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Polymer dipoles relaxation and potential energy (New Simulation Model)

Abbass A. Hashim

Centre for Automation and Robotics Research (CARR)

Material and Engineering Research Institute

Sheffield Hallam University, City Campus, Pond Street, Sheffield, S1 1WB, UK

1. Introduction

Development of polymer electronic devices is one of the most interesting and required in the recent year industrial technology.

Polymeric material has various characteristics which can be controlled and monitored. These characteristics are including light-weight, mechanical flexibility, high-dielectric strength, fracture tolerance, high chemical resistance, easy processibility, and low manufacturing cost. Moreover they can be configured into almost any conceivable shape and their properties can be tailored to suit many applications.

Nowadays and near future exactly, electronic-polymer represents an important piece of the electro-chemistry and many technological advances. Thus come from the combination of different materials in electrochemical cells.

New electro-active polymeric materials are always in the priority market requirements, with different properties, such as electroluminescence, semiconductor behaviour, electronic and ionic properties, electrochromism, etc (De Paoli & Gazotti, 2002; Noh et al., 2006).

Wide range of electronic components from micro to nano scales are in research from academics, industry, and national laboratories to present and discuss the recent research and commercial advances and needs. The researchers focus on the topics related to materials development, characterization, processing, manufacturing, analysis, device designing, implementation and applications.

The need for such nanoscale and microscale are demanding requirements for polymers as dielectrics, which have been used for insulators and charge-storage applications. Exploring polymer-based dielectrics with extremely low-k and high-k has recently become an important area of research and development. The influence of environment on electrical insulation and space-charge properties is the access topic for research on dielectric polymers (Taylor, 2006).

Recently, a significant highly competition in the markets of the tiny electronic chips technology specially and an extremely demands in the mobile industry and computer manufacturing. The future of electronic chip technology depends on the development of dielectric materials with low dielectric constant (K less than 2.2). The channel length of chip device approaches $0.1 \mu\text{m}$, the traveled signal delay on an integrated circuit chip is

dominated by the interconnect wiring. To reduce the size of the interconnect wiring; crosstalk between two adjacent lines dictates the minimum allowable spacing. This crosstalk is directly dependent upon the dielectric constant of the insulating material (Modafe et al., 2006).

Large number of capacitors employed in electronic systems, integration of capacitors is of great importance. The development of microelectronics requires decoupling capacitors with higher capacitance and shorter distance from their serving devices. In particular, the high-K materials are required for making embedded capacitors for integrated electronic devices (Müller et al., 2006; Lee et al., 2009; Popielarz & Chiang, 2007)

Microelectronic embedded capacitors are considered as a promising enabling technology. Development of an organic capacitor substrate compatible high dielectric constant material is currently unavailable (Hwang et al., 2008).

Polymer-ceramic nano-composites create potentially high-K materials. This approach could combine the low-temperature processibility of the organic polymer matrix and the high dielectric constant of the ceramic filler.

The devices working at high operating frequencies, such as fast computers, cellular phones, etc., require new high-dielectric constant (high- K) materials that combine good dielectric properties with both mechanical strength and ease of processing. The unique combination of dielectric and mechanical properties is hard to achieve in a one component material. Pure polymers are easy to process into mechanically robust components but generally suffer from a low dielectric constant. These polymers can be used for this application but the dielectric constant has to be improved to create material with high- K (Fan et al., 2002).

Typical high- K materials, such as ferroelectric ceramics, are brittle and require high-temperature processing, which is often not compatible with current circuit integration technologies. The model solution would be a high- K material that is mechanically robust and processable at ambient temperatures. This has raised a great interest in hybrid materials, such as ferroelectric ceramic/polymer composites, that may combine desired properties of the components (Popielarz et al., 2001).

Moreover electronic devices are transducing polymers (TPs), smart polymers (SPs), and electro-responsive polymers (ERPs) respond to an electric, magnetic, mechanical thermal, optical, chemical, and other stimulation, and respond with a change that includes mechanical, electrical, optical, and thermal stimuli, and many others, or vice versa. The transducing nature of these polymers led to their use as sensors and actuators. The development of miniature electronics and MEMS/NEMS has attracted a great deal of attention to the development of more sophisticated transducing polymers with enhanced performance in macroscale to microscale, and even down to nanoscale (Kassiba et al., 2007).

The research subjects mentioned above are promising research field in which needs to understand the variation in the dielectric properties due to the polymer thin film internal structure modification. This study is attempting to view the tiny structural changes in polymer structure and its affect on the dielectric phase properties as a result of any kind of treatments; chemical, thermal or mechanical.

2. Polymer dielectric properties

Understanding of polymer molecular dipole influences and field interactions on dielectric characteristics is a fundamental issue for the development of electronic device applications.

Three types of interaction of the polymer molecule with the electrical field (polarization) can be determined. These are possibly to be classified as electronic, atomic and orientation polarization. In the area of thin film, molecule orientation polarization is the most essential.

