Preparation and characterization of chitosan/ *Aloe Vera* gel film for fresh fruit preservation

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Abstract. Biofilm preservation has become a topic of interest among many scientists. A recent study involved the production of chitosan-based biofilms containing varying amounts of aloe vera gel (5%, 10% and 15% w/w). Evaluation of film formation, water absorption, swelling ratio, solubility, antibacterial ability, colouration, and biodegradability indicated that chitosan film added with 10% aloe vera (CS-10%AV) had the best properties. The study also found that using CS-A10 film to preserve tomatoes helped maintain the fruit's colour and shape for longer while retaining vitamin C and acids for an extended period.

1 Introduction

Food technology is an essential industry for human life. The development of the food industry led to the rapid growth of the food packaging industry. The increasing demand for food consumption has enormously boosted the need for food packaging. Unfortunately, this increased waste from synthetic materials used to package and preserve food. These wastes significantly contribute to environmental pollution [1-2]. As a result, the development of ecologically friendly food packaging materials became a priority in addressing this issue [1,3]. One of the most promising solutions was using biodegradable films made from natural materials. These biofilms provided a viable alternative to non-biodegradable plastics and could help to reduce both the demand for conventional plastics and environmental pollution [4-6].

One crucial development in the food packaging industry was the use of biodegradable preservation films, primarily composed of biopolymers [7-8]. Numerous studies showed that biologically derived preservation films protected the food inside, prevented moisture loss, and helped maintain flavour and increase shelf life [1,9-11]. Some common biopolymers used in food packaging and preservation include polyvinyl alcohol, polyglycolic acid, polyethene terephthalate, polybutylene adipate terephthalate, starch, cellulose and cellulose

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derivatives, chitosan, etc. [1,12]. These materials were known to be excellent film-forming substances, environmentally friendly, biodegradable, reasonably priced, and readily available [1].

Chitosan is a natural polysaccharide extracted from crustaceans and insects' shells. It comprised monomers called 2-acetamido-2-deoxy-β-D-glucose with β(1→4) linkages [13-16]. This polymer was highly waterproof, antibacterial, antioxidant, biocompatible, non-toxic to animals and humans, and environmentally safe [13,17]. However, its antibacterial properties were limited, which affected its use in preserving fruits and vegetables. To overcome these limitations, it was necessary to combine chitosan with other materials that had high antibacterial and antioxidant properties to optimize its application. Aloe vera (AV) is a widely recognized herbal with high antibacterial properties [18]. It was a popular plant known for its diverse applications in food, pharmaceuticals, and cosmetics [18-19]. AV gel, obtained from its leaves, contains soft and smooth tissues with parenchyma cells that are highly viscous and jelly-like. The chemical composition of AV gel contains bioactive compounds such as carbohydrates, proteins, fibres, soluble sugars, vitamins, minerals, amino acids, organic acids, and phenolic compounds [18-19]. AV gel was used in the food industry to process functional foods as a natural preservative or raw material for edible films and coatings [19]. Recently, this material has received significant attention due to its ability to extend the shelf life of food items and its high barrier, antioxidant, and antibacterial properties [18]. Therefore, adding AV gel to chitosan film could help improve its antioxidant and antibacterial properties without affecting its biodegradability and safety.

Thus, this study aimed to investigate the possibility of acquiring a biofilm based on chitosan and AV gel. The properties evaluated include film-forming ability, water uptake, swelling ratio, solubility, biodegradation, and colour of the obtained films, along with the evaluation of their surface morphology. The best film was selected for applying to preserve ripe tomatoes after harvest.

2 Materials and methods

2.1 Materials

The main ingredients in this study include chitosan and AV gel. The chitosan (CS) was obtained from the Chitosan Vietnam Company in Vietnam and was a light yellow powder with a purity of 95% and 80% deacetylation. Additionally, other chemicals were used in the study, such as sodium hydroxide, phenolphthalein, acetic acid, glycerol, disodium hydrogen phosphate dodecahydrate buffer, monopotassium phosphate buffer, citric acid, hydrochloric acid, ammonium chloride, potassium iodide, sulfuric acid, soluble starch, potassium iodate, yeast extract, peptone, agar, and sodium chloride.

2.2 Methods

2.2.1 AV gel preparation

After purchasing, AV leaves were washed and drained to obtain AV gel, and the green leaves were removed with a knife and the white part inside. Puree the collected white piece with a blender for 10 minutes to form a gel-like solution. Store the resulting gel solution at 5ºC in the combined film collection section. To determine the pH of the obtained AV gel solution, use an Analytical handheld pH meter (Al 15, Taiwan). Use the acid-alkaline titration method to determine the total acid content in the obtained AV gel [1].

