Available online at http://www.journalcra.com

International Journal of Current Research
Vol. 5, Issue, 09, pp.2576-2581, September, 2013

INTERNATIONAL JOURNAL OF CURRENT RESEARCH

ISSN: 0975-833X

RESEARCH ARTICLE

RELAXATION DYNAMICS AND A.C. CONDUCTIVITY IN POLY (VINYL LCOHOL)/HYDROXYPROPYL CELLULOSE

[1]Mimouna M. Abutalib and [*2]Osiris W. Guirguis

[1]Physics Department, College of Science (Girls Branch), King Abdulaziz University, Jeddah, Saudi Arabia
[2]Biophysics Department, Faculty of Science, Cairo University, Giza 12613, Egypt

ARTICLE INFO

Article History:
Received 14th July, 2013
Received in revised form
06th August, 2013
Accepted 19th August, 2013

Key words:
Poly(vinyl alcohol),
Hydroxypropyl cellulose,
PVA/HPC blends,
Fast neutrons irradiation,
Dielectric properties.

ABSTRACT

The frequency-dependent dielectric properties (dielectric permittivity, dielectric loss factor and a.c. electrical conductivity) of blends of poly(vinyl alcohol)/hydroxypropyl cellulose (PVA/HPC) prepared by the solution-cast technique is investigated in the frequency ranges from 1 to 100 kHz to examine a wide range of molecular mobility of the amorphous phase of the blend systems. The dielectric properties on certain chosen blends before and after irradiation with different fast neutron fluencies in the range from 10^5 to 10^8 n/cm^2 are also studied. The obtained results noticed that, measureable changes in the behavior and values of the dielectric properties under investigation are detected. These changes may be attributed to degradation and/or cross-linking process either by the presence of HPC with different concentrations or by the irradiation with different fast neutron fluencies or both of them, which suggests that the observed dispersion depends on composition and irradiation of blend matrix.

INTRODUCTION

Polymers have numerous applications for biomedical purposes, as for instance: in prostheses, medical devices, dental materials, contact lenses, and pharmaceutical excipients. Their applications as drugs, drug-conjugates, enzyme-conjugates or gene delivery systems entered less in clinical practice (Popescu *et al.,* 2011). Blending, natural polymer with synthetic polymer seems to be an interesting way of polymeric composites (Çaykara *et al.,* 2006). Compatibility among components has a marked influence on the mechanical properties of polymer blends (Folkes and Hope, 1993). Intermolecular interactions regulate the compatibility among the component polymer molecules (Popescu *et al.,* 2011; Çaykara *et al.,* 2006; Folkes and Hope, 1993). The selection and use of polymers can potentially form hydrogen bonds when two polymers mixed, as well as the study of blends properties, are of importance to find further applications of the resulting blend materials for biomedical and pharmaceutical devices (Kulkarni and Sa, 2009). Cellulose was blended with poly (N-vinyl-2-pyrrolidone) (Çaykara *et al.,* 2006; Masson and Manley, 1991), poly (methyl methacrylate) (Nishioka *et al.,* 1993; Osiris *et al.,* 2012), and poly (2-hydroxy ethyl methacrylate) (Nisko and Hirose, 1992). Poly (vinyl alcohol) can be blended with poly (N-vinyl-2-pyrrolidone) (Nisko *et al.,* 1994), hydroxypropyl cellulose and hydroxypropyl methyl cellulose (Sakellariou *et al.,* 1993; Osiris and Manal, 2011; 2012; 2013; Abutalib *et al.,* 2013). Here, hydrogen bonding interaction is an important aspect of miscibility since intermolecular interactions regulate the compatibility among the component polymer molecules (Folkes and Hope, 1993). Poly (vinyl alcohol) (PVA) is a promising semi-crystalline material for biomedical applications. It is soluble in water and can be formed into hydrogels by freezing and thawing or crystallizing from an aqueous theta solution (Ebru Oral *et al.,* 2007). Poly (vinyl alcohol) represents one of the most widely investigated biodegradable synthetic polymers for ocular applications (Sintzel *et al.,* 1996). PVA can be made into a hydrogel by either chemical or physical cross-linking. PVA hydrogels was reinforced by crystallinity to develop a synthetic articular cartilage material for use in reconstructive joint surgery (Bray and Merrill, 2007; Kobayashi and Oka, 2004; Kobayashi *et al.,* 2005). In consideration of the prognosis in such patients, Kobayashi *et al.* (2003) developed artificial meniscus using polyvinyl alcohol hydrogel.On other hand, chemical or physical cross-linking can be achieved using functional cross-linking agents, such as gluteraldehyde, or by using ionizing radiation (Bodugoz-Senturka *et al.,* 2008). Ionizing radiation such as gamma or electron-beam produces two types of free radicals on PVA chains (Bhat *et al.,* 2005). In addition, chemically cross-linked PVA hydrogel has been gaining increasing attention in the field of biomedics (Kim *et al.,* 2003). In the present study, poly(vinyl alcohol) (PVA) was selected as the hydrogel component based on its favorable water-soluble, desirable physicochemical properties and its biocompatibility (Osiris and Manal, 2011).

