Food Science and Technology 12(2): 115-127, 2024 http://www.hrpub.org
DOI: 10.13189/fst.2024.120201

Preservation of Tomatoes *(Lycopersicum esculentum)* with Composite Biofilms Based on Starch and Microcrystalline Cellulose (MCC)

Tran Y Doan Trang[1,*], Do Thi Hanh[2], Ha Thi Dzung[2], Ta Thi Huong[1], Pham Huong Quynh[1], Ha Thi Nha Phuong[3], Nguyen Thi Thu Hien[1], Hoang Thanh Duc[1]

[1]HaUI Institute of Technology, Hanoi University of Industry, Vietnam
[2]Faculty of Chemical Technology, Hanoi University of Industry, Vietnam
[3]Faculty of Chemical and Environmental Technology, Viet Tri University of Technology, Vietnam

Received September 4, 2023; Revised March 25, 2024; Accepted April 22, 2024

Cite This Paper in the Following Citation Styles

(a): *[1] Tran Y Doan Trang, Do Thi Hanh, Ha Thi Dzung, Ta Thi Huong, Pham Huong Quynh, Ha Thi Nha Phuong, Nguyen Thi Thu Hien, Hoang Thanh Duc , "Preservation of Tomatoes (Lycopersicum esculentum) with Composite Biofilms Based on Starch and Microcrystalline Cellulose (MCC)," Food Science and Technology, Vol. 12, No. 2, pp. 115 - 127, 2024. DOI: 10.13189/fst.2024.120201.*

(b): *Tran Y Doan Trang, Do Thi Hanh, Ha Thi Dzung, Ta Thi Huong, Pham Huong Quynh, Ha Thi Nha Phuong, Nguyen Thi Thu Hien, Hoang Thanh Duc (2024). Preservation of Tomatoes (Lycopersicum esculentum) with Composite Biofilms Based on Starch and Microcrystalline Cellulose (MCC). Food Science and Technology, 12(2), 115 - 127. DOI: 10.13189/fst.2024.120201.*

Abstract This research collected starch films with varying Microcrystalline Cellulose (MCC) contents, ranging from 0% to 20% by weight. Subsequently, and these starch-MCC composite films were assessed for their film-forming ability, solution properties, water absorption capacity, solubility, biodegradability, and surface morphology. The film with the best characteristics was selected for further testing in preserving ripe tomatoes. The results showed that adding 4% MCC to the film reduced water absorption and enhanced biodegradability and solubility to an optimal level while improving adhesion during production compared to the initial film. Electron microscopy observations revealed no significant impact on the surface morphology of the composite film when 4% MCC was added. Applying this starch film in preserving ripe tomatoes demonstrated its ability to maintain the freshness, shine, and color of the fruit for up to 12 days. The research reveals that using the St-4% MCC composite film effectively maintains the freshness and quality of ripe tomatoes. Compared to untreated fruits, this coating notably slows down decay, retains color, and mitigates firmness loss. Moreover, the film reduces weight loss and preserves total soluble solids, total acidity, and ascorbic acid content, indicating its potential as a promising preservation technique for maintaining fruit quality and nutritional value during prolonged storage.

Keywords Microcrystalline Cellulose (MCC), Starch, Composite Film, Preservation, Ripe Tomato

1. Introduction

Tomatoes (*Lycopersicum esculentum*) are a crucial vegetable in the human diet, and they are cultivated extensively worldwide [1-6]. In 2021, tomato production in the US reached 95.90 tons per hectare, Spain reached 84.73 tons/ha, and China reached 59.21 tons/ha [7]. However, the distribution of tomatoes varies across regions, necessitating transportation and storage. Tomatoes have relatively thin skins and high-water content, posing challenges for transportation and preservation, particularly when dealing with ripe tomatoes. This results in significant yearly post-harvest tomato loss, causing economic losses [2]. Therefore, it is essential to investigate methods for preserving ripe tomatoes after harvest to ensure its freshness and nutritional value are retained.

Packaging is crucial to maintaining the quality of fruits and facilitating their transportation and preservation [2,8]. Plastic packaging is a common choice in the food packaging and preservation industry. However, it relies on petroleum-based materials, which are non-renewable and increasingly scarce. Given the current situation, petroleum resources are depleting rapidly, creating numerous challenges related to their scarcity. Furthermore, plastic packaging is a material that either does not biodegrade or does so with difficulty [5,9-10]. This can have a significant impact on the environment due to the rapid increase in plastic waste. Consequently, there is a growing trend toward using natural and highly biodegradable materials as alternatives to synthetic ones in food packaging and preservation [5,9,11].

Usually, bio-packaging is crafted using various natural polymers, including glycerin, polyvinyl alcohol (PVA), chitin, chitosan, and starch [3]. Among these options, starch stands out as the most environmentally friendly, abundantly available, cost-effective, renewable, and accessible biopolymer [9,11]. It represents an ideal and promising source of raw materials for biopackaging production. Numerous studies have concentrated on the film-forming properties of starch and its applications in various fields such as food packaging, pharmaceutical packaging, biomedical materials, and more. However, starch-based films exhibit the drawback of being relatively hydrophilic and possessing unstable mechanical properties [9,11-12]. Therefore, it is necessary to incorporate a reinforcing agent into the starch matrix to enhance both mechanical properties and water resistance [9,11]. Lignocellulose, derived from lignocellulose sources, ranks among the primary ingredients added to starch to address the aforementioned shortcomings. Lignocellulose is a naturally occurring, abundant, low-cost, and renewable raw material characterized by high mechanical strength [11,13-17]. Studies have shown that adding lignocellulose to starch films significantly improves their mechanical properties [14-15,18-19]. Consequently, lignocellulose can be considered an ideal reinforcing material for overcoming the drawbacks associated with starch films.

Microcrystalline cellulose (MCC), a derivative of cellulose found in lignocellulose, serves as a safe and cost-effective material commonly employed in food applications [9,11]. It has low density and high mechanical strength, making it ideal for reinforcing biopolymers in film formation [20-21]. In comparison to conventional cellulose fibers, MCC possesses a greater specific surface area and smaller fiber size, allowing it to establish more uniform chemical cross-links within the starch network [11,13,15-17,22-23]. Thai Lan scientists utilized MCC derived from rice embryos to enhance the mechanical properties and biodegradability of composite biofilms [9]. Similarly, MCC obtained from soybean hulls was incorporated into starch, resulting in enhanced flexibility in the composite film [11]. While extensive research has been conducted on the physical and mechanical properties of MCC-reinforced starch films, studies pertaining to their application in fruit preservation remain limited.

This research aims to collect samples of starch films with different levels of MCC contents and evaluate properties such as film-forming ability, solution characteristics, water absorption capacity, viscosity, color, thickness, solubility, biodegradability, and surface morphology. The composite film samples exhibiting the best characteristics will be selected for preservation experiments on ripe tomatoes.

2. Materials and Methods

2.1. Materials

The composite film was composed of two primary components: corn starch and microcrystalline cellulose (MCC). The corn starch was procured from Tan Nhat Huong Trading Co., Ltd (Ho Chi Minh, Vietnam). Commercial MCC, a fine and opaque white powder with a purity of 99%, was acquired from Shandong Zunhong Biotechnology Co., Ltd. (China). Glycerol with a purity of 99% from Xilong Co. Ltd. (China) was employed as a plasticizer to facilitate the film formation.

2.2. Methods

2.2.1. Preparation of the Composite Films Based on Starch and MCC (St-MCC)

Composite biofilms (St-MCC) were prepared using a modified method by Jie Chen et al. [24]. Corn starch was added to distilled water at 1/40 g/ml, and 40 wt% glycerol was added to the mixture. The mixture was then stirred and heated at 70 ℃ for 10 minutes. Subsequently, the temperature was raised to 95 ℃, and continuous stirring for 30 minutes was carried out to facilitate the gelatinization process.

