Optical band gap of Polyvinyl alcohol and gelatin composite films

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Abstract: Polyvinyl alcohol (PVA) and its composition with gelatin with different concentrations were prepared in order to investigate the effect of additives on the optical properties of PVA host at room temperature. The optical parameters such as optical energy band gap and Urbach energy were evaluated by measuring of the absorbance in the wavelength range 190-600 nm. It was found that the optical absorption is due to direct transition, the variation in the optical energy band gap and Urbach energy of the films have been observed on doping up to 10% gelatin in to PVA matrix. The changes in the optical parameter of composite films with increase in concentrations of starch in the film have been observed.

Keywords: PVA, Gelatin, Optical energy band gap, Absorption coefficient, Extinction coefficient

1. INTRODUCTION

As well known, synthetic polymer materials have been widely used in every field of human activity [1] during last decades. These artificial macromolecular substances are usually originating from petroleum and mostly of the conventional ones are regarded as nondegradable. The use of synthetic polymer materials has caused significant environmental problems. Solid waste from these materials is a major contributor to environmental pollution because it takes up to a thousand years to degrade. Therefore a great deal of attention has been given to the development of various biodegradable materials to overcome this serious problem. The development of environment friendly polymer materials can be classified into two categories based on the raw material used: degradable synthetic polymers and renewable natural polymers [2]. Both synthetic polymers and natural polymers that contain hydrolytically or enzymatically labile bonds or groups are degradable. Renewable natural polymer resources include starch, cellulose, gelatin and chitosan. These materials have been tested alone and in combination with plastic to enhance the plastic properties and biodegradation potential of the product. In the present study we select gelatin among these four renewable polymer resources in order to obtain optical properties of pristine PVA and its composite films with addition of gelatin.

Gelatin was one of the first macromolecules employed in the production of biomaterials [3]. This biopolymer still attracts the attention of researchers because it is produced abundantly practically worldwide, has a relatively low cost and possesses excellent functional and filmogenic properties [3,4]. For this reason gelatin has been studied in film technology both alone [5-10] and in the blend with other biopolymers [11]. Gelatin based films present good mechanical strength, despite their reduced water vapour barrier [11]. On the other hand, these films present high susceptibility at room temperature and relative humidity conditions due to the hydrophilic nature of gelatin.

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Recently, several attempts have been made to blend gelatin with biodegradable synthetic polymers in order to modify the physical properties of gelatin. Low density polyethylene is the most studied synthetic polymer blended with biopolymers [12]. However, this polymer needs chemical treatment to become biopolymer compatible [13]. Polyvinyl alcohol (PVA) is a well known synthetic polymer and has recognized as biodegradable material [14,15]. PVA possesses excellent mechanical properties [16] and it is one of the best option to be blended with gelatin. PVA is a versatile polymer, and it may be the only synthetic polymer with a backbone that consists primarily of -OH bonds that is absolutely biodegradable. PVA is also an interesting watersoluble synthetic polymer with a broad range of application. PVA has the advantage of good film formation, strong conglutination and high thermal stability.

Optical properties of polymers constitute an important aspect in the study of electronic transition and the possibility of their application as optical filters, a cover in solar collection, selection surfaces

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and green house. The information about the electronic structure of crystalline and amorphous semiconductors has been mostly accumulated from their optical properties. The significance of amorphous semiconductors lies within its energy band gap [17].

Polymer absorbed amount of incident light and consequently optical transitions start when energy of photons is absorbed at a quantity higher than or equal to the forbidden energy gap. If the required energy is almost equal to the difference between the lowest level of conduction band and the highest level of covalent band, electrons will transfer from covalent band to the conduction band. At high absorption coefficient level $\alpha(\nu) > 10^4 \text{ cm}^{-1}$, the absorption coefficient: $\alpha(\nu)$ for noncrystalline materials can be related to the energy of the incident photon according to Mott, Devis and Tauc formula [18-22]

$$\alpha(\nu)h\nu = \beta(h\nu - E_g)^r \qquad \text{for } h\nu > E_g$$

= 0 \qquad for $h\nu < E_g \qquad \dots (1)$

where β is a constant, E_g is the optical energy band gap, $h\nu$ the photon energy and $\alpha(\nu)$ is the absorption coefficient, which was determined near the absorption edge at different photon energies for all the investigated films using the relation

$$\alpha(\nu) = \frac{2.303A_b}{d} \qquad \dots (2)$$

where A_{b} is the absorbance and d is the thickness of film.

