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# Electrical conduction mechanisms and dielectric properties of thermally evaporated *N*-(*p*-dimethylaminobenzylidene)-*p*-nitroaniline thin films

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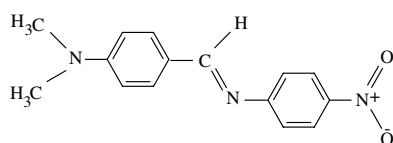
## Abstract

The electrical conductivity (DC, AC) and dielectric properties' dependence on temperature (293–393 K) and on frequency (0.1 Hz–100 kHz) of thermally deposited thin films of *N*-(*p*-dimethylaminobenzylidene)-*p*-nitroaniline (DBN) have been reported. The DC conductivity indicates a thermally activated carrier hopping rate; it increases with increasing temperature. The electronic parameters such as activation energy and room temperature conductivity are in the regime of semiconductors. The obtained experimental results of the AC conductivity have been analysed with reference to various theoretical models. The analysis shows that the correlated barrier hopping (CBH) model is the appropriate mechanism for the electron transport in DBN film. Application of the CBH model reveals that the electronic conduction takes place via bipolaron hopping processes in the whole temperature range of study. Both the dielectric constant and the dielectric loss showed a decrease with increasing frequency while they increased with increasing temperature. The barrier height,  $W_M$ , between charged defect states was calculated according to the theory of hopping of carriers over a potential barrier.

## 1. Introduction

Organic semiconductors have a growing interest for study as active components widely used in electronics and optoelectronics. Due to their flexibility, low cost and ease of production they represent a valid alternative to conventional inorganic semiconductor technology in a number of applications such as flat panel displays, plastic integrated circuits and solar energy conversion.

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**Figure 1.** The molecular structure of the DBN compound.

Because of the new commercial applications of this technology, there comes the need for their deep scientific understanding to achieve optimum device performance [1].

Aromatic Schiff bases exhibit thermochromic and photochromic properties, and have many technological applications [2–6]. The DBN compound is a member of the Schiff base family, and it has donor–acceptor groups linked by a  $\pi$  conjugated system, which makes it a stable structure and a promising compound for electrical and technological applications.

Thin film technology is well established and widely used in the fabrication of electronic devices. The technique has been successfully used to fabricate thin film resistors, capacitors, photoelectronic devices, etc [7]. The use of this technique in fabricating electronic devices makes it necessary to understand the electrical properties of the material in thin film form.

In the device applications of thin films, new and complex organic materials are being used and developed, and their electrical properties are being studied. A good amount of work on DC conduction [8–12], AC conduction [8, 13–20] and optical properties [21, 22] has been reported.

Measurement of AC conductivity of semiconductors has been extensively used to understand the conduction process [23] and it is also a powerful tool for obtaining information about the defect states in amorphous semiconductors [24, 25]. Various models such as the quantum mechanical tunnelling (QMT) model [26, 27] and correlated barrier hopping (CBH) model [24, 28] have been proposed to explain the AC conduction mechanisms.

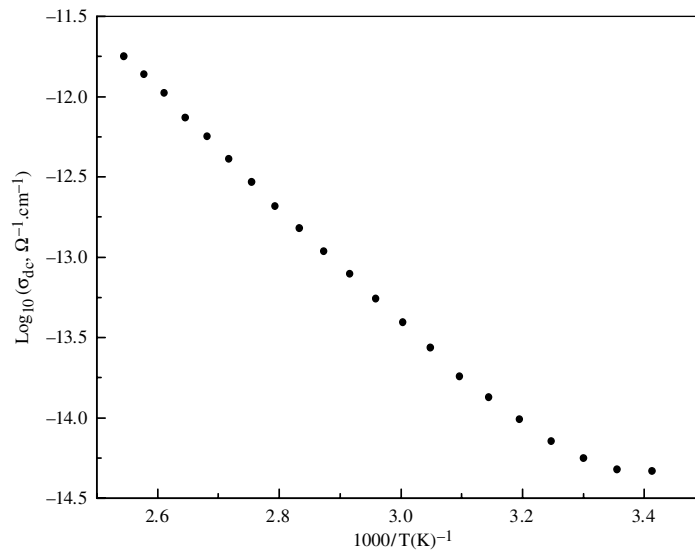
Dielectric relaxation studies are important in understanding the nature and the origin of dielectric losses, which, in turn, may be useful in the determination of structure and defects in solids. The dielectric behaviour of thin film devices depends not only on their material properties, but also on the substrate used for fabrication and the type of the metal electrodes. Fringing effects at the edges of thin film dielectrics are usually negligible because the thickness of the dielectric is usually very small compared to its lateral dimensions. The magnitude of geometric and measured capacitance may differ if the electric field at the metal insulator interface varies with the insulator over the region.