For the preparation of the MCC solution, MCC content of 4, 8, 12, 16, and 20 wt% was added to 20 ml of distilled water and stirred for 10 minutes. The resulting mixture was then sonicated for 30 minutes to achieve a homogeneous suspension.

The two prepared solutions were combined and continuously stirred for 30 minutes at 95 ℃. The mixture was gradually cooled to 45 ℃ using cold water while being continuously stirred. Finally, 100ml of the film-forming solution was poured into an 11 cm diameter mold and agitated thoroughly to ensure even distribution of the film-forming solution on the mold surface. Afterward, the films were collected for characterization studies following drying at 60 ℃ in a vacuum oven.

2.2.2. Viscosity

Film-forming solutions were prepared and cooled to a temperature of 25 ±3 ℃. The viscosity of the film-forming solutions was measured using a DV1MLVTJ0 viscometer (Brookfield, USA) at 25 ℃ with the set speed of 100 rpm. The results were measured three times and the average value was calculated.

2.2.3. Color

The color of the sample was measured using the handheld colorimeter LC100 (Lovibond, China). White color was chosen as the standard color before performing the color measurement. Color comparisons were randomly performed at three different positions of the sample and the average value was determined [25].

2.2.4. Thickness

To determine the thickness of the film, 30g of the film-forming solution for each formula was prepared and poured into molds with an 11 cm diameter. The molds were then dried at 60 ℃ until reaching a consistent weight. Subsequently, the thickness of the dried film was assessed at five random points using an electronic micrometer 293-240-30 (Mitutoyo, Japan) [26]. The resulting measurements were averaged, with the resolution 0.001 mm.

2.2.5. Water Absorption Capacity

The water absorption capacity of the composite films produced was determined according to the method of ASTM D570-98 [27]. Composite film samples measuring 2x2 cm were prepared and initially weighed (m_1). These prepared samples were then placed in a beaker containing 100 ml of room temperature water. After 5, 10, 20, 30, 40, 50, 60, and 90 minutes, the samples were retrieved from the beaker, and any excess water was removed by blotting with dry paper. Subsequently, the samples were dried until they reached a constant weight at 60 ℃. The final weights of the samples were recorded (m_2) [10-11]. The water absorption capacity of the film was calculated at each time point according to Eq. (1):

$$H = \frac{m_2 - m_1}{m_1}.100 \tag{1}$$

where, m_1: the initial mass of samples, g; m_2: the mass of samples after absorption, g; H: the water absorption capacity of the films, %.

2.2.6. Solubility

Samples measuring 2x2 cm were prepared and weighed as m_1. Each sample was then immersed in a beaker containing 100 ml of water for a duration of 24 hours at room temperature (22±5 ℃). Subsequently, the samples were taken out of the beaker and dried until a constant weight was achieved at 60 ℃ for 12 hours (m_2) [11,27]. The solubility of samples was determined using Eq. (2):

$$S = \frac{m_1 - m_2}{m_1}.100 \tag{2}$$

where, m_1: the initial mass of samples, g; m_2: the mass of samples after immersion, g; S: The solubility of samples, %.

2.2.7. Biodegradability

The landfill method carried out the biodegradability of the samples [27]. The biofilms were measured in the initial size and mass (m_1). The soil used in the study was collected from the same location and at the same sampling time. Bury the samples to a depth of about 5 cm in the prepared soil pots. After 2, 5, 10 and 15 days, the samples were removed from the soil, washed, dried, and weighed (m_2) after drying to constant weight at a temperature not exceeding 50 ℃. The biodegradability of samples was calculated by the Eq. (3):

$$B = \frac{m_1 - m_2}{m_1}.100 \tag{3}$$

where, m_1: the initial mass of sample before landfill, g; m_2: the mass of sample after landfill, g; B: the biodegradability, %.

2.2.8. The Surface Morphology

The surface morphology of the studied samples was examined using an electron microscope Optika B-820 (Italy) to assess the dispersion of MCC molecules on the starch background. Magnification of up to x200 was achieved. Film samples were obtained as described in Section 2.2.1 and were prepared with dimensions of 5x5 cm.

2.2.9. Application of the Composite Film St-M for Preservation of Ripe Tomatoes

The tomatoes used in this study were bright red tomatoes of uniform ripeness and size, surface without signs of spoilage or mildew. These tomatoes were of the VT10 variety and were meticulously handpicked from local farmers' plots in Hanoi, Vietnam. Upon harvesting, the fruits underwent a thorough washing with water and were subsequently air-dried on filter paper in the laboratory. Tomatoes were divided into an unpreserved group (control group) and a group preserved with composite film St-M (coating group). The experimental setup was replicated thrice, with each iteration comprising five tomatoes per group. The film-forming solution was prepared and cooled to room temperature. The film-forming solution was brushed three layers onto the surface of the tomatoes of the coating group with a brush, dried and stored under normal conditions, maintaining a temperature range of 25-28 ℃ and a relative humidity of 75%, for a period of 12 days. Throughout the storage duration, assessments were conducted on the morphology, color, spoilage status, weight loss, soluble solid content, vitamin C concentration, and total acid content of the tomatoes on days 0, 3, 6, 9, and 12 of storage.

2.2.10. The Decay Rate

The fruit samples were divided into two groups: the control group and the coated group. Fruits were visually inspected to identify signs of decay on the surface, including changes in color, indications of bacterial or fungal damage, softening, browning spots, or areas of injury, throughout the 12-day storage period. The two groups of fruits were stored separately in sealed boxes under normal temperature conditions (25±3 ℃). Each group comprised 10 fruits of consistent size and color. Observed signs were recorded, and the decay rate was calculated by determining the ratio of damaged fruits to the total number of fruits used in the experiment [25,28].

2.2.11. Firmness

The firmness of the fruit was assessed and measured using the TX700 fruit hardness tester (Lamy-Rheology, France). Firmness was quantified as the maximum force (cp) needed to depress the probe to a depth of 2 mm. Three fruits were tested within each group, with firmness readings taken at five random points on each fruit. The result was determined as the average of all measurements within each fruit group [28].

2.2.12. The Weight Loss

To determine the level of weight loss, three fruits from each sample group were tagged and weighed.The weight loss percentage (WLP) was calculated by the Eq. (4) [28]:

$$\mathrm{WLP} = \frac{m_2}{m_1}.100 \tag{4}$$

where, m_1: the initial mass of tomatoes, g; m_2: the mass of tomatoes after storage, g; WLP: the lose weight of fruit, %.

2.2.13. The Vitamin C Content of the Fruit

The vitamin C content of the fruit was determined by the iodometric titration method [29]. 10 ml of fruit juice was added to a 250 ml erlenmeyer flask. This was followed by the addition of 5ml of sulfuric acid solution and a few drops of starch indicator. The solution was then gently shaken. The mixture was titrated with a 0.01N I_2 solution until a blue color persisted for 15 seconds. The amount of I_2 used was recorded. The vitamin C content was calculated according to Eq. (5):

$$\mathrm{VTMC} = \frac{n.0.88.1000}{V} \tag{5}$$

where, VTMC: the vitamin C content in the fruit, mg/l; n: the volume of the 0.01N I_2 solution used for titration, ml; V: the volume of the test fruit solution, ml.

2.2.14. The Total Soluble Substances Content (TSS)

TSS of fruit was determined by a hand-held refractometer Atago ATC-1E (Atago - Japan) [30].

