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Studies on dielectric properties, AC-conductivity and impedance spectroscopy of γ – ray irradiated PVA(1-x)PSx polymer films

KEYWORDS

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ABSTRACT *The effects of gamma radiations on the properties of $PVA_{(1-x)}PS_x$ (x = 0.0, 0.25,0.50, 0.75 and 1.0) polymer blend films have been investicated. The changes in the properties of polymer films induced by gamma-irradiation are investicated by using X-ray diffraction, AC electrical conductivity and impedance spectroscopic techniques. The maximum AC conductivity was found to be 1.054×10^{-6} S cm^{-1} at room temperature for PVA polymer film irradiated to 1.5 kGy dose. The conductivity increases from 1.054×10^{-6} S cm^{-1} to 12.582×10^{-6} S cm^{-1} when the temperature is raised to 135°C. The bulk resistance (Rb) has been calculated from the Cole–Cole spectroscopic plot. The magnitude of the bulk resistance decreases with an increase in temperature from 30°C to 110 °C also decreases with increasing γ -ray irradiation dosage.*

Introduction

Radiation grafting and crosslinking were applied, in collaboration with local industry, to prove the possibility of improving the polymer properties. Polymeric film dosimeters are most commonly used as monitors of absorbed dose for routine use in radiation processing of gamma rays and electron beams (Mclaughlin et al. 1988, Abdel-Rehim and Abdel- Fattah 2003). Many dyed poly vinyl alcohol (PVA) films have been developed and investigated for the possibility of their being used to measure neutrons in nuclear reactors and doses of X-rays , γ-rays and electron beams (Chung and Miller 1994). Gamma ray (γ-ray) irradiation of polymer materials is a controlled technique that mainly induces various effects, in view of chemical modification, such as displacing atoms, carbonization, production of free radicals, cross-linking and chain scission that gradually and continuously modify or degrade the structural (Kumar et al. 2003), morphological, optical (Rizk et al. 2008, mechanical and electrical properties of polymer films and blends. Poly vinyl alcohol)/poly(ethylene glycol) (PVA/PEG) copolymer was prepared using casting technique. Structural property studies and transmission of the PVA/PEG samples were performed on both nonirradiated and irradiated samples. The color intensity was greatly increased with increasing the gamma dose and was accompanied by a significant increase in the blue and green color components (Nouh et al. 2012). The effects of ultraviolet (UV) radiation and thermal annealing on the optical and mechanical properties of polyvinyl alcohol (PVA) were studied by El-Kader et al. (2001). A colorless polyvinyl butyral film (PVB) based on radiation-sensitive dye of leucomalachite green (LMG) was investigated as a high-dose dosimeter for gamma radiation processing applications in the dose range of 3-150 kGy (Soliman et al. 2014).

The dielectric properties of blended polymer play an important role in device applications such as high performance electrical cable insulation, capacitors, electronic packaging and components. Impedance spectroscopy is used to determine the frequency dependent electrical properties of a material. Most of the work has been reported on the effects of irradiation on the properties of commercially available Polyvinyl Alcohol (PVA) and Polystyrene (PS) films, but no studies were reported on the effects of gamma irradiation on the properties of $PVA_{(1-x)}PS_x$. It is evident from the literature that the studies on the AC conductivity at different frequency ranges and impedance analysis at different temperature ranges of gamma irradiated $PVA_{(1-x)}PS_x$ films have also not been reported so far.

The prepared $PVA_{(1-x)}PS_x$ (x = 0.0, 0.25,0.50, 0.75 and 1.0) films are irradiated by gamma rays and to study the effect of γ irradiation on dielectric constant, dielectric loss, AC conductivity and impedance spectroscopy behaviour of the polymer blend films at the different temperatures and frequencies.

