Gamma radiation-induced changes in the conduction mechanisms in prepared PVA/Lithium chloride

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Received: 20th October, 2010 ; Accepted: 30th October, 2010

ABSTRACT

The electrical conductivity, current-voltage (I-V) and current–time (I – T) characteristics of pure PVA and different Wt % of LiCl -doped polyvinyl alcohol (PVA) films have been studied under different conditions. The electrical conductivity increased with increasing dopant concentration up to 60 % of weight and then showed a decrease beyond this concentration. The increase in conductivity is attributed to formation of charge transfer complexes while the decrease beyond 60 wt. % may be due to segregation. The variation of electrical conductivity with temperature showed two regions of activation energies for undoped films and three regions for doped films. I-V characteristics indicated that Schottky emission is the dominant charge transport mechanism in both undoped and LiCl-doped films. The effect of gamma irradiation at different radiation doses (20 and 50 kGy) on pure polyvinyl alcohol films and PVA loaded with different Wt % of LiCl was investigated. Measurements of *I-V* characteristics have been carried out at temperature range of 30-140°C. Therefore, the charge carrier's concentration in each of the conduction bands, the trapping parameter θ, the trap density N_t is determined as a function of different Wt % of LiCl is determined, leading to the electron mobility μ_0, effective electron drift mobility μ_e, as well as electrical transport properties and trapping energy calculation. The change that occurred in the dielectric constant values at different temperatures after irradiation can be attributed mainly to the changes in the intra- and intermolecular interactions.   © 2011 Trade Science Inc. - INDIA

INTRODUCTION

For many years, the electrical conductivity process has been the subject of extensive investigations in disordered solids. For polymeric materials, the electrical transport plays an important role in different device applications such as high performance capacitor, electrical cable insulation, electronic packing and components. From the fundamental point of view, dielectric properties have been widely used to realize the dynamical process in complicated system[1]. It has been successfully used in obtaining general information about the localized defect states in the forbidden gap. The charge injected into the insulator in response to an applied volt-

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age using a constant voltage source is trapped at these localized defects. The presence of these trapping sites in the forbidden gap strongly affects the current–voltage (J–V) characteristic of the material. The form of J–V characteristics depends on the type of distribution of traps. The magnitude of the current flow is much reduced due to trapping effect[2]. The appearance of SCLC regime is inhibited until a sufficiently large field is applied where the transition from Ohmic regime to an SCLC regime depends markedly on the distribution of the trapping level. Recently, several authors[3] have studied the J–V characteristics in polymer diodes where the SCLC (space charge limited current) theory could be successfully applied. They found that the barrier height for the formed metallic polypyrrole films Si Schottky diode (0.84 eV) is larger than that for the conventional Schottky diodes.

The commercial and scientific interest in studying polymer systems with desirable applications has led to significant information in the field of electrical insulators[4–8]. Frequently, electrical mechanisms (SCLC) have some electrical insulators, which are manufactured from polymer materials. For safety-related equipments, detailed qualifications to prevent accidents during service are given[9]. Also, Polymeric ion conductors with mechanical flexibility are key materials for developing all solid electrochemical devices such as rechargeable batteries, capacitors and sensors[10-12]. Polyvinyl alcohol, which is a semi crystalline polymer[13,14], plays an important role in the applications due to its OH group and hydrogen bonds[15]. Hydrogen – bonded polymer such as in PVA polymer used in gas sorption, diffusion[16,17] and can selectively adsorb metal ions[18]. In this respect, authors have been engaged in the study on life models for insulating materials subjected to electrical and thermal stresses[19], the influence of dose rate, irradiation surroundings[20], period of exposure[21], and type of materials[22]. Polyvinyl alcohol (PVA) was used to study the effect of LiCl on the variation of jet diameter with axial coordinate after the onset of whipping instability during electro spinning and it was found that the addition of LiCl significantly accelerated jet thinning and solidification[23]. This produces intermolecular cross-linking induced by irradiation blends with super molecular structure and improved properties. The treatment of such compositions with energy radiation leads to additional and unpredictable changes in their structure and properties. Irradiation with γ ray has a significant effect on polymer properties and some physical properties are usually modified. The metal ion doped polymers represent a new class of organic materials. Inorganic additives like transition metal salts have a considerable effect on many of properties; such as the electrical properties of the polymers. The present work focused mainly on the electrical properties of PVA filled with transition metal chloride (LiCl) at different filling ratios and its conduction mechanism. The object of this paper is to shed more light on the conduction phenomena for irradiated pure PVA and different Wt % of LiCl -doped polyvinyl alcohol (PVA) film. The study aimed to measure the electrical current density as a function of an applied voltage at different temperatures. A number of important parameters, namely free carrier density, trap density, effective density of state, Fermi level, etc. were determined. In addition, the dependence of the potential barrier and the localized trap levels on the irradiation dose will be discussed.