2.2.15. Total Acid Content

The total acid content was determined by the titration method [31]. 5 ml of fruit juice was added to a 250 ml erlenmeyer flask, followed by the addition of 5 drops of 1% phenolphthalein. The mixture was titrated with a 0.1N NaOH solution until a faint pink color appeared for 15 seconds. The amount of 0.1N NaOH consumed was recorded. The total acid content was calculated according to Eq. (6):

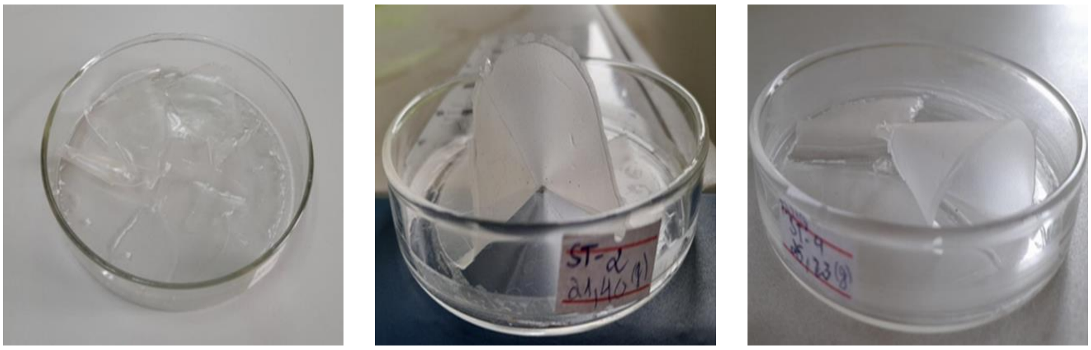
$$A_e = \frac{n.\,1000}{V} \tag{6}$$

where, A_e: the total acid content, mEq/l; n: the volume of the 0.1N NaOH solution used for titration, ml; V – the volume of the test fruit solution, ml.

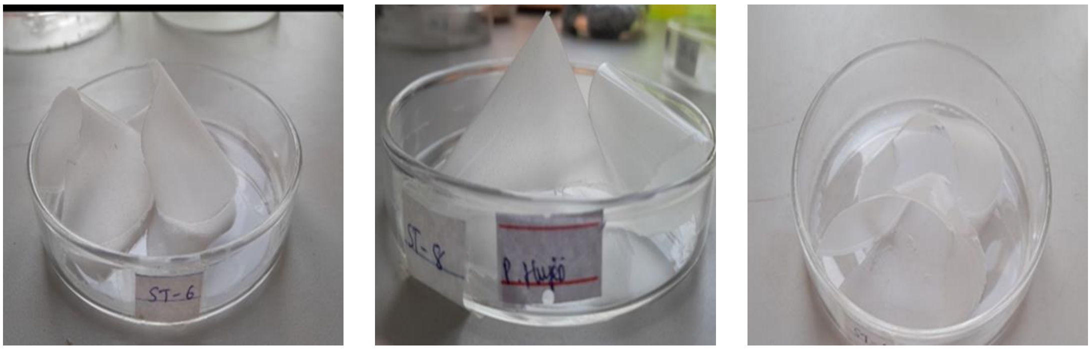
3. Results and Discussion

3.1. The Formation of the Composite Film St-MCC

The films were created using the die-casting method, with varying amounts of MCC ranging from 0% to 20%. Figure 1 displays a visual representation of the results, showing that adding MCC resulted in opaque white composite films, with turbidity increasing proportionately to the amount of MCC added. In contrast, films made solely from starch were transparent white, as seen in Figure 1a. The MCC-containing films were also thicker than the starch films despite having the same film formation mass. When attempting to remove the film from the mold, the films without MCC proved challenging to separate, resulting in the St-0% MCC film becoming torn and no longer intact. However, the composite films containing MCC were much easier to separate from the plate, and the film acquisition process resulted in significantly less loss than pure films. These results demonstrate that adding MCC to starch produces much more efficient film fabrication than creating starch films.



(a) St – 0% MCC (b) St – 4% MCC (c) St – 8% MCC



(d) St – 12% MCC (e) St – 16% MCC (f) St – 20% MCC

Figure 1. Visual images of the St-MCC composite film

In this experiment, the properties of the film-forming solution were also studied. The results showed that the solution without MCC had high viscosity and exhibited strong adhesion and consistency when viewed with the naked eye. On the other hand, the solution containing MCC had slightly lower viscosity, adhesion, and flexibility but still showed considerable improvement compared to the solution without MCC (Fig. 2). This property of the composite solution, known as St-M, has reduced production losses and improved economic efficiency.

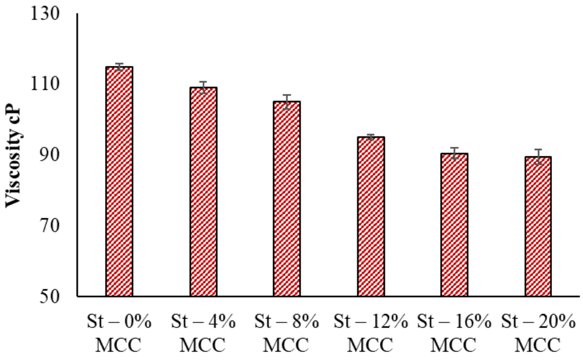


Figure 2. Viscosity of the composite film St-MCC

Starch and MCC have molecular formulas that include free hydroxyl groups. These groups can create bonds between starch, glycerol, and MCC, resulting in a sustainable three-dimensional network of links. As a result, the starch molecules were almost entirely connected to the MCC molecules, and the viscosity of the mixtures, referred to as St-M, changed significantly.

3.2. Color

When applying films for packaging or preserving food, the color of the film will significantly affect the sensory properties of the product, thereby influencing the level of consumer preference for the product. Therefore, evaluating the color of the film will contribute to correctly determining the selection of product targets to apply appropriately for the composite film, especially in food packaging. The color difference of films supplemented with MCC compared to films without MCC (St-0% MCC) has been evaluated and recorded (Fig. 3). It is noted that the film containing only starch is fairly transparent and colorless, while films containing MCC are more opaque. As the MCC content increases, the color difference ΔE increases and fluctuates from 7.95 (St-4%MCC) to 25.08 (St-20%MCC). It has been pointed out that when ΔE is greater than 3.5, it means that the human eye can perceive color differences between samples. The ΔE values of films containing MCC are all greater than 3.5, indicating significant color changes compared to the base film. The color comparison results are consistent with the visual images observed (Figure 1). Similar results were also observed when supplementing MCC into synthesized starch films [32].

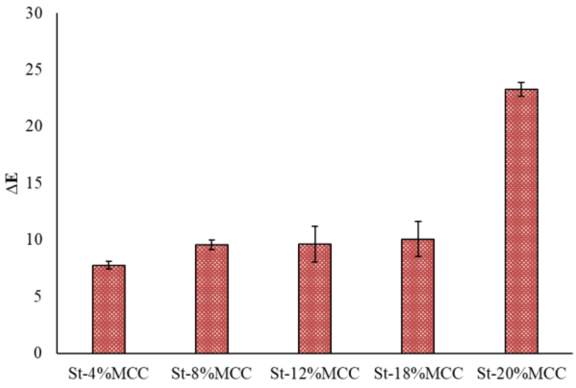


Figure 3. The color difference of films St-MCC compared to films St without MCC

3.3. Thickness

The thickness of the film affects its permeability and durability. Additionally, the thickness of the coating layer also affects the color intensity and visual aesthetics of the fruit. Therefore, it is necessary to investigate the thickness of the film with different levels of MCC supplementation. The results of the average thickness of the research films are presented in Figure 4. When comparing the thickness of the films with different levels of added MCC, it is observed that the addition of MCC does not affect significantly the thickness of the obtained dried films. In this experiment, the films had an average thickness ranging from 0.030 to 0.035 mm. The measurement error at different positions of the same film sample is relatively small. This indicates that the thickness of the film at different positions is relatively uniform. Uniform film thickness during casting will facilitate the preservation process with the film, avoiding local irregularities in thickness. Therefore, with this finding, it can be seen that the starch film supplemented with MCC is suitable for preservation applications.

