



# Characterization of Biocomposite Film Coating for Food Paper Packaging

Nurul Fatin Alia Mustapha<sup>1</sup>, Zatul Iranati Md. Sharif<sup>1</sup>, Junaidah Jai<sup>1\*</sup>, Noorsuhana Mohd Yusof<sup>1</sup>, Sitinoor Adeib Idris<sup>1</sup>

<sup>1</sup>Faculty of Chemical Engineering, Universiti Teknologi MARA, 40450 Shah Alam, Selangor.

\*Corresponding author E-mail: [junejai@salam.uitm.edu.my](mailto:junejai@salam.uitm.edu.my)

## Abstract

Paper and paperboard are commonly laminated with petroleum-based plastic such as polyethylene (PE) to enhance its barrier and mechanical properties. However, it cannot be degraded naturally by the environment. In an effort to counter this issue, natural sources biopolymer film was developed from cassava starch and carboxymethylcellulose (CMC) as alternatives in replacing PE. Different formulation viscosity was prepared by varying the contents of CMC in cassava starch from 0% to 50%. Characterizations analysis showed that sample BF 3 containing 10% w/w starch of CMC had the highest tensile strength (15 MPa), exhibited hydrophobic properties with contact angle higher than 90° and had the lowest moisture content. Fourier Transform Infrared (FTIR) analysis on BF 3 confirmed the presence of C-OH bending at C-6 of glycosidic ring indicated the presence of intermolecular and intramolecular hydrogen bond formation. A comparison study of BF 3 with commercially coating material –used in food packaging showed that BF 3 exhibit similar hydrophobic properties but higher tensile strength than the PE coating. Besides, thermogravimetric analysis identified that BF 3 had less residue and this indicated that BF 3 film degraded faster than the commercial. Thus, it can be stated that the develop biofilm could replace the currently used petroleum based derivative plastic in the food industry.

**Keywords:** Biocomposite film; Cassava starch; Carboxymethylcellulose; Food packaging; Paper coating

## 1. Introduction

The main roles of food packaging are to contain the food and to guarantee save delivery of the products to the customers in a great condition [1]. In addition, food packaging can significantly contribute in reducing the total municipal solid waste by extending the shelf-life of foods, thus prolonging their usability. In term of protection, food packaging acts as a preservative by providing protection from 3 major classes of outside influences which are chemical, biological and physical [1].

Currently, the packaging material field is still dominated by petroleum-derived polymers despite of worldwide concerns about the environment. Plastic is a more commonly used material when packaging films are required. Synthetic polymers are advantageous to be used due to their low cost, low density, inert quality, excellent barrier properties, good mechanical strength, high transparency, ability to be heat-sealed and easy to be printed on. The most widely used plastics in packaging include low density polyethylene (LDPE), linear low density polyethylene (LLDPE), high density polyethylene (HDPE), polypropylene (PP), ethylene vinyl acetate (EVA), polystyrene (PS), polyethylene terephthalate (PET) and polyvinyl chloride (PVC) [2].

However, this petrochemical product creates a negative impact to the environment. Most synthetic polymers are non-biodegradable and yet they are superior candidates as food packaging materials. These products cause landfill depletion, environmental pollution and high energy consumption of the manufacturing process. The incineration process of PVC and polvinylidene chloride causes

problems due to their chlorine content [2]. The diffusion of additives from polymers into food products could also endanger human health. Therefore, there has been a shift towards increasing the usage of biodegradable materials in recent years [3].

LDPE is the most commonly used plastic-coated paper for grease and leak proof properties which are known not to biodegrade at a reasonable time. In this study, the potential of biopolymers coating from renewable and natural sources to replace LDPE coatings is studied. Biofilm for paper coating can be prepared by using our everyday foods main components which are protein, lipids and carbohydrates. Using biopolymer as a base for the packaging system offers several advantages such as having excellent physical, mechanical, barrier and optical properties as well as a good carrier for various additives. Besides, it exhibits outstanding environmentally friendliness, biodegradability, biocompatibility and edibility [4]. In addition, by using agriculturally derived alternatives will reduce the usage of petroleum and its derivative, thus increasing the agricultural source of income. Generally, fats or lipids are used for water transmission reduction, polysaccharides act as a barrier for oxygen and other gas transmission while proteins provide mechanical stability.

Starch is a major carbohydrate that can be found in plant tubers and seed endosperm in the form of granules which are classified as polysaccharides [5]. Each of them contains millions of amylopectin molecules together with smaller amylose molecules [6]. Amylose molecules are responsible for the formation of the film capacity of starch. Nieto [7] confers that all starches containing amylose form films when cast under the right conditions. Starch generally cost less compared to other alternative materials with

high tensile strength. This is because starch is available abundantly in nature besides being readily available as an inexpensive commercial product. Cassava is highly abundant in Malaysia as the main source of starches as it is a much shorter term crop in comparison to the other sources of starch [8]. They bring to the fore the fact that cassava starch has a very good potential as a domestic demand. A study on cassava starch by Parra *et al.* [9] showed that films based from cassava starch have good flexibility and low water permeability. The films are also flavorless, colorless and tasteless. This study is agreed by Maran *et al.* [10] where authors stated that cassava starch is a potential starch material for edible film and coating because it exhibits film-forming properties with great biodegradability.