2.2.2 Fabrication of chitosan-AV gel film (CS-AV)

Chitosan was dissolved in a 1.5% (v/v) acetic acid solution at a 1/25 g/ml ratio. The mixture was stirred for 2 hours using a heating magnetic stirrer Velp (Arec. X, Italy). After the chitosan was completely dissolved, 0%, 5%, 10%, and 15% (w/v) of AV gel were added to the mixture and stirred continuously until the mixture was uniform. Next, 10% by mass of glycerol was slowly added, and the mixture was stirred for 10 minutes. The film-forming mixture was then sonicated for 10 minutes at 360W using an ultrasound machine, Ultrasonic SUS304 (Fuyang, China). The mixture was moulded onto an 11 cm diameter petri dish to form discs, which were then dried at room temperature until the film was dry and could be peeled from the mould [20-23].

2.2.3 Water uptake and swelling ratio

Samples were prepared with sizes of 2x2 cm and weighed. Place the prepared sample in the beaker containing water. After 5, 10, 20, 30, 40, 50, 60, and 90 minutes of immersion, take the sample out of the beaker and dry it with a dry towel/cotton/brush. The films were weighed and weighed [4]. The water uptake (W) and swelling ratio (S) of the film were calculated at the moment, and the Eq. (1) and (2):

$$W = \frac{m_2 - m_1}{m_1}.100 \quad (1)$$

$$S = \frac{S_1}{S_2}.100 \quad (2)$$

where, W – the water uptake of sample (%), m_1 – the initial mass of sample (g), m_2 – the mass of sample after imerising (g), S – the swelling ratio (%), S_1 – the area of sample before immersing (cm^2), S_2 – the area of sample after immersing (cm^2).

2.2.4 Solubility of film

The studied sample was placed in a beaker containing 100 ml of water so that the sample was completely submerged in water. After immersing for 24 hours, remove the film and dry it to a constant weight at 60ºC. The dried sample was weighed. The solubility of the sample (So) was calculated according to Eq. (3):

$$S_o = \frac{m_1 - m_2}{m_1}.100 \quad (3)$$

where, S_o – the solubility (%), m_1 – the initial mass of sample (g), m_2 – the mass of sample after immersing 24 hours (g).

2.2.5 Color

To begin with, prepare film samples that measure 2x2 cm. Next, utilize a handheld colourimeter Nippon Denshoku (NR-12A, Japan) to measure the colour of the film. Take note of the L*a*b values displayed on the instrument. Calculate the ΔE value, the square root of the sum of squares of the three recorded values from the colourimeter. Calculate the difference in E values between the original film and subsequent films to determine the degree of colour difference between the original film and subsequent films.

2.2.6 Biodegradability

The biodegradability of the research samples was investigated using the landfill method. The samples were prepared with the same size and weighed for initial mass. Then, the samples were buried about 5-7 cm deep in soil taken from the same location as the sample. After 1,

2, 5, 10, 20, 30, and up to 50 days, the soil was loosened, and the condition of the buried film was observed. If the sample was still intact, the film was removed from the soil, cleaned with a wet towel, dried, and weighed. The biodegradability of the film during the burial process was calculated using Eq. (4):

$$H = \frac{m_1 - m_2}{m_1}.100\% \quad (4)$$

where, H – the biodegradability of the sample (%), m_1 – the initial mass of sample (g), m_2 – the mass of sample after buring (g).

2.2.7 Antibacterial ability

To evaluate antibacterial ability, all instruments were sterilized using a 100L autoclave (KT100, Mitacom, Vietnam). Two strains of microorganisms selected for antibacterial research were E.Coli (Gram-positive) and St. aureus (Gram-negative). Microbial strains were cultured on Luria (LB) agar medium and autoclaved for 45 minutes at 121ºC. To culture microorganisms, 2 mL of solution containing bacteria concentrated between 105 and 106 CFU/mL was inoculated onto petri dishes containing LB agar. Wells with a diameter of 5 mm were made on agar plates. A micropipette introduced 100 µL of the prepared film-forming solutions into the agar wells. The locations of the agar wells containing the studied films were marked. Then, the cages were incubated for 24 hours at 37ºC. After 24 hours, measure the diameter of the antibacterial ring on the agar plates [24]. Antibacterial ability was calculated according to Eq. (5):

$$\Delta D = D - d \quad (5)$$

where, ΔD - diameter of the sterile ring (mm), D – diameter of the sterile zone after culturing (mm), d - agar well diameter (mm).

