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Effect of High-Dose Gamma Irradiation on the Dielectric Properties of PVA Polymer Films

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Abstract

In this study, the effect of gamma irradiation on the dielectric properties of poly(vinyl alcohol) (PVA) polymer films were investigated. PVA films were prepared via the solution casting method and irradiated at doses of 250, 500, and 1500 kGy. Dielectric measurements were carried out in the temperature range of 293–373 K. The results revealed that the real part of dielectric permittivity (ε′) decreases with increasing frequency and increases with temperature for all samples. The influence of irradiation was found to be non-linear: ε′ decreased at 250 kGy due to dominant cross-linking, increased at 500 kGy owing to enhanced defect density and chain scission, and exhibited reduced temperature sensitivity at 1500 kGy because of structural stiffening. Electric modulus and AC conductivity analyses confirmed that the relaxation processes are thermally activated and strongly dependent on radiation dose.

Keywords: polymer, gamma radiation, cross-linking, chain scission.
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1. Introduction

Polymers possessing high dielectric properties are widely used in many fields of modern science and technology. Such polymeric materials are extensively applied in insulating systems and energy storage devices [1–3]. One of these polymers is polyvinyl alcohol (PVA). This polymer exhibits several advantageous properties such as easy film-forming ability, high flexibility, and chemical stability. In addition, the

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hydrophilic nature of PVA and its water solubility are highly important in terms of easy processing and reducing fabrication costs [4]. On the other hand, as is well known, this polymer can be prepared in composite form with various fillers and can therefore be utilized in different application areas. Composites based on carbon-derived nanofillers have been widely investigated in recent years [5].

PVA is a semicrystalline polymer. The dielectric properties of PVA are strongly related to the mobility of polymer chains, the number of hydroxyl (–OH) functional groups present in the polymer backbone, and the degree of crystallinity. When an external AC electric field is applied, dipolar polarization dominates within the polymer matrix, while interfacial polarization occurring at crystalline–amorphous boundaries also contribute to the dielectric response. Moreover, polymer chain mobility strongly depends on factors such as temperature and filler incorporation, which in turn significantly affect the dielectric behavior of the polymer [6,7].

Radiation processing is one of the principal techniques used in materials science to modify polymeric materials through exposure to ionizing radiation. Gamma irradiation involves high-energy photons capable of inducing significant changes in the structure and properties of polymer films [8,9]. In general, when polymer matrices are exposed to gamma radiation, both chain scission and cross-linking processes occur simultaneously. Depending on which mechanism dominates, the physical properties of the polymer may vary. Consequently, radiation exposure may lead to an increase or decrease in crystallinity, as well as the formation of defects and recombination centers [10].

Although numerous studies have been conducted on PVA, the effects of high-dose gamma irradiation on this polymer have not yet been fully clarified. In the present study, the dielectric properties of PVA polymer films irradiated at doses of 250, 500, and 1500 kGy were investigated as a function of radiation dose. It was determined that the effect of radiation on the dielectric properties is non-linear. This behavior is associated with the dominance of either chain scission or cross-linking mechanisms depending on the irradiation dose.

2. Materials and Methods

In this study, medium-molecular-weight PVA was used for the preparation of polymer films. The films were fabricated using the solution casting method [11]. First, PVA was dissolved in distilled water to prepare a 5 wt.% solution. The prepared solution was then filtered and dried under ambient conditions at room temperature. The samples obtained were subsequently irradiated with gamma photons at different doses.

Gamma irradiation of the samples was carried out using a ^{60}Co gamma source. The dielectric properties were analyzed using a MINIPIE E7-20 dielectric spectroscope.

3. Results and Discussion

Figure 1 illustrates the frequency dependence of the real part of dielectric permittivity (ε′) for gamma-irradiated PVA polymer films measured in the temperature range of 293–313 K. Figures 1A, 1B, 1C, and 1D correspond to the pristine, 250 kGy, 500 kGy, and 1500 kGy irradiated samples, respectively. As seen from the plots, all samples exhibit a similar dielectric dispersion behavior, where ε′ decreases with increasing frequency. This trend is attributed to the ability of dipolar entities to orient along the direction of the applied electric field at low frequencies. However, as the frequency increases, dipoles become unable to follow the rapidly alternating field, resulting in a reduction in dielectric permittivity. In addition, for all samples investigated, the magnitude of ε′ increases with increasing temperature. This temperature-induced enhancement in dielectric permittivity is associated with thermally activated processes, particularly the increase in segmental mobility of polymer chains at elevated temperatures, which facilitates dipolar orientation and interfacial charge accumulation.

