

PREPARATION AND CHARACTERISATION OF CARBOXYMETHYL CELLULOSE/CARRAGEENAN/JACKFRUIT SEED STARCH BLEND FILM FOR PACKAGING APPLICATIONS

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Abstract: *In this study, carboxymethyl cellulose, carrageenan-based films and a blend film with 1:1 concentration were prepared using the solvent casting method and the influence of various concentrations of jackfruit seed starch (0, 0.5, 1.0, 1.5, and 2 g) on its functional properties were investigated. The effect of jackfruit seed starch on strength properties such as tensile strength and elongation, as well as water-related characteristics such as water contact angle, water vapour permeability, and moisture content, were evaluated and compared with the results of the control blend films. FT-IR, TGA, SEM, XRD were performed to identify the functional group and its potential interactions, thermal stability, surface morphology and crystallinity of prepared films. The results suggests that addition of jackfruit seed starch from 0 to 1.5 g enhanced the tensile strength from 26.62 MPa to 33.26 MPa, but thereafter the tensile strength drops to 32.58 MPa and the elongation decreases. When compared to the carboxymethyl cellulose carrageenan control film, some physical parameters of the film, such as water vapour permeability and moisture content, decreased while thickness and contact angle increased significantly. The results of scanning electron microscopy exhibited rough and heterogeneous morphology for films with more starch content, while the control films exhibited smooth and homogenous structure. The presence of starch increased the crystallinity of all films, which attributed to improved thermal and mechanical stability of the prepared films.*

Keywords: Carboxymethyl Cellulose, Carrageenan, Jackfruit Seed Starch; Packaging Film

1. INTRODUCTION

While plastic has numerous functional applications, humans have developed a dependency on single-use or disposable plastic, which has severe environmental effects. People have been more concerned in recent years about the safety of plastic as a food packaging material, as well as its long-term environmental impact. As a result, scientists are concentrating their efforts on developing new biodegradable packaging materials that provide similar properties to petroleum-based products but with lower environmental implications and manufacturing costs (Ballesteros et al., 2017; Jainan, Deenu & Kamthai, 2018). Biodegradable films made from natural origins have been proposed as a viable substitute to synthetic packaging materials (Pinru & Woramongkolchai, 2020; Bao, Xu & Wang, 2009). Agricultural waste and products can be used as a matrix material, which can help to reduce pollution levels in the environment (Tavares et al., 2020).

Cellulose is the most common natural resource on the planet, and modified cellulose have extraordinary filmogenic characteristics. Carboxymethyl cellulose (CMC), a modified cellulose which is soluble in water, it is an anionic polysaccharide made by partially replacing the hydroxyl groups in cellulose with carboxymethyl groups (Boontranurak et al., 2020). Because CMC is made up of an aquaphobic polysaccharide network structure connected by numerous carboxylic groups with a high water affinity, it is water soluble (Su et al., 2010). CMC is a promising biopolymer material with good film-forming ability, biodegradability, biocompatibility, non-toxic, and hydrophilic in nature (Ghanbarzadeh & Almasi, 2011). Carrageenan is a widely used polysaccharide derived from Rhodophyceae seaweed and modified by sulphate. It is made up of repeating unit of disaccharides with alternate α -(1-3)-D-galactose and β -(1-4)-3,6-anhydro-D-galactose (Sun et al., 2018). During the drying process of the film forming solution, the carrageenan forms a compact film due to the creation of a double-helical random coil structure (Shojaee-Aliabadi et al., 2013). To enhance its film-forming property, tensile strength, and brittleness, carrageenan was mixed with various other polymers such as starch (Ghanbarzadeh, Almasi & Entezami, 2010) and sodium alginate (Paula et al., 2015) and reported to have excellent outcomes.

Starch is a low-cost, biodegradable polysaccharide polymer made from a renewable and abundant resource (Tanetrungroj & Prachayawarakorn, 2020). The starch from jackfruit seeds (*Artocarpus heterophyllus*) is cheap and plentiful all year (Kahar et al., 2019). Starch films reported to have good barrier against gas and lipids characteristics in general (Tongdeesooontorn et al., 2011). When compared to traditional polymeric materials, they have inferior mechanical characteristics and higher water vapour permeability. It is combined with natural polymers such as cellulose and CMC to boost the mechanical property and water vapour barrier property, as well as to maximize its inherent qualities (Pongsawatmanit et al., 2018).

