Preparation and Characterization of Poly(vinyl alcohol) and Gelatin Blend Films

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ABSTRACT: Poly(vinyl alcohol) (PVA) is a water-soluble polymer that has been studied intensively because of several interesting physical properties that are useful in technical applications, including biochemical and medical applications. In this article, we report the effects of the addition of gelatin on the optical, microstructural, thermal, and electrical properties of PVA. Pure and PVA/gelatin blend films were prepared with the solution-casting method. These films were further investigated with Fourier transform infrared (FTIR), differential scanning calorimetry (DSC), X-ray diffraction (XRD), ultraviolet–visible (UV–vis) spectroscopy, and dielectric measurements. The FTIR spectrum shows a strong chemical interaction between PVA and gelatin molecules with the formation of new peaks. These peaks are due to the presence of gelatin in the blend

INTRODUCTION

Polymeric materials have attracted scientific and technological researchers because of their widespread applications. This is mainly due to the light weight, good mechanical strength, and optical properties, which make them multifunctional materials. Moreover, these polymers are traditionally considered excellent host materials for blending.¹ In recent years, polymer blends and composites have been subjects of interest for both theoretical and experimental studies because of certain physical and chemical properties needed for specific applications that may be obtained by blending. It has been observed that polymer blending has a significant effect on the physical properties, including optical, thermal, and electrical properties. These changes in physical properties depend on the chemical nature of the guest material and the way in which it interacts with the host polymer. Systematic investigations reported in the literature show that many polymers have two coexistent phases, crystalline and amorphous. When such polymers are blended with a suitable material,

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films. The DSC results indicate that the addition of gelatin to PVA changes the thermal behavior, such as the melting temperature of PVA, and this shows that the blends are compatible with each other. This also shows that the interaction of gelatin and PVA molecules changes the crystallite parameters and the degree of crystallinity, and this supports the XRD results. The UV–vis optical study also reflects the formation of the complex and its effect on the microstructure of the blend film. Moreover, the addition of gelatin also gives rise to changes in the electrical properties of PVA/gelatin blend films. © 2008 Wiley Periodicals, Inc. J Appl Polym Sci 109: 1328– 1337, 2008

Key words: blends; dielectric properties; films; FTIR

they may interact either in the amorphous fraction or in the crystalline fraction of the polymers, and in both cases, the polymeric properties will be altered. Hence, complete information about the effects of additives on a specific polymer helps in tailoring that polymer for a particular application.^{2,3}

Poly(vinyl alcohol) (PVA) has been used for more than 40 years because of its unique chemical and physical properties.⁴ These properties come from its hydroxyl groups. The hydrogen bonding between hydroxyl groups plays an important role in the properties of PVA, such as its high water solubility, wide crystallinity range, and high crystal modulus.⁵ The important features of semicrystalline PVA are the presence of crystalline and amorphous regions and its physical properties, which result from the crystal-amorphous interfacial effect. These two regions are well separated by portions of an intermediate degree of ordering, and this enhances the macromolecules, producing several crystalline and amorphous phases.² PVA is normally a poor electrical conductor; it can become conductive upon blending with some polymer. The poor conducting nature of PVA is thought to be due to the high physical interactions between polymer chains via hydrogen bonding with hydroxyl groups as well as the complex formation.^{2,3,6-9} Optical absorption spectroscopy [ultraviolet-visible (UV-vis) and infrared] is an

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established tool for investigating the effect of blending on the microstructure of a polymer, particularly on the band structure and electronic properties. Thermal analysis is another useful method for defining suitable processing conditions for polymers. Differential scanning calorimetry (DSC) is a major tool for studying the crystallization kinetics. Thermal stability includes the glass transition and melting temperature of polymers. The thermal treatment or thermal history of a semicrystalline polymer will be reflected in its melting behavior. For many semicrystalline polymers, double and multiple melting peaks have been observed in DSC scans, and various interpretations have been proposed for the origin of multiple melting peaks. Many authors have explained this double melting behavior with the melt-crystallization model. This model suggest that the low-temperature and high-temperature peaks in the DSC melting curve can be attributed to the melting of some amount of original crystal and to the melting of crystals formed through the melt-crystallization process during a heating scan, respectively. The melting proceeds through the melting of the original crystal, recrystallization, and melting of recrystallized and perfected crystal.^{10–12}