To our knowledge, no report is available on the study of DC and or AC conduction mechanisms and dielectric properties of thermally evaporated *N*-(*p*-dimethylaminobenzylidene)-*p*-nitroaniline (DBN) films. Combining DC and AC measurements at various temperatures is certainly of great importance in order to obtain a better understanding of the occurring electrical transport mechanism. We report on the influence of some environmental conditions such as temperature and applied frequencies on the (DC, AC) conductivity and dielectric properties of thermally vacuum-deposited DBN films.

## 2. Experimental details

The powder used in this study was prepared by the direct condensation of *p*-dimethylaminobenzaldehyde (purity of 99%) and *p*-nitroaniline (purity of 99%) obtained from Aldrich Company. The molecular structure of the DBN compound is depicted in figure 1.

Thin films of different thickness were prepared by thermal evaporation under vacuum ( $10^{-5}$  Torr) onto cleaned glass substrates kept at room temperature using an Edwards E306A



**Figure 2.** DC electrical conductivity of DBN thin film as a function of reciprocal temperature.

coating unit. The films were sandwiched between two gold electrodes of thickness 50 nm which act as ohmic contacts with the DBN films. The films were placed in a holder that was specially designed to minimize stray capacitance. The film thickness was monitored with a quartz crystal oscillator (Edwards FTM6), and accurate thickness measurements were determined interferometrically [29]. In all the devices used for the study, the thickness of the dielectric film was kept at least above 900 nm to minimize the inherent discrete defects and pinholes. The temperature of the sample was measured using a NiCr–NiAl thermocouple connected to the temperature controller.

The AC measurements were performed using a programmable automatic LCR meter (Stanford, model SR720). The LCR meter allows the direct measurements of capacitance, resistance and dissipation factor in both the parallel and series modes. The AC conductivity, dielectric constant,  $\epsilon_1$  (the real part of the dielectric constant) and the dielectric loss,  $\epsilon_2$  (imaginary part of the dielectric constant) were calculated from the measured values of capacitance,  $C$ , frequency loss factor,  $\tan \delta$ , film thickness,  $d$ , and cross-sectional area,  $A$ , of the capacitor.

The measurements of the DC conductivity were carried out using a high impedance programmable electrometer (Keithley model 617). These measurements were obtained using the same sample holder in which the dielectric measurements were carried out.

### 3. Results and discussion

#### 3.1. DC conductivity

The temperature dependence of the DC conductivity of the DBN thin film is presented in figure 2. Perusal of this relation reveals that the conduction is through a thermally activated process having two conduction mechanisms. The first one extends from room temperature up to 320 K and the second one is at temperatures higher than 320 K. The conductivity exhibits an activation energy dependence on temperature of the form

$$\sigma = \sigma_0 \exp(-E_a/k_B T) \quad (1)$$

**Table 1.** DC and AC electrical conductivity parameters.

Frequency (kHz)	Activation energy (eV)		Pre-exponential factor	
	$E_{a1}$	$E_{a2}$	$\sigma_{01}$	$\sigma_{02}$
0 (dc)	0.70	0.39	$1.81 \times 10^{-3}$	$2.12 \times 10^{-9}$
0.1	0.41	0.24	$5.28 \times 10^{-5}$	$1.00 \times 10^{-9}$
1	0.28	0.15	$2.10 \times 10^{-6}$	$2.17 \times 10^{-8}$
10	0.20	0.10	$8.61 \times 10^{-7}$	$1.52 \times 10^{-8}$
100	0.15	0.07	$6.48 \times 10^{-8}$	$3.76 \times 10^{-8}$