The electrical field tries to align the dipolar molecules along its own direction. This effect is hindered by thermal motion of the molecules. As a consequence orientation polarisation decreases when the temperature increases.

The electronic and atomic polarisation is independent of the temperature. The orientation polarisation is not observed when the thermal motion of the molecules is hindered, since rigidly bounded molecules or polar groups are move under the effect of the electrical field. This situation occurs at low temperatures. The orientation polarisation becomes possible when a certain, relatively high temperature is attained during the heating of the material. Increasing the temperature, the thermal energy of various polar molecules and groups successively increases, permitting orientation processes. However, the polarisation increases only in a relatively narrow range of temperature, because at higher temperature the intense thermal motion favours the disorientation of the dipoles and the polarisation decreases.

If an electric field is applied on a dielectric material, the variation of the field is followed by polarisation with a certain delay. This delay is a phenomenon common to all the three types of polarisation, and it is called "relaxation effect", characterized by the relaxation time, the time interval during which polarisation decays to the e -th fraction of its initial value. The relaxation frequencies (times) are different for the various types of polarisation. The decay of electronic polarisation is the most rapid because of the low inertia of electrons, while that of atomic polarisation is slower. The relaxation time of atomic polarisation is in the range of the period of infrared electromagnetic radiation, while the relaxation time of electronic polarisation is in the range of visible light. The relaxation time of the orientation polarisation depends on different parameters of the polar molecules (for example: size, molecular environment, etc.) and appears in the range from the radio frequencies and microwaves to several weeks.

3. Temperature dielectric relaxation

Mort and Pfister (Mort & Pfister, 1982) have shown that several distinct dielectric relaxation processes can exist in solid polymers. This is observed more clearly when the dielectric loss is studied as a function of the temperature at a given frequency.

As the temperature increases, the molecular mobility of the polymer increases leading to more dipole orientation. By convention, the dielectric relaxation processes are labelled α , β and so on, beginning at the high temperature end. The same relaxation processes are generally responsible for dispersions when mechanical properties are considered, although a particular molecular rearrangement may produce a stronger dielectric than the mechanical effect, or vice versa.

Some polymers are wholly amorphous and most of the solid materials have two phases of relaxation in solid materials. In such cases there is always a high temperature α - relaxation associated with the micro-Brownian motion of the whole chain and, at least one low-temperature (β , γ , etc) subsidiary relaxations. The relative strength of α and β - relaxations depends on the dipole group orientation which is limited by the mobility of the β - process prior to the α -process of the higher mobility. The potential barrier U of the dipole stable

levels was studied since this leads to the values of ϵ' , ϵ'' and consequently the mode of the relaxation (Blythe & Bloor, 1980).

4. Theoretical Model

The specimen may be regarded in terms of a series circuit. If the equivalent series components of capacitance and resistance are C_s and R_s , respectively, the total impedance will be given by (Blythe & Bloor, 1980):

$$Z = R_s + \frac{1}{j\omega C_s} \quad (1)$$

By comparing the out of phase and in phase currents after application of the alternating voltage, the imaginary part of the complex dielectric constant ϵ'' , can be calculated using the measured values of R_s and C_s (Blythe & Bloor, 1980):

$$\epsilon'' = R_s \frac{C_s}{C_o(1+\tan^2\delta)} \quad (2)$$

C_o is the free space capacitance and $\tan \delta$ is the dissipation factor.

The temperature dependence of the dipole relaxation time constant τ often follows Arrhenius' law (Smith, 1955):

$$\tau = \tau_o \exp\left(\frac{U}{kT}\right) \quad (3)$$

Where τ_o is the free relaxation time constant at the high temperature range and τ is the time constant defined by the circuit parameters:

$$\tau = R_s C_s \quad (4)$$

Equations (2) can be modified using $\tan \delta = R_s C_s \omega$:

$$\epsilon'' = \frac{\tau_o e^{(U/kT)}}{C_o \left[1 + \tau_o^2 \omega^2 e^{(2U/kT)} \right]} \quad (5)$$

In the most cases the applied frequency makes $\tau_o^2 \omega^2 e^{(2U/kT)} \ll 1$, and equations (5) can be simplified as:

$$\epsilon'' = \frac{\tau_o}{C_o} e^{(U/kT)} \quad (6)$$

Most of the parameters are temperature independent, except the term $\exp\left(\frac{U}{kT}\right)$, which show greater temperature dependence at high temperature range.

5. The U Energy Bands Model

Experimental data have prove that the effect of the term $\exp\left(\frac{U}{kT}\right)$ in the ϵ'' -curves (equation 9) is more pronounced than in the ϵ' -curves (equation 8) and the small variation in U with temperature is controlled the relaxation processes. However, the U/T curves are more compatible with ϵ''/T curves when the polynomial fitting method was used.