Carrageenan and carboxymethylcellulose blend films had better functional characteristics than its respective control films according to experimental analysis (Hamdan et al., 2019). As a result, there is a greater opportunity to investigate the properties of combination blend films for use as a packaging material. An experiment involving the addition of starch to the carrageenan enhanced the functional characteristics of the control film (Shahbazi, Majzoobi & Farahnaky, 2018). There has been no research on the combination of carboxymethylcellulose carrageenan and starch film, despite the fact that it has a great potential for use as a sustainable packaging material. As a result, the goal of this study is to see how different concentrations of JFS affect the polymer matrix of CMC and carrageenan, as well as to see if this film is suitable for packing applications.

2. MATERIALS AND METHODS

2.1 Materials

Carrageenan (iota-carrageenan) was purchased from Himedia Laboratories Pvt. Ltd., India and sodium carboxymethyl cellulose from Merck India Pvt. Ltd., India. The jackfruit seed was extracted from the ripened jackfruit bought from the local market and manually deseeded. The seed's mucous was entirely removed by intensive washing. The white seed coats and brown spermoderm layers of the seeds were manually removed. The extraction of starch from jackfruit seed was made with reference to experimental procedure outlined by Naknaen, Tobkaew & Chaichaleom (2017).

2.2 Film forming process

The CMC and CAR control films were formulated by solubilising 3 g of each material in 100 mL of deionised water respectively. The film-forming solution CC was prepared by dispersing 1.5 g of CMC and 1.5 g CAR in 100 mL of deionized water with 70^o C temperature, at 600 rpm revolution for 2 hours using a hot plate magnetic stirrer. The films CCS1, CCS2, CCS3 and CCS4 were prepared by adding JFS with weight of 0.5, 1.0, 1.5, and 2 g respectively to CC film forming solution and continued stirring for 30 minutes. Each of these blend solutions of 100 mL volume was poured onto a glass petri dish of 15 cm diameter. It was allowed to dry for 24 hours at 60^o C temperature using a ventilated hot air oven and the film was peeled off thereafter. The prepared sample films were kept in the environmental stability chamber with 65% RH and 23^o C temperature for 24 hours before carrying out each testing.

2.3 Functional Groups

Functional group identification, and its physio-chemical interaction between film forming materials present in the films were determined by Fourier transform infrared spectroscopy (8400 S spectrophotometer, Shimadzu, Japan) and the samples were scanned under a frequency range from 4000 cm⁻¹ to 400 cm⁻¹.

2.4 Surface Morphology

The surface morphology of the film was investigated by scanning electron microscopy (SEM) analysis (Tescan, Vega III). The films were sputtered with gold under the vacuum condition and examined at 5 k magnification with 10 microns resolution.

2.5 Thermal property

The thermal properties of the films was determined by a thermogravimetric analyser (TGA, Netzsche 5). A film sample of 2 mg was paced in an aluminium plate and the sample then heated at 10° C/min rate and the changes in mass was scanned within the temperature range of 25° C to 600° C.

2.6 Crystallinity

Crystalline phases of the films were ascertained by using an X-ray diffraction (XRD, Philips X-Pert PRO, 127, Netherlands) instrument with the 40 kV energy, 30 mA current and Cu K α irradiation ($\lambda = 1.54056 \text{ \AA}$).

2.7 Film Thickness and Moisture Content

The film thickness was measured with a digital micrometre (Mitutoyo, Japan) with a precision of 0.1 μm . The thickness value is determined by taking measurements in three random places and calculating the mean of obtained results.

The moisture retention capacity of the film was assessed by employing moisture content test using gravimetric analysis. The pre-cut film sample (2 \times 2 cm) was preweighed and kept at 1050 C for 24 hrs until a constant weight was attained. All the tests were conducted thrice, and the means were taken with standard deviation. Moisture content was calculated by following the equation (Soradech et al., 2012)

$$\text{Moisture content (\%)} = \frac{W_0 - W_1}{W_0} \times 100 \quad (1)$$

Where W₀ is the initial weight and W₁ is the final dry weight.