EXPERIMENTAL

Materials and samples preparation

The samples were prepared by casting method using PVA of μW 17000 g in the grain form (provided by Avondale laboratories Beaumount close Banbury Oxon, England) and LiCl was obtained from adding dilute HCl to $LiCo_3$, then the solution formed was dried in electric oven at 40°C for four days until the water were completely evaporated . Then, a proper mass fraction of the PVA and LiCl separately dissolved in distilled water, added together under a good stirring at 40°C for 4 hours to ensure that the solution is homogeneous[2]. After casting the mixture in a petry dish, it was exposed to ^{60}Co irradiator at a constant dose rate for two doses, 20 kGy and 50 kGy at room temperature. Then the dishes were placed in the electric oven at 40°C until water was evaporated. Finally, films were prepared using blow co-extrusion techniques with blow ratio 7. The thickness of the blow co-extruded film amount is about of thickness 0.5 mm and diameter 5 mm. These films were sandwiching by silver paint (electrodes) having the following LiCl mass fraction 0, 20, 40, 60, 80 wt % and doses 20kGy and 50kGy.
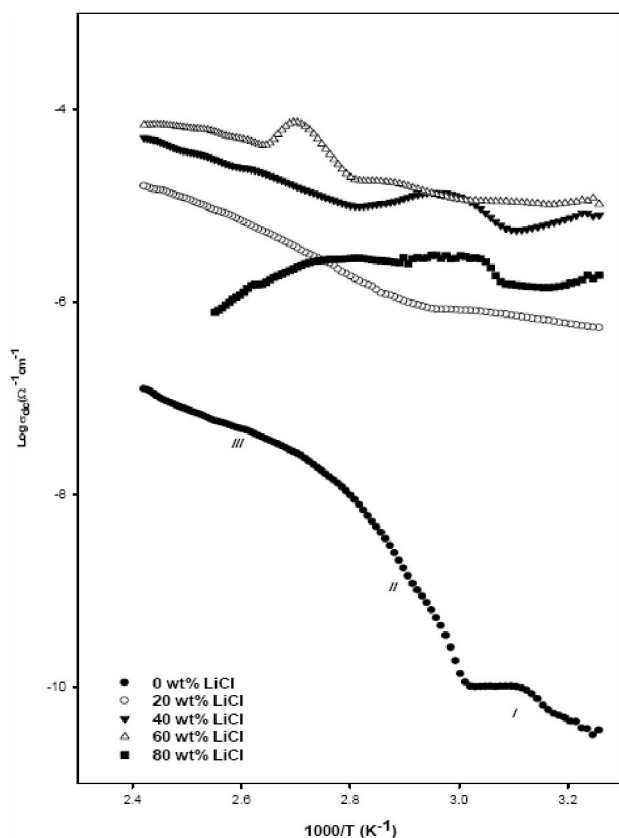
Figure 1 : Variation of the electrical conductivity for pure PVA and PVA loaded different Wt % of LiCl with reciprocal of temperature at 1v and at 20 kGy γ radiation
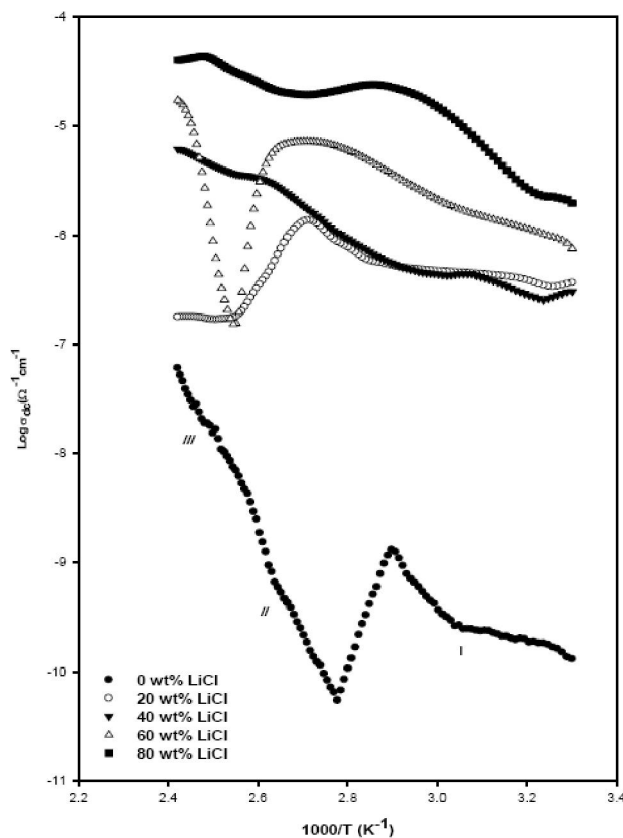
Figure 2 : Variation of the electrical conductivity for pure PVA and PVA loaded different Wt % of LiCl with reciprocal of temperature at 1v and at 50 kGy γ radiation