Gelatin is a well-known material widely used in biomedical applications.^{13–16} There are various studies on the compatibility of gelatin-based blends and composite films, but still the compatibility of PVA/ gelatin blend films is an important parameter from the point of view of the behavior of the two polymers in the blend. In view of this, it is very important to note that the blending modifies the structure of the polymer and hence its properties because the changes in the polymer properties mainly depend on both the polymers and the way in which they interact with each other. In this investigation, the effects of the addition of gelatin on the microstructure and optical, thermal, and electrical properties of PVA were studied with Fourier transform infrared (FTIR), UV-vis, X-ray diffraction (XRD), DSC, and dielectric measurements.

EXPERIMENTAL

Materials and sample preparation

PVA used in this work was obtained in a powder form from S.D. Fine Chemicals, Ltd. (Mumbai, India), with a molecular weight of 125,000 g/mol and degree of saponification of 86–89%. The gelatin used in this work was purchased from Rankem Ranbaxy Fine Chemicals, Ltd.(Mumbai, India) and used without further purification.

PVA and PVA/gelatin blend films with different concentrations of gelatin (2, 5, 7, 10, and 15% w/w) were prepared by the solution-casting method. A known quantity of PVA powder was added to dou-

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bly distilled water and kept at 40°C with constant stirring to get complete dissolution. The required quantity of gelatin was also dissolved in distilled water and added to the polymeric solution with continuous stirring. This solution was set aside to get a suitable refractive index. The solution so obtained was poured onto a cleaned glass Petri dish and kept in an oven at 60°C for slow evaporation of the solvent for a few hours. The water was evaporated completely, and thin films of PVA and PVA/gelatin blends remained on the Petri dish. These films were peeled from the plate and kept in vacuum desiccators for further study.

Characterization

The refractive index for PVA/gelatin blend solutions of different compositions was measured with an Abbe refractometer (Milton Roy Company, Ivyland, PA).

FTIR spectroscopy for the pure PVA and PVA/ gelatin blend films prepared by the solution-casting technique was performed with an ATR–FTIR spectrophotometer (Paragon 500, PerkinElmer, Beaconsfield, Buckinghamshire, UK) in the wave-number range of 400–4000 cm⁻¹ with a resolution of 4 cm⁻¹. The FTIR spectrum was taken in the transmittance mode.

The thermal analysis was performed with a Perkin-Elmer DSC-7 differential scanning calorimeter at a heating rate of 10°C/min under a nitrogen atmosphere in the temperature range of 50–350°C.

The X-ray diffractograms of the samples were recorded with a Bruker (Rigaku Corporation, Tokyo, Japan) D8 advance X-ray diffractometer with Ni-filtered Cu K α radiation (wavelength = 1.54060 Å) with a graphite monochromator. The scan was taken in the 2 θ range of 5–60° with a scanning speed and step size of 1°/mm and 0.01°, respectively.

According to the method suggested by Manjunath et al.¹⁷ for any polymer, the resolution of the peak (R) is given by

$$R = (m_1 + 2m_2 + \dots + m_{n-1})/(h_1 + h_2 + \dots + h_n) \quad (1)$$

where m_1 , m_2 , and so on are the heights of the minima between two peaks and h_1 , h_2 , and so on are the heights of the peaks from the base line.

The factor 1 - R gives the lateral order or index of crystallinity, and the crystallinity percentage is given by

Crystallinity (%) =
$$(1 - R) \times 100$$
 (2)

UV–vis spectra were obtained with a PerkinElmer Lambda/25/35/45 UV–vis spectrophotometer in the range of 190–700 nm.



Figure 1 Refractive-index variations for PVA/gelatin blends of different compositions.

Dielectric properties were measured under normal temperature and pressure (NTP) conditions with Agilent 4282A precession inductance, caparitance and resistance (LCR) meter (Agilent Technologies Ltd., Santa Clara, CA) within the frequency range of 20 Hz to 1 MHz and with a direct-current (DC) bias potential varying from 0 to 40 V. The samples were tested only as a function of frequency. The guard electrode contact method was adopted. The diameter of the films was around 40 mm. The films were silver-coated from both sides to achieve better electrode contact.¹⁸ The relative dielectric constant was confirmed by the measurement of the out-of-plane capacitance with the following formula: $C_p = \varepsilon \times C_0$, where C_p is the parallel capacitance of the dielectric that is placed between two electrodes and C_0 is the parallel plate capacitance of two electrodes without dielectric material. If the dielectric completely fills the space between the plates, the capacitance increases by the dimensionless factor ε , which is the dielectric constant.¹⁹ The specialties of this impedance analyzer were inbuilt DC bias potential variation facilities, which enabled us to apply simultaneous DC potential along with alternating-current (AC) signal for measuring the overall electrical parameters. The fixture assembly attached to this instrument was designed to take observations under NTP conditions only.

