INVESTIGATION ON PROPERTIES OF COMPOSITE FILM USING CARBOXYMETHYLCELLULOSE

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A project report submitted in partial fulfilment of the requirements for the award of Bachelor of Engineering (Honours) Petrochemical Engineering

Faculty of Engineering and Green Technology
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DECLARATION

I hereby declare that this project report is based on my original work except for citations and quotations which have been duly acknowledged. I also declare that it has not been previously and concurrently submitted for any other degree or award at UTAR or other institutions.

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APPROVAL FOR SUBMISSION

I certify that this project report entitled **"INVESTIGATION ON PROPERTIES OF COMPOSITE FILM USING CARBOXYMETHYLCELLULOSE"** was prepared by **LOH WIN KEI** has met the required standard for submission in partial fulfilment of the requirements for the award of Bachelor of Engineering (Honours) Petrochemical Engineering at Universiti Tunku Abdul Rahman.

Approved by,

Signature : ____ _________

Supervisor : Dr. Chng Lee Muei

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ABSTRACT

Recently, researchers have been focusing on the development of sustainable packaging to minimize the environmental issues created by conventional packaging. In this study, Carboxymethylcellulose (CMC), is used as an organic matrix for the formation of biocomposite film, one of the potential sustainable packaging due to its good film-forming ability, biodegradable and low-cost. With this purpose, the biocomposite film was prepared with nanomer PGV as reinforcement inorganic filler to increase the stiffness and strength of a polymer matrix and PEG as plasticizer mainly to improve the flexibility and the water vapor barrier properties of the biocomposite films. These all ingredients were mixed at various content (CMC content: 1 g, 2 g and 3 g, PEG content: 0.5 g, 0.6 g and 0.7 g, PGV content: 0.1 g, 0.2 g and 0.3 g) to form homogeneous solution before the film is formed via a simple casting method. The properties of the film were then characterized with Fourier Transform Infrared Spectroscopy (FTIR) analysis, Thermogravimetric analysis (TGA), tensile test and water solubility test.

Results from the analysis show that the increase of CMC content improve the thermal stability and mechanical properties of the film. Besides, the effect of increasing PEG content on the mechanical properties, thermal stability and water solubility were not so significant. Apart from that, the addition of PGV content could improve the water resistance of the film as PGV nanoclay acts as a filler and physical blockage for water resistance. Increasing PGV content also improve the mechanical properties due to the presence of hydroxyl group in both CMC and PGV could form hydrogen bond between CMC and PGV. The optimum film was formed with a mixture of 3 g of CMC, 0.6 g of PEG and 0.2 g PGV, where the tensile strength of

1431 MPa, elongation at break of 27.3%, residue percentage of 33.3076% after third step thermal decomposition and took 17 minutes to dissolve in water. Compared to the commercial PE film with tensile strength of 126 MPa, elongation at break of 432.25%, residue percentage of 17.2132% after third step thermal decomposition and took more than 24 hours to dissolve in water. From the comparison between 3CMC-0.6PEG-0.2PGV film and commercial PE film, it could be observed that 3CMC-0.6PEG-0.2PGV film are quite weak in terms of elongation at break and water resistance. Findings of this study provide a significant and clear direction for development of sustainable packaging for commercialization to fulfill future market demand.

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LIST OF SYMBOLS / ABBREVIATIONS

ATR-FTIR	Attenuated Total Reflectance-Fourier Transform Infrared Spectroscopy
CMC	Carboxymethylcellulose
DP	Degree of Polymerization
DSC	Diffential Scanning Calorimetry
FE-SEM	Field-Emission Scanning Electron Microscopy
MMT	Montmorillonite
PBS	Polybutylene Succinate
PEG	Polyethylene Glycol
PHA	Polyhydroxy Acids
PLA	Polylactic Acid
PVA	Polyvinyl Alcohol
SEM	Scanning Electron Microscope
TGA	Thermogravimetric Analysis
UTS	Ultimate Tensile Strength
WVP	Water Vapor Permeability
XRD	X-Ray Diffraction

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CHAPTER 1

INTRODUCTION

1.1 Background of Study

Carboxymethylcellulose (CMC) is an anionic and water-soluble cellulose derivative with carboxymethyl substituents ($-CH_2COOH$) bound to some of the hydroxyl groups of cellulose. The molecular structure of CMC is shown in **Figure 1.1**, where R is the carboxymethyl group. Due to the highly hygroscopic nature of CMC, it is soluble in water at any temperature. However, the water solubility of CMC would affect by three factors which are the degree of polymerization (DP), degree of substitution and the uniformity of substitution distribution. In particular, decrease in DP and increase in both carboxymethyl substitution and substitution uniformity would result in high solubility of CMC in water (Ergun, Guo and Huebner, 2016).

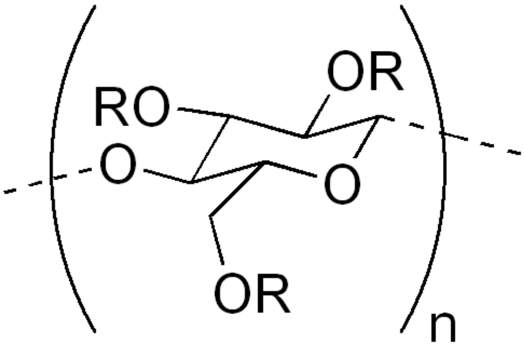

Figure 1.1: Molecular structure of CMC (John, 2014).

According to Grand View Research (2017), plastic films and sheets have attained significant popularity in applications across various industries. The global demand for plastic films and sheets in 2016 was 46,343.9 kilotons and it is estimated to be rising at a rate of over 3% every year due to the increasing application and popularity of plastic films (Smithers, 2021). However, most of the conventional plastic films in the market are derived from petroleum, which petroleum is a non-renewable resource. In addition, since the plastic films are non-biodegradable, a large amount of waste will be created from the high usage of plastic film.

Along with the recent growth in environmental awareness, bioplastic has been widely used in various industrial applications to replace conventional plastic. This is because bioplastics which are made from bio-based materials are non-toxic and can be degraded in a certain period of time. Thus, using bioplastics not only can protect the environment, but it helps to reduce environmental issues by reducing waste and limiting the use of non-renewable resources. Apart from packaging films, bioplastics also become a necessity in many industrial applications including agriculture, biomedical, structural, electrical, and other consumer products (Ashter, 2016).

Generally, biodegradable film materials can be divided into four categories which are polysaccharides, proteins, lipids, and resins. CMC, which is cellulose derivative is one of the examples of natural polysaccharides and it is the most highly available and renewable biopolymer as compared to other natural polysaccharides including starch, chitosan, pectin and alginate (Niladri, et al., 2012). The additional properties of CMC such as good film-forming ability, biodegradability, non-toxicity, and low price have made it important to be applied in packaging film (Inyoung, et al., 2017). Other than that, adding CMC to the biodegradable composite film formulation could improve the film network structure which subsequently enhance the film strength and the gas barrier property of the film (Inyoung, et al., 2017).

Apart from biodegradable packaging film application, CMC is also widely applied in various field to improve product quality and processing properties owing to its hydrophilic character, high viscosity, good adhesive performance and good film-forming ability. **Figure 1.2** shows the application of CMC in various industries.

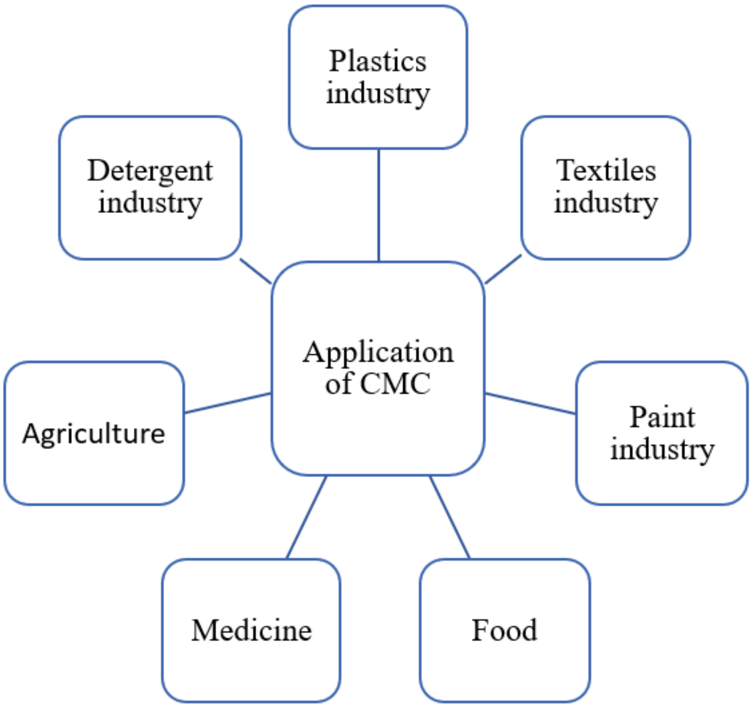

Figure 1.2: Application of CMC.

1.2 Problem Statement

The use of petroleum-derived plastics has created a lot of issues for the environment and even wildlife. By the end of the 20th century, plastics were known as one of the persistent polluters of many environmental niches due to their non-biodegradable properties. This property of plastics has been contributed to the accumulation of plastic waste in landfills and aquatic lives could be killed when plastic was dumped into the river causing the aquatic lives mistakenly consume plastics debris for food (Moore, 2021). Although the plastic pollution can be controlled via reuse and recycle, the efficiency is still very low as the rate of plastics recovery is slower as compared to plastics waste generation (Liu, 2006). Therefore, using bioplastics to replace the conventional plastics is one of the effective methods to reduce the environmental problems because bioplastics could be degraded by microorganisms after a certain period. Before looking into the properties of the film, in order to convince people to replace conventional plastic with bioplastic, the cost of bioplastics should not be higher than the conventional plastic. CMC, which is a material with low cost, widely available with biodegradable properties is very suitable for the purpose to develop sustainable packaging (Yaradoddi, et al., 2020).

Although CMC has good film-forming ability and other advantages such as being non-toxic, inexpensive, and abundant in nature, CMC itself is not suitable to produce biodegradable film due to its hydrophilic nature and highly crystalline structure properties. In particular, the hydrophilic nature of CMC results in the film having poor moisture-barrier properties, whereas the highly crystalline structure of CMC makes the film become brittle and low flexibility (Liu, 2006). To overcome the inherent properties of CMC as well as to reduce the environmental issues given by the conventional plastics, this research will be focusing on the synthesis of composite film with CMC as the host material.

The concentration of CMC in the film could greatly influence the tensile strength and elongation at break of the composite film. As per report from Tongdeesoontorn, et al. (2011), the moisture sorption and tensile strength of the starch-CMC composite film increased with the increase of CMC concentration, whereas the elongation at break of the film decreased as the CMC concentration increase. However, the maximum tensile strength of the film is only found at optimum level of CMC concentration, where an excess amount of CMC will result in decrease in tensile strength. In addition, according to the research by Kapil, et al. (2019), increasing the CMC content could increase the thermal stability of the composite owing to the stable properties of CMC.

The characteristics and properties of the composite film are important as there are some specific properties are required for different types of applications. The most common properties of the composite film are barriers to moisture, high tensile strength, high elasticity, biodegradability and stability. For example, one of the important properties of edible packaging film is permeability. Since the microbial growth problem will happen on most of the food with high moisture content, edible packaging film should have a high barrier to moisture to inhibit the moisture change between the food and environment, at the same time prevent the deterioration of food. The incorporation of MMT such as PGV nanoclay into CMC matrix could improve the water vapor barrier property of the film. This is because clay acts as filler to fill the void space and become physical blockage to increase the water resistance of the film (He, Fei, and Li, 2019).

Apart from that, the mechanical property of the film is important as well. In particular, a composite film with high tensile strength is not easy to brittle or crack, which allow it to be applied in wide application and it can protect the goods to be packed as well (Theeranun and John, 2010). Other than increasing the CMC content, another approaches to increase the film tensile properties is the incorporation of nanoclay into the composite film, as nanoclay induced the chemical interaction between the clay particle and polymer chains to improve the tensile properties (He, Fei, and Li., 2019; de Melo Fiori, et al., 2019,).

Therefore, the formulation of a mixture for the formation of composite film that is comparable with conventional plastics such as high tensile strength and elongation at break as well as a high barrier to moisture is vital to determine its usage to diminish the plastic waste to solve the environmental issues. However, the information on the formulation using CMC as host material is scarce for commercialization, projecting the importance of this study.

1.3 Objectives

The focus of the study is to achieve the following objectives:

i. To characterize the carboxymethylcellulose, nanoclay PGV and polyethylene glycol.
ii. To investigate the effect of mixture formulation on the properties of biocomposite film.
iii. To compare the biocomposite film and commercial plastic film.

1.4 Scope Study

i. The characteristic of carboxymethylcellulose, nanoclay PGV and polyethylene glycol were analyzed.
ii. The composite films are prepared using different compositions of CMC, nanoclay PGV and plasticizer PEG to investigate the effect of mixture formulation on properties of biocomposite film. By varying the compositions, it may result in different properties of the biocomposite film.
iii. Then, the properties of the synthesized biocomposite film are determined using Fourier transform infrared spectroscopy (FT-IR), tensile strength tester, thermogravimetric analysis (TGA) and water solubility.
iv. The properties of a commercial film are analyzed with FTIR, tensile strength tester, TGA and water solubility. The results are then compared with the synthesized biocomposite film.

CHAPTER 2

LITERATURE REVIEW

2.1 Properties and Applications of CMC

CMC is a cellulose derivative that is produced from the reaction of monochloroacetic acid on alkali cellulose. It is generally used in the form of sodium salt (cellulose gum) which is present in white, granular, odourless, and tasteless free-flowing powder (Hollabaugh, H. Burt, and Anna, 1945). CMC has wide acceptance in the industry due to its high viscosity, non-allergenic, non-toxic, good adhesive, and good film-forming characteristics. The numerous hydroxyl and carboxylic groups that are presented in CMC enable it to have the ability to absorb moisture and be soluble in water easily (Ali, et al., 2018). In particular, CMC soluble in water would form high viscous solutions which the solution can be used as a thickener, suspender, and stabilizer in the industry (Hollabaugh, H. Burt, and Anna, 1945). **Table 2.1** listed the application of CMC and its roles in the industry. Apart from the application in **Table 2.1**, CMC is currently used by the researcher to synthesize biocomposite film due to its polymeric structure and high molecular weight (Ali, et al., 2018).

Table 2.1: The roles of CMC in a different application.

Application	Description
Construction industry	CMC acts as a stabilizer as well as a hydrophilic agent in cement and building materials. It helps the sand to disperse well in the cement and intensifies its adhesive action (Marketizer.com, 2012).
Detergent industry	CMC acts as an inhibitor to prevent the grease to redeposit on the fabric after the grease is removed by the detergent (Patel Industries, n.d.).
Paper industry	CMC is a coating on paper to make the surface of the paper smooth and shine. As a result, it gives a lot of advantages such as reducing the consumption of printing ink, improving the color of the paper and improving the resistant to grease on paper (Jining Fortune Biotech, 2016).
Agriculture	CMC acts as a suspending agent in pesticides to remain the pesticides mixture in suspension in an extended period of time. CMC in pesticides also acts as a glue to attach the insecticide to the leaves of plants (Marketizer.com, 2012).
Cosmetics industry	CMC acts as a thickener, stabilizer and suspending agent in dental impression materials, toothpastes, creams, lotions, shampoos, and other hair care products (Marketizer.com, 2012).
Paint industry	CMC acts as a thickener and suspending agent in oil paints and varnishes to ensure the pigment is suspended in the fluid (Sinocmc, 2019).
Oil industry	Crude or purified CMC acts as a colloid thickener in removing the drill from the hole to avoid sediments (Celsol.co, n.d.).
Plastics industry	CMC is used to increase the viscosity of plastics (latex) in the plastics industry (Marketizer.com, 2012).
Textiles industry	Crude CMC acts as a sizing agent in the textiles industry to protect the yarn and give better fabric. It also acts as a thickening agent in textile varnishes (Sinocmc, 2019).

Pharmaceuticals	The high purity and low viscosity of CMC act as a coating on tablets. Owing to its gel-forming ability, it is used to transport the drug and disintegrate tablets. Next, it also acts as a stabilizer for suspensions, emulsions, sprays, and bio-adhesive tablets which attach internally to the mucus of a body part (Pacific Biolabs, n.d.).
Food	CMC acts as a stabilizer in bakery products, suspending agent in fruit juices, a protective agent to cover the surface of fruits as well as an auxiliary agent for gel formation in gelatines and dairy products (Vassilis, 2021).
Medicine	Owing to its gel-forming ability, CMC is used in thorax operations to prevent air leaks and fluid ingress in the lungs. It is also used in the field of orthopedics to lubricate the joints of the bones by injection (Pacific Biolabs, n.d.).

2.2 CMC Films Drawbacks and its Improvements

CMC films which is polysaccharide-based film are transparent and resistant to oils and fats; however, the hydrophilic nature of the cellulose derivative films has resulted in low water vapor barrier properties in the CMC films, which subsequently limited the usage of the film due to poor mechanical properties. In addition, CMC films are tough which has caused the films to be brittleness. Another drawback of the CMC films without any additives is their vulnerability to degradation since CMC is derived from natural polysaccharides (Nanou, et al., 2013).

Despite the drawbacks of the CMC films as mentioned above, the performance of the CMC films can be improved by adding film additives such as plasticizers. Plasticizers are small-molecular-weight hydrophilic agents which can improve the flexibility of the film. The plasticizers work by locating themselves in the polymeric network and then competing for chain-to-chain H-bonding along the polymer chains. Generally, plasticizers can be divided into three types which are mono-, di-, or oligosaccharides, polyols, as well as lipids and derivatives (Theeranun and John, 2010). Among all the plasticizers, glycerol is one of the most well-known plasticizers in film-making due to its compatibility and stability with hydrophilic bio-polymeric packaging chains (Ali, et al., 2018).

Apart from that, the properties of CMC films can be improved by blending CMC with other biopolymers to make composite films. This is because the films made from blends of biopolymers generally exhibit more excellent properties than the films that are made from biopolymers alone (Jufang, et al., 2019). Composite films enable the film to combine the properties of the polymers. In other words, the aim of the composite film is to take advantage of the complementary functional properties of other polymers to overcome the limitations of CMC (Theeranun and John, 2010).

2.3 Properties of Additives in Biocomposite Film

As discussed above, pure CMC film is low resistance to water due to its hydrophilic nature. In the aspect of mechanical properties, pure CMC film is tough and brittleness which is not suitable for any application. Thus, incorporating other biopolymers or adding additives are very important to improve the water barrier properties and mechanical properties of CMC film. **Table 2.2** shows the properties of some biopolymers and additives that are being used to enhance the properties in CMC film.

Table 2.2: Properties and functions of biopolymers and additives used in CMC film formation.

Biopolymer/ Additives	Properties	How It Function
Polyvinyl alcohol (PVA)	• Water-soluble • High tensile strength • Good chemical resistance • Biodegradability • Good gas barrier	The reaction of CMC and PVA is usually enhanced by adding citric acid. Citric acid is a natural organic acid with one hydroxyl group and three carboxyl groups which enable it to be used as cross-linking agent. When the cross-linking happened between CMC and PVA, the covalent bonds hold them together and result in the improvement of the mechanical properties of the film (Kapil, et al., 2019).
Chitosan	• Good film-forming ability • Biodegradability • Nontoxicity • Perform antimicrobial properties • Insoluble in water	Chitosan is always being used to produce composite film due to its antimicrobial properties of chitosan. The chitosan-based film is often applied in the food packaging industry to extend the shelf life of the food. Basically, the mechanisms of antimicrobial in chitosan involve three steps. First, the positive charge of chitosan having electrostatic interaction with the negative charge of microbial cell membranes results in cell leakage. Then, chitosan penetrates into the microorganism nuclei and inhibit the synthesis of mRNA and protein. Lastly, chitosan forms an external barrier

		which the barrier blocks the nutrients for the growth of microorganism (Goy, Britto and Assis, 2009).
Starch	• Good film-forming ability • Biodegradability • Low water resistance • Poor mechanical properties	Similar to PVA as mentioned above, reaction of CMC and starch can be enhanced by adding citric acid as cross-linking agent. During the cross-linking process, the covalent bonds hold the CMC chains and starch chains together and resulted in the improvement of film mechanical properties (Kapil, et al., 2019).
Gelatin	• Good film-forming ability • Biodegradability • Poor mechanical properties • Low water-vapour barrier	When gelatin incorporates into the CMC film, cross-linking will happen between gelatin chains and CMC chains. While in the presence of cross-linking agents, the cross-linking between the biopolymer chains can be increased and hence enhance the mechanical properties of the biocomposite film (Theeranum, et al., 2010).
Clay mineral	• Good thermal stability • Able to withstand high stress • Able to swell in water	The incorporation of clays into CMC could improve the mechanical properties of the composite film. This is because clay minerals act as fillers in the CMC composite film, a strong interaction between the clay mineral and hydroxyl groups of CMC are formed after the evaporation of the solvent (De Oliveira, et al., 2015).

Plasticizer	• Low molecular weight • Biodegradable • Nontoxic • Water-soluble	A plasticizer is a low molecular weight and polar in nature. When mixing with the biopolymer, the plasticizer located itself between the polymer chains and compete for chain-to-chain H-bonding. As a result, plasticizers improve the flexibility of the film (GODWIN, 2000).
Antioxidants	• Able to slower the oxidation reaction of the film	Two types of antioxidants can be used in film formation which are primary and secondary antioxidants to slow the oxidation reaction of the film. Primary antioxidants act as free radical acceptors to delay the process of initiation of autooxidation. Whereas secondary antioxidants prevent the happening of oxidation by capturing and deactivating oxygen, absorbing UV radiation, and decomposing hydroperoxides to non-radical species (Theeranum, et al., 2010).

2.3.1 CMC-PVA Composite Film

Polyvinyl alcohol (PVA) is one of the water-soluble synthetic polymers that has been highly used by researchers to blend with CMC to synthesize composite film. The properties of PVA such as high tensile strength, chemical resistance, biodegradability, biocompatibility, excellent adhesive, and gas barrier have made it become the best option in composite film (Kapil, et al., 2019). Addition of PVA to CMC film formulation can greatly improve the mechanical properties of the film.

El-Sayed, et al. (2011) had prepared the PVA-CMC composite films by casting method and the properties of the film such as compatibility and thermal stability were studied. The differential scanning calorimetry (DSC) result showed that the decrease in PVA composition caused a rapid decrease in degree of crystallinity structure in the film. It also showed that the CMC and PVA were compatible due to the presence of –OH and CH_2OCH_2COONa in CMC and the presence of –OH groups in PVA capable of hydrogen bonding. Besides, the TGA analysis result showed that the thermal stability of the PVA-CMC composite film increased with the CMC composition, which the degradation temperature was higher as compared to pure CMC film (El-Sayed, et al., 2011).

Tajeddin, et al. (2016) studied the properties of CMC-PVA composite film with glycerol as plasticizer. The various formulations of nanocomposite films were prepared by casting method and their mechanical properties were studied. Tajeddin, et al. (2016) reported that the role of CMC in this composite film was to improve the mechanical and barrier properties of the starch-based film. Adding PVA could significantly help to increase the resistance to water and improve the tensile strength. While the glycerol acted as plasticizers due to its stability and compatibility with the hydrophilic nature of the biopolymer chains.

The X-ray diffraction (XRD) result showed that there was no peak for clay, which means that the clay minerals were well distributed throughout the matrix. Next, as the clay content increased from 0.5% to 3%, the tensile strength of the CMC-PVA-clay composite film increased from 14 MPa to 25.01 MPa, whereas the modulus of the film increased from 96.14 MPa to 184.14 MPa. This indicated that both tensile strength and modulus of the composite film increased with the clay content. However, the elongation at break of the film decreased from 17.87 MPa to 6.85 MPa as the clay content increased from 0.5% to 3% indicating that the addition of clay to the film lead to low ductility and brittleness of the composite film. Other than that, the water absorption of pure CMC-PVA was 22.03% and it was decreased to 20.75%, 19.04% and 16.95% as the clay content increased from 0.5%, 1% and 3%. Hence, the addition of clay minerals as fillers resulted in the reduction of permeability of the films (Tajeddin, et al., 2016).

PVA-CMC composite film is actively used in food packaging. For example, PVA-CMC films with clove oil which were produced by Muppalla, et al. (2014) were used as meat products packaging. Muppalla, et al. (2014) reported that increase in PVA concentration resulted in the increase of tensile strength and moisture barrier of the film, which these improvements enable the films to be stored in the refrigerator for a longer time. Moreover, adding additives such as antimicrobial and antioxidant agents enable the foods to be stored longer by preventing the growth of fungal.

2.3.2 CMC-Chitosan Composite Film

Chitosan is a biopolymer that is produced by the fusion of chitin with alkaline. Chitosan is the second most abundant material in nature subsequent to cellulose (Thomas, et al., 2016). It has the good film-forming ability and other excellent properties such as biocompatibility, biodegradability and nontoxicity (Jahit, et al., 2016).

Thomas, et al. (2016) studied the thermal stabilities of chitosan-CMC composite film with glycerol as plasticizer. The composite film was prepared with chitosan, CMC and glycerol cross-linked with formaldehyde. Thermogravimetric analysis was carried out to measure the thermal weight loss of the chitosan-CMC composite film at a heating rate of $10℃\ \mathrm{min}^{-1}$ in nitrogen atmosphere. The result showed that chitosan/CMC composite film has better thermal stability.

Jannatyha, et al. (2020) carried out an investigation on nanochitosan-CMC composite film that was prepared by casting method. Addition of nanomaterials to the composite film is one of the methods to improve the properties of the film because the high surface area of nanomaterials could improve water resistance and mechanical properties of nanocomposite films. The XRD result showed that the crystallinity of composite film was higher at high concentration of nanochitosan, while the pure CMC film without the addition of nanochitosan showed a more amorphous structure. Besides, the thickness of the film increased with the concentration of nanochitosan due to the enhancement in solid volume, where the thickness increased from 0.013 mm to 0.017 mm as the concentration of nanochitosan increased from 0.1% to 1%. Jannatyha, et al. (2020) reported that the tensile strength of the nanochitosan-CMC composite film was increased from 8.41 MPa to 18.03 MPa when the content of nanochitosan increased from 0.1% to 0.5%. However, the tensile strength decreased from 18.03 MPa to 11.34 MPa when the concentration of nanochitosan increased from 0.5% to 1.0%. According to Jannatyha, et al. (2020), the decrease of tensile strength at concentration 1.0% of nanochitosan may be due to the self-agglomeration of nanochitosan. Despite there was decrease of tensile strength at concentration of 1.0% of nanochitosan, in overall, the tensile strength of nanochitosan-CMC composite film was still higher than the pure CMC film.

Next, the elongation at break of the nanochitosan-CMC composite film decreased from 140.78% to 28.08% and then increased to 71.25% as the concentration of nanochitosan increased from 0.1% to 0.5% and 1.0%. As compared to pure CMC film which has the elongation at break of 210.73%, the nanochitosan-CMC composite films with three different concentrations of nanochitosan were considered to have lower percentage of elongation at break. On the other hand, the water vapor permeability (WVP) decreased from 0.45 to 0.17 $g\ s^{-1}m^{-1}Pa^{-1} \times 10^{-10}$ as the concentration of nanochitosan increased from 0.1 % to 1%, this indicated that the addition of nanochitosan could significantly help to decrease the WVP and increase the water resistance of the film. Besides, nanochitosan-CMC composite film showed antimicrobial properties, and the antimicrobial activities were stronger at higher concentration of nanochitosan.

As per report from Theeranum, et al. (2010), the use of chitosan coatings could keep the freshness of the fruit for a long time after harvesting as well as delay enzymatic browning in fresh products with a negative charge such as fats, cholesterol, basic ions, and proteins. This is because the cationic property of chitosan could react with negative charge of substances and provide antimicrobial and antioxidant properties. The antibacterial properties of chitosan-CMC composite film have made it suitable to use as food packaging which can decrease the rate of deterioration of the food by inhibiting the growth of microorganisms on the food surfaces (Luyu, et al., 2021). Other than that, chitosan-CMC composite film can be used for biomedical devices due to the effect of antimicrobial in chitosan (Aliya, et al., 2017).

2.3.3 CMC-Starch Composite Film

Starch is one of the popular materials in bioplastic film production due to its abundance in nature, excellent film-forming abilities, biodegradability, and most importantly is a renewable resource. Starch is always used to blend with other materials in bioplastic film production because the film produced by starch alone exhibits several limitations such as high-water sensitivity and poor mechanical properties. These disadvantages have limited the usage of starch film for packaging purposes (Ghanbarzadeh, et al., 2010).

