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Development of antioxidant film based on gelatin and carboxymethylcellulose incorporated with *Tecoma stans* (L.) Juss. ex Kunth Petals extract for biodegradable food packaging

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Abstract

The aim of this research was to develop a novel combination of antioxidant blended film for use as biodegradable packaging in the food industry. The antioxidant film was prepared based on gelatin (G) and carboxymethyl cellulose (CMC) incorporated with *Tecoma stans* (L.) Juss. ex Kunth petals extract (TKE) at various concentrations (0.5, 1.0, and 2.0 mg·L^{-1}) via solution casting method. The structural and surface morphology of G/CMC-TKE film were characterized using FTIR spectrometry technique and scanning electron microscopy (SEM), including determining antioxidant activity, water solubility, water vapor permeability, and biodegradability. According to FTIR analysis, the significant interaction between the gelatin and CMC chain is associated with hydrogen bonding. Adding TKE into the blended films significantly increased their roughness, thickness and antioxidant activity while decreasing their water solubility and water vapor transmission. Likewise, the biodegradability of the films containing antioxidants exhibited greater degradation values than the pure G/CMC film, and all of the biofilms was entirely degraded (>80%) in 14 days. The G/CMC-TKE 2.0 demonstrated the best antioxidant (74.47%), biodegradable activity (95.85% in 14 days), and the lowest water solubility (61.80%) and water vapor transmission rate (3.2483 g·m^{-2}·day^{-1}), which could be a feasible candidate for the food active packaging.

1. Introduction

Several million tons of petro-based synthetic plastic film are utilized in various applications, especially in the food packaging industry. Synthetic plastic packaging was exploited and accumulated in the ecosystem at an incredible rate of several tons annually. To overcome this problem, environmentally friendly materials have attracted much attention as an alternative to lowering the environmental impact of petroleum-based plastic food packaging [1]. Bio-based active packaging is a feasible candidate to replace synthetic plastic due to its versatility, non-toxic, low cost, and eco-friendly. Numerous natural polymers (protein, polysaccharide and lipid) have the potential to be utilized as raw material for bioactive packaging film preparation [2].

Gelatin is one of the biomaterials derived from proteins commonly used in film preparation as food packaging materials due to outstanding characteristics, including low cost, great film-forming ability and adaptability. Additionally, gelatin is a renewable material that is biodegradable, edible, and can be combined with bioactive compounds to create a functional packaging film [3]. Generally, gelatin film exhibited high water absorption due to the presence of several hydrophilic groups, which may weaken the mechanical properties and water vapor transmission of the film. For this reason, blending gelatin with other biopolymers, such as chitosan, starch and gum. is a viable way to eliminate the shortcomings of gelatin-based films [4-7]. Carboxymethyl cellulose (CMC), an anionic derivative of cellulose, is the most widely utilized in the food packaging industry for improving the properties of the composite film due to its excellent film formability, high viscosity, biocompatibility, good gas barrier properties, and stable internal network structure [8,9].

Recently, active packaging production has attracted significant attention because active packaging is regarded as one of the most effective methods for extending the shelf life and ensuring the safety of packaged foods. Antioxidants, antimicrobial agents, or other active chemicals could be added to food packaging to preserve food quality and extend shelf life by preventing microbial growth and avoiding unwanted biological or chemical changes [10]. Instead of synthetic additives, plant-based materials with both antioxidant and antibacterial could be applied to the film-forming solution to improve the qualities of composite films, particularly antioxidant and antibacterial properties.

Tecoma stans (L.) Juss. ex Kunth (Bignoniaceae), sometimes referred to as yellow-elder, yellow trumpet bush, trumpet-flower, yellow-bells, trumpet bush, ginger-thomas, esperanza, and timboco, has established itself naturally in tropical and subtropical regions of Africa, Asia, and Oceania. It is cultivated as an ornamental plant with

eye-catching clusters of cup-shaped, bright yellow flowers with a light perfume, evergreen leaves, and abundant fruits and seeds. Phytochemical compounds present in plant flowers have been reported such as phenolic compounds, carotenoids, saponins, flavonoids, and alkaloids, which exhibit antioxidant and antibacterial activity [11,12]. The plant parts have been used in several therapies, including treating diabetes and digestive problems, kidney problems, jaundice, skin infections, toothaches, headaches, joint pains, sore eyes, and heart pain. From previous research, extracts from *Tecoma stans* have been discovered to inhibit the development of yeast infection and exhibit antioxidant constituents, anticancer activity, and antibacterial activity in human pathogenic bacteria [13,14]. The plant benefits perfumery as a flavoring, cosmetic, and lubricating ingredient. In addition, plant leaves, and flowers have recently been applied for silver nanoparticle synthesis, which appears to be a more simple, affordable, and environmentally friendly alternative to the conventional process [15-17].