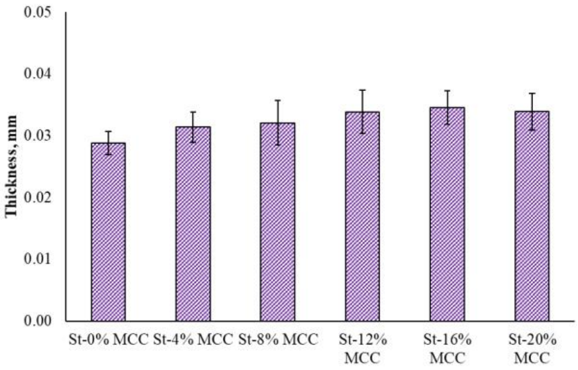


Figure 4. The thickness of the biofilms

3.4. Water Absorption Capacity

Maintaining a humid environment is crucial in film applications for preserving fresh fruit, so the water absorption capacity plays a significant role in achieving this [33]. Figure 5 presents the water absorption capacity of the composite films studied. The findings indicate that the composite films' water absorption was lower than starch films. Furthermore, as the MCC content ratio increased, the water absorption decreased. However, when the MCC content ratio reached 20%, the film's water absorption capacity increased compared to other composite films. Adding MCC to a starch solution strengthened the bond between MCC, starch, and glycerol, forming a dense network between the ingredients [24]. This network reduced the water absorption of the St-M composite films. It increased the MCC content to 20 wt.% and saturated the bonds between the components, making the MCC molecules single. MCC, being hydrophilic, could absorb more water, making the St-20% MCC film more hydrophilic than composite films with 12 wt.% and 16 wt.% MCC content. These results demonstrated that adding MCC to the starch film improved the water absorption properties of blank film.

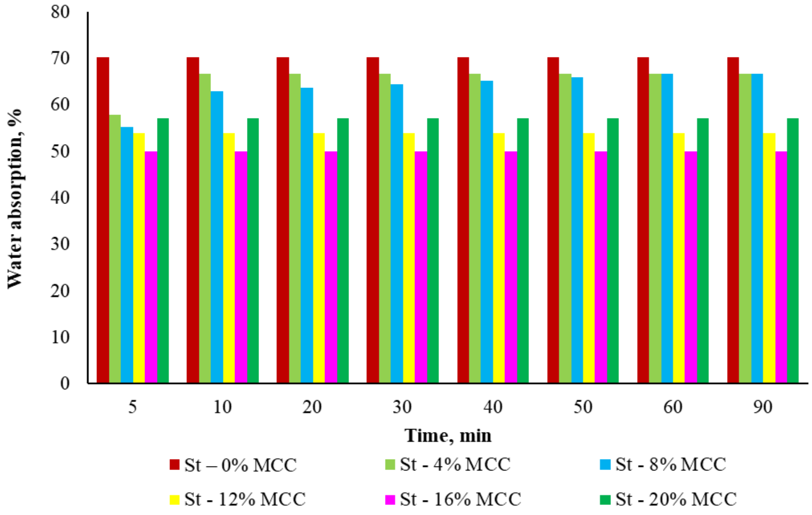


Figure 5. Water absorption capacity of the studied composite films

3.5. Solubility

Many food products, mainly fresh fruits, have high water content. This makes it challenging to use films with high solubility for food preservation. As a result, it is essential to study the solubility properties of these films. Figure 6 displays the solubility of the films that were analyzed. The findings indicate that composite films have higher solubility than films without MCC. Increasing the MCC content ratio results in a higher solubility for composite films. However, adding MCC at 4% and 8% content resulted in a solubility that was not significantly different from the St-0% MCC film. When MCC content increased to 12 wt.%, the solubility of the composite film increased significantly. With such a high level of solubility of the MCC-supplemented film, it can be seen to provide favorable conditions for the preservation of fresh fruits. Fruit preservation films with high solubility will dissolve during storage due to the fruit's respiration, which generates moisture. However, if the film is not soluble, it will inconvenience the usage process. This means that after preservation, consumers can easily remove the film by rinsing it in water.

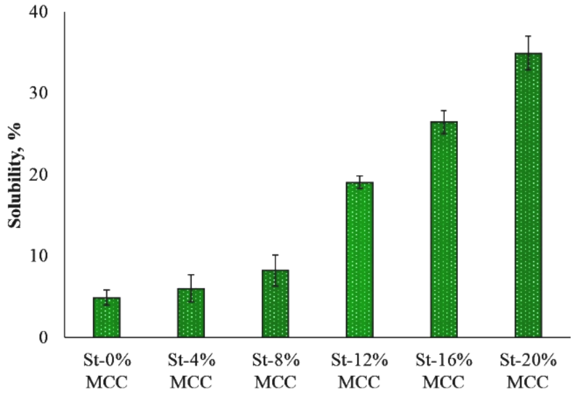


Figure 6. The solubility of the studied composite films

3.6. Biodegradability

The results of the biodegradation of the studied films through the landfill method are displayed in Figure 7. After two days of burial, all films showed similar levels of biodegradability, ranging from 18% to 27%. However, after five days of burial, all samples started to crack and tear, losing their initial integrity. The film without MCC and composite films exhibited a self-decomposition ability of 29.58% to 45.45%. Figure 4 shows that adding MCC reduced the biodegradability of the composite films not much further compared to the starch films. After ten days of burial, almost 80% of all films were decomposed into tiny pieces and mixed with the soil. The films were almost completely decomposed after 20 days of burial, with no small fragments detected in the ground. This demonstrated that the composite films St-M had good biodegradability, similar to the blank starch films. With the rapid degradation of the research films, it will facilitate the post-use disposal process. Burial is a simple and easily implementable method. Additionally, the research films' ability to completely degrade within 20 days will help minimize the accumulation of large volumes of non-biodegradable plastic waste as seen today.

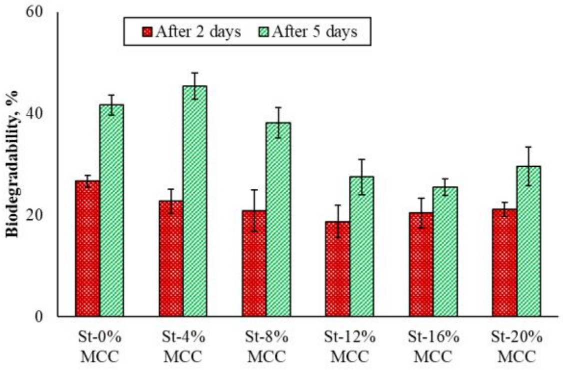
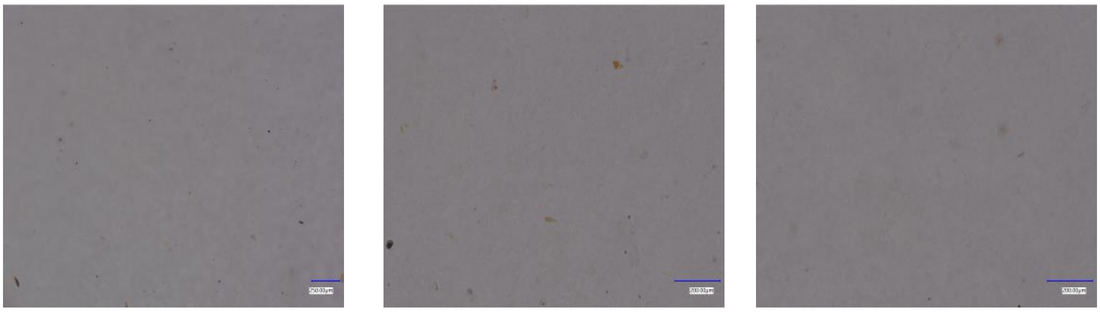


