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# Study the Frequency Dependence of Dielectric Properties of Gamma Irradiated $PVA_{(1-x)}PS_x$ Polymer Blends

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## Abstract

**$PVA_{(1-x)}PS_x$ (x = 0.0, 0.25, 0.50, 0.75 and 1.0) polymer blend films were prepared by simple solution cast method using mixer of dimethyl sulphoxide (DMSO) and double distilled water (DDW) as a common solvent. Gamma irradiation studies have been carried out to investigate the potential for improvements in the electrical properties of pure and blend polymer film. The induced changes in the dielectric constant ($\varepsilon_r$), dielectric loss ($\tan\delta$) and AC conductivity ($\sigma_{ac}$) properties for the unirradiated and irradiated films were studied, in the frequency range 50 Hz - 5 MHz at room temperature (27˚C). Further, it was seen that the frequency dependent dielectric constant, dielectric loss and AC conductivity were found to increase with increasing $\gamma$ irradiation dose for all the $PVA_{(1-x)}PS_x$ (x = 0.0, 0.25, 0.50, 0.75 and 1.0) polymer blends. The maximum $\sigma_{ac}$ at 1 kHz frequency was observed to be $2.4421 \times 10^{-6}$ s/m (at room temperature) for PVA film irradiated at 1.5 kGy.**

## Keywords

**Polymer Composite Films, Gamma Irradiation, Dielectric Properties**

## 1. Introduction

In recent years the effects of gamma radiation on polymers have been investigated by a number of researchers [1]-[4]. Gamma ray ($\gamma$-ray) irradiation becomes an advanced approach to optimize the physical properties of polymer materials such as structural, thermal, optical, dielectric and electrical properties. The property modification depends upon the irradiation dosage and material characteristics. $\gamma$ radiation is reported to alter the structural pattern within the polymer matrix through the formation of new functional groups, rearrangements of linking-

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bonds, and scission and/or cross-linking of chains, etc. [5]-[7]. Among them, the major governing effects observed to alter the structural pattern in polymer films are scissions and cross-linking or joining of chains by formation of bonds [8] [9]. The scissions and cross-links of the polymers are more easily determined due to its nature. Hence, the polymers have been catalogued as to whether they undergo one or the other of the two processes [10]. The radiation not only alters the structure of the polymer, but it can also produce significant changes in the thermal, optical dielectric and electrical properties of irradiated polymer films [11] [12]. When the polymer films are irradiated with $\gamma$-rays, the radiation enhances the presence of trapped charges and to create defects in the polymer matrix. Thus the electronic transport in polymer films is highly affected by ionizing gamma radiation.

The physical and chemical properties of unirradiated and $\gamma$-ray irradiated blends based on miscible polymers have been extensively investigated [13]-[18]. The structural, optical and electrical properties of the polymers are drastically affected by the presence of another polymer. Polyvinyl Alcohol (PVA) and Polystyrene (PS) are the important polymers and are available in the form of powders, fibers and films. PVA and PS are non-toxic and water soluble polymers which have good film forming and high hydrophilic properties. PVA is a semicrystalline whereas PS is an amorphous in nature. They are a unique pair of polymers and they are chemically different; however, they display compatibility over different composition ranges at room temperature. Interest in such studies has gained significance of potential applications in integrated electronics, phosphors, surface modifications and used industrially for sizing, adhesives and emulsification.

The literature review revealed that very little work had been reported on $\gamma$-ray irradiated induced effect on structural, morphology [19], thermal, photo [20], dielectric and AC conductivity of PVA and PS polymer films. However, to the best of our knowledge, no reports are available on optical and electrical properties of gamma irradiated $PVA_{(1-x)}PS_x$. The detailed structural analysis of unirradiated and $\gamma$-ray irradiated PVA, PS films and $PVA_{0.5}PS_{0.5}$ blind film have already been reported elsewhere [21]. Thus, in the present investigation we study the frequency dependence of dielectric properties of gamma irradiated $PVA_{(1-x)}PS_x$ (x = 0.0, 0.25, 0.50, 0.75 and 1.0) films.

