Study physical characteristics of Polyvinyl Alcohol/Carboxymethyl cellulose blend films

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Abstract

The pure polyvinyl alcohol (PVA), pure CarboxyMethyl Cellulose (CMC), and CMC/PVA blend films with different amounts (0.1, 0.2, 0.25, 0.3 and 0.4 g) of both polymers were prepared by simple solution casting method in this study. FTIR and UV-Vis spectroscopies were used to characterize the physical properties of as-prepared samples. FTIR spectroscopy revealed that the original bonds for both PVA and CMC polymers were appearing in blend polymer, but some bonds referring to CMC polymer seemed in high amounts of this polymer. Optical characteristics showed that the absorbance and absorption coefficient of PVA polymer is improved with increased amounts of CMC polymer. The direct relationship between absorbance and absorption coefficient with amounts of CMC polymer will be established. Furthermore, the energy gap of pure CMC (5.24 eV) and pure PVA (4.74 eV) polymers films were estimated. Tuning E_g toward lower value due to change CMC polymer concentration of blend polymer (PVA/CMC) film was achieved. From the findings of optical properties, it concludes that the blend film has the highest absorbance and absorption coefficient and lowest transmittance in the UV region compared to pure PVA and CMC polymers.

Keywords: Blend polymer; CMC polymer; FTIR spectroscopy; PVA polymer; UV-Visible spectroscopy.

1. Introduction

Generally, natural polymeric materials (amber, wool, silk, cellulose, natural rubber etc.) were used in flocculation, textiles, papers, foods, and medicines as well as in several technical operations, such as solar cells, energy storage devices, nano-dielectric structures, and optoelectronic devices [Khoramabadi *et al.* 2020; Pashameah *et al.* 2022; Atta *et al.*, 2021]. Plants' fundamental structural component is cellulose, which is both renewable and biodegradable. It is the world's most abundant natural biopolymer. Natural cellulose fibers are primarily formed by photosynthesis in plants such as grasses, reeds, stalks, and woody vegetation. There are numerous advantages, including renewable, low cost, low density, low energy consumption, high specific strength, nonabrasive, and relatively reactive surface etc., [Al-Bermany *et al.*, 2013]. Cellulose derivatives like carboxymethyl cellulose (CMC) are biocompatible [Miyamoto *et al.*, 1989], and have been used in medication delivery formulations [Hashim & Hadi, 2017]. Despite their effective applications and inexpensive cost, cellulose derivatives have been neglected [Coviello *et al.*, 2007; Malafaya *et al.*, 2007].

The CMC polymer is one of the significant derivatives of cellulose, which is a cellulosederived ionic linear polysaccharide. CMC is employed primarily because of its high viscosity, lack of toxicity, and lack of allergen city. CMC hydro-gel has high water content, superior biodegradability, and a wide range of uses. It can be employed as filler in bio-composite films due to its polymeric structure and high molecular weight [Almasi *et al.*, 2010] CMC can enhance the mechanical and barrier properties of pea starch-based films [Ma *et al.*, 2008]. Furthermore, polyvinyl alcohol (PVA-semicrystalline) is a type of polymer that comes in powders, fibers, and film. This water-soluble polymer is extensively utilized in warp sizing agents, paper coating agents, adhesives, and hydrophilic applications [Atta *et al.*, 2021; Bhat *et al.*, 2005]. PVA has recently been the focus of intense research due to its low cost of production leads to a wide range of industrial applications [Jasim *et al.*, 2020]. PVA polymer with amazing mechanical characteristics compared to CMC-semicrystalline polymer, which has high biocompatibility and biodegradability while suffering from meagre mechanical properties.