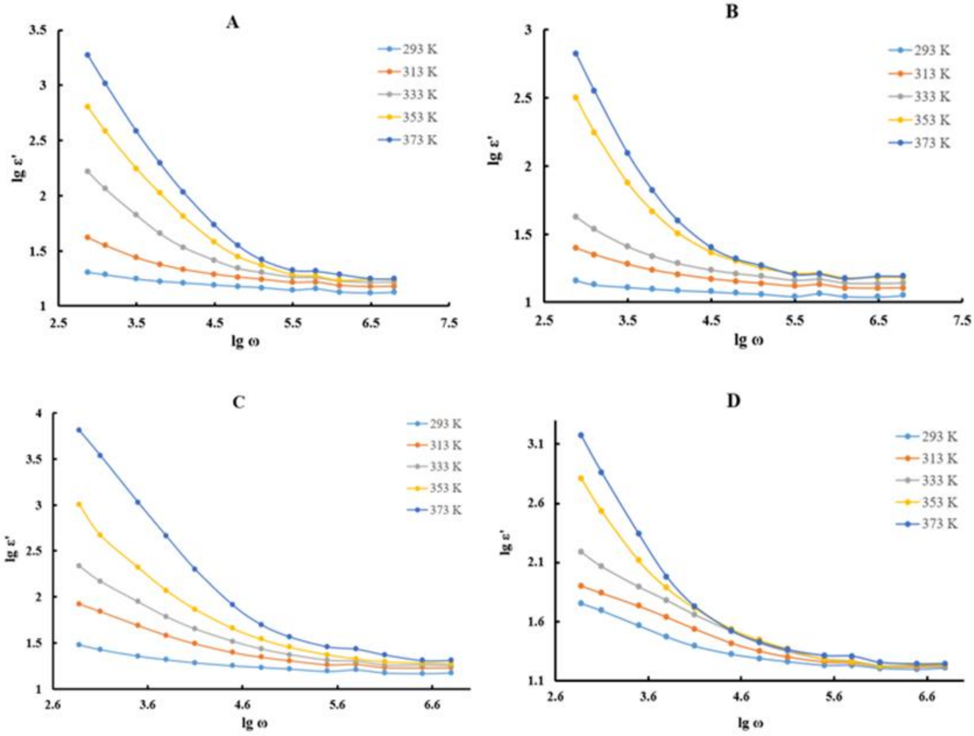

Fig. 1. Frequency dependence of the real part of dielectric permittivity for gamma-irradiated PVA polymer films measured in the temperature range of 293–313 K: (A) pristine PVA, (B) PVA irradiated at 250 kGy, (C) PVA irradiated at 500 kGy, and (D) PVA irradiated at 1500 kGy

The influence of gamma irradiation on dielectric behavior is also clearly evident in Figure 1. Figure 1B shows the ε′–log ω dependence for the sample irradiated at 250 kGy. Compared with the pristine sample (Figure 1A), a reduction in dielectric permittivity is observed. This decrease can be attributed to the predominance of radiation-induced cross-linking at relatively low irradiation doses. The formation of cross-linked networks restricts the segmental motion of polymer chains, thereby limiting dipolar polarization and reducing ε′. Figure 1C presents the frequency dependence of dielectric permittivity for the sample irradiated at 500 kGy. In contrast to the 250 kGy sample, a pronounced increase in ε′ is observed at this irradiation dose. This behavior can be explained by the simultaneous occurrence of cross-linking and chain scission processes under gamma irradiation. Chain scission introduces additional free volume and enhances local chain flexibility. Moreover, the concentration of radiation-induced radicals and structural defects increases significantly. These defects act as charge trapping and accumulation centers, promoting interfacial polarization under an applied electric field, which ultimately leads to an increase in dielectric permittivity. Figure 1D shows the ε′–frequency relationship for the PVA film irradiated at 1500 kGy. At lower temperatures, the dielectric permittivity values are higher than those of the other samples. This enhancement is mainly associated with the high concentration of structural defects and amorphous regions formed at high irradiation doses. The increased density of defect sites and trapping centers facilitates space-charge accumulation, thereby contributing to higher dielectric permittivity.

To determine whether the relaxation processes occur within the bulk volume of the polymer film and to quantitatively evaluate these processes, the electric modulus formalism (M*(ω)) is considered more appropriate [12]. The complex electric modulus is defined as the reciprocal of the complex dielectric permittivity, as expressed in Eq. (1). This approach effectively suppresses the influence of electrode polarization and interfacial charge accumulation at low frequencies, thereby enabling the analysis of relaxation phenomena occurring within the bulk of the material.

$$M^*(\omega) = \frac{1}{\varepsilon^*(\omega)} = M'(\omega) + jM''(\omega), \tag{1}$$

here, ε*(ω) represents the complex dielectric permittivity, while $M'(\omega)$ and $M''(\omega)$M″(ω) denote the real and imaginary parts of the electric modulus, respectively.