Ghanbarzadeh, et al. (2010) had prepared the corn starch-CMC composite films with glycerol as plasticizer and citric acid as additive. The physical and mechanical properties of the film were studied. The use of plasticizers could help to increase the flexibility and overcome the brittleness of the film. The citric acid is a main organic acid with one hydroxyl group and three carboxyl groups. The addition of citric acid could improve the water resistance and mechanical properties of the film due to the multi-carboxylic structure of citric acid. It also helped to prevent recrystallization and retrogradation during the film forming process due to the formation of stronger hydrogen bonds with the hydroxyl groups of starch molecules. While blending the CMC with corn starch could improve the mechanical and barrier properties of the starch-based film due to the chemical similarities of starch and fibres. Ghanbarzadeh, et al. (2010) reported that the water vapor permeability (WVP) of the control film without CMC was $2.62 \times 10^{-7}\ \mathrm{g\,Pa^{-1}h^{-1}m^{-1}}$, the WVP value started to decrease to 2.45×10^{-7} and 2.34 $\times\ 10^{-7}\ \mathrm{g\,Pa^{-1}h^{-1}m^{-1}}$ as the CMC content increased from 10% to 15%. This was because the corn starch formed hydrogen bonds with the hydroxyl and carboxyl groups of the CMC macromolecules, and the diffusion of water molecules through the film could be reduced by this strong structure.

Next, for the solubility property, Ghanbarzadeh, et al. (2010) reported that the water solubility of the composite film decreased from 21.33% to 17.65% as the CMC content increased from 5% to 20%. This indicated that as compared to the starch film without CMC that had a water solubility of 23.76%, the addition of CMC at high level to the starch film could decrease the water solubility of the film where the percentage of water solubility of the starch-CMC composite films were lower than the starch film without CMC. Besides, the optical properties of the film increased with the CMC content, and the whiteness index of the film increased by increasing the CMC content. For the tensile properties, an increase in CMC content resulted in an increase of ultimate tensile strength (UTS) of the film. The UTS of the composite film increased from 6.57 MPa to 16.22 MPa as the CMC content increased from 0% to 20%. Ghanbarzadeh, et al. (2010) also reported that the addition of CMC to the starch film could improve the film's strength without significantly affecting the flexibility of the film.

Tongdeesoontorn, et al. (2011) studied the effect of CMC concentration on physical properties of cassava starch-CMC film. The SEM results showed that the morphology of CMC-starch biocomposite film was similar to the pure CMC film and pure starch film, in which three of the films showed a dense and smooth cross-section that indicated a homogeneous structure. While the FTIR results showed that the absorption in the $\mathrm{COO^-}$ band of CMC-starch biocomposite film increased at around $1592\ \mathrm{cm^{-1}}$, indicating that the antisymmetric and symmetric vibrations of C=O and C-O bonds were enhanced due to the disruption of intermolecular H-bonds between carboxylic groups of CMC and starch. Besides, Tongdeesoontorn, et al. (2011) reported that the increase in CMC concentration resulted the increase in tensile strength but decrease in elongation at break of the film. It was reported that the film containing 30% of CMC performed better physical properties as compared to pure cassava film and pure CMC film. Besides, the water solubility of the film decreased with the increase of CMC concentration. Tongdeesoontorn, et al. (2011) explained that the decrease in solubility of the film might be due to the strong bond formed by the hydroxyl group and carboxyl group on CMC with the hydroxyl groups on starch. Hence, the strong bonds between the CMC and starch improved the interactions between molecules and subsequently resulted in the improvement in cohesiveness of the biopolymer matrix and water resistance of the composite film.

CMC-starch biocomposite film is currently used as edible film. The edible film is coated on the food to prevent water loss and protect it against adverse effects. As per report from Siying, et al. (2020), a previous study showed that CMC-based edible films are suitable to be used as carriers for some probiotic strains which can significantly protect the oxidation-sensitive foods by maintaining the quality of packaged foods and extending their shelf life.

2.3.4 CMC-Gelatin Composite Film

Gelatin is a natural hydrocolloid biopolymer. It could be obtained by hydrolysis of collagen from bones, skin and connective tissue. Due to its abundance, effective cost, biodegradability, biocompatibility, excellent film-forming ability and good mechanical properties, gelatin is widely used in various industries such as food, edible films, pharmaceutical, cosmetic and coating industries (Mohamed, et al., 2019). Gelatin could form a transparent film with an excellent gas barrier. However, the poor mechanical properties of the gelatin film such as low water-vapour barrier have limited its usage in food packaging. As per report from Mohamed, et al. (2019), the addition of gelatin could enhance the properties of CMC film as it contains amino, carboxyl and hydroxyl groups side chains which may be cross-linking with the functional groups of CMC and subsequently result in better stability, mechanical properties, and water resistance.

Mohamed, et al. (2019) studied the effect of CMC and gelatin to the properties of CMC-gelatin film. In this study, glycerol was used as plasticizer and the film was prepared by casting method. Mohamed, et al. (2019) reported that the addition of gelatin lead to the increase in film thickness. Besides, the SEM result showed that the gelatin was well distributed and the firmness of the CMC-gelatin composites film was improved.

Apart from that, both increase of CMC and gelatin content resulted in increase of tensile strength of the CMC-gelatin composite film. In other word, CMC-gelatin composite film has higher mechanical property as compared to pristine CMC or gelatin film. However, high ratio of gelatin could decrease the breaking length of the film as the high amount of gelatin between CMC layers reduce the bonding and increase the spacing between CMC layers. Next, the hydrogen bonding between CMC and gelatin had improved the air permeability of the composite film. The thermal stability of the film was improved when blending CMC with gelatin in the ratio of 50:50. For the water vapor permeability (WVP), there were no significant effect on WVP in the presence of gelatin. This might be due to the addition of gelatin resulted in the increase of spacing between the fabrils and subsequently promote higher water vapor diffusivity through the CMC film. In addition, the addition of glycerol as plasticizer could affect the moisture sorption as well (Mohamed, et al., 2019).

Jahit, et al. (2016) studied the effect of drying temperature to the properties of gelatin-CMC-chitosan composite film. The composition of gelatin, CMC and chitosan were varied and two set of films solutions were dried at room temperature and 45 ℃ respectively. Jahit, et al.

(2016) reported that the FTIR results showed that blending of CMC with gelatin and chitosan lead to decrease in intensity of the COO- group peak as compared to the pristine CMC film. Besides, based on the XRD result, the addition of chitosan to the film formulation lead to the increase in film's crystallinity structure, whereas the film formulation without chitosan showed more amorphous structure.

For vapour permeability value (WVP), there was no significant effect on WVP in the presence of CMC. However, the addition of chitosan could increase WVP value of the composite film. This was because addition of chitosan may lead to the increase of crystallinity and decrease of amorphous properties of the film, in which the higher crystallinity resulted in an increase of the WVP value of the film. For the film biodegradability, it was correlated positively with crystallinity of the film. Higher crystallinity of the film may increase the rate of biodegradable as compared to the amorphous films. Thus, it was clear that increasing the content of chitosan in the film could increase the crystallinity of the film and lead to an increase in biodegradability of the film (Jahit, et al., 2015)

CMC and gelatin are nontoxic and edible materials. Gelatin is often applied in food industries for reconstructing, stabilizing, emulsifying as well as forming foam and film purposes. On the other hand, CMC is also widely applied in food production due to its high viscosity property, high solubility in water and ability to form film. Blending the CMC with gelatin can significantly improve the mechanical properties and water barrier of the CMC film, hence enabling the CMC-gelatin film to be used as edible films and coatings to maintain the food quality and increase the food durability (Tabari, 2017). For the antimicrobial activity of the CMC-gelatin composite film, previous studies stated that the CMC-gelatin film does not show an antimicrobial property by itself and the antimicrobial property showed only when mixed with other substances. A study by Mohamed, et al. (2019) indicated that the increase of shellac concentration leads to reduce in microbial growth, hence adding shellac to the CMC-gelatin composite film formulation can enhance the antimicrobial properties in the film.

2.3.5 CMC-Clay Mineral Composite Film

Clay minerals are nanoparticles that are often used for biopolymer modification to overcome the drawbacks of biopolymer such as low mechanical strength, high brittleness, and poor water vapor barrier properties. Clay minerals such as montmorillonite (MMT) are naturally hydrophilic materials, it swells when in contact with water. The layers of MMT are negatively charged due to the impurities in MMT structure, the negative charges were being counterbalanced by cations such as sodium when blending with CMC. The clay mineral usually would undergo a process of cation exchange, replacing the original cations for organic ones such as alkylammonium. This procedure renders the clay organophilic and increases the affinity of clay mineral with organic materials such as polymers (de Oliveira, et al., 2015). As per report by de Melo Fiori, et al. in 2019, blending biopolymers with lamellar silicates could make the film have higher thermal stability, mechanical strength and better barrier properties. The nanoparticles may even accelerate the biodegradation of the film. Besides, the nanoparticles were found that it could improve the barrier properties by filling the void spaces and serving as physical blockages for water or oxygen molecules. However, such nanocomposite film has the drawbacks like less flexibility and ductility, which could limit its usage as packaging materials.

de Melo Fiori, et al. (2019) studied the properties of the CMC-clay minerals composite film using two types of clay minerals which were nanomer PGV and bentonite with polyethylene glycol (PEG) as plasticizers. PEG is a biodegradable, nontoxic, and water-soluble compound that shows plasticizing effect when mixed with other polymers. The addition of PEG into the film formulation could help to improve the toughness, ductility and flexibility of the film since the clay mineral tends to make the film become brittleness. The clay swelling percentage for both PGV and bentonite clay was high, which indicate the high affinity to the water molecule. Next, the SEM results showed that the clay was dispersed well in the film and no clay agglomerates was found.

de Melo Fiori, et al. (2019) reported that the addition of clay could improve the mechanical properties of the film including the modulus, yield strength, ultimate strength and elongation at break. However, PGV presented better mechanical properties improvement in film as compared to bentonite clay. The mechanical properties of CMC-PGV clay-PEG showed 260% higher modulus, 247% increase in strength, and 300% in elongation as compared to pure CMC film. For the water vapor permeability (WVP), the addition of PGV clay resulted in 27%

decrease in WVP value while Ben clay showed an 8% decrease in the WVP value as compared to the pure CMC film. This indicated that the addition of clay minerals could help to improve the water vapor barrier properties of the CMC film. de Melo Fiori, et al. (2019) also reported that the addition of PEG not only acts as plasticizers, but could also decrease the WVP as well.

de Oliveira, et al. (2015) has carried out an investigation on the properties of CMC-synthetic clay composite film prepared by casting method. In this investigation, laponite was used as synthetic clay to produce the nanocomposite film. de Oliveira, et al. (2015) reported that the addition of laponite clay had not affected the transparency of the film, and the surface of the film become more regular with the addition of clay. Besides, the increase of clay content in the film formulation could induce the chemical interactions between clay particles and polymer chains, which favor the organization of the macromolecules and subsequently promoted the formation of crystalline structure. While the FE-SEM data showed that the laponite clay was homogeneous disperse in the polymeric matrix.

For the mechanical properties of the CMC-laponite nanocomposites, de Oliveira, et al. (2015) reported that the yield strength at break, the elastic modulus and the elongation at break increased with the laponite clay content. However, the strain at break decreased significantly when the clay content was more than 10%. For the water vapor permeability, the sample with 17% of laponite showed a lower water vapor permeability value, which was 42% lower than the pure CMC film. Next, CMC-laponite nanocomposites showed a significant improvement in thermal stability as compared to pure CMC film. This was because the well dispersion of clay particle in the film promoted a protective mechanical barrier against the carbon chain decomposition, which enable the film to be degraded at higher temperature as compared to pure CMC film.

Laponite synthetic clay is widely applied in industries such as engineering, agriculture, pharmaceuticals and ceramics (de Oliveira, et al., 2015). It is always found in the composition of paints, cosmetics and house cleaning products. Laponite is also often used to mix with other biopolymers to form film such as cellulose/laponite composite film prepared by Perotti, et al. (2013) and Yuan, et al. (2014). The CMC-clay mineral composite film can be used in food packaging. The reason is nanocomposites films are able to withstand the stress of thermal food processing, transportation, and storage (Arora and Padua, 2010).

2.4 Application of Biocomposite Film in Current Market

According to Research and Markets (2022), the global market size of biocomposite films in year 2021 is around USD 1.0 billion, and it is projected to increase by USD 0.4 billion in 2026. The increasing global market size for biocomposite films is due to the public getting more aware of the importance of protecting the environment by using biodegradable films to replace plastic films.

Actually, biodegradable films can be produced by various types of natural materials such as collagen, gelatin, starch, cellulose, chitosan, CMC and others. In the current market, the most widely used materials for food packaging film are Polyhydroxy Acids (PHA), cellulose, Polybutylene Succinate (PBS), Polylactic Acid (PLA), and starch blends. Among all these materials, PLA is the most preferred owing to its high availability and more economical.

PLA-based biocomposite films are highly applied as packaging film especially for food packaging. For example, fresh food, frozen food, bread, and dried snacks packaging. There is some researches indicated that adding chitosan to the biocomposite films can give antimicrobial properties which can prolong the shelf life of the food (Research and Markets, 2022). Apart from that, PLA-based biocomposite films are widely used in supermarkets and shopping malls to pack the goods. In most of the supermarket in Malaysia, PLA-based biocomposite films have been used to replace conventional plastic bags.

On the other hand, CMC-based biocomposite is widely applied in biomedical fields. For example, CMC-based composite is used for wound healing, artificial organs, development of tissue and bone-tissue engineering and other purposes. Whereas CMC-based biocomposite films or hydrogels are highly used in pharmaceutical industry. In addition, owing to its high biocompatibility and ability to bind against pharmaceutically active compounds such as drugs, it is always used for drug delivery and drug emulsification (Rahman, et al., 2021).

CHAPTER 3

METHODOLOGY

3.1 Research Flow Chart

The brief outline of the research experimental methodology was illustrated in **Figure 3.1**. First, the CMC solution was prepared separately because CMC powder tends to form lumps and took time to dissolve completely in water. At the same time, PGV solution was prepared under magnetic stirring and sonication to obtain a well dispersed solution. Then, CMC solution and PGV solution were mixed together followed by adding PEG. After that, the solution was poured in a petri dish and dried for 24 hours in a fume hood. The films were then undergone characterization which include tensile test, water solubility, attenuated total reflectance-fourier transform infrared spectroscopy (ATR-FTIR) and Thermogravimetric Analysis (TGA).

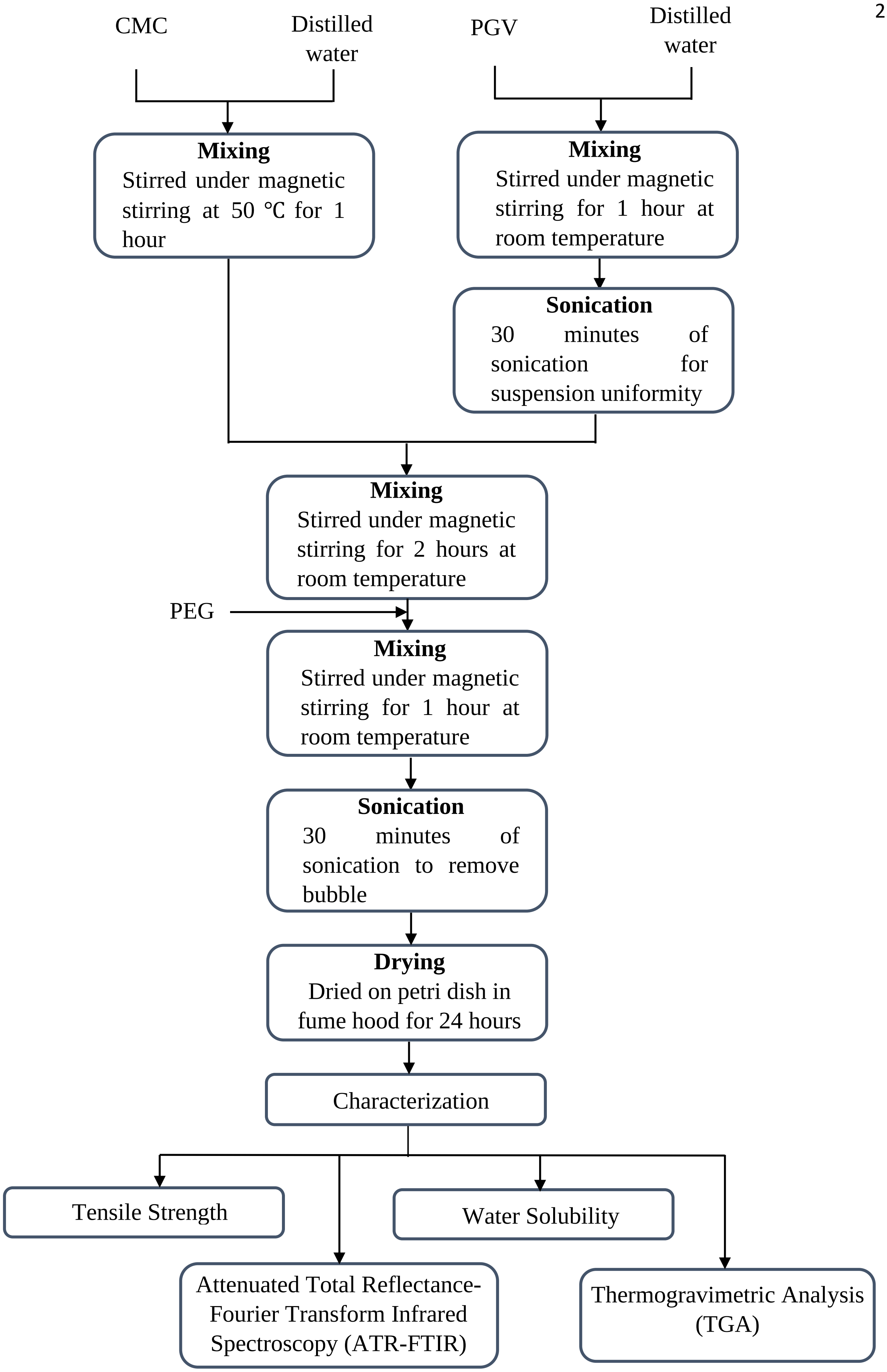

Figure 3.1: Research flow chart for the CMC-PGV-PEG composite film.

3.2 Chemical Used in the Research

Table 3.1 shows the chemical used in this research with their source.

Table 3.1: Chemical used in this research.

Chemical	Source
Carboxymethylcellulose sodium salt powder (CMC)	Alfa Aesar
Polyethylene glycol (PEG)	Chemiz
Nanomer PGV (PGV)	Aldrich

3.3 Preparation of CMC Solution (CMC film)

Carboxymethylcellulose (CMC) solution was prepared by dissolving 2 g of CMC in 80 ml of distilled water. The mixture was stirred at 50 ℃ for 1 hour under magnetic stirring until the CMC completely dissolve in the water. Then, the solution was poured into a petri dish and dried at room temperature for 24 hours in a fume hood. The dried films were peeled and stored at room temperature before analysis.

3.4 Preparation of CMC Film Plasticized with PEG

Carboxymethylcellulose (CMC) solution was prepared by dissolving 2 g of CMC in 80 ml of distilled water. The mixture was stirred at 50 ℃ for 1 hour under magnetic stirring until the CMC completely dissolve in the water. Subsequently, 0.6 g of PEG was added to the mixture and stirred at room temperature for 1 hour under magnetic stirring. The mixture was then sonicated for 30 minutes to remove bubble. Then, the solution was poured into a petri dish and dried at room temperature for 24 hours in a fume hood. The dried films were peeled and stored at room temperature before analysis.

3.5 Preparation of CMC-PGV-PEG Composite Film

Carboxymethylcellulose (CMC) solution was prepared by dissolving 2 g of CMC in 80 ml of distilled water. The mixture was stirred at 50 ℃ for 1 hour under magnetic stirring until the CMC completely dissolve in the water. Subsequently, 0.2 g of PGV was dissolved in 20 ml of distilled water and stirred for 1 hour at room temperature under magnetic stirring, followed by sonication for 30 minutes for uniform suspension. Then, the CMC solution and PGV solution were mixed together and stirred for 2 hours under magnetic stirring at room temperature. 0.6 g of PEG was added to the mixture and stirred at room temperature for 1 hour under magnetic stirring. The mixture was then sonicated for 30 minutes to remove bubble. Then, the solution was poured in a petri dish and dried at room temperature for 24 hours in a fume hood. The dried films were peeled and stored at room temperature before analysis. The preparation steps were repeated by varying the composition of CMC, Nanomer PGV, and PEG as shown in **Table 3.2**.

Table 3.2: Compositions of film formulations.

Sample	CMC Content (g)	PGV Content (g)	PEG Content (g)
CMC	2	-	-
2CMC-0.6PEG	2	-	0.6
1CMC-0.2PEG-0.6PGV	1	0.2	0.6
2CMC-0.2PEG-0.6PGV	2	0.2	0.6
3CMC-0.2PEG-0.6PGV	3	0.2	0.6
2CMC-0.1PEG-0.6PGV	2	0.1	0.6
2CMC-0.2PEG-0.6PGV	2	0.2	0.6
2CMC-0.3PEG-0.6PGV	2	0.3	0.6
2CMC-0.2PEG-0.5PGV	2	0.2	0.5
2CMC-0.2PEG-0.6PGV	2	0.2	0.6
2CMC-0.2PEG-0.7PGV	2	0.2	0.7

3.6 Characterization of Film

3.6.1 Attenuated Total Reflectance - Fourier Transform Infrared Spectroscopy (ATR-FTIR)

Attenuated Total Reflectance- Fourier Transform Infrared Spectroscopy (ATR-FTIR) analysis is conducted to identify the functional group that presented in the sample. ATR-FTIR analysis was carried out using Perkin Elmer Spectrum ex1. The sample was directly put under the sensor and the analysis conducted in the wavelength range of 400-4000 cm^{-1} (The Michael Faraday Laboratories, 2007).

3.6.2 Tensile Strength

The mechanical properties of the film which is tensile strength were investigated using tensile test. An ASTM D638 plastic tensile strength test was performed in a light-weight tensile tester. The samples were first cut into “dumbbell” shapes and the thickness of the samples were measured with nanometer. Then, the sample was loaded into tensile grips and the extensometer was attached to it. The test was started after the load and speed were adjusted. The tensile test ended once the sample rupture. The test was repeated for three times to get the average results (Andrew, 2010).

3.6.3 Thermogravimetric Analysis (TGA)

Thermogravimetric Analysis (TGA) is carried out to determine the thermal stability of the film. During analysis, aluminum and alumina pans are used as reference. The analysis is conducted under continuous nitrogen flow of 30 mL/min. The sample is heated from 30 ℃ to 900 ℃ at a heating rate of 10 ℃/min, where the analysis condition is referred and modified from the study by De Oliveira, et al. (2015).

3.6.4 Water Solubility

Water solubility test is carried out to determine the water resistance of the film. The film samples were kept in a desiccator with dry calcium sulphate until the film samples reached constant weight. Then, 0.5 g of the film samples were immersed in 50 ml of distilled water in a beaker at room temperature. The time taken for the film to dissolve in the water is recorded (de Melo Fiori, et al., 2019). The water solubility test was carried out in three replicates to obtain the average result.

CHAPTER 4

RESULTS AND DISCUSSION

4.1 Overview

In this research paper, there were nine different composition of CMC composite films were prepared by casting method. Each of the film was undergoing characterization test which include Attenuated Total Reflectance- Fourier Transform Infrared Spectroscopy (ATR-FTIR), tensile test, Thermogravimetric Analysis (TGA) and water solubility test. The results obtained from characterization are discussed in detail in this chapter.

4.2 Characterization of CMC, PEG, PGV and CMC Composite Film

4.2.1 Physical Observation

Figure 4.1 (a), (b) and (c) show the pure CMC, PEG and PGV chemical without any treatment. While **Figure 4.2 (a), (b) and (c)** show the pure CMC film, CMC-PEG film and 2CMC-0.6PEG-0.2PGV film. When CMC is dissolved in water, the CMC solution presented as clear and viscous solution. It became a transparent and glossy film with some stickiness when the moisture content in the solution evaporated. When PEG added to the CMC, the CMC-PEG film became less transparent and less glossy as compared to pure CMC film and there is no stickiness in CMC-PEG film. Then when PGV is added to form the CMC-PEG-PGV film, the film became less transparent as compared to CMC-PEG film, which means the film having the lowest film transparency among the films. Also, there is no stickiness and glossy are observed in CMC-PEG-PGV film.

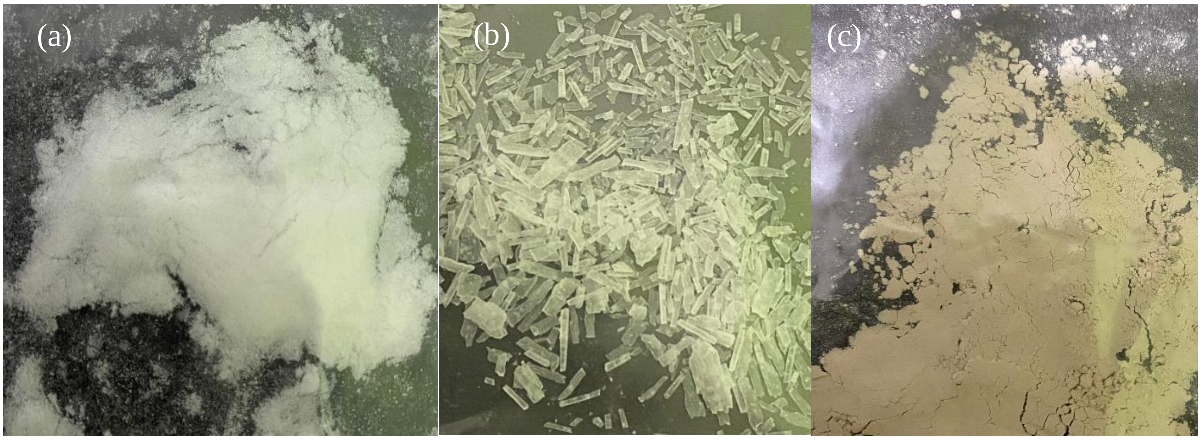

Figure 4.1: The sample of pure (a) CMC, (b) PEG, (c) PGV chemicals.

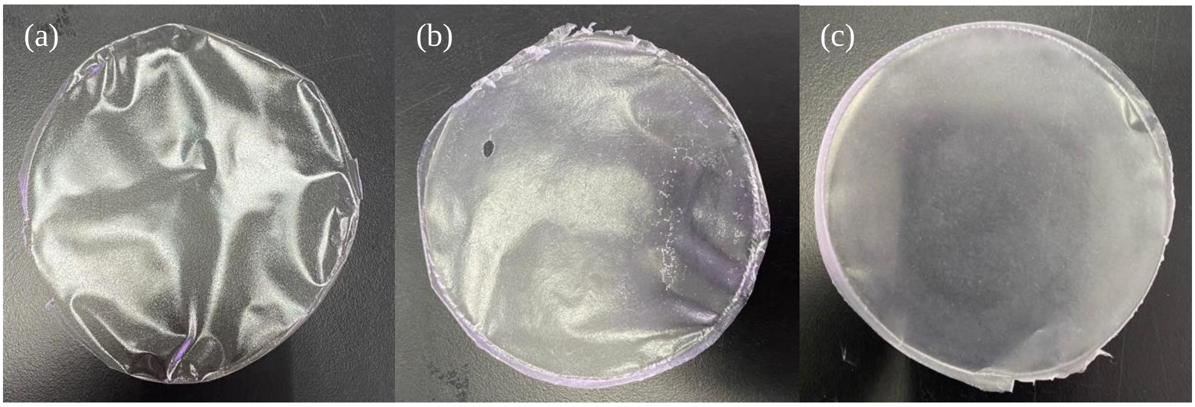

Figure 4.2: The sample of (a) CMC film, (b) CMC-PEG film, (c) 2CMC-0.6PEG-0.2PGV film.

4.2.2 Analysis of Attenuated Total Reflectance- Fourier Transform Infrared Spectroscopy (ATR-FTIR)

Figure 4.3 and 4.4 show the spectrum of ATR-FTIR of PEG, CMC, PGV, CMC composite film and commercial PE film. The functional groups for each sample is presented in **Table 4.1**. In pure CMC film, the absorption bands at 3376 cm^{-1} corresponds to the stretching of hydroxyl (-OH) group. The stretching of carboxyl (-COOH) group is found at 1590 and 1414 cm^{-1}. The peak at 996 cm^{-1} are attributed to the presence of C-O bonds (Mohkami and Talaeipour ,2011). In PEG, the absorption band at 3416 cm^{-1} indicated the presence of hydroxyl (-OH) group. The bands at 2882, 1467, 1359 and 1341 cm^{-1} are allocated to C-H bending. The band at 1279 cm^{-1} is due to O-H deformation, whereas 1098 cm^{-1} is attributed to the presence of C-O-H bending (Shameli, et al., 2012). In PGV, the peak at 3615 cm^{-1} indicated the presence of H-O-H stretching, which is free water molecules in the interlayer region of clay. The presence of H-O-H stretching in PGV has proven that PGV nanoclay is hydrophilic. Then, the absorption band at 3393 cm^{-1} is due to the presence of O-H stretching, while the absorption band at 1633 cm^{-1} is attributed to the O-H deformation. The bands at 984 and 511 cm^{-1} correspond to the Si-O stretching while the band at 452 cm^{-1} is attributed to the Si-O bending (Celestino, et al., 2018). The CMC and 2CMC-0.6PEG-0.2PGV film exhibit similar functional group, where the band at 3358 cm^{-1} correspond to O-H stretching, the bands at 1416 and 1588 cm^{-1} are due to -COOH stretching, and the peak at 1023 cm^{-1} correspond to C-O bonds. Besides, the C-H bending that found in PEG also present in 2CMC-0.6PEG-0.2PGV film at the band of 2889 cm^{-1}, while the Si-O bending was found in 2CMC-0.6PEG-0.2PGV film at the band of 511 and 453 cm^{-1}. The C-O-H bending was not found in 2CMC-0.6PEG-0.2PGV film as it was overlapping with C-O bonds, whereas H-O-H was not found in 2CMC-0.6PEG-0.2PGV film as the moisture was condensed in the film formulation process. From **Figure 4.4**, it could be observed that the O-H stretching curve for 2CMC-0.6PEG-0.2PGV film was deeper as compared to CMC and PGV, which indicated that the hydrogen bonds might be formed between the hydroxyl group in CMC and PGV. Since there is no new functional group is found in the 2CMC-0.6PEG-0.2PGV film, it could be concluded that there is no chemical reaction happened during the mixing process in the formation of composite film (De Oliveira, et al., 2015). On the other hand, the commercial PE film, consists of the bands at 2916 and 2849 cm^{-1} which correspond to the C-H bending, while the bands at 1465 and 721 cm^{-1} are attributed to the presence of C=C stretching and C-H bending (Asgari, Moradi and Tajeddin, 2014).