Therefore, this study tries to apply the blending technique of PVA/CMC polymers which can improve the physical characteristics of produced materials [Khoramabadi et al., 2020]. Several studies have been done in this area. R. G. Kadhim et al. [Kadhim et al., 2017] have studied the structural, electrical, and optical characteristics of a CuO nanoparticle to enhance the physical properties of (PVA/CMC) blend. The absorbance of (PVA/CMC) blend film increases as the number of CuO nanoparticles increases. The energy gap (Eg) and optical constants (extinction coefficient, refractive index, absorption coefficient, imaginary and real dielectric constants) of the (PVA/CMC) blend increases as the weight percentages of CuO increase. A. J. Kadham et al. [Kadham et al., 2018], prepared (PVP/CMC/MgO nanoparticles) blend film and investigate the optical properties of the film. The results of the experiments revealed that when the concentration of MgO nanoparticles increases, the absorption and refractive index, and extinction coefficient of the blend film was improved. Furthermore, [Khoramabadi et al., 2020], introduce a review paper that includes the benefit of synthesis of (PVA/CMC) composites and their properties and potential applications (such as electrochemical and energy storage devices, food packaging, and agriculture). [Pashameah et. al., 2022], investigated the production and characterization of polymer (PVA/CMC/MnO₂ NPs) nanocomposite; they mentioned to polymer nanocomposite compounds can use in multifunctional applications like (nano-dielectric systems, optoelectronic devices and solar cells).

The current study aims to investigate the effect of applying the blend technique on enhancing the structural and optical properties of (PVA/CMC) composites-based films by taking different amounts of mixing ratios between two polymers, which needs more investigation.

2. Theoretical Part

Absorbance (A) is defined as the ratio of the material's absorbed light intensity (I_A) to the incoming light intensity (I_o) [Soliman *et al.*, 2020]

$$A = \frac{I_A}{I_o}$$
(1)

The absorption coefficient α is well-defined as a material's ability to absorb light of a specific wavelength [Hashim & Hadi, 2017]:

$$\alpha = 2.303 \frac{A}{t} \tag{2}$$

Where t is the sample thickness from the following equation can calculate the energy bond gap [Jawad & Al-kadhemy, 2021]:

$$\alpha h \upsilon = B(h \upsilon - E_g)^x \tag{3}$$

Where E_g : is the optical energy band gap, hu: is the photon energy, B: is a constant and x: is constant may take values (1/2, 3/2) for allowed and forbidden direct transition, respectively and (2, 3) for allowed and forbidden indirect transition, respectively.

3. Experimental Part

Sodium Carboxymethyl cellulose (CMC) was attained as a powder form (AVONCHEM) with an average molecular weight 67000 g/mole made in the UK. PVA was obtained as a powder form (THOMAS BAKER) with an average molecular weight 14000 g/mole made in India. Synthesis of all films was carried out by solution casting method. To obtain pure PVA, pure CMC, and PVA/CMC blend films, these procedures will be taken. Firstly; the appropriate amounts of PVA and CMC powders were chosen (0.5) g dissolved in (15 ml and 25 ml) of distilled water under stirring and heat (30°C) for (2 hr. and 24 hrs.), correspondingly. For blend films, initially (0.1, 0.2, 0.25, 0.3 and 0.4 g) of PVA polymer and (0.1, 0.2, 0.25, 0.3 and 0.4 g) of CMC polymer were dissolved separately in 8 ml and 17 ml of distilled water, respectively. The solutions were stirred and heated at (30 °C) for (2 hr. and 24 hrs.), respectively. The blend solutions were obtained by mixing the two solutions to get (PVA 0.1 g/CMC 0.4 g, PVA 0.2 g/CMC 0.3 g, PVA0.25 g/CMC 0.25 g, PVA0.3 g/CMC 0.2 g, PVA0.4 g/CMC 0.1 g). The mixed solutions were continually shaken for (30 min.) at room temperature until they had a uniform viscous appearance. Secondly, all films were attained by leaving the solutions in a petri dish at room temperature $(25 \,^{\circ}\text{C})$ for one weak to dry and get homogeneous pure PVA, pure CMC and PVA/ CMC blend films.

Using the UV-Visible spectrophotometer type (T80 Series UV/VIS spectrometer) that tests optical properties within the range (200-1100) nm, the absorption and transmission spectra of as-prepared samples are measured. Fourier transformer Infrared spectroscopy (FTIR) was conducted for all films using FTIR spectrometer (type: Bruker-Tensor 27 with ATR unit). Moreover, using a digital micrometre type (Tasha) made in Japan with measurement accuracy (0.001) mm and measurement range (0-150) mm, the thickness of all as-synthesis film wear was measured.