The electron transition between the valance and conduction bands can be either direct or indirect. In both cases it can be allowed as permitted by the transition probability or forbidden when there is no such probability. The exponent r in the equation (1) is an index, determined by the type of electronic transition causing the optical absorption $r = \frac{1}{2}$ for direct-allowed transition, r=2 for indirect-allowed transition, $r = \frac{3}{2}$; for direct forbidden transition, and r=3 for indirect torbidden transition.

Figure 1a and Figure 1b represents the direct and indirect absorption transition of an electron from valance band to conduction band. The straight line values of $(\alpha h\nu)^2$ versus $h\nu$ plots reveal the nature of transition in PVA composite films is direct-allowed (i.e. $r = \frac{1}{2}$ for our case). Intercept of the linear portion of the plot on th $h\nu$ axis provides the value of direct optical band gap energy $E_{\rm g}$, of the PVA and its composite films with starch and gelatin.



2. EXPERIMENTAL

Polyvinyl alcohol molecular weight 1,25,000 supplied by Central Drug House Pvt. Ltd., New Delhi (India) and gelatin were procured from Hi Media Laboratories, Mumbai. The film of pristine PVA and different concentrations of gelatin up to 10 wt% were prepared by solution cast method [23-27]. The thickness of the films was of the order of 110 μ m. The prepared films were subjected to investigate their optical as well as structural properties using UV-Visible absorption spectroscopy and FTIR analysis. **UV-Visible Spectroscopy**

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Optical absorption spectra of the films were recorded using a PC based dual beam UV-Visible spectrophotometer (Model: UV5704SS) supplied by EC India Ltd., in the wavelength range 190-600 nm with a resolution less than 2nm and wavelength accuracy ± 0.5 nm at normal incidence.

Fourier Transform Infrared (FTIR) Spectroscopy

FTIR spectra of the films were determined on Shimadzu (Affinity 1) spectrometer, at a resolution of 4 cm^{-1} , and 12 scan in absorbance mode in the wave number region 500-4000 cm⁻¹.

2. RESULTS AND DISCUSSION

Ultraviolet-visible (UV-VIS) absorption spectra of the prepared samples have been carried out at room temperature in the wavelength range 190-600 nm, using a PC based dual beam spectrophotometer. The variation in the optical absorption with wavelength (λ), for all the prepared films with addition of gelatin in PVA matrix is shown in figure 2. It is clearly observed from these curves that there is very less absorption in the visible region as compared to the ultraviolet region for all the samples. The observed

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Figure 2:- The UV-visible absorption spectra of PVA filled with different concentration of gelatin SN - 2456-7 The Urbach energy E_{μ} is linked to the absorption

coefficient $\alpha(\nu)$ in the lower energy region of the fundamental edge and can also be described by the Urbach's relation [28,29].

 $\alpha(v) = \alpha_0 \exp \exp \left[\frac{hv}{F_0}\right]$... (3)

In order to evaluate Urbach energy (E_{μ}) , the natural logarithm of absorption coefficient $\alpha(\nu)$ was plotted as a function of photon energy $(h\nu)$. Magnitude of E₁ was estimated by taking the reciprocal of the slopes of the linear portion of the curves. The Urbach energy values obtained using equation (3) for pure PVA and PVA/gelatin composite films are listed in Table 1. For pure PVA, value of E_u is 0.20 eV [Fig 3].