Whereas, all U polynomial fitting curves are shifted on the temperature scale relative to the ϵ'' -curves. Such a shift can add to modified equation (9) in terms of the correction shift parameter or polynomial error (P):

$$\epsilon'' = \frac{\tau_o}{C_o} \exp\left[\frac{-1}{kT} \sum_{i=0}^n a_i T^i\right] \quad (10)$$

where $T = T_o \pm P$

$$\epsilon'' = \frac{\tau_o}{C_o} \exp\left[\frac{-1}{k(T_o \pm P)} \sum_{i=0}^n a_i T^i\right] \quad (11)$$

Where T_o is the real temperature, T is the calculated temperature and n is the polynomial degree.

ϵ' and ϵ'' can be plotted against the temperature using equations (2) and (3). U can be deduced from $R_s C_s$ measurements using equations (4) and (5) as follow:

$$\ln R_s C_s = \ln \tau_o + \frac{U}{kT} \quad (12)$$

6. Implementation of the new model

6.1 Dielectric properties of a chosen example

The dielectric properties of polar polymers depend on the position, direction and the length of the dipolar group with respect to the chain. Two main relaxations were observed, namely; the high- temperature α relaxation which is associated with chain backbone movement due to the rotation of the Ethers groups (C-O-C) around the main chain (crankshaft motion), and the low-temperature β -relaxation due to the hundred rotation of the acrylate groups $-COOR$ around the C - C bond linking it to the chain (Blythe & Bloor, 1980; McCrum et al, 1967; Boyer, 1982). It is not known, however, how many chains are involved in the movement

Figure 1 shows the ϵ''/T -curves for PMA, in which an α -relaxation process can be identified clearly at a temperature of 36°C. The PMA molecules show no degree of freedom at 0°C due to the acrylate and hydrogen side groups, but the degree of freedom increases as the temperature approaches 20°C. The molecules absorb enough energy, giving the chain backbone enough kinetic energy to vibrate with the field frequency and show the α -peak. The joint motions of the side groups and the main chains gives rise to the α -relaxation. The

β -relaxation originates from the movements of the acrylate group ($-COOCH_3$) at $19^\circ C$ (Blythe & Bloor, 1980). This group ($-COOCH_3$) has enough space to rotate at room temperature. It should be noted that, the acrylate group ($-COOCH_3$) controls the dielectric properties of the PMA, making the β and α processes close to each other and overlapping in a single wide peak. The curves in Figure 1 represent the average results of two repeated dielectric spectroscopy measurements with an error of ± 0.035 in the ϵ'' curve which are within the range of the data point resolution.

The structure of PMA is one of the most simplest and regular structure in polar polymers. PMMA and PMA have the same acrylate groups with different r groups. This fact means that PMA has more polarity than PMMA due to the effect of the hydrogen atom in its r -group while in PMMA, $r = CH_3$ is more stable.

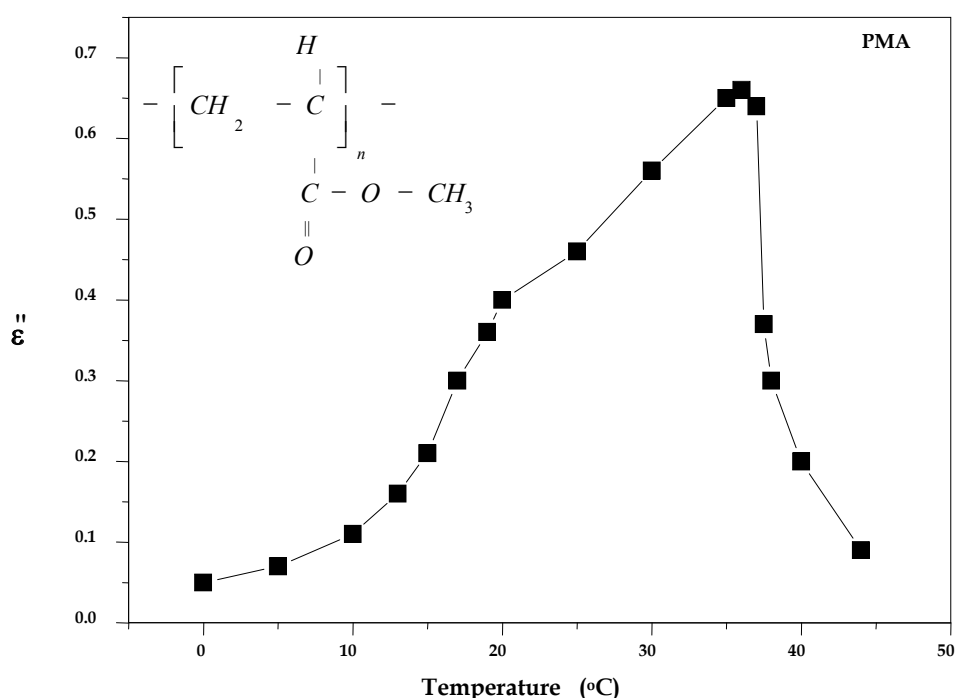


Fig. 1. Temperature dependence of ϵ'' for PMA thin film.