RESULTS AND DISCUSSION

Refractive index

The refractive index is a suitable state parameter directly correlated to the material density. The refractive-index values in a polymer are around 1.3– 1.6, which are almost comparable to those of silica glass. Figure 1 shows the variation of the refractive index with the gelatin concentration in PVA/gelatin blend films. The refractive index of PVA/gelatin blends decreases with an increase in the amount

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of gelatin in the blends. The refractive-index measurements can have a correlation with the electrical properties of blend films. Hence, there was a need to measure the refractive index for the different weight percentages of the crosslinking agent. The obtained results demonstrate the capability of a sensor system such as PVA to monitor the cure kinetics of polymeric materials.

Infrared spectroscopy

The FTIR spectra of pure PVA and PVA/gelatin blend films were obtained, and the results are shown in Figure 2. In FTIR spectra, the bands at 916 and 850 cm⁻¹ confirm the presence of a skeletal vibration of PVA. The band corresponding to the C—H asymmetric stretching vibration occurs at 2928 cm⁻¹, and the band at 1733 cm⁻¹ corresponds to the C=O stretching vibration of the carbonyl group; the band at 1640 cm⁻¹ corresponds to an acetyl C=O group of PVA. The C—C and C—O stretching vibration band was observed and confirmed at 1144 cm⁻¹. The band for C—O stretching and O—H bending



Figure 2 FTIR spectra for PVA/gelatin blend films of different compositions: (a) 100/0, (b) 98/2, (c) 95/5, (d) 93/7, and (e) 85/15.



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Figure 3 DSC thermograms for PVA/gelatin blend films of different compositions: (a) 100/0, (b) 95/5, (c) 90/10, (d) 85/15, and (e) 0/100.

was observed at 1096 cm⁻¹. The band at 1089 cm⁻¹ corresponds to the C–O stretching vibration of the acetyl group present on the PVA backbone. The corresponding bending and wagging of CH₂ vibrations are at 1438 and 1378 cm⁻¹, respectively, and the band at 1247 cm⁻¹ corresponds to C–H wagging.^{9,20,21} The peak at 3348.6 cm⁻¹ corresponds to hydroxyl stretching vibrations of PVA, and the peak at 1331.2 cm⁻¹ represents the stretching mode of the carboxylic group.²²

In the case of PVA/gelatin blends, the FTIR spectrum shows shifts in the bands with changes in the intensities. The new peaks were obtained at 1508.3, 1794.0, 1870.3, and 3676.3 cm⁻¹. These peaks are due to the presence of gelatin in the blends. This indicates the considerable interaction between PVA and gelatin molecules.

DSC

Figure 3 shows the DSC thermograms for PVA/gelatin blends of different compositions. In the DSC curves, the endothermic peaks corresponding to the melting of PVA can be seen at 220–230°C. However, the melting peak corresponding to the phase change

of the gelatin cannot be clearly seen at 200-220°C. The endothermic peaks corresponding to the glass transition of gelatin^{23,24} can be observed in the same range as the melting temperature of PVA. Therefore, the endothermic peaks of the PVA/gelatin blends (except pure gelatin) are regarded as the melting temperature of PVA. However, the area under the PVA melting peak decreases as the gelatin content increases, and this corresponds to the decrease in crystallinity observed in the XRD results. The decrease in the melting temperature was observed at a PVA/gelatin ratio of 85/15. On the basis of these facts, the depression of the melting temperature indicates the strong interaction between PVA and gelatin molecules. Thus, we can say that PVA/gelatin blends are miscible blends. Hence, they are compatible with each other.