The absorption coefficient $\alpha(\nu)$ and extinction coefficient k have been obtained using equation 2 and 4 respectively,

 $k = \frac{\alpha \lambda}{4\pi}$ (4)Further the evaluated values of E, can be correlated to the number of carbon atoms per molecule through the following expression according to Fink et al. [30]. $E_g = \frac{34.3}{\sqrt{M}}$.. (5)

Where M is the number of carbon atoms in carbonaceous cluster.



Figure 3:- Optical energy band gap of PVA film filled with different concentration of gelatin

It has already established in the findings of Devi et al. [31] that the value of E_{g} is 5.32 eV for pure PVA. The values of E_g so determined (Fig. 3) for PVA/gelatin composite films are tabulated in Table 1.

After comparing the absorption spectra recorded for PVA/gelatin composite films, shown in Figure 2. It is clearly observed that as the composition of gelatin in PVA increases it absorbs more ultraviolet radiation. It is also seen that the peak appears at 273 nm wavelength, which is the characteristic of undoped PVA [31] and its height decreases with increase in gelatin concentration. This may be ascribed to the cross-linking between PVA, which has free hydroxyl -OH groups and gelatin, which has C-terminal amino acid COOH groups. The following reaction between them may take place:

$$\begin{array}{c} \begin{array}{c} CH_2 - CH - CH_2 - CH \\ | \\ OH \\ OH \\ OH \\ OH \\ COOH \end{array} \begin{array}{c} CH_2 - CH - CH_2 - CH \\ | \\ O \\ OH \\ COH \\ CH_2 - CH - CH_2 - CH \\ | \\ O \\ OH \\ OH \\ CH_2 - CH - CH_2 - CH \\ | \\ OH \\ OH \\ CH_2 - CH - CH_2 - CH \\ | \\ OH \\ OH \\ CH_2 - CH - CH_2 - CH \\ | \\ OH \\ OH \\ CH_2 - CH - CH_2 - CH \\ | \\ OH \\ OH \\ CH_2 - CH - CH_2 - CH \\ | \\ OH \\ CH_2 - CH - CH_2 - CH \\ | \\ OH \\ CH_2 - CH - CH_2 - CH \\ | \\ OH \\ CH_2 - CH - CH_2 - CH \\ | \\ OH \\ CH_2 - CH - CH_2 - CH \\ | \\ OH \\ CH_2 - CH - CH_2 - CH \\ | \\ OH \\ CH_2 - CH - CH_2 - CH \\ | \\ OH \\ CH_2 - CH - CH_2 - CH \\ | \\ OH \\ CH_2 - CH - CH_2 - CH \\ | \\ OH \\ CH_2 - CH - CH_2 - CH \\ | \\ OH \\ CH_2 - CH - CH_2 - CH \\ | \\ OH \\ CH_2 - CH - CH_2 - CH \\ | \\ OH \\ CH_2 - CH - CH_2 - CH \\ | \\ OH \\ CH_2 - CH - CH_2 - CH \\ | \\ OH \\ CH_2 - CH - CH_2 - CH \\ | \\ OH \\ CH_2 - CH - CH_2 - CH \\ | \\ OH \\ CH_2 - CH - CH_2 - CH \\ | \\ CH_2 - CH - CH_2 - CH \\ | \\ CH_2 - CH - CH_2 - CH \\ | \\ CH_2 - CH - CH_2 - CH \\ | \\ CH_2 - CH - CH_2 - CH \\ | \\ CH_2 - CH - CH_2 - CH \\ | \\ CH_2 - CH - CH_2 - CH \\ | \\ CH_2 - CH - CH_2 - CH \\ | \\ CH_2 - CH - CH_2 - CH \\ | \\ CH_2 - CH - CH_2 - CH \\ | \\ CH_2 - CH - CH_2 - CH \\ | \\ CH_2 - CH - CH_2 - CH \\ | \\ CH_2 - CH - CH_2 - CH \\ | \\ CH_2 - CH - CH_2 - CH \\ | \\ CH_2 - CH - CH_2 - CH \\ | \\ CH_2 - CH - CH_2 - CH \\ | \\ CH_2 - CH - CH_2 - CH \\ | \\ CH_2 - CH - CH_2 - CH \\ | \\ CH_2 - CH - CH_2 - CH \\ | \\ CH_2 - CH - CH_2 - CH \\ | \\ CH_2 - CH - CH_2 - CH \\ | \\ CH_2 - CH - CH_2 - CH \\ | \\ CH_2 - CH_2 - CH \\ | \\ CH_2 - CH$$