2.8 Water Barrier Property

The water vapour permeability (WVP) of prepared sample films was measured gravimetrically following the ASTM E 95-96 standard as described as follows (Ayana & Turhan, 2009). A clean dry glass vial is filled with 1 g of anhydrous CaCl₂ (0% RH) and then covered with the film material to be tested. A sealant was used to prevent any leakage of any moisture across the joints. The prepared vials were placed in a desiccator containing distilled water. The changes in weight of the vial were measured at regular intervals of time and WVP is calculated using the equation below:

$$WVP = \frac{w \times l}{t \times A \times \Delta P} \quad (2)$$

where, w is the amount of absorbed water (g), l is the average thickness (m), t is the time (s), A is the area of the exposed film surface (m²) and ΔP is the partial vapour pressure difference across the film (Pa).

2.9 Surface Wettability

The surface wettability of the prepared samples was determined by using a contact angle meter (Holmarc, India). The film was mounted on to the sample base and a drop (0.1 μl) of water was released to the film surface by the micro-syringe. At room temperature, the angle of contact (θ) of water droplet was determined.

2.10 Mechanical Properties

The tensile strength (TS) values of the samples were used to determine the sample film's mechanical strength. Elongation at break (EAB) values was used to assess dimensional stability or stretchability. For the experimental investigation, a universal testing machine (H10K-S, Tinius Olsen, UK) with a 100 N load cell and a 150 mm gauge length was used, and the ASTM D882–12 standard was followed. Three samples were used to obtain measurements, and the average values were recorded. The following formulae were used to compute the TS and EAB values:

$$\text{Tensile strength (MPa)} = \frac{F}{A} \quad (3)$$

$$\text{Elongation at break (\%)} = \frac{\Delta L}{L_0} \times 100 \quad (4)$$

Where, F was the Force (N) at maximum load, and A was the initial cross-sectional area (mm^2) of the film specimen. ΔL was the difference in length of film at the break, L_0 is the original length of the film.

2.11 Statistical Analysis

All tests were run at least three times, and data was collected using the determined mean and standard deviation. The analysis of variance (ANOVA) tool in SPSS software 13.0 was used to do statistical analysis on a completely randomised design. To discover the differences in the means, Tukey B Test was used, and $p < 0.05$ was regarded statistically significant.

3. RESULTS AND DISCUSSION

3.1 Functional Groups

The figure 1 represents the FT-IR spectra of prepared films. The spectra of all samples show a wide band around 3450 cm^{-1} and is corresponding to the hydrogen bonding by the OH stretching vibration, which is considered to be the characteristic peak of materials with high moisture content. In CMC film spectra, the small peak at 2920 cm^{-1} was due the C-H stretch linked to the hydrogen atoms methane ring (Ma, Chang & Yu, 2008). The transmission peak at 1224 cm^{-1} was assigned to the development of the sulphate ester group, while the bands at 924 cm^{-1} and 842 cm^{-1} were assigned to the C-O stretch of 3, 6 anhydro galactose and the C-O-SO₃ stretch of D galactose 4 sulphate stretching of the polysaccharide skeleton, respectively (Akhtar et al., 2018).