Experimental

Polyvinyl alcohol (PVA) and polystyrene (PS) monomers (98% with molecular weight less than 3500) were obtained from Sigma Aldrich. The dimethyl sulphoxide (DMSO) and double distilled water (DDW) were used as a solvent. The preparation of $PVA_{(1-x)}PS_x$ films were already reported elsewhere and initial K.Prabha et al) 2015a). The prepared polymer film sheets were cut into small films and subjected to ^{60}Co γ-radiation of two different dosages viz. 1.0 and 1.5 kGy at a dose rate 1.19 Gy per second at room temperature. γ-ray irradiated and unirradiated films were cut into circular pieces with 6.5 mm radius and both surfaces of the film were coated with a good graphite layer. Electrical analysis was made on these circular pieces of the samples using Hioki LCR HiTESTER 3532-50 meter in the frequency range of 50 Hz 5 MHz at 27 °C. The dielectric constant (ε_r) and AC conductivity (σ_{ac}) of the prepared film was calculated by applying the same method described earlier (K.Prabha et al) . For electrical impedance characterization, graphite coated films were kept in an oven at 80 °C for 1 h to achieve perfect contact with the electrode. The impedance measurements (i.e., Z and phase angle) were carried out at an input signal of 1 V with an accuracy of ± 1 % using Hioki LCR HiTESTER model number 3532-50 at three different temperatures, viz. 30, 70 and 110 °C. The temperature was controlled to an accuracy of ± 1 °C(Meena et al 2014).

Results and Discussion

The electrical measurements[(dielectric constant (ε_r), dielectric loss (tan δ) and AC conductivity (σ_{ac})] of $PVA_{(1-x)}PS_x$ (x = 0.0, 0.25,0.50, 0.75 and 1.0) polymer films which were irradiated to different doses (0,1.0 kGy and 1.5 kGy) were done in the frequency range from 50 kHz to 5 MHz and temperature range of 27 – 138 °C. The obtained results were illustrated in Figures 1 - 3. From the figures it reveals that the ε_r increases with the irradiation dosage from, 0, 1 kGy to 1.5 kGy. Also tanδ increases with increasing temperature as well as irradiation dosage. This may be due to formation of charged dangling bonds on the surfaces of polymer when irradiated with γ-ray. The increase of dielectric constant (ε_r) with temperature is due to the enhancement of polarization in the polymer film and likewise as the temperature grows, the chaotic thermal oscillations of molecules are intensified (Sakthivel et al. 1997). The outcome of induced radiation on $PVA_{(1-x)}PS_x$ polymer may increase the disorder of the dipole groups so the dielectric properties increase (Mohamed and Gadon 2000).

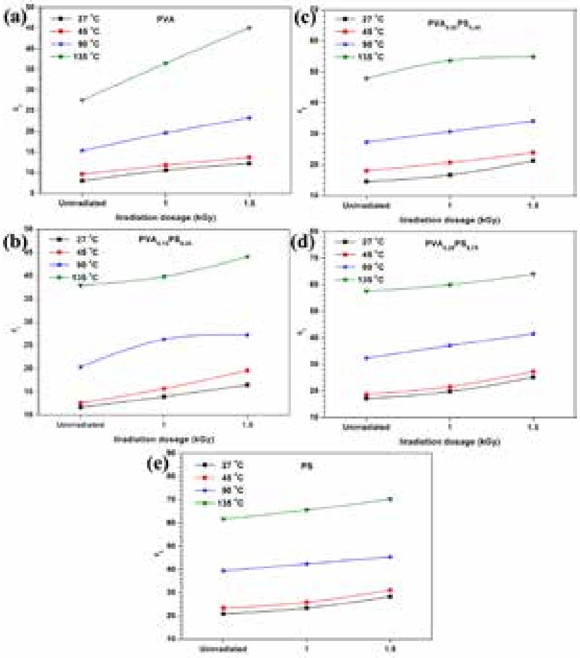

Figure 1: The variation of dielectric constant (ε_r) with irradiation dosages at constant 1 kHz frequency **(a)** PVA, **(b)** $PVA_{0.75}PS_{0.25}$, **(c)** $PVA_{0.50}PS_{0.50}$ **(d)** $PVA_{0.25}PS_{0.75}$, **(e)** PS films at various temperatures.