Ghanbarzadeh, Almasi and Entezami [11] stated that the single based edible film from single sources of biopolymer exhibited good properties in some aspects but was poor in other aspects. Films from starch have poor mechanical strength and lack of efficient barrier against low polarity compound as stated by Fennema, Kamper and Kester [12]. Even though biopolymers have the potential as a fully bio-based paper coating, Rastogi and Samyn [13] stated that most single sources of biopolymers exhibit difficulties in processing. This is because of the hydrophilicity properties, brittleness, melt instabilities and crystallization behavior of the biopolymer chains. Referring to this problem that has become a hindrance in full scale of industrial application, the authors suggested to mix the biopolymers with others biopolymers, plasticizers or compatibilizers in order to improvise the performance of the bio-based coating. Thus, plasticizer was suggested to be used by Mali *et al.* [14] for edible film with starch-based in order to overcome film brittleness. Sorbitol and glycerol are the commonly used plasticizer for film-based starch.

Other than that, blending biopolymers from polysaccharide with protein, polysaccharide with lipid or polysaccharide with polysaccharide sources known as biocomposite films may attribute to better performance of the biopolymers. A study by Alves *et al.* [15] on corn starch film has proven that the films by themselves are unsuitable for packaging purposes as they exhibit poor mechanical properties and are highly water sensitive. Meanwhile, a study by Hong [16] on composite films found that by blending the corn starch with carboxymethylcellulose (CMC), it showed that the film mechanical properties had been improved twice compared to the corn starch film itself. CMC is suitable for film blending with others biopolymers due to its non-toxicity, non-allergen and high viscosity properties besides acting as a filler to improve barrier and mechanical properties [11], [17], [18]. Thus, the objectives of this study was to develop a film solution for coating in food paper packaging by using cassava starch and carboxymethylcellulose as a composite biopolymer and to determine the strength, surface wettability and thermal stability of the developed film.

## 2. Materials and methods

### 2.1. Materials

Commercial cassava starch (Cap Kapal ABC.Co, Thailand) was applied as a polymer base for the coating film. Glycerol (Merck, Germany) was used as the plasticizer. Carboxymethylcellulose, CMC from the oil palm empty fruit bunch was used as a composite biopolymer together with cassava starch as the filler and thickener was kindly provided by Waris NOVE Sdn. Bhd., Pahang, Malaysia. The degree substitution of the CMC was 0.74-0.95 with a viscosity of 200-2217 cP. Distilled water was used as the solvent of the filmogenic solution. Unbleached kraft paper or brown paper (Muda Packaging, Malaysia) was used as the paper food packaging material.

### 2.2. Film preparations

Film-forming solutions was prepared by cassava starch gelatinization (5% w/v of distilled water) at a temperature of  $75^{\circ}\text{C} \pm 5^{\circ}\text{C}$  for 20 mins under continuous stirring at 500 rpm before being plasticized with glycerol (30% w/w of starch). Meanwhile, CMC was dissolved in 100 mL distilled water at room temperature. The amount of CMC added into the film forming solutions varied at concentrations of 0, 5, 10, 20, 30, 40 and 50% w/w of the total solid cassava starch and was labelled as control, BF 2, BF 3, BF 4, BF 5, BF 6 and BF 7, respectively. Then, both of the solutions were mixed and slowly stirred at 250 rpm for 20 minutes at  $40^{\circ}\text{C}$  to remove any air bubble and to ensure the mixture was completely mixed. The film-forming solutions were then casted at volumes of 30 mL on flat kraft paper (30 cm length  $\times$  15 cm width) using an automatic film casting machine at a speed of 1 cm/s for blade distance of 1.25 mm, respectively. After that, the wet-coated paper was dried at  $50^{\circ}\text{C}$  for two and half hours and conditioned in a desiccator at  $25 \pm 1^{\circ}\text{C}$  for 24 hours prior to sample analysis.

### 2.3. Viscosity measurements

A viscosity profile of film-forming solutions of cassava starch/carboxymethylcellulose was measured at various percentage of the composition of carboxymethylcellulose. The best range of viscosity in film-forming solutions for food packaging is 500 to 2000 cP [19]. The viscosity for the film-forming solution was measured by using the Viscometer (Brookfield Engineering Laboratories, Inc, USA). The shear viscosities of the film-forming solutions were measured at room temperature with a cylindrical spindle (LV 25) at viscometer speed of 150 rpm.

### 2.4. Fourier Transform Infrared analysis

The disruption on the functional group of starch-based film affected by the different content of CMC on the starch-based film was analyzed by using the Fourier Transform Infrared (FTIR) spectroscopy (Perkin Elmer, USA) at wave number ranging from 400 to  $4000\text{ cm}^{-1}$ . The sample preparation of the biopolymer films was done by poured 30 mL of film-forming solutions in disposable Petri dish (90 mm  $\times$  15 mm). Next, the solutions were dried at  $50^{\circ}\text{C}$  until the weight of the samples was constant. The sample port was cleaned by using acetone to remove any variation caused by contamination on the scanning. The spectrum yields from the sample scanning were collected and studied.