Measurements

The *I-V* characteristics measurements of pure PVA and different Wt % of LiCl -doped polyvinyl alcohol (PVA) films has been preformed at temperature rang 30-140C using a High Resistance Meter Electrometer (Keithly, type 6514). This electrometer has an in-built capability of output in depend voltage source of V. The same equipment (power supply) was used to apply the voltage across the sample and to measure the current through the film. To ensure the proper connection to the sample, an indigenously designed sample holder with silver electrode has been used. In this arrangement the film is sandwiched between two circular electrodes from silver. For electrical measurements, the samples were sandwiched between two copper electrodes coated with gold to perform good contact, avoiding air gaps between the electrodes and the samples. A specially designed cell was used for mounting the samples into an oven. The temperature of the samples was monitored using a K-type thermocouple attached with a digital thermometer type Tri-Sense, Cole-Parmer, USA,

with temperature resolution of 0.1 1C.

Dielectric measurements were performed by impedance spectroscopy using a Hioki LCR meter, 3531 Z Hi-Tester, Japan, operating at a frequency range (42 Hz–5 MHz); with impedance accuracy ranges from 0.15% up to 4%. The bridge was connected via a standard interface (RS-232C interface, Hioki, Japan) to a Pentium personal computer for instrument control and data processing. Silver electrodes 10 mm in diameter were used on both sides of the samples. The capacitance (C) and the loss tangent (tan δ) were obtained directly from the bridge from which the permittivity (ε') and dielectric loss (ε'') were calculated. For the denoted samples ε' and ε'' were measured over a frequency range from 1kHz to 5 MHz at room temperature.

RESULTS AND DISCUSSION

DC electrical conductivity

Figure 1 shows the relation between the electrical conductivity σ_{dc} ($\Omega^{-1}m^{-1}$) and the γ-irradiation dose at
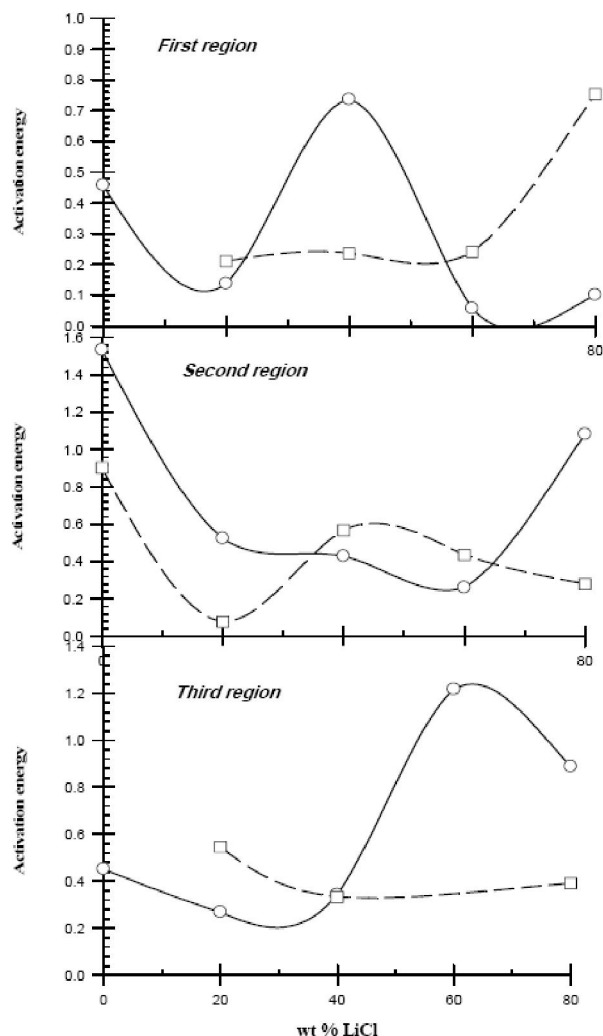
Figure 3 : The variation of the activation energy of different wt% of Lithium chloride at 20 and 50kGy γ - dose

Figure 4 : The temperature dependence of e′ for pure PVA and its LiCl loaded contents at 20kGy γ -irradiated