The real and imaginary components of the electric modulus can be calculated using Eqs. (2) and (3) [13]:

$$M'(\omega) = \frac{\varepsilon'(\omega)}{(\varepsilon'(\omega))^2 + (\varepsilon''(\omega))^2} \tag{2}$$

$$M''(\omega) = \frac{\varepsilon''(\omega)}{(\varepsilon'(\omega))^2 + (\varepsilon''(\omega))^2}, \tag{3}$$

where $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ are the real and imaginary parts of the dielectric permittivity, respectively.

To evaluate the electrical response of the samples and to analyze the relaxation behavior, it is essential to examine the frequency dependence of the imaginary part of the electric modulus at different temperatures.

Figure 2 presents the variation of M″ as a function of frequency for pristine and gamma-irradiated PVA polymer films. For both unirradiated and irradiated samples, distinct relaxation peaks are observed in the M″–frequency spectra. The position of these peaks shifts toward higher frequencies with increasing temperature. This shift indicates the thermally activated nature of the relaxation processes and reflects a reduction in relaxation time with temperature [14, 15].

The relationship between relaxation time and peak frequency is given by Eq. (4):

$$\tau = \frac{1}{2\pi\nu_{\max}} = \frac{1}{\omega_{\max}}, \tag{4}$$

where τ is the relaxation time and $\nu_{\max}$ is the frequency corresponding to the relaxation peak. As evident from this expression, relaxation time is inversely proportional to the peak frequency.

Figure 2A shows the frequency dependence of M″ for the pristine PVA polymer film. The broad half-width of the relaxation peaks suggests a wide distribution of relaxation times within the sample. However, with increasing temperature, the peak half-width decreases, indicating that the relaxation process becomes more dominant and better defined.

Figures 2B, 2C, and 2D correspond to the M″–log ω plots of PVA films irradiated at doses of 250 kGy, 500 kGy, and 1500 kGy, respectively. With gamma irradiation, an increase in the M″ magnitude is observed, which is associated with radiation-induced defects and cross-linking that modify the relaxation dynamics.

For the sample irradiated at 500 kGy, the relaxation peaks become broader compared to the other doses, indicating a redistribution of relaxation times within the polymer matrix. This behavior may be attributed to the simultaneous occurrence of chain scission and cross-linking processes, leading to increased structural heterogeneity. In contrast, for the sample irradiated at 1500 kGy, the peak half-width narrows again relative to the 500 kGy sample. This suggests partial structural reorganization and an increase in the contribution of dominant relaxation mechanisms. The structural stiffening induced by high-dose irradiation leads to a more constrained but relatively uniform relaxation process.

The temperature dependence of relaxation time generally follows Arrhenius-type behavior and can be described by Eq. (5) [6]:

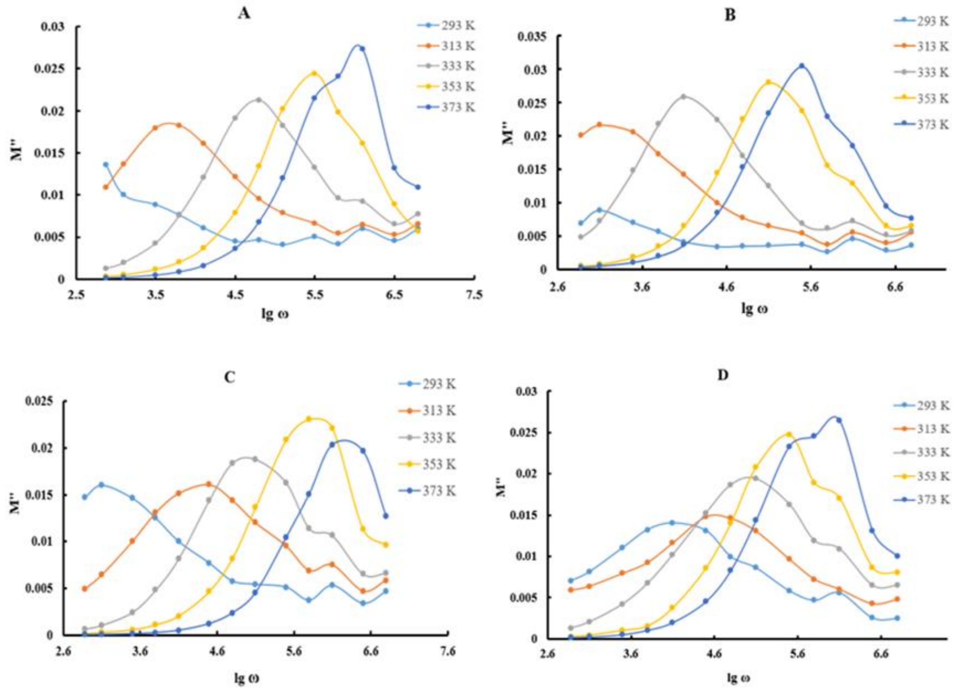

Fig. 2. Frequency dependence of the imaginary part of electric modulus (M”) for pristine and gamma-irradiated PVA polymer films at different temperatures: (A) pristine PVA, (B) PVA irradiated at 250 kGy, (C) PVA irradiated at 500 kGy, and (D) PVA irradiated at 1500 kGy

$$\tau = \tau_0 ex\,p\left(\frac{E_a}{kT}\right), \tag{5}$$

where E_a is the activation energy representing the energy barrier that must be overcome for the relaxation process to occur, k is the Boltzmann constant, and Tis the absolute temperature.

This relation indicates that the relaxation mechanism is governed by a thermally activated process characterized by a specific energy barrier. Based on this expression, the variation of activation energy for PVA polymer films before and after gamma irradiation was evaluated, and the obtained values are summarized in Table1. As seen from Table, the activation energy for pristine PVA was calculated as 0.88±0.04eV. This value is associated with the segmental mobility of polymer chains and the correlated hopping motion of charge carriers within the polymer matrix. Following initial gamma irradiation at a dose of 250 kGy, the activation energy increased significantly to 1.29±0.13eV. This increase can be attributed to radiation-induced cross-linking, as well as an increase in crystallinity and crystallite size. Under these conditions, dipolar motion becomes more restricted, and the hopping probability of charge carriers decreases, resulting in a higher energy barrier for relaxation. However, with further increase in irradiation dose, a decreasing trend in activation energy was observed. For the sample irradiated at 500 kGy, E_adecreased to 0.81±0.09eV. A more pronounced reduction was recorded for the sample irradiated at 1500 kGy, where the activation energy reached 0.54±0.04eV. This reduction is associated with dominant chain scission processes at high irradiation doses. Radiation-induced bond breaking increases defect density, reduces crystallite size, and enhances the amorphous phase fraction. Consequently, the energy barrier that dipoles and charge carriers must overcome for orientation and transport decreases, facilitating relaxation.

To evaluate the linearity of the Arrhenius dependence, the regression coefficient (R^2) was calculated from the lnτ versus 1/T plots. The R^2 values, ranging between 0.96 and 0.99, confirm the strong linearity of the fitted data. These results indicate that the relaxation process is thermally activated and that the Arrhenius model is well applicable to PVA polymer films.

Sample	PVA	PVA- 250 kGy	PVA- 500 kGy	PVA- 1500 kGy
E_a (eV)	0.88±0.04	1.29±0.13	0.81±0.09	0.54±0.04
R^2	0.99	0.97	0.96	0.98

Figure 3 presents the frequency dependence of AC electrical conductivity for gamma-irradiated PVA polymer films measured in the temperature range of 293–373 K.

As observed from the conductivity spectra, the AC conductivity (σ_ac) increases with increasing frequency for all samples. In the low-frequency region, conductivity exhibits a relatively weak dependence on frequency, which is attributed to the localized motion of charge carriers confined within potential wells. As frequency increases, the probability of charge carriers responding to the applied electric field also increases, leading to an enhancement in conductivity [17].

In addition, conductivity increases with temperature for all the samples investigated. This behavior is related to thermally activated charge carriers gaining sufficient energy to escape localized states. The influence of temperature becomes particularly pronounced at higher frequencies, where charge carriers can respond more effectively to the alternating field [17].

Figure 3B shows the conductivity–frequency dependence for the sample irradiated at the initial dose of 250 kGy. Compared to the pristine sample (Figure 3 A), a slight decrease in conductivity is observed. This reduction is attributed to radiation-induced cross-linking, which imposes constraints on polymer chain mobility. The formation of cross-linked structures reduces the hopping probability of charge carriers, thereby lowering effective conductivity. At an irradiation dose of 500 kGy (Figure 3 C), conductivity increases significantly. The increase in radiation dose leads to a higher density of structural defects within the PVA matrix. These defects act as additional hopping sites for charge carriers, facilitating charge transport.