XRD studies

The microstructural variations of PVA upon the incorporation of gelatin were investigated with wide-angle X-ray diffraction (WAXD). The observed WAXD patterns for pure PVA and PVA/gelatin blend films are shown in Figure 4. It shows a relatively sharp and broad peak centered at $2\theta = 19.56^{\circ}$



Figure 4 XRD for PVA/gelatin blend films of different compositions: (a) 100/0, (b) 98/2, (c) 95/5, (d) 93/7, (e) 90/10, and (f) 85/15.

XRD Results for the PVA/Gelatin Blend Films				
PVA/gelatin blend composition (w/w)	20	$d = (n\lambda/2\sin\theta)$ (Å)	Crystallinity (%)	
100/0	19.56	4.5347	69.82	
98/02	18.96	4.9406	67.25	
95/05	17.21	5.1680	63.49	
93/07	16.72	5.2966	59.36	
90/10	16.33	5.4209	56.42	
	39.48	2.2810		
85/15	16.25	5.4488	51.82	

 TABLE I

 XRD Results for the PVA/Gelatin Blend Films

 λ = wavelength; *n* = order of diffraction; θ = phase angle.

(d = 4.5347 Å), indicating the semicrystalline nature of PVA, which contains crystalline and amorphous regions. The crystalline phase of this polymer may be regarded as an amorphous matrix in which small crystallites are randomly distributed. However, it is more natural to treat a crystalline region as a certain sufficiently imperfect crystalline lattice in which the free volume is filled with an amorphous phase. Sites saturated with crystal defects, which are due to chain folded crystals, may play the role of an amorphous region.²⁵

In the case of PVA/gelatin blends, various sharp peaks were observed. The unchanged nature of a broad peak at $2\theta = 18.96^{\circ}$ (*d* = 4.9406 Å) for 2 wt % gelatin indicates that the crystallinity remains the same as that of pure PVA. For 95/05 PVA/gelatin blend films, many multiple peaks were observed with respect to the main peaks. A broad hump was observed at $2\theta = 39.480$ (*d* = 2.281 Å) for 10 wt % gelatin. Table I show 20 values, *d*-spacings, and crystallinity percentages obtained by XRD. From these data, one can notice that, at small angles, the firstorder diffraction shifts to lower angles in comparison with pure PVA. These results also show clearly that the gelatin interacts with the polymer chain, mainly with hydroxyl groups, as observed in FTIR spectra and decreases the crystallinity with an increase in its content.²⁶ The decrease in the crystallinity phase of the semicrystalline PVA matrix with gelatin is the result of the local ordering in the polymeric structure due to complex formation. These crystalline regions are connected to amorphous regions in which the molecular chains of the polymer are irregularly folded.

UV-vis spectra

PVA is one of the most important polymeric materials because it has many optical uses that result from its lack of color, its clarity, and its high transmittance.^{27–35} Figure 5 shows the transmittance spectra of pure PVA and PVA/gelatin blend films of differ-

ent compositions. One can observe a nearly zero transmission in the UV range of 190-330 nm for all samples. At longer wavelengths, the transmission decreases as the percentage of gelatin is increased in the blend. Table II presents the analyses for these spectra. The absorption band at 196 nm in pure PVA shows maximum absorption, and the band around 208 nm indicates the presence of unsaturated bonds, C=O and/or C=C, mainly in the tail-head of PVA.¹ The sharp absorption edge around 231 nm in pure PVA indicates the semicrystalline nature of PVA. This polymer contains a single bond in the main chain and double bonds in branches as observed from absorption spectra in the UV (190-330 nm) region.²² Figure 5 shows a shift in both absorption bands and band edges toward the higher wavelength with a different absorption intensity for PVA/gelatin blend films. The shift arising in the absorption edge in the blends is due to the variation in the crystalline nature of the polymer matrix. The absorption intensity is higher in the blends than in pure PVA, and this may be due to the breaking of some intrahydrogen and/or interhydrogen bonds.³⁶ As the gelatin concentration increases, interhydrogen/



Figure 5 UV–vis transmittance spectra for PVA/gelatin blend films of different compositions: (a) 100/0, (b) 95/5, (c) 93/7, (d) 90/10, and (e) 85/15.