Figure 5 : The temperature dependence of e′ for pure PVA and its LiCl loaded contents at 50 kGy γ –irradiated

different temperature in the range 30-140°C for doped PVA samples at different concentrations of LiCl (20, 40, 60, and 80 wt %) at 20 kGy. It was clear from the interaction of this figure that there is a direct proportion between the dc conductivity with temperature and metal ions concentration. However, the sample loaded with 80-wtpercentage of LiCl showed a decrease in σ when compared with other loaded samples at 356 K. For the induced changes in σ at 20 kGy to occur it depended on metal salts and γ-ray and can attributed to the creation of induced charge carriers in the PVA matrix. Figure 2 plotting the variation in log σ with 1/T for the 20, 40, 60, and 80 wt % LiCl-doped PVA samples at 50 kGy, showing that the induced changes in σ increase with wt % and temperature. However, the electrical conductivity for pure PVA samples is less than that for the counterparts in 20, 40, 60, and 80 wt %
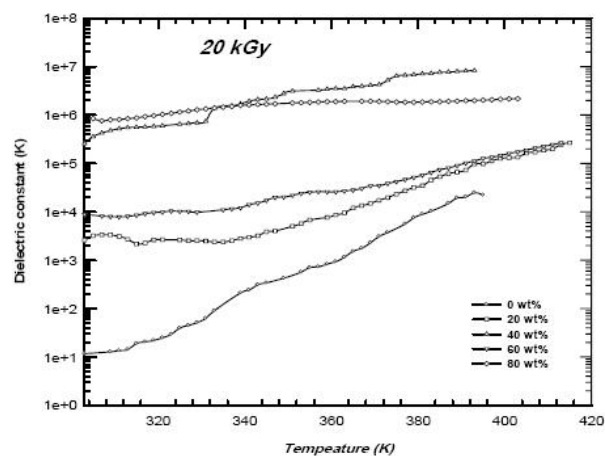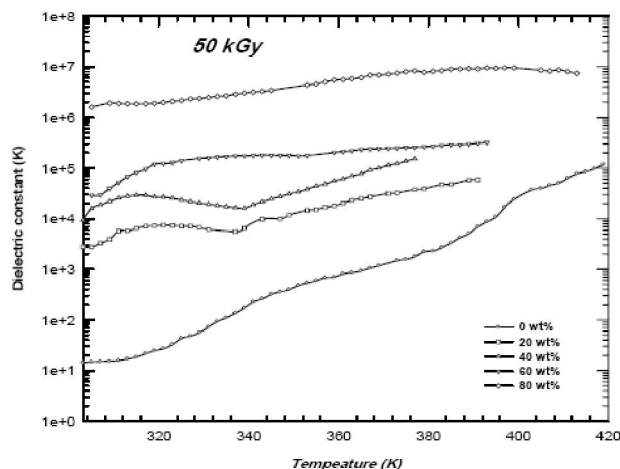
LiCl-doped PVA in region I, II and III. Lithium ions are coordinated through ionic bonds with the hydroxyl group belonging to the different chains in PVA[25]. The dc conductivity σ_{dc} (T) values of the films in both areas exhibit the validity of applying Arrhenius equation.

$$\sigma_{dc}(T) = \sigma_0 \exp\left(\frac{\Delta E}{KT}\right) \qquad (1)$$

where σ_0 is the temperature-independent constant, K is the Boltzmann constant and ΔE is the activation energy.

In other words, the addition of lithium increases the volume required for ionic carriers to drift in the polymer. This leads to an increase in the ionic mobility and a reduction in the activation energy. This is in complete accordance with the results observed for 80 wt %. It has been thought that the non-uniform doping cause's

TABLE 1 : Values of the Activation Energy Calculated from DC Conductivity measurements

Samples	Activation Energy (eV)					
	At 20kGy			At 50kGy		
	I	II	III	I	II	III
PVA	0.458035	1.533807	0.451555	3.503351	0.903772	
PVA+20 wt%LiCl	0.138362	0.523872	0.268377	0.211044	0.077846	0.546399
PVA+40 wt%LiCl	0.737048	0.428774	0.342751	0.236907	0.567733	0.332775
PVA+60 wt% LiCl	0.057899	0.260908	1.217277	0.240833	0.435308	3.793409
PVA+80 wt% LiCl	0.101805	1.083193	0.888374	0.753537	0.281117	0.391613

TABLE 2 : Values of μ ($cm^2V^{-1}s^{-1}$) and n (cm^{-3}) for pure, 20, 40, 60,80wt% LiCl doped PVA samples at 20kGy and 50kGy γ–dose Irradiation

Samples	20kGy		50kGy	
	μ	n	μ	n
PVA	3.72E-07	6.06E+15	4.73693E-08	8.88E+14
PVA + 20 wt % LiCl	1.22914E-05	7.32E+16	1.24091E-05	9.69E+16
PVA + 40 wt % LiCl	7.55682E-06	8.10E+16	4.58182E-05	4.30E+17
PVA + 60 wt % LiCl	1.37534E-05	1.82E+17	0.015757955	2.61E+20
PVA + 80 wt % LiCl	6.32466E-07	1.19E+16	0.0002975	1.67E+19

heterogeneity, which Leads to the formation of non-stoichiometric charge–transfer complexes between the polymer and dopant, this is accounted for high content of Li[26]. The decrease in the activation energy upon doping with lithium chloride may be attributed to the increase in crystallinity of the polymer due to the alignment of the entangled chains in the amorphous region because of electrostatic interaction between the negatively charged hydroxyl groups and positive lithium ions. Figure 3 & 4 shows the variation in activation energy ΔE values is increased especially at regions I, II and III, where these can be attributed to the formation of molecular aggregates of lithium with the dopant concentration, these activation energy values can be obtained from the slopes of the curves for all doses. TABLE 1 illustrates the values of the activation energies below (ΔE_1) and above (ΔE_2) glass transition for the samples, measured as a function of the dopants wt% and the absorbed dose.

