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# The Effects of Caulerpa Racemosa Composition on the Properties of Corn Starch Biodegradable Film

**Wahida Abdul Rahman[1*], Nur Badiah Raja Roslan[1], Noor Aishatun Majid[1]**

*[1]Faculty of Applied Sciences, Universiti Teknologi MARA Cawangan Perlis, Kampus Arau, 02600 Arau, Perlis, Malaysia*

*[*]Corresponding author's e-mail: wahida811@uitm.edu.my*

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## ABSTRACT

*Biodegradable films derived from Caulerpa Racemosa (CR) seaweed and cornstarch were successfully developed via the solution casting method. The effect of CR composition on the properties of cornstarch biodegradable films has been successfully evaluated. FTIR analysis confirmed the existence of good compatibility between CR as a filler and starch as the matrix. The solubility and moisture uptake test revealed a slightly increasing trend when the composition of CR is more than 2.5 %. When the composition of CR increases, the hydrophilicity behaviour of the films increases due to a large number of hydroxyl groups (-OH) that improve its binding water capability. Mechanical test results revealed that increasing CR composition increases the flexibility and elasticity of the films produced. Films with 2.5 % CR exhibit the highest tensile strength and Young's modulus compared to other films. Apart from that, the soil burial test revealed that increasing CR compositions increases the weight loss of the film. Among the overall biodegradable blended films produced, a film with 2.5 % CR composition has been chosen as the best due to its good physical, mechanical, and biodegradability properties. Therefore, different compositions of seaweed and starch can be used to customize a film with the desired application.*

*Keywords: Caulerpa Racemosa; seaweed; hydrophilicity; biodegradable; solution casting*

## INTRODUCTION

The manufacturing and applications of synthetic films in packaging caused many severe environmental problems due to non-biodegradable properties and derived from non-renewable materials [1]. Therefore, the biodegradable films made from biopolymers play a significant role in reducing the impact of non-biodegradable plastic wastes. The biodegradable film is derived from renewable agricultural and biomass feedstock, and therefore it complies with the eco-efficient and sustainable materials. Starch was considered among the promising materials to produce biodegradable films because of its high abundance, low cost, and renewability [2]. However, starch-based polymers displayed poor water barrier and mechanical properties compared to non-natural polymers due to their high hydrophilicity and water sensitivity [3]. Therefore, the starch-based polymer can be strengthened by fillers, mixing with other biopolymer materials, or chemical modification to form an effective interlock with the hydroxyl group [4].

The emerging focus on renewable chemicals and polymers had led to a particular focus on marine biomass, which is seaweed [5]. Seaweeds are an example of biopolymer-based polysaccharides. Seaweeds are an excellent source of polysaccharides, with exceptionally high protein content, consisting of the essential amino acids, minerals, nutrients, and trace elements [6]. Seaweeds are divided into three groups which are red, brown, and green seaweed. Previous studies have reported that the development of biodegradable film from red seaweed, known as Kappaphycus alvarezii with calcium carbonate as the filler, has promoted considerable mechanical strength and other required properties for several industrial demands. Another species of green seaweed is *Caulerpa Racemosa* (CR), which is widely dispersed in tropical and semi-tropical areas such as the South China Sea. CR, also known as sea grape, belongs to *Chlorophyta, Ulvophyceae, Bryoposodales, Caulerpaceae* as marine green macroalgae [7]. In many Southeast Asian countries, including Indonesia, Malaysia, Philippines, Singapore, Vietnam, and Taiwan, seaweed of the genus *Caulerpa* is used as food [8].

Alginate, carrageenan, and agar are standard seaweed derivatives product that exhibits interesting film-forming properties. However, during the extraction of seaweed derivatives activities, the yield of seaweed derivatives extracted is too small, and the extraction method is not economical and not environmentally friendly because of the excessive chemical and energy consumption. Thus, raw seaweed is a good alternative for biodegradable film to overcome this problem. In addition, biopolymer or bioplastics made from agriculture or marine raw materials are still in the preliminary research stage. There are only a few reports about the development of biodegradable film by using natural green seaweed as a filler. Therefore, this study has attempted to develop biodegradable film from starch by using raw CR as the filler via the solution casting method. The effect of CR filler on the chemical, physical and mechanical properties of the biodegradable starch film have been evaluated in this paper.

## METHODOLOGY

### Preparation of raw material

The raw material used in this study was the green seaweed, *Caulerpa Racemosa* (CR). Green seaweed has been collected from the market at Simpang Empat, Perlis. CR was washed by using tap water and dried in the oven at 80 °C for 24 hours.