Figure 2 shows a broad (β, α) peak in the PMMA ϵ''/T curve, which is observed in the temperature range of $(30 - 100)^\circ C$. Since PMMA is a non-crystalline polymer, this peak is assigned to an α -relaxation (glass transition) as reported earlier (Bistac & Schultz, 1997) (Bistac & Schultz, 1997) (Krause et al., 1965) (Utte et al., 1995). When this curve is compared with the ϵ''/T (PMA) curve, they are found to be very close with a shift of $28^\circ C$ towards high temperature in the PMMA data. This means that the $-COOCH_3$ group has a sufficient kinetic capability at room temperature to show a dielectric relaxation and the degree of freedom approaches its maximum value at $65^\circ C$. The temperature shift is due to the dipole length differences between both types. Stereochemistry indicates that the PMA structure

involves many big loop spaces when compared with *PMMA*. The small peak in Figure 2 at 120°C is thought to originate from the space charge ρ peak (Krause et al., 1965).

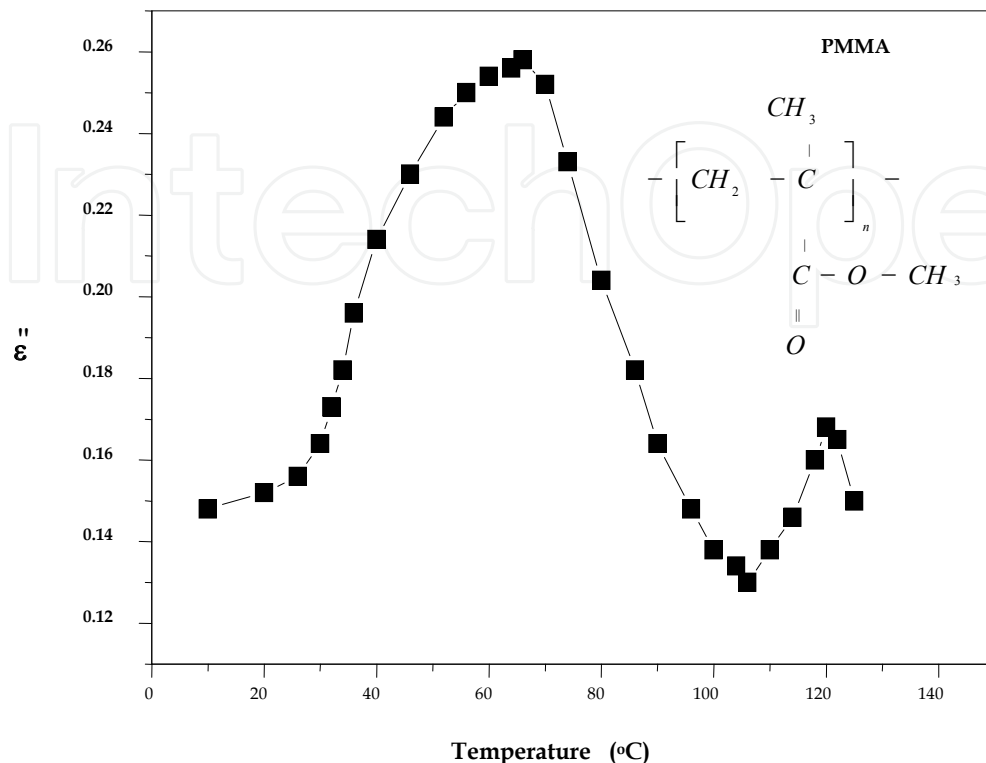


Fig. 2. Temperature dependence of ϵ'' for PMMA thin film.

Figure 3 shows the ϵ''/T -curve of PMαClA, in which the structural differences are also in the *r* group, which is chlorine (*Cl*), associated with high polarity. The α -relaxation peak shape at 140°C depends on the substitution and vibration of the chlorine loop associated with the crankshaft motion. The high polarity of the chlorine atom increases the hardness of this polymer and pushes the relaxation processes towards higher temperature range. β -relaxation occurs at temperatures around 80°C and is due to the weak rotation of the $-\text{COOCH}_3$ group and the field loops overlapping which produces strong links between the neighbored chains. The comparison of β -peaks in Figure 1, 2 and 3 indicates that there is an inverse proportional relationship between the polarity value and the strength of the β -peak.