where  $\sigma_0$  is the pre-exponential factor,  $E_a$  is the activation energy, and  $k_B$  is Boltzmann's constant. Table 1 lists the calculated values of the activation energy  $E_a$  ( $E_{a1}$  and  $E_{a2}$  at high and low temperatures, respectively) and the pre-exponential factor  $\sigma_0$ . A smaller value of  $\sigma_0$  indicates the presence of the contribution of localized states and conduction occurs by phonon-assisted hopping between these localized states. The electrical conduction of the films follows a mechanism in which the electron or hole hops from one localized site to the next. Whenever it is transferred to another site, the surrounding molecules respond to this perturbation with structural changes and the electron or hole is temporarily trapped in the potential well leading to atomic polarization. The electron resides at this site until it is thermally activated to migrate to another site [20]. Another aspect of this charge hopping mechanism is that the electron or hole tends to associate with local defects, so the activation energy for charge transport may also include the energy of freeing the hole from its position next to the defect [30, 31].

### 3.2. Temperature and frequency dependence of AC conductivity

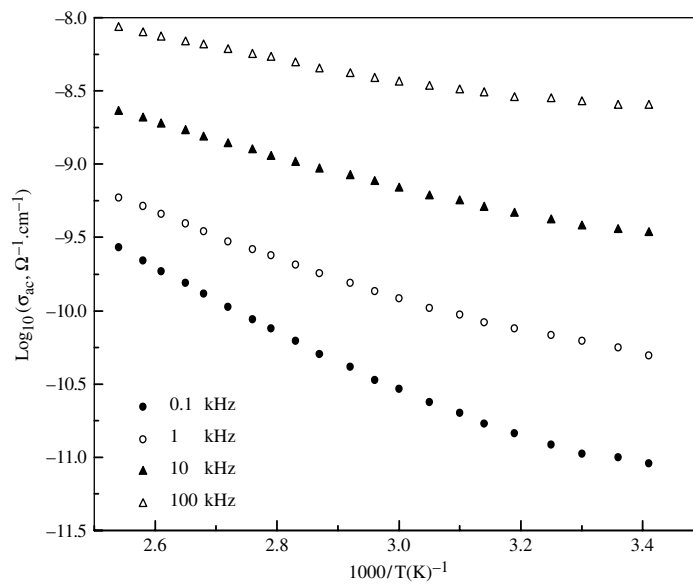
The total conductivity  $\sigma_{\text{tot}}(\omega)$  at a particular angular frequency,  $\omega$ , and at a certain temperature can be written as [25]

$$\sigma_{\text{tot}}(\omega) = \sigma_{\text{ac}}(\omega) + \sigma_{\text{dc}} \quad (2)$$

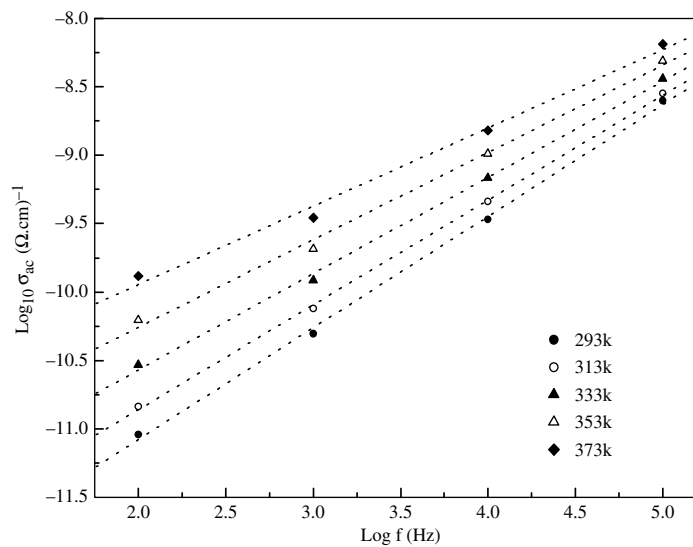
where  $\sigma_{\text{ac}}(\omega)$  is the AC conductivity and  $\sigma_{\text{dc}}$  is the DC conductivity. It is noted that equation (2) is valid when the AC and DC conductivities arise from completely separate mechanisms; otherwise the DC conductivity represents the AC conductivity in the limit  $\omega \rightarrow (0)$  [23].