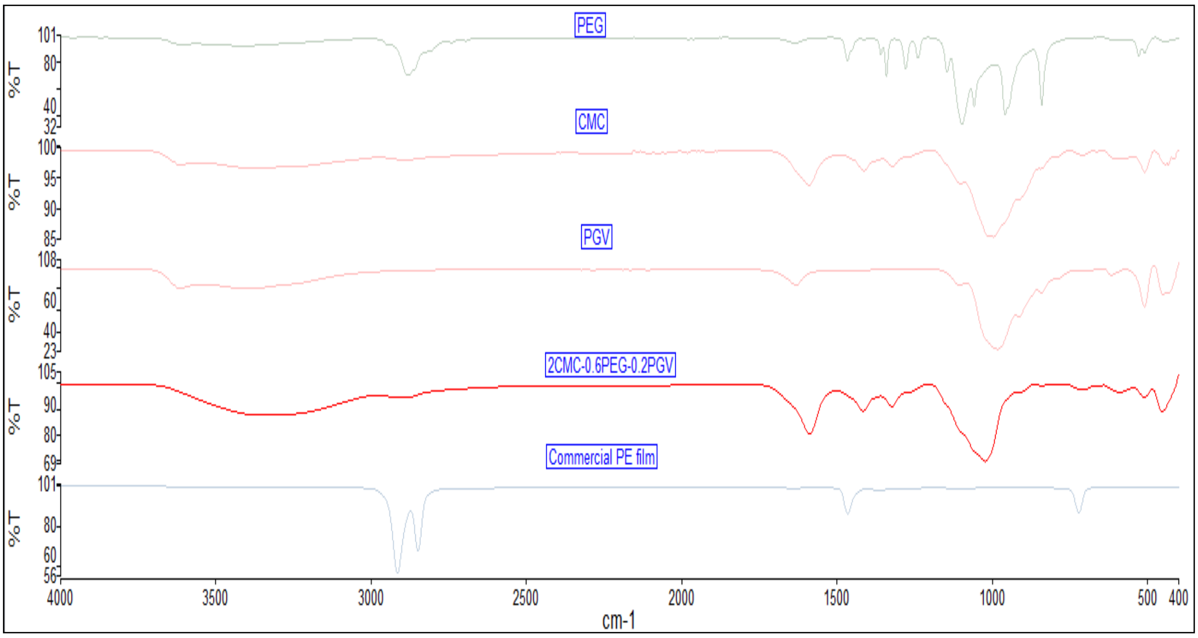

Figure 4.3: ATR-FTIR of PEG, CMC, PGV, CMC composite film and commercial PE film.

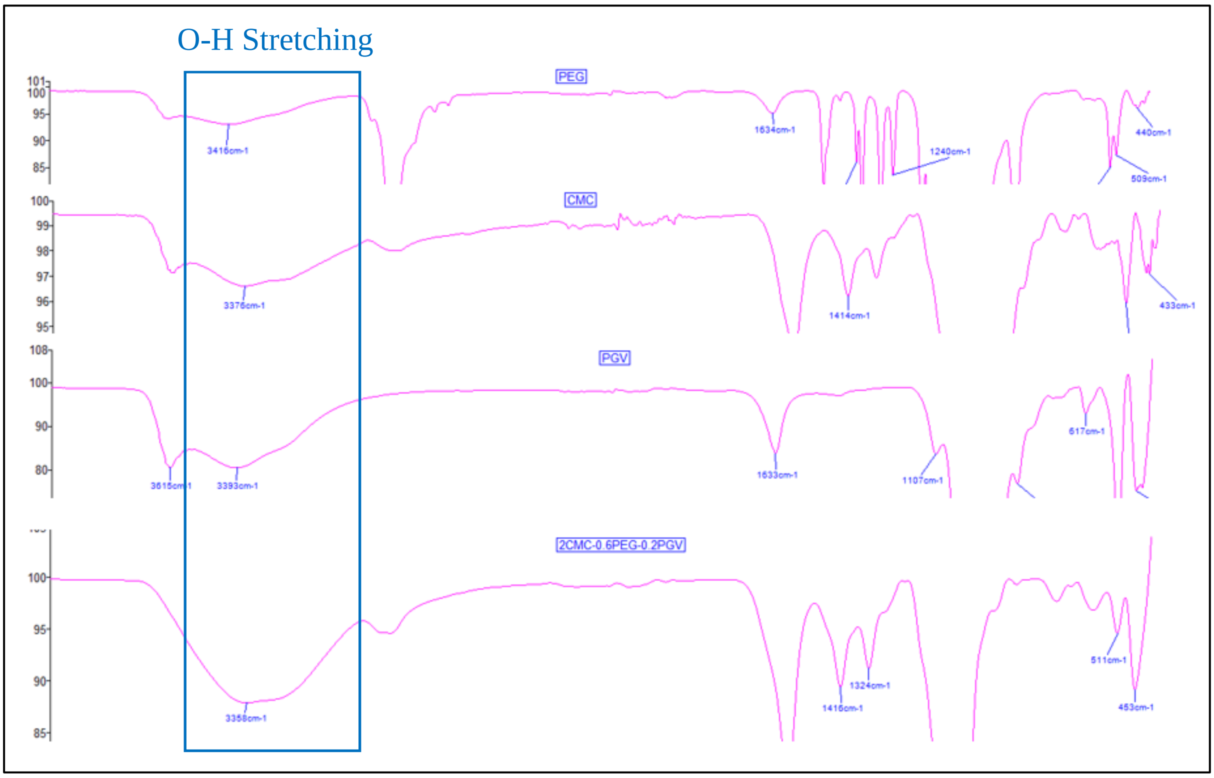

Figure 4.4: Comparison of O-H stretching curve for 2CMC-0.6PEG-0.2PEG with other samples.

Table 4.1: Summaries of functional group of PEG, CMC, PGV, CMC composite film and commercial PE film.

Film	**Wavenumber** (cm^{-1}) **-OH**	**Wavenumber** (cm^{-1}) **C-H**	**Wavenumber** (cm^{-1}) **-COOH**	**Wavenumber** (cm^{-1}) **-CO**	**Wavenumber** (cm^{-1}) **H-O-H**	**Wavenumber** (cm^{-1}) **-COH**	**Wavenumber** (cm^{-1}) **Si-O**
CMC	3376	-	1590,1414	996	-	-	-
PEG	3416, 1279	2882, 1467, 1359, 1341	-	-	-	1098	-
PGV	3393, 1633	-	-	-	3615	-	984, 511, 452
2CMC-0.6PEG-0.2PGV	3358	2889	1416, 1588	1023	-	-	511, 453
Commercial PE plastic	-	2916, 2849		-	-	-	-

4.2.3 Mechanical Properties

Table 4.2 shows the summary of film's tensile properties. The average tensile modulus of pure CMC film, 2CMC-0.6PEG film and 2CMC-0.6PEG-0.2PGV film were 993.67 MPa, 579.67 MPa and 1054.33 MPa, respectively. The average ultimate strength of pure CMC film, 2CMC-0.6PEG film and 2CMC-0.6PEG-0.2PGV were 47.12 MPa, 29.88 MPa, and 51.78 MPa. While the average elongation at break was 41.80%, 36.63%, and 27.53% for pure CMC film, 2CMC-0.6PEG film and 2CMC-0.6PEG-0.2PGVm respectively. The result showed that the addition of PEG to the CMC film resulted in decreases of tensile modulus, ultimate strength, and elongation at break of the film. On the other hand, the tensile modulus, ultimate strength, and elongation at break of the film increased with the addition of PGV. The studies from de Melo Fiori, et al. (2019) indicated that the addition of PEG could increase the elongation at break of the film as PEG affect the structure between the polymeric chain and disrupt the interaction between the polymeric chain to make the chain become more flexible. However, this study did not show the similarity which may be due to inhomogeneous dispersion of PEG. Another reason is the film do not consist of a plasticizer or additives for chain interaction as conducted by de Melo Fiori et al. (2019). Apart from that, de Oliveira, et al. (2015) and de Melo Fiori, et al. (2019) showed that the addition of PGV could increase the tensile properties of the film, which is similar to this study. This is because the nanoclay particles were dispersed uniformly in the polymer matrix and form hydrogen bonds between the PGV nanoclay and CMC (Rhim, Hong and Ha, 2009).

Table 4.2: Summary of film tensile properties at different composition.

No	Film	Average tensile modulus (MPa)	Average ultimate strength (MPa)	Average elongation at break (%)
1	CMC (control)	993.67	47.12	41.80
2	2CMC-0.6PEG	579.67	29.88	36.63
3	2CMC-0.6PEG-0.2PGV	1054.33	51.78	27.53

4.2.4 Thermogravimetric Analysis (TGA)

Figure 4.5 to 4.7 show the TGA and DTG thermograms of the pure CMC film, 2CMC-0.6PEG film and 2CMC-0.6PEG-0.2PGV film. The TGA and DTG thermograms indicated that the decomposition of both pure CMC film and CMC composite films takes place in multi-step thermal decomposition, in which the pure CMC film showed two-step decomposition, whereas the 2CMC-0.6PEG film and 2CMC-0.6PEG-0.2PGV film decomposed through three-step decomposition (Ezati, et al., 2020).

At the temperature range of 70 ℃ to 110 ℃, all the films experienced first weight loss which was probably due to the evaporation of moisture on the film surface for pure CMC film and the evaporation of volatile components that decompose easily such as PEG for 2CMC-0.6PEG film and 2CMC-0.6PEG-0.2PGV film (Ezati, et al., 2020; He, Fei and Li, 2019). For the pure CMC film, the second step of thermal decomposition took place at around 260 ℃ to 310 ℃. At this step, the pure CMC film experienced the main weight loss due to the rapid thermal degradation of polymers (He, Fei and Li, 2019). The residue percentage after the second step thermal decomposition of pure CMC film was 42.4367%. For the CMC composite film, the second step of thermal decomposition is due to the degradation of polymer chain while the third step of thermal decomposition is due to further degradation causing a breakdown in the polymer backbone (Kapil, et al., 2019). The second step thermal decomposition of 2CMC-0.6PEG film and 2CMC-0.6PEG-0.2PGV film took place at around 260 ℃ to 320 ℃, while the third step of thermal decomposition took place at around 350 ℃ to 400 ℃. The residue percentage after third step thermal decomposition of 2CMC-0.6PEG film and 2CMC-0.6PEG-0.2PGV film were 34.3475% and 32.5351% respectively. According to Saha, et al. (2016) and Mohamed, et al. (2016), the film’s thermal stability will increase with the addition of MMT into the CMC matrix owing to the good heat insulation capacities of MMT. In other words, with the addition of MMT, the residue percentage would increase after the thermal decomposition. However, in this study, the incorporation of PGV nanoclay which is one of the types of MMT into the CMC matrix did not show improvement of residue percentage in 2CMC-0.6PEG-0.2PGV film. This may be due to the inhomogeneous dispersion of PGV nanoclay in the polymer matrix.

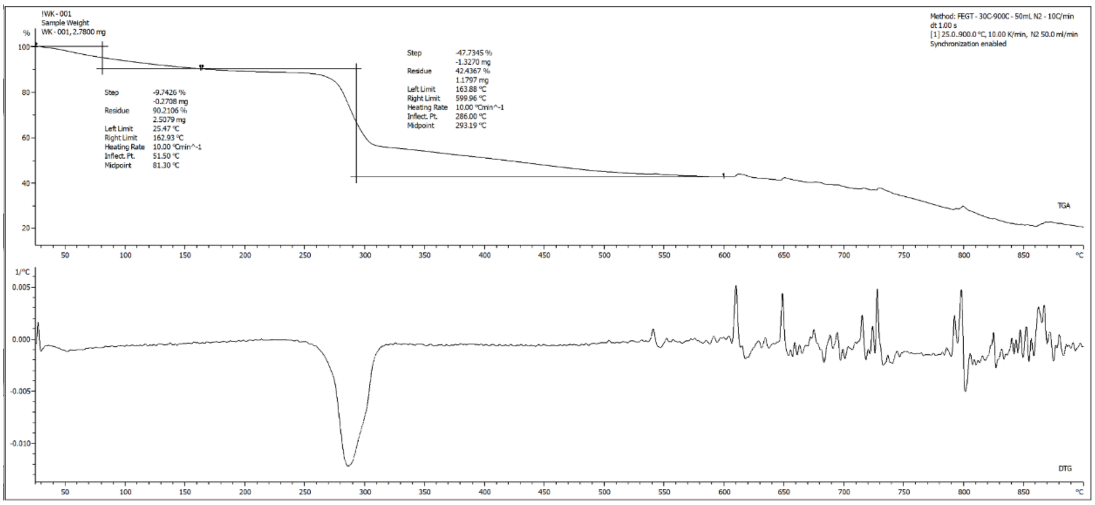

Figure 4.5: TGA (top) and DTG (bottom) thermograms of pure CMC film.

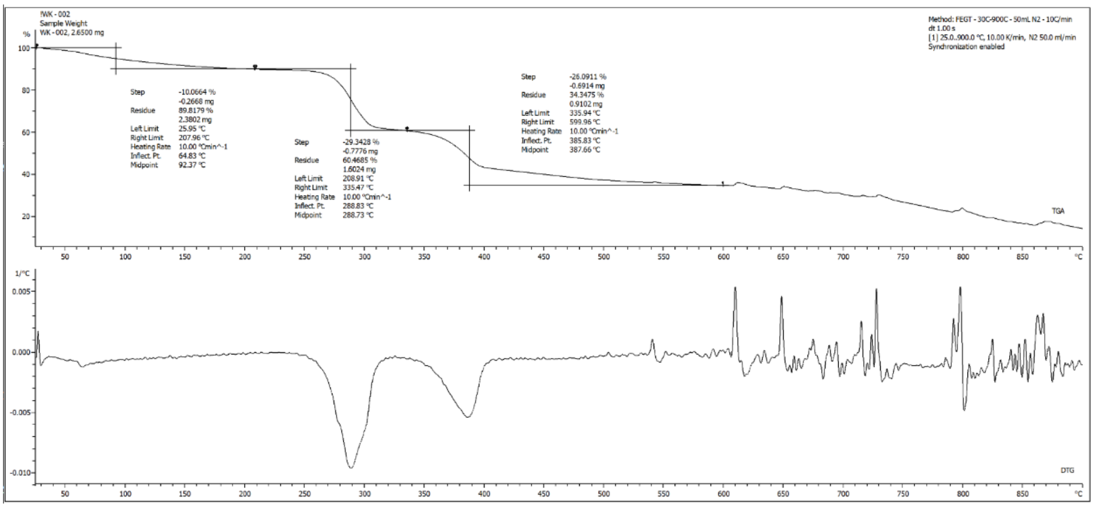

Figure 4.6: TGA (top) and DTG (bottom) thermograms of 2CMC-0.6PEG film.

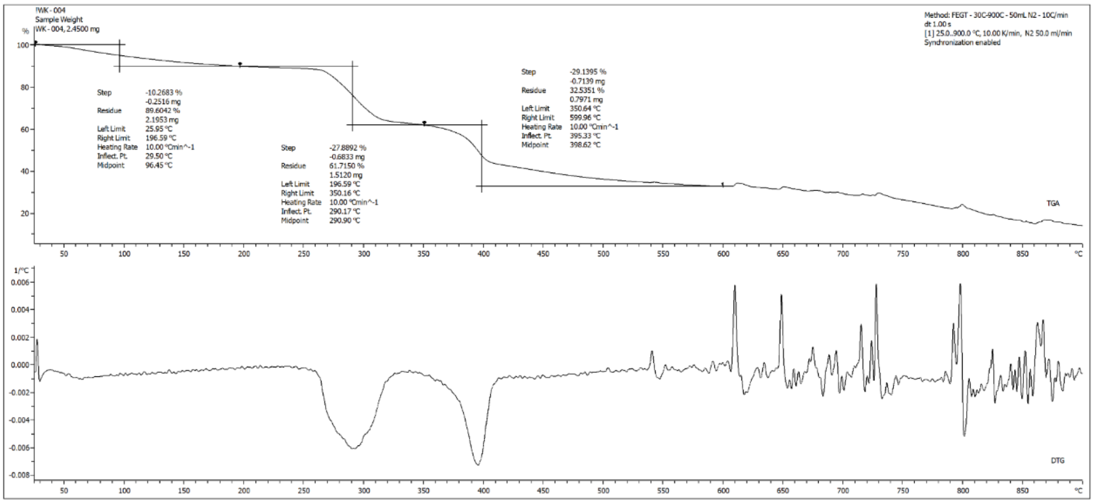

Figure 4.7: TGA (top) and DTG (bottom) thermograms of 2CMC-0.6PEG-0.2PGV film.

4.2.5 Water Solubility

Table 4.3 shows the time taken for films to dissolve in water. CMC film, 2CMC-0.6PEG film and 2CMC-0.6PEG-0.2PGV film took 3, 7 and 15 minutes, respectively to dissolve in water. It could be observed that the pure CMC film was dissolved in water in the shortest time. The addition of PEG to CMC increased the time taken to dissolve in water by 4 minutes. The addition of PGV nanoclay further increased the time taken to dissolve in water by 8 minutes as compared to 2CMC-0.6PEG film. According to the study from de Melo Fiori, et al. (2019), a similar trend of result was obtained which indicate that the addition of PEG could slightly decrease the WVP of the film as well. Besides, the study by de Melo Fiori, et al. (2019) and de Oliveira, et al. (2015) presented the addition of nanocomposite to the CMC film resulted in a decrease in WVP of the film. This is because the nanocomposite such as PGV nanoclay act as a filler to fill the void space and acts as physical blockage to increase the water resistance of the film.

Table 4.3: Time taken for film samples to dissolve in water.

No	Film	Time taken to dissolve in water (mins)
1	CMC (control)	3
2	2CMC-0.6PEG	7
3	2CMC-0.6PEG-0.2PGV	15

4.3 Effect of CMC Content to Film Properties

4.3.1 Functional Group Analysis via Attenuated Total Reflectance-Fourier Transform Infrared Spectroscopy (ATR-FTIR)

Figure 4.8 shows the ATR-FTIR of pure CMC film, 2CMC-0.6PEG film, and CMC composite film with various CMC content. It could be observed that there is no new functional group presented in the CMC composite film, which means that there is no chemical reaction occurred during the mixing or sonication process. Besides, as the CMC content increases, the O-H stretching bands at around 3000 – 3700 cm^{-1} become more deeper, which indicating that the number of hydrogen bonds increases as the CMC content increases. Also, the -COOH stretching at around 1500 – 1700 cm^{-1} become deeper as the CMC content increases from 1 g to 2 g and 3 g, which indicates that there is more carboxyl group presented in the film as the CMC content increases, thus promoted more interaction with others functional groups in PGV and PEG.

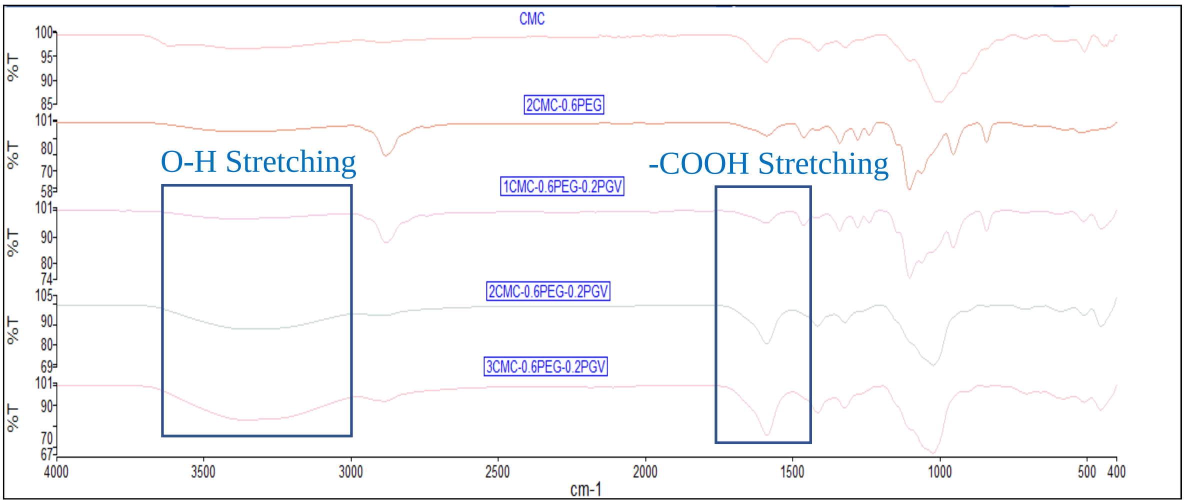

Figure 4.8: ATR-FTIR analysis of pure CMC film and CMC composite film at various CMC content.

4.3.2 Mechanical Properties

Table 4.4 summarized the tensile properties of the film at various CMC content. While **Figure 4.9, 4.10 and 4.11** compare the tensile modulus, ultimate strength, and elongation at break of the film samples at various CMC content. The data indicated that the average tensile modulus, ultimate strength and elongation at break of the pure CMC film were 993.67 MPa, 47.12 MPa and 41.80% respectively. As the CMC composition increased from 1 g to 2 g and 3 g, the average tensile modulus increased from 1002.67 MPa to 1054.33 MPa and 1431 MPa, whereas the average ultimate strength increased from 29.15 MPa to 51.78 MPa and 56.74 MPa. For the average elongation at break, it reduced from 30.63% to 27.53% and 27.30% as the CMC content increased from 1 g to 2 g and 3 g. According to the study from Tongdeesoontorn, et al. (2011) and Ghanbarzadeh, et al. (2010), similar trend of result was obtained. This trend can be explained by more strong hydrogen bond being formed between inorganic PGV nanoclay and the hydroxyl group of organic biopolymer CMC as the CMC content increased, where the hydrogen bond in the film resulted in improvement of tensile properties and reduction of the flexibility of the film (Delhom, et al., 2010; Perotti, et al., 2011).

Table 4.4: Summaries of film tensile properties at various CMC content.

No	Film	Average Tensile modulus (MPa)	Average Ultimate strength (MPa)	Average Elongation at break (%)
1	CMC (control)	993.67	47.12	41.80
2	2CMC-0.6PEG	579.67	29.88	36.63
3	1CMC-0.6PEG-0.2PGV	1002.67	29.15	30.63
4	2CMC-0.6PEG-0.2PGV	1054.33	51.78	27.53
5	3CMC-0.6PEG-0.2PGV	1431.00	56.74	27.30

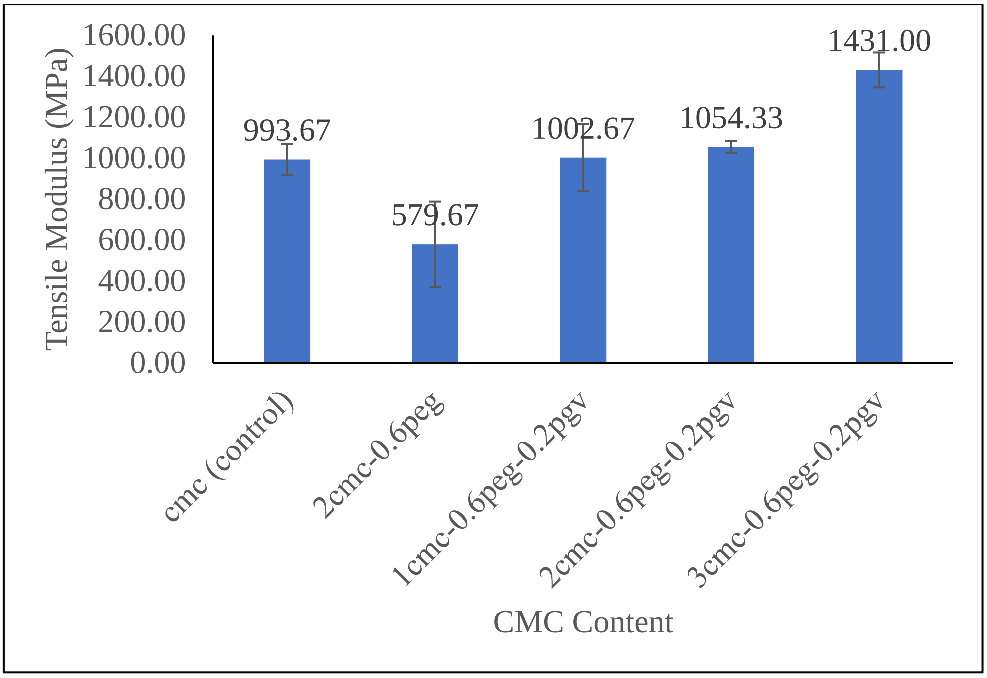

Figure 4.9: Tensile modulus of the film at various CMC content.

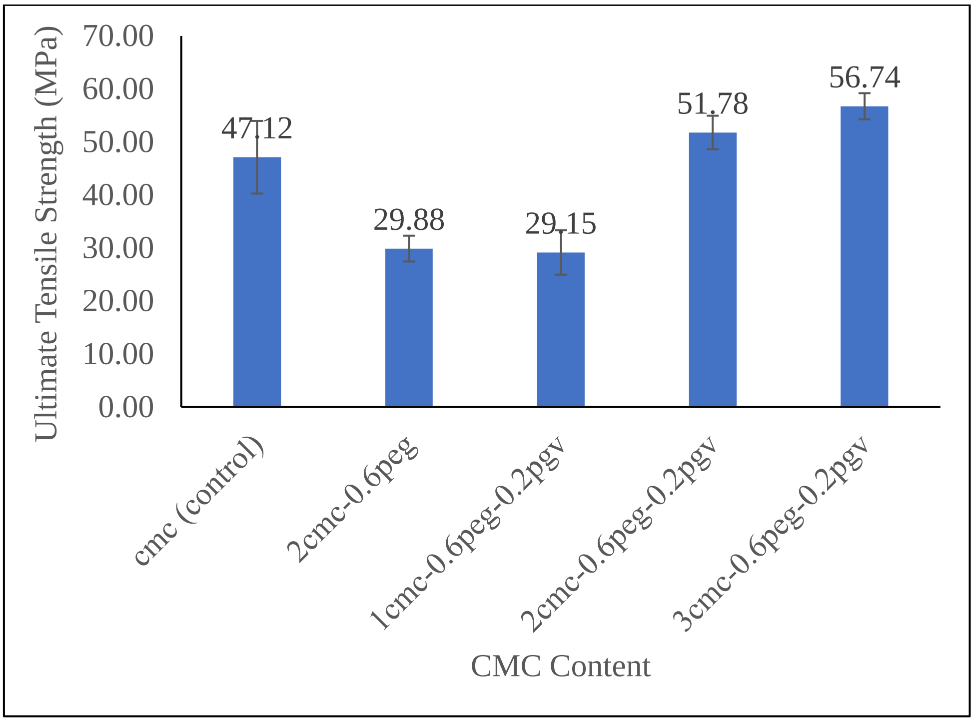

Figure 4.10: Ultimate strength of the film at various CMC content.