### Films preparation

The solution casting method has been used to prepare the film. About 10 g corn starch was dissolved in 200 mL of distilled water and heated for 15 minutes at 60 °C with continuous stirring. Raw CR with various compositions of 0 %, 2.5 %, 5 %, 10 % and 15 % were added into the solution. Next, 3 grams of glycerol were added, and the mixture was stirred at a constant temperature at 70 °C for two hours. The mixture was cast, cooled, and dried at room temperature on glass plates [2].

### Characterization

The films prepared were characterized using Attenuated Total Reflection Fourier Transform Infrared Spectroscopy (ATR – FTIR) (UiTM Perlis, model: Perkin Elmer – Spectrum One). The absorbance spectrum for each film was analyzed between 400 and 4000 $cm^{-1}$ [4]. The solubility test was conducted according to the method Moey [9]. The films were cut into 2 cm x 2 cm and were immersed for 30 minutes at 25 °C with constant agitation in 80 mL of deionized water. Then, the films were filtered using filter paper and dried at 60 °C in an oven to a stable weight. Film solubility (S %) has been calculated by using the formula below:

$$\textbf{Film Solubility, S}(\%) = \frac{\mathbf{W_i - W_o}}{\mathbf{W_i}} \times \mathbf{100\%}$$

Where; $W_i$ is the initial dry weight, and $W_o$ is the final dry weight.

The films' moisture uptake was determined using the method described by Tran et al. [10]. The films were cut into strips of 20 x 20 mm. Then, the samples were dried in the hot air oven at 105 °C for 2 hours. For each sample, two replicates were conducted. The moisture uptake was measured by using the formula below:

$$\textbf{Moisture Uptake, MU}(\%) = \frac{\mathbf{M_i - M_f}}{\mathbf{M_i}} \times \mathbf{100\%}$$

Where; $M_i$ is the initial weight of the film and $M_f$ is the final weight of the dried film.

For the soil burial test, the films were dried until constant mass at 60 °C. The films were cut to 2 cm x 3 cm. Then, the film was buried in the natural organic soil at a depth of 4 cm from the box's surface. The water was tossed into the soil to maintain a 40 % moisture value. After that, the film set was scooped out, then was rinsed with distilled water and dried on the filter paper at 60 ° C until a constant mass was obtained. The films were evaluated at specific times, which are 3, 6, 9, and 12 days before being buried and after. The biodegradability of the film was measured using the equation below as the weight loss (WL %):

$$\textbf{Weight Loss, WL}(\%) = \frac{\boldsymbol{m_o - m_t}}{\boldsymbol{m_o}} \times \mathbf{100\%}$$

Where $m_o$ is the first mass, and $m_t$ is the remaining dried mass at t.

Before tensile testing, the films were cut to 8 cm in length, 1.5 cm in width, and 0.20 ± 0.04 mm and were conditioned at 50 % RH. Tensile strength (TS), percentage of elongation at break, and Young's modulus were determined using a tensile tester operated according to the ASTM D882-02 standard. The films were replicated at least three for each sample to obtain the mean values [2].

## RESULTS & DISCUSSION

### FTIR Analysis

FTIR analysis was carried out to determine the chemical characteristics of the raw CR (Figure 1) and CR/corn starch biodegradable films (Figure 2). According to Figures 1 and 2, prominent peaks at 3268 $cm^{-1}$, 2900 $cm^{-1}$ and 1004 $cm^{-1}$ are attributed to the O-H stretching, -CH stretching, C-O and C-C stretching from cellulose and hemicellulose of CR seaweed. A sharp peak at 1620 $cm^{-1}$ (Figure 1) was attributed to the N-O asymmetric stretching indicative ester group in CR seaweed [11]. In Figure 2, the intensity of some peaks at 1600 $cm^{-1}$ and 1300 $cm^{-1}$ keeps reducing as the composition of the CR seaweed decreases. These peaks are attributed to the N-O asymmetric stretching and S=O stretching (sulfones) in green seaweed.

Moreover, medium to strong absorption peaks between 1200 and 970 $cm^{-1}$ have been observed for all spectra associated with the C-C and C-O pyranoid ring-stretching characteristic of green seaweed [12]. Previously, a peak at 1004 $cm^{-1}$ has been shifted to the right (999 $cm^{-1}$) in Figure 2 for all spectra. This phenomenon might be due to the glycosidic bond formation between CR seaweed filler and cornstarch matrix. This result was also similar with the findings by Hermawan et al.[13] that stated the existence of peak around 900 $cm^{-1}$ might be due to the glycosidic linkage (C-O) of 3,6-anhydro-D-galactose, C-O-S stretching in a (1-3)-D-galactose and C-O-C stretching in 3,6-anhydrogalactose which representing the seaweed. A peak attributed to free –OH stretching in Figure 1 was shifted from 3268 $cm^{-1}$ to 3285 $cm^{-1}$ in Figure 2 for all spectra. This phenomenon occurred due to the formation of new hydrogen bonding between CR seaweed and cornstarch matrix. This result is consistent with the findings by other researchers [1,14,15]. The formation of new hydrogen bonding improves the films' tensile strength, especially for a film with 2.5 % CR.