2.2.8 Morphology of the filmby optical microscopy

To observe the surface morphology of the film, a scanning electron microscope model (Optika, Italy) was used with 100-500 magnification.

2.2.9 Weight loss of fruit

The fruits were weighed before being covered with film and then stored at 28-30ºC for 12 days. The weight of the fruit was recorded every two days, and the natural weight loss was calculated by comparing the difference in weight with the initial weight before coating.

2.2.10 Total soluble solids content, vitamin C content, total acid content

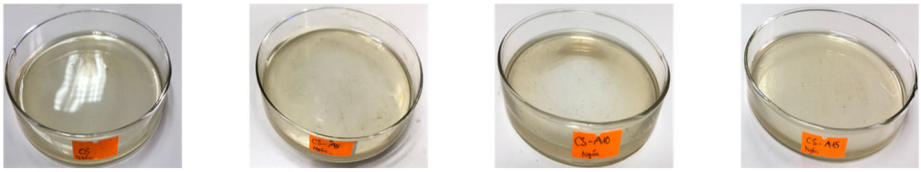
After storage for 3, 6, 9 and 12 days, fruit extracts were collected. The fruit extracts were measured for total soluble solids content by refractometer Brix (Master-T, Atago, Japan) [25], vitamin C content by iodometric titration [26], and total acid content by using the acid-alkaline titration method [20].

3 Results and discussion

3.1 Formation of CS-AV film

Chitosan was only soluble in acidic conditions; therefore, it was necessary to investigate the AV gel material's pH and acid content for materials added to chitosan films. Measuring the

obtained AV gel's pH and acid content showed that the AV gel's total acid content was only 0.135%, and the pH of the gel solution was 6.3. This indicated that additional acid was required to dissolve chitosan easily. Investigating the properties of AV gel, a film-forming formula from two components, chitosan, and AV gel, is proposed in section 2.2.2. Visual images of the films are shown in Figure 1.



(a) Chitosan (CS) film (b) CS- 5% AV (CS-A5) (c) CS - 10% AV (CS-A10) (d) CS - 15% AV (CS-A15)

Fig. 1. The film from CS with the various content of AV.

The data presented in Figure 1 indicates that adding AV gel to the film-forming mixture results in small black spots on the film as the dose increases. This result revealed that the gel collection process did not produce the minor components, leading to low uniformity of the collected AV gel. Therefore, tiny particles appeared on the composite film when AV gel was added to the CS film. However, these particles were not too obvious, so the aesthetics of the hybrid films were not significantly affected.

Furthermore, upon examining the film's surface, it was observed that the chitosan-AV - AV gel films retained their smoothness and did not have any cracks or air bubbles. Additionally, the composite film was easy to collect after drying without losing weight, tearing, or cracking. During the moulding process on the plate, there was no layering phenomenon, and the solution was relatively homogeneous.

The film's colour was also evaluated, and it was discovered that the addition of AV gel did not significantly alter the colour of the resulting film. This is because the AV gel viscosity was white, and when added to the chitosan film, the colour of the resulting film remained almost unchanged.

In conclusion, the initial results suggest that films can be successfully created from chitosan and AV gel by adding glycerol as a plasticizer.

3.2 Some propeties of the CS-AV film

The physical properties of the film are one of the essential characteristics of food packaging because the film acts as a barrier that separates the food from external factors like air, humidity, and microorganisms, thus protecting it. Hence, it becomes essential to evaluate parameters like water uptake, swelling ratio, and solubility of the films to ensure their effectiveness in keeping the food safe and fresh.

3.2.1 Water uptake

Protecting food, especially fruit, was usually done in a moist environment. Therefore, the water uptake of the film must be ensured so that the film does not absorb too much water. A moderate amount of water absorption is preferable as it helps keep the film moist enough to protect the fruit. However, if the film absorbs too much water, it will take water from the fruit, causing it to wilt faster. Therefore, it was necessary to investigate the water uptake of the developed films.

According to the results (shown in Figure 2), the water uptake of the CS film reached equilibrium after 60 minutes. The films that were supplemented with AV gel achieved water

uptake balance sooner - expressly, 30 minutes for the film CS containing 5% AV and 40 minutes for the film CS containing 10% and 15% AV. Therefore, adding AV gel to the chitosan film helped it achieve water uptake balance faster than without AV.