## 2. Experimental Analysis

### 2.1. Materials

Polyvinyl alcohol (PVA) and polystyrene (PS) monomers with 99.9% purity were obtained from Sigma Aldrich and used without any purification. The dimethyl sulphoxide (DMSO) and double distilled water (DDW) were used as a solvent.

### 2.2. Preparation of PVA-PS Blend

For the casting of typical $PVA_{(1-x)}PS_x$ (x = 0.25) blend film, 7.5 wt. % of pure PVA and 2.5 wt.% of PS (3:1) were dissolved separately in 7 ml of DDW-DMSO mixture (1:1) and stirred continuously until complete miscibility at room temperature. Then the PS solution was slowly added to the PVA solution and continuously stirred for 1 h at 60˚C. The homogenous mixer of hydrogel was transferred into a petri dish and allowed it to cool to room temperature. In order to obtain smooth film the petri dish was placed under ambient conditions in a fume hood over a week and then the film was carefully peeled off from the petri dish. The prepared polymer film was then dried at 70˚C for about 2 h in a vacuum oven to remove the residual solvent. In order to prepare PVA, $PVA_{0.5}PS_{0.5}$, $PVA_{0.25}PS_{0.75}$ and PS films, the ratio of wt. % of PVA:PS was taken respectively, 1:0, 1:1, 1:3 and 0:1 and the same procedure was adopted. The thickness of the films was controlled by the volume of a polymer solution in the petri dish. The measured average thickness of the $PVA_{(1-x)}PS_x$ (x = 0.0, 0.25, 0.50, 0.75 and 1.0) films were given in **Table 1**.

### 2.3. Characterization

The prepared $PVA_{(1-x)}PS_x$ (x = 0.0, 0.25, 0.50, 0.75 and 1.0) film sheets were cut into small films with dimensions 5 cm × 5 cm and subjected to $^{60}Co$ $\gamma$-rays with two different dosages viz. 1.0 and 1.5 kGy at a dose rate 1.19 $Gy \cdot s^{-1}$ at room temperature. The thickness of the films was measured before and after $\gamma$ irradiation at 5 randomly selected places with Mprobe reflectrometer. $\gamma$-ray irradiated and unirradiated films were cut into circular disc with 6.5 cm radius and both surfaces of the film were coated with graphite layer. Electrical analysis

**Table 1.** The average thickness of the prepared $PVA_{(1-x)}PS_x$ (x = 0.0, 0.25, 0.50, 0.75 and 1.0) films (before $\gamma$-ray irradiation).

| Sample name | Average thickness |
|---|---|
| | μm |
| PVA | 0.435 |
| $PVA_{0.75}PS_{0.25}$ | 0.515 |
| $PVA_{0.5}PS_{0.5}$ | 0.580 |
| $PVA_{0.25}PS_{0.75}$ | 0.605 |
| PS | 0.795 |

was made on these circular pieces of the samples using Hioki LCR HiTESTER 3532-50 meter in the frequency range of 50 - 5 MHz at room temperature (27˚C). The dielectric constant ($\varepsilon_r$) and AC conductivity ($\sigma_{ac}$) of the prepared film was calculated by using the following relations.

$$\varepsilon_r = \frac{C_{film}}{C_{air}}, \tag{1}$$

$$\sigma_{ac} = \varepsilon_o \varepsilon_r \omega \tan\delta, \tag{2}$$

where $C_{film}$ and $C_{air}$ is the capacitance of the film and air capacitance respectively [22]. $\varepsilon_o$ is the permittivity of free space ($8.854 \times 10^{-12}$ F/m), $\omega$ is the angular frequency ($\omega = 2\pi f$; f = 50 Hz - 5 MHz in the present study) and $\tan\delta$ is the measured dielectric loss of the sample.