PVA/gelatin blend	Transmission	Maximum absorption			
composition (w/w)	percentage at a visible wavelength (300 nm)	λ_{max}	Optical density	Absorption edge	
100/0	78.072	190	0.9837	231	
		250	0.2075		
95/05	74.55	190	1.8264	244	
93/07	70.11	190	1.7206	241	
90/10	39.181	190	3.3077	255	
		192	3.1468		
85/15	27.762	190	1.4782	247	
		192	1.4733		

	TABLE II				
Data for the Optical Absorption and	nd Transmission	Percentage of the	PVA/Gelatin		
Blend Films					

intrahydrogen bonding increases, and hence absorption increases. This is in accordance with the Beers law; that is, the absorption is proportional to the number of absorbing molecules. The shift in the absorption edge in the PVA/gelatin blend films reflects the variation in the energy band gap, which arises because of the variation in the crystallinity with the polymer matrix.^{1,26}

Electrical parameter	100/0	98/02	95/05	93/07	90/10	85/15
$\frac{1}{C_n}$	1.46679 pF	0.0818 pF	0.92084 pF	0.97803 pF	10.6816 pF	4.4193 pF
С _р	10 V	20 V	30 V	40 V	40 V	20 V
	20 Hz	20 Hz	20 Hz	20 Hz	20 Hz	20 Hz
Series capacitance	1.96613 pF	0.95899 pF	1.09733 pF	1.61905 pF	1.06291 pF	157.49 pF
	30 V	40 V	30 V ¹	20 V ¹	10 V	20 V
	20 Hz	20 Hz	20 Hz	50 Hz	20 Hz	20 Hz
3	1.4563	1.1888	1.1481	1.2945	1.4938	0.9374
	0 V	0 V	0 V	0 V	10 V	0 V
	20 Hz	20 Hz	20 Hz	20 Hz	20 Hz	20 Hz
$\tan^{-1}A$	1.8142	0.9336	0.67086	0.708207	0.88	1.84988
	40 V	10 V	20 V	20 V	30 V	10 V
	20 Hz	20 Hz	1 MHz	20 Hz	1 MHz	1 MHz
Quality factor	3.01	2.6	22.5	4.33	3.63	4.27
	40 V	20 V	30 V	30 V	20 V	40 V
	1 kHz	20 Hz	1 kHz	20 Hz	20 Hz	20 Hz
Resistance in parallel	18.493 MΩ	-5.294 MΩ	9.093 MΩ	18.379 MΩ	27.218 MΩ	16.569 MΩ
I.	40 V	40 V	40 V	40 V	10 V	30 V
	1 MHz	1 MHz	1 MHz	1 MHz	1 MHz	1 MHz
Resistance in series	3.339 MΩ	-1.469 MΩ	3.202 MΩ	12.647 MΩ	6.112 MΩ	93.9 MΩ
	40 V	30 V	40 V	30 V	30 V	20 V
	1 MHz	10 kHz	1 MHz	1 kHz	1 MHz	100 kHz
Impedance	98.237 MΩ	96.528 MΩ	28.79 MΩ	30.004 MΩ	97.632 MΩ	90.839 MΩ
1	40 V	30 V	10 V	40 V	10 V	30 V
	10 kHz	10 kHz	100 kHz	100 kHz	100 kHz	100 kHz
Reactance	94.110 MΩ	98.637 MΩ	28.660 MΩ	29.939 MΩ	93.396 MΩ	89.662 MΩ
	40 V	40 V	10 V	40 V	40 V	40 V
	10 kHz	10 kHz	100 kHz	100 kHz	100 kHz	100 kHz
Phase angle	176.28°	174.847°	105.154°	113.369°	1176.989°	166.589°
	20 V	20 V	10 V	10 V	20 V	15 V
	1 kHz	50 Hz	10 kHz	100 kHz	1 kHz	10 kHz
Conductance	0.10348 μS	0.0556 μS	0.10998S	0.12260 S	0.03428 µS	0.01467 μS
	10 V	30 V	40 V	10 V	10 V	10 V
	50 Hz	1 kHz	1 MHz	1 MHz	50 Hz	1KHz

TABLE III Comparative Electrical Parameters Recorded by the LCR Meter as Function of Frequency at Various DC Bias Potentials for the PVA/Gelatin Blend Films

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Dielectric properties

Role of the DC bias potential

The application of a DC bias potential (influence of a direct field) from 10 to 40 V along with an AC signal clipped the peaks of the AC.^{37,38} The clipped signal polarized the chain molecules, and comparative electrical parameters were recorded with an LCR meter as a function of frequency at various DC bias potentials for pure PVA and PVA/gelatin blend films under NTP conditions. The comparative electrical parameters recorded by the LCR meter as a function of frequency at various DC bias potentials of pure PVA and PVA/gelatin blend films under NTP conditions. The comparative electrical parameters recorded by the LCR meter as a function of frequency at various DC bias potentials of pure PVA and PVA/gelatin blend films are shown in Table III.