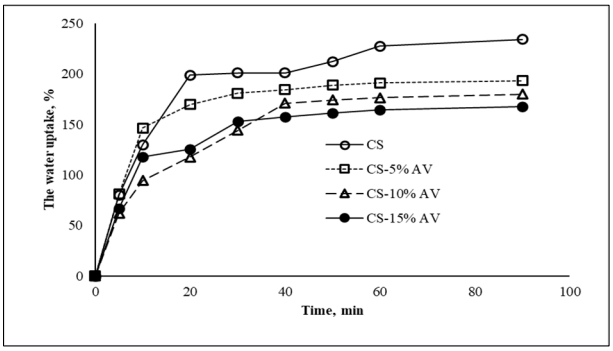


Fig. 2. Wate uptake of studied films over time.

It was observed that the water absorption of the films with AV gel added to them was significantly lower than those without AV gel. As per the ascending order of water absorption, the films CS-10% AV, CS-15% AV, CS-5% AV, and film CS were arranged. This indicates that the water uptake of films with added AV gel was significantly improved compared to those without AV gel.

3.2.2 Swelling ratio

The higher the swelling property of the material, the larger the occupied area. Therefore, finding a suitable way to arrange food using film for packaging was necessary. The swelling ratio of the biofilm is shown in Figure 3.

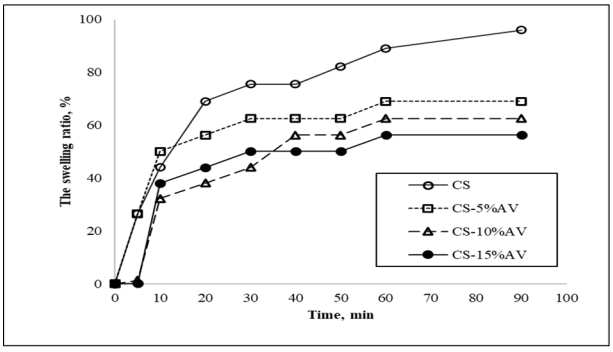


Fig. 3. The swelling ratio of studied films.

According to the results, the films that contained AV gel had less swelling ability compared to the CS film without AV. The swelling of films in the order of increasing from CS-15% AV and CS-10% AV were less than the swelling of CS-5% AV and CS. Two films that contained 10% and 15% AV had swelling properties that changed over time. Within the first 40 minutes, the swelling property of the film containing 10% AV was lesser, but after 40 minutes, the swelling property of this film increased and became higher than that of the film CS-15% AV. The results of Figure 3 also showed that the film CS with 10% and 15%

AV had a more suitable swelling ratio than the film containing 5% AV and pure chitosan films.

3.2.3 Solibility

The solubility of films is a crucial aspect to consider. Generally, the higher the solubility, the lower the quality of the film. This is especially problematic for packaging products that contain a lot of moisture, as it becomes difficult to apply the film. Therefore, it was essential to investigate the solubility of the studied film. The solubility of the film is presented in Figure 4.

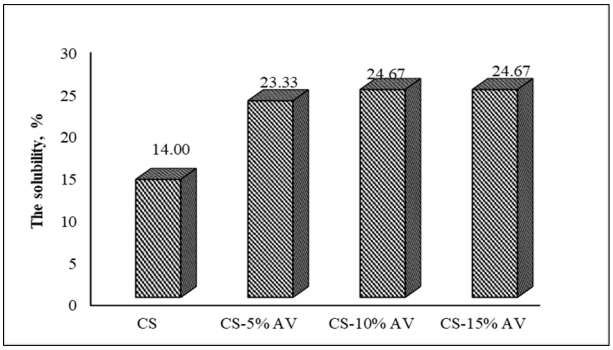


Fig. 4. Solubilyty of the studied films.

Based on the results presented in Figure 4, it was observed that the solubility of CS film was lower than those of the films CS-5% AV, CS-10% AV, and CS-15% AV. This means that adding AV gel increases the solubility of the film. However, there was no significant difference in solubility between the films supplemented with 10% and 15% AV. Notably, the solubility of the studied films only reached 24.67%, which is relatively low. Nonetheless, this level of solubility is suitable for food packaging applications.

3.2.4 Color

The color of the film had a significant impact on the sensory properties of the product. Therefore, it was necessary to survey to propose suitable application areas.

Table 1. Color comparison results of the studied films.