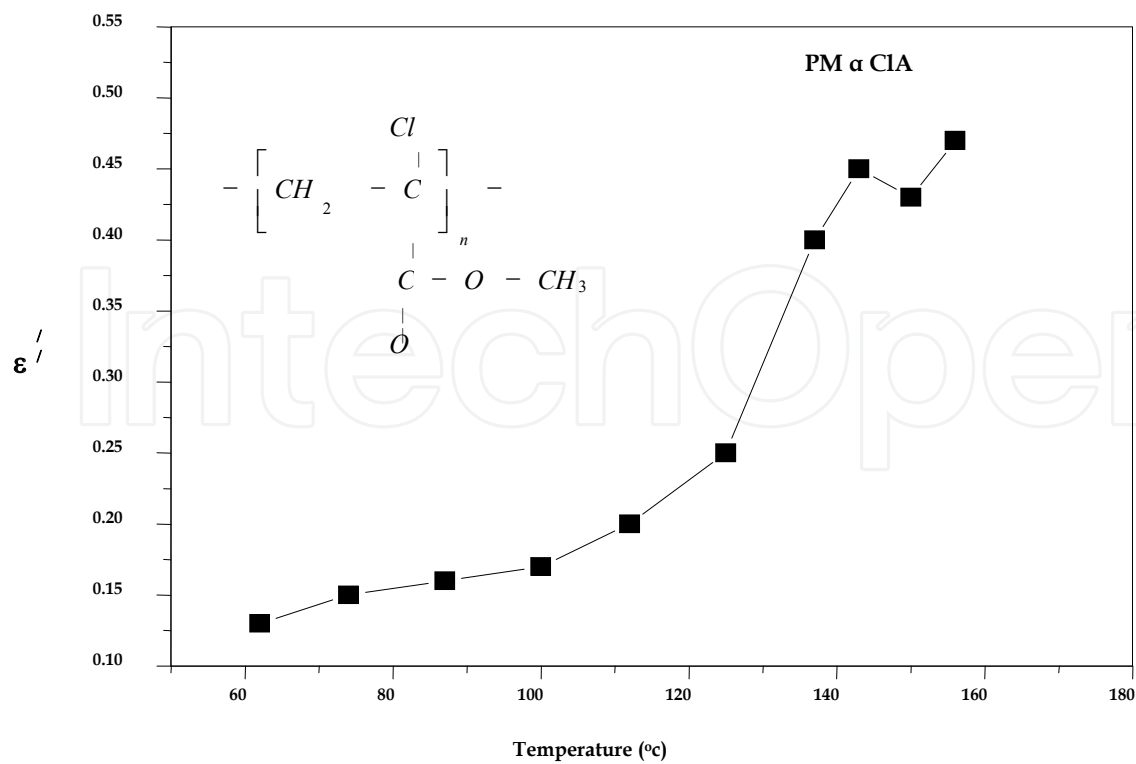


Fig. 3. Temperature dependence of ϵ' for PM α CIA thin film.

The PVAc polarity is high as shown in Figure 4, and is increased by two fold when the temperature is doubled. The oxygen atoms substitute the carbon atoms in the acrylate group, leading to a change in the dipole length and polarity value due to the strong intermolecular interaction, which gives a sharp α -relaxation peak at 75°C.

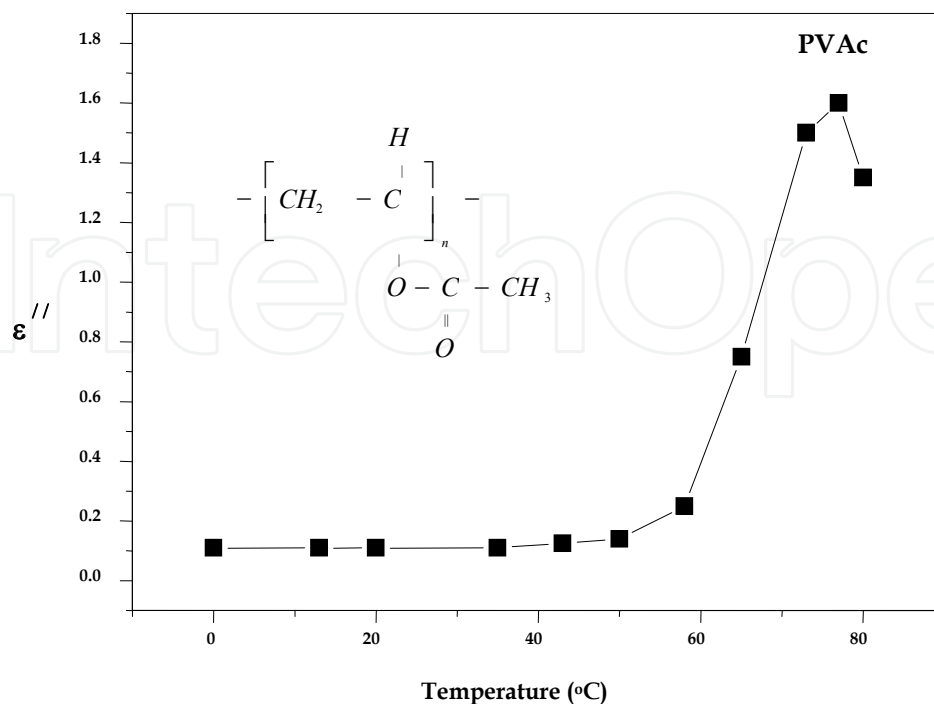


Fig. 4. Temperature dependence of ϵ'' for PVAc thin film.