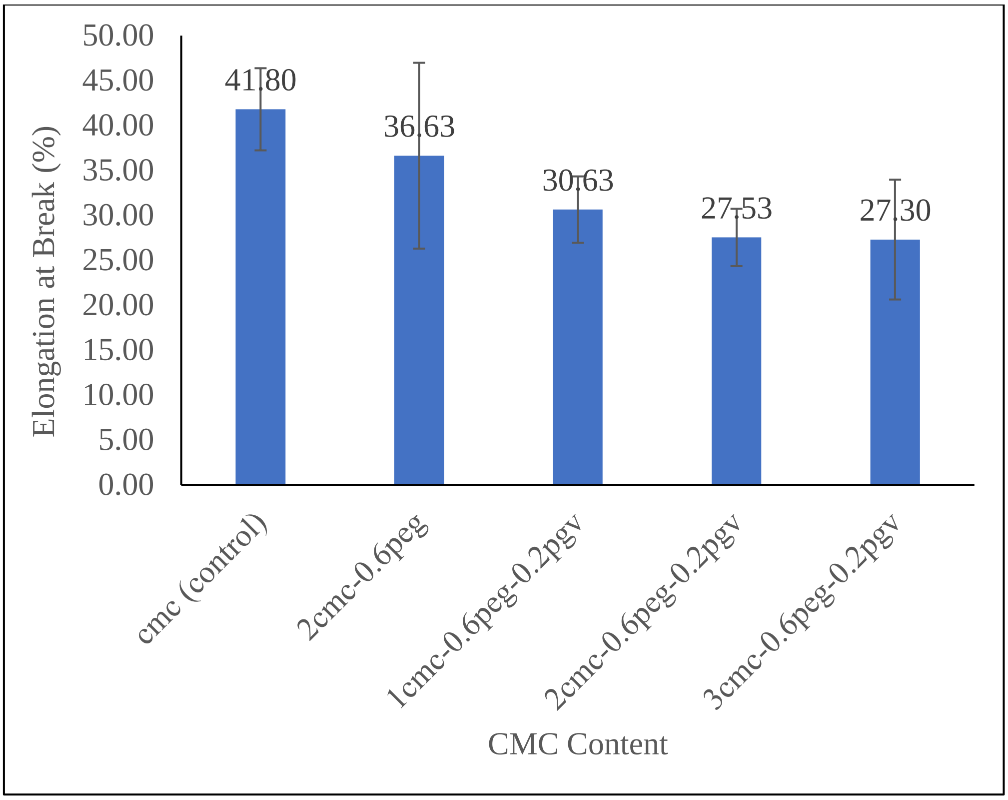

Figure 4.11: Elongation at break of the film at various CMC content.

4.3.3 Thermogravimetric Analysis (TGA)

From **Figure 4.12, 4.13 and 4.14**, it could be observed that as the CMC content increased from 1 g to 2 g and 3 g, the residue percentage was slightly increased from 29.3055% to 32.5351% and 33.2076% after the third step thermal decomposition. The increase of residue percentage after thermal decomposition is completed may be due to the CMC acting as a stabilizer in the film. Similar results were obtained in the study by Kapil, et al. (2019) and Shehap (2008), proving that increasing the CMC content could improve the thermal stability of the film. Increasing CMC content could provide more thermal stability to the film to decrease the weight loss during heating.

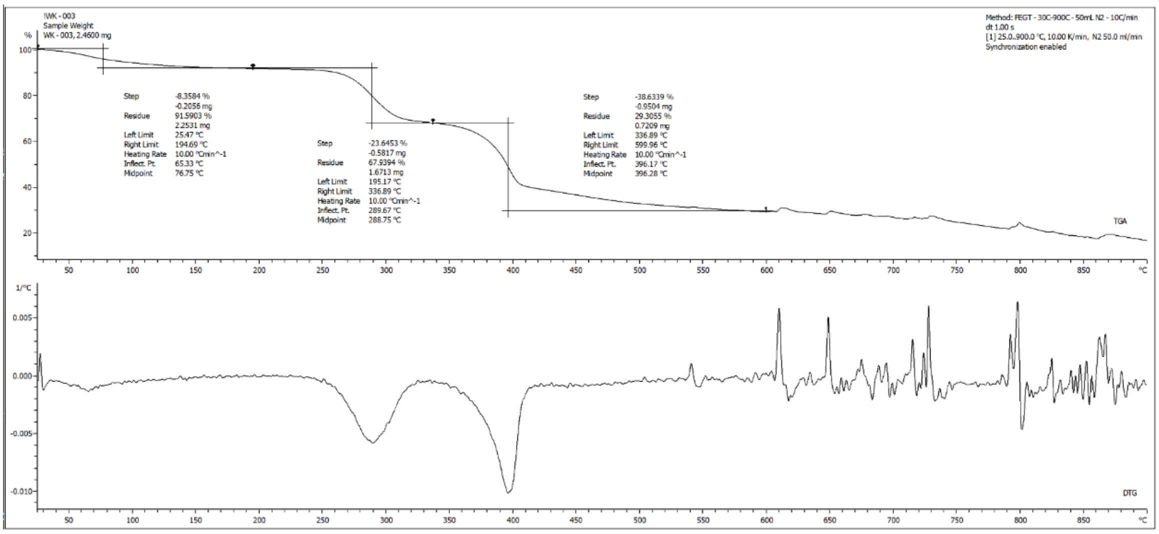

Figure 4.12: TGA (top) and DTG (bottom) thermograms of 1CMC-0.6PEG-0.2PGV film.

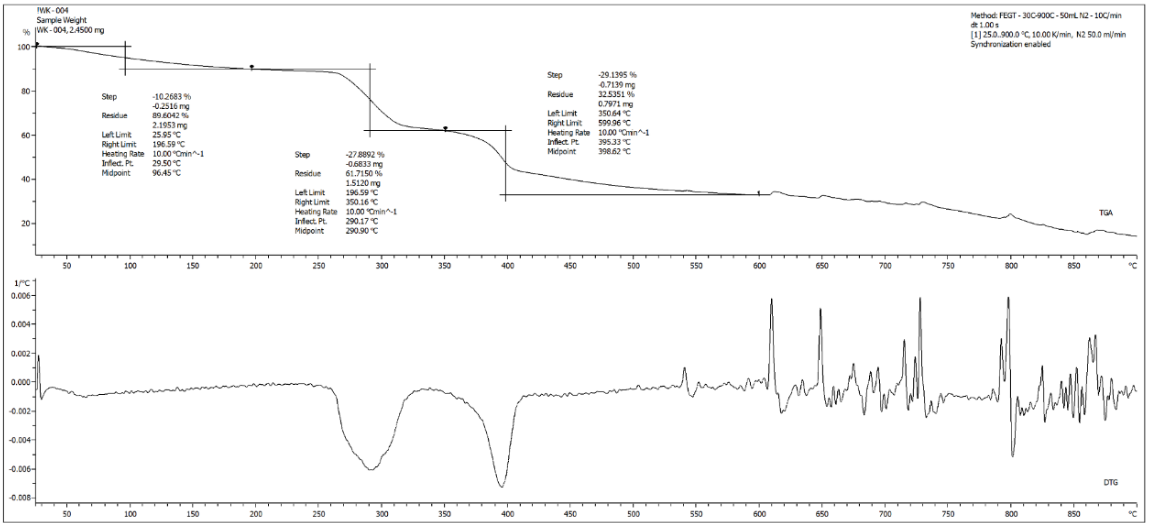

Figure 4.13: TGA (top) and DTG (bottom) thermograms of 2CMC-0.6PEG-0.2PGV film.

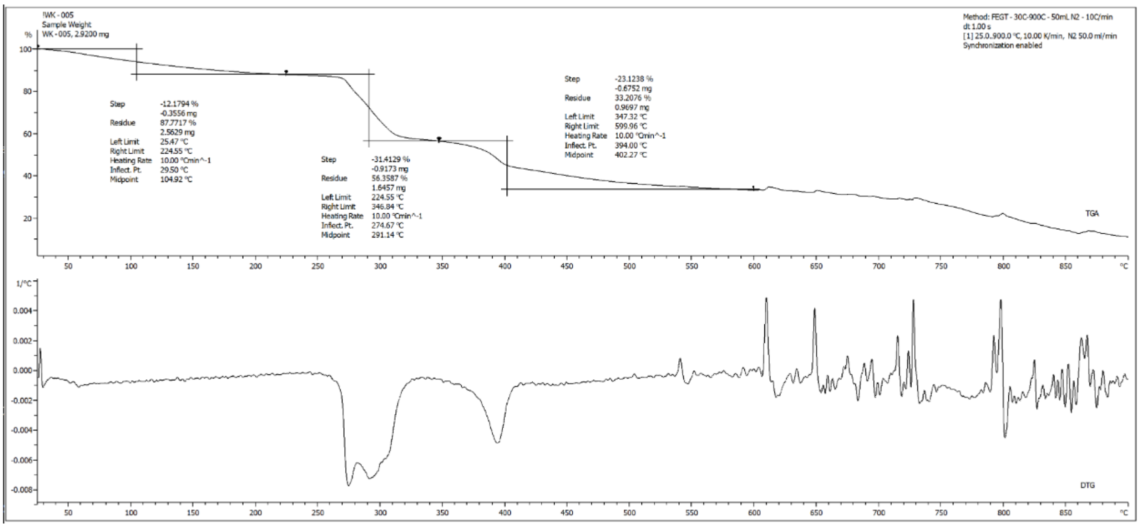

Figure 4.14: TGA (top) and DTG (bottom) thermograms of 3CMC-0.6PEG-0.2PGV film.

4.3.4 Water Solubility

Figure 4.15 shows the time taken for the film at various CMC content to dissolve in water**.** The pure CMC film takes 3 minutes to dissolve in water, which it takes the shortest time to dissolve in the water among all the film samples. Adding PEG to the film increases the time taken for the film to dissolve in water by 4 minutes as compared to pure CMC film. As the CMC content increased from 1 g to 2 g and 3 g, the time taken for the film to dissolve in water increased from 13 minutes to 15 minutes and then decreased to 11 minutes. According to the Tongdeesoontorn et al. (2011), the water solubility of CMC-cassava starch film decreased as the CMC content increased due to the intermolecular interaction between CMC and cassava starch. The water solubility of CMC-cassava starch film decreased as the hydroxyl group of CMC form strong hydrogen bonds with hydroxyl groups of starch. In this study, the water solubility of the film decreased when there was interfacial interaction through hydrogen bond between hydroxyl group of CMC and inorganic PGV.

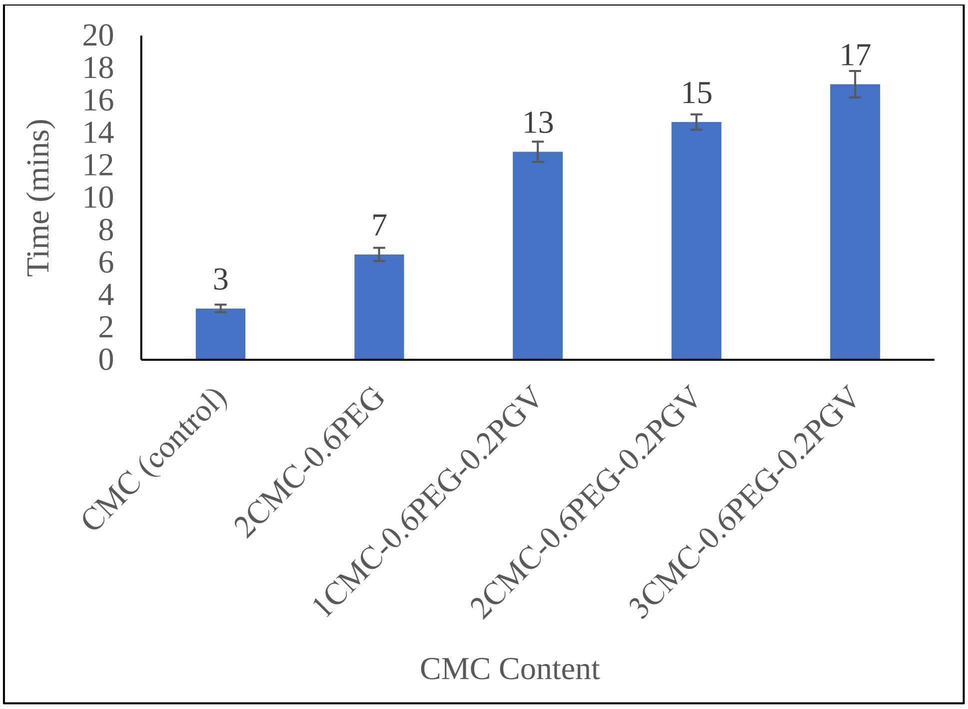

Figure 4.15: Time taken for the film at various CMC content to dissolve in water.

4.4 Effect of PEG Content to Film Properties

4.4.1 Functional Group Analysis Using Attenuated Total Reflectance- Fourier Transform Infrared Spectroscopy (ATR-FTIR)

Figure 4.16 shows the ATR-FTIR of pure CMC film, 2CMC-0.6PEG film, and CMC composite film with various PEG content. It could be observed that there is no any new functional group presented in the CMC composite film, which means that there is no chemical reaction occurred during mixing or sonication process. Besides, as the PEG content increases from 0.5 g to 0.6 g, the C-H bending at around 2800 cm^{-1} was reduced and the peak appears again when the PEG content increases from 0.6 g to 0.7 g.

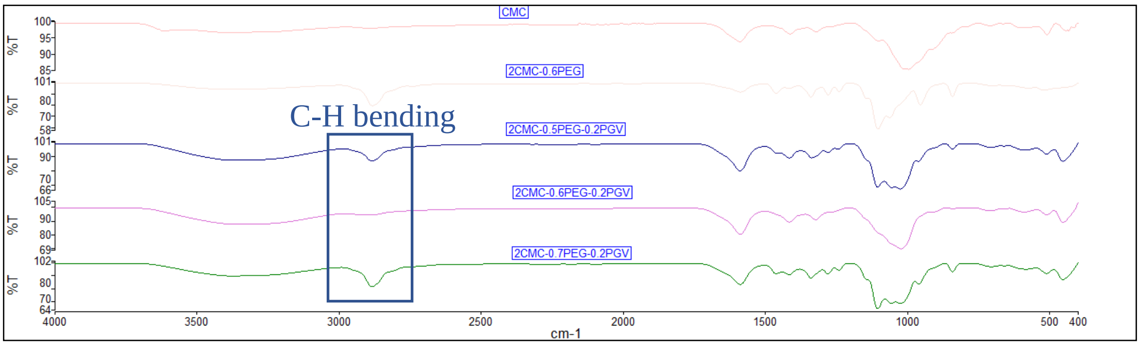

Figure 4.16: ATR-FTIR of pure CMC film and CMC composite film with various PEG content.

4.4.2 Mechanical Properties

Table 4.5 summarized the tensile properties of the film at various PEG content. While **Figure 4.17, 4.18 and 4.19** showed the graph of tensile modulus, ultimate strength, and elongation at break of the film samples at various PEG content. For the pure CMC film, the average tensile modulus, ultimate strength and elongation at break were 993.67 MPa, 47.12 MPa and 41.80% respectively. When the PEG was added to the film, the average tensile modulus, ultimate strength and elongation at break has an obvious decrement to 579 MPa, 29.88 MPa and 36.63% respectively. It can be told that adding PEG to the CMC film without the presence of PGV could decrease the tensile modulus, ultimate strength and elongation at break of the film. As the PEG content increased from 0.5 g to 0.6 g and 0.7 g, the tensile modulus of the film decreased from 1162.67 MPa to 973.67 MPa and 828.67 MPa, whereas the average elongation at break of the film decreased from 29.37% to 27.53% and 25.17%. For the average ultimate strength of the film, the value increased from 50.73 MPa to 51.78 MPa and then decreased to 42.13 MPa as the PEG content increased from 0.5 g to 0.6 g and 0.7 g. According to the study by de Melo Fiori et al. (2019), increasing PEG content in the film resulted in increase of elongation at break of the film as PEG which is low molecular weight acts as an internal spacer between the polymeric chain. The presence of PEG disrupts the interaction between the polymeric chain eventually causing the chain become more flexible. However, this study shows the opposite result, where the elongation at break of the film decreased when the PEG content increased, this may be due to PEG not dispersed well in the mixture during mixing. Thus, the linking between polymer chains are not well distributed for better mechanical properties when PEG content increased. In short, the amount of PEG in composite film need to control at its optimum ratio to ensure uniform distribution and homogeneous interaction with CMC and PGV structure.

Table 4.5: Summaries of film tensile properties at various PEG content.

No	Film	Average Tensile modulus (MPa)	Average Ultimate strength (MPa)	Average Elongation at break (%)
1	CMC (control)	993.67	47.12	41.80
2	2CMC-0.6PEG	579.67	29.88	36.63
3	2CMC-0.5PEG-0.2PGV	1162.67	50.73	29.37
4	2CMC-0.6PEG-0.2PGV	973.67	51.78	27.53
5	2CMC-0.7PEG-0.2PGV	828.67	42.13	25.17

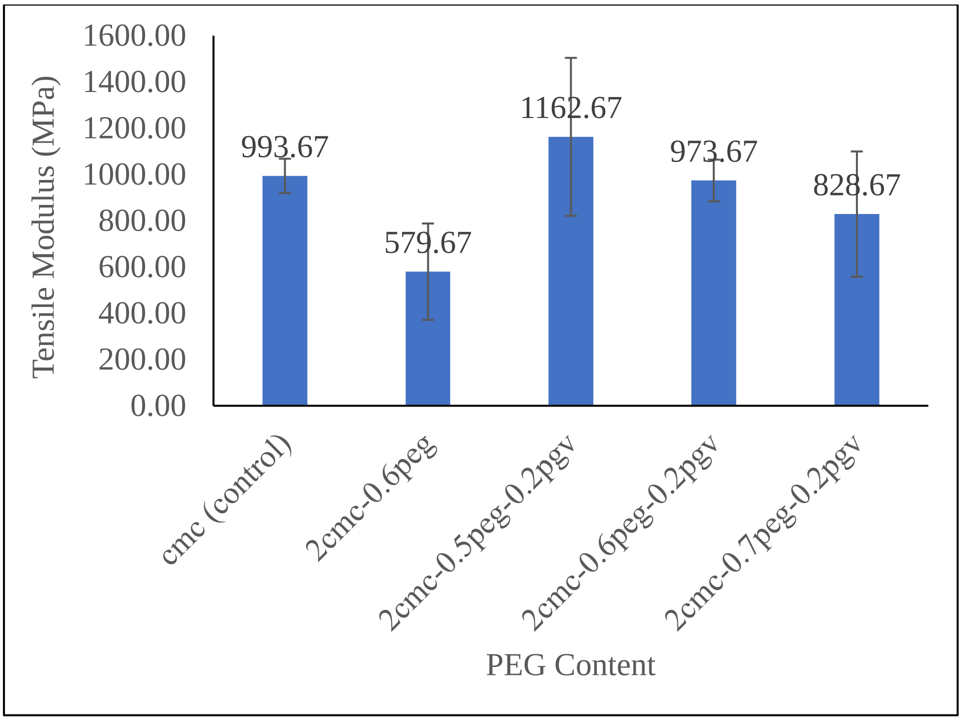

Figure 4.17: Tensile modulus of the film at various PEG content.

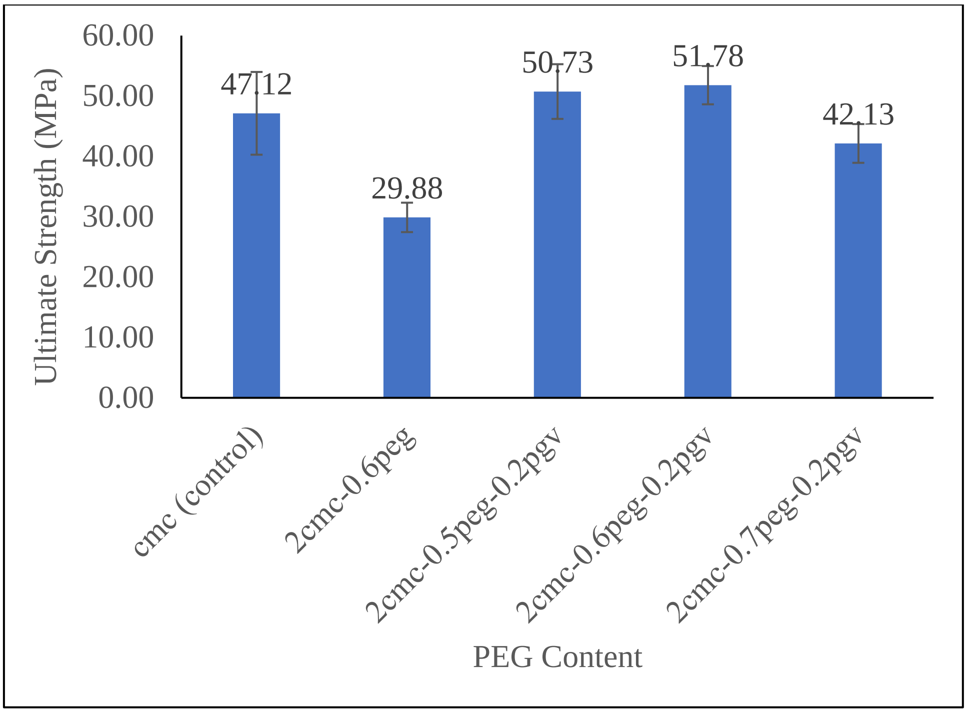

Figure 4.18: Ultimate strength of the film at various PEG content.

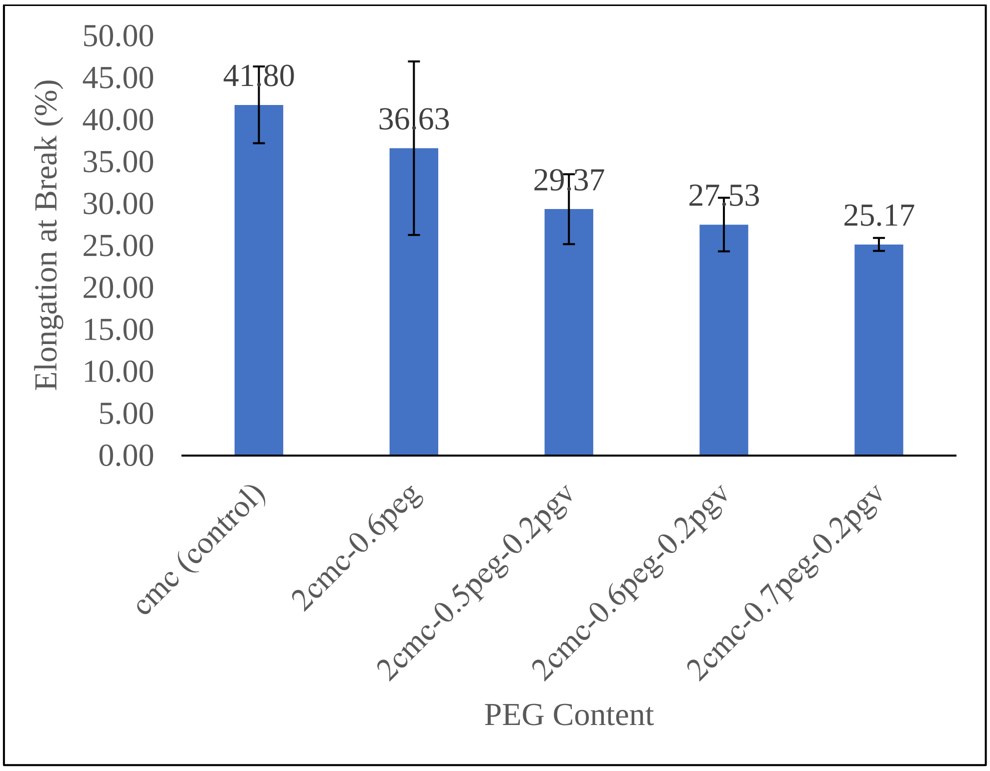

Figure 4.19: Elongation at break of the film at various PEG content.

4.4.3 Thermogravimetric Analysis (TGA)

From **Figure 4.20, 4.21 and 4.22**, it could be observed that when PEG content increased from 0.5 g to 0.6 g and 0.7 g, there were very small changes in residue percentage after the third step thermal decomposition which were 32.8505%, 32.5351% and 33.8598% respectively. It could be said that the addition of PEG to the film formulation gives almost no effect on the thermal property of the film. Basu, et al. (2018) obtained the similar trend in their study indicating that the thermal property of CMC film was not influenced by the presence of PEG.

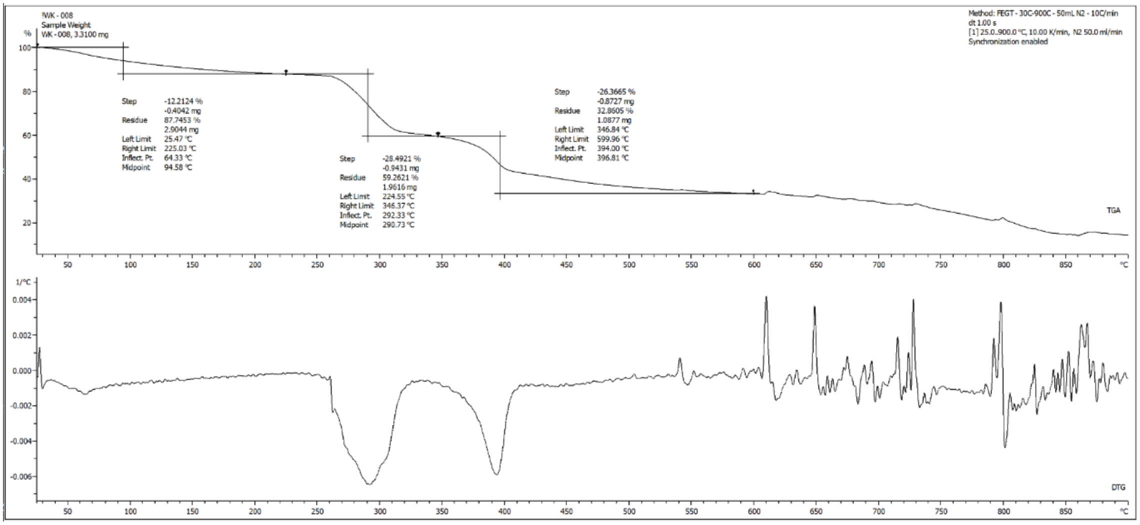

Figure 4.20: TGA (top) and DTG (bottom) thermograms of 2CMC-0.5PEG-0.2PGV film.

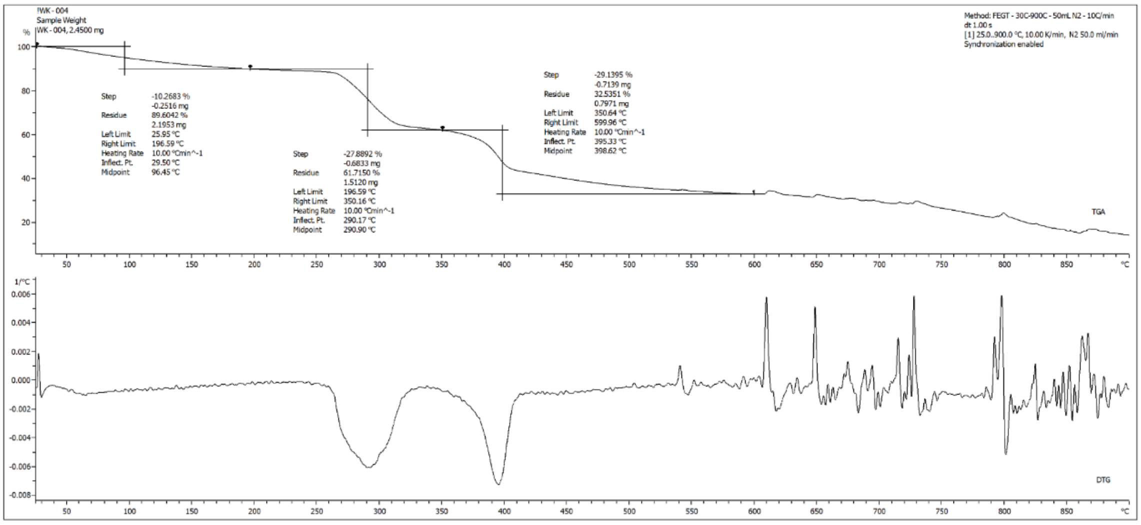

Figure 4.21: TGA (top) and DTG (bottom) thermograms of 2CMC-0.6PEG-0.2PGV film.

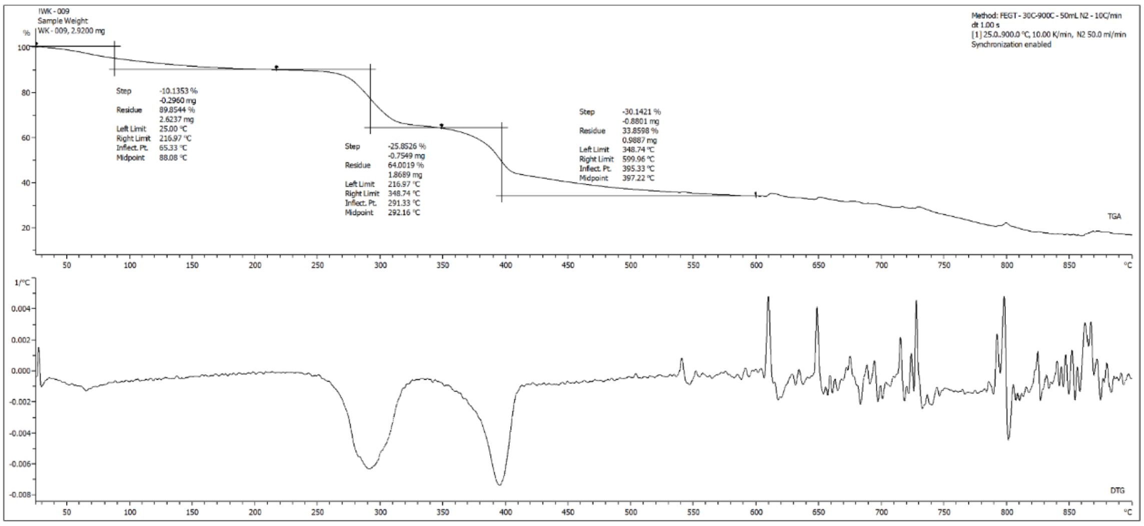

Figure 4.22: TGA (top) and DTG (bottom) thermograms of 2CMC-0.7PEG-0.2PGV film.