The J–V characteristics showed three regions with different slopes depending on the applied voltage. At low-voltage region, the conduction is ohmic ($J\alpha V$) due to the intrinsic conductivity of the material, indicating that the thermally activated carriers controlled the current. Such behavior suggests an ohmic regime below

the threshold voltage and an SCLC regime for higher voltage levels. It can be considered that the general equation for current flow is;

$$J=qn_0\mu_0E \tag{2}$$

where; J is the current density, q is the electronic charge, μ_o is the electron mobility of the free carrier in the conduction band and E is the electric field strength and equals to V/d; (where V is the applied voltage and d is the sample thickness) and n_o is the concentration of the free thermally activated carriers in the conduction band.

The J-V characteristic curves at different temperature for the un-irradiated and irradiated films are plotted in figure 1. It was observed that the current increased by increasing the applied voltage for all samples at each dose. The current also increases gradually with increasing gamma irradiation dose 20 - 50 kGy.

In addition, γ-irradiation seems capable to make some sort of variation in the amorphous regions reflecting a change in the structure of PVA–LiCl system. On the other hand, the increase in activation energy at a higher concentration for 40-wtp percentage at 20 kGy and for 80 wt% at 50 kGy can be explained as being due to the formation of molecular aggregates of lithium chloride. The charge carriers become more localized, which gives the increase in the trapped carrier density. These molecular aggregates may be formed due to the inhomogeneous distribution of the dopant as well as the nonbonding part of the dopant[27].

In the present study, the band model is used to justify the mechanism of charge transport through the polymeric material assuming the localized energy states to be uniformly distributed in energy within a range of the energy gap. However, the localized states which are due to the lack of order may be considered trapping levels similar to the localized states arising from impurities, dislocation, et. Thus, it can be concluded that the current density in the second and the third regions obeys the equation commonly used.

$$J = \left(\frac{9}{8}\right)\left[\theta\mu_o\varepsilon\varepsilon_0\left(\frac{V_{tr}^2}{d^3}\right)\right] \tag{3}$$

where ε is the dielectric constant of the materials, ε_0 permittivity, d is the films' thickness, μ_o is the electron mobility in the conduction band, V_{tr}^2 is the ohmic to square region transition voltage and θ is the trapping

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Figure 6 : Current relaxation curves for different wt % LiCl - doped PVA samples 0.01 - 0.1 cm thick at 20 kGy γ - doses at 10 volt and 50°C where (a) is in charge and (b) is discharge or reverse polarity