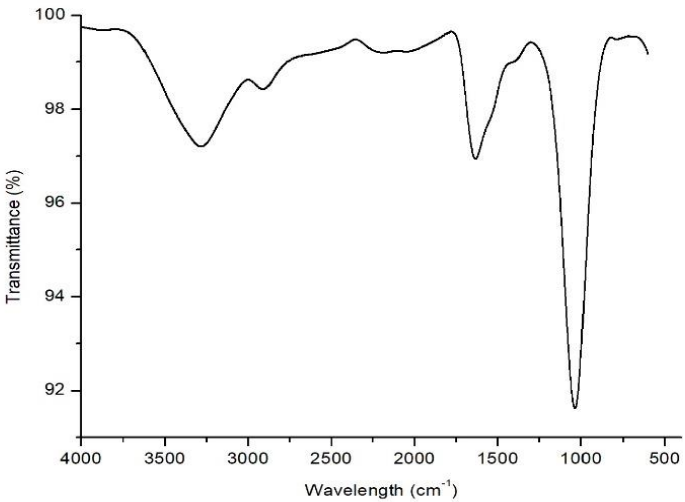


**Figure 1: FTIR Spectrum of Raw Caulerpa Racemosa (CR)**

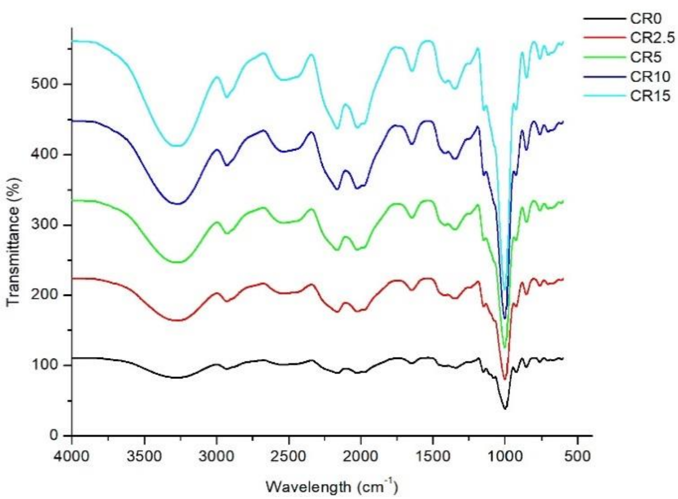


**Figure 2: FTIR Spectrum of Starch Film with different CR Compositions**

## Solubility and Moisture Uptake Test

Figure 3 shows the solubility of the starch–seaweed blend films which indicates the film's water resistance when immersed and continuously stirred in water. It has been found that film formulation of 2.5 % and 10 % results in lower solubility in water. Lower solubility might be due to the higher water-resistance of starch, which helps prevent water absorption that can contribute to substrate disintegration and dissolution [16]. Moisture uptake testing was used to determine the amount of moisture absorbed under the conditions specified. Figure 3 also shows the highest value for moisture uptake of the film with 15 % CR composition, which would be attributed to the hydrophilic characteristic of seaweed due to its large number of hydroxyl groups (OH) that improve its binding water capability. The binding site for the film with water also decreased as the composition of the seaweed decreased. As the CR seaweed loading decreased, the trend of the moisture uptake kept reducing, respectively. The declines trend might be due to the fewer sites of a hydroxyl group from seaweed that act as water-binding capability. Therefore, when the water-binding capability is reduced, the moisture uptake is also reduced. Similarly, this trend was reported by Hermawan et al. [13] and Huq et. [17]. They concluded that as the filler decreased, the sites for water-binding capability, which refers to the formation of new hydrogen bonding of the films with water, also reduced.