### 2.5. Thickness measurements

The thickness of the coated paper was measured prior to the characterization study using the Universal Testing Machine to analyze tensile strength. The samples were conditioned in the same atmosphere in a desiccator prior to the measurement. The equipment used was Digital Micrometers (Mitutoyo, USA). The micrometer was free from vibration and measurement was done on a stable horizontal surface. The sample was placed between the open measuring faces and the micrometer was operated in a way that the upper face lowered onto the top surface of the samples. The micrometers showed the reading values. Five readings at different positions on the paper were measured and the mean values of the thickness were calculated.

### 2.6. Tensile strength measurements

Tensile strength measurement studied if the strength of the film-coated paper was sufficient to apply as a food packaging. The mechanical test was conducted using the Tinius Olsen Universal Testing Machine, Model H50KT (USA). Samples were cut into the dimensions of  $75 \times 25 \times 2.5$  mm (length  $\times$  width  $\times$  thickness) before placing them onto the sample holder. The bottom and upper gaps were tightened. The crosshead speed was set at 50 mm/min with a gauge length of 50 mm. The test was repeated three times.

The data were tabulated and plotted using Microsoft Excel. The sequence of procedure is according to ASTM D3500.

## 2.7. Wettability analysis

Wettability analysis was done to identify the hydrophobicity surface of the CS/CMC films by measuring the contact angle of water in contact with the film-coated paper using Video Contact Angle Goniometer (AST Products, INC.). Wettability of film was measured using the contact angle method as suggested by Liu *et al.* [20]. The samples were cut in rectangular strips with a dimension of 1 cm × 1 cm (width × length). The rectangular strip samples were then directly placed on the horizontal moveable stage and 10  $\mu$ L of water was dropped onto the film surface using a 500  $\mu$ L precision micro syringe. For each sample, three measurements were taken and the average values were calculated.

## 2.8. Thermogravimetric analysis

Thermogravimetric analysis was done to tabulate data on the thermal stability of the sample based on its reduction in weight. At the same time, the derivative thermogravimetric analysis was tabulated to know the decomposition of each material used in the formulation of the biodegradable antimicrobial film in term of degradation of the sample's weight per time. The aim of this analysis was to identify the biodegradable properties of the film. The degradation of the samples was investigated by using the Thermo Gravimetric Analyzer (TGA) (Mettler Toledo, USA). Samples weighing 20 mg were placed in a tarred platinum TGA pan. The samples were heated from 27 to 500°C with a heating rate of 10°C  $\text{min}^{-1}$  in an ambient air environment. The samples were monitored during the heating process by recording the weight of the samples every 1 minute. TGA was equipped with precision balance at the bottom of the sample pan. Inert gas was purged to control the sample environment by flowing over the sample and exiting through an exhaust. The sequence procedure followed ASTM D6370 standard test method for compositional analysis through thermogravimetric analysis.

## 3. Results and discussions

### 3.1. Viscosity of film-forming solutions analysis

The trend of changes in viscosity values of film-forming solutions as weight percentage of CMC increased is presented in Fig. 1. As can be seen, viscosity values of solutions gradually increased at each increment of CMC's weight percentage added into the solution. The lowest viscosity value of 349.24 cP was found at 5% (w/w of starch) of CMC and the highest viscosity value at 2173.67 cP was found at 50% (w/w starch) of CMC. However, low viscosity of film-forming solutions allowed the solutions to penetrate into food packaging easily and too viscous solution only allowed the solutions to penetrate only small partway into the paper which violate the concept of paper coating as agreed in study done by Shogren [21].

Referring to Fig. 1, control solution has the lowest viscosity at 230.37 cP. It is also noted that the weight percentage of CMC added into gelatinized cassava starch range from 10 to 40% (w/w starch) are satisfied the viscosity range from 500 cP to 2000 cP [19] with viscosity of 752.91 (BF 3) to 1901.33 cP (BF 6). Therefore, CMC was chosen to mix with cassava starch as biocomposite polymer in order to control the viscosity of film-forming solutions and to improve the ultimate tensile strength of film-coated on paper. This study was done as suggested by Hong [16] on composite films of corn starch with CMC where they found that the film mechanical properties had been improved twice compared to film based from the corn starch only. It was also can be seen that the film-forming solutions with 10 to 40% (w/w starch) of CMC could homogeneously distribute on the paper surface. This is be-

cause, at this range viscosity, the solutions able to penetrate into the paper at certain distance and form a layer of film on the paper surface.