4.4.4 Water Solubility

Figure 4.23 shows the graph of time taken for the film at various PEG content to dissolve in water**.** As compared to the pure CMC film in this study, adding PEG to the film could increase the time taken to dissolve in water by 4 minutes. As the content of PEG increased from 0.5 g to 0.6 g and 0.7 g, the films take 14, 15 and 16 minutes to dissolve in water, which the time taken for the film to dissolve in water is slightly increased with the PEG content. The study by de Melo Fiori, et al. (2019) and Giyatmi, et al. (2020) showed the similar trend which stated that the addition of PEG not only act as plasticizers, it could also decrease the WVP as well but the effect was not significant.

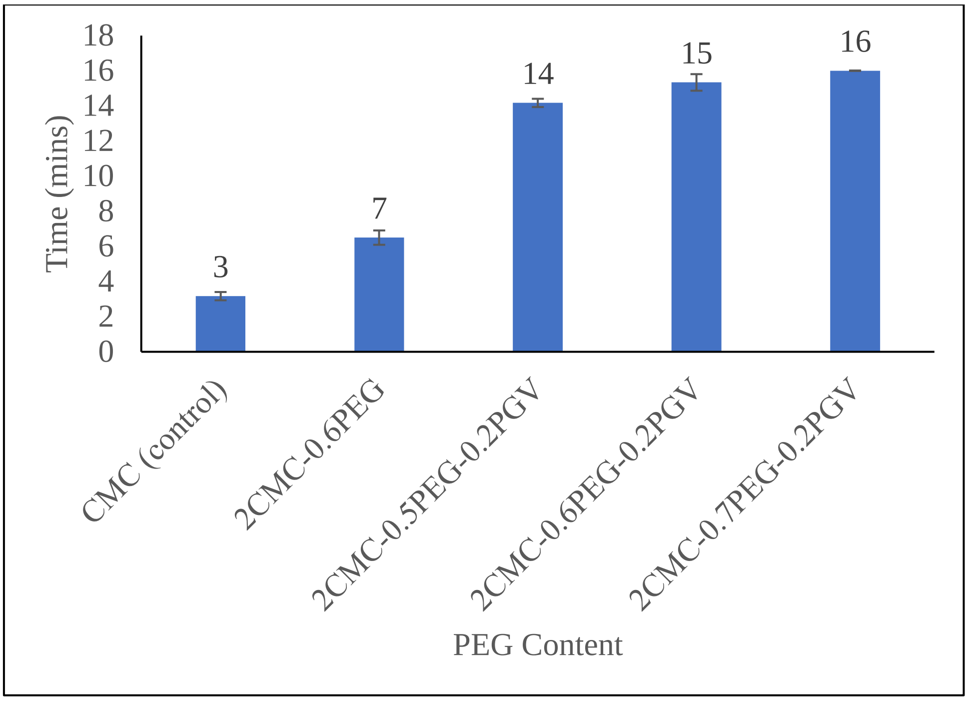

Figure 4.23: Time taken for the film at various PEG content to dissolve in water.

4.5 Effect of PGV Content to Film Properties

4.5.1 Functional Group Analysis using Attenuated Total Reflectance- Fourier Transform Infrared Spectroscopy (ATR-FTIR)

Figure 4.24 shows the ATR-FTIR of pure CMC film, 2CMC-0.6PEG film, and CMC composite film with various PGV content. It could be observed that there is no new functional group presented in the CMC composite film, which means that there is no chemical reaction occurred during mixing or sonication process. Besides, as the PGV content increases from 0.1 g to 0.2 g and 0.3 g, the Si-O stretching bands at around 400 – 500 cm^{-1} become slightly deeper, which indicating that the number of Si-O bonds increases as the PGV content increases.

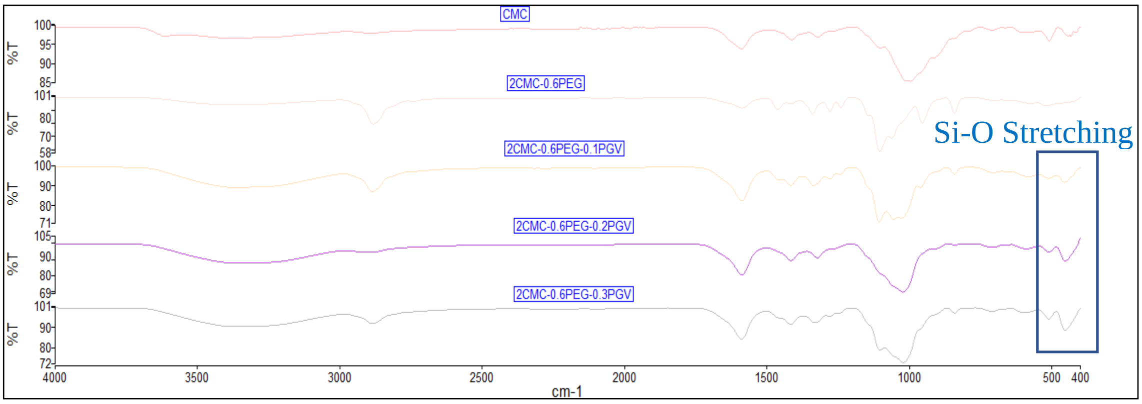

Figure 4.24: ATR-FTIR of pure CMC film and CMC composite film with various PGV content.

4.5.2 Mechanical Properties

Table 4.6 summarized the tensile properties of the film at various PGV content. While **Figure 4.25, 4.26 and 4.27** show the graph of tensile modulus, ultimate strength, and elongation at break of the film samples at various PGV content. For the pure CMC film, the average tensile modulus, ultimate strength, and elongation at break were 993.67 MPa, 47.12 MPa and 41.80% respectively. As the PGV content increased from 0.1 g to 0.2 g and 0.3 g, the tensile modulus of the film increased from 858.33 MPa to 1032 MPa and 1450 MPa. For the average ultimate strength, the value firstly increased from 48.69 MPa to 51.78 MPa and then decreased to 42.61 MPa as the PGV content increased from 0.1 g to 0.2 g and 0.3 g. While the average elongation at break of the film decreased from 31.07% to 27.53% and 24.17% when PGV content increased from 0.1 g to 0.2 g and 0.3 g, respectively. The previous study from de Oliveira, et al. (2015) and de Melo Fiori, et al. (2019) obtained the similar trend for tensile modulus and elongation at break of the film, which increases the content of PGV resulting in increases of tensile modulus and decreases of elongation at break of the film. The increment in tensile strength was due to the ultra-high interfacial interaction was occurred between the nanoclay and the host polymer through hydrogen bonds when the nanoclay particles dispersed uniformly in the polymer matrix (Rhim, Hong and Ha, 2009). He, Fei and Li (2019) also indicated that the decrement in elongation at break was owing to the film becoming rigid, brittle, and less flexible when more PGV was added.

Table 4.6: Summaries of film tensile properties at various PGV content.

No	Film	Average Tensile modulus (MPa)	Average Ultimate strength (MPa)	Average Elongation at break (%)
1	CMC (control)	993.67	47.12	41.80
2	2CMC-0.6PEG	579.67	29.88	36.63
3	2CMC-0.6PEG-0.1PGV	858.33	48.69	31.07
4	2CMC-0.6PEG-0.2PGV	1032.00	51.78	27.53
5	2CMC-0.6PEG-0.3PGV	1450.00	42.61	24.17

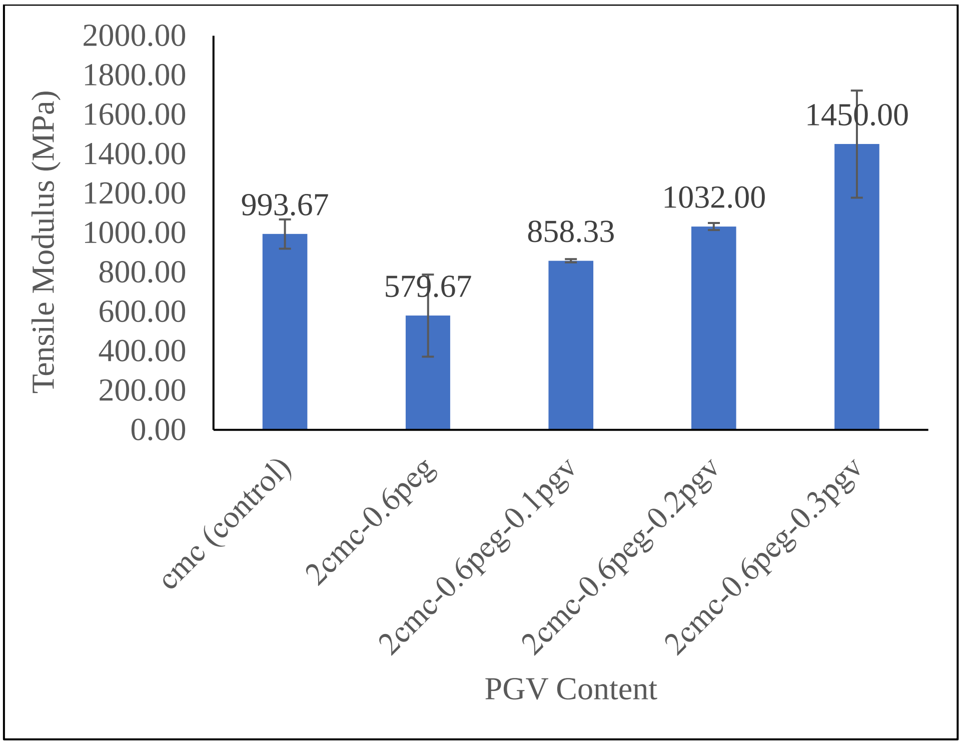

Figure 4.25: Tensile modulus of the film at various PGV content.

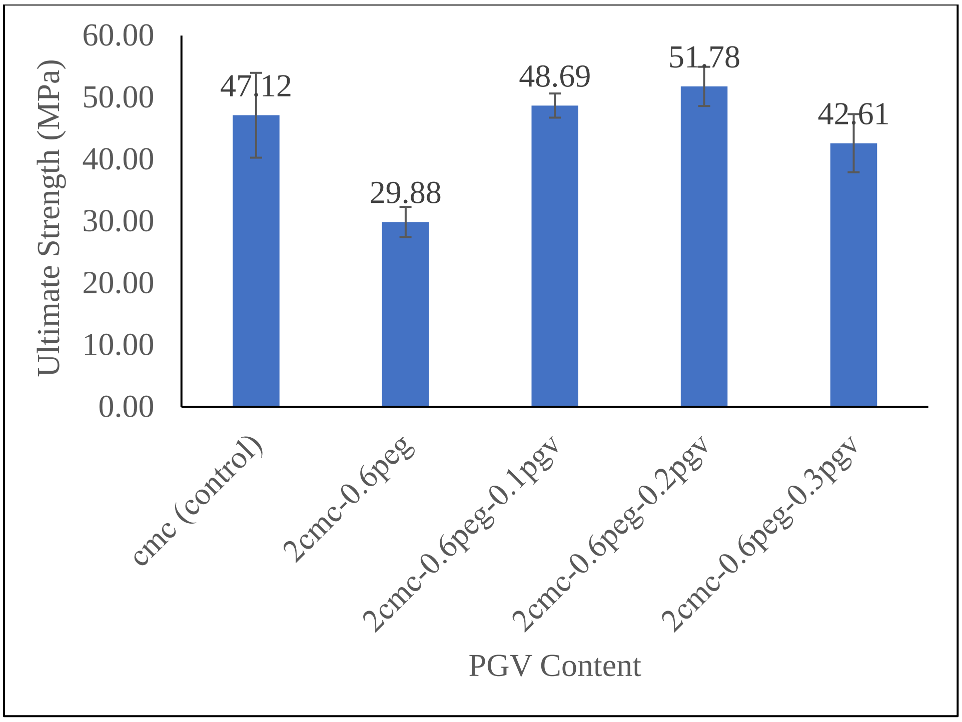

Figure 4.26: Ultimate strength of the film at various PGV content.

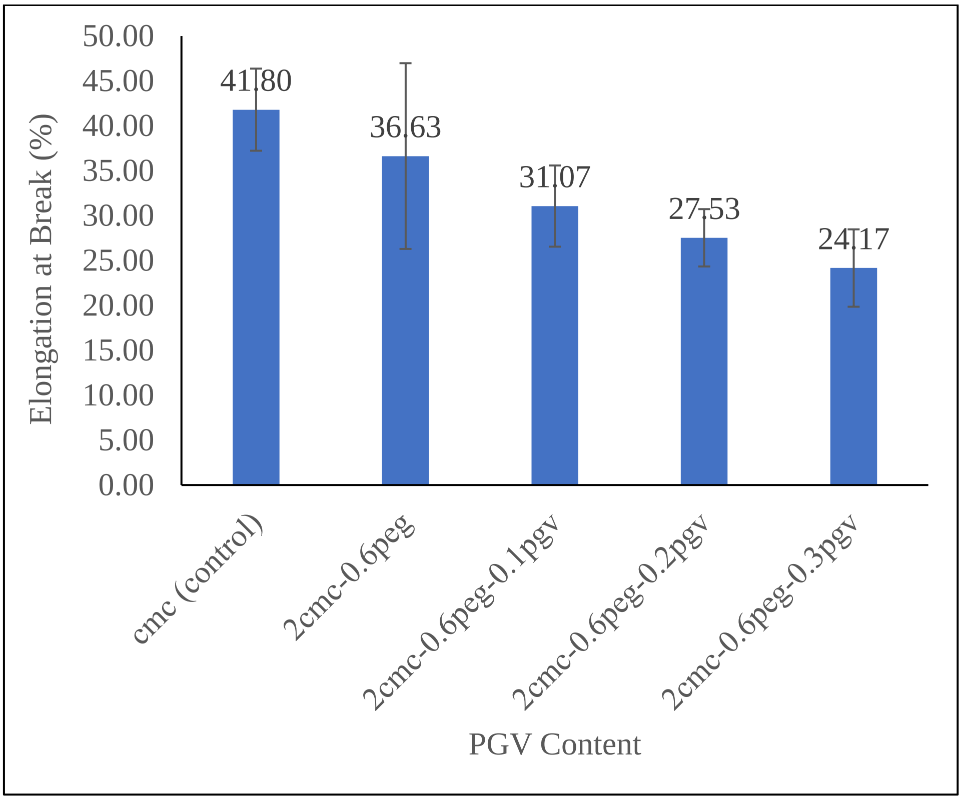

Figure 4.27: Elongation at break of the film at various PGV content.

4.5.3 Thermogravimetric Analysis (TGA)

From **Figure 4.28, 4.29 and 4.30**, it could be observed that as the PGV content increased from 0.1g to 0.2 g and 0.3g, the residue percentage have fluctuated between 33.6998% to 32.5351% and 33.9057% after the third step of thermal decomposition. The result showed almost no effect of adding PGV to the film. According to Saha, et al. (2016) and Mohamed, et al. (2016), the film thermal stability would increase with the addition of MMT into the CMC matrix owing to the good heat insulation capacities of MMT. However, in this study, the opposite result was obtained which the incorporation of PGV nanoclay (one of the types of MMT) into the CMC matrix did not show an improvement in thermal stability of CMC composite film. This may be due to the inhomogeneous dispersion of PGV nanoclay in the polymer matrix, where white powder is observed on film.

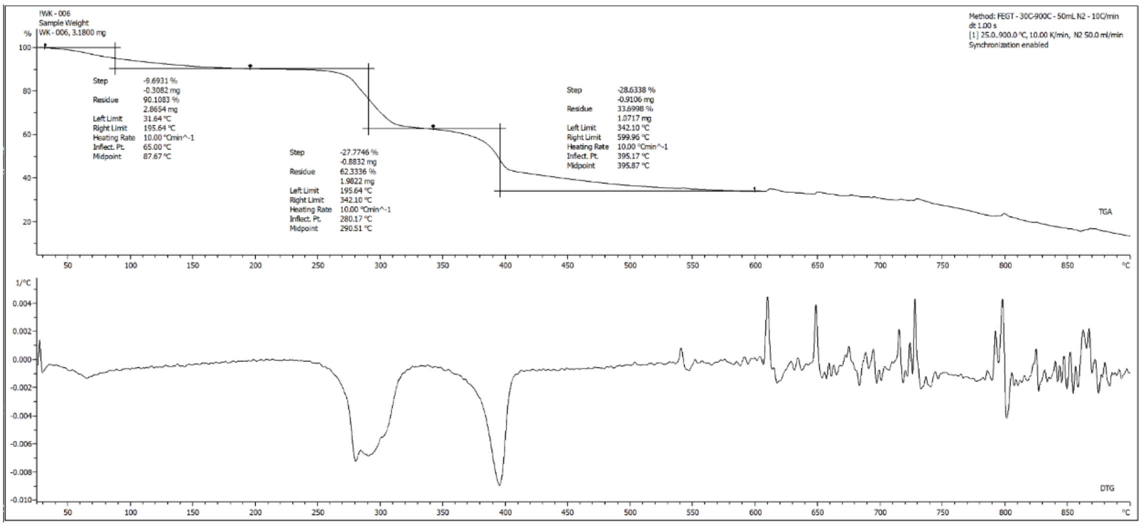

Figure 4.28: TGA (top) and DTG (bottom) thermograms of 2CMC-0.6PEG-0.1PGV film.

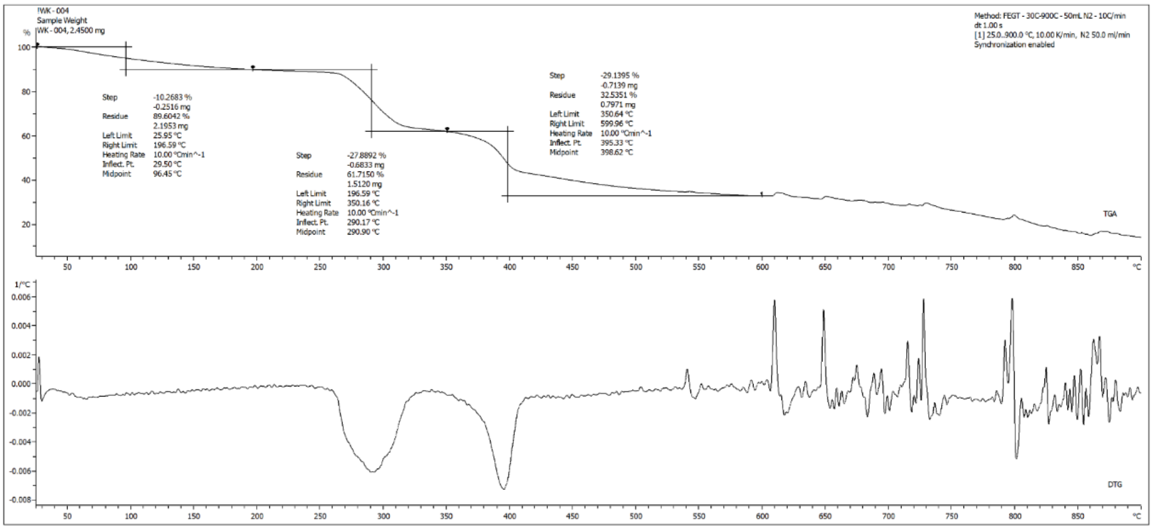

Figure 4.29: TGA (top) and DTG (bottom) thermograms of 2CMC-0.6PEG-0.2PGV film.

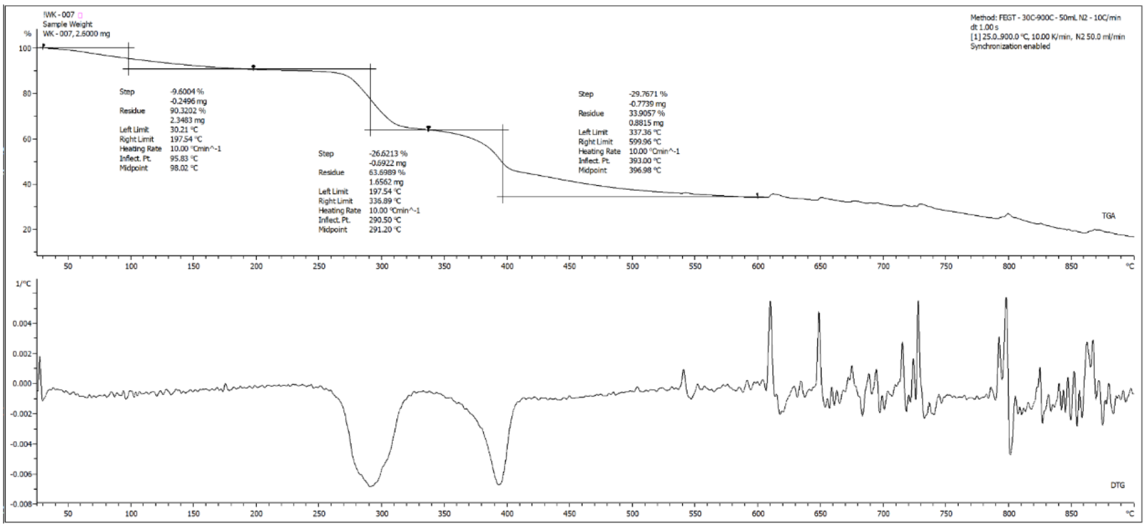

Figure 4.30: TGA (top) and DTG (bottom) thermograms of 2CMC-0.6PEG-0.3PGV film.

4.5.4 Water Solubility

Figure 4.31 shows the graph of time taken for the film at various PGV content to dissolve in water**.** As the content of PGV nanocomposite increased from 0.1 g to 0.2 g and 0.3 g, the time taken for the film to dissolve in water increases from 11 to 15 and 17 minutes. The result showed that the time taken for the film to dissolve in water increasing with the PGV content. Similar trend was obtained in the study from de Melo Fiori, et al. (2019) and de Oliveira, et al. (2015). The studies presented that increasing of nanocomposite resulted in decrease of WVP of the film due to the nanocomposite act as filler to fill the void space and acts as physical blockage to increase the water resistance of the film. In addition, the study from He, Fei and Li (2019) indicated that water vapor barrier properties of the film could be enhanced if the nanofiller is well dispersed in the polymer matrix. In this study, PGV decreased the water solubility of the film by filling the void space of the polymer matrix and acts as physical barrier for the passage of water (de Oliveira, et al., 2015).

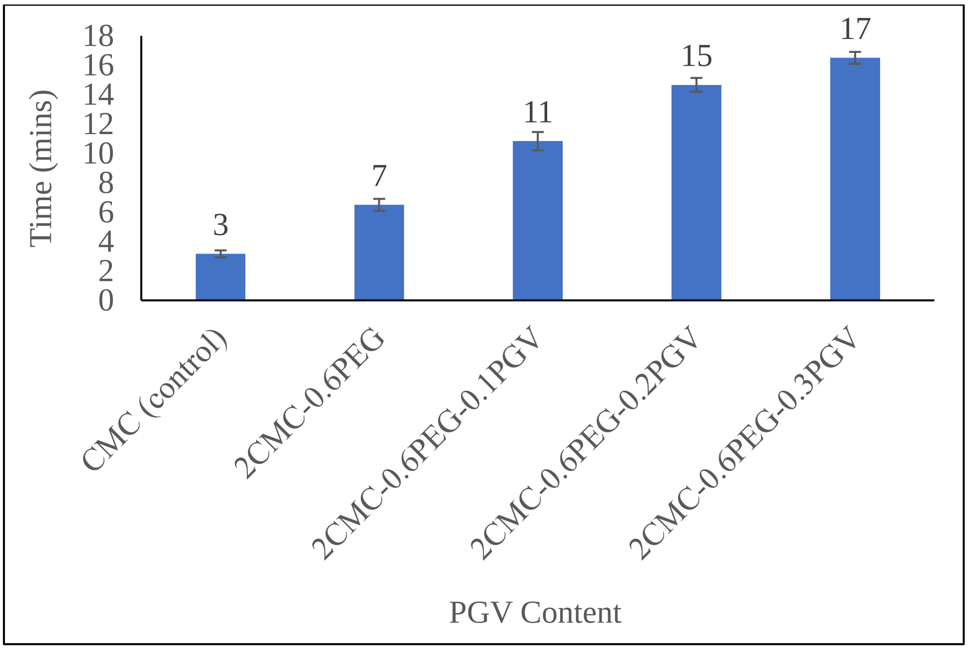

Figure 4.31: Time taken for the film at various PGV content to dissolve in water.

4.6 Comparison with Commercial PE Film

A comparison of properties among three film was shown in **Table 4.7**. As compared to the other two CMC composites, 2CMC-0.6PEG has a lower tensile strength which is 579.67 MPa and took a shorter time which is 7 minutes to dissolve in water. While 1CMC-0.6PEG-0.2PGV performed moderately in all the properties which are 1002.67 MPa of tensile strength, 30.63% of elongation at break, 34.3475% of residue after the third step of thermal decomposition and took 13 minutes to dissolve in water. In this study, the optimum film was 3CMC-0.6PEG-0.2PGV as it performed better in tensile strength and water resistance. The tensile strength of the film was 1431 MPa and it took a longer time which is 17 minutes to dissolve in water. However, as compared to the commercial PE film, the water resistance and elongation at break of 3CMC-0.6PEG-0.2PGV were still too far from the commercial PE film. Hence, in order to be applied as commercial film, the water resistance and elongation at break of 3CMC-0.6PEG-0.2PGV is needed to be improved.

Table 4.7: Comparison of the film with different properties.

Film properties	**2CMC-0.6PEG**	**1CMC-0.6PEG-0.2PGV**	**3CMC-0.6PEG-0.2PGV**	**Commercial PE film**
Tensile strength	579.67 MPa	1002.67 MPa	1431.00 MPa	126 MPa
Elongation at break	36.63%	30.63%	27.30%	432.25%
Thermal stability	34.3475% Residue	29.3055% Residue	33.3076% Residue	17.2132% Residue
Water solubility	7 mins	13 mins	17 mins	> 24 hours

CHAPTER 5

CONCLUSION AND RECOMMENDATION

5.1 Conclusion

Characterization test of CMC, nanoclay PGV and PEG were successfully conducted via ATR-FTIR analysis and physical observation. The presence of hydroxyl group in CMC and PGV could form a strong hydrogen bond when mix and form film together. The pure CMC film had the highest transparency followed by plasticized CMC film (2CMC-0.6PEG film) and CMC film incorporated with nanoclay (2CMC-0.6PEG-0.2PGV).

The second objective of this study was to investigate the effect of mixture formulation on the properties of biocomposite film. This objective was achieved by preparing mixture at various formulation that successfully formed the film through casting method. The CMC content was varied from 1 g to 2 g and 3 g, the PEG content was varied from 0.5 g to 0.6 g and 0.7 g, and the PGV content was varied from 0.1 g to 0.2 g and 0.3 g.

There are four characterization tests have been done on the CMC composite film which are ATR-FTIR analysis, tensile test, TGA and water solubility test. In ATR-FTIR analysis, there is no new functional group in the CMC-PEG-PGV film, where all the functional groups in CMC composite film could be found in CMC, PEG and PGV powder. In other words, there is no chemical reaction happened during the mixing process. In the tensile test, the tensile strength of the film could be improved by increasing the CMC content and PGV content. This is because increasing the CMC and PGV content could form more hydrogen bonds between inorganic PGV nanoclay and hydroxyl group of organic CMC. The hydrogen bonds could significantly improve the tensile strength of the film. In TGA analysis, this study showed the thermal stability of the film could be improved by increasing the CMC content. This is because CMC acts as a stabilizer to provide thermal stability to the film by decreasing the weight loss during heating. In the water solubility test, the time taken for the film to dissolve in water could be increased by increasing the CMC and PGV content. As the CMC content increased, there is more hydrogen bond was formed through interfacial interaction between the hydroxyl group of CMC and inorganic PGV, hence the water resistance of the film was increased. Besides, PGV nanoclay act as filler to fill the void space in the polymer matrix and acts as physical blockage to increase the water resistance of the film.

The last objective which is to compare the CMC composite film and commercial plastic film was achieved by comparing the properties of the optimum CMC composite film and commercial PE film. In this study, the optimum film was 3CMC-0.6PEG-0.2PGV with tensile strength of 1431 MPa, elongation at break of 27.3%, residue percentage of 33.3076% after third step of thermal decomposition and took 17 minutes to dissolve in water. Compared to the commercial PE film with tensile strength of 126 MPa, elongation at break of 432.25%, residue percentage of 17.2132% after third step of thermal decomposition and took more than 24 hours to dissolve in water. From the comparison between 3CMC-0.6PEG-0.2PGV film and commercial PE film, it could be observed that 3CMC-0.6PEG-0.2PGV film has a lower elongation at break and water resistance. Elongation at break of the film is important for the commercial film so that the film would not break easily when the materials with some weights were inside the film. In addition, the water resistance of the commercial film was important especially for its in packaging the frozen food and drinks. Hence, to be used as a commercial biocomposite film, the 3CMC-0.6PEG-0.2PGV film still need improvement in elongation at break and water resistance properties.