Polysaccharides fabricated into hydrophilic matrices remain popular biomaterials for controlled-release dosage forms and the most abundant naturally occurring biopolymer is cellulose (Osiris and Manal, 2012; 2013; Abutalib *et al.,* 2013; Kamel*et al.,* 2008). Hydroxypropyl cellulose (HPC) belongs to the group of cellulose ethers which has been used already for a year by paper of conservators as glue and sizing material (Hofenk-de Graaff, 1981). Also, HPC can be used for production of time controlled delivery systems and is also used as a topical ophthalmic protectant and

---

Corresponding author: *Osiris W. Guirguis, Biophysics Department, Faculty of Science, Cairo University, Giza 12613, Egypt.*

lubricant (Chandra Mohan Eaga *et al.,* 2009). In pharmaceuticals, HPC is used as a disintegrants and a binder for the wet granulation method of making tablets (Ishikawa *et al.,* 2001). In the present work, blends of poly (vinyl alcohol)/hydroxypropyl cellulose (PVA/HPC) (100/0, 96/4, 92/8, 88/12, 84/16 and 0/100 wt/wt%) are prepared to enhance the dielectric properties of the poly(vinyl alcohol) (PVA) to be used as a biopolymer. Three prepared chosen PVA/HPC blends (100/0, 92/8 and 88/12 wt/wt%) are irradiated to four low-level fast neutron fluencies (10^5, 10^6, 10^7 and 10^8 n/cm^2). The induced changes in the dielectric permittivity (ε'), dielectric loss factor (ε'') and a.c. electrical conductivity (σ_{ac}) caused by both different concentrations of HPC and irradiation with fast neutron fluencies as functions of frequency in the range from 1 to 100 kHz are investigated.

EXPERIMENTAL WORK

Materials and sample preparation

Poly (vinyl alcohol) (PVA) granules with molecular weight of 125 kg/mole was supplied from El-Nasr Company, Cairo, Egypt. Hydroxypropyl cellulose (HPC; Pharmacoat 606) with molecular weight of 95 kg/mole was supplied by Shin Etsu Chemical Co., Tokyo, Japan. Thin transparent films of poly(vinyl alcohol) /hydroxypropyl cellulose (PVA/HPC) blend (100/0, 96/4, 92/8, 88/12, 84/16 and 0/100 wt/wt%) are prepared by using solution-cast technique (Osiris and Manal, 2011;2012;2013; Abutalib *et al.,* 2013; Suhas Nalle *et al.,* 2010; El-Zaher and Osiris, 2005). This method depends on the dissolution, separately, the weighted amounts of the poly (vinyl alcohol) (PVA) granules and hydroxypropyl cellulose (HPC) powder in double distilled water. Complete dissolution is obtained using a magnetic stirrer in a 50 ^{o}C water bath. To prepare thin films of the homopolymers (PVA and HPC) and the blend of their samples (PVA/HPC) with different weight percentages (96/4, 92/8, 88/12 and 84/16 wt/wt%), the solutions are mixed together at 50 ^{o}C with a magnetic stirrer. Thin films of appropriate thickness (about 0.01 cm) are cast onto stainless steel Petri dishes (10 cm diameter). The prepared films are kept at room temperature (about 25 ^{o}C) for 7 days until the solvent completely evaporated and then kept in desiccators containing fused calcium chloride to avoid moisture. The prepare thin films of the blend of PVA/HPC are irradiated with fission neutrons with mean energy of approximately 4.2 MeV from Americium-Beryllium (^{241}Am-Be) neutron source of activity 5 Ci (185 GBq) and with emission rate 0.87 x 10^7 n/s at the Physics Department, Faculty of Science, Al-Azhar University, Cairo, Egypt. During irradiation the samples are fixed in positions that neutrons were incident approximately normal. The irradiation temperature is adjusted to be about 25 ^{o}C. The films are exposed to four different fast neutron fluencies covers the range from 1 x 10^5 to 1 x 10^8 n/cm^2 (Abutalib *et al.,* 2013; Abdel Zaher and Osiris, 2012). The fast neutron fluence is measured using a calibrated TLD-700 Thermoluminescence detector at the location of the sample.

Dielectric measurements

The dielectric properties such as: dielectric permittivity (ε'), dielectric loss factor (ε'') and a.c. electrical conductivity (σ_{ac}) of the prepared PVA/HPC blended samples before and after irradiation with different fast neutron fluencies are measured in the frequency range 1-100 kHz at room temperature (about 25 ^{o}C) using a Hioki 3531Z Hiester Programmable Automatic RLC Meter (Japan). The accuracy of the dielectric measurements was within ±1%. To determine the dielectric permittivity ($\acute{\varepsilon}$), the relaxation dielectric loss factor (ε'') and the a.c. conductivity (σ_{ac}), the following equations are used (El-Bassyouni *et al.,* 2013):

$\acute{\varepsilon} = [100/8.85 \times 10^{-12}]\ (d/A)\ C$ (1)

$\varepsilon'' = \acute{\varepsilon} \tan \delta$ (2)

$\sigma_{ac.} = \varepsilon''\ f / 1.8 \times 10^{10} \quad (\mathrm{S.cm^{-1}})$ (3)

where, d = thickness of the sample (cm), A = Area of electrodes (cm^2) and C = capacitance (Farad), f is the applied frequency (Hz) and tan δ is the dielectric loss tangent obtained directly from the RCL meter.