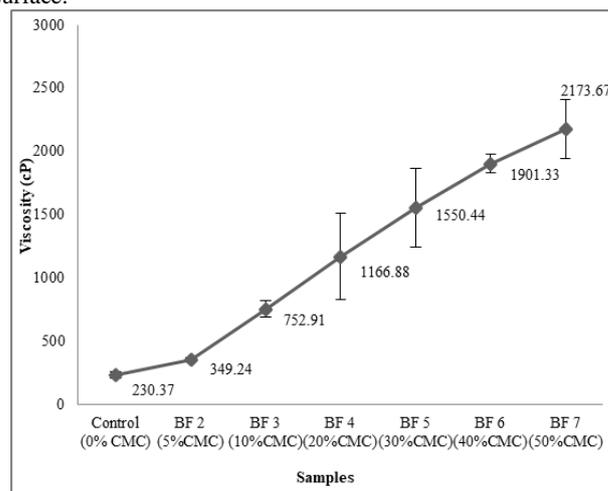


Fig. 1: Effect of CMC on viscosity of film-forming solutions

### 3.2. Fourier transforms infrared analysis

The band assignments of FTIR analysis spectra of cassava starch based film added with different volumes of CMC are shown in Fig. 2. The biopolymer film without presence of CMC (control) had the lowest O-H stretch band assignments which were at 3276.93  $\text{cm}^{-1}$ . Blending CMC with starch biopolymer film significantly shifted the O-H stretch region toward higher wavenumber at 3296.82  $\text{cm}^{-1}$ . This increase in wavenumber indicated that CMC form a complex polymer chain with the starch where the force constant of the bond was increased and thus enhance the films strength.

The O-H stretch regions were shifted to higher and lower wavenumber at different weight of CMC but the trend was not uniformed. It significantly shifted toward lower wavenumber as CMC's weight increased from 5 to 30% (w/w starch) and slightly shifted toward higher wavenumber a CMC's weight increased from 30 to 50%. Thus, it can be suggested that as CMC's weight increased from 5 to 30% (w/w starch) the intramolecular hydrogen-bound hydroxyl group increased and hydrogen bond formation was also increased but the opposite behaviors were observed when CMC weight added exceeded 30% (w/w starch). This is in agreement with results reported by Turhan, Şahbaz, & Güner [22]. Authors stated that the unbounded or free hydroxyl group absorbs strongly in the regions of 3650-3500  $\text{cm}^{-1}$  and presence of hydrogen-bound hydroxyl group may cause a shift of absorption to lower wavenumber down to 3200  $\text{cm}^{-1}$ . The change of wavenumber between 'free' and 'bound' hydroxyl absorption is a measure of hydrogen bonding interactions and the strength of hydrogen bonds in the polymer matrix. Although all OH groups were bonded, some acted only as proton donors while others were both donor and acceptor.

It can also be seen in Fig. 2 that the intensity of water been adsorbed in the amorphous regions of starch increased as the weight percentage of CMC added was increased. However, BF 3 (10% w CMC/w starch) had lower band intensity compared to BF 2 but it had the highest wavenumber at that particular band assignment regions. Besides, peak at 1231.23  $\text{cm}^{-1}$  which was assigned as C-OH bending at C-6 of glycosidic ring only can be found in BF 3. This showed that intermolecular hydrogen bond formation also occurred for BF 3. Thus, it can be stated that sample BF 3 absorbed lesser water during formulations and storage compared to other samples and exhibit stable dense biopolymers due to formation of intermolecular and intramolecular hydrogen bond in polymer chains.

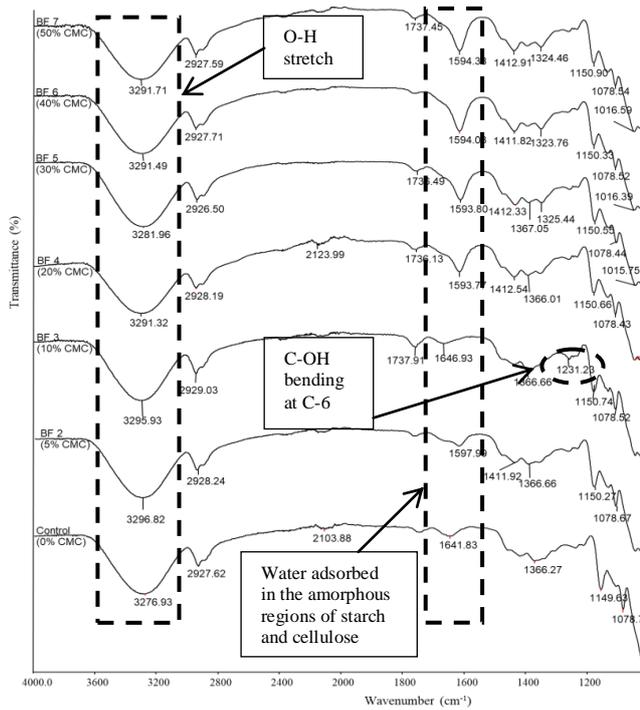


Fig. 2: FTIR spectra on the effect of CMC on cassava starch based film

### 3.3. Coating thickness analysis

The trend of changes in thickness of dried film-coated on paper as weight percentage of CMC increased is shown in Fig. 3. Altogether, it can be seen biopolymer film thickness increased with increasing CMC contents. The relationship between viscosity of film-forming solutions (Fig. 1) and biopolymer film thickness Fig. 3 is approximately linear, with the amount of CMC contents increasing up to 50% (w/w total solid). The lowest thickness of the coating film was 0.0161 mm (and the highest thickness was 0.0284 mm which had similar trend as viscosity behavior at different weight percentage of CMC. The result is in agreement with study done by Cisneros-Zevallos and Krochta [23] on dependence of thickness with viscosity of solution where authors identified that film thickness increased with viscosity of solutions.