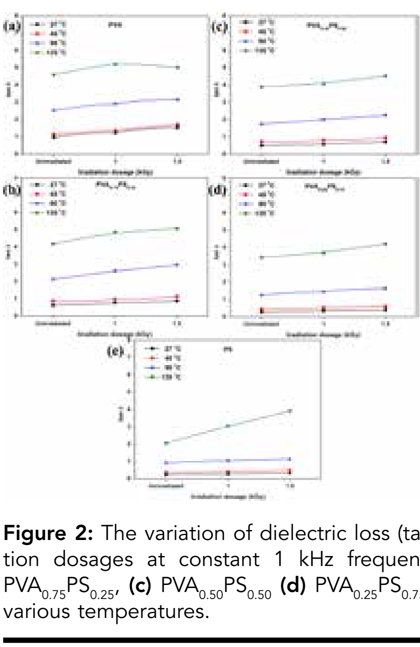

Figure 2: The variation of dielectric loss (tan δ) with irradiation dosages at constant 1 kHz frequency **(a)** PVA, **(b)** $PVA_{0.75}PS_{0.25}$, **(c)** $PVA_{0.50}PS_{0.50}$ **(d)** $PVA_{0.25}PS_{0.75}$, **(e)** PS films at various temperatures.

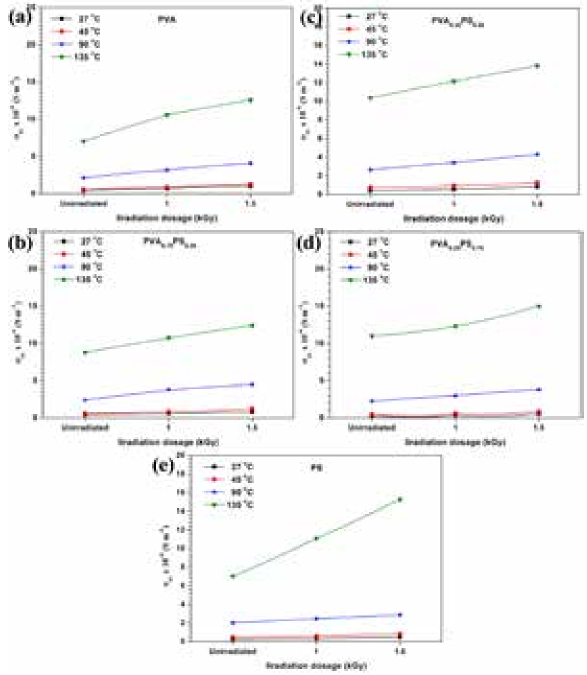

Figure 3: The variation of AC conductivity (σ_{ac}) with irradiation dosages at constant 1 kHz frequency **(a)** PVA, **(b)** $PVA_{0.75}PS_{0.25}$, **(c)** $PVA_{0.50}PS_{0.50}$ **(d)** $PVA_{0.25}PS_{0.75}$, **(e)** PS films at various temperatures.

The impedance spectroscopic studies were carried out for $PVA_{(1-x)}PS_x$. Using these values Col-Col plot is plotted. The Col-Col plots for $PVA_{(1-x)}PS_x$ at different temperatures viz. 30 °C, 70 °C and 110 °C for different irradiation dosages is shown in the Figure 4. The Col-Col plots consist of a high frequency depressed semicircle represented by a frequency dependent capacitor C_g parallel to a bulk resistor (R_b) and a low frequency spike represented by a constant phase element (CPE). In this case the migration of ions may occur through the free volume of $PVA_{(1-x)}PS_x$ matrix polymer which can be interpreted by a resistor. Similar behavior was observed for all the polymer blend at different irradiation dosages. The immobile polymer chains become polarized in the alternating field which can be interpreted by a condenser. The ionic migration and bulk polarization are physically in parallel and therefore the semicircle at high frequency can be observed. The inclined straight line at the low frequency region could be the effect of electrode and electrolyte interface. All the figures show two well defined regions; a high frequency semicircle related to the parallel combination of a resistor and a capacitor and a low frequency spike representing the formation of double layer capacitance at the electrode interface due to migration of ions at low frequency. From these Figures it is evident that all the semicircles almost fit well. The effect of the temperature and irradiation on the impedance behavior of the samples becomes clearly visible from Figures 4(a – e). The bulk resistance (R_b) has been calculated from the Cole–Cole spectroscopic plot. The bulk resistance of the polymer film at different temperatures was obtained by the intersection of a semicircle with the real axis. It was calculated for all the compositions of $PVA_{(1-x)}PS_x$ polymer films. The calculated bulk resistance values at different temperatures (30 °C, 70 °C and 110 °C) and at different radiation dosages are given in Table 1. It can be seen from the Table 1 that the magnitude of bulk resistance decreases with an increase in temperature from 30 °C to 110 °C and also