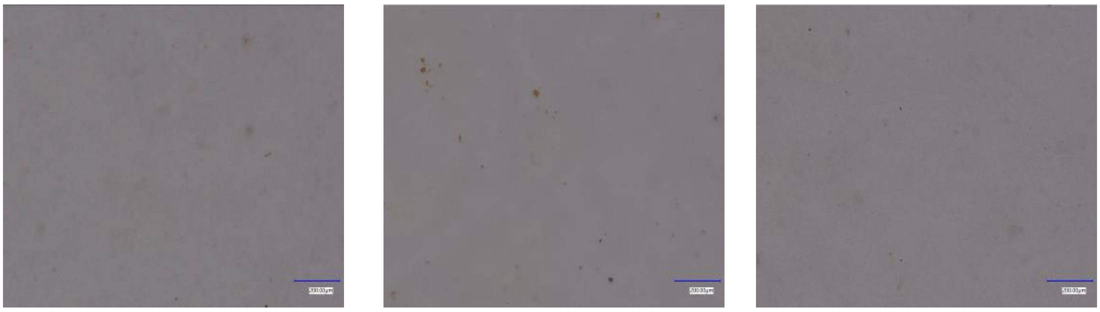
Figure 7. The biodegradability of the studied films

3.7. Morphological Analysis

Figure 8 displays the surface morphology of composite films created using different ratios of starch and MCC. The film without MCC had a relatively smooth surface (Figure 8a), which can be attributed to the solubility of starch in water and the formation of cross-links between starch, water, and glycerol molecules [24]. However, adding MCC to the starch films altered the surface morphology of the resulting films, as shown in Figures 5b-f. The MCC molecules were evenly distributed on the St-M films, even when the MCC content was up to 20 wt.%. But when the MCC content increased to 16 and 20 wt.%, MCC self-aggregation occurred, leading to large particle clusters and reduced surface smoothness (Figures 8e, f).



(a) St – 0% MCC (b) St – 4% MCC (c) St – 8% MCC



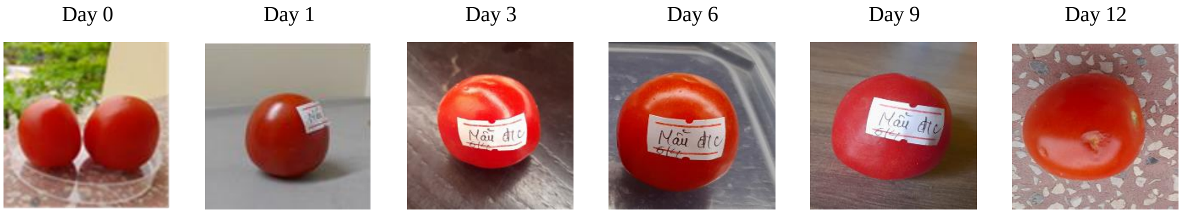
(d) St – 12% MCC (e) St – 16% MCC (f) St – 20% MCC

Figure 8. Surface morphology of the studied films

According to the survey, composite films with 4% and 8% MCC content are better than those with higher MCC content. In particular, the film St-4% MCC has the highest biodegradability; its solubility is comparable to that of the blank starch film and higher than those with a higher content of MCC. Besides, the water absorption capacity of the film St-4% MCC is improved than that of the film St-0% MCC. Therefore, the St-4% MCC film is the best choice for preserving fresh fruit.

3.8. Preservation of Ripe Tomatoes with a Composite Film

Biofilm has a preservative effect that helps fruits maintain freshness and moisture [26,34-35]. Ripe tomatoes were chosen as the subjects to study the preservation application using St-4% MCC film. The purchased fruits were washed, drained, and not subjected to any sterilization process. The study included two groups of fruits: the control group, which did not use any composite films or preservation measures, and the coating group, which had three layers of film applied to the fruit's skin (Figure 9). As observed in Figure 6, the group of fruits not preserved with film still maintained their colour; however, the gloss of the fruit decreased significantly from day 3, and by day 6, the fruit surface appeared damaged, the hardness on day 9 began to decline sharply, and on the day 12, fungal growth appeared on the fruit surface (Figure 9a). Meanwhile, for the coating tomato group, the colour and gloss of the fruit were maintained until day 12. The fruit began to soften on day 9, and on day 12, the fruit surface started to show signs of surface damage (Figure 9b). Thus, it can be seen that the film St-4% MCC helped preserve the freshness of the fruit, prolonging the shelf life of the fruit compared to the control group. Similar results were also observed when preserving tomatoes with a chitosan coating combined with MCC [36].



(a) Film coating

(b) Uncoated (control)

Figure 9. Tomatoes ripen with and without film coating

Fruit decay rate. The fruit samples were visually assessed for external condition through indicators such as color changes observed by the naked eye, the presence of signs of damage, brown spots, or soft mushy areas on the fruit surface. These indicators were recorded to evaluate the decay rate of the two research fruit groups (Figure 10A). The data obtained shows that the group of fruits not preserved with the film had a significantly rapid increase in decay rate from the 9th day of preservation, while the decay rate of the group preserved with the film was much lower. The outer coating layer of the fruit creates a barrier that separates the fruit from deteriorating agents such as fungi or microorganisms in the ambient air. Therefore, the process of fruit decay is limited more effectively in the coated group compared to the non-coated group.

Color. One of the factors affecting the quality of preserved fruits is their color. Therefore, monitoring and studying the color of fruits throughout the preservation process is necessary. The results of fruit color over the preservation period are illustrated in Figure 10B. Typically, a ΔE value ranging from 0 to 1 indicates a color difference that is imperceptible to the naked eye, or in other words, a negligible color difference. ΔE values ranging from 1 to 3 indicate a color difference that can be observed by the naked eye. When ΔE exceeds 3.5, the color difference becomes distinctly noticeable [2]. For ripe tomatoes, the color usually transitions from bright red to dark red. The change in color during fruit storage is primarily due to the pigments present in the fruit, specifically the degradation of these pigments and enzymatic reactions causing the transition from bright red to dark red. Therefore, the less the color of the fruit changes, the longer the fruit's shelf life. From the data obtained, it is observed that after 12 days of preservation, the color change in the control group is significantly higher compared to the coated sample. Additionally, it is noted that when using the film, tomatoes tend to change color more slowly compared to the control sample. This indicates that the degradation of pigments and enzymes is limited. This may be attributed to the enzymes' ability to act on and influence the coating layer.

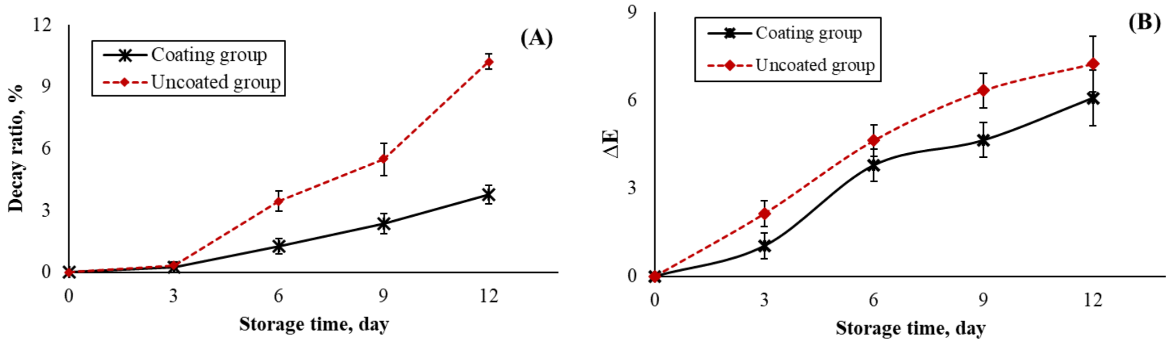
Firmness. Firmness is also one of the key parameters for evaluating fruit quality. Figure 10C depicts the extent of hardness reduction in fruit groups over the storage period. The results indicate that as the storage time increases, the degree of hardness reduction in the fruit also increases. Compared to the control fruit sample without preservation, fruits coated with the research film exhibit lower levels of hardness reduction, implying that fruits preserved with the film have higher hardness compared to the control group. In the initial 9 days of storage, there is a slight decrease in hardness, but after 9 days, the hardness of the fruit tends to decrease rapidly. This result can be explained by aging processes, cell degradation, and pectin hydrolysis, leading to softening of the fruit [37-38]. For fruits coated with a supplementary starch film containing MCC, this film limits fruit exposure to external air, thereby restraining fruit respiration processes and slowing down biochemical reactions. Consequently, fewer products of these processes are generated, meaning that the fruits remain fresh for a longer period, or in other words, the fruits maintain their freshness for a longer duration.