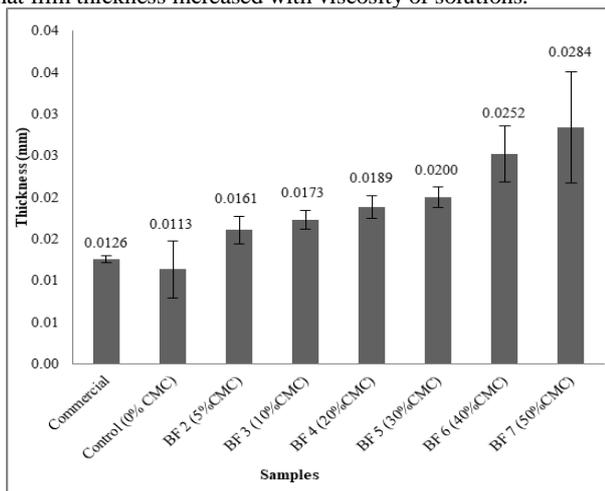


Fig. 3: Effect of CMC on coating thickness of biopolymer films coated on kraft paper

Referring to Fig. 3, coating thickness of the biopolymer film coated on the kraft paper is higher than commercial except for control samples. However, thickness of the biopolymer films also can be controlled by controlling distance of blade during casting process. In Fig. 3, it can also be seen that the weight percentage of CMC added into gelatinized cassava starch between 5 to 30% (w/w starch) are satisfied the thickness range from 0.005 to 0.02 mm as suggested in previous studies [24], [25] with thickness range from

0.0161 to 0.020 mm. From the results, it can be suggested that for industrial scale production, the thickness of the biopolymer film could be controlled by varied CMC's composition added in the formulations according to the type of applications required.

### 3.4. Tensile strength analysis

The trends for influence of CMC contents on tensile strength when the factors were varied over the range of 0% to 50% w/w of starch are shown in Fig. 4. Referring to Fig. 4, it can be seen that the weight of CMC from 0% to 10% (w/w starch) added in the formulations significantly enhanced the tensile strength with increasing value of tensile strength from 11.27 to 15.34 MPa. Meanwhile further increased in the percentage of CMC weight added up to 30% (w/w starch) reduced the tensile strength value from 15.34 to 12.57 MPa and further increase in weight of CMC added increased the tensile strength from 12.57 to 14.40 MPa. There is no obvious trend in tensile strength measurement with changes of weight percentage of CMC.

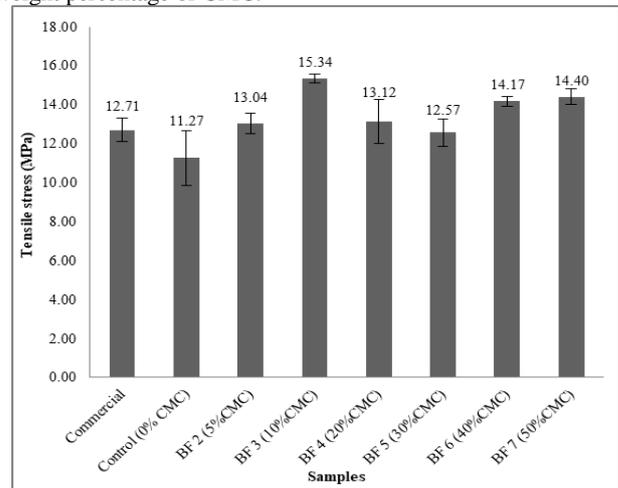


Fig. 4: Effect of CMC on tensile strength of biopolymer films coated on kraft paper

BF 3 containing 10% CMC had the highest value of tensile strength at 15.34 MPa. BF 3 exhibit stable dense biopolymers compared to other samples attributed by stable dense biopolymers due to formation of intermolecular and intramolecular hydrogen bond in polymer chains as previously discussed in FTIR analysis of the biopolymer film. Hydrogen bond in BF 3 may contribute to the strength performance of BF 3. This is in agreement with that reported by Tashiro and Kobayashi [26] where they stated that hydrogen bonds contributed about 20% the strain energy to the cellulose. Similar finding was also found in study by Fan, Dai and Huang [27]. Authors highlighted that formation of intramolecular and intermolecular hydrogen bond had significant effect on mechanical properties of polymer. This finding also supported by Hong [16] in the study on composite films by blending the corn starch with carboxymethylcellulose (CMC). He identified that the film mechanical properties had been improved twice compared the corn starch film itself. In advanced materials fields, some researchers employed FTIR analysis in conjunction with mechanical study to study the molecular response to the stress/load, such as for spruce wood and cellulose paper materials. As can be seen in Fig. 4, BF 3-coated on kraft paper has higher value of tensile stress compared to commercial coated kraft paper. Thus, it can be stated that the biopolymer film strength was comparable with commercial packaging which satisfied the main roles of food packaging to contain the food and to guarantee save delivery of food to user in great condition as suggested by previous researcher [1].