## 3. Results and Discussion

The $\gamma$-ray irradiation can modify the dielectric properties of $PVA_{(1-x)}PS_x$ (x = 0.0, 0.25, 0.50, 0.75 and 1.0) films. **Figures 1-3** illustrate the change in dielectric constants ($\varepsilon_r$), dielectric loss ($\tan\delta$) and AC conductivity ($\sigma_{ac}$) for the unirradiated and irradiated samples of different doses (1.0 kGy and 1.5 kGy) within the frequency range 50 Hz - 5 MHz at room temperature as well as, at different temperature range from 27˚C - 138˚C at constant 1 kHz frequency.

From **Figure 1**, it was observed that the dielectric constant increases with the increasing temperature for all the pure and different concentration of PS in $PVA_{(1-x)}PS_x$ blends. Also the dielectric constant increases with the increase of radiation dosage from 1.0 kGy to 1.5 kGy. For example, at 27˚C (room temperature) and at 1 kHz frequency, the dielectric constant of the pure PVA, $PVA_{0.75}PS_{0.25}$, $PVA_{0.5}PS_{0.5}$, $PVA_{0.25}PS_{0.75}$ and PS (unirradiated films) is 8.0329, 11.6976, 14.6452, 17.1556 and 20.9068, respectively. Whereas for the $\gamma$ irradiated films (1.5 kGy) it is 12.2268, 16.5231, 21.2954, 25.1157 and 28.3029, respectively. The variation in $\varepsilon_r$ for the unirradiated $PVA_{(1-x)}PS_x$ films showed a maximum value at lower values of frequencies and then decreases rapidly up to 10 kHz for all doses as the frequency increases. Then, it remains almost constant up to 5 MHz. The increasing $\varepsilon_r$ value with the $\gamma$-ray irradiation dosage is attributed to the formation of few defects sites in the band gaps of $PVA_{(1-x)}PS_x$ films as a result of occurrence of chain scisson and rearrangements of linking-bonds. These defects taps the charge carriers exist in the band gap of the polymer/polymer blend films. Thus the $\gamma$-ray irradiation increases the ability of the $PVA_{(1-x)}PS_x$ polymer/blend films to store charges. In both the unirradiated and irradiated $PVA_{(1-x)}PS_x$ films, $\varepsilon_r$ decreases with the increasing frequency. The reduction in value of the dielectric constant with the gain in frequency can be excused by the fact that at low frequency, the electric dipoles can follow the electric field, but with an increase in frequency, the electric dipoles can no longer follow the electric field, resulting in a reduction in the value of the dielectric constant [23]. It is recognized that the first appearance of a dielectric into an electric field causes its polarization. Polystyrene is a solid amorphous dielectric material containing no polar groups. In such polymers, the main kind of polarization is electronic polarization caused by deformation of the electronic shells and appearance of induced dipoles. The contribution of the other kinds of polarization like ionic, space charge and orientation is negligibly low because of the absence of free ions and polar molecules in the amorphous PS dielectric material. Hence the addition of PS in $PVA_{(1-x)}PS_x$ (x = 0.25, 0.50, 0.75) blend films, induced dipoles are arises and enhance the $\varepsilon_r$.

The variation of the dielectric loss ($\tan\delta$) of $PVA_{(1-x)}PS_x$ films (unirradiated and irradiated) as a function of