Sample	CS	CS-5% AV	CS-10% AV	CS-15% AV
ΔE	105.90	104.,28	111.84	92.69
Comparison with original film	-	1.62	5.94	13.21

Table 1 demonstrates that the increase in AV content resulted in a greater degree of colour variation when compared to the original film (the CS film). Notably, the sample containing 15% AV by weight exhibited the most significant colour difference compared to the other samples. AV comprises both colouring compounds and tannins. During the drying process of the film, the natural colouring compounds became more active due to the impact of temperature, and the tannins were transformed into grey-brown fibres or particles. Dissolving chitosan and AV did not form a homogeneous mixture, causing the colouring molecules to form distinct coloured "stains" on the film. This, in turn, led to an increased variation in the colour of the films that were supplemented with high AV content.

3.2.5 Biodegradability

The biodegradability of the film was a significant property for self-destructing biofilms. To evaluate the biodegradation properties, the landfilling method was used. The results indicated that the films without AV had the lowest biodegradability, while those containing AV had the highest. Moreover, the decomposition ability increased as the AV content increased. After 50 days of burial, the biodegradability of the films was 46.67% (film CS), 60.67% (films CS-5% AV and CS-10% AV), and 80% (films CS-15% AV), respectively. These results demonstrated that adding AV to chitosan accelerates the biodegradation process.

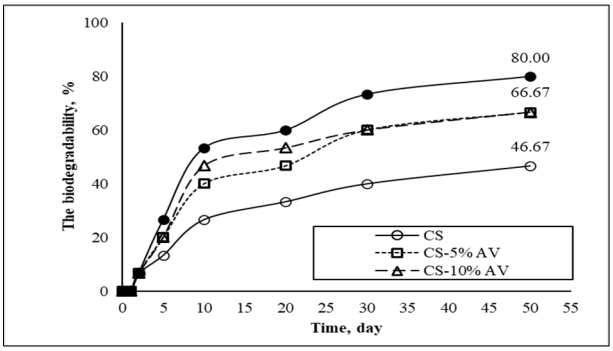


Fig. 5. The biodegradability of the studied film.

3.2.6 Antibacterial activity

Microorganisms are one of the main reasons for food spoilage, especially fresh fruit. To improve the preservation process, it is essential to use films that have antibacterial properties. Therefore, we prioritize preservation films that possess such properties. In our research, we used two microorganism strains, Escherichia Coli and Staphylococcus aureus, to test the antibacterial ability of the studied film. Figure 6 and Table 2.

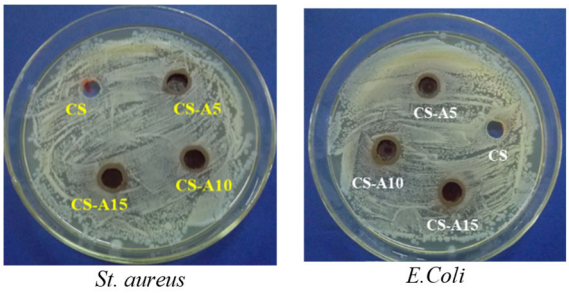


Fig. 6. Antibacterial activity *St. aureus* and *E. Coli*.

Table 2. Antibacterial activity of the samples.

Samples	**Antibacterial diameter, mm**	
	E. Coli	***St. aureus***
CS	0.00	0.00
CS-5% AV	0.89	1.58
CS-10% AV	1.67	2.07
CS-15% AV	1.78	2.13

The results showed that the antibacterial activity of the film increased when AV was added to chitosan, compared to the original film. The more AV added to the film, the greater its antibacterial ability became. This is because AV possesses an antibacterial ability that improves the overall antibacterial ability of the film. However, adding AV to the film up to 15% did not significantly increase its antibacterial ability. Therefore, we recommend using the film CS that contains 10% AV as the most reasonable option.

3.2.7 Mophology

An optical microscope was used to examine the surface morphology of the films studied, as shown in Figure 7. The results indicated that as the AV content increased, the "AV fibres" on the film became more pronounced. Additionally, the results demonstrated that the film CS, which contained 15% AV, had a more noticeable colour difference when compared to the pure film. In Figure 7d, brown spots were clearly visible when the AV content was increased to 15%.