Figure 3 shows the AC conductivity as a function of the reciprocal temperature at four fixed frequencies for a film sample having a thickness of 950 nm. It is evident that the temperature dependence of the conductivity has a small temperature dependence up to 325 K and a much larger dependence at temperatures higher than 325 K. It is observed from figures 2 and 3 that the value of  $\sigma_{\text{dc}}$  at a given temperature is much less than that of  $\sigma_{\text{ac}}$  at the same temperature. Such a trend indicates that the temperature dependence of AC conductivity for the DBN films is also represented by equation (1). Figure 3 shows that at a given temperature, increasing the frequency increases the AC conductivity of DBN thin films.

The activation energies and the pre-exponential factors  $\sigma_0$  have been calculated for frequencies in the range 0.1–100 kHz and are listed also in table 1. As is observed from this table, the activation energies decrease with increasing frequency. The small values of the AC activation energy and the increase of  $\sigma_{\text{ac}}(\omega)$  with increase of frequency confirm that hopping conduction is the dominant current transport mechanism. Thus, the increase of the applied frequency enhances the electronic jumps between the localized states; consequently, the activation energy,  $E_a$ , decreases with increasing frequency.



**Figure 3.** The temperature dependence of the AC electrical conductivity at four different frequencies of DBN thin film.

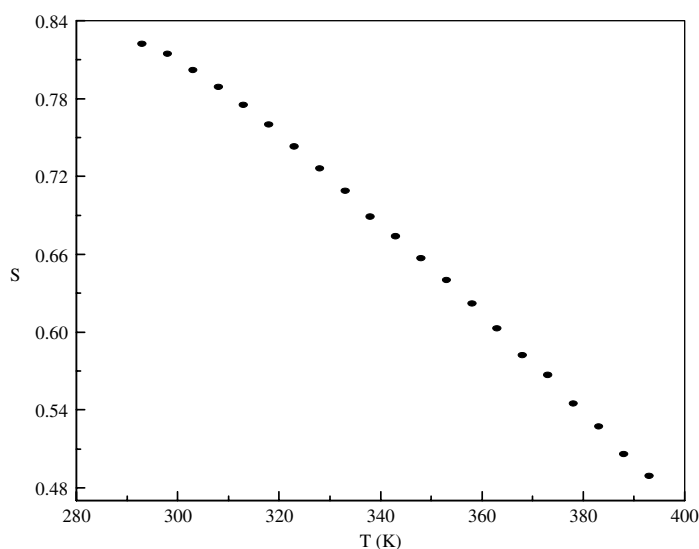


**Figure 4.** The frequency dependence of the AC electrical conductivity  $\sigma_{ac}$  at various temperatures.

The frequency dependence of  $\sigma_{ac}(\omega)$  at various temperatures is shown in figure 4. It is clear from these curves that the AC conductivity,  $\sigma_{ac}(\omega)$ , has a frequency dependence given by the equation [23]

$$\sigma_{ac}(\omega) = A\omega^s \quad (3)$$

where  $A$  is a constant depending on temperature,  $\omega$  is the angular frequency and  $s$  is an



**Figure 5.** Temperature dependence of the frequency exponent,  $s$ , as determined from the straight-line relationship between  $\log \sigma_{ac}$  and  $\log f$ .

exponent, generally less than or equal to unity. It is clear from figure 4 that the variation in the logarithmic AC conductivity is almost linear with the variation in logarithmic frequency and that  $\sigma_{ac}(\omega)$  increases with increasing both frequency and temperature. The frequency exponent  $s$  is obtained by the least squares straight-line fit of the experimental data and is plotted as a function of temperature in figure 5. The exponent  $s$  decreases as the temperature increases. The observed frequency dependence of  $\log \sigma_{ac}$  reveals that the mechanism responsible for AC conduction could be due to hopping [19, 24]. An AC conductivity, having a frequency dependence ( $\sigma_{ac} \propto \omega^s$ ) with  $s < 1$ , has been observed in organic semiconductor thin films [8, 32, 33].