The novelty of this research is the first production of antioxidant film composed of carboxymethylcellulose and gelatin incorporated with *Tecoma stans* (L.) Juss. ex Kunth Petals extract. There is no prior research utilizing *Tecoma stans* extract as an ingredient for active food packaging production. Therefore, this research aimed to study the preparation of gelatin and carboxymethyl cellulose blended film, which contains *Tecoma stans* extract cultivated in Thailand. An initial investigation was conducted into the physical appearance and morphology, structure, antioxidant activity, mechanical capabilities, water solubility, and water vapor transmission rate. Additionally, the biodegradability of these films was examined for their potential use as biodegradable food packaging applications.

2. Experimental

2.1 Preparation of *Tecoma stans* (L.) Juss. ex Kunth (TKE)

The *Tecoma stans* (L.) Juss. ex Kunth petals were collected in July-September from the area of Valaya Alongkorn Rajabhat University, Pathum Thani province, located in the central region of Thailand. The petals were dried and ground into powder using a blender. The petal powder was then weighed for 10 g and extracted twice with 200 mL of ethanol at concentrations of 75% v/v. The mixture was centrifuged for 30 min at room temperature and then heated at 60°C for 30 min before extracting using an ultrasonic bath for 20 min. The supernatant was then filtered through filter paper No 1. The filtrates were evaporated at 40°C using a rotary evaporator. The crude extract was then kept in the dark at 4°C. The crude extract was kept in the dark at 4°C before use.

2.2 Development of antioxidant G/CMC biofilms

The gelatin (G) was purchased from QReC, New Zealand. Carboxy-methylcellulose (CMC) was obtained from Krungthep Chemie, Thailand. The G/CMC films were prepared using the solution casting method The 1% w/v of gelatin (15 mL) was mixed with 1% w/v CMC (35 mL) using a magnetic stirrer at 60°C for 30 min. After stirring, a clear solution was cast on Teflon-coated plates and allowed to dry at 40°C to 50°C in a hot air oven for 6 h to 8 h. Before properties analysis, the G/CMC dried films were peeled off the plate and stored in a desiccator. For the antioxidant films, the films were prepared following the same procedure with the addition of 5 mL of *Tecoma stans* (L.) *Kunth* petals extract at various concentrations (0.5, 1.0, and 2.0 mg·L^{-1}). The prepared antioxidant films were designated G/CMC-TKE 0.5, G/CMC-TKE1.0, and G/CMC-TKE0 2.0, respectively. The thickness of the film sample was determined using a vernier caliper. The thickness was measured at five random spots of each film, and their average value was utilized.

2.3 Antioxidant properties

The antioxidant capacity of the G/CMC and G/CMC-TKE blend films at various antioxidant content was assessed using the DPPH radical scavenging ability method The DPPH test examines the bleaching of DPPH from a purple to a clear solution for assessing its electron-donating capabilities. The antioxidant test of the G/CMC blend films was carried out according to Adilah *et al.* (2018) [18] with slight modification. Approximately 0.10 g of each film sample was soaked and continuously shaken in 10 mL of pure ethanol for 10 min to extract the antioxidant from the blend films. A 3 mL of film extract was mixed with 1 mL of 0.1 mM ethanolic DPPH solution, followed by incubation at room temperature in a dark area for 30 h. The absorbance of the mixture was investigated at 517 nm against ethanol as a blank solution, and the DPPH radical scavenging activity of the blend film was measured in three repetitions and determined using the following Equation (1).

$$DPPH\ radical\ scavenging\ activity\ (\%) = \frac{(A_{DPPH} - A_{sample})}{A_{DPPH}} \times 100 \quad (1)$$

Where, A_{DPPH} is the absorbance of the DPPH solution, and A_{sample} is the absorbance of the film sample extract

