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Effects of Gamma Radiation on Electrical Conductivity of PVA-CH Composites

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Abstract. The effects of radiation on polymer composite PVA-based organic blends containing chlorine have been studied for their potential applications in electrochemical devices. The polymer composite PVA-Chloral Hydrate (CH) were blended separately with 23, 34, 45 and 57% CH. The composite films were prepared by solvent-casting method and each film has been irradiated with γ -rays at different doses up to 12 kGy. The electrical properties have been studied using an impedance analyzer of LCR meter in the frequency range from 20 Hz to 1 MHz. The conductivity-dose relation study revealed that increase in conductivity of the irradiated PVA-CH blends with increasing dose up to 12 kGy. The increase in the conductivity with dose is attributed to the increase of ionic carriers in the composites induced by radiation scission of CH molecules and also due to hydrolysis of water.

Introduction

The physical, chemical and biological properties of materials may be modified by treatment with ionizing radiation in the form of gamma rays, energetic electrons and ion beams. This radiation processing technique has become very important in the present material processing technology because it is less expensive and is environmental friendly it offer many advantages when compared with traditional chemical processes. The mechanisms of radiation processing of materials include polymerization, cross-linking, grafting, curing and scission.

Gamma irradiation processing is widely known for cross-linking plastic materials. High energy electron beam cross-linking is used to produce heat-shrinkable plastic films for packaging foods and other consumer products, heat-shrinkable plastic tubing and encapsulations for industrial products, wire insulations, plastic foam and hydrogels for medical applications. Natural and synthetic polymers have been treated with ionizing radiation to modify their mechanical, structural, optical and electrical properties for various applications such as dosimetry and electrochemical, optical and electronic devices [1-3]. When polymers are exposed to high energy radiation such as gamma rays, there may be cross-linking or scission of the long polymer chains. Both processes may occur simultaneously, but usually one dominates [4].

In general, pure polymers are insulators having very low electrical conductivity at room temperature. Their structural, physical and chemical properties can be changed with addition of salts, blends and plasticizers to form solid composite polymers [5]. Electrochemical methods of energy storage and conversion are interest for many practical applications. There has been an ever increasing demand for portable batteries used in electronic devices. The focus has been on relatively low-cost and environmentally friendly electrode materials and electrolytes used in batteries and fuel cells. Therefore, much research has been devoted to work on electrodes and electrolytes for battery, super capacitor and fuel cell applications [6]. PVA is a water soluble polymer which is in widespread use in several interesting physical properties and very useful in technical applications in biochemical and medical. PVA is a potential material having a good charge storage capacity and dopant-dependent electrical and optical properties [7].

PVA has the different internal structure which may be considered as amorphous or semi crystalline. The semi crystalline structure of PVA shows an important feature rather than the

amorphous one, electrical conductivity material PVA can be tailored to a specific requirement by addition of suitable doping material and irradiation [8,9]. Polyvinyl alcohol (PVA) is a transparent, water-soluble and biodegradable polymer with many technological, excellent chemical resistance, good mechanical properties and pharmaceutical and biomedical applications [11,12]. PVA is good insulating material with low conductivity and importance to microelectronic industry. Its electrical conductivity depends on the thermally generated carriers and also on the addition of suitable dopants [12].

The hydroxyl groups present in its main backbone are responsible for the strong intra- and intermolecular hydrogen bonds, endowing PVOH with many good properties, such as high tensile strength, excellent adhesive properties, abrasion resistance, chemical resistance and gas barrier properties [13, 16]. The aim of this study is to investigate the effect of ionizing radiation on PVA-based blended with CH for potential applications in electrochemical devices. The quantitative and qualitative aspects are studied in terms of their electrical properties.

MATERIALS AND METHODS

Preparation of PVA Polymer Composite

The PVA stock was supplied by SIGMA ($M_w = 70,000$ g/mol, 99 – 100% hydrolyzed). The PVA solutions were made by dissolving PVA powder in double distilled water at temperature of 90 °C in a water-bath. The solution was magnetically stirred throughout at that temperature for 2 hours and then left to cool at room temperature. To each 50 ml of the well mixed solution, 23, 34, 45 and 57% of chloral hydrate (CH) product of Scharlau Chemie were added, stirred for 12 hours and poured onto a 15×15 cm² horizontal glass plate and dried at room temperature for about 72 hours. The films were peeled off and cut into 2 x 4 cm pieces, dried, stored and ready for irradiation. The average thickness of the film was found to be 70 μ m, which was measured within an accuracy of ± 1 μ m by a digital micrometer (Mitutoyo). PVA composite films were irradiated with 1.25 MeV γ -rays from a J.L Sherperd type γ -ray Co⁶⁰ source at a mean dose rate of 163.75 Gy min⁻¹. $T_{1/2}$ is the half-life of the source, which is 5.272 years. Conductivity measurements were made using HP 4284A LCR meter which operates in the frequency range from 20 Hz to 1 MHz



Fig. 1. PVA-CH composite films at different doses

RESULTS AND DISCUSSION

Effects of radiation on conductivity of the PVA-CH composites.