RESULTS AND DISCUSSION

Dielectric permittivity

The investigation of dielectric permittivity of polymeric materials is one of the most methods of unravelling the origin of dielectric dispersion and relevant phenomena (Abouelhassan, 2010; Frübing, 2001). Figure (1) illustrates the frequency dependence of the dielectric permittivity (ε') for PVA/HPC blended samples. The inset of the figure shows the variation of ε' by increasing HPC content at 10 kHz. It is shown from the figure that the values of ε' for pure PVA are intermediate values between 96/4, 88/12 and 92/8 wt/wt% PVA/HPC blended samples. It is shown from Fig. (1) that, the values of the dielectric permittivity (ε') decreased as the applied frequency increased and they showed anomalous dispersion (Abou-Aiad *et al.,* 2002; Parada *et al.,* 1998). This behavior usually arises due to the occurrence of dielectric dispersion from the lag of the polymer molecules behind the alternations of the applied field (Abouelhassan, 2010; Osiris *et al.,* 1999). Moreover, the decrease in ε' may be attributed to the decreasing in the contribution of the dipolar orientational polarization which is primarily responsible for the high dielectric permittivities typical of high polymers to which PVA belongs (Osiris *et al.,* 1999; Garrett and Grubb, 1988; Salama *et al.,* 1988). The addition of HPC up to 8 wt% may cause an increase in the number of ionisable charge carriers which affects the inherent ability of the dipoles to orient themselves (Tager, 1972). Further addition of HPC up to 16 wt% may cause the distribution of HPC molecules in the inter-aggregates space which decrease the polymer chain elongation and consequently may result in a decrease in the dielectric permittivity. Also, It is clear from the inset of the figure that the values of ε' at 10 kHz for all investigated compositions of the blend were somewhat far from the line connecting the values of pure PVA and HPC (the ideal line) except 96/4 wt/wt% blend.

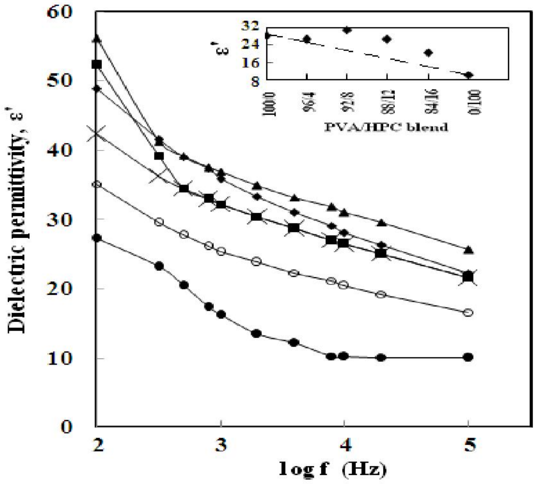

Figure 1. Frequency dependence of the dielectric permittivity (ε') for PVA/HPC blended samples: (♦) 100/0, (■) 96/4, (▲) 92/8, (x) 88/12, (o) 84/16 and (●) 0/100 wt/wt%. The inset of the figure shows the variation of ε' as a function of HPC concentration at 10 kHz.

On other hand, the decrement can be attributed to the fact that at low frequency the dielectric permittivity of the polar material is due to the contributions of multicomponents of the polarizability (deformation and relaxation components) (Abouelhassan, 2010; Frübing, 2001). In addition, as the frequency increases the dipoles are completely unable to follow the field, and hence the orientation polarization stops. The decrease of ε' tends to assume small values at higher frequencies which may be attributed to the polarization due to ions and electrons as a result of the defects present in the polymer matrix. In conclusion, the magnitude of the frequency dispersion depends on the composition, i.e. decreases as the concentration of HPC increases. Figures (2a-c) show the variation of ε' with frequency for PVA/HPC blended samples (100/0, 92/8 and 88/12 wt/wt%, respectively) before and after irradiation with neutron fluency at room temperature. It is clear from the figures that the values of ε' decrease with increasing frequency approaching a nearly constant value at higher frequencies. This confirms that already previously reported in the literatures (Abouelhassan, 2010; Osiris *et al.,* 1999).

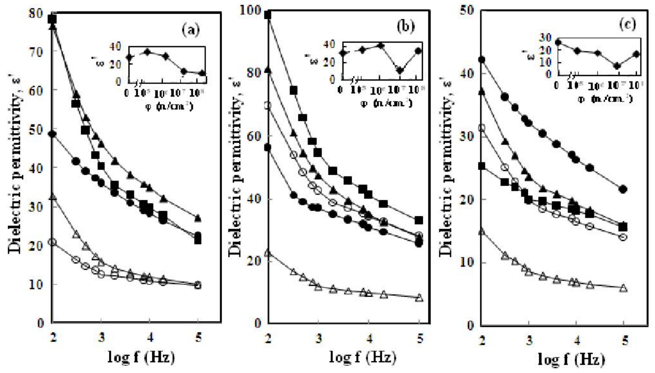

Figure 2. Frequency dependence of the dielectric permittivity (ε') for unirradiated and irradiated PVA/HPC blended samples with different fast neutron fluencies: (a) 100/0, (b) 92/8 and (c) 88/12 wt/wt%. (●) unirradiated, (▲) 1 x 10^5, (■) 1 x 10^6, (Δ) 1 x 10^7 and (○) 1 x 10^8 n/cm^2. The insets of the figures show the variation of ε' as a function of neutron fluency (φ) at 10 kHz.

From Figs. (2), it is noticed that the change in ε' is more at low frequency range for all the irradiated samples. This low frequency dispersion may be attributed to the presence of large polymer domain caused by order segments entangling on themselves giving rise to heterogeneous regions in the polymer blend system. At frequencies near ultraviolet, the contribution to polarization is mainly electronic. As the frequency is lowered the contribution to polarization from atomic or ionic polarization becomes significant. It is reasonable to suggest that there is a contribution from all sources to the polarization measured in these studies, where the maximum value of frequency is only 100 kHz. Thus, the large value of dielectric permittivity (ε') at low frequency suggests that there is a contribution from all four known source of polarization, namely, electronic, ionic, dipolar and space charge polarization at which the latter is known to contribute strongly at low frequencies.