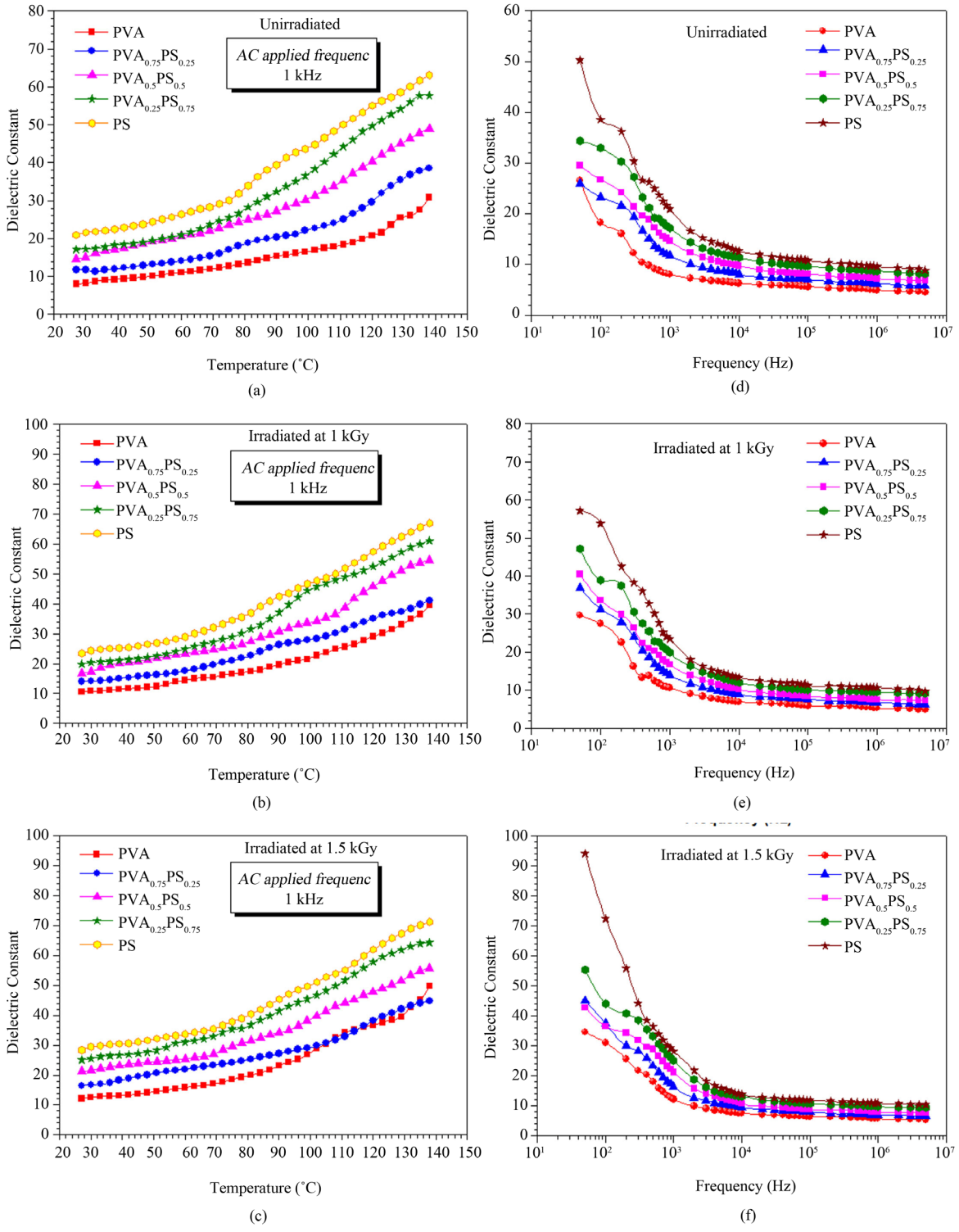


**Figure 1.** The variation of dielectric constant ($\varepsilon_r$) with temperature (a) unirradiated, (b) irradiated at 1 kGy, (c) irradiated at 1.5 kGy and the variation of $\varepsilon_r$ with frequency (at 27˚C) (d) unirradiated, (e) irradiated at 1 kGy, (f) irradiated at 1.5 kGy of $PVA_{(1-x)}PS_x$ (x = 0.0, 0.25, 0.50, 0.75 and 1.0) films.

temperature at constant 1 kHz frequency and as a function of frequency at constant 27˚C is shown in **Figure 2**. From the **Figure 2**, it reveals that the tan$\delta$ increases with increasing temperature whereas decreases with increasing