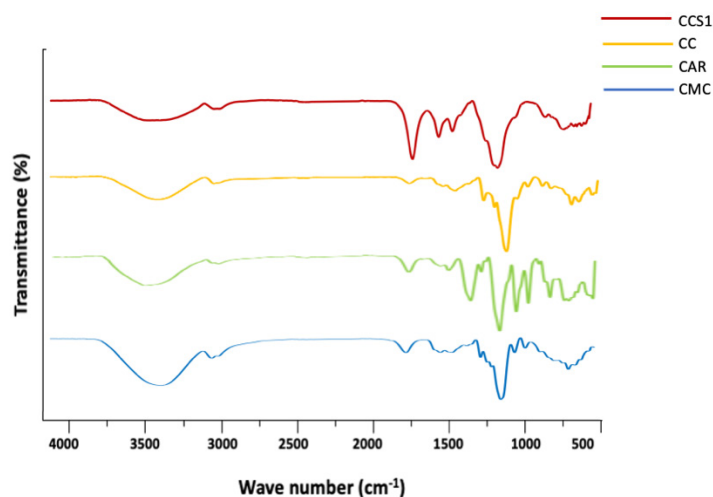


Figure 1: FTIR spectra of CMC, CAR, CC, and CCS

The asymmetric stretching of C-H groups is credited to the FT-IR spectra of CAR control film, which peaks at 1937 cm^{-1} . The asymmetric stretching of ester sulphate groups (O=S=O) is responsible for the absorption band around 1226 cm^{-1} (Li et al., 2022). C-O-C stretching of 3,6 anhydro galactose and C-O-S stretching of galactose 4 sulphate are represented by the distinctive peaks at 923 cm^{-1} and 845 cm^{-1} , respectively (Kanmani & Rhim, 2014). CMC-Carrageenan film (CC) does not show any variations in the peak as the FTIR spectra of CMC and carrageenan show similar functional groups.

The starch film (CCS1) is having an intensity of the broad OH bond at 3800 cm^{-1} - 3000 cm^{-1} indicates the presence of hydroxyl groups. A single peak found at about 1646 cm^{-1} is assigned to the strongly bound moisture presence in the JFS structure due to its hydrophilic character (Nzengu et al., 2018). The peak at 1409 cm^{-1} and 1433 cm^{-1} is related to the C-H bending of CH₂ and peak at 1240 cm^{-1} , 1299 cm^{-1} , and 1333 cm^{-1} are linked to O-H bending of primary or secondary alcohol (Van Soest et al., 1995). The intensity bands at 982 cm^{-1} were associated to C-O stretching

In the blend film CS1, there is a displacement of peak from 1648 cm^{-1} to 1597 cm^{-1} by the addition of JFS suggesting a probable interaction with CMC and CAR (Tongdeesoontorn et al., 2011). The inclusion of JFS lowered the water affinity of the produced films by lowering certain characteristic peaks, particularly

peaks associated with the O-H group. As a result of the effect, the water-related properties of the sample films may have a direct influence.

3.2 Surface Morphology

Scanning electron microscopy (SEM) was used to analyse the microstructures of the film to understand the film's smoothness, cracks, and homogeneity. The film sample CC in figure 2(a) shows a smooth homogenous surface without any cracks or pores. This indicates that the CMC and CAR were blended well and do not form any agglomerates suggesting a good intermolecular interaction between CMC and CAR (Ma et al., 2017; Suriyatem, Auras & Rachtanapun, 2019). By the addition of JFS, the film surface shows an increasing heterogeneity nature. The film surface shows a good nonporous structure without much agglomerate for JFS concentration of 1.5 g and above. This suggests a good molecular bonding, which could contribute to improved mechanical and barrier properties (Tavares et al., 2020). The surface of the films with 1.5 g and greater starch concentrations shows some white spots, indicating uneven JFS mixing with the solution. This could be because the solution reached a saturation threshold with 50% JFS concentration (1.5 g).