In order to explain the behaviour of  $\sigma_{ac}$  with both frequency and temperature, different theoretical models have been proposed to correlate the conduction mechanism of AC conductivity with  $s(T)$  behaviour [34]. Theories proposed for AC conduction in amorphous semiconductors [27, 35] have mostly assumed that carrier motion occurs through quantum mechanical tunnelling (QMT) between localized states near the Fermi level. For the QMT model, the frequency exponent,  $s$ , is temperature independent but frequency dependent. This conclusion is further strengthened from the  $s$  versus  $T$  plot in figure 5. It is observed that  $s$  decreases with increasing temperature in contrast with the QMT model, which predicts a temperature-independent  $s$  of a value of 0.81 [23].

The temperature dependence of  $s$  can be met in the tunnelling models if the carrier forms a small or large polaron. The small-polaron tunnelling model [7, 25] is also not a suitable mechanism for explaining the results of DBN films because it predicts an increase in  $s$  with increase in temperature, in sharp contrast with the experimental observation shown in figure 5. The large-polaron tunnelling model is also not applicable for the present film system, since this model predicts a minimum in the temperature dependence of  $s$ , which is not observed in figure 5.

In the classical hopping over a barrier (HOB) model [25, 36] the value of frequency exponent,  $s$ , is 1, and this rules out the applicability of this model to the DBN film system.

**Table 2.** AC electrical conductivity parameters obtained by fitting the experimental data to the correlated barrier hopping (CBH) model for DBN thin films.

$s$	$W_m$ (eV)	$N$ (cm <sup>-3</sup> eV <sup>-1</sup> )	$R_\omega$ (cm <sup>-1</sup> )	$W$ (eV)	$\tau$ (s)	$W_M$ (eV)
0.81	1.30	$1.17 \times 10^{18}$	$4.21 \times 10^{-7}$	0.49	$1.61 \times 10^{-5}$	0.07

We invoke the correlated barrier hopping (CBH) model to explain the observed behaviour. In the CBH model, the electrons in charged defect states hop over the Coulomb barrier, the height of which is given as

$$W = W_m - (ne^2/\pi\epsilon_1\epsilon_0R) \quad (4)$$

where  $n$  is the number of polarons involved in the hopping process ( $n = 1$  or  $2$  for the single and bipolaron hopping processes, respectively),  $e$  is the electron charge,  $R$  is the distance between the hopping sites,  $\epsilon_1$ ,  $\epsilon_0$  are the dielectric constants of the material and free space, respectively and  $W_m$  is the maximum barrier height (for a bipolaron  $W_m$  is approximately equal to the band gap width).

The relaxation time,  $\tau$ , for electrons to hop over a barrier of height  $W$  is given by

$$\tau = \tau_0 \exp(W/k_B T) \quad (5)$$

where  $\tau_0$  is the characteristic relaxation time, which is of the order of an atomic vibrational period. In the pair approximation model, a major contribution to the AC conductivity arises from hopping within the pairs of sites for which  $\omega\tau = 1$ .

According to the CBH model [23, 25, 28], the AC conductivity can be expressed as

$$\sigma_{ac}(\omega) = n\pi^3 N^2 \epsilon_1 \epsilon_0 \omega R_\omega^6 / 24 \quad (6)$$

where  $N$  is the density of localized states at which carriers exist. The temperature dependence of  $\sigma_{ac}$  originates from the hopping length,  $R_\omega$ , in equation (6), where

$$R_\omega = ne^2/\pi\epsilon_1\epsilon_0/[W_m + k_B T \ln(\omega\tau_0)]. \quad (7)$$

The frequency dependence of  $\sigma_{ac}(\omega)$  in this model arises from the factor  $\omega R_\omega^6$ . The temperature dependence of the frequency exponent  $s$  arises due to the temperature dependence of  $R_\omega$  and is evaluated to be