2.4 Properties characterization of blend films

2.4.1 Color of biofilms

The color of biofilm samples was investigated using a chroma meter (Konica Minolta, CR-400, Tokyo, Japan) at 5 points/3 times per sample (n =3). The color parameter was reported as lightness (L), redness (a), and yellowness (b). The total color difference (ΔE) was calculated using Equation (2):

$$\Delta E = \sqrt{\Delta L^2 + \Delta a^2 + \Delta b^2} \quad (2)$$

2.4.2 Structural characterization (FTIR)

The functional group interaction and structure of biofilm were investigated using an Attenuated Total Reflectance-FTIR spectro-photometer at a wavenumber of 4000 cm^{-1} to 400 cm^{-1}. Before the measurement, the biofilm samples were cut and placed directly on the beam exposure stage

2.4.3 Mechanical properties

The mechanical properties of biofilms were investigated using a Universal Testing Machine (Lloyd LR 50k, Lloyd Instruments, UK) with a crosshead speed of 50 mm·min^{-1} and a gauge length of 50 mm. Mechanical properties such as tensile strength (TS), elongation at break (EB), and elastic modulus (EM) of each film was measured according to the standard method of ASTM D638.

2.4.4 Water solubility

The gravimetric method was used to determine the water solubility of biofilms with various antioxidant content. The film samples were cut into 4 cm^2 × 4 cm^2 squares and drying the film in a hot air oven at 100 ± 5°C for 2 h before accurately weighing (W_1). Each film sample was immersed in 10 mL of distilled water at room temperature for 15 min. Then, place the swollen films on the already weighed filter paper and re-dried at 100 ± 5°C for 2 h to determine the final weight of the dry film sample (W_2). The water solubility of the film was measured in three triplicates and calculated using the following Equation (3).

$$\% \, Water \, solubility = \frac{W_1 - W_2}{W_1} \quad (3)$$

Where; W_1 is the initial weight of dry film (g), W_2 is the final weight of dry film after soaking (g)

2.4.5 Water vapor transmission rate (WVTR)

The water vapor permeability of biofilms was investigated gravimetrically in triplicate according to the standard method ASTM E96-01 of the American Society for Testing and Material [19]. The films were cut into a circular shape and used to close on the top of the permeation cells (2.8 cm external diameter × 1.5 cm depth) containing 1.00 g of anhydrous sodium chloride as a desiccant. The permeation cells were sealed with paraffin, weighed to determine the initial weight, and placed in a desiccator containing silica gel. The weight of the cells was monitored every 24 h interval to 120 h (5 day) in three replicates [20]. The water vapor transmission rate (WVTR) of the blend films was calculated by using Equation (4).

$$WVTR = \frac{\Delta G}{tA} \quad (4)$$

Where; ΔG is the difference in weight of the permeation cell at an indicated period of time (g), t is time (day), A is the exposure area/surface area of the top of the permeation cell (m^2)

2.4.6 Surface morphology

The surface morphology of the antioxidant biofilms was investigated using Field Emission Scanning Electron Microscope (FESEM, JEOL/JSM-7610F, Tokyo, Japan) at the voltage of 1 kV. The films were cut into 1 cm × 1 cm and fixed on stubs then coated with gold using sputter-coater before surface analyzing.

2.5 Biodegradability test

The biodegradation in soil of antioxidant biofilm at various conditions was performed as described by Ai. *et al*. with a slight modification [21]. The film samples were cut into 4 cm × 4 cm and allowed to dry at 60°C in a hot air oven for 30 min. The dry film samples were kept in a net bag, and then accurately recorded the total weight of film and net bag (A_0). The sample films were buried in a nursery bag containing 500 g of soil at a depth of 10 cm. The nursery bags were placed in an outdoor environment and taken at regular times (1, 3, 5, 7, and 14 day). The contaminated soils were carefully eliminated from the film sample before drying at 60°C in a hot air oven for 2 h, and the dry weight of each sample was measured (A_1). The weight loss of each sample was calculated from Equation (5), indicating the degradation of the sample films.

$$Biodegradability = \frac{A_0 - A_1}{A_0} \times 100 \quad (5)$$

3. Results and discussion

3.1 Physical appearance and morphology of G/CMC antioxidant films

The antioxidant biofilms (G/CMC-TKE) were successfully prepared by the solution casting method. The physical appearance of the prepared G/CMC films with various content of *Tecoma stans* (L.) Juss. ex Kunth petals extract was shown in Figure 1.