5.2 Recommendation

There was some recommendation to improve the mechanical properties and water resistance of the film. First of all, XRD and SEM could be conducted on all the CMC composite films to analyze the crystalline structure and distribution of the particle in the film. The SEM analysis could investigate whether the distribution of PGV nanoclay was homogenous (De Oliveira, et al., 2015).

Based on the SEM and TEM analysis, some improvement needed to be done to achieve a homogeneous mixture. In order to ensure the particle is distributed homogeneously in the mixture, the mixing time and sonication time should be increased. This is because when the sonication duration was longer, the sonication intensity of the input energy is higher, thus the nanoparticles could be separated and better dispersion quality of solution could be achieved (Gou, Zhuge and Liang, 2012).

Last but not least, as mentioned in the conclusion that the film, which is 3CMC-0.6PEG-0.2PGV film was still low in properties of elongation at break and water resistance as compared to commercial PE film. Hence, it was recommended that the PEG content could be increased to improve the elongation at break of the film. This is because increasing PEG content could affect structure between the polymeric chain and eventually disrupts the initial linkage but build new interaction between the polymeric chain to make the film become more flexible (de Melo Fiori, et al., 2019). Besides, it was recommended that the PGV content could be increased to improve the water resistance of the film. This is because PGV nanoclay could significantly improve the water resistance of the film by filling the void space in the polymer matrix and acts as physical barrier to block the passage of water (de Oliveira, et al., 2015).

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APPENDICES

APPENDIX A: TENSILE PROPERTIES OF THE FILM

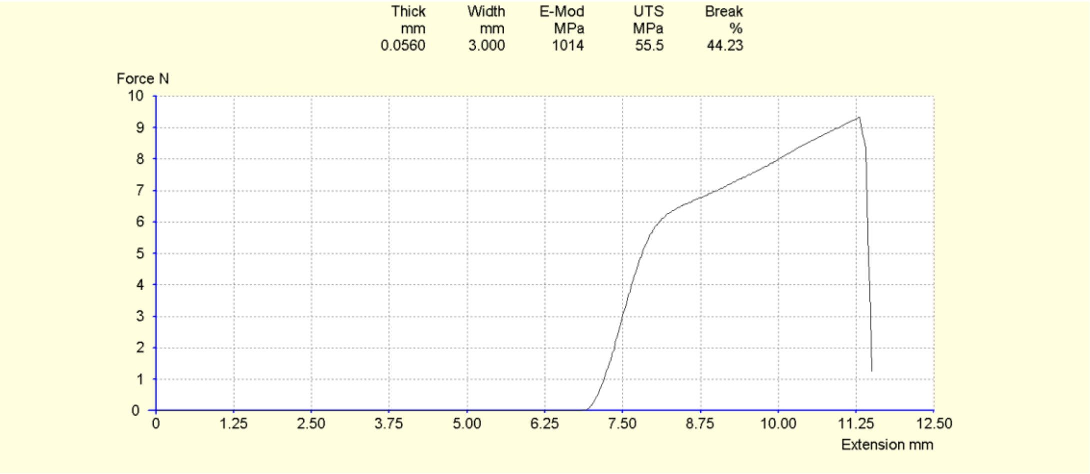


Figure A.1: First test of tensile properties of pure CMC film.

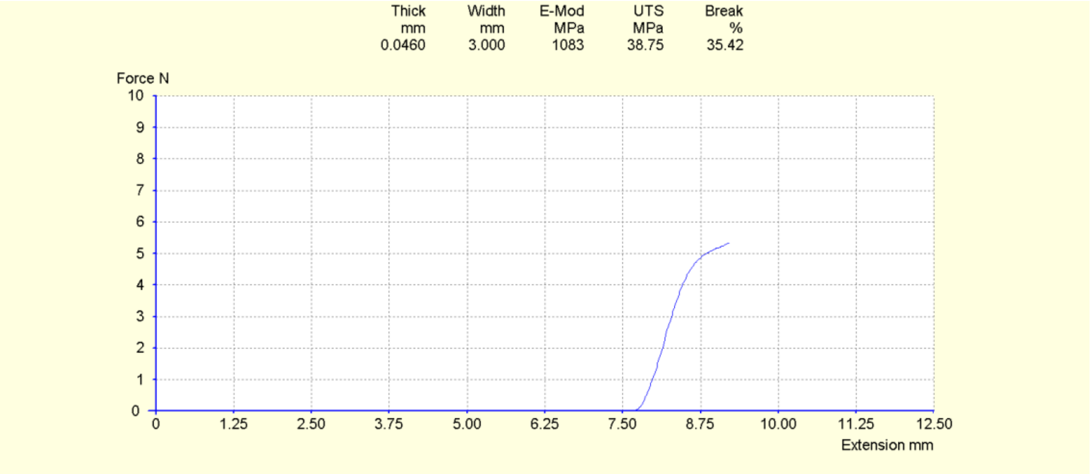


Figure A.2: Second test of tensile properties of pure CMC film.

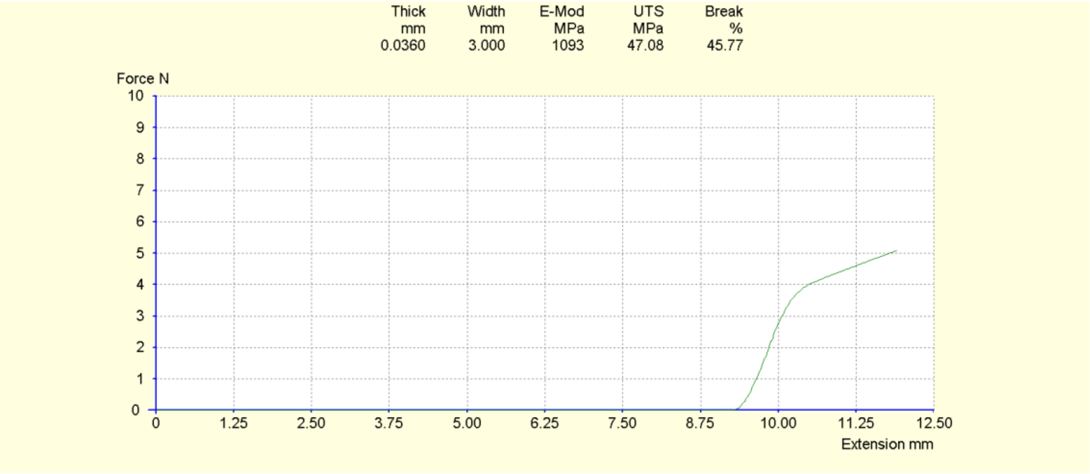


Figure A.3: Third test of tensile properties of pure CMC film.

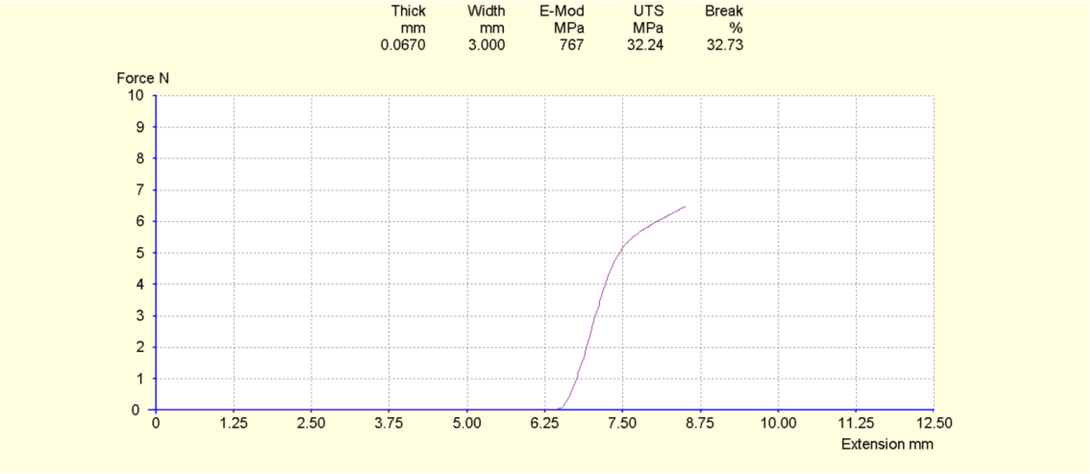


Figure A.4: First test of tensile properties of 2CMC-0.6PEG.

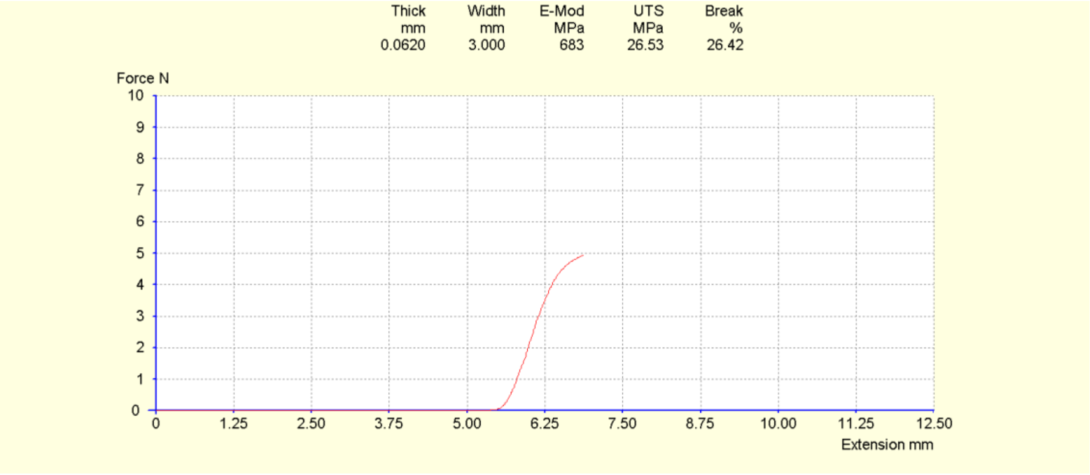
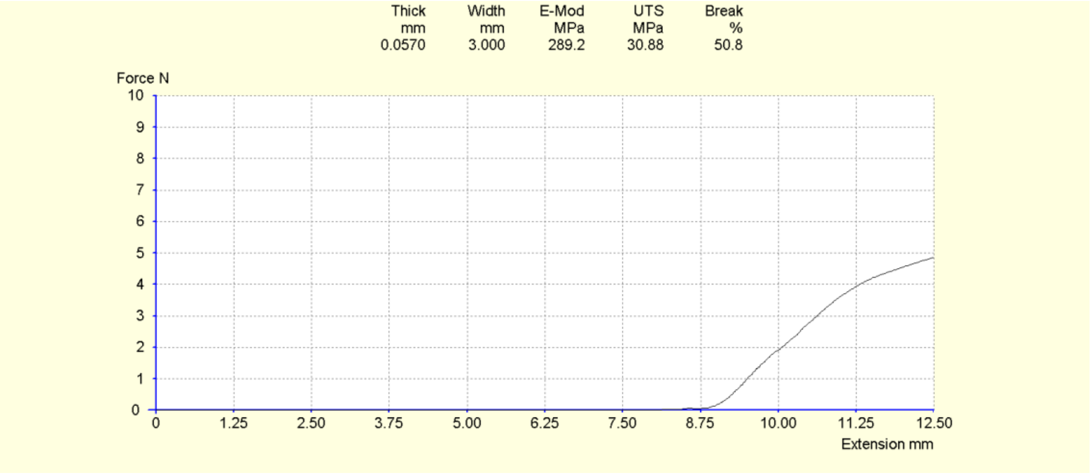
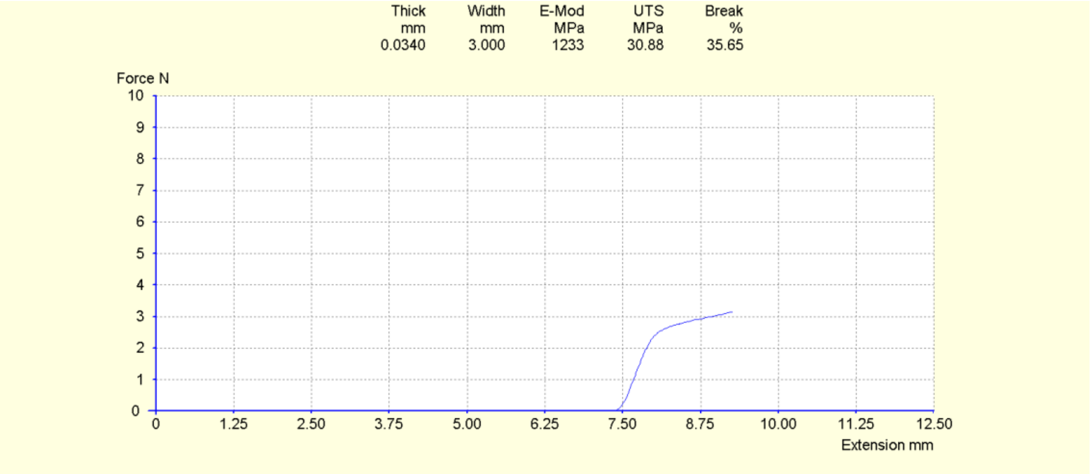


Figure A.5: Second test of tensile properties of 2CMC-0.6PEG.



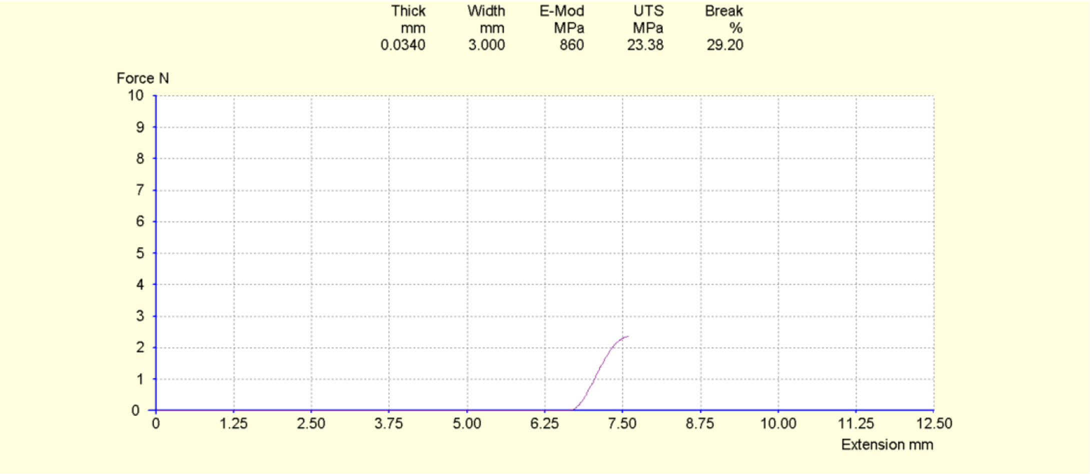
Thick mm	Width mm	E-Mod MPa	UTS MPa	Break %
0.0570	3.000	289.2	30.88	50.8

Figure A.6: Third test of tensile properties of 2CMC-0.6PEG.



Thick mm	Width mm	E-Mod MPa	UTS MPa	Break %
0.0340	3.000	1233	30.88	35.65

Figure A.7: First test of tensile properties of 1CMC-0.6PEG-0.2PGV film.



Thick mm	Width mm	E-Mod MPa	UTS MPa	Break %
0.0340	3.000	860	23.38	29.20

Figure A.8: Second test of tensile properties of 1CMC-0.6PEG-0.2PGV film.

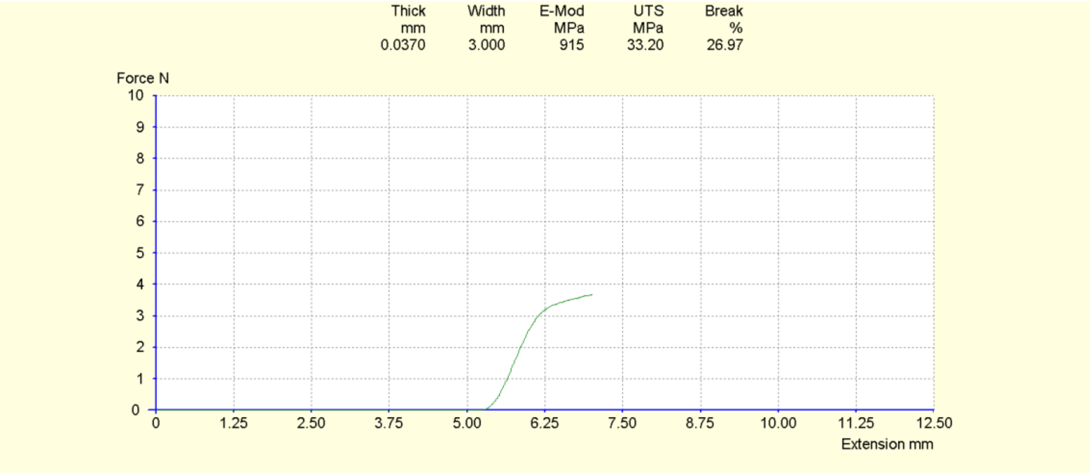


Figure A.9: Third test of tensile properties of 1CMC-0.6PEG-0.2PGV film.

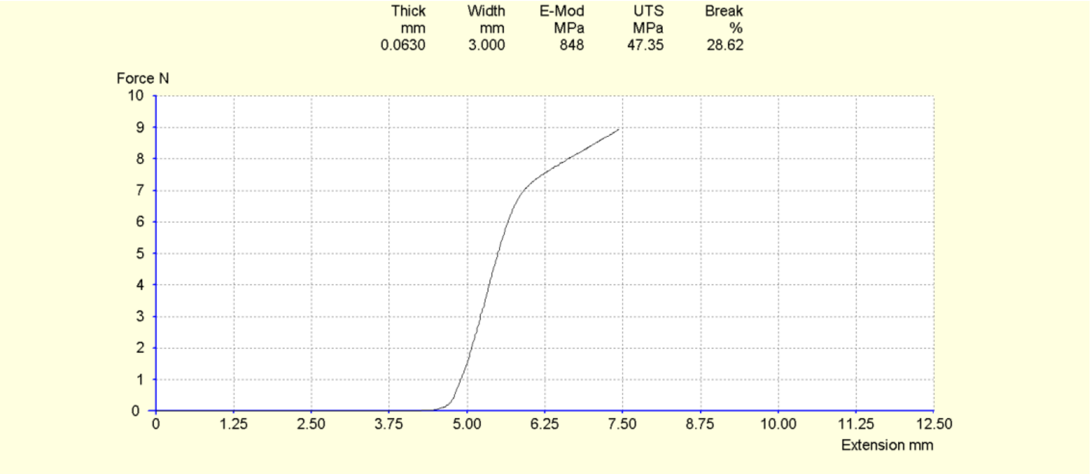


Figure A.10: First test of tensile properties of 2CMC-0.6PEG-0.2PGV film.

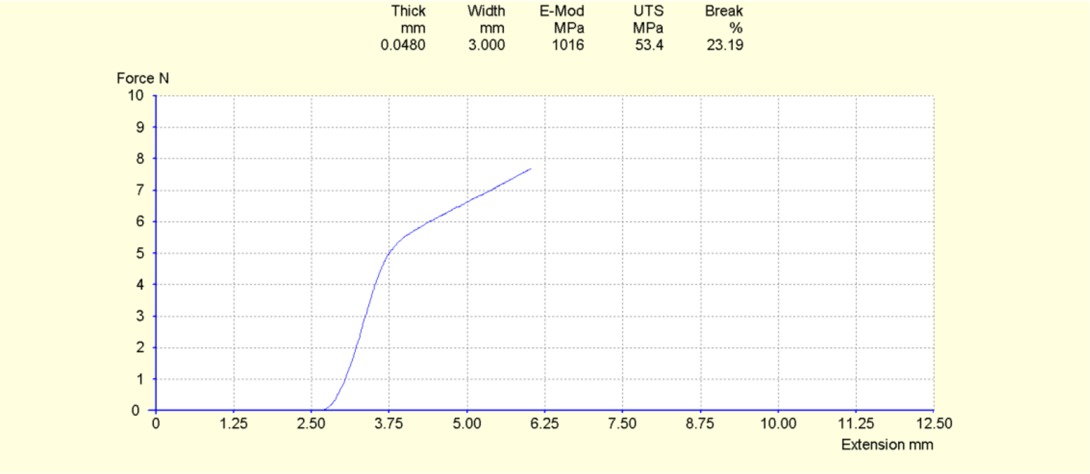


Figure A.11: Second test of tensile properties of 2CMC-0.6PEG-0.2PGV film.

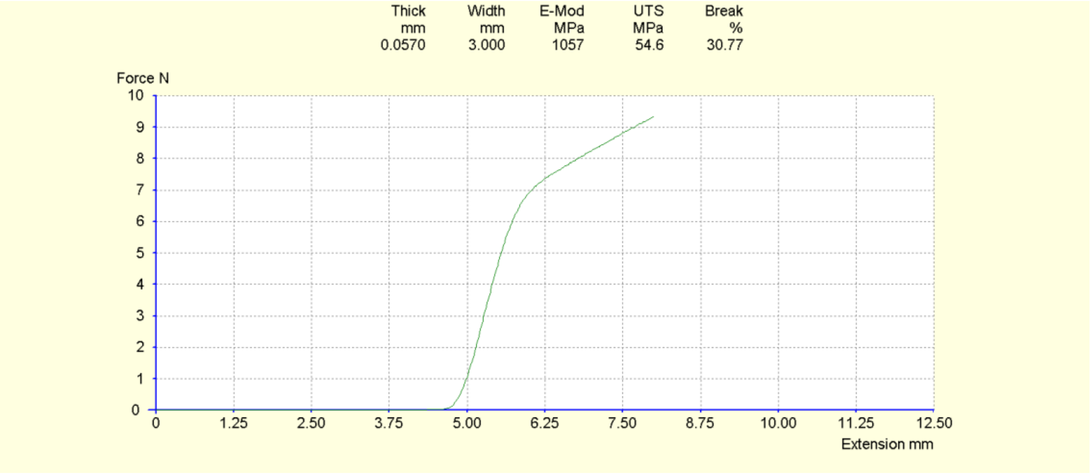


Figure A.12: Third test of tensile properties of 2CMC-0.6PEG-0.2PGV film.

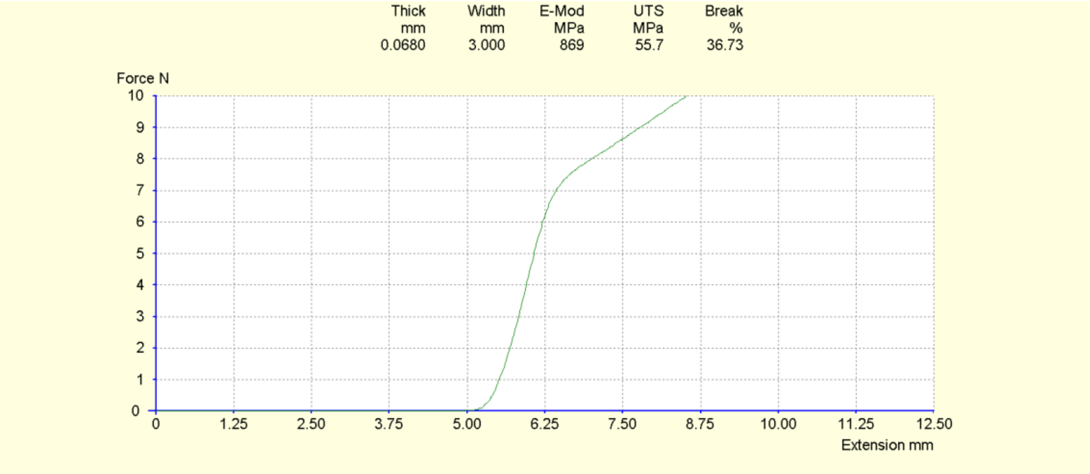


Figure A.13: First test of tensile properties of 3CMC-0.6PEG-0.2PGV film.

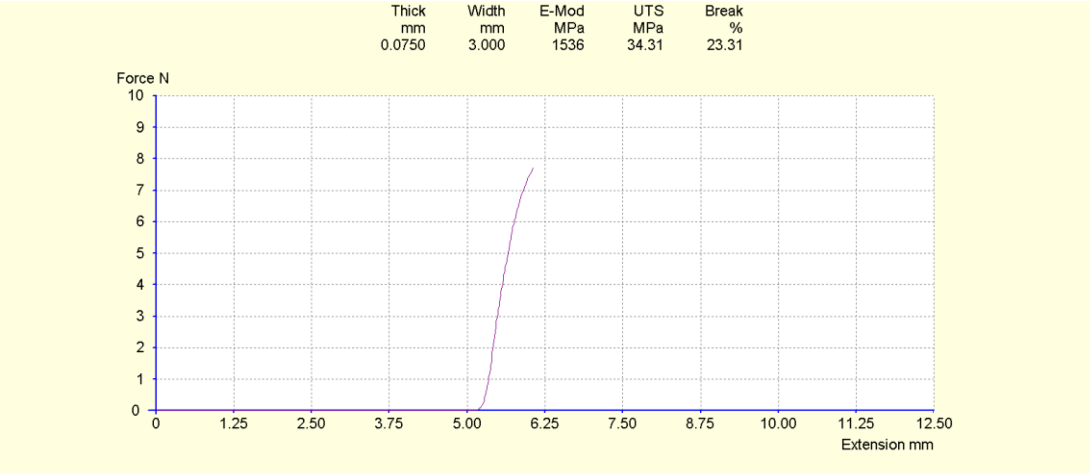


Figure A.14: Second test of tensile properties of 3CMC-0.6PEG-0.2PGV film.

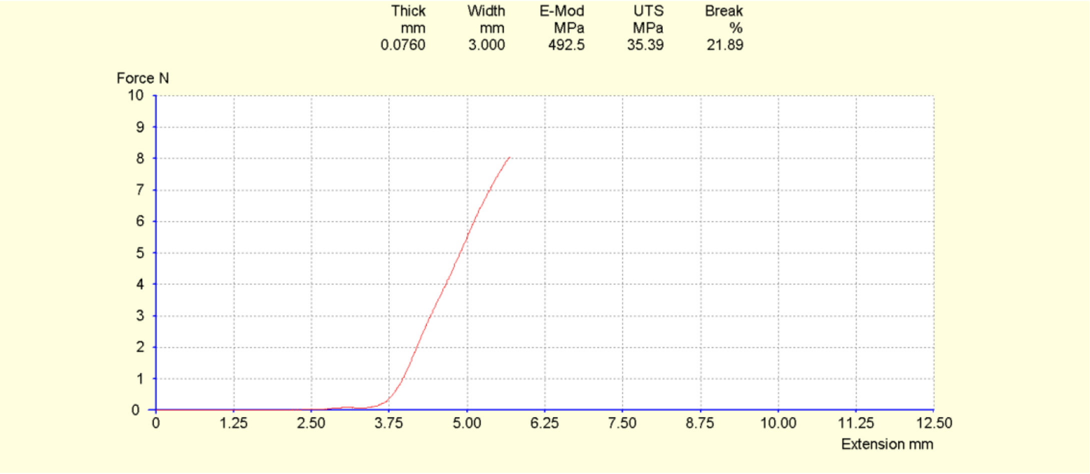


Figure A.15: Third test of tensile properties of 3CMC-0.6PEG-0.2PGV film.

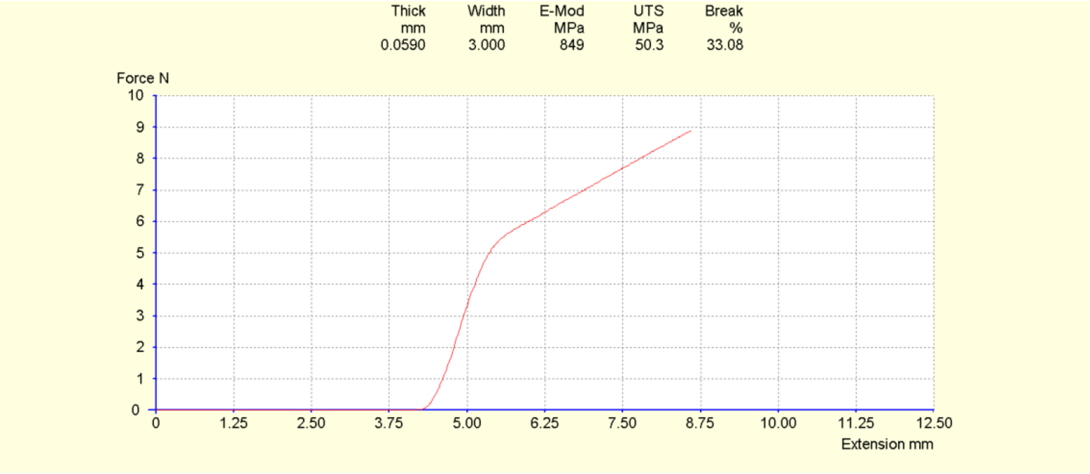


Figure A.16: First test of tensile properties of 2CMC-0.6PEG-0.1PGV film.