Dielectric polarization of the blend films

The dielectric properties of alcohols and diols have attracted the attention of many researchers. There is great agreement that these molecules are associated with various species. The precise nature of these species and the parameters governing the various associative equilibria are still very much sought. The dielectric studies of alcohols and diols have revealed that dielectric dispersion can be attributed to the OH-group reorientation, whereas the intermediate dispersion is suggested to be a contribution from molecular rotation due to an end group of the polymer and/or smaller polymer reorientation. Altogether, numerous studies have been carried out on various types of alcohols as pure liquids, in solution, and even in the solid state.³⁹

In this study, the electrical properties of PVA/gelatin blend films were studied. Variations in ε , the dielectric loss (tan⁻¹*A*), and other electrical parameters give rise to structural changes in the polymer matrix due to the presence of gelatin.⁴⁰ Figure 6 shows a plot of ε for the PVA film. The ε value is





Figure 6 ε as a function of frequency for PVA film.

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Figure 7 Tan⁻¹*A* as a function of frequency for PVA film.

maximum (1.4563) at a frequency of 20 Hz and at a

DC bias potential of 0 V. The second maximum

value of ε (1.1818) can be observed at a frequency of

20 Hz and DC bias potential of 0 V. ε for the PVA

film increases with an increase in the frequency, and

this increase rate is particularly high up to 100 Hz

and then rapidly goes down at 1 kHz. This may be

due to polarization of the O-H chain molecule of

PVA. The dipole orientation seems to be responsible

for the changes occurring in ε of typically high-po-

larity polymers such as PVA and gelatin.⁴¹ Figure 7

Figure 8 ε as a function of frequency for the 98/2 PVA/ gelatin blend film.



Figure 9 Tan⁻¹*A* as a function of frequency for the 98/2 PVA/gelatin blend film.

at a DC bias potential of 0 V. ε decreases at 50 Hz and increases with a further increase in the frequency. $Tan^{-1}A$ of the 98/02 PVA/gelatin blend film is shown in Figure 9. The maximum $\tan^{-1}A$ value (0.93364) can be observed at a lower frequency (20 Hz) and at a DC bias potential of 10 V. At a frequency of 50 Hz, there are sudden decreases in $\tan^{-1}A$ at various DC bias potentials, and with further increases in the frequency and DC bias potential, the tan⁻¹A value increases. The maximum ε value of the 95/05 PVA/gelatin blend film (1.1481) can be observed at a frequency of 20 Hz and at a DC bias potential of 0 V. There is a sharp decrease in the ε value at frequencies of 50 and 100 Hz, and with a further increase in the frequency, ε decreases, as shown in Figure 10. $Tan^{-1}A$ of the 95/05 PVA/ gelatin blend film is shown in Figure 11. The maximum value (tan⁻¹A = 0.67086) can be observed at a frequency of 1 MHz and a DC bias potential of 20 V. At a frequency of 50 Hz, the $\tan^{-1}A$ value decreases, and after a frequency of 50 Hz, there is a sharp



Figure 11 Tan⁻¹*A* as a function of frequency for the 95/5 PVA/gelatin blend film.

increase in the $\tan^{-1}A$ value with a further increase in the frequency and DC bias potential. The ε value of the 93/07 PVA/gelatin blend film is shown in Figure 12. ε (1.2945) is maximum at a frequency of 20 Hz and at a DC bias potential of 0 V. The ε value decreases at 50 Hz and increases at a frequency of 100 Hz. With a further increase in the frequency and DC bias potential, the ε value decreases. Tan⁻¹A of the 93/07 PVA/gelatin blend film is shown in Figure 13. The $\tan^{-1}A$ value (0.708207) is maximum at a frequency of 20 Hz and at a DC bias potential of 20 V. At a frequency of 50 Hz, the $\tan^{-1}A$ value decreases. With a further increase in the frequency and DC bias potential, the tan⁻¹A value increases. ε of the 90/10 PVA/gelatin blend film is shown in Figure 14. The ε value (1.4938) is maximum at a frequency of 20 Hz and at a DC bias potential of 10 V. With a further increase in the frequency and DC bias potential, the ε value decreases. This behavior can be attributed to the decrease in the chain space



Figure 10 ε as a function of frequency for the 95/5 PVA/ gelatin blend film.