From Figure 1, the prepared G, CMC and G/CMC membranes were clear and colorless while the color of transparent G/CMC-TKE membranes varied to pale yellow depending on the TKE content. The color parameter index of the G, CMC, the blend G/CMC film, and G/CMC-TKE at various concentrations were illustrated in Table 1. The color results of biofilms were represented in terms of L (lightness-darkness), a (redness-greenness), b (yellowness-blueness), and the total color difference (ΔE) using the blend G/CMC without TKE as a reference film. The results indicated that the pure gelatin film displayed the maximum lightness value (82.11) when compared to the pure CMC

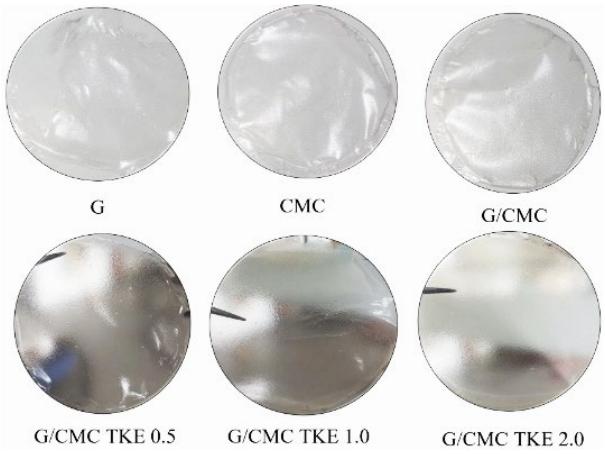

Figure 1. The physical appearance of G, CMC film and G/CMC incorporated with TKE at various concentrations.

(80.21) and the blended G/CMC (75.14) which showed a remarkable drop in film lightness. The incorporation of TKE into the G/CMC biofilm resulted in a marginal reduction of film lightness but increased the a and b values of G/CMC films, which was due to the intrinsic greenish-yellow color of the TKE, resulting in biofilm with a darker, greener, and yellower appearance. In addition, the color difference (ΔE) showed greater values in the G/CMC films incorporated with the TKE at various concentrations because of the colorant natural compounds found within the extract. The thickness of G, CMC, G/CMC and G/CMC-TKE films with various compositions were displayed in Table 2. The addition of TKE resulted in a slight increase in the thickness of the G/CMC (0.123 ± 0.002). This was mostly owing to the increase in solution casting composition.

The surface morphology of the films with various compositions was characterized using SEM, as exhibited in Figure 2. The surface SEM images of pure G, CMC, and G/CMC blend films were smooth and homogeneous following the cross-section image of biofilms which depicted a homogeneous phase and showed small pallets in G/CMC film. In contrast, the surface of the G/CMC blend films incorporated with TKE was rougher and covered with numerous dimples. Furthermore, the greater TKE in these blended films, the more noticeable microspheres were on the surface of the SEM image. Cross-sectional SEM images revealed that pure G and CMC had a homogeneous phase, with a few insoluble pallets visible in the G/CMC blend film. After adding TKE, the films that contained a greater quantity of TKE exhibited heterogeneous sections on the surface of these films. It can be indicated that the TKE affected the morphology of the G/CMC films. This is due to the phenolic compounds and bioactive elements in TKE possessing hydrophobic characteristics (hydrocarbon chain). The hydrophobic sections which have a propensity to reject water in the biofilm, could be responsible for the aggregation of microdroplets leading to a rougher film surface [22].

3.2 Antioxidant activity

The TKE was acquired from the solvent extraction method with an extraction yield of 18.86 ± 1.22 % per dry weight of petals. The antioxidant activity of TKE and biofilms was assessed using the DPPH radical scavenging activity techniques, which are displayed in Figure 3. The DPPH radical scavenging activity of G, CMC and G/CMC film were 1.55% and 2.60% and 2.11%, respectively, indicating quite low antioxidant activity. The antioxidant activity of TKE before being incorporated into the G/CMC blend film was 76.24%. The results in Figure 3 demonstrated that the G/CMC incorporated with the TKE films significantly affected their antioxidant activity. The DPPH scavenging activity of the films was correlated to TKE content. The G/CMC TKE 2.0 film displayed the highest antioxidant activity (74.47%). This indicated that the increase in the antioxidant activity of the G/CMC film with TKE was dependent on the concentration of TKE. From the antioxidant activity result, it can be implied that the antioxidant activity of the blend films was mainly due to an antioxidant compound (carotenoid, phenolic and flavonoids) from *Tecoma stans* (L.) Juss. ex Kunth petals extract. The utilization of the antioxidant compounds from *Tecoma stans* (L.) Juss. ex Kunth petal extract has not been reported in previous studies for antioxidant film production. The biofilms incorporated TKE with high antioxidant activity can be applied as active food packaging to protect oxidation-sensitive foods for sustaining the quality and increasing the shelf life of food. The biofilms incorporated with antioxidant compounds can improve the quality and shelf life of oxidation-sensitive foods. This is achieved through the neutralization of free radicals and reactive oxygen species, which are known to contribute the food oxidation. By reducing the oxidation processes that result in spoilage, these compounds effectively extend the shelf life of the food product. [16,23].