Dielectric loss factor (ε'')

All dielectric materials have two types of losses. One is the conduction loss representing the flow of actual charges through the dielectric material, while the other is a dielectric loss due to movement or rotation of the atoms in an alternating field. Moreover, dielectric loss of pure polymers is due to the perturbation of the phonon system by the application of an electric filed, the energy transferred to the phonon is dissipated in the form of heat. Treatment of polymers by blending with appropriate additives modifies the perturbation of phonons during application of electric field. The dependence of dielectric loss factor (ε'') on frequency for PVA/HPC blended samples is shown in Fig. (3). The inset of the figure represents the variation of ε'' with HPC concentration at 10 kHz. From the figure, the values of ε'' for all the blended samples are lowers than that of the pure PVA one over the whole frequency range under investigation. Also, it is seen from the inset of the figure that the values of ε'' for all the investigated blends were somewhat far from the line connecting the values of PVA and HPC homopolymers (the ideal line) except for the blend 92/8 wt/wt%. As known water can influence the dielectric relaxation of polymers in various ways (Suzuki *et al.,* 1981). It usually enhances the local relaxation process by dipole-dipole coupling and it can act as an effective plasticizer in polar polymers. If water can form a hydrogen bonded super-structure, it produces a very specific dielectric response, often termed, “anomalous low frequency dispersion” (Dissado and Hill, 1983). The essence of this kind of response is that both ε' and ε'' decrease with increasing frequencies as noticed from the obtained results. On the other hand, the arrangement of the polar groups is considered as a very important factor affecting the polymeric units’ mobility. So that, if the electric field of this polar groups compensate one another, the polar bonds will be symmetrically arranged and consequently the polymeric chains will posses sufficient flexibility which increases the segmental movement of the polymer (Abouelhassan, 2010; Osiris *et al.,* 1999; Tager, 1972). This could be interpreted the decrease in the value of ε'' with the increasing of HPC contents. Figures (4a-c) show the dependence of the dielectric loss factor (ε'') on frequency for unirradiated and irradiated PVA/HPC blended samples (100/0, 92/8 and 88/12 wt/wt%, respectively). The plots show a steep decrease in ε'' at low frequencies. This is a general sequence attributed to decrease in the stir effect hindering the micro-Brownian motion. At high frequencies the variation of ε'' with frequency is marginal and tends to be a constant value.

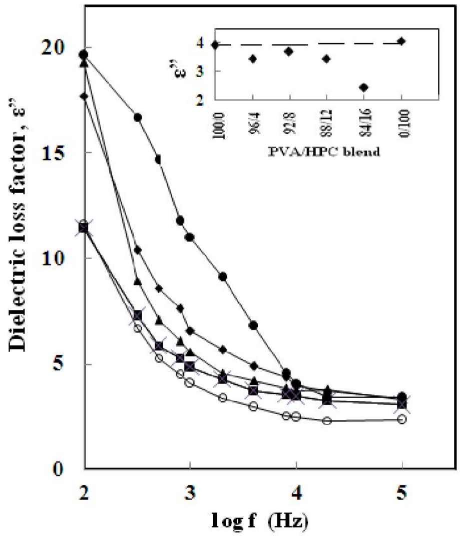

Figure 3. Frequency dependence of the dielectric loss factor (ε'') for PVA/HPC blended samples: (♦) 100/0, (■) 96/4, (▲) 92/8, (x) 88/12, (o) 84/16 and (●) 0/100 wt/wt%. The inset of the figure shows the variation of ε'' as a function of HPC concentration at 10 kHz.

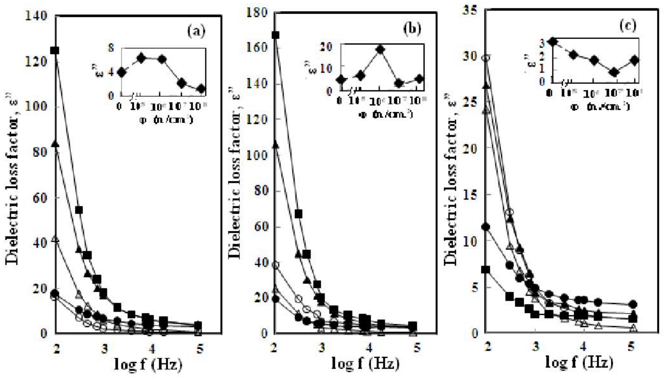

Figure 4. Frequency dependence of the dielectric loss factor (ε'') for unirradiated and irradiated PVA/HPC blended samples with different fast neutron fluencies: (a) 100/0, (b) 92/8 and (c) 88/12 wt/wt%. (●) unirradiated, (▲) 1 x 10^5, (■) 1 x 10^6, (Δ) 1 x 10^7 and (○) 1 x 10^8 n/cm^2. The insets of the figures show the variation of ε'' as a function of neutron fluency (φ) at 10 kHz.

