation between the neighboring sites and W the hopping barrier potential after lowering of the effective barrier (W_m) due to coulomb wells overlap.

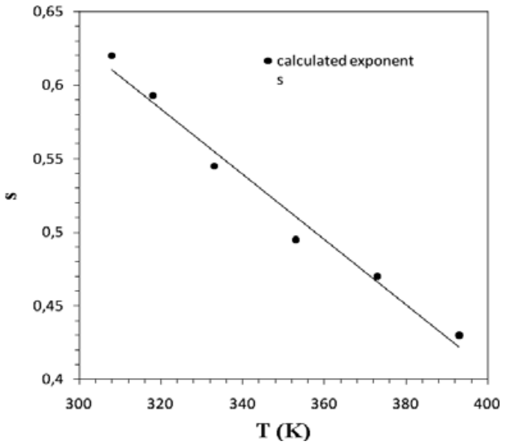

Fig. 8. Temperature dependence of exponent s measured for Al / ClAlPc / Al devices in the frequency range 10^3 – 10^5 Hz

Fig. 8 shows the temperature dependence of the exponent s in the frequency range 10^3 – 10^5 Hz. It can be seen the agreement of the linear decrease of s with temperature with the CBH model described by equation (6). The effective potential barrier (W_m) of sample can be calculated from the slop of Fig 7 that for our sample was 0.35 eV.

4. Conclusion

The AC electrical behavior of sandwich devices manufactured totally in vacuum using thermally evaporated nanostructure thin films of Chloroaluminium Phthalocyanine with aluminum electrodes (Al / ClAlPc / Al) is examined over the range of frequency 10^2 to 10^5 Hz and the temperature range 308 to 408 K. Capacitance increase with increasing temperature and decrease with increasing frequency and dissipation factor ($\tan\delta$) decrease with increasing frequency and has a pronounced maximum with increasing temperature at a given constant frequency and temperature of the maximum point of curves increase with increasing frequency.

At low frequency the capacitance and the dissipation factor are more temperature dependent than frequency; such behavior has been shown to be in good agreement with the model of Simmons et al assuming existence of two schottky barriers set up due to the electrodes and also the capacitance due to the bulk of ClAlPc so the schottky contact of Al/ClAlPc in this condition. In this research work the over the range of frequency < 10^3 Hz the band theory and over the range of frequency > 10^3 Hz the hopping mechanism with power law $\sigma_{AC} = A\omega^s$ is applicable in explaining the conduction which s is decreasing with increasing with temperature.

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