4. Results and Discussions

FTIR Spectra of pure PVA, pure CMC and PVA/CMC blend films with different amounts of mixing are recorded in the range (4000-400) cm⁻¹ to identify the functional groups of asprepared samples as shown in Figure (1) and Tables (1 and 2). The vibrational spectrum of pure PVA is exposed in Figure (1A), it exhibits the characteristics of the monomer vinyl alcohol and it indicates the modes of vibration. The broad bond absorbed out at 3280.12cm⁻¹ assigned to O-H stretching from intermolecular hydrogen bonds. The vibrational bond noticed at 2914.05 cm⁻ ¹ and 2938.75 cm⁻¹ refers to the C-H stretching form alkyl groups. The noticeable peak 1732.61cm⁻¹ is due to C-O carbonyl stretching. Furthermore, three peaks in the region (1328.80, 1373.5 and 1420.45) cm⁻¹ are assigned to C-H and CH₂ Bending. All FTIR spectrum peaks in the region (1300-1461) cm⁻¹ are allocated to C-H and CH₂ bending; it will be appeared discriminate three peaks (1328.80, 1373.5 and 1420.45) cm⁻¹. At interval (1050-1300) cm⁻¹, the peaks (1024.02, 1046.54, 1087.13 and 1241.93) cm⁻¹ are noticed and referred to C-O stretching, while IR peak at 1241.39 cm⁻¹ is expected to C-H wagging mode. Finally, IR two peaks 835.00 cm⁻¹ and 918.58 cm⁻¹ result from C-C starching and CH₂ rocks, respectively. All these bonds will be compared with results of previous studies [Abbas et al., 2014; Kharazmi et al., 2015; Yao et al., 2022].

Furthermore, Figure (1B) demonstrates the FTIR spectrum of pure CMC polymer film, and information on locations of IR peaks appeared with types of bonds given in Table (1). The seven peaks positions at (3291.55, 2920.14, 1585.21, 1417.49, 1265.30, 1020.30 and 897.28) cm⁻¹ correspond to O-H stretching, C-H stretching vibration of the CH, CH₂ and CH₃, Asymmetrical COO- (stretching carboxyl group), scissoring –CH₂, stretching at ether linkage C-O, bending C-O-C, and 1, 4- β Glycoside of cellulose, respectively. The current results are strongly matched with the result of previous investigates [Jawad *et al.*, 2021; El-Sakhawy *et al.*, 2018, Sohaimy & Isa, 2020].

Further FTIR Spectra are planned for PVA/CMC blend films for different amounts of both polymers as illustrated in Figures (1 C to G) and Table (2), for all amounts mixed between two polymers. The IR two peaks referred to O-H stretching (3200-3550) and C-H stretching (2920) cm⁻¹ for both CMC and PVA polymers are observed. The bonds (1585.2 cm⁻¹, Asymmetrical COO-stretching of carbonyl group, CMC polymer) appeared for all blend films. Moreover, noticeable bonds of C=O carbonyl stretch from vinyl alcohol, acetate 3groups (PVA polymer), 1733.61 cm⁻¹ looked with (PVA0.25 g /CMC 0.25 g, PVA0.3 g /CMC0.2 g and PVA0.4 g/ CMC 0.1 g) blend films, i.e. with increasing PVA polymer this bond will appear. In all blend polymer films, the two IR absorption bands refer to C-H scissoring (1417cm⁻¹) and C-H bending (1300-1461cm⁻¹) (PVA polymer). The new band at 1456.66 cm⁻¹ (PVA 0.4 g /CMC 0.1 g) film is corresponding to C-H bending. The bond C-O stretch (1050-1300) cm⁻¹ has appeared in all blend films. While the bond C-C rocking noted for PVA 0.1g/ CMC 0.4g only and IR peak 835.00cm⁻¹ corresponds to CMC polymer that acts 1, 4-β Glycoside of cellulose seemed only in (PVA 0.2g /CMC 0.3 g) and (PVA0.25 g / CMC0.25 g) and disappeared in the rest of films. The present FTIR findings strongly agreed with the outcomes of previous studies [Jawad et al., 2021; Thuy et al., 2021].