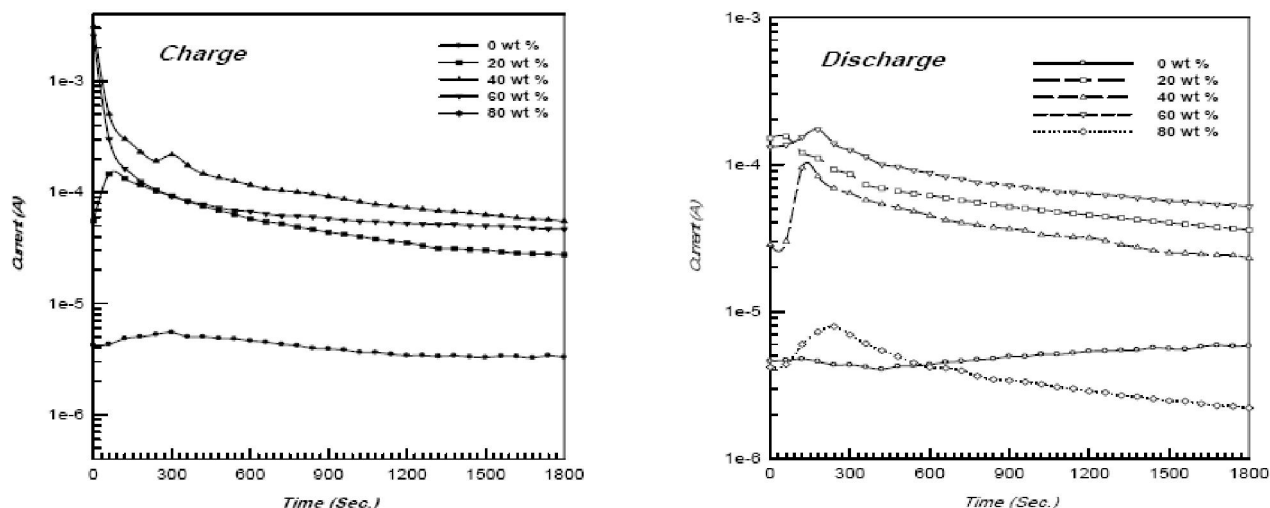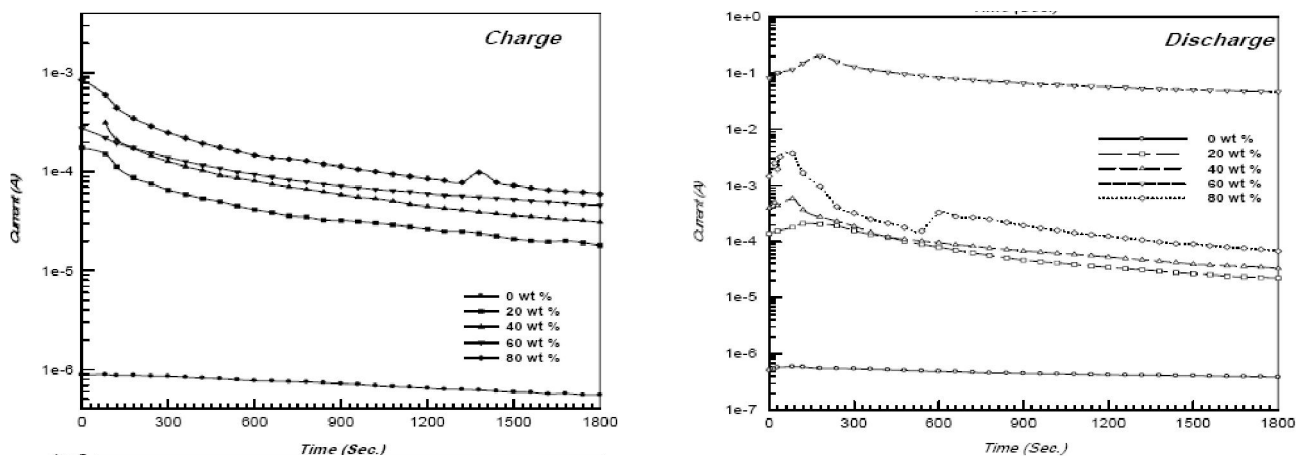**Figure 7 : Current relaxation curves for different wt% LiCl- doped PVA samples 0.01 -0.1 cm thick at 50 kGy γ - doses at 10 volt and 50°C where (a) is in charge and (b) is Discharge or reverse polarity**

parameter.

The latter can be considered as the ratio between the free electron numbers (n_o) on the conduction band and the total density (n_0+n_t) where n_t is the density of the trapped electrons. The concentration of traps is defined as:

$$n_t = \left(\frac{2\varepsilon\varepsilon_0}{qd^2}\right) \times V_{TFL} \qquad (4)$$

where V_{TFL} is the voltage at which the current significantly increases with temperature (third region).

On the other hand, when a trap level exists, the electron mobility is reduced by $1/\theta$, as trapped electrons do not follow an electrical field. The free carrier mobility μ_0 can now be estimated from eq. (5). It may be mentioned that when a trap level exists, the electron

mobility (μ_0) is reduced by $1/\theta$; and the effective electron drift mobility (μ_e) in an insulator with traps is therefore:

$$\mu_e = \mu_o\theta \qquad (5)$$

The free carrier density in the conduction band (n_t) was calculated by eq. (4). The free carrier mobility (μ_0) could be obtained for both the un-irradiated and irradiated films by eq. (5) and is given in TABLE 2.

The data of *J-V* characteristic curves at different temperature for the un-irradiated and irradiated films show that the carrier concentration is changed due to two main factors, the first is the presence of the Li , which it is responsible for ions to be increased, the second is the ionization due to γ - irradiation of the polymer, which it is rupture the chemical bonds and create

energetic free electrons, ions, and radicals, which are able to migrate through the network, leading to a change in electrical conductivity . The value of drift mobility is very low and decreases with the increase of subsequent addition of lithium chloride to PVA, indicating predominant ionic conduction.[32] besides, it can be deduced that conduction enhancement is apparently due to the gradual increase of bulk-generated free carriers, density n_o. Moreover, μ_o mobility smaller than 1 cm^2 V^{-1} s^{-1}, which is the case in TABLE 2, indicate that polaron hopping between localized sites is involved in the process of carrier transport.