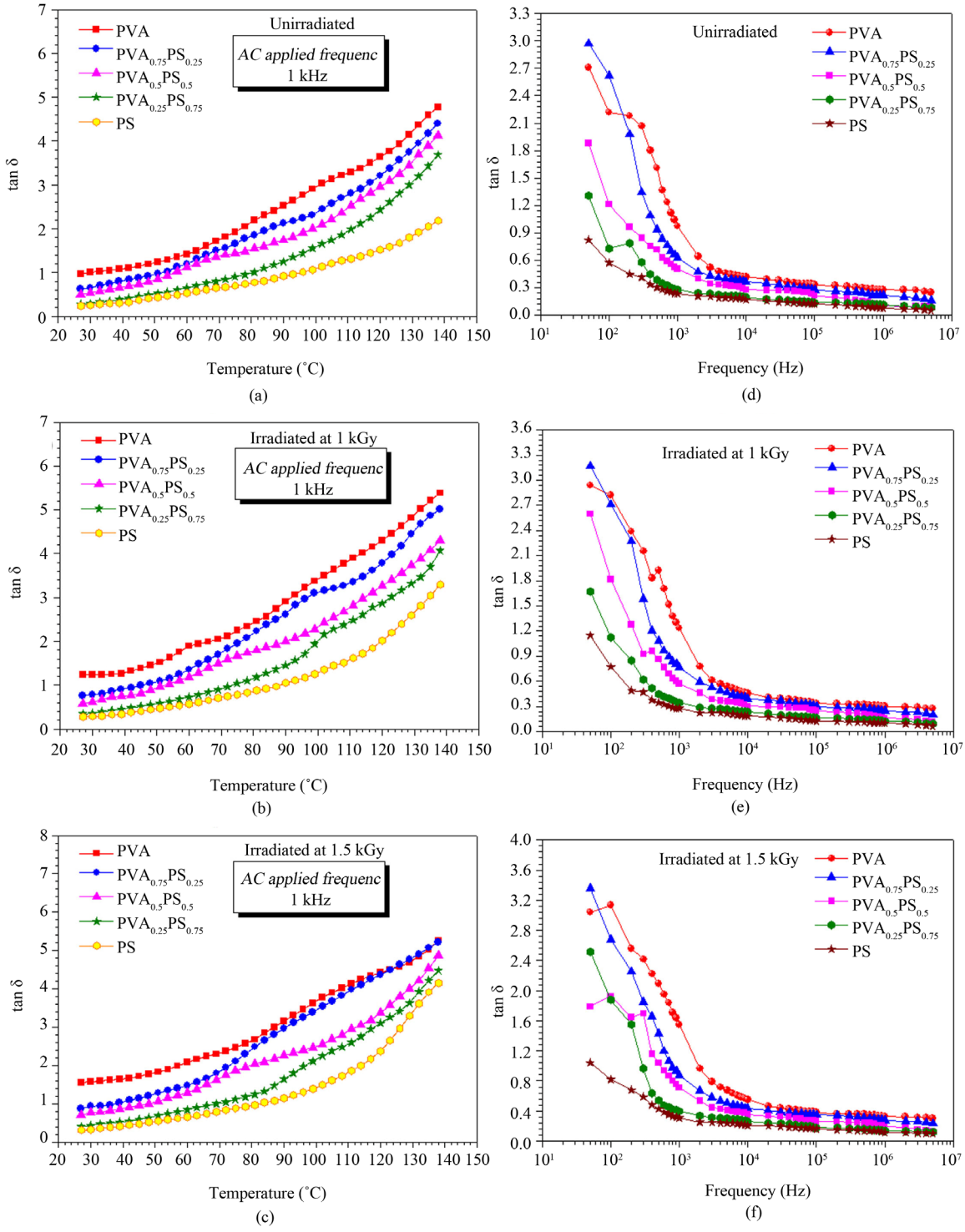


**Figure 2.** The variation of dielectric loss (tan$\delta$) with temperature (a) unirradiated, (b) irradiated at 1 kGy, (c) irradiated at 1.5 kGy and the variation of tan $\delta$ with frequency (at 27˚C) (d) unirradiated, (e) irradiated at 1 kGy, (f) irradiated at 1.5 kGy of $PVA_{(1-x)}PS_x$ (x = 0.0, 0.25, 0.50, 0.75 and 1.0) films.