Pure PVA film		Pure CMC films	
Assignments Wavenumber (cm ⁻¹)		Assignments	Wavenumber (cm ⁻¹)
O-H stretching (3200-3550)cm ⁻¹	3280.12	O-H stretching	3291.55
C-H Asymmetric stretch form alkyl groups (2800-3000)cm ⁻¹	2938.75 2914.06	C-H stretching vibration of 2920.14 the CH,CH ₂ and CH ₃	
C=O carbonyl stretch (1550-1780)cm ⁻¹	1732.61	C=O stretching of carbonyl 1585.2 group	
CH,CH ₂ bending (1300-1461)cm ⁻¹	1420.54 1373.56 1328.80	CH ₂ ,scissoring 1417.49	
CH-wagging C-O stretch	1241.93 1087.13 1046.54 1024.02	C-O stretching of ether 1265. linkage	
CH 2rocky C-C Stretching (625-970)cm ⁻¹	945.02 918.58 835.00	1,4-β Glycoside of Cellulose 897.28	

Table 1. FTIR characteristics of pure PVA and pure CMC films

Table 2. FTIR characteristics for PVA/CMC blend films

Assignments			Wavenumber(cm ⁻¹)		
0-н	PVA0.1g/CMC0.4g	PV0.2g/CMC0.3g	PVA0.25g/CMC0. 25g	PVA0.3/CMC0. 2g	PVA0.4g/CMC0 .1g
Stretching	3290.96	3275.61	3275.57	3275.99	3275.32
C-H stretching	2920.65	2920.48	2920.96	2920.71	2920.37
C=O stretching carbonyl group	1585.2	1585.2	1585.2	1585.2	1733.61 carbonyl stretching
CH,CH bending	1417.00 1321.02	1418.58 1374.28 1319.02	1417.85 1374.55 1321.02	1417.70 1320.96	1456.66 1319.36
CH-wagging C-O stretch	1263.14 1052.12	1243.62 1021.68	1247.30	1260.12 1053.35	1259.16 1021.50
CH 2 rocky C-C stretching	913.90	834.16	827.08		



Fig. 1. FTIR spectra for (A) pure PVA, (B) pure CMC, (C) PVA0.1 g/CMC0.4 g, (D) PVA0.2 g/CMC0.3 g, (E) PVA0.25 g/CMC0.25 g, (F) PVA0.3 g/CMC0.2 g, (G) PVA0.4 g/CMC0.1 g

UV-visible absorption spectra are studied for pure PVA and pure CMC films as illustrated in Figure (2). It is clear that the maximum absorption wavelength for pure PVA film is located at 280 nm with maximum absorbance. (0.12). This absorption peak may be attributed to $\pi \rightarrow \pi^*$ electronic transition, this matched with the results of Refs. [Taleb et al., 2009; Behera, 2015; Ram & Mandal, 2004; Kaiqiang & Quyet et al., 2020]. Whereas the behavior of the absorption spectrum of pure CMC film has a wide spectrum, and absorbance is greater than pure PVA film. It reveals that the pure PVA and pure CMC films have mostly early zero absorbance and higher transmittance in the visible region because they are colorless polymers. The transmittance (Figure 3) in the UV region shows a higher decrease than in the invisible region. These results are agreement with aforementioned Ref. [Jawad et al., 2021]. The absorption coefficient (α) was calculated from Equation (2) and plotted against wavelength for pure PVA and pure CMC films in Figure (4). The results indicate that resembles the absorption spectrum in behavior but with greater value, i.e. increasing the absorbance leads to increase the value of α . The advantage of measure absorption coefficient to conclude the transition nature when α greater than 10⁴ cm⁻¹, it will be expected direct electronic transition. Whereas, when the value of α less than 104 cm⁻¹, indirect transition will be evaluated [Taleb *et al.*, 2009]. It will be concluded from Figure (4), the absorption coefficient less than 104 cm⁻¹, so indirect electronic transition will be deduced. Furthermore, the value of energy band gap (Eg) can be determined against photon energy (hu) as illustrated in Figure (5). the value of (E_g) for pure PVA film is assessed about (4.74) eV, that powerfully agreed with many literature [Taleb et al., 2009; Al-Kadhemy et al., 2020]. The estimated value of (Eg) for pure CMC film is found (5.24) eV, which is matched with both earlier investigates [Jawad et al. 2021].