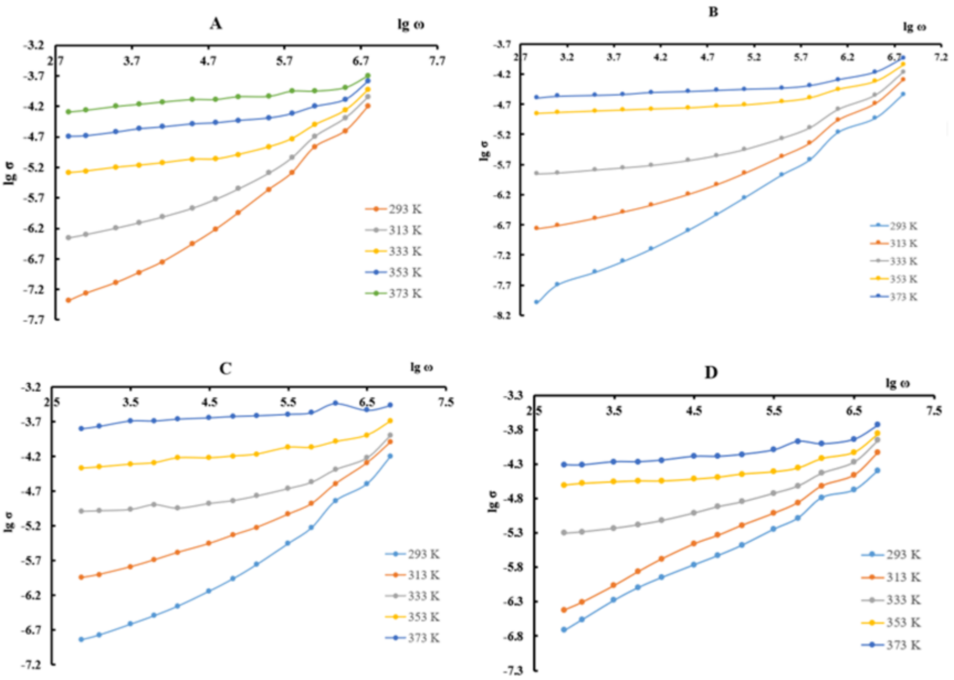

Fig. 3. Frequency dependence of AC electrical conductivity (σ_ac) of pristine and gamma-irradiated PVA polymer films measured in the temperature range of 293–373 K: (A) pristine PVA, (B) PVA irradiated at 250 kGy, (C) PVA irradiated at 500 kGy, and (D) PVA irradiated at 1500 kGy

A further increase in conductivity is observed for the sample irradiated at 1500 kGy (Figure 3). Similar to other samples, conductivity increases with temperature; however, the rate of increase is slower compared to lower irradiation doses. As a result, at elevated temperatures, the conductivity of the 1500 kGy sample becomes lower than that of the 500 kGy sample.This behavior is explained by the formation of both shallow and deep trapping centers at high irradiation doses. While shallow traps assist hopping conduction, deep traps tend to immobilize charge carriers. With increasing temperature, the probability of carrier trapping at deep defect sites increases, thereby limiting long-range charge transport despite thermal activation

3. Conclusion

Gamma irradiation was found to significantly modify the dielectric and electrical properties of PVA polymer films. For all samples, the real part of dielectric permittivity (ε′) decreased with increasing frequency and increased with temperature, confirming dipolar polarization and thermally activated segmental motion. A non-linear dose dependence was observed. At 250 kGy, ε′ and AC conductivity decreased due to dominant cross-linking that restricted polymer chain mobility. At 500 kGy, both ε′ and conductivity increased markedly because of chain scission and increased defect density, which enhanced interfacial polarization and charge carrier hopping. At 1500 kGy, although defect concentration remained high, structural stiffening reduced temperature sensitivity of dielectric response. Electric modulus analysis revealed clear relaxation peaks shifting toward higher frequencies with temperature, indicating bulk, thermally activated relaxation processes. Arrhenius analysis showed that activation energy increased from 0.88 eV (pristine) to 1.29 eV (250 kGy) and then decreased to 0.81 eV (500 kGy) and 0.54 eV (1500 kGy), reflecting the transition from cross-linking-dominated to chain-scission-dominated structural modification. Overall, the study demonstrates that gamma irradiation effectively tunes dielectric permittivity, relaxation dynamics, activation energy, and charge transport mechanisms in PVA films, highlighting their potential for dielectric insulation and energy storage applications.

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