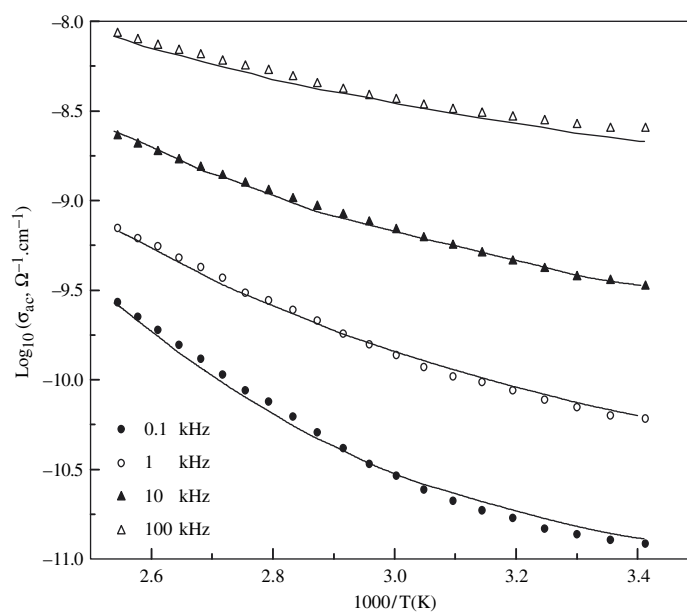
$$s = 1 - 6k_B T/[W_m + k_B T \ln(\omega\tau_0)]. \quad (8)$$

Thus in the CBH model a temperature dependent exponent,  $s(T)$ , is predicted, with  $s$  decreasing as the temperature increases and  $s$  increasing towards unity as the temperature tends to zero, in marked contrast to the QMT or simple HOB mechanism. The temperature dependence on  $s$ , shown in figure 5, is consistent with equation (8), indicating the dominance of a CBH model as a transport mechanism for the DBN film system. The values of  $N^2$  and the maximum barrier height,  $W_m$ , are adjusted to fit the calculated curves (as shown in figure 6 using equation (6) with  $n = 2$ ) with the experimental results. Fitting is made at 10 kHz and the same values are used for the other frequencies. The various parameters used in the fitting procedure are given in table 2. As is clear, the fitting is reasonably good, indicating that the conduction mechanism is through a bipolaron hopping mechanism.

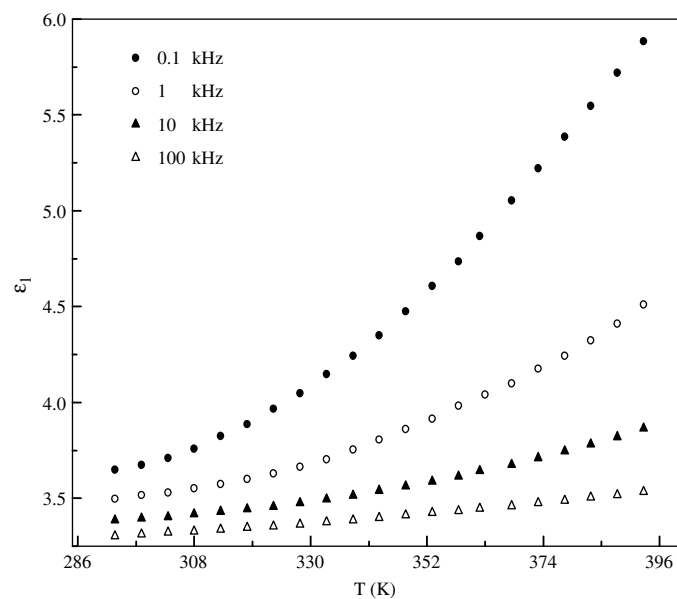
### 3.3. Dielectric properties

The temperature and frequency dependence of the dielectric constant,  $\epsilon_1$ , and dielectric loss,  $\epsilon_2$ , are studied for film samples in the temperature range 293–393 K and frequency range 0.1–100 kHz. The results are shown in figures 7 and 8, respectively. A comparative study of these