The weight loss percentage (WLP). The primary cause of fruit weight loss is dehydration. During post-harvest handling and storage, fruits are prone to water loss due to respiration and transpiration processes [34,39]. Higher respiration rates and CO_2 diffusion within the fruit peel lead to more pronounced water loss [40]. Fruit weight loss results in the outer skin becoming shriveled and less appealing in appearance. Therefore, fruits with less weight loss remain fresher for longer and have an extended shelf life. Figure 10D shows that the weight loss of the control group increased rapidly from day 6, while the weight loss of the coating group increased more slowly. At the same time, the value WLP of the coating group on day 12 was also lower than that of the control group. This proves that the studied film St-4% MCC has the effect of helping to reduce the weight loss of the fruit. This may be attributed to the water-absorbing properties of the starch film supplemented with MCC. During storage, the research film can absorb water from the fruit, retaining it on the film surface, thus limiting water loss to the surrounding environment. In other words, the coating film acts as a barrier preventing water loss and maintaining moisture around the fruit. Consequently, fruits experience less dehydration, leading to prolonged freshness.

The total soluble solids percentages (TSS). The soluble solids in fruits mainly include components such as sugars, acids, soluble proteins, minerals, vitamins, and pigments. During fruit growth and aging, soluble solids tend to increase as a result of the hydrolysis of insoluble polysaccharides into soluble monosaccharides [2]. Therefore, if the total soluble solids (TSS) of fresh fruits are maintained or similar to those of the original product, it means that the fruits have a longer shelf life [2]. In this experiment, the control sample significantly increased TSS after six days. Meanwhile, the coating sample maintained TSS comparable to baseline until day 12. For the uncoated sample, TSS increased significantly, whereas in the first 9 days of storage, the coated samples showed a tendency to increase in TSS, but the rate of increase was much lower compared to the control sample (Fig.10E). After 12 days of storage, the control sample showed a TSS increase of up to 28.86% compared to the initial level, while the samples coated with starch-MCC film only increased by 14.29% compared to the initial level. This indicates that the starch-MCC film effectively inhibited the respiration process and thus slowed down the hydrolysis of polysaccharides into simple sugars. This finding is consistent with the results of Ali et al., who reported that preserving sweet cherry fruits with a gum guar and ginseng coating also increased the TSS content during storage [41].

The total acidity (TA). The acid content in the fruit is primarily composed of organic acids. Typically, during post-harvest storage of fruits, these organic acids are utilized as substrates for respiration processes or converted into sugars [41], resulting in a gradual decrease in acid content in the fruit. The maintenance of these acid levels indicates a slower respiration process and limited conversion into sugars. Data in Figure 10F shows that the TA of both fruit samples decreased with storage time. The data shows that for the uncoated fruit group, the TA loss rate occurred relatively quickly; from day 9, the TA loss reached 62.03%. Meanwhile, for the coating group, the TA loss occurred significantly slower; specifically, the TA loss was only 36.73% (day 9) and 42.94% (day 12). Thus, it can be seen that the coating group maintains the TA content in the fruit longer; this means that the respiration process of the coated by film St-4% MCC fruit group is limited. This can be attributed to the starch coating combined with MCC forming a semi-permeable membrane on the fruit surface, altering the surrounding air composition, specifically increasing CO_2 levels and decreasing O_2 levels. This altered air composition effectively slows down ripening and biochemical processes in the fruit [42].

The ascorbic acid loss (AA). Fruits contain quite a large amount of ascorbic acid (vitamin C). However, when the storage time is increased, the fruit loses this vitamin content relatively much. Figure 10G shows that, for fruit not preserved by film, the loss of AA increased rapidly from day 9, reaching 50.89% (day 9) and 63.17% (day 12). Meanwhile, AA loss increased significantly for the coating fruit group from day 12, only 20.20% (day 9) and 36.62% (day 12). Thus, it can be shown that fruit preserved with research film will help maintain vitamin C content in the fruit better. Vitamin C is a relatively sensitive component, especially to air exposure. The more the fruit comes into contact with air, the easier it is for vitamin C to degrade. In the results of this experiment, the research membrane acted to create a barrier between the fruit and the air, thereby limiting the degradation of vitamin C present in the fruit.



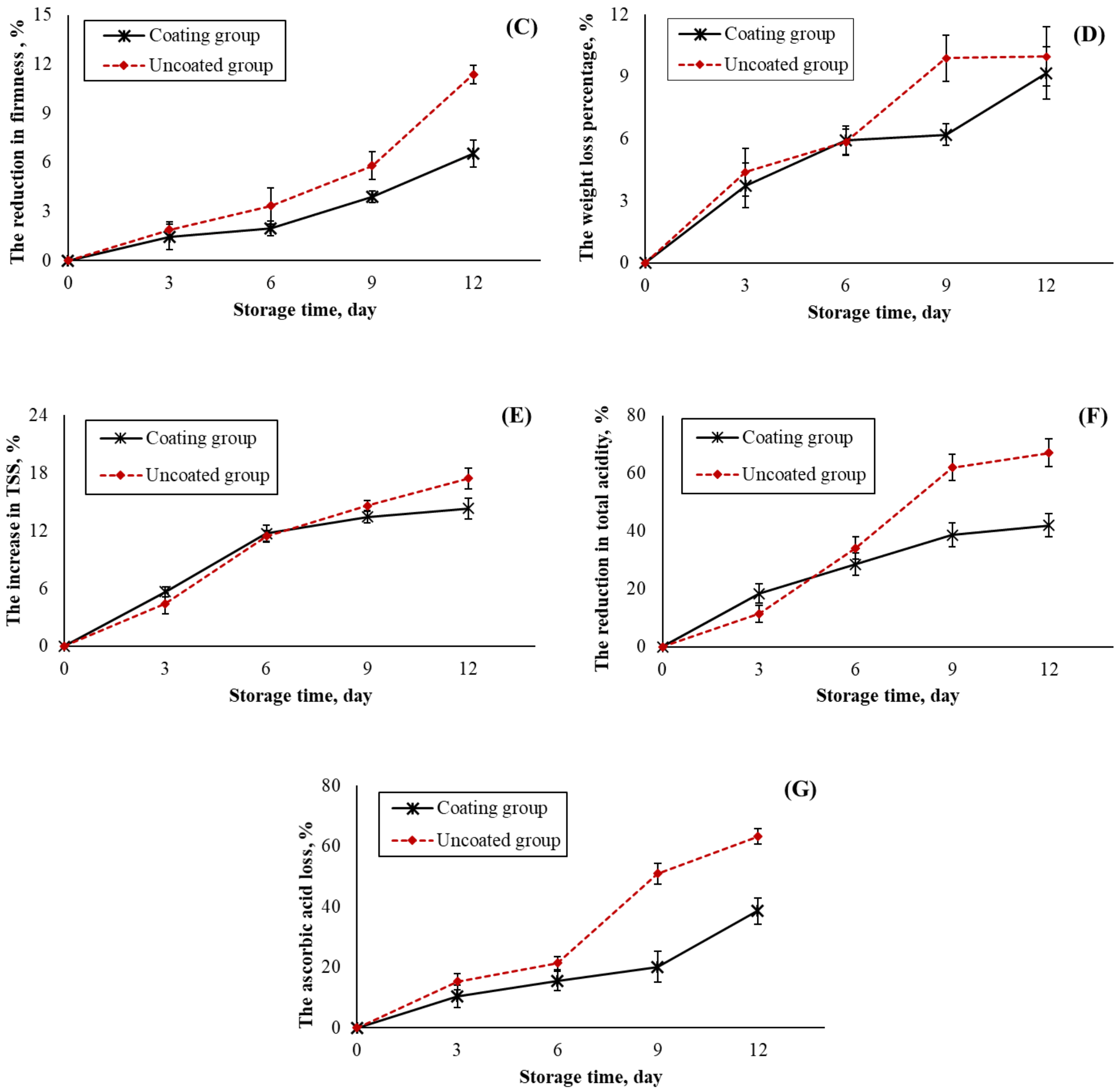


Figure 10. (A) Decay rate, (B) the change of color (C) Firmness, (D) The weight loss percentage, (E) The total soluble solids percentages, (F) The reduction in total acidity, (G) The ascorbic acid (AA) loss of fruit in the storage time