Dielectric constant

The dielectric constant was calculated from the capacitance c using the following formula:

$$\varepsilon' = {Cd}\big/{\varepsilon_0 A} \qquad (6)$$

where d is the thickness of the film, A is the cross-sectional area of the film surface, and ε_o is the free-space permittivity.

The dielectric properties of pure and LiCl-doped PVA samples, at 20 kGy and 50kGy γ-dose irradiation, as function of temperature were studied. The values of $\in' (\omega)$ are very high at low frequency and high temperature but at high frequency, there are relatively constant values with frequency. Figure (5) shows the variation of dielectric constant ε' with temperature for pure PVA samples and its different wt% of LiCl at 20kGy γ- Irradiated dose. A broad glass transition temperature T_g peak appeared at about 358 - 360 K for pure and doped samples, where it is clearer as dopant increase than pure PVA. Thus, a relaxation is observed, at low temperatures, as a gradual decrease in $\in'$ and a broad peak in $\in''$ at low frequency have been monitored. The polymer chain possesses a variety of internal motions and branch segments of the main chain diffuse from one configuration to another with changing dipole moments. To each such internal diffusion, there is a characteristic relaxation time.

In figure 6 at 50 kGy γ-irradiation dose, we did not remark this peak for the glass transition, where it disappeared, and may be attributed to structural changes in the polymer matrix, probably molecular and crystallographic rearrangement. In fact, gamma irradiation can produce different valence states of the transition metal

ions in the matrix of the chelated compounds. And may be for that reason, we find in figure 6 a new peak, at low range of temperature for the samples at 50kGy, which begins to be more clear and pronounced and takes more wide range on temperature scale with increasing the dopant content up to 60 wt% and at 80wt% start to decreases. At the same time, irradiation can affect the electronic state of the ligand molecules in the polymer matrix[21,22] and, also we can remark that with increasing of γ-irradiation, the magnitude of the dielectric constant increases. Also, the ε' is strongly dependent on temperature and the dopant wt%. This dependence may be due to the creation of carbonyl groups, which are highly polar, cause, a remarkable increase in dielectric constant[28,29]. Besides cross linking and degradation, evolution of hydrogen may take place due to the irradiation of polymer material. The induced radiation effect on pure and doped polymer may decrease the disorder of the dipolar groups so the permittivity and the dielectric parameter increase. The subsequent addition of transition metal halides to PVA matrix entails a considerable increase of ε' at frequency of 1 kHz within the temperatures range used. At relatively higher concentrations of dopants, the obtained ε' (T) results, indicate the important role of metal halide additions, which cause structural variations in the polymeric network.

Current–time characteristics

The current–time curves for pure PVA and LiCl-doped samples at 20kGy and 50kGy γ-irradiation at 10 V and temperature 50°C were measured. The order of magnitude of the drift mobility (μ) of charge carriers can be determined from the approximate relation, as follows:

$$\tau \cong t_{max} = {d^2}\big/{\mu V} \qquad (7)$$

where d is the thickness of the sample, V is the applied voltage, and t_{max} is the relaxation time corresponding to the peak current in I–t curve.

Similar plots were obtained, and, therefore, the current–time curves for 20kGy samples are taken as a representative. The observed relaxation phenomena can be attributed to two mechanisms. The first is the clean-up effect[30] that is concerned with the effective decrease in the density of the mobile ions within the specimen

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due to their arrival at the electrode. The second mechanism is the effective drop of the electric field in the bulk of the specimen due to the space–charge effect of ions drifting to the electrode. The peaks observed in the reversed polarity curves. Figure 6 & 7 can be explained as follows. A fraction of the ions that arrive at an electrode accumulate there as immobilized ions without discharging at the electrode. These ions can become mobile again after the reversal of the polarity.

CONCLUSIONS

It is worthy to mention that the irradiation of polymers and the addition of dopant give rise to Changes in electrical characteristics such as conductivity and dielectric constant. Irradiation of Polymers generally increases their electrical conductivity and decrease the activation energy.

The decrease in the activation energy upon doping with lithium chloride may be attributed to the increase in crystallinity of the polymer due to the alignment of the entangled chains in the amorphous region because of electrostatic interaction between the negatively charged hydroxyl groups and positive lithium ions.

The increase of drift mobility with γ-dose indicates that the conduction is apparently due to thermally activated mobility. This seems to be consistent with electrical conductivity data.

The increase of the dielectric constant with temperature indicates that the dependence is governed mainly by the change in the intra- and intermolecular interactions. The subsequent addition of transition metal halides to PVA matrix entails a considerable increase of ε' at frequency of 1 kHz within the temperatures range used. This may be related to the effect of local environment on the inherent ability of the dipoles to orient. At relatively higher concentrations of dopants, the obtained ε' (T) results indicate the important role of metal halide additions, which cause structural variations in the polymeric network.