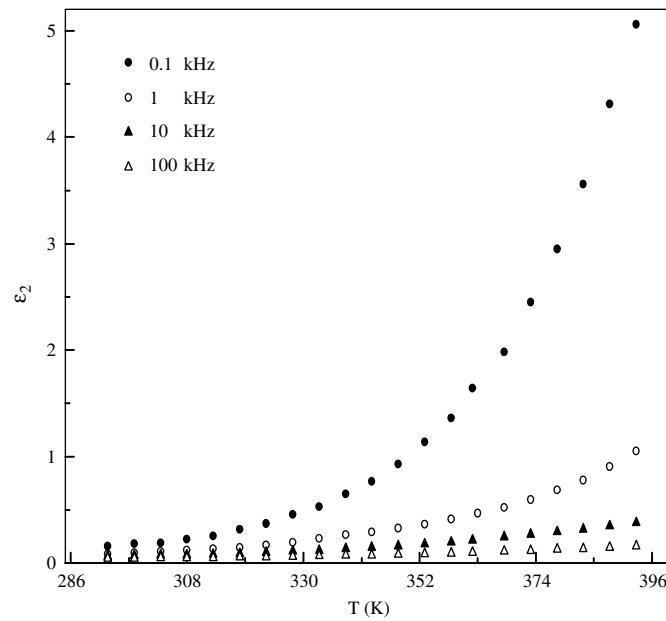
**Figure 6.** The CBH model fitting to the temperature dependence of the AC electrical conductivity,  $\sigma_{ac}$ , at various frequencies for DBN thin film.



**Figure 7.** The temperature dependence of the dielectric constant,  $\epsilon_1$ , at different frequencies.

figures indicates that  $\epsilon_1$  and  $\epsilon_2$  increase with the increase of temperature (the increase being different at different frequencies) and they decrease with increasing frequency. This type of behaviour has also been reported in organic films [8, 17, 32].

The variation of  $\epsilon_1$  with the temperature is related to the charge carriers which in most cases cannot orient themselves with respect to the direction of the applied field; therefore,



**Figure 8.** Dielectric loss,  $\varepsilon_2$ , as a function of temperature at four fixed frequencies.

they possess a weak contribution to the polarization and the dielectric constant  $\varepsilon_1$ . As the temperature increases, the bound charge carriers get enough thermal excitation energy to be able to respond to the change in the external field more easily. This in turn enhances their contribution to the polarization leading to an increase of the dielectric constant  $\varepsilon_1$  of the sample [37]. When the frequency is increased, the dipoles will no longer be able to rotate sufficiently rapidly, so their oscillation will begin to lag behind this field, which explains the observed decrease in  $\varepsilon_1$  with frequency.

It is shown that the dielectric loss,  $\varepsilon_2$ , increases as the temperature increases for the considered frequencies; at low temperatures, this increase is linear and the variation of  $\varepsilon_2$  with frequency is small, this linear region extending to higher values of temperature with increasing frequency, while at higher frequencies, it increases nonlinearly. This behaviour can be clarified by plotting  $\ln \varepsilon_2$  versus  $\ln f$  for various temperatures as shown in figure 9. It is observed that a series of straight lines with different slopes is obtained, in which  $\varepsilon_2$  decreases as the frequency increases and it increases as the temperature increases.

The frequency dependence of  $\varepsilon_2$  follows the relation [38]

$$\varepsilon_2 = (\varepsilon_s - \varepsilon_\infty) 2\pi^2 N (ne^2/\varepsilon_s)^3 k_B T \tau_0^m W_M^{-4} \omega^m. \quad (9)$$

According to the proposed theory,  $\varepsilon_2$  should follow a power law with frequency, i.e.