Polymer composites of an amorphous phase are considered heterogeneous disordered systems. The electrical transport mechanism in amorphous materials is very complex and usually assumed by hopping charge carriers such as ions (cations and anions) and polarons or bipolarons. The conductivity depends on the several factors such as the concentration and type of blends added [3, 5, 12]. The effect of radiation on conductivity of polymer composites comprises of three stages: generation of more ionic carriers by radiation scission of organic plasticizers; the immobilization of the carriers by trapping at localized sites or other recombination mechanisms; and finally the motion of these carriers under the influence of density gradients (diffusion) and any existing electric fields (drift) [2, 3, 5].

The electrical conduction is via ionic hopping between localized sites over a potential barrier with the hopping frequency depending on the absorbed dose [17]. The number of charge carriers

localized in the trapping sites are quantized in energy and depends on dose; therefore dose sensitivity is an important parameter to quantify the effect of ionizing radiation on the conductivity.

Dose-Dependent AC Conductivity

The total conductivity $\sigma_{ac}(\omega)$ was determined from the measured parallel conductance $G_p(\omega)$. Figure 2 shows the variation of the total conductivity as a function of frequency for the PVA-CH films at different doses. All the PVA-CH samples exhibit a frequency-independent conductivity at low frequencies and a frequency-dependent conductivity at high frequencies. There is no electrode polarization effect in all the samples since there is no appearance of tails of the conductivity data at the low frequency range, but instead it shows a constant value at low frequencies. Some charges are weakly bound and partially free to move independent of frequency and contributed to dc conductivity [3].

The switch over from the frequency-independent region to frequency-dependent region is the signature of the onset of conductivity relaxation, which shifts towards higher frequencies as the dose increases. The strong frequency dispersion may be due to increase connectivity of conductive inhomogeneities in the amorphous structure, which allows the transport of charge carriers from one site to another via ionic hopping mechanism or even by phonon-assisted tunneling process [3, 10, 17]. An increase of conductivity with dose is attributed to charge carriers in the composites, induced by radiation scission of CH molecules and hydrolysis of water in the samples to produce more ions of Cl^- , OH^- , and H^+ that are trapped in localized sites in the PVA matrix. The chloral hydrate acts as an ionic carrier source, which increases with concentration of CH.

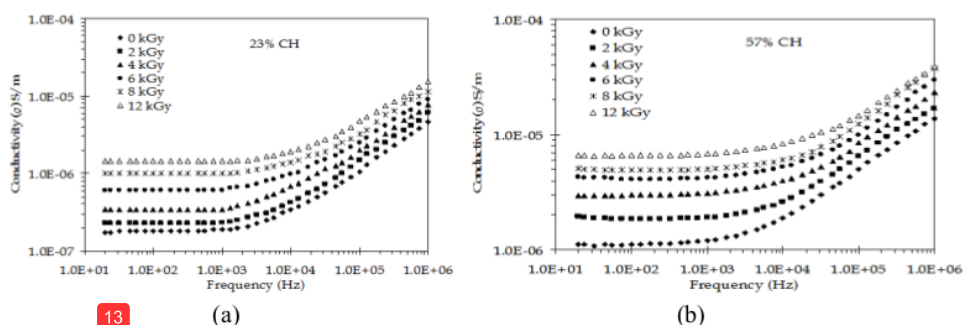


Fig. 2. Total conductivity as a function of frequency for PVA-CH composites at different doses with (a) 23% and (b) 57% CH composition.

It was found at the lowest frequency of 20 Hz that the conductivity for 23% CH, increases from $1.73 \times 10^{-7} \text{ S m}^{-1}$ at zero doses to $1.46 \times 10^{-6} \text{ S m}^{-1}$ at 12 kGy. While for 57% CH, the conductivity was increased from $1.11 \times 10^{-6} \text{ S m}^{-1}$ to $6.64 \times 10^{-6} \text{ S m}^{-1}$ at 12 kGy. From these results it could be concluded that the conductivity increases with the increase of absorbed dose and CH composition. The influence of dose was about 8.4 times for 23% CH and 5.9 times for 57% CH at 20 Hz, whereas the influence of CH composition was about 6.4 times. Most polymers have insulating properties. An insulator is a dielectric material, which is electrically non-conducting and exhibits or may be made to exhibit an electric dipole structure [4].

Polymers having the dipole moment aligned parallel to the chain contour exhibit conductivity due the fluctuation of the end-to-end vector of the chains. Parameter relating to the electrical insulating property is the dielectric permittivity, which may reveal the presence of dipoles in the bulk materials. This parameter is useful in conductivity study to understand the structure-property behavior of composite polymers [12]. The effects of radiation below 20 kGy on PVA based composites result in the formation of more ionic carriers by radiation scission of organic plasticizers, which may be immobilized by trapping at localized sites in the binder matrix and can

be made in motion under the influence of density gradients (diffusion) and any existing electric fields.