### 3.5. Wettability analysis

The trend of contact angle reductions over 120 seconds corresponds to the value obtained at increasing volume of CMC added into the formulations and the actual wettability trend for the biopolymer film is given in Fig. 5. Behavior of water droplets on surface of BF 2 (5% CMC w/w starch) and BF 3 (10% CMC w/w starch) clearly changed from control (0% CMC w/w starch) with presence of CMC. Referring to Fig. 5, only BF 2 and BF 3 exhibit hydrophobic properties as the contact angle remained higher than 90° over 120 seconds while BF 6 and BF 7 exhibit hydrophilic properties as the contact angle remained lower than 90°. As for BF 4 and BF 5, its showed hydrophobic properties only at initial time of water been dropped and it start to spread or adsorb into film with time and exhibit hydrophilic properties at the end of 120 seconds. Based on the contact angle against water on different sources, the materials were classified as hydrophilic for contact angle values lower than 90° or hydrophobic for contact angle values higher than 90° as reported by number of researchers [6], [28], [29]. This is in agreement with similar finding reported by Hong [16]. At lower content, CMC act as filler which allowed it to disperse well in starch matrix and thus improved the water barrier of film. Meanwhile, at higher content of CMC, the excessive filler allowed formation of cluster which defeated the effectiveness of CMC as filler.

However, it is noteworthy that the weight percentage of CMC added significantly change the contact angle at zero time compared to weight percentage of glycerol added into the biopolymer film. The results are in agreement with the FTIR analysis as previously discussed where the 'bound' hydroxyl group is higher as CMC was added which may attributed to the changes in contact angle. Indeed, BF 3 exhibit slower rate of water spreads or absorbed into the film-coated paper after a defined elapsed period of time compared to BF 2. This may attributed by film surface of BF 3 denser than BF 2. This study found that BF 3 exhibits the highest hydrophobic surface properties and lesser wettability when exposed to water. Therefore, BF 3 had efficient water barrier properties as the absorption was delayed as water was slowly been absorbed. Even though the commercial paper exhibit small changes in contact angle of water as time increased compared to BF 3, however BF 3 had higher contact angle for the whole time. Higher contact angle when water in contact with the solid surface showed that the surface more hydrophobic. Thus, it can be concluded that BF 3 exhibited more hydrophobic surface toward water compared to the commercial paper and it's improved the uncoated kraft paper's water barrier properties significantly.

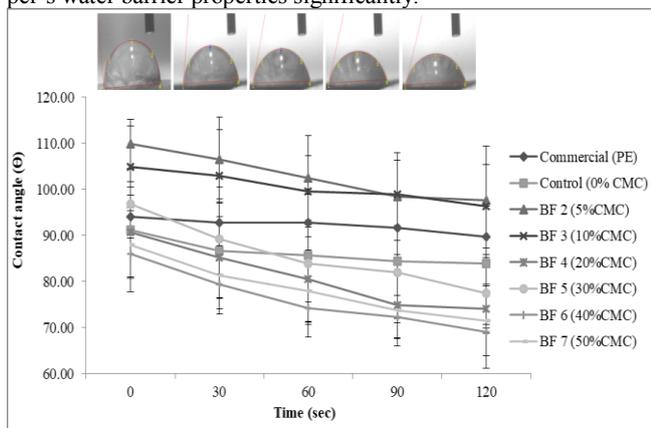


Fig. 5: Effect of CMC on wettability of biopolymer films coated on kraft paper

### 3.6. Thermogravimetric analysis

Thermogravimetric analysis (TGA) was performed on formulated biopolymer film and commercial film coated on kraft paper for food packaging where the resulting thermographs are represented in Fig. 6. The onset degradation temperature, weight loss of sam-

ple and residue of sample are summarized and compared in Table 1. In general, there is no obvious trend in degradation rate with changes of weight percentage of CMC. For all the samples included the commercial sample, the thermal evaporation temperature started at below 50°C indicating all the samples absorb a small amount of ambient moisture which in agreement with study done by Sichina [30]. This is a common phenomenon for kraft paper as its made from pulp as suggested by Beyler and Hirschler [31]. As can be seen in Table 1, the level of moisture content for BF 3 was the lowest compared to commercial and other samples which only 0.67% at onset temperature of 37°C. This may attributed by the lower intensity of water adsorbed in the amorphous regions of starch and cellulose at wavenumber 1642  $\text{cm}^{-1}$  as previously discussed in FTIR analysis.