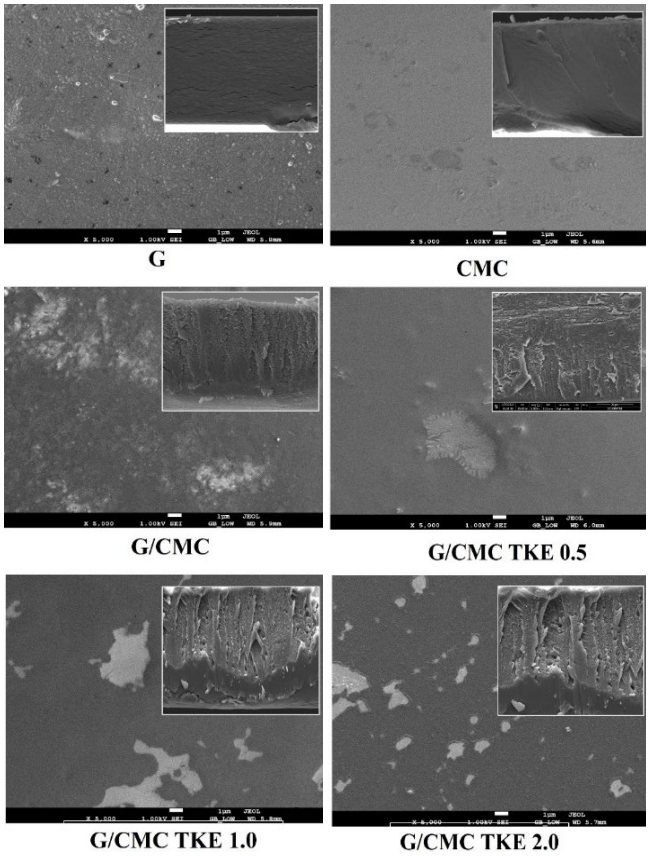

Figure 2. The SEM images of biofilm morphology: G, CMC, G/CMC, and G/CMC-TKE at various concentrations. Insets are cross-section images of biofilms.

Table 1 Surface color parameter of biofilms.

Films	L	a	b	ΔE
G	82.11 ± 0.12	0.27 ± 0.03	1.69 ± 0.05	7.95 ± 0.02
CMC	80.21 ± 0.11	–3.28 ± 0.05	5.07 ± 0.14	5.15 ± 0.03
G/CMC	75.14 ± 0.06	–3.17 ± 0.06	4.19 ± 0.04	Ref
G/CMC TKE 0.5	73.59 ± 0.03	–5.20 ± 0.02	6.39 ± 0.05	3.37 ± 0.06
G/CMC TKE 1.0	72.79 ± 0.02	–6.28 ± 0.06	9.67 ± 0.04	6.72 ± 0.02
G/CMC TKE 2.0	71.15 ± 0.05	–7.21 ± 0.02	15.17 ± 0.03	12.36 ± 0.02

Table 2. Thickness, water solubility and water vapor transmission rate of the G, CMC, G/CMC, and those incorporated with TKE.

Films	Thickness (mm)	Water solubility (%)	WVTR ($g \cdot m^{-2} \cdot day^{-1}$)
G	0.128 ± 0.003	81.90 ± 1.81	4.5476 ± 0.06
CMC	0.112 ± 0.002	62.25 ± 1.86	9.9072 ± 0.02
G/CMC	0.123 ± 0.002	69.07 ± 1.09	5.6845 ± 0.10
G/CMC TKE 0.5	0.126 ± 0.002	66.73 ± 1.35	3.7355 ± 0.01
G/CMC TKE 1.0	0.127 ± 0.002	65.48 ± 1.02	3.5731 ± 0.02
G/CMC TKE 2.0	0.127 ± 0.002	61.80 ± 1.18	3.2483 ± 0.02