The electrical conduction of these materials is via ionic hopping with the hopping frequency increasing with the increase of the absorbed dose. An increase in conductivity leads to a drop in the dipole relaxation time of most segmental species of composite polymers, resulting in increase in the degree of dipole orientation and consequently enhances the conductivity. Therefore, dose sensitivity is an important parameter in quantifying the effect of ionizing radiation on the conductivity.

Frequency Exponent

The total frequency-dependent conductivity $\sigma_{ac}(\omega)$ at a given dose is the sum of dc and ac components of the conductivity and can be expressed as

$$\sigma_{ac}(\omega) = \sigma_{dc}(0) + \sigma(\omega) \quad (1)$$

where $\sigma_{dc}(0)$ is frequency-independent dc conductivity at low frequency region, which is a constant and $\sigma(\omega)$ is frequency-dependent ac conductivity at high frequency region, which increases with increasing frequency [14]. The conductivity $\sigma_{ac}(\omega)$, in Figure 3 obeys Jonscher's universal power law (UPL) [15], which is applicable to very wide materials irrespective of their chemical and physical structures and the type of charge carriers (Duta, 2001) written in the form

$$\sigma(\omega) = A\omega^s \quad (2)$$

where A is a constant dependent on the dose and composite composition, $\omega = 2\pi f$ is the angular frequency and s ($0 < s < 1$) is the frequency exponent, which is dependent on dose or $s(D)$. The value of s at various doses for PVA-CH composites has been evaluated from the gradient of linear slope of $\log \sigma(\omega)$ vs. $\log \omega$, as illustrated in Figure 3 for 23, 57% CH respectively. The value of s decreases with the increase of dose and CH composition as shown in Figure 4. When irradiated to 12 kGy, s value decreases from 0.62 to 0.5 for 23% CH and from 0.40 to 0.30 for 54% CH. The results confirm the domination of ionic conductivity by ionic hopping when $s < 1$. As the dose increases more ions are generated and trapped at localized a site that allows more ions hopping between the localized sites. As the CH composition increases more ions are present in the samples. An increase of the conductivity by ionic hopping leads to a decrease in the value of s . The dose dependence of s is very complex and has not been studied until now. Most measurements found in the literatures were based on temperature dependence of the conductivity of composite polymers with or without being treated with ionizing radiation.

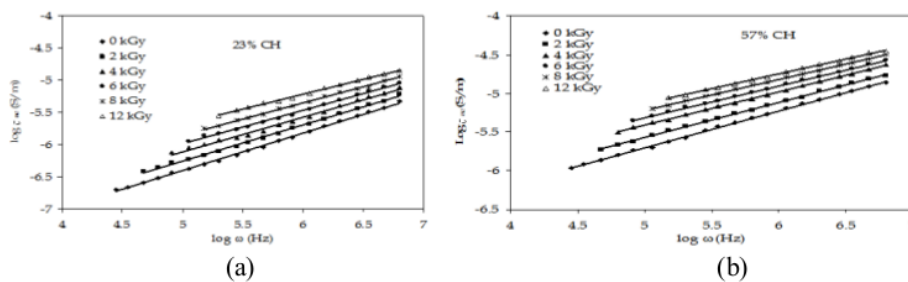


Fig. 3. Variation of $\log \sigma(\omega)$ versus $\log \omega$ at different doses for PVA-CH composites at (a) 23% and (b) 57% chloral hydrate.

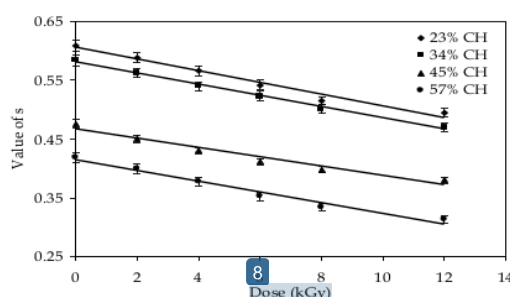


Fig. 4. Frequency exponent s as a function of dose for ac conductivity of PVA-CH composites at different CH compositions.

Conclusion

The effects of ionizing radiation enhanced the electrical conductivity¹⁸ dyed PVA-CH, blends in treated unirradiated blends. Results from conductivity study revealed that the conductivity consists of frequency-independent dc component and frequency-dependent ac component. The increase of dc conductivity with dose is attributed to the increase of free ion generated by the radiation. The power law, $A\omega^s$ where $s \leq 1$, of ac conductivity has been attributed to the hopping of ions trapped in the localized sites of the PVA matrix and resulted in the frequency exponent s decreasing with increasing dose. The increase of dc conductivity with dose is attributed to the increase of trapped ions generated by radiation in the polymer matrix. This suggests that conductivity is governed by the formation of more ion charge carriers as induced by gamma radiation by the ions.

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