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decreases with increase of radition dosage. However, there is no systematic variation in bulk resistance with respect to concentration of PS in host $PVA_{(1-x)}PS_x$ polymer matrices

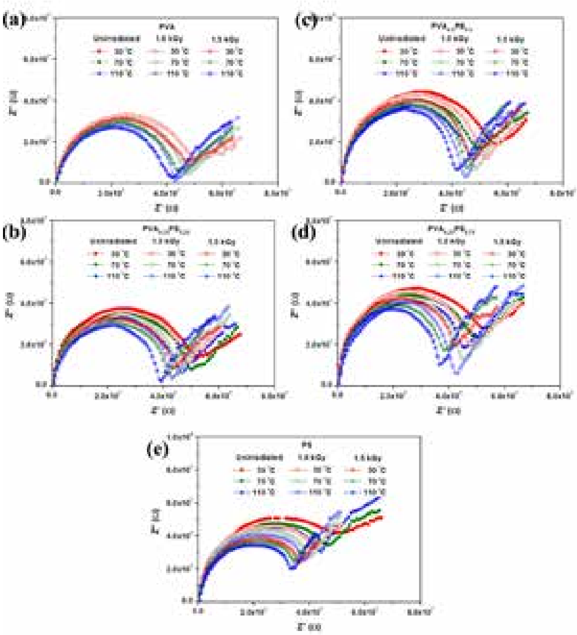

Figure 4: Impedance plots (Cole–Cole) of $PVA_{(1-x)}PS_x$ polymer films with different dosage of irradiation at different temperatures (a) PVA (b) $PVA_{0.75}PS_{0.25}$ (c) $PVA_{0.5}PS_{0.5}$ (d) $PVA_{0.25}PS_{0.75}$ (e) PS

Table 1: Bulk resistance of $PVA_{(1-x)}PS_x$ (x = 0.0, 0.25, 0.50, 0.75 and 1.0) polymer films at different temperature

Sample Name	Irradiation dosage	Bulk resistance (MΩ)		
		30°C	70 °C	110 °C
PVA	Unirradiated	55.14	48.08	43.37
$PVA_{0.75}PS_{0.25}$		58.92	50.87	47.83
$PVA_{0.50}PS_{0.50}$		62.78	54.08	49.67
$PVA_{0.25}PS_{0.75}$		65.59	56.36	50.97
PS		72.56	62.11	57.51
PVA	1.0 kGy	53.11	47.33	43.15
$PVA_{0.75}PS_{0.25}$		49.05	46.90	43.06
$PVA_{0.50}PS_{0.50}$		54.50	49.52	45.13
$PVA_{0.25}PS_{0.75}$		52.76	47.68	43.27
PS		49.58	45.41	41.98
PVA	1.5 kGy	49.05	44.42	41.86
$PVA_{0.75}PS_{0.25}$		44.31	41.78	39.24
$PVA_{0.50}PS_{0.50}$		48.74	46.07	42.35
$PVA_{0.25}PS_{0.75}$		43.81	40.32	38.18
PS		40.39	38.04	36.53

Conclusion

$PVA_{(1-x)}PS_x$ polymer blends has been prepared by solution casting technique using mixed DMSO and water as solvent.Dielectric parameters like delectric constant and loss factors increases with temperature and radiation dose due to formation of charged dangling bonds.Also the impedance studies revealed that bulk resistance of the polymer composites highly depend on the temperature as well as on dose rates. But there is no systematic variation with the composition of $PVA_{(1-x)}PS_x$ polymer films.

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