It is recognized from Figs. (4) that HPC concentration and neutron irradiation plays a predominant role in both morphological and microstructure change occurring in the polymer matrix (Suzuki *et al.,* 1983). Such blended sample behaves as a system in which one phase relaxes like the amorphous one while the existence of crystalline phase restrict the general long-range segmental motions in the amorphous phase (Abo-Ellil *et al.,* 2000). Therefore, the variation of magnitude of the dielectric loss may be attributed to the change in the microstructure induced by the addition of either HPC and/or fast neutron fluencies.

a.c. electrical conductivity (σ_{ac})

Figure (5) represents the variation of a.c. electrical conductivity with frequency for PVA/HPC blended samples. It is clear from the figure that as the concentration of HPC increases up to 16 wt%, the conductivity value decreases. Moreover, σ_{ac} is monotonically increases with increasing frequency. The plot of the pure PVA sample is the highest while the 84/16 wt/wt% blended sample takes the lower conductivity values. The inset of the figure illustrates that the a.c. conductivity values at 10 kHz decrease with increasing the HPC contents. It is found from Fig. (5) that, the decrease in σ_{ac} may be due to that the addition of HPC causes a decrease in the mobility of charge carriers due to scattering of ionized molecular aggregates which may be formed due to inhomogeneous distribution as well as the non-bonding parts of the blend. Furthermore, this behavior is well defined experimentally for a wide range of materials, the conduction of which occurs by hopping mechanism (Sharma *et al.,* 1991). Figures (6a-c) show the dependence of σ_{ac} on frequency for unirradiated and irradiated PVA/HPC blended samples (100/0, 92/8 and 88/12 wt/wt%, respectively). It is clear from Fig. (6a) for pure PVA sample that the conductivity increases for 10^5 and 10^6 n/cm^2 compared with the unirradiated one. Further increase in neutron fluency up to 10^8 n/cm^2 causes a decrease in the electrical conductivity.

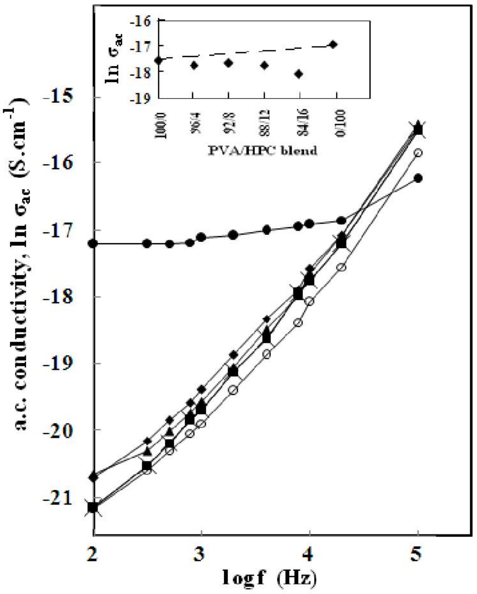

Figure 5. Frequency dependence of the a.c. conductivity, ($\ell n\ \sigma_{ac}$) for PVA/HPC blended samples: (♦) 100/0, (■) 96/4, (▲) 92/8, (x) 88/12, (o) 84/16 and (●) 0/100 wt/wt%. The inset of the figure shows the variation of $\ell n\ \sigma_{ac}$ as a function of HPC concentration at 10 kHz.

It is seen from Fig. (6b) for 92/8 wt/wt% blend that, over the whole frequency range, the values of σ_{ac} for the irradiated sample with neutron fluence 10^6 n/cm^2 are the highest values and then decreases with increasing the neutron fluencies. In addition, from Fig. (6c) for 88/12 wt/wt% blend, it is clear that, over the whole frequency range, the irradiated sample with neutron fluence 10^6 n/cm^2 has minimum values in the low frequency range while irradiated sample with neutron fluence 10^7 n/cm^2 has minimum values in the high frequency range. From Figs. (6), as discussed, the decrease in the values of σ_{ac} with neutron fluencies may be due to the predominance of cross-linking process caused by higher fluencies. On the other hand, the increase in σ_{ac} may be attributed to the occurrence of degradation process as a result of irradiation with higher fluencies of fast neutrons (Abo-Ellil *et al.,* 2000). In general, the conductivity of the amorphous materials where the charge carriers experience on approximately random potential energy on diffusing is found to obey the following power law (Dutta *et al.,* 2000):

$$\sigma_{ac}(\omega) = A\ \omega^S \quad (4)$$

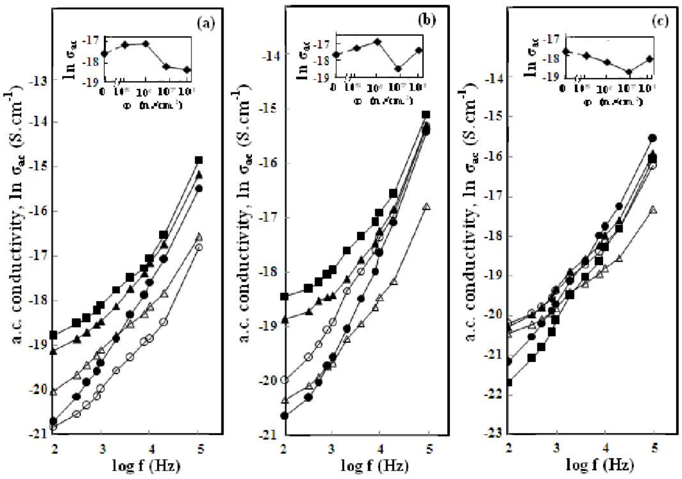

Figure 6. Frequency dependence of the a.c. conductivity ($\ell n\ \sigma_{ac}$) for unirradiated and irradiated PVA/HPC blended samples with different fast neutron fluencies: (a) 100/0, (b) 92/8 and (c) 88/12 wt/wt%. (●) unirradiated, (▲) 1 x 10^5, (■) 1 x 10^6, (Δ) 1 x 10^7 and (○) 1 x 10^8 n/cm^2. The insets of the figures show the variation of $\ell n\ \sigma_{ac}$ as a function of neutron fluency (φ) at 10 kHz.