4. Conclusions

In this study, composite films based on starch and MCC with the support of glycerol as a plasticizer were obtained, and some of their properties were studied. Films containing MCC have more turbidity, higher solution viscosity, and reduced adhesion properties than blank starch films. Composite films containing MCC have reduced water absorption and increased solubility compared to films without MCC. The biodegradability of film St-4% MCC was the highest, reaching 45.45% after five days of burial in the soil. The surface of the films with MCC was rougher when adding 20 wt% MCC. Application of film St-4% MCC for post-harvest ripe tomato preservation shows potential results. The study demonstrates that the St-4% MCC composite film effectively preserves the freshness and quality of ripe tomatoes. The coating significantly delays decay, maintains color, and reduces firmness loss compared to untreated fruits. Additionally, the film minimizes weight loss, total soluble solids, total acidity, and ascorbic acid content, showcasing its potential as a promising preservation method for maintaining fruit quality and nutritional value over an extended storage period.

Acknowledgments

The authors wish to thank the supports from the HaUI Institute of Technology (HIT), Faculty of Chemical Technology and Faculty of Mechanical Engineering (Hanoi University of Industry, Vietnam).

Author Contributions

Tran Y Doan Trang, Ha Thi Dzung and Do Thi Hanh: performed the experiments; Tran Y Doan Trang and Ha Thi Nha Phuong: analysed and interpreted the data; Tran Y Doan Trang and Ta Thi Huong: wrote the paper; Tran Y Doan Trang, Nguyen Thi Thu Hien and Pham Huong Quynh: conceived and designed the experiments; Tran Y Doan Trang, Do Thi Hanh and Hoang Thanh Duc: contributed reagents, materials, analysis tools or data.

Declaration of Interests Statement

The authors declare no conflict of interest.

REFERENCES

[1] Samya B., H. Hajjaj, "Lycopene Extracted from Tomato - A Review," Food Science and Technology, vol. 12, No. 1, pp. 1-14, 2024. DOI: https://doi.org/10.13189/fst.2024.120101.

[2] Li Y., Zhou Y., Wang Z., Cai R., Yue T., L. Cui, “Preparation and characterization of chitosan–nano-ZnO composite films for preservation of cherry tomatoes,” Foods, vol. 10, no. 12, 3135, 2021. DOI: https://doi.org/10.3390/foods10123135.

[3] Tran Y.D.T., Ha T.D., Ta T.H., Le Q.D., Do T.H., and T.N.P. Ha, “Preparation and characterization of chitosan/*Aloe Vera* gel film for fresh fruit preservation,” E3S Web of Conferences, vol. 443, 02003, https://doi.org/10.1051/e3sconf/202344302003.

[4] Yun X., Qi X., Zhang Y., Song S., T. Dong, “Application of SiOx-coated poly (ε-caprolactone) film for preservation of cherry tomato,” Polymers and Polymer Composites, vol. 28, no. 5, pp. 309-319, 2020. DOI: https://doi.org/10.1177/096739111987455.

[5] Yeo S.P., Siti A.O., N.N. Nur, “The effects of gamma radiation on bioplastics,” Food Science and Technology, vol. 7, no. 4, pp. 53 – 56, 2019. DOI: https://doi.org/10.13189/fst.2019.070402.

[6] Muazu I., Edinah K.S., Samuel N., Elijah M.A., Justus O., W.N. Victoria, “Identification and in vitro characterization of plant growth-promoting *pseudomonas* spp. Isolated from the Rhizosphere of Tomato (*Lycopersicum esculentum*) Plants in Kenya,” Universal Journal of Agricultural Research, vol. 10, no. 6, pp. 667 – 681, 2022. DOI: https://doi.org/10.13189/ujar.2022.100608.

[7] Our World in Data, Tomato Yield, 1961 to 2021. https://ourworldindata.org/grapher/tomato-yields?tab=chart.

[8] Laila A.N., Joydeep D., D. Sergey, “Nanocomposite zinc oxide-chitosan coatings on polyethylene films for extending storage life of okra (Abelmoschus esculentus),” Nanomaterials, vol. 8, no. 7, pp. 479–492, 2018. DOI: https://doi.org/10.3390/nano8070479.

[9] Suklaw N. and U. Ratanakamnuan, “Mechanical properties and biodegradability of starch-based biocomposite films reinforced with microcrystalline cellulose from rice embryo,” Journal of Physics: Conference Series, vol. 2175, 012034, 2022. DOI: https://doi.org/10.1088/1742-6596/2175/1/012034.

[10] Tran Y.D.T., L.A. Zenitova, “Effective treatment of oil spills by adsorbent formed from chitin and polyurethane foam,” Current Applied Science and Technology, vol. 20, no. 2, pp. 321–333, 2020. DOI: https://doi.org/10.14456/cast.2020.19.

[11] Merci A., Marim R.G., Urbano A., S. Mali, “Films based on cassava starch reinforced with soybean hulls or microcrystalline cellulose from soybean hulls,” Food Packaging and Shelf Life, vol. 20, 100321, 2019. DOI: https://doi.org/10.1016/j.fpsl.2019.100321.

[12] Garcia P.S., Baron A.M., Yamashita F., Mali S., Eiras D., M.V.E. Grossmann, “Compatibilization of starch/poly (butylene adipate-co-terephthalate) blown films using itaconic acid and sodium hypophosphite,” Journal of Applied Polymer Science, vol. 135, no. 33, 46629, 2018. DOI: https://doi.org/10.1002/app.46629.

[13] Li C., Dongli Z., Zhengbiao G., Zhaofeng L., Yan H., L. Caiming, “Preparation of acetylated nanofibrillated cellulose from corn stalk microcrystalline cellulose and its reinforcing effect on starch films,” International Journal of Biological Macromolecules, vol. 11, pp. 959-966, 2018. DOI: https://doi.org/10.1016/j.ijbiomac.2018.01.056.

[14] Kargarzadeh H., Johar N., I. Ahmad, “Starch biocomposite film reinforced by multiscale rice husk fiber,” Composites Science and Technology, vol. 151, pp. 147-155, 2017. DOI: https://doi.org/10.1016/j.compscitech.2017.08.018.

[15] Nascimento P.H.A., Marim R.G., Carvalho G.M., and S. Mali, “Nanocellulose produced from rice hulls and its effect on the properties of biodegradable starch films,” Materials Research, vol. 19, no. 1, pp. 167–174, 2016. DOI: https://doi.org/10.1590/1980-5373-MR-2015-0423.

[16] Tian Y., Zhang K., Zhou M., Wei Y., Cheng F., Lin Y., et al. “High-performance starch films reinforced with microcrystalline cellulose made from eucalyptus pulp via ball milling and mercerization,” Starch – Stärke, vol. 71, no 3-4, 1800218, 2018. DOI: https://doi.org/10.1002/star.201800218.

[17] Xuejiao H., Fei X., X. Xiaopeng, “Surface-modified microcrystalline cellulose for reinforcement of chitosan film,” Carbohydrate Polymers, vol. 201, pp. 367–373, 2018. DOI: https://doi.org/10.1016/j.carbpol.2018.08.085.

[18] Chen Q-J., Zhou L-L., Zou J-Q. and X. Gao, “The preparation and characterization of nanocomposite film reinforced by modified cellulose nanocrystals,” International Journal of Biological Macromolecules, vol. 132, pp. 1155-1162, 2019. DOI: https://doi.org/10.1016/j.ijbiomac.2019.04.063.