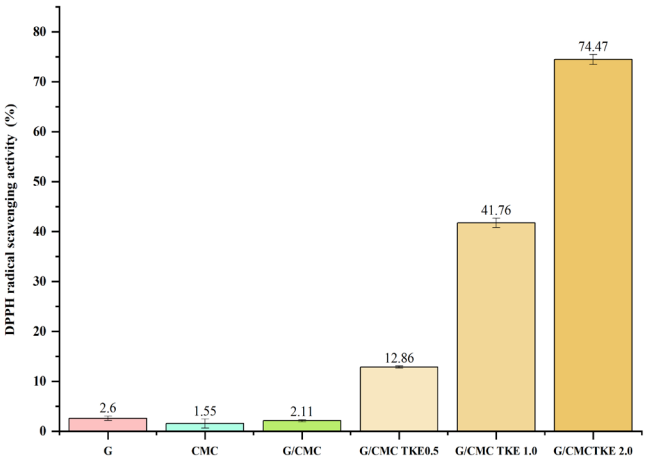

Figure 3. DPPH radical scavenging activity of biofilms; G, CMC, and G/CMC films incorporated with 0.5 $mg \cdot L^{-1}$ to 2.0% $mg \cdot L^{-1}$ of *Tecoma stans* (L.) *Kunth* petals extract.

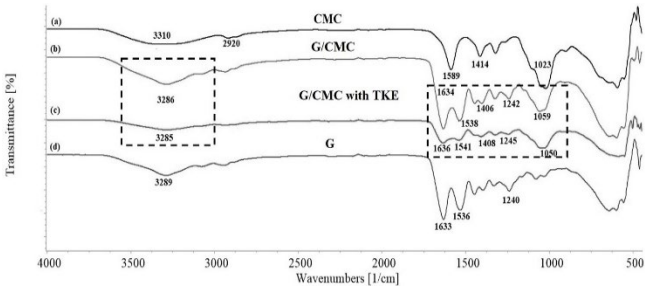

Figure 4. FTIR spectra of blend films: G, CMC, G/CMC, and G/CMC with TKE.

3.3 Structural characterization

The structural conformation of the blend films was investigated by FT-IR spectroscopy technique which can typically provide information regarding the interactions between the polymer chains. The structural changes in the polymer chain can be detected by a change in the absorption band of the functional group in their FTIR spectrum. The FTIR spectra of G, CMC, G/CMC, and G/CMC-TKE 2.0 films was displayed in Figure 4.

From the spectrum of CMC film (Figure 3(a)), the absorption peaks appeared at 3310 cm^{-1} and 2920 cm^{-1}, corresponding to the O-H stretching of the hydroxyl group and C-H stretching vibration of alkane groups in the CMC molecule. The adsorption bands at 1589, 1414, and 1023 cm^{-1} clearly showed evidence of the asymmetric, symmetric C=O stretching vibration of the carboxyl group and C-O stretching of ether bond, respectively [24]. The characteristic FTIR spectra of protein usually exhibited the absorption band of Amide I: C=O stretching (1700 cm^{-1} to 1600 cm^{-1}), Amide II: N-H bending (1600 cm^{-1} to 1500 cm^{-1}), and Amide III: C-N stretching (1300 cm^{-1} to 1200 cm^{-1}). For the gelatin film (Figure 3(d)), the adsorption band at 3289 cm^{-1} was correlated to the N-H and O-H stretching of the amino acid group in the peptide chain of gelatin. The Amide I, II, and III adsorptions of gelatin film were presented at 1633, 1536, and 1240 cm^{-1}, respectively [25]. For the blend film, the remarkable change in the spectrum of the G/CMC (Figure 3(b)) was observed by the appearance of the bands at 1634 cm^{-1} and 1059 cm^{-1}, which were the characteristic band of Amide I in gelatin and asymmetric C=O stretching vibration in CMC. This indicates that gelatin and CMC were strongly associated with the blend films. All variations in the peak of the G/CMC blend film suggest some identified interactions between the distinctive groups of gelatin and CMC, which most indicates strong molecular compatibility between gelatin and CMC. The spectra of G/CMC film with and without the extract (Figure 3(b-c)) showed a strong absorption band at 1059 cm^{-1} and 1050 cm^{-1} illustrating the presence of the C-O group. The locations of peaks in the spectra of the G/CMC film with extract (Figure 3(c)) were similar to the control (Figure 3(b)), due to the relatively low quantity of extract in the systems. According to the peak at 3285 cm^{-1} of the G/CMC film with extract became flattened and slightly shifted to a lower wavenumber as the increasing of extract, which was caused by the OH group in the extract, might be interacted to G/CMC film by hydrogen bonding [26,27].