The significance of finding these electrical parameters for the polymeric materials arises from its important role in some device applications such as high-performance capacitors, electrical insulator and electronic packing. Special emphasis is put on the polymer-irradiation interaction process, which not only increases electronic mobility but also reduces its potential-barrier giving rise to desired properties of the irradiated polymer.

REFERENCES

[1] P.Dutta, S.Biswas, Subodth Kumar De; Material Research Bulletin, **37**, 193-200 **(2002)**.
[2] J.Chutia, K.Barua; J.Phys.D, **13**, L9-L13 **(1980)**.
[3] P.W.M.Blom, H.C.F.Martens, H.E.M.Schoo, H.C.J.M.Vissenberg, J.N.Huiberts; Synth.Met.B, **122(1)**, 95-98 **(2001)**.
[4] V.P.Thalacker; Radiation Physics and Chemistry, **35(1-3)**, 18-29 **(1990)**.
[5] M.Nagao, Y.Murakami, N.Hozumi, M.Fukuma; 'Annual Report', 496-499 **(2001)**.
[6] R.Bartnikas; IEEE Transaction, Dielectrics and Electrical Insulation, **4(5)**, 544-577 **(1997)**.
[7] L.Castellani, F.Peruzzotti, A.Zaopo, P.L.Cinquemani, S.Foulger, J.C.Filippini, V.Lachevre; IEEE Transaction, Dielectrics and Electrical Insulation, **7(6)**, 818-824 **(2000)**.
[8] B.A.Briskman; Nuclear Instruments and Methods B, **185(1-4)**, 116-122 **(2001)**.
[9] T.Zaharescu, F.Ciuprina, P.Notingher; In: Proc. 8th IEEE 2004 IEEE International Conference on Solid Dielectrics (ICSD) Conf. on Conduction and Breakdown in Solid Dielectrics, Toulouse, France, 215 **(2004)**.
[10] S.Tonge, D.F.Shriver, J.H.Lai (Eds.); Polymers for Electronic Applications, CRC Press, Boca Rotan, FL, Chap. 5, **(1989)**.
[11] D.F.Shriver, P.G.Bruce (Eds.); Solid State Electrochemistry, Cambridge University Press, Cambridge, Chap. 5, **(1995)**.
[12] C.A.Vincent, B.Scrosati (Eds.); Modern Batteries, 2nd Ed., Arnold, London, Chap. 7, **(1997)**.
[13] Y.Takahashi; J.Polym.Sci.Polym.Phys., **35**, 193 **(1997)**.
[14] A.Tawansi, H.M.Zidan, A.H.Oraby, M.E.Dorgham; J.Phys.D: Appl.Phys., **31**, 3428 **(1998)**.
[15] J.Pacansky, S.Schneider; J.Phys.Chem., **94**, 3166 **(1990)**.
[16] M.J.Reimers, M.J.Cibulsky, T.A.Barbari; J.Polym. Sci., Polym.Phys., **31**, 537 **(1993)**.
[17] R.Lei, X.Jie, X.Jun, Z.Ruiyum; J.Appl.Polym.Sci., **53**, 325 **(1994)**.
[18] M.Todd Alam, Mat Celina, A.Roger Assink, L.Roger Clough, T.Kenneth Gillen; Radiation Physics and Chemistry, **60(1-2)**, 121-127 **(2001)**.

[19] V.Placek, B.Bartonícek, V.Hnat, V.B.Otahal; Nuclear Instruments and Methods B, **208**, 448-453 **(2003)**.

[20] S.Ilie, M.Tavlet; Nuclear Instruments and Methods B, **185(1-4)**, 318-322 **(2001)**.

[21] M.Tavlet; Nuclear Instruments and Methods B, **208**, 476-479 **(2003)**.

[22] Ni Li, Xiao-hong Qin, En-long Yang, Shan-yuan Wang; Materials Letters, **62(8-9)**, 31, 1345-1348 **(2008)**.

[23] R.D.Gould; Thin Solid Films, **125(1-2)**, 63-69 **(1985)**.

[24] F.H.Abd El-Kader, G.Attia, S.S.Ibrahim; J.Appl. Polym.Sci., **50**, 1281 **(1993)**.

[25] A.F.Basha, M.Amin, K.A.Darwish, H.A.Abdel Samed; Indian J.Polym.Mater., **5**, 115 **(1988)**.

[26] N.Venugeopl Reddy, V.V.R.Narasimha Rao; J.Mater.Sci.Lett., **11**, 1036 **(1992)**.

[27] S.Cygan, J.R.Laghari; IEEE Trans Nucl.Sci., **36**, 1386 **(1989)**.

[28] S.A.Gaafar, F.H.Abd El-Kader, M.S.Risk; Physica Scripta, **49**, 366 **(1994)**.

[29] M.Koski, S.M.Ceda; Jpn,J.Phys.Soc., **29**, 1012 **(1970)**.