frequency. Also, as the concentration of PS increases in $PVA_{(1-x)}PS_x$ blends the tan $\delta$ decreases in both temperatures and frequencies. However, the tan $\delta$ increases with increasing irradiation dosage when compared to

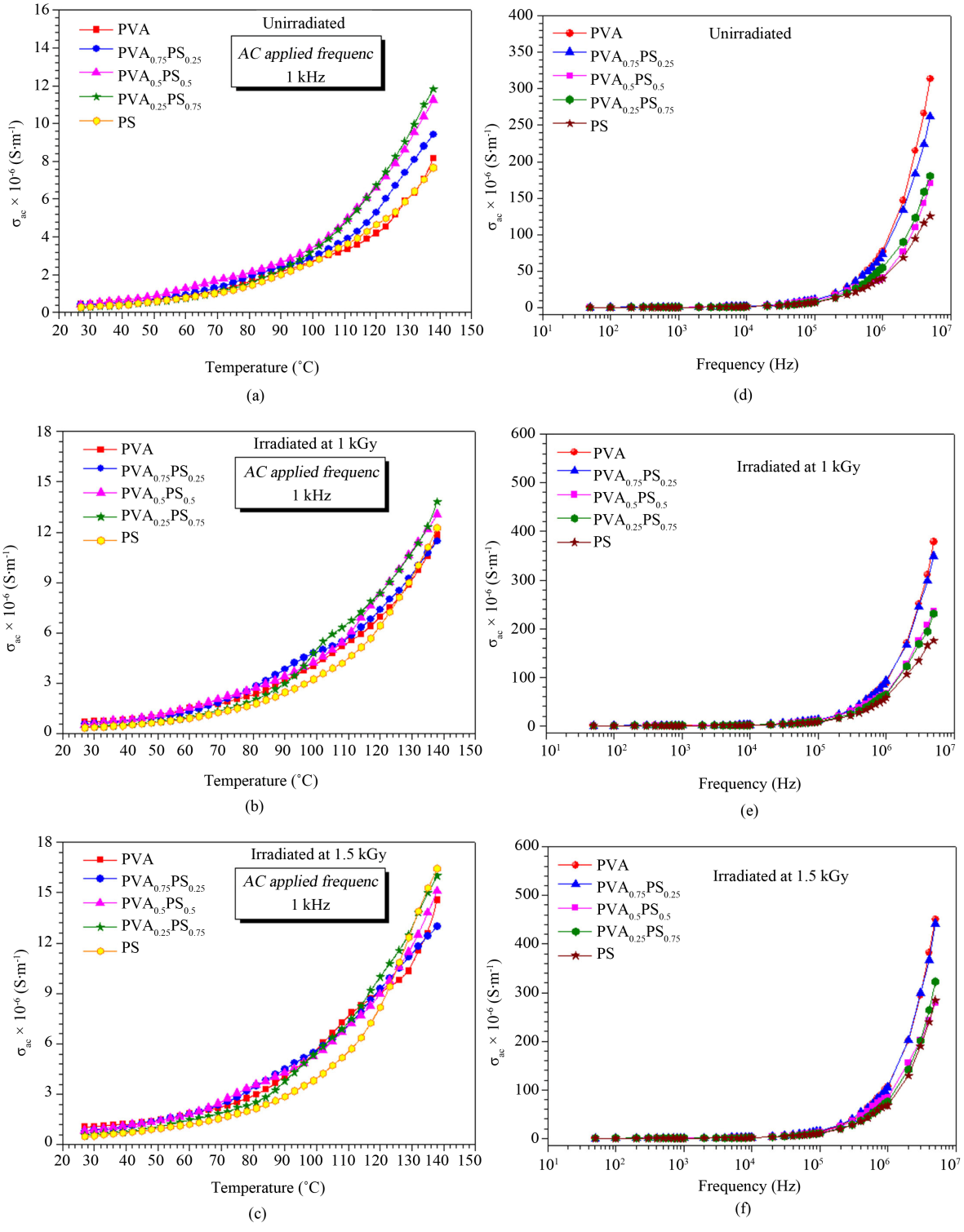


**Figure 3.** The variation of AC conductivity ($\sigma_{ac}$) with temperature (a) unirradiated, (b) irradiated at 1 kGy, (c) irradiated at 1.5 kGy and the variation of $\sigma_{ac}$ with frequency (at 27˚C) (d) unirradiated, (e) irradiated at 1 kGy, (f) irradiated at 1.5 kGy of $PVA_{(1-x)}PS_x$ (x = 0.0, 0.25, 0.50, 0.75 and 1.0) films.

unirradiated films.

The variation of AC conductivity ($\sigma_{ac}$) with the temperature at 1 kHz frequency and with frequency at room

temperature for pure PVA/PS and blended ($PVA_{(1-x)}PS_x$) films of unirradiated and irradiated is represented in **Figure 3**. From the figure, it is clear that the $\sigma_{ac}$ increases as the temperature, frequency and irradiation dose increases whereas decreases with the increasing concentration of PS from x = 0.0 to x = 1.0. It can be noticed that the $\sigma_{ac}$ for the $PVA_{(1-x)}PS_x$ films have a minimum value of low frequency range up to 10 kHz and then increases rapidly while increasing frequency. The $\sigma_{ac}$ of the polymer film increases with frequency which is the common characteristic of disordered materials. Furthermore, almost all the prepared $PVA_{(1-x)}PS_x$ films show similar behaviour up to 10 kHz, which is typical for hopping conduction. In this range there is not much change in AC conductivity with frequency. However, $\sigma_{ac}$ slightly increases with increasing frequency within this frequency range. Thereafter an increase in frequency leads to rapid increase in AC conductivity.