Hence, the conjugate C=O groups are most probably forward with the addition of gelatin to PVA structure. This may be the reason responsible for the decreased peak height with increased gelatin concentrations. Table 1, indicates that the value of E_g for PVA/gelatin composite films decreases from 5.14 eV to 4.84 eV with increasing gelatin concentrations from 2 wt% to 10 wt% into PVA lattice (Fig. 3). The variation of the obtained values of E, may reflect the role of gelatin in modifying the electronic structure of PVA matrix due

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to appearance of various polaronic and defect levels [33]. Table: 1:- Optical energy band gap (E_a), Urbach energy (E_n) , Absorption coefficient (α), Extinction coefficient (k) and number of carbon atoms in carbonaceous cluster (M) carried out for PVA and PVA/gelatin composite films

Composition	E _g (eV)	Eu (eV)	α (cm ⁻¹) at 235 nm	k x10 ⁻⁶ at 235 nm	М
Pure PVA	5.32	0.20	211.50	395.38	42
PVA+2%Gelatin	5.14	0.46	260.92	489.83	45
PVA+4%Gelatin	5.10	0.54	320.37	601.43	45
PVA+6%Gelatin	5.08	0.58	343.59	645.03	46
PVA+8%Gelatin	4.94	0.69	596.39	1119.59	48
PVA+10%Gelatin	4.84	0.74	558.60	1048.65	50

The decrease in the E_g with filling may be explained on the basis of the fact that the incorporation of small amounts of gelatin forms charge transfer complexes in 56-7 the host matrix [31,34]. These charge transfer complexes increase the electrical conductivity by - 24 providing additional charges, this cause a decrease of the optical energy gap. As the gelatin concentration is increased the gelatin molecules start bridging the gap separating the two localized states and lowering the potential barrier between them, thereby facilitating the ò transfer of charge carrier between two localized states [35]. Figure 3 showed the variation in the E_{a} and E_{u} for pure and gelatin doped PVA films. 5.4

E

5.3

5.2

logarithm of the absorption coefficient $\alpha(\nu)$ was plotted as a function of photon energy $(h\nu)$ for the studied PVA/gelatin films. The magnitude of Urbach energy E_n were estimated by taking the reciprocal of the slopes of the linear portion of these curves and substituted in the equation (3). The Urbach energy for PVA/gelatin composite films were found in the range 0.46 eV-0.74 eV. The larger is the value of E_{μ} , the greater is the compositional, topological or structural disorder [21,30]. Hence on loading gelatin into PVA lattice causes more structural disorder of PVA film.

The absorption coefficient $\alpha(\nu)$ and extinction coefficient k have also been obtained for PVA/gelatin composite films using equations 2 and 4 respectively at 235 nm wavelength and depicted in Table 1. These parameters show an increasing trend for all investigated films. The gelatin content in PVA matrix caused an increase of k value, which is a result of increasing the number of refractions due to the increase of the linear polarizability. Further, the evaluated values of E_e, correlated to the number of carbon atoms per molecule through the expression given by equation (5). The calculated values of M for all investigated composite films are also tabulated in Table 1. The value for M of PVA/gelatin composite films ranging between 45-50. The increase in the value of M is attributed to the increased conjugation in monomer units [32] of PVA matrix after the composition of gelatin molecules.