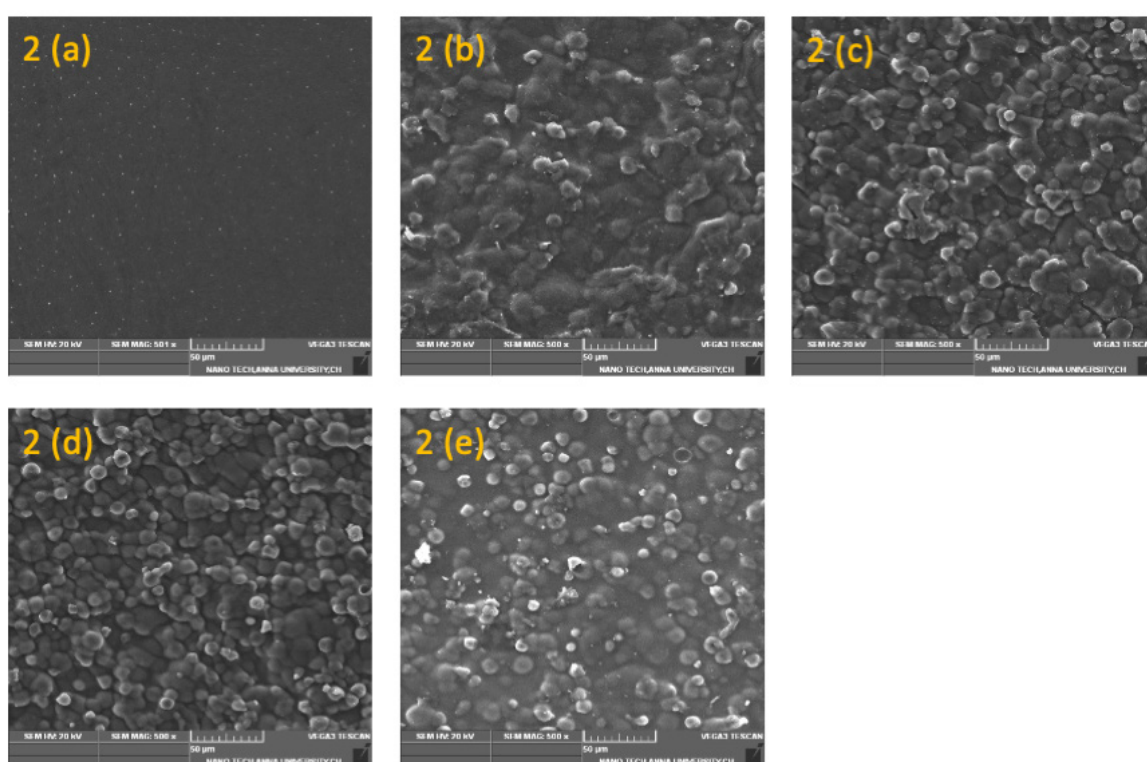


Figure 2: SEM surface structure representing 2(a) (CC), 2(b) CCS1, 2(c) CCS2, 2(d) CCS3 and 2(e) CCS4, respectively.

3.3 Thermal Property

Thermogravimetric analysis (TGA) was carried out to evaluate the thermal stability of the films by the addition of JFS. The CC film sample shows three weight loss changes when the film was exposed to heating until 6000 C as shown in figure 3. The evaporation of water absorbed by the material, produced by the breakdown of O-H bond, causes the first weight loss between 600 C and 1200 C (Kanmani & Rhim, 2014; Akhtare et al., 2018). Between 1700 C and 2400 C, the second stage of mass loss starts, which was connected to structurally bound moisture content. The degradation range of most of the polysaccharide thin films occurs in the range of 2400 C –3600 C (Suriyatem, Auras & Rachtanapun, 2019; Jaramillo et al., 2016).

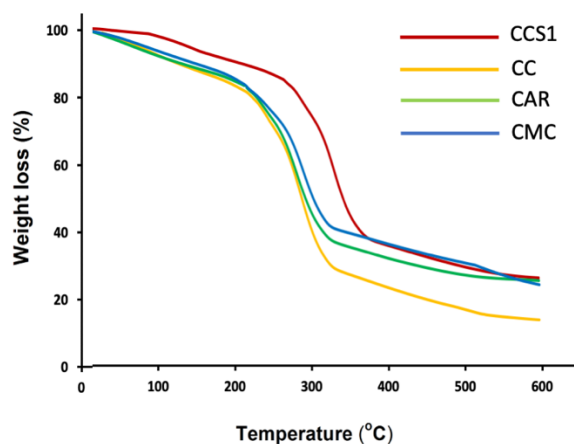


Figure 3: TGA of CMC, CAR, CC and CCS1

3.4 Crystallinity

The X-ray Diffraction (XRD) image of CMC and CAR control films and CC blend films are shown in figure 4. The control film CC displayed a wide peak about 20.00 which reflects the amorphous nature of CMC as well as CAR (Nanaki et al., 2009; Hazirah, Masp & Sarbon, 2016). The broad peak at 51.00 represents a low crystallinity (Ghanbarzadeh & Almasi, 2011). Similar characteristic peaks in XRD pattern were observed in films with a minor peak shift to 21.00 by the addition of starch component (Hazirah, Masp & Sarbon, 2016).