6.2 Simulation of the U Energy Bands

The simulation of the dipoles' stable energy levels U is illustrated in Figure 5. This simulation method can show the energy bands and the stable energy levels available for specific dipoles. The specific dipole group depends on the temperature range, the polymer structure and the solid state of the main polymer (Blythe & Bloor, 1980). Applying the polynomial fitting method to the data produces a smooth curve, which shows the shape of the dipole energy bands.

Figure 5 shows the U/T (PMA)-curve for the data produced from equation 9. Using the polynomial fitting approximation for $i = 4$ gives a compatible shape for U/T with ε''/T -

curve of Figure 1. The peak in region II flips from negative to positive signed to the dipole vector directed with or opposite to the electrical field. The simulation curve shows the density of states of the energy bands available for the main dipoles.

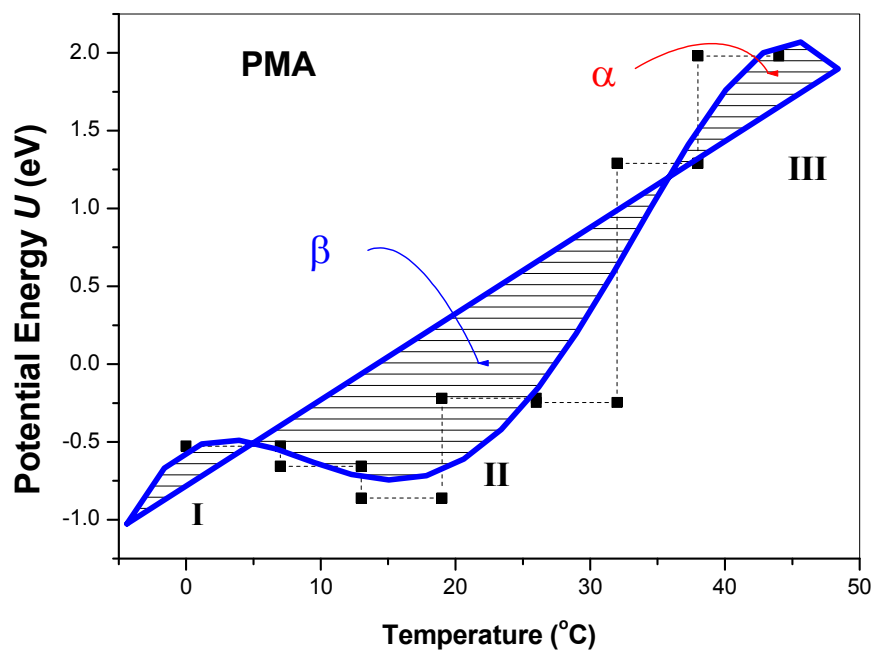


Fig. 5. The potential barrier U versus temperature, the dot line indicates the experimental data and the solid line indicates the simulation model for the PMA thin film.

The γ -relaxation does not appear in all figures because, the r groups have, in most cases, one carbon atom. γ -relaxation is observed clearly when the side chain r is increased in length beyond 3 or 4 carbon atoms (Blythe & Bloor, 1980) and it depends also on the resolution of the measurement method. Taking this into account, the band marked I in Figure 5 (PMA), is the continuation of the second relaxation (β -relaxation). The density of states observed in region (I) is due to the temperature relaxation arising from the relaxation of the hundreds of $-COOCH_3$ groups around themselves. Region II is present in the temperature range (5 – 35)°C and related to β -relaxation indicating that there is a high density of states available for the main dipole produced from the rotation of group of $-COOCH_3$ around the $C - C$ backbones.

The positive and negative values of U are related to the orientation of the dipole groups to the direction of the applied field. Region III appears at 36°C indicating α - relaxation associated with the chain backbone movement (crankshaft motion).

Figure 6 shows four different density of states for PMMA. Comparing the first three regions I, II, III with those in Figure 1b₂ shows clearly the separation between the relaxation processes. Region V shows the high density of states available to the space charges.