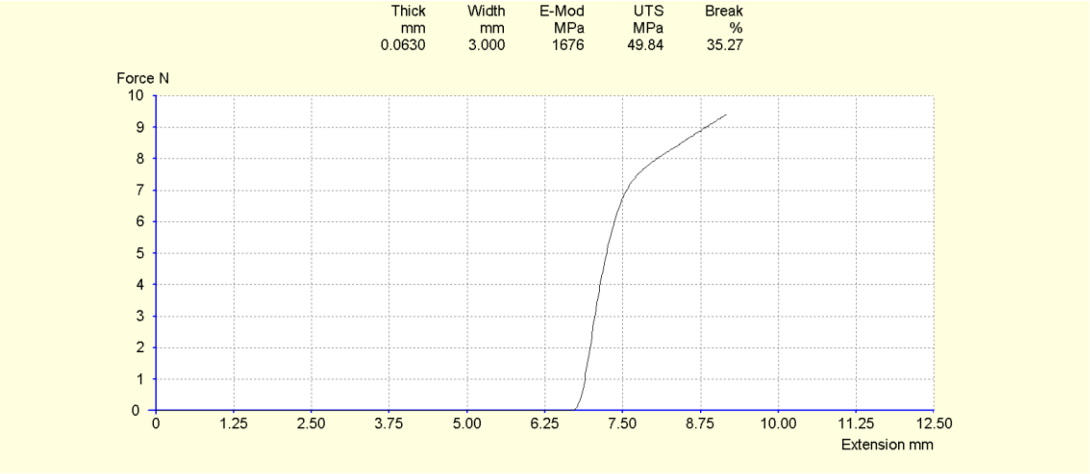


Figure A.17: Second test of tensile properties of 2CMC-0.6PEG-0.1PGV film.

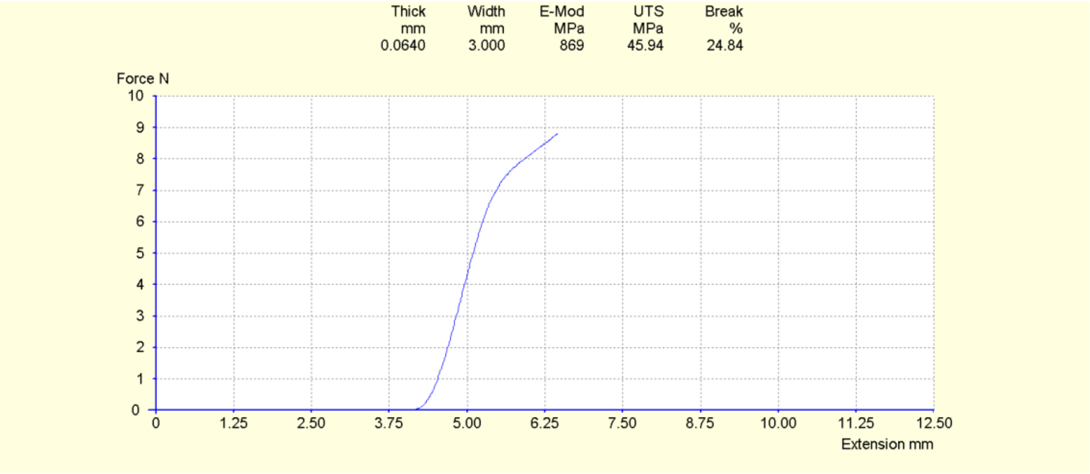


Figure A.18: Third test of tensile properties of 2CMC-0.6PEG-0.1PGV film.

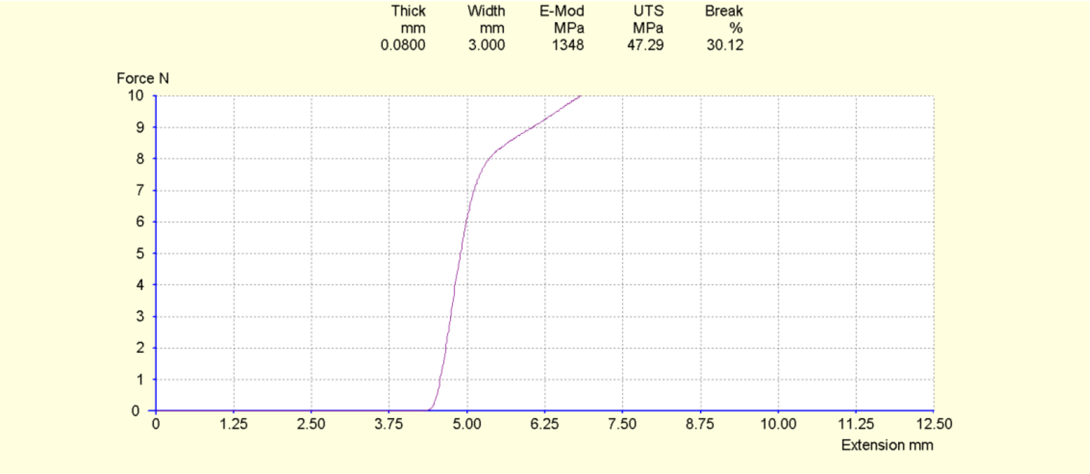


Figure A.19: First test of tensile properties of 2CMC-0.6PEG-0.3PGV film.

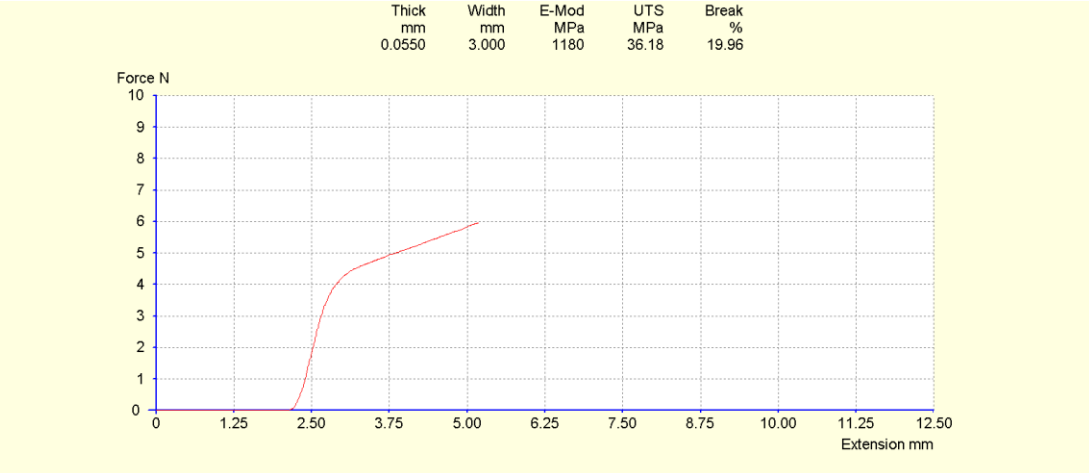


Figure A.20: Second test of tensile properties of 2CMC-0.6PEG-0.3PGV film.

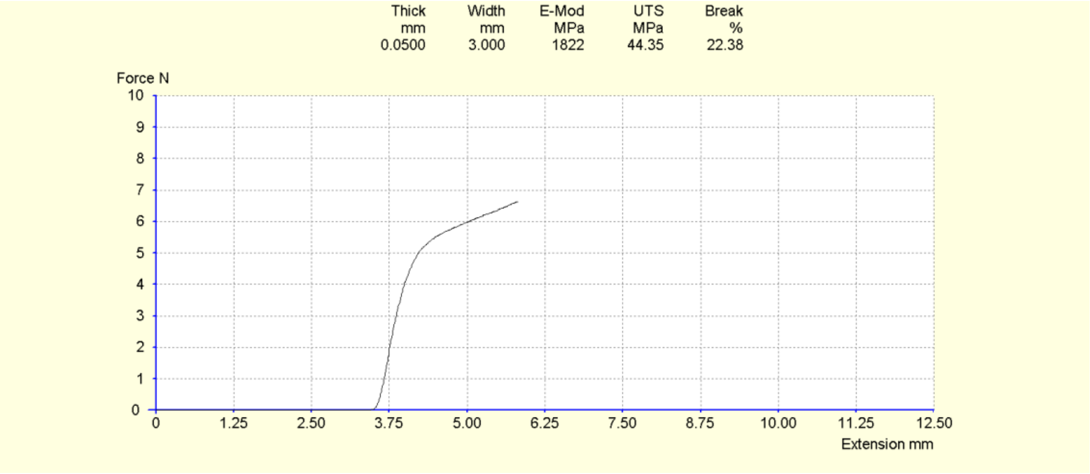


Figure A.21: Third test of tensile properties of 2CMC-0.6PEG-0.3PGV film.

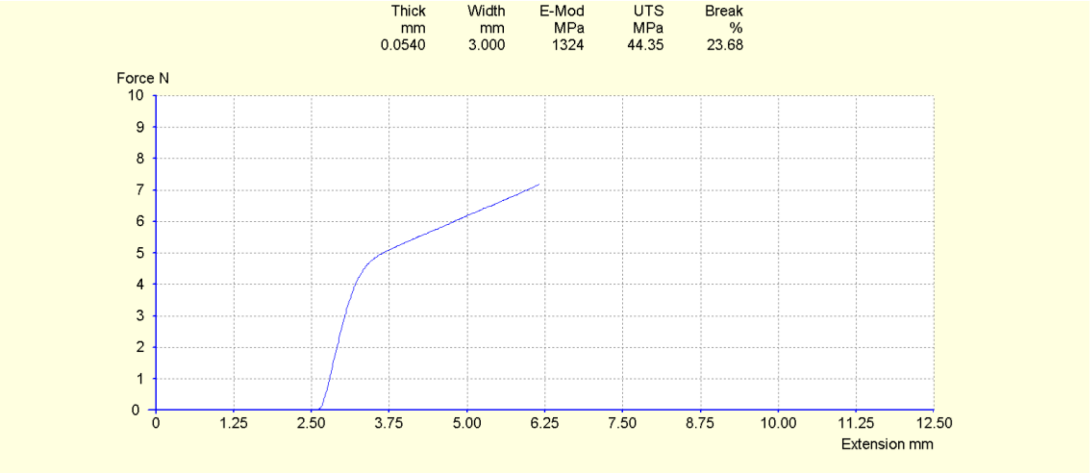


Figure A.22: First test of tensile properties of 2CMC-0.5PEG-0.2PGV film.

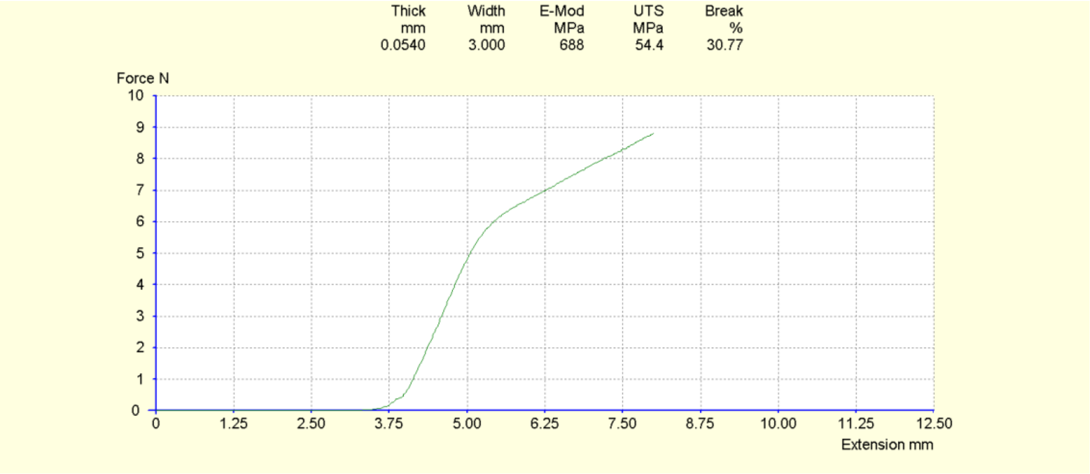


Figure A.23: Second test of tensile properties of 2CMC-0.5PEG-0.2PGV film.

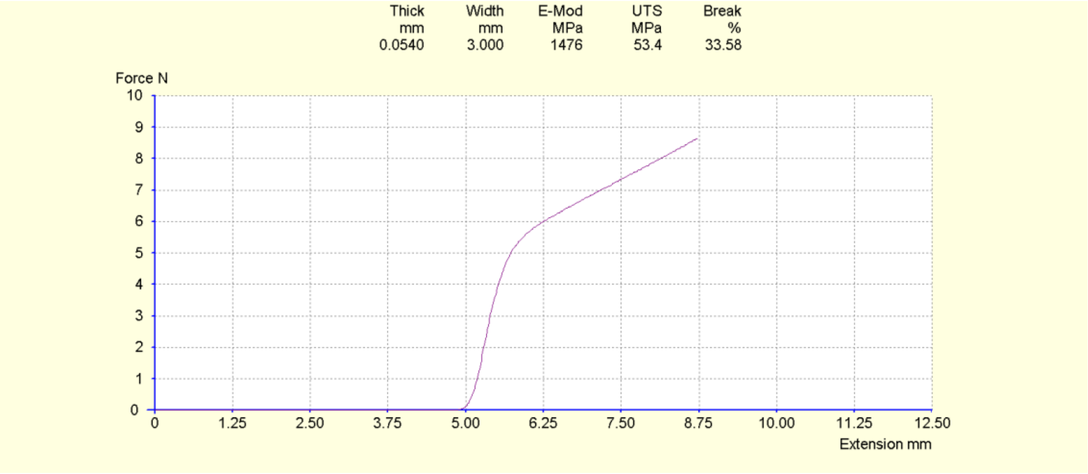


Figure A.24: Third test of tensile properties of 2CMC-0.5PEG-0.2PGV film.

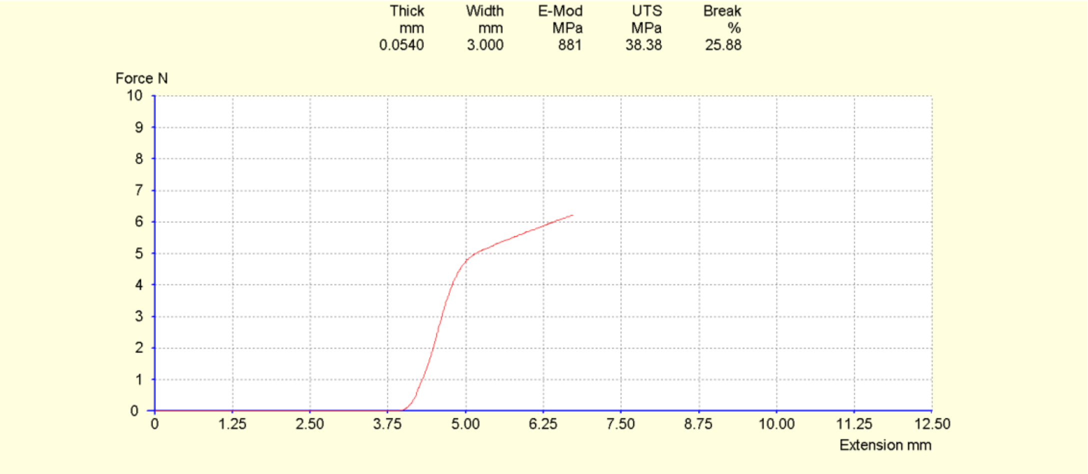


Figure A.25: First test of tensile properties of 2CMC-0.7PEG-0.2PGV film.

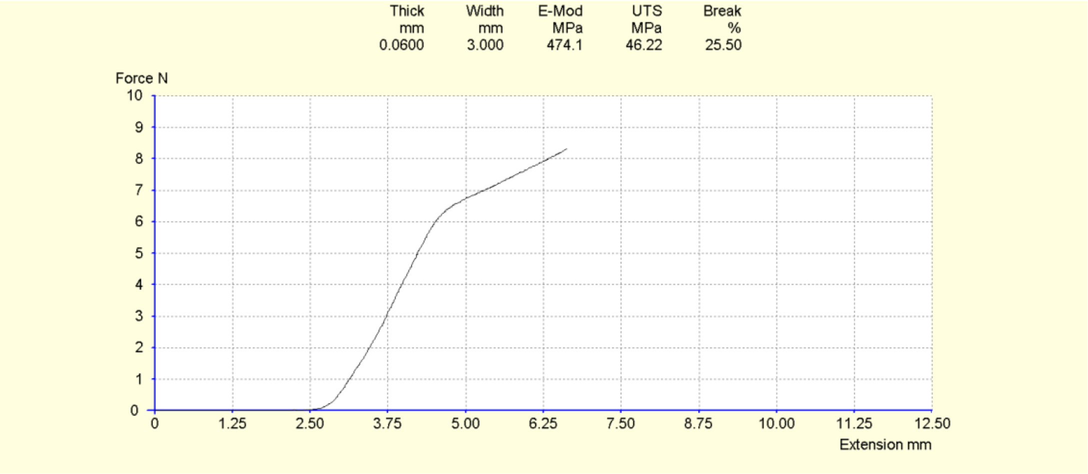


Figure A.26: Second test of tensile properties of 2CMC-0.7PEG-0.2PGV film.

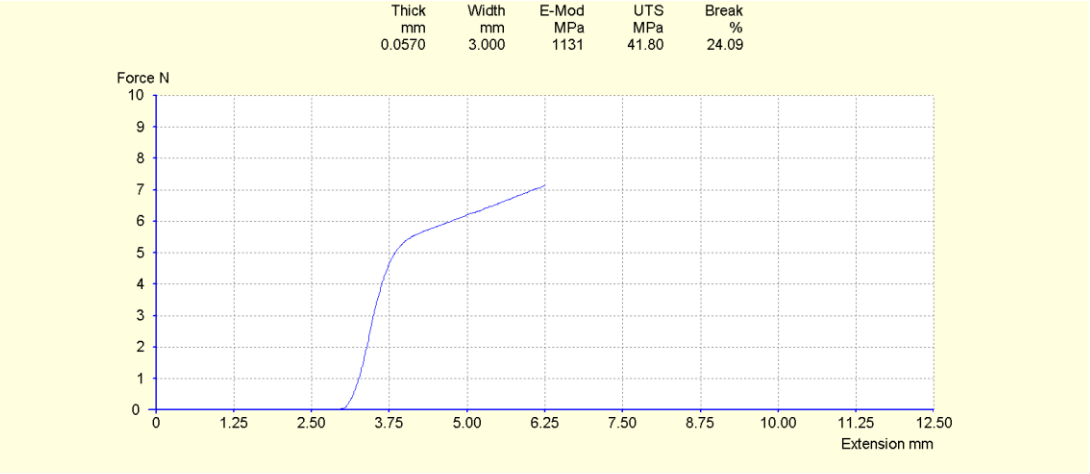


Figure A.27: Third test of tensile properties of 2CMC-0.7PEG-0.2PGV film.

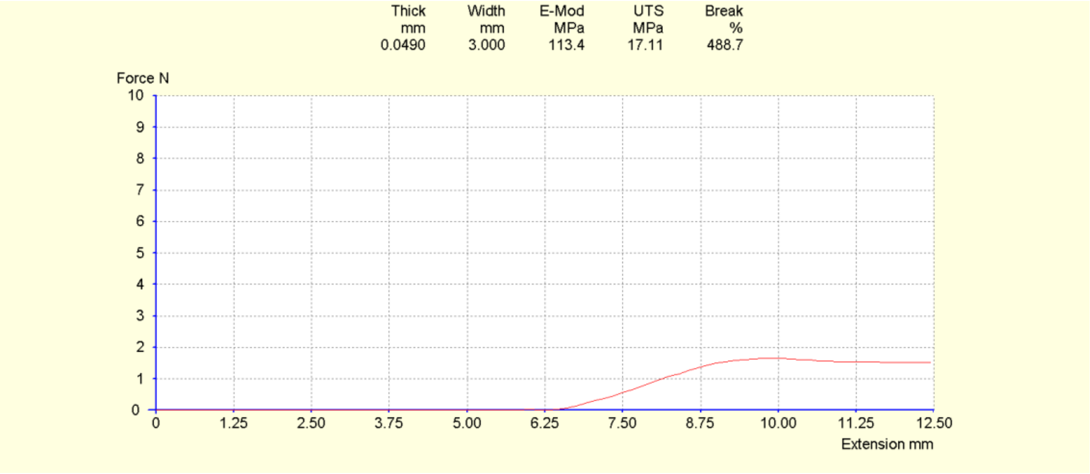


Figure A.28: First test of tensile properties of commercial PE film.

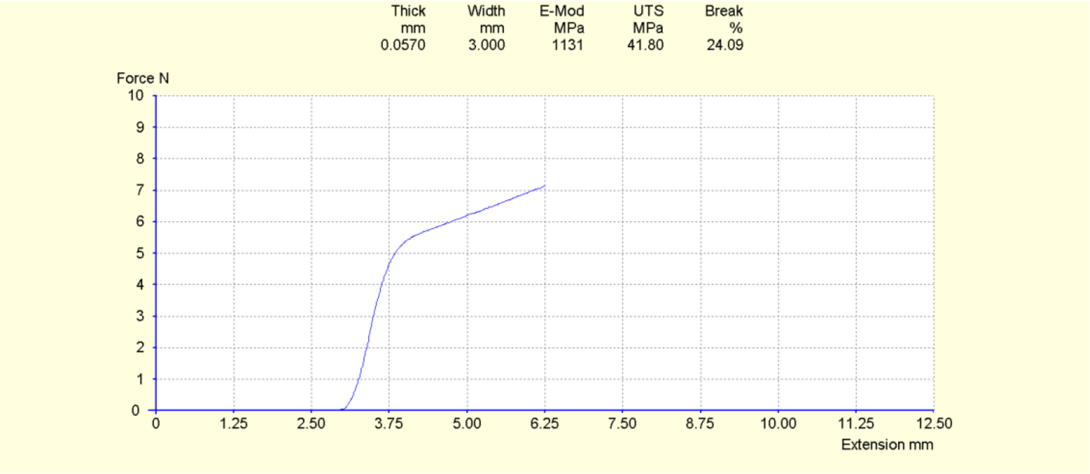


Figure A.29: Second test of tensile properties of commercial PE film.

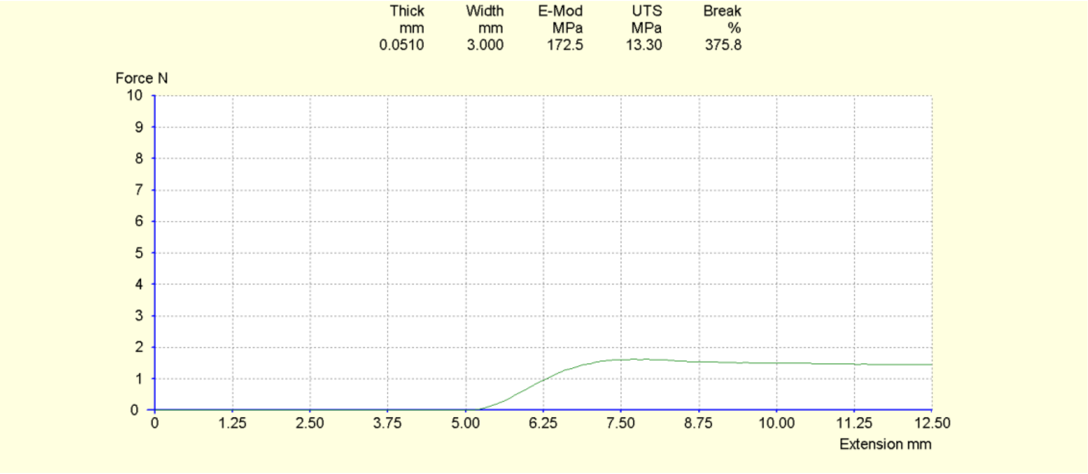


Figure A.30: Third test of tensile properties of commercial PE film.

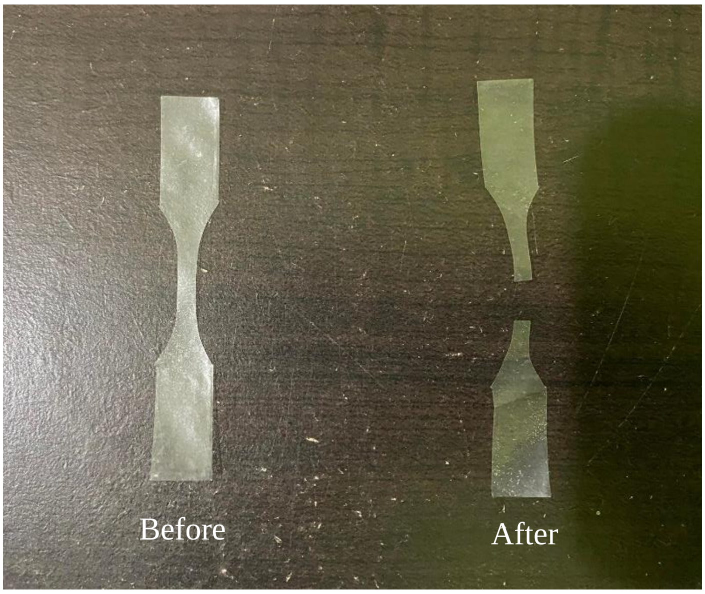


Figure A.31: Sample of the film before and after tensile test.

APPENDIX B: PHYSICAL APPEARANCE OF THE FILM

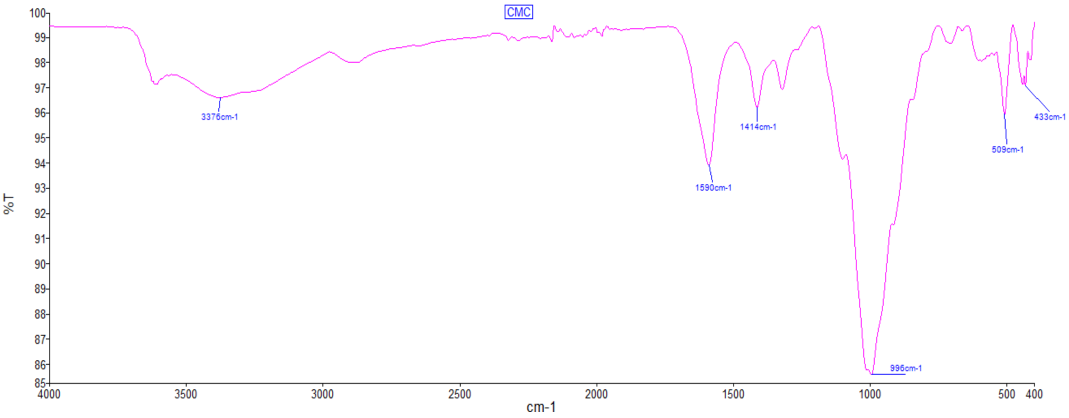


Figure B.1: ATR-FTIR analysis of CMC.

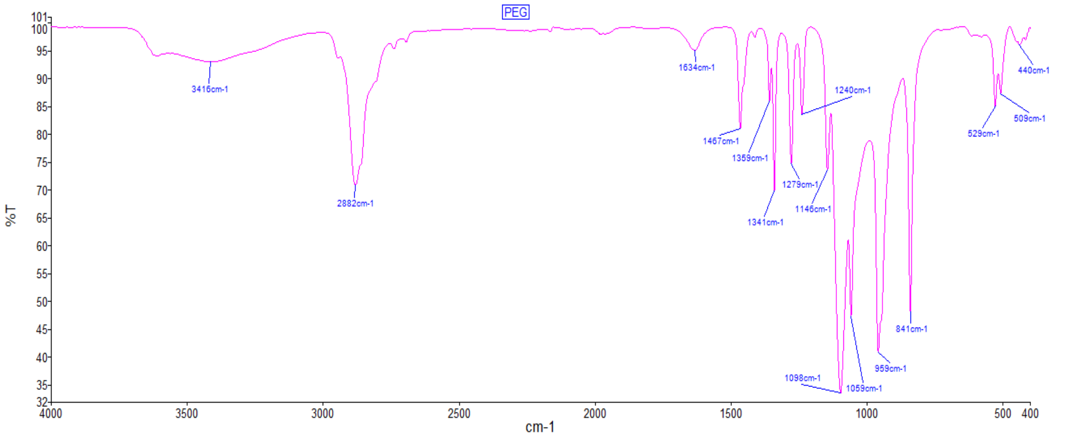


Figure B.2: ATR-FTIR analysis of PEG.