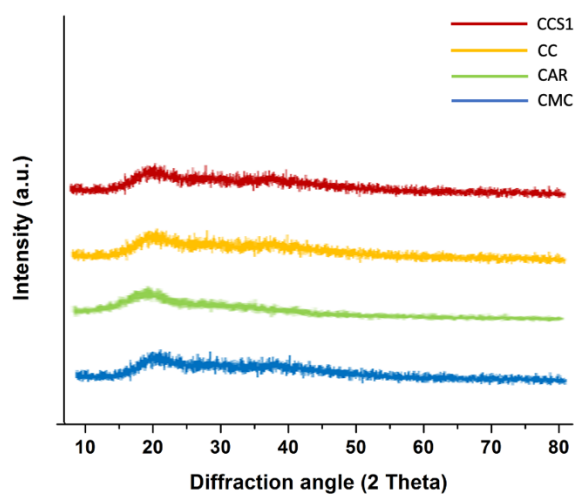


Figure 4: XRD of CMC, CAR, CC and CCS1

3.5 Film Thickness and Moisture Content

From the observed values from table 1, the thickness of the control film (CC) is 140.39 μm and the addition of JFS increased the film thickness significantly. The sample film CS1 had a thickness of 148.58 μm and it gradually increases with the increasing concentration of JFS. The highest thickness value was observed for CS4 sample with 158.62 μm . From the inferred data in table 1, it can be concluded that increasing concentrations of starch positively influences the thickness of the prepared films (Shahbazi, Majzoobi & Farahnaky, 2018).

Moisture content data from table 1, gives the hydrophilic nature of the film and how it was affected by the addition of starch. The inclusion of JFS altered the moisture retention capacity of the film, according to the findings. The film's moisture content decreased from 22.08 % to 10.19 %, and the changes were found to be gradual and significant. The control film CC shows the highest moisture content of 22.08%

(Shojaee-Aliabadi et al., 2013; Ballesteros et al., 2017) compared to all the films with starch content, CCS1, CCS2, CCS3 and CCS4. The presence of active hydrogen and ester bonds created by the hydroxyl and carboxyl groups of CMC with the hydroxyl groups of starch which was responsible for improving the interactions between molecules and the cohesion of the matrix of the polymer blend and there-by decreasing the water affinity of the film (Tongdeesoontorn et al., 2011).

Table 1: Film thickness and moisture content of prepared sample films.

Film Sample	Concentration of CMC/CAR/JFS (g)			Film Thickness (μm)	Moisture Content (%)
	CMC	CAR	JFS		
CC	1.5	1.5	0	140.39 \pm 1.12 ^e	22.08 \pm 0.53 ^a
CCS1	1.5	1.5	0.5	148.58 \pm 1.23 ^d	20.42 \pm 0.73 ^b
CCS2	1.5	1.5	1.0	153.44 \pm 0.92 ^c	17.55 \pm 1.10 ^c
CCS3	1.5	1.5	1.5	155.36 \pm 1.01 ^b	14.69 \pm 0.77 ^d
CCS4	1.5	1.5	2.0	158.62 \pm 0.79 ^a	10.19 \pm 0.36 ^e

All data represent mean \pm standard deviation; the different superscript letter (a-e) in the same column indicate significant difference ($p < 0.05$). Values are expressed as mean \pm SD ($n = 3$).

3.6 Moisture Barrier Property

Water vapour permeability (WVP) is an imperative parameter for a packaging film as it determines the amount of water vapour transferred between the surrounding atmosphere and the product it packed inside, and hence affects the shelf life of the food product. A biopolymer film with lower WVP is better considered for packaging food products as it limits less passage of water vapour. The control film shows a WVP of 2.85×10^{-10} g/m \cdot S \cdot Pa. By the addition of 0.5 g of JFS the WVP decreases to 2.94×10^{-10} g/m \cdot S \cdot Pa, similar observations were also reported in WVP of the film with starch and CMC content (Tavares et al., 2019). Further addition of JFS of 1.0, 1.5, 2 g the WVP decreases gradually from 2.94×10^{-10} g/m \cdot S \cdot Pa to 3.85×10^{-10} g/m \cdot S \cdot Pa. The blending of starch into the matrix reduced the WVP of the film due to the chemical interaction between the carboxylic group of CMC with the hydroxyl groups of the starch (Li et al., 2008). This reduced the number of available hydroxyl groups, thus restricted the mobility of water molecules (Kristo & Biliaderis, 2007).