Fig. 2. Absorption spectra of pure PVA and pure CMC films



Fig. 3. Transmission spectra for pure PVA and pure CMC films



Fig. 4. Absorption coefficient of pure PVA and pure CMC films



Fig. 5. The allowed indirect transition for pure PVA and pure CMC films

Moreover, the absorption spectrum of PVA/CMC blend polymer is illustrated in Figure (6). the results revealed that direct relationship between the absorbance and the concentration of CMC polymer. The (PVA0.1 g/CMC0.4 g) blend film displays the highest absorbance in both UV and visible regions. These findings may be explained as the absorbance of pure CMC film is higher than pure PVA film, as noted in Figure (2). So, increasing the concentration of CMC molecules in blend film leads to an increase in the absorbance of the (PVA0.1 g/CMC0.4 g) film. The findings of the transmittance spectra of PVA/CMC blend polymer with different mixing amounts demonstrated in Figure (7); which are confirmed the results of UV-Vis spectra of all blend films. When CMC amounts increased, the value of transmittance decreased due to the increasing absorbance of CMC molecules. That can be attributed to the accumulation of CMC inside the polymer matrix [Jawad *et al.*, 2021].

For further inspection of the optical properties of PVA/CMC films, Figure (8) demonstrates the absorption coefficient calculated from Equation (2) and plotted against incident wavelength. It indicates is lowering absorption at low energy, which means that the transitions of electrons are low. The absorption coefficient becomes large at high energy, which shows the high likelihood of transfers of electrons. The findings of Figure (8) revealed that the absorption coefficient for PVA/ CMC blend film is increased by the increase in the additive CMC polymer [Jawad et al., 2021]. The benefit of the absorption coefficient is determining the transition electrons nature when the value of absorption coefficient is high in the higher energy predictable direct transition of electrons. Conservation of momentum and energy can be accomplished before photons and electrons, where the absorption coefficient is low so that indirect transition is expected [Jawad &Al-Kadhemy, 2021]. Conservation of momentum is through phonons alone. Figure (8) shows that the value of absorption coefficient for PVA/CMC blends films is lower than (10^4 cm^{-1}) indicating that it has indirect (E_g). The value of energy gap (E_g) can be calculated for the allowed indirect transition by using Equation (3) as shown in Figure (9) and Table (3). Figure (9) shows a remarkable decrease in the Eg value of (PVA 0.1 g/ CMC 0.4 g) blend film. The inverse relation between (Eg) for indirect transition and the amount of CMC polymer is predictable as confirmed in Table (3). This action may be attributed to the formation of new energy levels in the band gap, resulting in facilitating the crossing of electrons from the valence band to these local levels to [Jawad et al., 2020].



Fig. 6. Absorption spectra of PVA/CMC blend films



Fig. 7. Transmission spectra of PVA/CMC blend films



Fig. 8. Absorption coefficient of PVA/CMC blend film



Fig. 9. The allowed indirect transition for CMC/PVA blend film

Table 3. Energy band gap (Eg) for pure CMC, pure PVA and PVA/CMC blend films

Samples	E _g (eV)
Pure CMC film	5.24
Pure PVA film	4.74
PVA 0.1g/CMC0.4g	3.30
PVA0.2g/CMC0.3g	4.76
PVA0.25g/CMC0.25g	4.75
PVA0.3g/CMC0.2g	4.72
PVA0.4g/CMC0.1g	4.71

5. Conclusions

In this study, the solution casting method was used to prepare pure PVA and CMC and PVA/CMC blend films. Different amounts (PVA0.1 g/ CMC 0.4 g, PVA0.2 g/ CMC 0.3 g, were mixed to improve the physical properties of as-synthesis films. FTIR analysis revealed that increased CMC amounts led appearance of some peaks related to it in the polymer blend. The original peak for both PVA and CMC are presented into blend polymer. From optical properties, it will be concluded that the PVA 0.1g/CMC 0.4g blend film has the highest absorbance and absorption coefficient and lowest transmittance in the UV region. The E_g modification of PVA/CMC blend films was achieved in this study. Where the low value of energy band gap of PVA/CMC blend films compared with E_g for pure PVA (4.74 eV) and pure CMC (5.24 eV) was revealed. That suggests a new area of various applications (such as solar cells and optoelectronic devices) of PVA/CMC blend films.

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