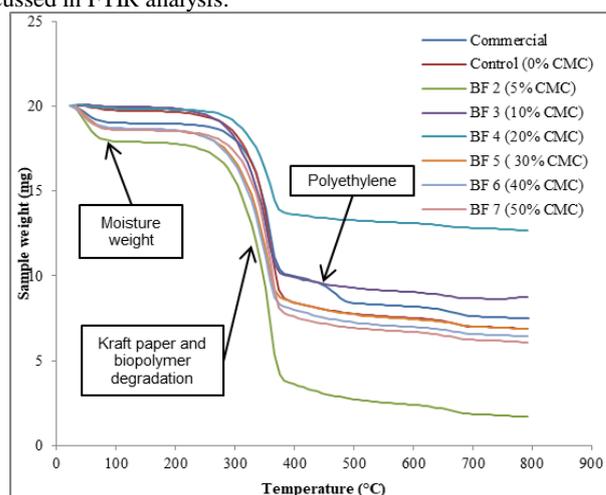


Fig. 6: TGA thermograph of commercial (PE) and biopolymer films coated on kraft paper

Table 1: Thermal Parameters of Commercial Paper Packaging in Comparison with Biopolymer Film Coated on Kraft Paper for Food Paper Packaging

Sample	Onset temperature (°C)				Weight loss (%)			
	1	2	3	4	1	2	3	4
Commercial	30	322	435	624	4.7	44.1	7.8	2.6
Control (0% CMC)	35	322	-	654	1.3	60.3	-	2.4
BF 2 (5% CMC)	48	317	-	620	10.3	73.5	-	2.6
BF 3 (10% CMC)	36	307	-	611	0.7	52.6	-	2.0
BF 4 (20% CMC)	26	320	-	593	1.2	33.1	-	1.6
BF 5 (30% CMC)	32	303	-	601	6.7	53.7	-	2.4
BF 6 (40% CMC)	21	304	-	600	6.5	57.1	-	2.3
BF 7 (50% CMC)	33	322	-	600	6.9	55.6	-	2.3

Fig. 6 shows that thermal degradation of BF 3-coated kraft paper started at 208.61°C and continued up to 700°C with two successive peaks at 307.44 and 611.56°C with weight loss of 52.6 and 2.02%, respectively. Thermal degradation of commercial (PE)-coated kraft paper started at 241.22°C and continued up to 700°C with three successive peaks which were 357.92, 471.01 and 676.52°C with weight loss of 44.06, 7.80 and 2.61%, respectively. This finding indicates that commercial, PE-coated exhibit better thermal stability than BF 3-coated on kraft paper. However, in term of disposal after been used, BF 3-coated had lower thermal breakdown points with higher weight loss than commercial paper packaging. Besides that, PE-coating of commercial packaging only degraded at onset temperature of 435.39°C. This is in agreement with study done by Sichina [30] where decomposition of polyethylene started at temperature higher than 400°C. Thus, from this study it can be concluded that BF 3 formulation can be used as alternative polymer replacing the commercial polyethylene

coating film in term of better water barrier properties, higher tensile strength and thermally degraded at lower temperature.

#### 4. Conclusion

The formulation of film coating for food paper packaging based on cassava starch was found the best at weight percentage of 10% (w/w starch). The biopolymer film coated on the Kraft paper with presence of 10% (w/w starch) of CMC had the highest tensile strength at 15 MPa. The film-coated paper also exhibits hydrophobic surface with reduction of contact angle from 104.95° to 96.33° for 120 seconds which attributed by the presence of intermolecular hydrogen bond and intramolecular hydrogen bond as confirmed by FTIR analysis at peak 3295.93 cm<sup>-1</sup> and at peak 1231.23 cm<sup>-1</sup>, respectively. Even though the PE-coated exhibited better thermal stability than the BF 3-coated, in term of disposal after being used, BF 3-coated will degrades faster than PE-coated. The advantageous of using biodegradable coated paper compared to PE-coated is that it allows degradation by environments and thus reduced the landfill depletion risks. It can be concluded that BF 3 formulation can be used as an alternative polymer coating replacing PE as it has better water barrier properties, higher tensile strength and low degradation temperature.

#### Acknowledgement

The authors would like to acknowledge Universiti Teknologi MARA for the financial support to carry out this research through 600-IRMI/DANA KCM 5/3/LESTARI (131/2017) and INQKA.