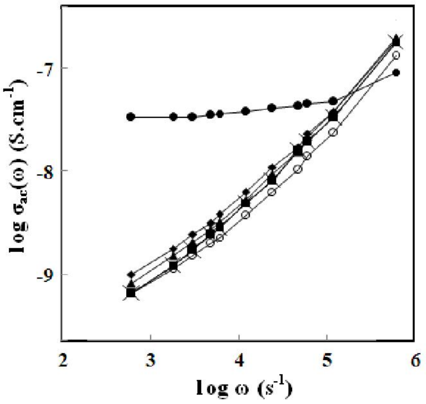

Figure 7. Frequency dependence ($\ell og\ \omega$) of a.c. conductivity ($\ell og\ \sigma_{ac}$) for PVA/HPC blended samples: (♦) 100/0, (■) 96/4, (▲) 92/8, (x) 88/12, (o) 84/16 and (●) 0/100 wt/wt%.

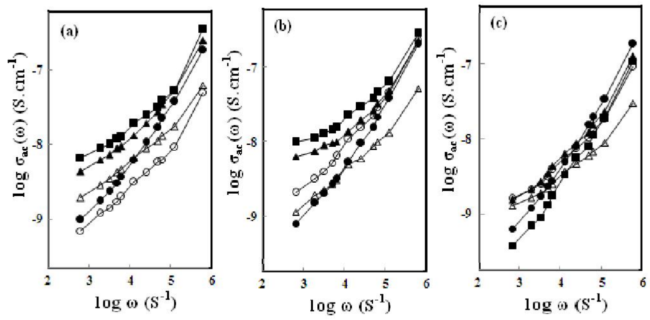

Figure 8. Frequency dependence ($\ell og\ \omega$) of a.c. conductivity ($\ell og\ \sigma_{ac}$) for unirradiated and irradiated 100/0 (a), 92/8 (b) and 88/12 wt/wt% (c), PVA/HPC blended samples with different fast neutron fluencies: (a) pure PVA, (b) 8 wt% and (c) 12 wt%. (●) unirradiated, (▲) 1 x 10^5, (■) 1 x 10^6, (Δ) 1 x 10^7 and (○) 1 x 10^8 n/cm^2.

where A is a complex constant and the index (S) is the exponent of the angular frequency (ω). With exponent S < 1 up to frequency 1 MHz is considered to signify the coupling of an ions movement with its environment (Osiris *et al.,* 1999). The value of S has been determined from the linear slope of $\ell og\ \sigma_{ac}(\omega)$ versus $\ell og\ \omega$. Figure (7) represents the frequency dependence ($\ell og\ \omega$) of a.c. conductivity ($\ell og\ \sigma_{ac}$) for PVA/HPC blended samples. It is clear from the figure that as the concentration of HPC increases up to 16 wt%, the values of conductivity value decreases. As mentioned before, σ_{ac} is monotonically increases with increasing frequency. The plot of the pure PVA sample is the highest while the 84/16 wt/wt% blended sample takes the lower conductivity values. In addition, Figs. (8a-c) show the relation between ℓog ω and ℓog σ_{ac} for unirradiated and irradiated 100/0, 92/8 and 88/12 wt/wt% PVA/HPC blended samples, respectively. As can be seen from the Figs. (7 and 8) for the blended samples before and after irradiation with fast neutron fleunce, the plots for each sample have yielded nearly straight lines for all considered samples up to 10 kHz. This type of behavior reveals that the exponent S is approximately independent of frequency; hence, the mechanism responsible for a.c. conduction mechanism could be a hopping one. It is found to be consistent with that observed in many hopping system (Saleh *et al.,* 1993). Two distinct mechanisms have been proposed for the relaxation phenomena: (1) quantum mechanical tunneling (QMT) of electrons or polarons through the barrier separating localized states and (2) correlated barrier hopping (CBH) model over the same barrier. According to the QMT model, the power S is either independent or an increasing or decreasing function of frequency and fast neutron irradiation. The obtained values of S were derived by calculating the slopes of the curves in Figs. (7 and 8) which use a single exponent S and represent the hopping conduction near the Fermi level and are recorded in Tables (1) and (2).

Table 1. Values of the exponent S for PVA/HPC blended samples.

Blend sample PVA/HPC (wt/wt%)	Exponent S
100/0	0.739
96/4	0.798
92/8	0.753
88/12	0.798
82/16	0.712
0/100	0.087

Table 2. Values of the exponent S for unirradiated and irradiated 100/0, 92/8 and 88/12 wt/wt% PVA/HPC blended samples with neutron fluencies

Neutron fluence (φ) (n/cm^2)	Glass transition temperature, T_g (K), for the blended samples (wt/wt%)		
	100/0	92/8	88/12
Unirradiated	342	371	314
1 x 10^5	343	347	354
1 x 10^6	334	356	354
1 x 10^7	357	366	365
1 x 10^8	370	347	339

From the data obtained the values of S change regularly with increasing the HPC concentration in the blend. Moreover, it is observed that the exponent S for the chosen irradiated blends decreases with increasing neutron fluency up to 10^6 n/cm^2 and then returns back towards the unirradiated value with increasing the neutron fluency up to 10^8 n/cm^2, but the S values are less than unity, i.e. $S < 1$. This means that, the results of frequency and fast neutron irradiation dependence of a.c. conductivity obtained were found to support the correlated barrier hopping (CBH) model, where the frequency dependence of a.c. conductivity is linear, In conclusion, the obtained S values suggest that the correlated barrier hopping mechanism depends on composition and the irradiated blend matrix.