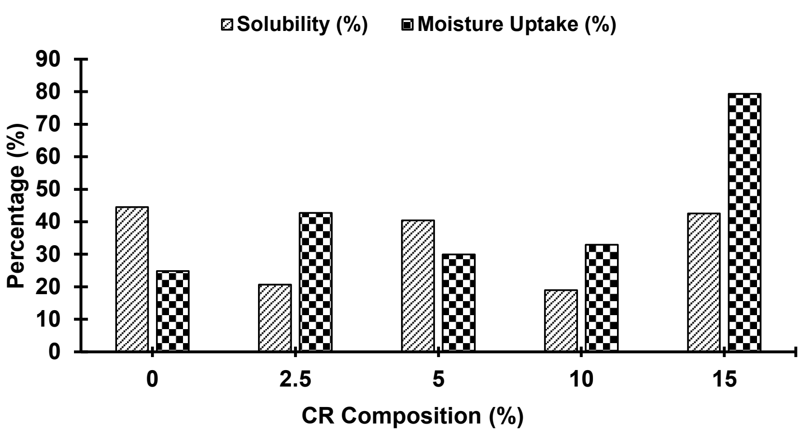


**Figure 3: Solubility & Moisture Uptake of CR/Cornstarch Films**

## Soil Burial Test

Figure 4 shows that the weight loss of all biodegradable films exhibits the increasing trend from Day 1 until Day 12. A longer burial time results in a higher weight loss for all the seaweed loadings, which indicates a higher number of microorganism activities in the films [15]. The introduction of seaweed into the cornstarch matrix has generally led to an aggressive biodegradation rate. Therefore, it has increased the weight loss of the films. As CR seaweed loading increased in the cornstarch matrix, the water-binding capability and hydrophilicity of the films increased. This result was supported by the moisture uptake test previously. This finding is similar to Jumaidin et al. [16] and Queiroz et al. [18], which concluded that the increasing site of water-binding capability would increase hygroscopic characteristics of the films and facilitate the growth of microorganisms during the degradation process. The film with 15 % CR exhibits the highest moisture uptake value, which promotes an appropriate environment for developing the microorganism in the soil. Therefore, this situation will increase the population of microorganisms in the soil and rapidly speed up the biodegradation rate of the films.

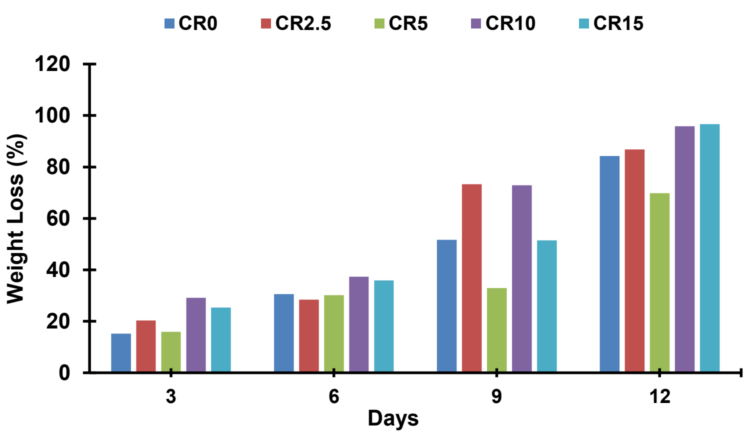


**Figure 4: Biodegradability of CR/Cornstarch Films**

## Tensile Test

The tensile test is essential for assessing the mechanical properties of biopolymer films, including tensile strength, elongation at break, and Young's modulus. The film with 2.5 % CR composition exhibits the highest tensile strength and Young's Modulus value (Figures 5 and 6). This phenomenon might be due to the new formation of the hydrogen bonding between the CR filler and cornstarch matrix. Therefore, this formation resulted in the compatibility between the CR filler and starch matrix that is confirmed by FTIR characterization in Figure 2 above. This finding is in agreement with Jumaidin et al. [16] that showed the same increasing trend of tensile strength and Young's modulus. However, the trend of tensile strength and Young's modulus showed a decreasing trend as seaweed content increased more than 2.5 %. When the composition of filler increases in the matrix, the filler will cause the restriction of stress transfer between the filler and the matrix itself. This phenomenon refers to the effect of matrix discontinuity due to the agglomeration of filler in the matrix that resulted in the decreasing value of tensile strength and Young's modulus. This finding also aligns withother researchers [13,17,19] that reported a decreasing trend of the tensile strength and Young's modulus in their composites as the filler content increased. Elongation at break measures the films flexibility before it breaks during the characterization process. The percentage elongation at break for 5 % CR exhibited the lowest value, which is 12.17 % (Figure 7). However, when the filler content increased more than 5 % in the matrix, the flexibility of the films improved based on the increasing value of elongation at break. This situation happened because the starch matrix provides ductility while seaweed exhibits delicate behaviour with subsequent loss of composite material toughness. This result agrees with Rahman et al. [20], which stated the same findings of the film characteristic when the filler content increased in the polymer matrix.