3.4 Mechanical properties

The mechanical properties of the sample films were examined using the Universal Testing Machine; the results are depicted in Figure 5. As indicated by the data in Figure 5, the pure G film demonstrated superior mechanical performance (higher TS and EM) in comparison to the CMC film, which corresponds to the previous research [28]. The plausible explanation for this phenomenon is the continuous fibrillar three-dimensional arrangement of proteins within gelatin molecules [29]. In turn, the G/CMC blend film demonstrated lower TS and EM values and exhibited greater elongation in comparison to the pure gelatin film. Following the film's physical appearance, the gelatin film was rather brittle, whereas the G/CMC film displayed enhanced flexibility and durability upon incorporation of CMC. In the case of G/CMC with TKE films, marginally higher TS and EM values were observed at higher TKE concentrations due to the high polyphenolic content of TKE, which facilitates hydrogen bonding between polymer chains [30,31]. Furthermore, an increase in TKE concentration resulted in a little increase in % E for the G/CMC TKE films; hence, it is possible to assume that the addition of TKE improved the elasticity of the biofilms.

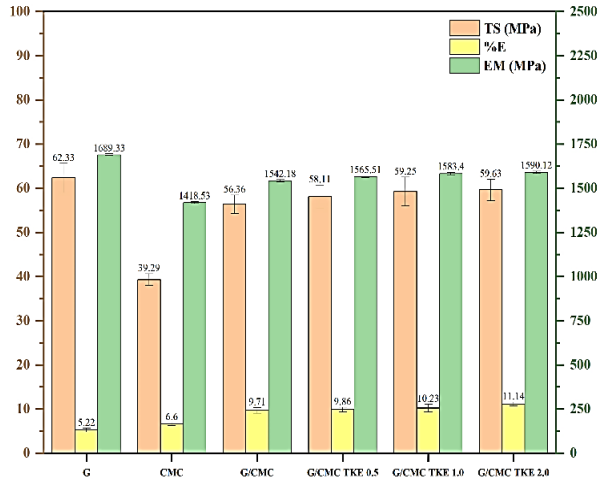

Figure 5. Mechanical properties of biofilm; G, CMC, and G/CMC with and without TKE.

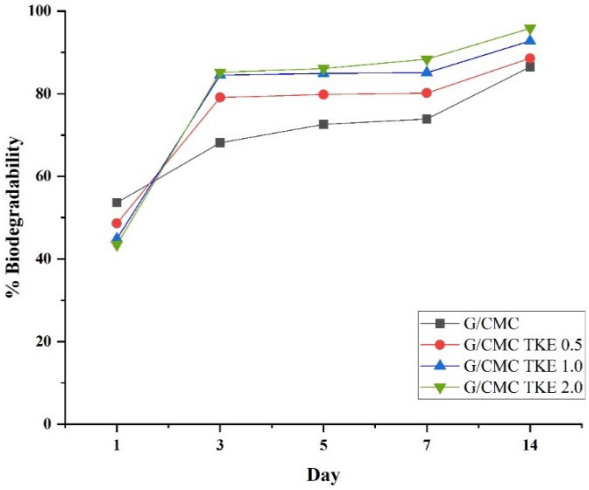

Figure 6. Biodegradability in the soil of G/CMC and G/CMC incorporated with the TKE in the period of 14 days.

3.5 Water solubility

The water solubility of the control film (G and CMC), the blend G/CMC film, and G/CMC-TKE at different concentrations were displayed in Table 2. The water solubility of the films is correlated with the OH groups present in the polymeric matrix, which can establish hydrogen bonds between the components. The highest water solubility was illustrated in the gelatin (G) control film (81.90%) due to the hydrophilic nature of gelatin and the abundance of hydroxyl groups (OH) in gelatin molecules. The blend of G with CMC exhibited a decrease in water solubility (69.07%) which could occur due to intermolecular hydrogen interactions between the hydrophilic groups present in the CMC and gelatin [22]. For the G/CMC-TKE films, adding TKE at various concentrations reduced the water solubility of blend films (approximately 3% to 10%). The G/CMC TKE2.0 displayed the lowest water solubility (61.80%). The reduction in water solubility is according to the interaction of hydrogen bonds between OH groups in the G/CMC chain and the phenolic hydroxyl group of the polyphenols in TKE. Thus, the hydrophilic groups in the polymer chain and the polyphenols in the extract formed hydrogen bonds that decreased the capacity of the OH groups to interact with water molecules, lowering the water solubility of blend films. Generally, films with low water solubility are recommended for food packaging applications [32]. This is because films with low water solubility prolong the shelf life of packaged foods. Through the reduction of water and moisture infiltration, these films effectively uphold the product's freshness, impede oxidative reactions, and obstruct microbial growth.