#### References

- [1] Coles, R., McDowell, D., & Kirwan, M. J. (2003). Packaging Technology Series: Food Packaging Technology. London, U.K.: Blackwell Publishing.
- [2] Marsh, K., & Bugusu, B. (2007). Food Packaging-Roles, Materials, and Environmental Issues. *Packaging, Food, and Environmental Issues*, 72(3), 39–56.
- [3] Sung, S., Sin, L., Bee, S., et al. (2013). Antimicrobial agents for food packaging applications. *Trends in Food Science and Technology*, 1–14.
- [4] Kanmani, P., & Rhim, J. (2014). Development and characterization of carrageenan / grapefruit seed extract composite films for active packaging. *International Journal of Biological Macromolecules*, 68, 258–266.
- [5] M. Chaplin, "Sources for starch," *Water Structure and Science*, 2018. [Online]. Available: <http://www1.lsbu.ac.uk/water/starch.html>.
- [6] Skurtys, O., Acevedo, C., Pedreschi, F., Enrione, J., Osorio, F., & Aguilera, J. (2010). Food hydrocolloid edible films and coatings. *Science & Engineering*, 318-380.
- [7] Nieto, M. B. (2009). Edible films and coatings for food applications: Structure and function of polysaccharide gum-based edible films and coatings. Springer. New York.
- [8] Lian, T. S., & Idris, K. (2000). *Present situation and future potential of cassava in Malaysia*. Kuala Lumpur, Malaysia.
- [9] Parra, D., Tadini, C., Ponce, P., & Lugao, A. (2004). Mechanical properties and water vapor transmission in some blends of cassava starch edible films. *Carbohydrate Polymers*, 58(4), 475–481.
- [10] Maran, J. P. P., Sivakumar, V., Sridhar, R., & Thirugnanasambandham, K. (2013). Development of model for barrier and optical properties of tapioca starch based edible films. *Carbohydrate Polymers*, 92(2), 1335–1347.
- [11] Ghanbarzadeh, B., Almasi, H., & Entezami, A. A. (2010). Physical properties of edible modified starch/carboxymethyl cellulose films. *Innovative Food Science and Emerging Technologies*, 11(4), 697–702.
- [12] Fennema, O. R., Kamper, S. L., & Kester, J. J. (1990). *U.S. Patent No. 4,915-971*. Wisconsin Alumni Research Foundation, Madison, Wis.
- [13] Rastogi, V. K., & Samyn, P. (2015). Bio-based coatings for paper applications. *Coatings*, 5, 887–930.
- [14] Mali, S., Sakanaka, L. S., Yamashita, F., & Grossmann, M. V. E. (2005). Water sorption and mechanical properties of cassava starch films and their relation to plasticizing effect. *Carbohydrate Polymers*, 60(3), 283–289.
- [15] Alves, V., Costa, N., Hilliou, L., Larotonda, F., Goncalves, M., & Sereno, A. (2006). Design of biodegradable composite films for food packaging. *Desalination*, 1, 331–333.
- [16] Hong, K. M. (2013). *Preparation and characterization of carboxymethylcellulose from sugarcane bagasse* (Degree Thesis). Universiti Tunku Abdul Rahman, Kuala Lumpur, Malaysia.
- [17] Suderman, N., Isa, M. I. N., & Sarbon, N. M. (2016). Effect of drying temperature on the functional properties of biodegradable CMC-based film for potential food packaging. *International Food Research Journal*, 23(3), 1075–1084.
- [18] Tabari, M. (2017). Effect of carboxymethyl cellulose on the mechanical and barrier properties of sago starch based films," *Preprints*, 1, 1–7.
- [19] Aqualon. (1999). Physical and chemical properties of sodium carboxymethylcellulose. *Hercules Incorporated*, 6.
- [20] Liu, Y., Cai, Y., Jiang, X., Wu, J., & Le, X. (2016). Molecular interactions, characterization and antimicrobial activity of curcumin-chitosan blend films. *Food Hydrocolloids*, 52, 564–572.
- [21] Shogren, R. L. (1999). Preparation and characterization of a biodegradable mulch: paper coated with polymerized vegetable oils. *Journal of Applied Polymer Science*, 73, 2159–2167.
- [22] Turhan, K. N., Şahbaz, F., & Güner, A. (2001). A spectrophotometric study of hydrogen bonding in methylcellulose-based edible films plasticized by polyethylene glycol. *Journal of Food Science*, 66(1), 59–62.
- [23] Cisneros-Zevallos, L., & Krochta, J. M. (2003). Dependence of coating thickness on viscosity of coating solution applied to fruits and vegetables by dipping method. *Journal of Food Science*, 68(2), 503–510.
- [24] Battersby, G. C., Santos, S. A., Zhong, B., & Gan, X. (2015). *U.S. Patent No. 9102125 B2*. Mantrose-Hauser Co., Inc.
- [25] Paunonen, S. (2010). *Influence of moisture on the performance of polyethylene coated solid fiberboard and boxes* (PhD Thesis). Norwegian University of Science and Technology, Trondheim, Norway.
- [26] Tashiro, K., & Kobayashi, M. (1991). Theoretical evaluation of three-dimensional elastic constants of native and regenerated cellulose: Role of hydrogen bonds. *Polymer*, 32(8), 1516–1526.
- [27] Fan, M., Dai, D., & Huang, B. (2012). Fourier Transform Infrared Spectroscopy for natural fibres. *Fourier Transform Materials Analysis*, 45–68.
- [28] Balan, T., Guezennec, C., Nicu, R., Ciolacu, F., & Bobu, E. (2015). Improving barrier and strength properties of paper by multi-layer coating with bio-based additives. *Cellulose Chemistry and Technology*, 49, 607–615.
- [29] Basiak, E., Debeaufort, F., & Lenart, A. (2016). Effect of oil lamination between plasticized starch layers on film properties. *Food Chemistry*, 195, 56–63.
- [30] Sichina, W. J. (2010). Characterization of polymers using TGA. *Thermal Analysis Perkin Elmer Instrument*, 1–5.
- [31] Beyler, C. L., & Hirschler, M. M. (2010). Thermal decomposition of polymers. *SFPE Handbook of Fire Protection Engineering*, 110–131.