[19] Zhao Y., Huerta R.R. and M.D.A. Saldaña, “Use of subcritical water technology to develop cassava starch/chitosan/gallic acid bioactive films reinforced with cellulose nanofibers from canola straw,” The Journal of Supercritical Fluids, vol. 148, pp. 55-65, 2019. DOI: https://doi.org/10.1016/j.supflu.2019.02.022.

[20] Deng S., Binauld S., Mangiante G., Frances J.M., Charlot A., Bernard J. et al., “Microcrystalline cellulose as reinforcing agent in silicone elastomers,” Carbohydrate

Polymers, vol. 151, pp. 899–906, 2016. DOI: https://doi.org/10.1016/j.carbpol.2016.06.035.

[21] Ummartyotin S. and C. Pechyen, "Microcrystalline-cellulose and polypropylene based composite: A simple, selective, and effective material for microwavable packaging," Carbohydrate Polymers, vol. 142, pp. 133–140, 2016. DOI: https://doi.org/10.1016/j.carbpol.2016.01.020.

[22] Maulida, Siagian M., P. Tarigan, "Production of starch-based bioplastic from cassava peel reinforced with microcrystalline celllulose avicel PH101 using sorbitol as plasticizer," Journal of Physics Conference Series, vol. 710, pp. 1–7, 2016. DOI: https://doi.org/10.1088/1742-6596/710/1/012012.

[23] Trache D., Hussin M.H., Chuin C.T.H., Sabar S., Fazita M.N., Taiwo O.F. and et al., "Microcrystalline cellulose: isolation, characterization, and bio-composites application - A review," International Journal of Biological Macromolecules, vol. 93, part A, pp. 789–804, 2016. DOI: https://doi.org/10.1016/j.ijbiomac.2016.09.056.

[24] Chen J., Long Z., Wang J. and et al., "Preparation and properties of microcrystalline cellulose/hydroxypropyl starch composite films," Cellulose, vol. 24, pp. 4449–4459 2017. DOI: https://doi.org/10.1007/s10570-017-1423-6.

[25] Khalil H.A., Abdelkader M.F.M., Lo'ay A.A., El-Ansary D.O., Shaaban F.K.M., Osman S.O., Shenawy I.E., Osman H.-E.H., Limam S.A., Abdein M.A., et al. "The combined effect of hot water treatment and chitosan coating on mango (*Mangifera Indica L. Cv*. kent) fruits to control postharvest deterioration and increase fruit quality," Coatings, vol. 12, 10083, 2022. DOI: https://doi.org/10.3390/coatings12010083

[26] Zárate-Moreno J.C., Escobar-Sierra D.M., R. Ríos-Estepa, "Development and evaluation of chitosan-based food coatings for exotic fruit preservation," BioTech, vol. 12, 10020, 2023. DOI: https://doi.org/10.3390/biotech12010020.

[27] ASTM D570-9 Standard test method for water absorption of plastics, ASTM International, USA, 2018.

[28] Efthymiou M.N., Tsouko E., Papagiannopoulos A. et al. "Development of biodegradable films using sunflower protein isolates and bacterial nanocellulose as innovative food packaging materials for fresh fruit preservation," Sci Rep, vol. 12, 6935, 2022. DOI: https://doi.org/10.1038/s41598-022-10913-6

[29] Ikewuchi C.J. and C.C. Ikewuchi, "Iodometric determination of the ascorbic acid (vitamin C) content of some fruits consumed in a university community in Nigeria," Global journal of pure and applied sciences, vol. 17, no.1, pp. 47-49, 2011.

[30] Kusumiyati, Hadiwijaya Y., Putri I.E., Mubarok S. and J.S. Hamdani, "Rapid and non-destructive prediction of total soluble solids of guava fruits at various storage periods using handheld near-infrared instrument," IOP Conference Series: Earth and Environmental Science, vol. 458, 012022, 2020. DOI: https://doi.org/10.1088/1755-1315/458/1/012022

[31] Catrin T., D.S. George, Chapter "pH and Titratable Acidity" part of the "Food Science Text Series," Food analysis, pp. 389-406, 2017. DOI: https://doi.org/10.1007/978-3-319-45776-5_36

[32] Nor A.O., Fatmawati A., H.M.Y. Nur, "Reinforced bioplastic film at different microcrystalline cellulose concentration," Materials Today: Proceedings, vol. 41, part 1, pp 77-82, 2021. DOI: https://doi.org/10.1016/j.matpr.2020.11.1010.

[33] Mansbridge J.N., Ratner B.D., Hoffman A.S., Schoen F.J., J.E. Lemons, "Tissue-engineered skin substitutes," in Biomaterials Science, 3rd ed, Eds.; Academic Press: Millbrae, CA, USA, 2013, pp. 1276–1288.

[34] Maringgal B., Hashim N., Tawakkal I.S.M.A., M.T.M. Mohamed, "Recent advance in edible coating and its effect on fresh/fresh-cut fruits quality," Trends in Food Science & Technology, vol. 96, pp. 253–267, 2020. DOI: https://doi.org/10.1016/j.tifs.2019.12.024.

[35] G. Kerch, "Chitosan films and coatings prevent losses of fresh fruit nutritional quality: A review," Trends in Food Science & Technology, vol. 46, no. 2, part A, pp. 159–166, 2015. DOI: https://doi.org/10.1016/j.tifs.2015.10.010.

[36] Tran Y.D.T., Ha T.D, Ta T.H., and H.Q. Pham, "Composite biofilm chitosan-microcrystalline cellulose for tomato preservation," IOP Publishing, Journal of Physics: Conference Series, vol. 2697, 012049, 2024. DOI: https://doi.org/10.1088/1742-6596/2697/1/012049.

[37] Posé S., Morris V.J., Kirby A.R., Quesada M.A. and J.A. Mercado, "Fruit softening and pectin disassembly: An overview of nanostructural pectin modifications assessed by atomic force microscopy," Ann. Bot., vol. 114, pp. 1375–1383, 2014. DOI: https://doi.org/10.1093/aob/mcu149.

[38] Sultan M., Hafez O.M., Saleh M.A., A.M. Youssef, "Smart edible coating films based on chitosan and beeswax-pollen grains for the postharvest preservation of Le Conte pear," RSC Advances, vol. 11, no. 16, pp. 9572-9585, 2021. DOI: https://doi.org/10.1039/D0RA10671B.

[39] Martínez-Romero D., Alburquerque N., Valverde J.M., Guillén F., Castillo S., Valero D., M. Serrano, "Postharvest sweet cherry quality and safety maintenance by Aloe vera treatment: a new edible coating," Postharvest Biology and Technology, vol. 39, no. 1, pp. 93–100, 2006. DOI: https://doi.org/10.1016/j.postharvbio.2005.09.006.

[40] Ali A., Muhammad M.T.M., Sijam K., Y. Siddiqui, "Effect of chitosan coatings on the physicochemical characteristics of *Eksotika* II papaya (*Carica papaya* L) fruit during cold storage," Food Chem., vol. 124, pp. 620–626, 2011. DOI: https://doi.org/10.1016/j.foodchem.2010.06.085

[41] Dong F., X. Wang, "Effects of carboxymethyl celluloseincorporated with garlic essential oil composite coatings for im-proving quality of strawberries," International Journal of Biologi-cal Macromolecules, vol. 104, pp. 821–826, 2017. DOI: https://doi.org/10.1016/j.ijbiomac.2017.06.091.

[42] Fagundes C., Moraes K., Pérez-Gago M.B., Palou L., Maraschin M., A.R. Monteiro, "Effect of active modified atmosphereand cold storage on the postharvest quality of cherry tomatoes," Postharvest Biology and Technology, vol. 109, pp. 73–81, 2015. DOI: https://doi.org/10.1016/j.postharvbio.2015.05.017.