3.6 Water vapor transmission rate (WVTR)

The water vapor transmission rate (WVTR) is the rate of water vapor permeating through the film. The WVTR can be computed using the slope of a graph illustrating the change in weight of permeation cells versus time, whereby the slope is then divided by the exposure area of the film (Equation (4)). The WVTR values of the G/CMC-based biofilms enriched with various concentrations of TKE were tabulated in Table 2. A comparison of the WVTR of biofilms showed that the pure gelatin film (G) exhibited lower WVTR (4.5476 $g \cdot m^{-2} \cdot day^{-1}$) than CMC (9.9072 $g \cdot m^{-2} \cdot day^{-1}$) and G/CMC film (5.6845 $g \cdot m^{-2} \cdot day^{-1}$). This is because gelatin contains several polar groups (OH) that can interact with water molecules and fix them to the active polar sites; this result is consistent with G film's high water solubility value. It is possible that gelatin-containing biofilm has the capacity to trap passing water vapor, hence preventing vapor permeability [28]. The WVTR of the G/CMC films containing the extract (TKE) was lower than the G/CMC film. The G/CMC film with the highest concentration of TKE (G/CMC TKE 2.0) displayed the lowest of WVTR value (3.2483 $g \cdot m^{-2} \cdot day^{-1}$). The addition of natural extracts into the blend of G/CMC reduces the availability of the OH groups of gelatin-carboxymethyl cellulose matrix due to cross-linking with the bioactive molecules. Therefore, the reduction in the water vapor transmission rate might be related to the interaction between the extract (TKE) and the biopolymer matrix, which hindered the formation of intermolecular interactions between the biopolymer matrix and the water molecules. The film with a low water vapor transmission rate is preferred for food packaging because it prevents moisture emigration for the purpose of decreasing spoilage [22,33].

3.7 Biodegradability

The decomposition of antioxidant biofilms after 14 days in the soil is depicted in Figure 6. G/CMC demonstrated the maximum biodegradability after one day (53.64 %), followed by G/CMC TKE0.5. (48.61%). However, after three days, the film containing antioxidants exhibited greater degradation values than the pure G/CMC film. Generally, antioxidants are often added to food packaging materials to prolong their shelf life by preventing oxidation. In some cases, the presence of antioxidants may slow down the initial biodegradation process. The protective effect of antioxidants could temporarily hinder microbial activity responsible for the decomposition of materials [34]. In addition, all of the biofilms were nearly entirely degraded (>80%) in 14 days, with the G/CMC TKE2.0 film exhibiting the greatest biodegradability (95.85%) and G/CMC displaying the lowest degradation (86.50%). The results indicated that the biodegradability of the film is affected by the presence of antioxidants. According to the SEM study, the film with a high concentration of TKE was a rougher surface, which could enhance the degradability of biofilms. The rough

surface of biofilms could be due to inhomogeneous TKE compound distribution. Phenolic substances are the most antioxidant in TKE. These compounds also possess potential antibacterial characteristics, which could impact the microbial communities in the soil that are accountable for biodegradation. Furthermore, the biodegradation of film was also dependent on the amount and type of microorganisms in the soil environment [35].

4. Conclusions

The gelatin/carboxymethyl cellulose-based blend films were successfully incorporated with different concentrations of *Tecoma stans* (L.) Kunth petal extract as a source of antioxidant components to develop a novel active functional food packaging. The blend films were prepared via the simple method (solvent casting). Enriching TKE into the blended films significantly enhanced their thickness and antioxidant activity while decreasing their water solubility and water vapor transmission. The SEM and FT-IR results obviously indicated the good compatibility and strong interaction of the prepared blend film. Furthermore, *Tecoma stans* (L.) Kunth petal extract could be suggested as a novel green antioxidant additive in functional food packaging applications to ensure food safety and extend shelf life. Additionally, the antioxidant-blend films demonstrated excellent biodegradability in soil, producing non-toxic and environmentally friendly packaging. Overall, the prepared biofilms provided a variety of benefits as inexpensive active food packaging materials.

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