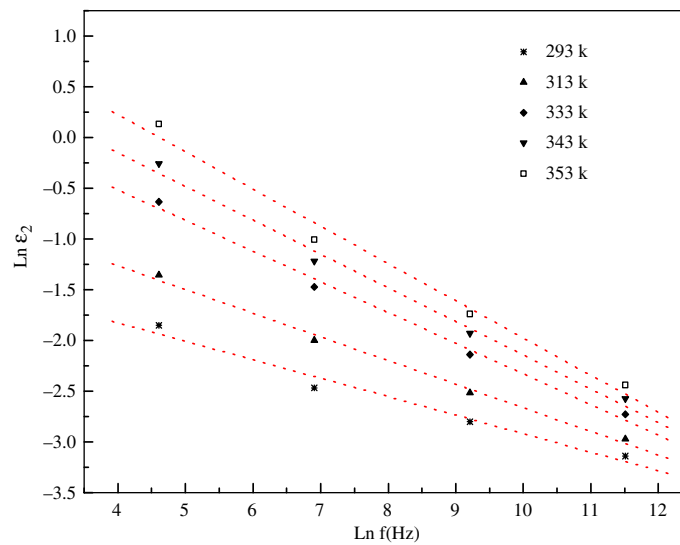
$$\varepsilon_2 = G\omega^m \quad (10)$$

with

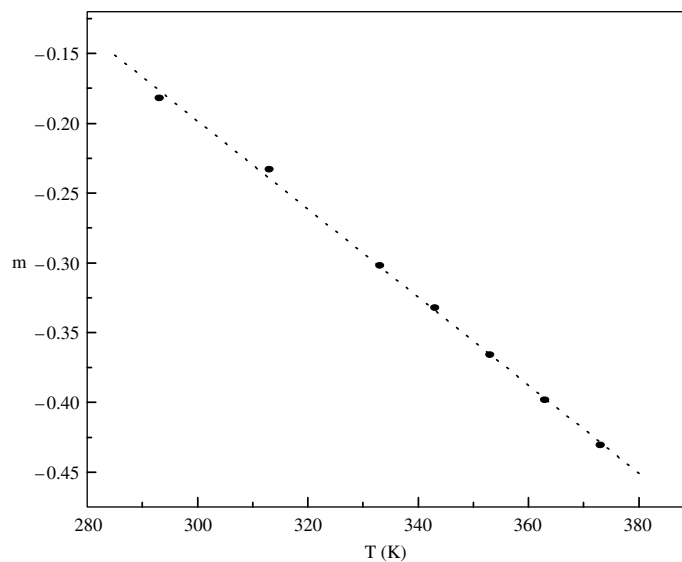
$$m = -4k_B T / W_M \quad (11)$$

where  $W_M$  is the barrier height,  $\varepsilon_s$  is the static dielectric constant,  $\varepsilon_\infty$  is the dielectric constant at infinite frequency,  $G$  is a constant depending on temperature and the other symbols have the same meaning as before.

The power,  $m$ , is calculated from the slopes of the curves of figure 9 and it was found that the values of  $m$  are negative, and it decreases linearly with increasing temperature as shown in



**Figure 9.**  $\ln \varepsilon_2$  versus  $\ln \omega$  at a certain fixed temperature.  
(This figure is in colour only in the electronic version)



**Figure 10.** Temperature dependence of the parameter  $m$ .

figure 10; this is in agreement with the prediction of the theory. The parameter,  $m$ , confirms the observed variations as a function of the measuring temperature. In fact, this result is satisfying if we consider the empirical law [39, 40]:  $\sigma_{ac}(\omega) = \omega \varepsilon_2(\omega) = A\omega^s$ . It is obvious that if  $s$  is temperature dependent [24, 28],  $m$  should consequently depend on  $T$ .

#### 4. Conclusions

The DC and AC electrical conductivity, dielectric constant and dielectric loss of thermally evaporated *N*-(*p*-dimethylaminobenzylidene)-*p*-nitroaniline thin films have been reported.

The DC electrical conductivity indicates semiconductor behaviour: it increases with increasing temperature. It was found that the correlated barrier hopping (CBH) model is the probable model for conduction mechanism in DBN thin film. Application of the CBH model reveals that the electronic conduction takes place by bipolaron hopping processes in the whole temperature range of study. The different parameters obtained from the fits of this model to the experimental data are reasonable. The values of dielectric constant,  $\epsilon_1$ , and dielectric loss,  $\epsilon_2$ , increase with increasing temperature and with decreasing frequency. Values of the maximum barrier height were estimated from the data of dielectric loss, and are in good agreement with the theory of hopping of charge carriers over a potential barrier between charged defect states. The value of the barrier height,  $W_M$ , is calculated according to the Giuntini *et al* model [38].

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