Table 2: Tensile strength (TS), Elongation at break (EAB), and Water vapour permeability (WVP) of film samples

Film Sample	TS (MPa)	EAB (%)	WVP (g/m \cdot S \cdot Pa)	CA
CC	26.62 \pm 1.17 ^d	38.33 \pm 0.81 ^a	$2.85 \times 10^{-10} \pm 8.5 \times 10^{-12}$ c	49.56 \pm 1.26 ^d
CCS1	29.48 \pm 1.04 ^c	23.45 \pm 1.21 ^b	$2.94 \times 10^{-10} \pm 9 \times 10^{-12}$ c	52.49 \pm 1.18 ^c
CCS2	31.03 \pm 0.75 ^{bc}	18.94 \pm 0.95 ^c	$3.26 \times 10^{-10} \pm 7.5 \times 10^{-12}$ b	53.72 \pm 0.93 ^c
CCS3	33.26 \pm 0.94 ^a	17.54 \pm 1.25 ^c	$3.32 \times 10^{-10} \pm 1.1 \times 10^{-11}$ b	56.54 \pm 1.00 ^b
CCS4	32.58 \pm 1.05 ^{ab}	17.37 \pm 0.93 ^c	$3.85 \times 10^{-10} \pm 1 \times 10^{-11}$ a	59.31 \pm 0.99 ^a

All data represent mean \pm standard deviation; the different superscript letter (a–d) in the same column indicate significant difference ($p < 0.05$). Values are expressed as mean \pm SD ($n = 3$).

3.7 Surface Wettability

The surface wettability of the sample films was evaluated by the contact angle study. As shown in table 2, wettability of the sample film is decreasing with the increasing amount of JFS. The control film shows a contact angle of 49.560, similar observations are also reported in studies of CMC and CAR based packaging films (Ballesteros et al., 2017; Yadav & Chiu, 2019). Incorporation of JFS of different concentrations resulted in an increased contact angle and reached to 59.310 for CCS4. The increased contact angle value was due to the functional groups of CAR, CMC, and JFS interact through hydrogen bonds, which provided more stiffness for the prepared films and lower hydroxyl groups on the surface (Shahbazi, Majzooobi & Farahnaky, 2018). This impact due to hydroxyl group deficiency can also be seen in the observed values of WVP results, where the WVP drops significantly when JFS concentration was increased gradually.

3.8 Mechanical Properties

Mechanical properties such as tensile strength (TS) and elongation at break (EAB) are very important parameters for a packaging film, as it determines the strength and stretchability of the material. Table 2 shows the TS and EAB values obtained for the film samples. The CC control film shows a TS value of 26.62 MPa and EAB of 38.33%, which concurred with similar experiment (Hamdan et al., 2019). By the addition of 0.5 g, to 1.5 g of JFS, the TS increased from 29.48 MPa to 33.26 MPa and the EAB value decreased from 23.45% to 17.54% respectively. The increased strength might be due to the increase in the intermolecular interaction between the CAR and the JFS, as starch molecules have affinity to create molecular bonding with CAR (Abdou & Sorour, 2014). The TS dropped for CCS4 with 2 g JFS, possibly due to increased solid content above the saturation point as seen in SEM examination. Similar experiments found that when the amount of starch in the mixture increased, the strength characteristics reduced (Roy & Rhim, 2020). The lower EAB values suggest that the film stiffened at the highest concentration of JFS and became more dimensionally stable when the JFS concentration was increased.

4. CONCLUSIONS

The carboxymethyl cellulose, carrageenan- based functional polymer films with various concentrations of jackfruit seed starch were prepared by the solvent casting method. The addition of jackfruit seed starch influenced the physical and functional properties of the prepared blend films. The tensile values improved by the addition of starch up to a concentration of 1.5 g, beyond that the tensile strength decreases. The SEM images explain this tendency in the mechanical property as the analysis show a complex heterogenous structure beyond 1.5 g concentration of jackfruit seed starch. The results show that the thickness and contact angle of the film were increased whereas the water vapour permeability and moisture content were reduced, which implies an increase in water barrier properties. The XRD analysis shows an increasing crystallinity for the film with starch content, whereas TGA analysis shows an improved thermal stability for film with starch content. The results shows that carboxymethyl cellulose carrageenan jackfruit seed starch ternary blend films can be used as an assuring substitute for synthetic polymer-based single use packaging material.

5. CONFLICT OF INTEREST STATEMENT

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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