When the films are irradiated by $\gamma$-rays, ions and free radicals are formed and partially trapped in the bulk of the material [24]. Thus the conductivity of the irradiated film showed higher conductivity than that of unirradiated film for both temperature and frequency dependent conditions. Similar behavior was observed for all the samples (x = 0.25, 0.50, 0.75 and 1). The electrical conductivity of the polymer material depends on the presence of free ions connected chemically with macromolecules. The molecular chain of the polymer does not participate in the transfer of electrical charge [25]. The increase in AC conductivity with temperature for the prepared $PVA_{(1-x)}PS_x$ films may be accounted for the liberation of charge carriers through the amorphous region. Also, it can be observed that the increase in AC conductivity with increase in irradiation dose is ascribed to the degradation of the polymer chains in $PVA_{(1-x)}PS_x$ films. The irradiation dosage (1.5 kGy) increases the $\sigma_{ac}$ by more than an order of magnitude for all the samples considered in the present study.

## 4. Conclusion

$PVA_{(1-x)}PS_x$ polymer blends were successfully prepared by using simple solution cast method. The changes in the electrical properties of the pure and polymer blend films after exposing to different dosage (1.0 and 1.5 kGy) of $\gamma$-ray irradiation have been investigated. The variation in $\varepsilon_r$ for the unirradiated $PVA_{(1-x)}PS_x$ films showed a maximum value at lower values of frequencies and then decreased rapidly up to 10 kHz for all doses as the frequency increased. The irradiation dosage (1.5 kGy) increased the $\sigma_{ac}$ by more than an order of magnitude for all the films.

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