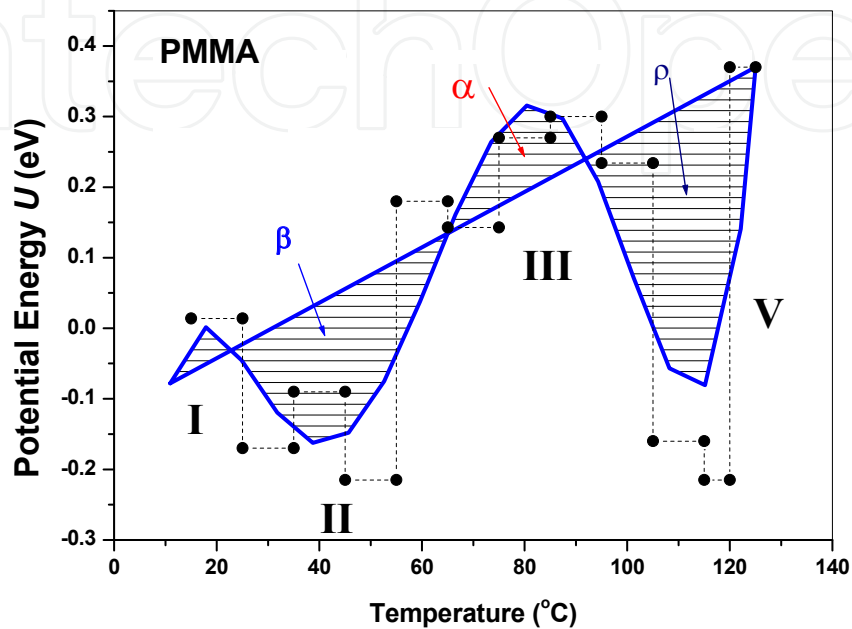


Fig. 6. The potential barrier U versus temperature, the dot line indicates the experimental data and the solid line indicates the simulation model for the PMMA thin film.

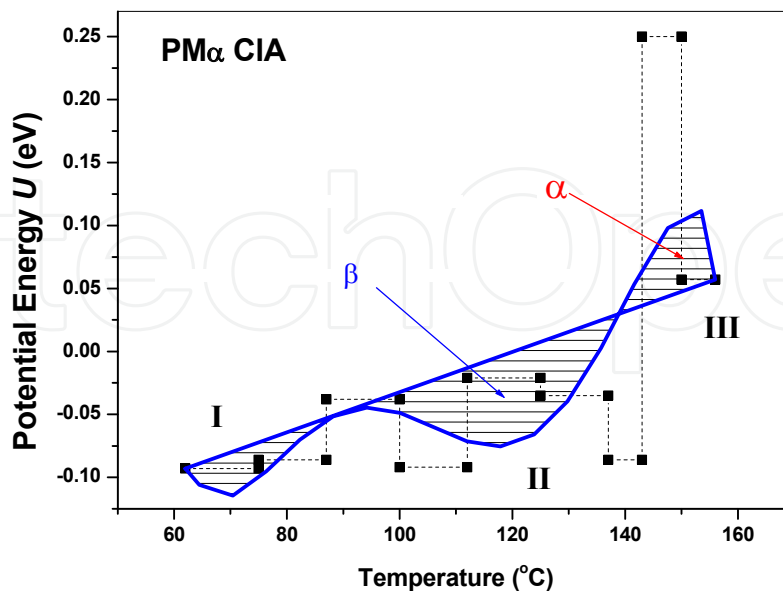


Fig. 7. The potential barrier U versus temperature, the dot line indicates the experimental data and the solid line indicates the simulation model for the PM α CIA thin film.

The band structure of *PMαClA* also shows three regions as for *PMA*. The variation in the width and the depth of the bands is shown in Figure 7 and is due to the chlorine in the *r* group which replaces the $CH_3 - C$ or $H - C$ dipoles by a $Cl - C$ dipole.

The high chain flexibility in the *PVAc* dominates the relaxation response very early i.e. at low temperatures. There is a wide range of density of states for α dipole groups and this is the reason for disappearance of the other relaxation mechanisms which are seen quite clearly in *PVAc* (Figure 8).

Figure 2, shows that the α -peak is around 65 – 70 °C and ρ is around 120 °C. However, in Figure 6, the α - peak is shifted forward to (80 – 85) °C and ρ peak is moved from 120 °C to around 110 °C, using the simulation model.

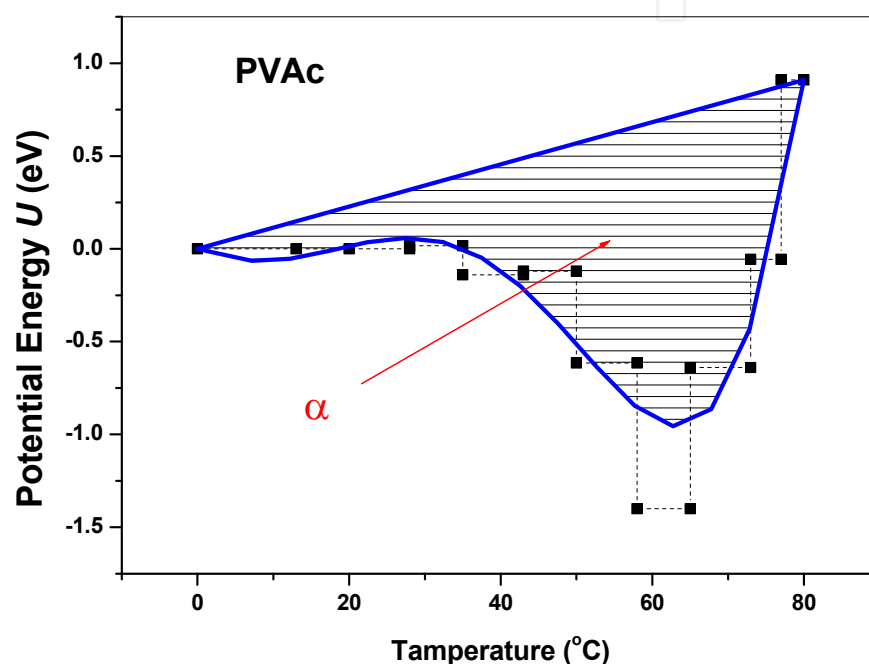


Fig. 8. The potential barrier U versus temperature, the dot line indicates the experimental data and the solid line indicates the simulation model for PVAc thin film.