Figure 12 ε as a function of frequency for the 93/7 PVA/ gelatin blend film.

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0.8 0.7 0.6 0.5 Dielectric loss -**-** 0v 0.4 - 10v 0.3 · 20\ 0.2 -**e**- 30\ - 40v 0.1 Π -0.1 20 Hz 0 50 Hz 100 Hz 1 kHz 10 kHz 100 kHz 1 MHz Frequency

Figure 13 Tan⁻¹*A* as a function of frequency for the 93/7 PVA/gelatin blend film.

and the free-volume dipoles in the PVA polymer network due to the increasing concentration of gelatin.⁴⁰ Tan⁻¹A of the 90/10 PVA/gelatin blend film is shown in Figure 15. The maximum value $(\tan^{-1}A)$ 0.88) can be observed at a frequency of 50 Hz and DC bias potential of 0 V. At a frequency of 50 Hz, the $\tan^{-1}A$ trend is decreasing up to a frequency of 1 kHz, and with a further increase in the frequency, there is a steady increase in the $\tan^{-1}A$ value. ε of the 85/15 PVA/gelatin blend film is shown in Figure 16. The ε value (0.9374) is maximum at a frequency of 20 Hz and at a DC bias potential of 0 V. The ε value decreases at a frequency of 50 Hz and suddenly increases at 100 Hz. The ε value varies with a further increase in the frequency. Tan⁻¹A of the 85/ 15 PVA/gelatin blend film is shown in Figure 17. The maximum $\tan^{-1}A$ value (1.84988) can be observed at a frequency of 1 MHz and at a DC bias potential of 10 V. The $\tan^{-1}A$ value decreases as the



Figure 15 Tan⁻¹*A* as a function of frequency for the 90/10 PVA/gelatin blend film.

frequency increases up to 1 kHz. With a further increase in the frequency and DC bias potential, the $\tan^{-1}A$ value increases.

Such samples behave as a system in which one phase behaves as an amorphous phase whereas the other phase behaves as a crystalline phase, which restricts the long-range segmental motion in the amorphous phase. Almost all mechanisms are related to different types of polarization taking place in such systems. Each of these mechanisms predominates in a given DC bias potential range at a certain field frequency. The fact that PVA and gelatin are considered to be highly polar suggests that polarization conduction becomes predominant at a low frequency and at low DC bias potentials.⁴² The plot of ε as well as tan⁻¹A is deduced by the application of higher DC bias potentials up to 40 V. As the percentage of gelatin increases, the blend films show good dielectric characteristics.



Figure 14 ε as a function of frequency for the 90/10 PVA/gelatin blend film.



Figure 16 ε as a function of frequency for the 85/15 PVA/gelatin blend film.

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Figure 17 Tan⁻¹*A* as a function of frequency for the 85/15 PVA/gelatin blend film.

CONCLUSIONS

The refractive index of PVA/gelatin blend films decreases slightly with an increase in the amount of gelatin. FTIR analyses of PVA/gelatin blend films highlight a strong chemical interaction between gelatin molecules and PVA chains, mainly with hydroxyl groups. This shows that gelatin interacts with PVA and forms a complex via intramolecular/intermolecular hydrogen bonding. The DSC results show a depression in the melting temperature of PVA, which indicates that the amorphous regions of PVA and gelatin are supposed to be compatible or interact with each other. This also suggests that the interaction of PVA and gelatin molecules changes the crystallite parameters and the degree of crystallinity, and this supports the XRD results. The XRD results show that because of the interaction of gelatin with PVA chains, mainly with hydroxyl groups (as observed in FTIR spectra), structural repositioning takes place that leads to a decrease in the crystallinity with an increase in the gelatin content. The UVvis optical studies also reflect the complex formation and its effect on the microstructure. These microstructural changes due to complex formation are reflected in the variation of the optical properties. It is worth mentioning that the addition of gelatin gives rise to changes in electrical characteristics such as ε and $\tan^{-1}A$.

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