Acknowledgement

The authors are very grateful to Prof. Dr. Nabawia A. Abdel Zaher, Professor, National Institute for Standard, Giza, Egypt, for her kind help to bring this work.

REFERENCES

Abo-Ellil, M.S.; Gaafar, S.A.; Abd El-Kader, F.H. and Kamel, N.A. (2000). Effect of fast neutrons on dielectric properties of pure and gelatin doped-poly (vinyl alcohol) films. Journal of Polymer Research 7: 149-53.

Abou-Aiad, T.H.; El-Sabee, M.Z.; Abd-El-Nour, K.N.; Saad, G.R.; El-Sayed A. El-Sayed and Gaafar E.A. (2002). Miscibility and the specific interaction of polyhydroxybutyrate blended with polyvinylacetate and poly(vinyl acetate-co-vinyl alcohol) with some biological applications. Journal of Applied Polymer Science 86: 2363-74.

Abouelhassan, S. (2010). Investigation of the dielectric properties and thermodynamic parameters of (50-x) P_2O_5-xAgI-40Ag_2O-10Fe_2O_3 ionic glass. Chinese Journal of Physics 48: 650-61.

Abutalib, M.M.; Osiris W. Guirguis and Nabawia A. El-Zaher (2013). Optical studies of fast neutron irradiated poly (vinyl alcohol)/hydroxypropyl cellulose blends. Materials Science (An Indian Journal), 9: 433-44.

Bhat, N.V.; Nate, M.M.; Kurup, M.B.; Bambole, V.A. and Sabharwal, S. (2005). Effect of γ-radiation on the structure and morphology of polyvinyl alcohol films. *Nuclear* Instruments and Methods in Physics Research Section *B* 237: 585-9.

Bodugoz-Senturka, H.; Choi, J.; Oral, E.; Kung, J.H.; Macias, C.E.; Braithwaite, G. and Muratoglu, O.K. (2008). The effect of polyethylene glycol on the stability of pores in polyvinyl alcohol hydrogels during annealing. Biomaterials 29: 141–9.

Bray, J.C. and Merrill, E.W. (1973). Poly (vinyl alcohol) hydrogels for synthetic articular cartilage material. *Journal* of *Biomedical* Materials Research 7: 431-43.

Çaykara, T. and Demirci, S. (2006). Preparation and characterization of blend films of poly (vinyl alcohol) and sodium alginate. Journal of Macromolecular Science, Part A: Pure and Applied Chemistry 43: 1113-21.

Chandra Mohan Eaga, Jagan Mohan Kandukuri, Venkatesham Allenki and Madhusudan Rao Yamsani, *In-situ* gels -a novel approach for ocular drug delivery. Der Pharmacia Lettre 1: 21-33.

Dissado, L.A. and Hill, R.M. (1983). A cluster approach to the structure of imperfect materials and their relaxation spectroscopy. Proceedings of the *Royal* Society A 390: 131-80.

Dutta, P.; Biswas, S.; Ghosh, M.; De, S.K. and Chatterjee, S. (2000). The dc and ac conductivity of polyaniline-polyvinyl alcohol blends. Journal of Synthetic Materials 122: 455-61.

Ebru Oral; Hatice Bodugoz-Senturk; Celia Macias and Orhun K Muratoglu (2007). Vitamin C hinders radiation cross-linking in aqueous poly(vinyl alcohol) solutions. Nuclear Instruments and Methods in Physics Research B 265: 92–7.

El-Zaher, N.A. and Osiris, W.G. (2005). Thermal and structural properties of poly(vinyl alcohol) doped with hydroxypropyl cellulose. Journal of Applied Polymer Science 96: 1914-23.

Folkes, M.J. and Hope, P.S. (1993). Polymer Blends and Alloys. Chapman and Hall, London.

Frübing, P. (2001). Dielectric Spectroscopy, University of Potsdam, Institute for Physics, Germany.

Garrett, P.D. and Grubb, D.T. (1988). Effect of drawing on the α relaxation of poly(vinyl alcohol). Journal of Polymer Science Part *B: Polymer Physics* 26: 2509-23.

Gehan T. El-Bassyouni; Osiris W. Guirguis and Wafa I. Abdel-Fattah (2013). Morphological and macrostructural studies of dog cranial bone demineralized with different acids. Current Applied Physics 13: 864-74.

Hofenk-de Graaff, J. (1981). In Central Research Laboratory for Objects of Art and Science. Gabriel Metsustroat and 1071 EA: Amsterdam.

Ishikawa, T.; Mukai, B.; Shiraishi, S.; Utoguchi, N.; Fujii, M.; Matsumoto, M.A. and Watanabe U. (2001). Studies of rapidly disintegrating tablet prepared by direct compression method. *Chemical* and Pharmaceutical Bulletin 49: 134-9.

Kamel, S.; Ali, N.; Jahangir, K.; Shah, S.M. and El-Gendy, A.A. (2008). Pharmaceutical significance of cellulose: A review. eXPRESS Polymer Letters 2: 758-78.