This shows that *PMMA* is most likely to be isotactic, and this is considered to be one of the advantages of our simulation model to determine the correct position of the dielectric relaxation peaks.

The low values of the potential energy U (activation energy), which were determined by this method, are due to the thick layer, the high molecular weight of the polymer, and the lower degree of freedom as well as the limited resolution of the measurement method (Kalogeras, 2003; Kalogares, 2005). For *PMMA*, the maximum potential energy value at 120 °C is found to be 0.17 eV and for $(\beta + \alpha)$ activation energy peak is found around 0.4 eV. Kalogeras *et al.*, found that the activation energy for *PMMA* at 120 °C using DRS method was about (≈ 0.1 eV) and for β - relaxation it was (0.4 – 0.9) eV while for α - relaxation it was (≥ 0.9 eV) (Kalogares *et al.*, 2005). For ultrathin films of *PMMA*, Wübbenhorst *et al.*, have found that α activation energy peak varied from 2 - 4 eV when the film thickness changed from 6.9 - 58.5 nm (Wübbenhorst *et al.*, 2003). This means that there is a direct proportion between the film

thickness and the activation energy. However, this proportion will be changed to inverse proportion when the film thickness is over the maximum limit of thickness-activation energy spectroscopy. In this case, α - relaxation is affected more than β and γ relaxations, and therefore explains the low value of $(\beta + \alpha)$ - activation energy peak in Fig.(2), which is most likely influenced by β relaxation more than α relaxation. Fig.(6), shows that β peak has a double size than the α peak. This confirms that α relaxation is less pronounced in PMMA dielectric spectroscopy due to the film thickness ($61 \mu m$).

Table 1, demonstrates the polynomial fitting coefficients and the polynomial error (P) for each polymers. The combination between these coefficients can be used to determine the dielectric relaxation before and after any kind of treatment to locate the variation in the relaxation model of the polymer under investigation. The coefficients values give a good indication and recognition for any possible structural changes.

Toluene is the basic solvent used, and interacts only weakly with the selected polymers. Its plasticizing effect is lower, due to the fact that part of the solvent molecules does not interact with the polymer chains, and is probably retained in the form of clusters (Spěvák & Schneider, 1987; Bosscher et al., 1982).

Finally using the simulation polynomial fitting technique, the U band values can be observed specifically and separately. Moreover the dielectric relaxations can be clearly detected and the temperature ranges for the relaxation processes can be obtained.

CONSTANT	PMA	PMMA	PM α ClA	PVAc
a ₀	-0.55749	-0.83145	21.1653	0.01961
a ₁	0.05066	0.12363	-1.10272	-0.02187
a ₂	-0.01037	-0.00632	0.0222	0.00152
a ₃	4.99027E-4	1.32219E-4	-2.17063E-4	-1.0469E-5
a ₄	-5.88414E-6	-1.17606E-6	1.0316E-6	-7.6449E-7
a ₅	—	3.71396E-9	-1.90824E-9	9.08388E-9
P	-9	-15	-10	-8

Table 1.

The energy bands encountered using the technique indicated all the density of states available for (α, β) dipoles. Comparing the different types of polymers, the effect of changing or replacing the side group on the energy band shapes is clearly demonstrated (Hashim et al., 2006).

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This book provides a timely overview of a current state of knowledge of the use of polymer thin film for important technological applications. Polymer thin film book covers the scientific principles and technologies that are necessary to implement the use of polymer electronic device. A wide-ranging and definitive coverage of this emerging field is provided for both academic and practicing scientists. The book is intended to enable readers with a specific background, e.g. polymer nanotechnology, to become acquainted with other specialist aspects of this multidisciplinary field. Part A of the book covers the fundamental of the key aspect related to the development and improvement of polymer thin film technology and part B covers more advanced aspects of the technology are dealt with nano-polymer layer which provide an up-to-date survey of current research directions in the area of polymer thin film and its application skills.

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University Campus STeP Ri
Slavka Krautzeka 83/A
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Phone: +385 (51) 770 447
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InTech China

Unit 405, Office Block, Hotel Equatorial Shanghai
No.65, Yan An Road (West), Shanghai, 200040, China
中国上海市延安西路65号上海国际贵都大饭店办公楼405单元
Phone: +86-21-62489820
Fax: +86-21-62489821

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