FTIR analysis

The structural features of samples have been successfully ascertained by the technique versatile and accessible for the study of the polymers, FTIR spectroscopy, in the range 4000-500 cm^{-1} are presented in Figures 4 a-b. All FTIR spectra exhibit the characteristic absorption bands typical of PVA which can be assigned to vibrations of the C=O, C-H, C-OH, C-Obonds [35].



0.7

0.6

0.5

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Figure 4b:- FTIR spectra of PVA film filled with 2% gelatin

For the films under study it can be noticed that some observable changes in the intensity and frequency of the bands appear in their spectral features. The PVA film sample give very strong broad band observed from 3300-3525 cm⁻¹ and centered at about 3450 cm⁻¹ (Fig. 4a) may be assigned to O–H stretching due to strong hydrogen bond of intramolecular and intermolecular type [36-39] and the free hydroxyl groups (Fig. 5a&b). The hydroxyl groups from residual water contribute also to the band. In the region (2828-3000 cm⁻¹) both the CH₂ symmetric and antisymmetric stretching vibration bands are notable [40], suggests that the hydrocarbon chains of polymer takes a trans zigzag conformation [41].



Figure 5b:- Intramolecular Hydrogen Bonding in PVA

The residual acetate groups, related from manufacturing, handling, and degradation of the polymer [42] at about 3070 cm⁻¹ associated with C–H stretching from alkyl groups. At 1732 cm⁻¹ and 1716 cm⁻¹ (antisymmetric and symmetric C=O stretching), 1376 cm⁻¹ (C–H wagging), 1258 cm⁻¹ (C–O bending) appear bands from the same groups. Other stretching vibrational bonds appear at 1650 cm⁻¹ and 1560 cm⁻¹ for C=O and C=C groups. Otherwise these observations related to C=O and C=C groups are in good agreement with those by UV spectroscopy as was reported in literature [43-44].

Figure 4b shows the FTIR scan of PVA and 2 wt% gelatin composite film. It is evident from the spectra that there has been a shifting of O-H stretching vibration peak 3487 cm⁻¹, as compared to O-H stretching vibration of pure PVA film, which was appeared at 3450 cm^{-1} (Fig. 4a). This is attributed to the increase in hydrogen bonding or strong intermolecular interaction between PVA and gelatin molecules. The peak at 2900 cm⁻¹ and 2955 cm⁻¹ indicates the presence of C-H stretching vibration peaks in the composite film [45]. The peak at 1653 cm^{-1} is the characteristic peak of the gelatin [46] could also be associated with C–O and C–N stretching [47] and was insensitive to the PVA content. On the other hand the shoulder appearing at 1686 cm^{-1} became a true peak at 1692 cm⁻¹, this band was not visible in the spectrum of pure PVA. The peaks appearing below 1000 cm⁻¹ may be assigned with C–N stretching. An additional peak appeared at 3000 cm⁻¹ in the PVA and gelatin composite film spectra, which is associated with N-H stretching and also belongs to the characteristics of gelatin. This confirmed the presence as well as the interaction of gelatin in the PVA matrix.

1. CONCLUSION

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On loading gelatin upto 10 wt% in PVA matrix, the optical energy band gap lies between 5.14-4.84 eV. The decreasing trend of the optical energy band gap with different gelatin concentrations may be attributed to the formation of charge transfer complexes, that suggests the presence of deep localized state in the band gaps.

The increase of extinction coefficient k with gelatin content in polymer matrix can be

attributed to increase the number of atomic refractions due to the increase of the linear polarizability.

- The UV-Visible spectra of PVA/gelatin composite films show that these composite films can be used to screen off ultraviolet radiation, beside the fact revealed from the results that PVA/gelatin composite films show better capability to absorb ultraviolet radiation. This indicates that gelatin may act as an excellent ultraviolet shield for PVA host especially at higher gelatin content.
- Addition of gelatin with PVA produce completely biodegradable composite materials with adequate optical properties, which could be a practical way to reduce environmental pollution.
- The FTIR analysis of PVA and doped PVA with gelatin films reveals the considerable interaction between PVA and additives.

19 **References**

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