Kim, S.J.; Lee, C.K.; Lee, Y.M.; Kim, I.Y. and Kim, S.I. (2003). Electrical/pH-sensitive swelling behavior of polyelectrolyte hydrogels prepared with hyaluronic acidpoly(vinyl alcohol) interpenetrating polymer networks. Reactive and Functional Polymers 55: 291-8.

Kobayashi, M. and Oka, M. (2004). Characterization of a polyvinyl alcohol-hydrogel artificial articular cartilage prepared by injection molding. Journal of *Biomaterials Science, Polymer Edition* 15: 741-51.

Kobayashi, M.; Chang, Y.S. and Oka, M. (2005). A two year in vivo study of polyvinyl alcohol hydrogel (PVA-H) artificial meniscus. Biomaterials 26: 3243-8.

Kobayashi, M.; Toguchida, J. and Oka, M. (2003). Preliminary study of polyvinyl-alcohol-hydrogel (PVA-H) artificial meniscus. Biomaterials 24: 639-47.

Kulkarni, R.V. and Sa, B. (2009). Electroresponsive polyacrylamide-grafted-xanthan hydrogels for drug delivery. Journal of Bioactive and Compatible Polymers 24: 368-84.

Masson, J. and Manley, R.S. (1991). Miscible blends of cellulose and poly (vinylpyrrolidone). Macromolecules 24: 6670-9.

Nabawia A. Abdel Zaher and Osiris W. Guirguis (2012). Optical studies of fast neutron irradiated composites of poly (vinyl alcohol) and bovine serum albumin. Materials Science (An Indian Journal) 8: 370-82.

Nishioka, N.; Yamamoka, M.; Haneda, H. and Kawakami, K. (1993). Thermal decomposition of cellulose/synthetic polymer blends containing grafted products. 1. Cellulose/ poly (methyl methacrylate) blends. Macromolecules 26: 4694-9.

Nisko, Y. and Hirose, N. (1992). Cellulose/poly (2-hydroxyethyl methacrylate) composites prepared via solution coagulation and subsequent bulk polymerization. 33: 1519-24.

Nisko, Y.; Suziki, H. and Sato, K. (1994). Molecular orientation and optical anisotropy induced by the stretching of poly (vinyl alcohol) poly (N-vinyl pyrrolidone) blends. Polymer 35: 1452-61.

Osiris W. Guirguis and Manal T.H. Moselhey (2011). Optical study of poly (vinyl alcohol)/hydroxypropyl methylcellulose blends. Journal of Materials *Science* 46: 5775-89.

Osiris W. Guirguis and Manal T.H. Moselhey (2012). Thermal and structural studies of poly(vinyl alcohol) and hydroxypropyl cellulose blends. Natural Science; 4: 57-67.

Osiris W. Guirguis and Manal T.H. Moselhey (2013). Optical properties of poly (vinyl alcohol)/hydroxypropyl cellulose blends. Materials Science (An Indian Journal) 9: 8-23.

Osiris W. Guirguis; Wedad A. Alharbi and Jamila H. Alzahrani (2012). Physical study of poly(methyl methacrylate)/rare earth composite luminescent materials. Materials Science (An Indian Journal) 8: 423-33.

Osiris, W.G.; Gaafar, S.A.; Mohamed, S.I. and Elsayed, A.A. (1999). Induced changes in the dielectric properties of pure and albumin-doped poly (vinyl alcohol) films due to irradiation with fast neutrons. Egyptian Journal of Biophysics (EJB) 5: 283-302.

Parada, L.G.; Casteros, L.C.; Meauria, E. and Katime, I. (1998). Miscibility in blends of poly(vinyl acetate-co-vinyl alcohol) with poly(N,N-dimethylacrylamide). Polymer 39: 1019-24.

Popescu, I.; Suflet, D.M.; Pelin, I.M. and Chitanu, G.C. (2011). Biomedical applications of maleic anhydride copolymers. Revue Roumaine de Chimie, 56: 173-88.

Sakellariou, P.; Hassan, A. and Rowe, R.C. (1993). Phase separation and polymer interactions in aqueous poly(vinyl alcohol)/hydroxypropyl methylcellulose blends. Polymer 34: 1240-8.

Salama, A.A.; Youssef, S.; Osiris, W.G. and Hashad A.M. (1988). Effects of neutrons on some physical properties of cellulose nitrate for application in radiation dosimetry. Polymer Degradation and Stability 22: 275-83.

Saleh, A.M.; Gould, R.D. and Hassan A.K. (1993). Dependence of AC electrical parameters on frequency and temperature in zinc phthalocyanine thin films. Physica Status Solidi (a) 139: 379-89.

Sharma, A.K.; Adinarayana, V. and Santhi Sagar, D. (1991). dc-conduction mechanism in Fe-doped polystyrene films. Materials Letters 12: 247-51.

Sintzel, M.; Bernatchez, S.; Tabatabay, C. and Gurny, R. (1996). Biomaterials in ophthalmic drug delivery. European *Journal* of Pharmaceutics and Biopharmaceutics 42: 358-74.

Suhas Nalle, Rupali Sarpate, Mallikarjuna Setty, Patan Inayat and Anand Deshmuk (2010). Controlled release nateglinide tablets using Na-CMC and HPC hydrophilic polymer. Research Journal of Pharmacy and Technology 3: 87-91.

Suzuki, T.; Adachi, K. and Kotaka T. (1981). Dielectric relaxations of water molecules occluded in polymeric media: Some hydrophobic polymer systems. Polymer Journal 13: 385-97.

Tager, A.A. (1972). Physical Chemistry of Polymers, Mir Publishers, Moscow.

****