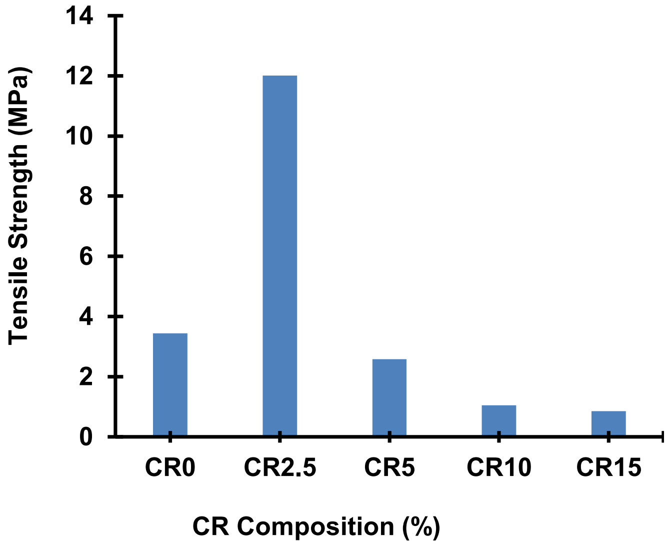


**Figure 5: Tensile strength result of films with various CR composition**

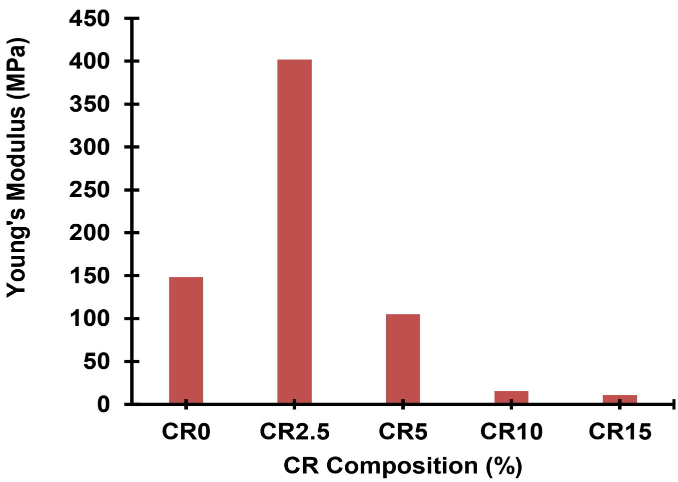


**Figure 6: Young's modulus result of films with various CR composition**

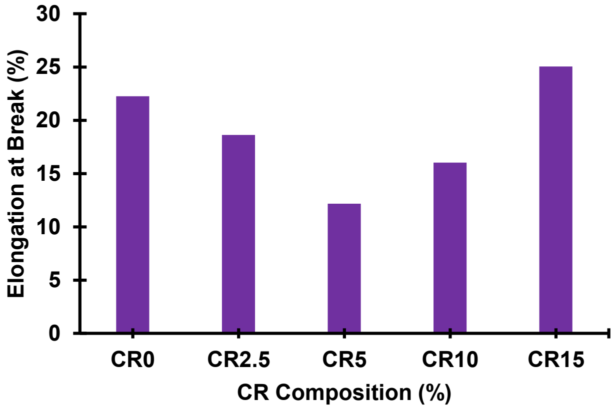


**Figure 7: Elongation at break result of films with various CR composition**

## CONCLUSION

This study investigated the effect of different loading of raw Caulerpa Racemosa seaweed on the functional group, physical and mechanical properties of the films. The FTIR analysis confirmed that the CR filler possessed good compatibility with the cornstarch matrix due to the formation of new hydrogen bonding between the filler and matrix. The increasing loading of seaweed filler in starch-based composites films significantly increased percentage of moisture uptake. Meanwhile, it has been found that film formulation of 2.5 % and 10 % results in lower solubility in water due to the higher water-resistance of starch, which helps prevent water absorption that can contribute to substrate disintegration and dissolution. In addition, the soil burial test indicated that the introduction of seaweed increases the material's weight loss, indicating faster biodegradation rate of composite films. However, the value of tensile strength and Young's Modulus exhibited decreasing trend as the CR content was more than 2.5 %. Elongation at break exhibited increasing trend as the CR content increased more than 5 %. This might be due to the increasing ductility behavior of the films produced. From this study, biodegradable films with 2.5 % raw *Caulerpa Racemosa* came across as being the best physical, mechanical, and good biodegradability among other films.

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