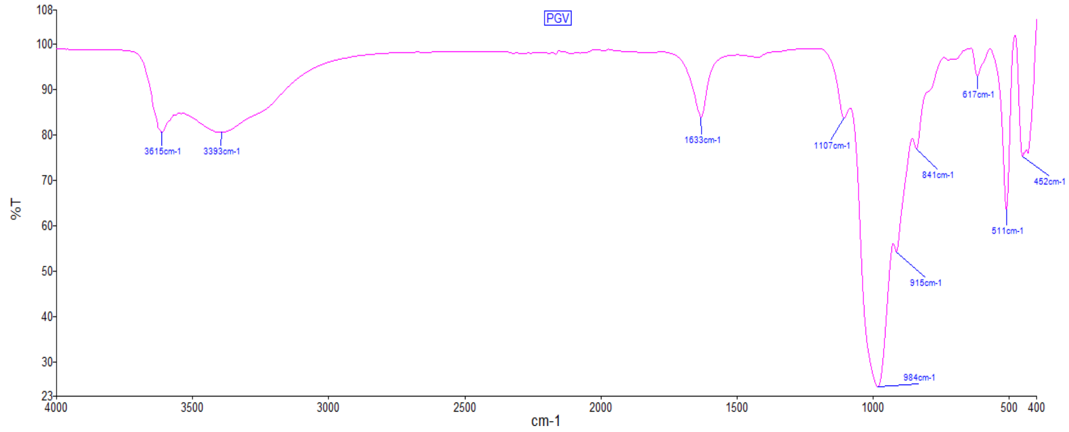


Figure B.3: ATR-FTIR analysis of PGV.

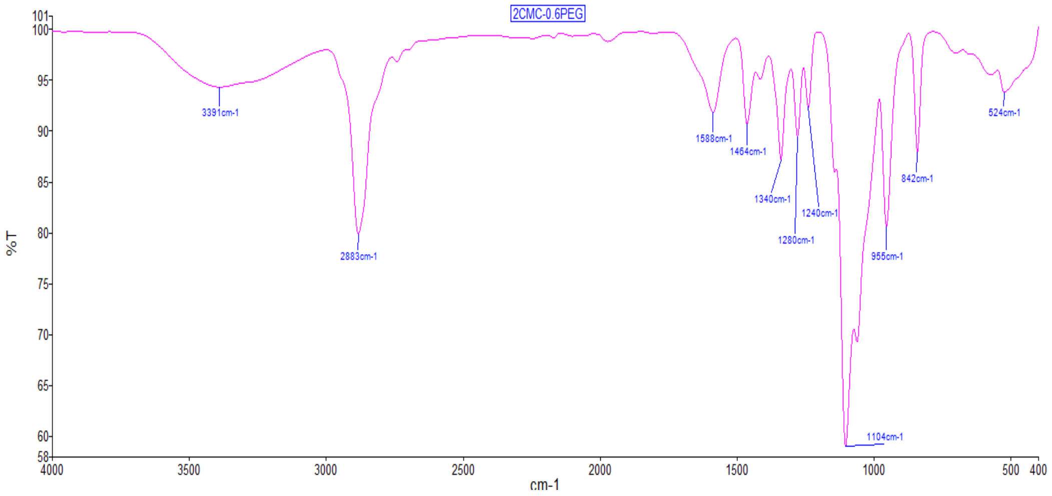


Figure B.4: ATR-FTIR analysis of 2CMC-0.6PEG film.

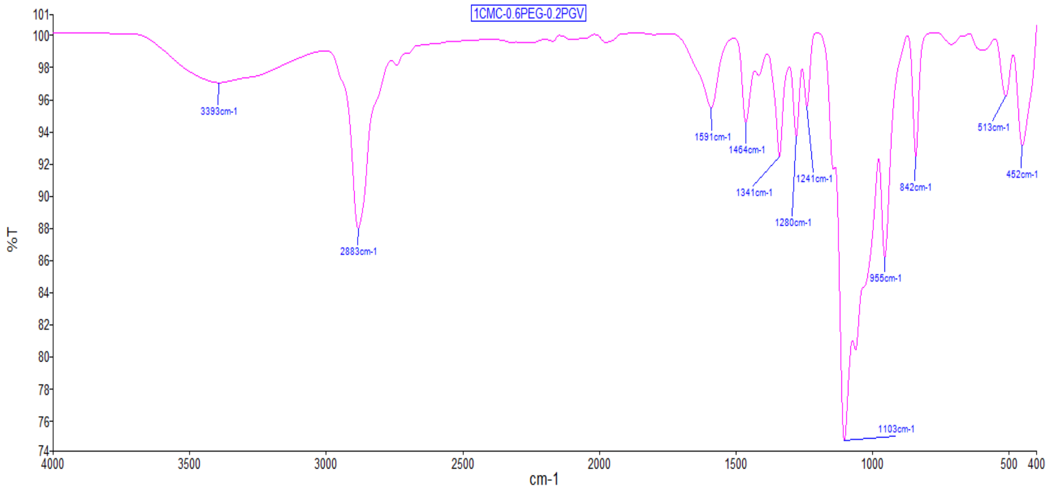


Figure B.5: ATR-FTIR analysis of 1CMC-0.6PEG-0.2PGV film.

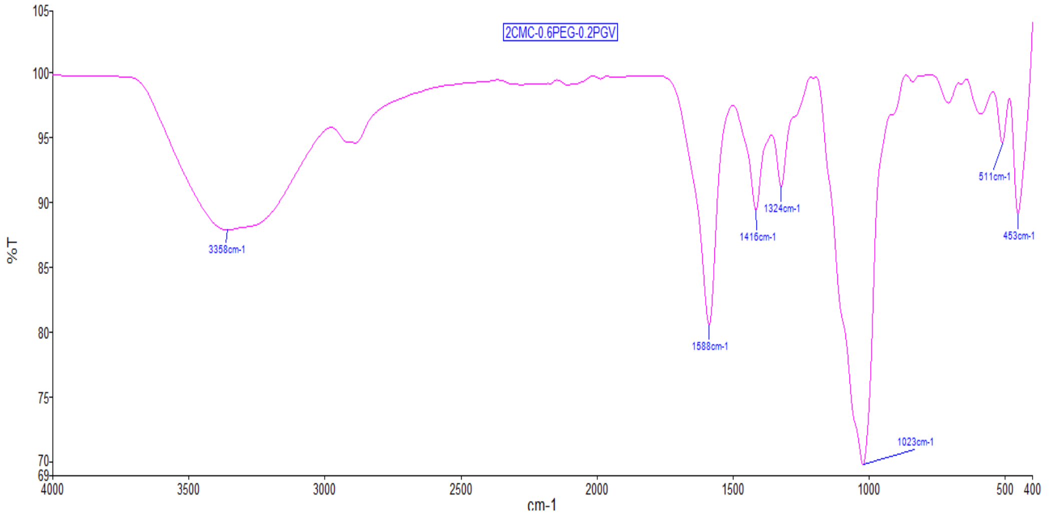


Figure B.6: ATR-FTIR analysis of 2CMC-0.6PEG-0.2PGV film.

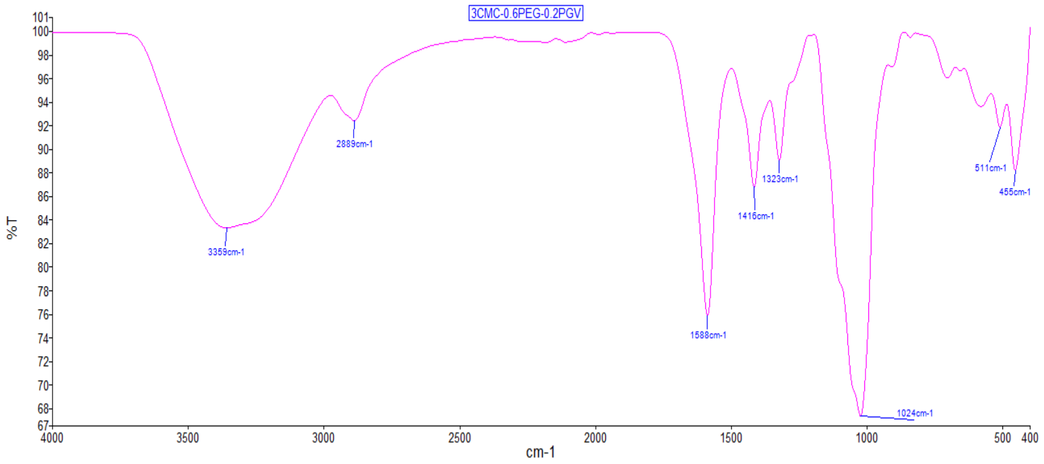


Figure B.7: ATR-FTIR analysis of 3CMC-0.6PEG-0.2PGV film.

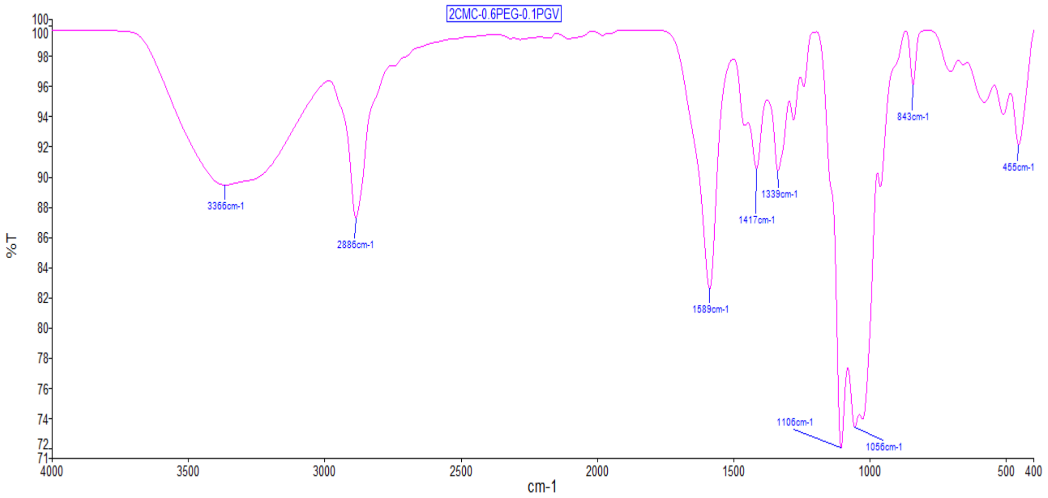


Figure B.8: ATR-FTIR analysis of 2CMC-0.6PEG-0.1PGV film.

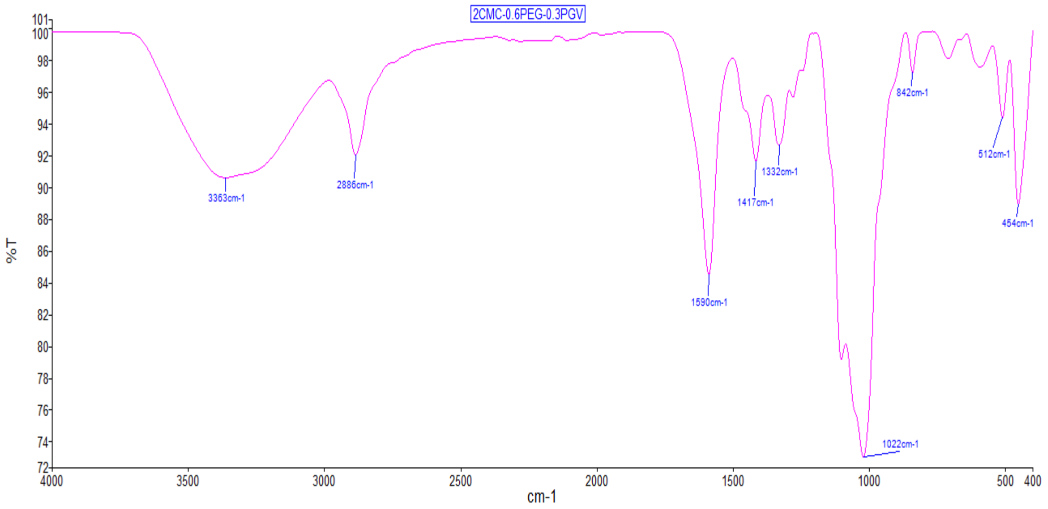


Figure B.9: ATR-FTIR analysis of 2CMC-0.6PEG-0.3PGV film.

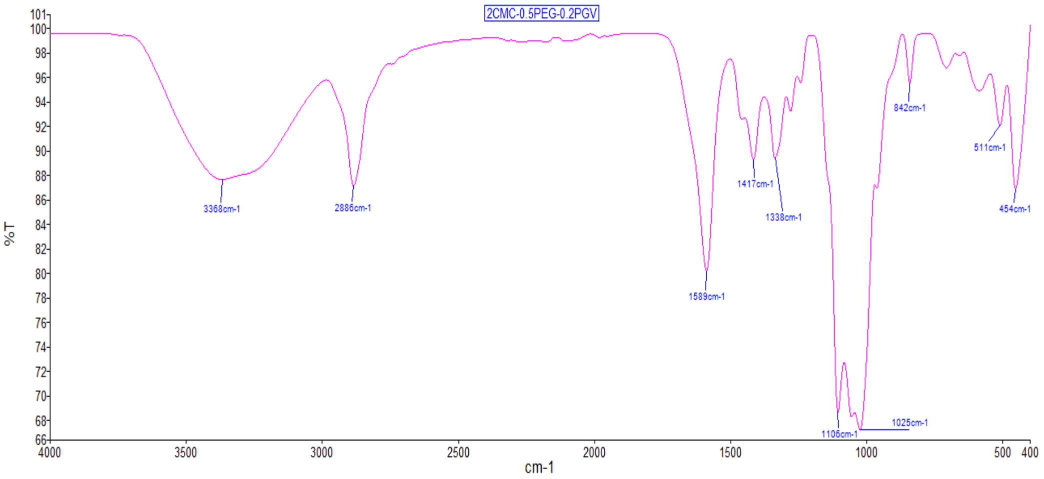


Figure B.10: ATR-FTIR analysis of 2CMC-0.5PEG-0.2PGV film.

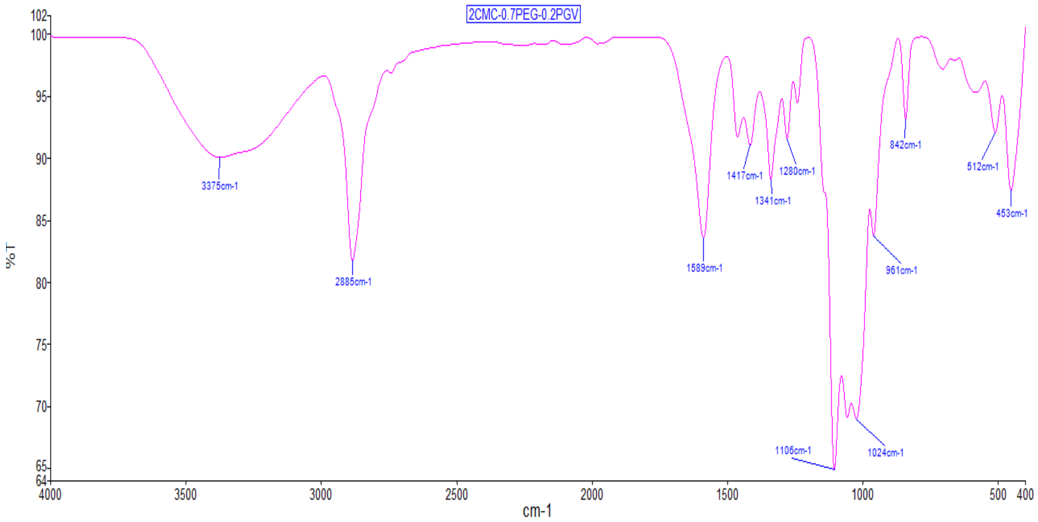


Figure B.11: ATR-FTIR analysis of 2CMC-0.7PEG-0.2PGV film.

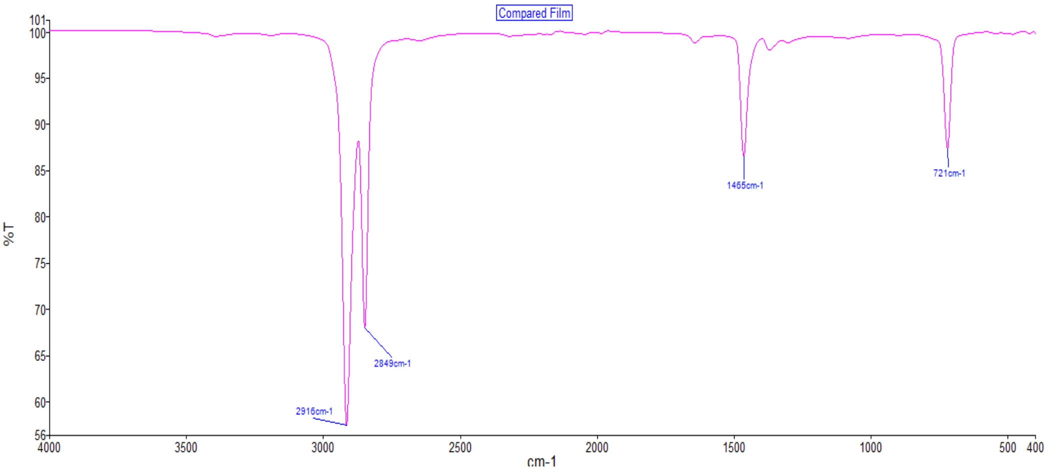


Figure B.12: ATR-FTIR analysis of commercial PE film.

APPENDIX C: PHYSICAL APPEARANCE OF THE FILM

Figure C.1: Sample of CMC film.

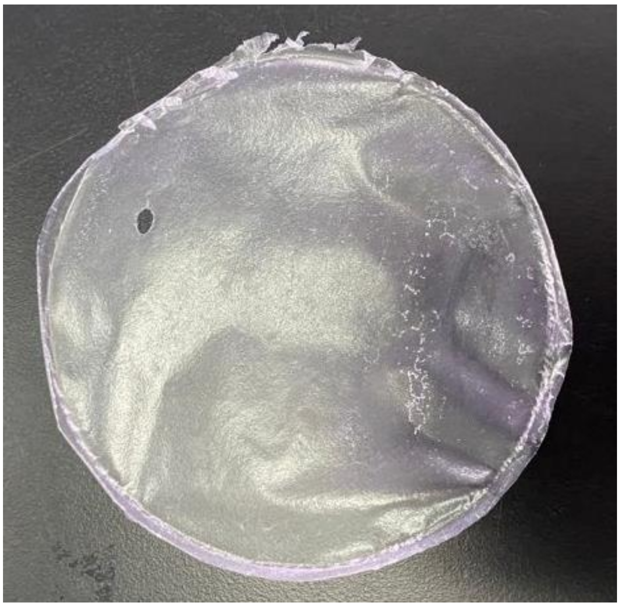


Figure C.2: Sample of 2CMC-0.6PEG film.

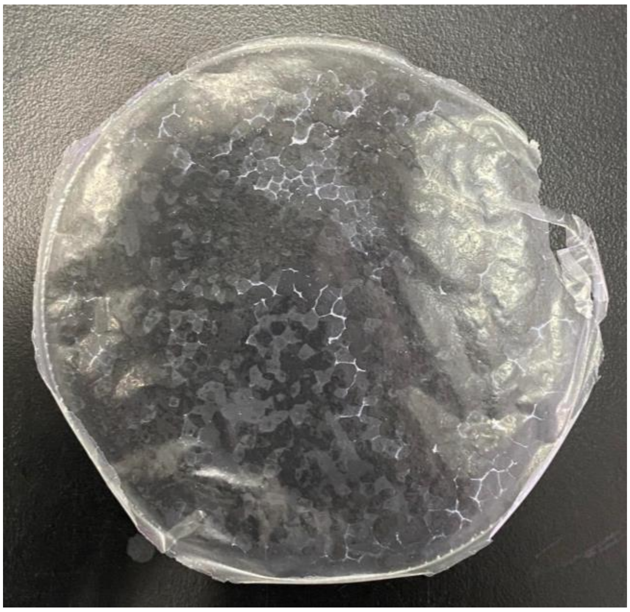


Figure C.3: Sample of 1CMC-0.6PEG-0.2PGV film.

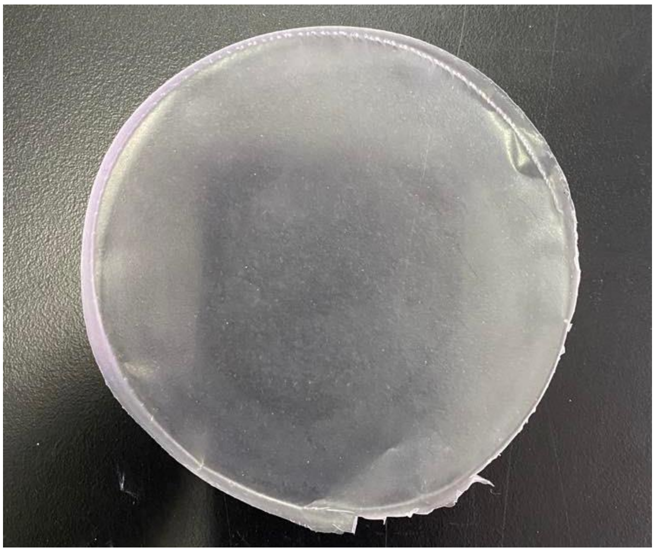


Figure C.4: Sample of 2CMC-0.6PEG-0.2PGV film.

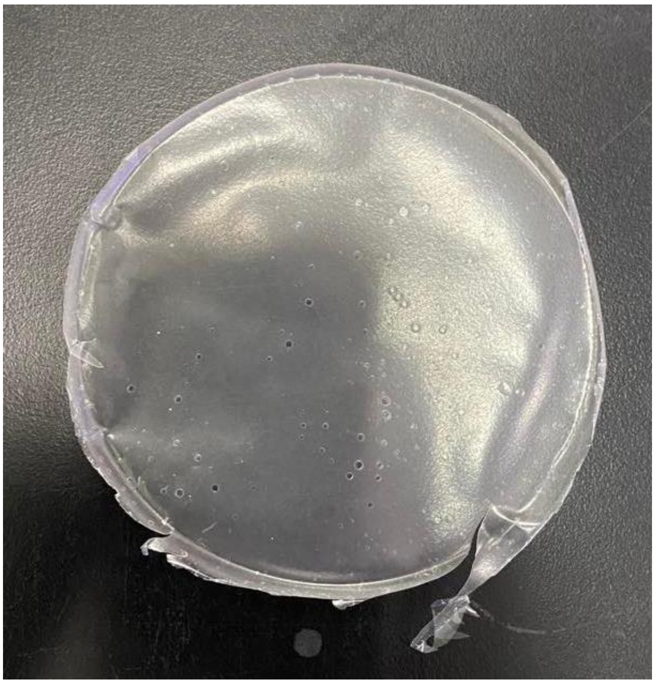


Figure C.5: Sample of 3CMC-0.6PEG-0.2PGV film.

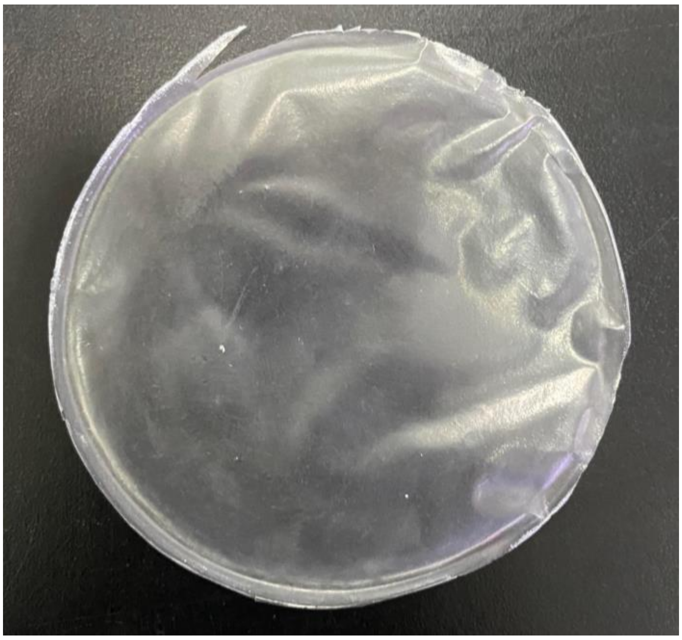


Figure C.6: Sample of 2CMC-0.6PEG-0.1PGV film.

Figure C.7: Sample of 2CMC-0.6PEG-0.3PGV film.

Figure C.8: Sample of 2CMC-0.5PEG-0.2PGV film.

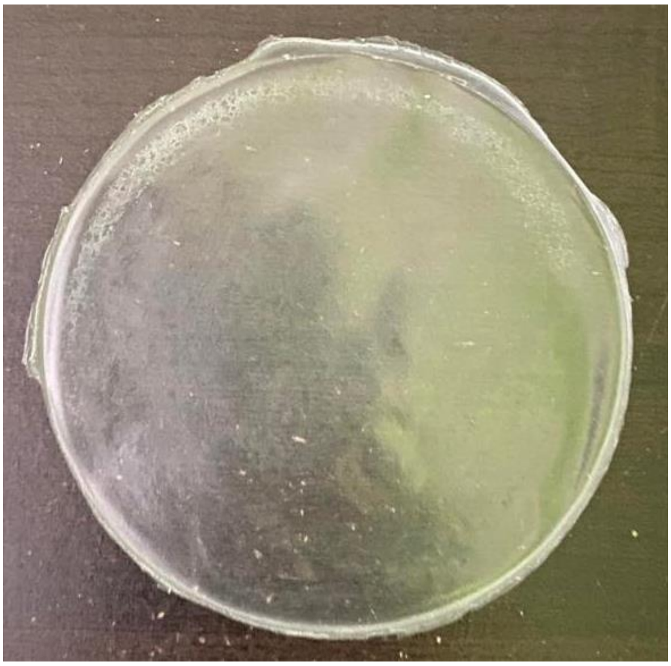


Figure C.9: Sample of 2CMC-0.7PEG-0.2PGV film.

Figure C.10: Sample of commercial PE film.

APPENDIX D: ANOVA Analysis

Table D.1: ANOVA analysis of the film tensile modulus at various CMC content.

Source of Variation	SS	df	MS	F	P-value	F crit
Between Groups	328016.7	2	164008.3	9.311307	0.014469	5.143253
Within Groups	105683.3	6	17613.89			
Total	433700	8				

Table D.2: ANOVA analysis of the film ultimate strength at various CMC content.

Source of Variation	SS	df	MS	F	P-value	F crit
Between Groups	1297.71	2	648.8548	38.54265	0.000377	5.143253
Within Groups	101.0083	6	16.83472			
Total	1398.718	8				

Table D.3: ANOVA analysis of the film elongation at break at various CMC content.

Source of Variation	SS	df	MS	F	P-value	F crit
Between Groups	20.77556	2	10.38778	0.303983	0.748601	5.143253
Within Groups	205.0333	6	34.17222			
Total	225.8089	8				

Table D.4: ANOVA analysis of the time taken for the film at various CMC content to dissolve in water.

Source of Variation	SS	df	MS	F	P-value	F crit
Between Groups	26.16667	2	13.08333	20.47826	0.002086	5.143253
Within Groups	3.833333	6	0.638889			
Total	30	8				

Table D.5: ANOVA analysis of the film tensile modulus at various PEG content.

Source of Variation	SS	df	MS	F	P-value	F crit
Between Groups	168302	2	84151	0.850044	0.473115	5.143253
Within Groups	593976	6	98996			
Total	762278	8				

Table D.6: ANOVA analysis of the film ultimate strength at various PEG content.

Source of Variation	SS	df	MS	F	P-value	F crit
Between Groups	168.1347	2	84.06734	4.114033	0.074992	5.143253
Within Groups	122.6057	6	20.43429			
Total	290.7404	8				

Table D.7: ANOVA analysis of the film elongation at break at various PEG content.

Source of Variation	SS	df	MS	F	P-value	F crit
Between Groups	26.60222	2	13.30111	0.944905	0.439799	5.143253
Within Groups	84.46	6	14.07667			
Total	111.0622	8				

Table D.8: ANOVA analysis of the time taken for the film at various PEG content to dissolve in water.

Source of Variation	SS	df	MS	F	P-value	F crit
Between Groups	5.166667	2	2.583333	18.6	0.002679	5.143253
Within Groups	0.833333	6	0.138889			
Total	6	8				

Table D.9: ANOVA analysis of the film tensile modulus at various PGV content.

Source of Variation	SS	df	MS	F	P-value	F crit
Between Groups	554953.6	2	277476.8	7.470679	0.02352	5.143253
Within Groups	222852.7	6	37142.11			
Total	777806.2	8				

Table D.10: ANOVA analysis of the film ultimate strength at various PGV content.

Source of Variation	SS	df	MS	F	P-value	F crit
Between Groups	130.767	2	65.38348	3.639263	0.092258	5.143253
Within Groups	107.7968	6	17.96613			
Total	238.5638	8				

Table D.11: ANOVA analysis of the film elongation at break at various PGV content.

Source of Variation	SS	df	MS	F	P-value	F crit
Between Groups	71.42889	2	35.71444	1.451807	0.306024	5.143253
Within Groups	147.6	6	24.6			
Total	219.0289	8				

Table D.12: ANOVA analysis of the time taken for the film at various PGV content to dissolve in water.

Source of Variation	SS	df	MS	F	P-value	F crit
Between Groups	50.16667	2	25.08333	64.5	0.0000878	5.143253
Within Groups	2.333333	6	0.388889			
Total	52.5	8				