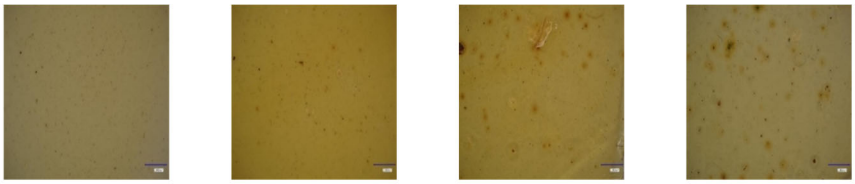


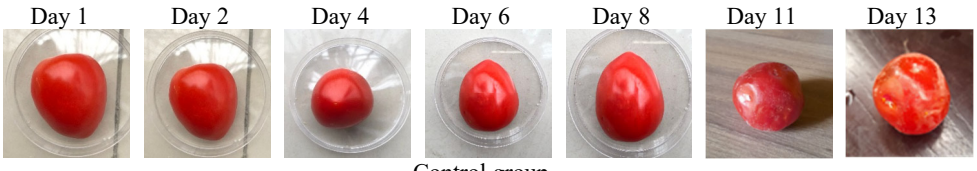
Fig. 7. Optical microscopy results of the studied films (magnification x100).

From the properties of the studied films, it was found that the film CS containing 10% AV had the lowest water uptake compared to the remaining films; the swelling ratio was commensurate with the film CS containing 15% AV but lower than the film CS and film CS-5% AV, the solubility was not too high compared to the original film but was on par with the other films, the colour was not significantly different from the pure film CS, good biodegradability, reaching 60.67%. After 50 days of burial, the antibacterial ability improved compared to the film chitosan and was commensurate with the remaining films. Thus, it was clear that the film CS containing 10% AV was superior to the other films. Therefore, film CS-10% Av was chosen to study the experimental application for fresh fruit preservation.

3.3 Presevention tomato by the CS-AV film

3.3.1 External morphology of the fruit

To assess the effectiveness of the film, both internal and external parameters were analyzed. The color and appearance of tomatoes were observed using film CS-AV and control tomatoes (without any preservation method). Figure 8 and Table 3.



Group using the film CS-AV

Fig. 8. External morphology of the tomatoes according to storage time.

Table 3. Appearance of tomatoes according to storage time.

Sample	Group using the film CS-AV	Control group
Before storage	The fruit was bright red and ripens evenly. Good gloss. No signs of damage. No wrinkles.	
After 1 day	The fruit was bright red, does not change ripeness. Good gloss. No signs of damage. No wrinkles.	The fruit was bright red, does not change ripeness. Good gloss. No signs of damage. No wrinkles.
After 3 days	The fruit was bright red, does not change ripeness. Good gloss. No signs of damage. No wrinkles.	The fruit was bright red, slightly soft. Good gloss. No signs of damage. No wrinkles.
After 5 days	The fruit was bright red, the hardness of the fruit decreased. Good gloss. No signs of damage. No wrinkles.	The fruit was bright red, slightly soft. Good gloss. No signs of damage. No wrinkles.
After 7 days	The fruit was bright red, the hardness of the fruit decreased. Good gloss. No signs of damage. No wrinkles.	The fruit was bright red, slightly soft. Good gloss. No signs of damage. Lightly wrinkle the fruit skin.
After 10 days	The fruit was bright red, the hardness of the fruit decreased. Good gloss. No signs of damage. Slightly wrinkled.	The fruit was bright red, very soft. Good gloss. There is slight waterlogging, and a few black spots on the fruit. Lightly wrinkle the fruit skin.
After 12 days	The fruit was bright red, very soft. Good gloss. No signs of damage. Slightly wrinkled.	The fruit was bright red, hơi mềm. Good gloss. There is slight waterlogging, and a few black spots on the fruit. Slightly wrinkled.

It was observed that the colour of fruits without film preservation was brighter red than those preserved with film. Although both gloss was similar, the unpreserved fruit became softer after 12 days, and black spots appeared on them. In general, using film CS-AV improved the external state of tomatoes after 12 days of storage.

3.3.2 The loss of vitamin C content of fruit

Fruits are rich in vitamin C, but this nutrient gradually decreases over time, along with other nutrients, which causes a reduction in fruit quality. The research depicted in Figure 9a shows that the vitamin C content of fruits declines gradually with time. Both stored samples showed an increase in vitamin C loss. However, the film CS-AV preserved fruits' vitamin C content better than unpreserved fruits for up to 10 days. After 12 days of storage, both samples showed similar levels of vitamin C decline.

3.3.3 Weight loss of fruit

Figure 9b displays the natural weight loss of fruit during storage. The data reveals that the natural weight loss of both tomato samples increased with a longer storage period. During the first nine days of storage, fruit preserved with film CS-AV experienced lower weight loss than the unpreserved fruit. However, after nine days of storage, both fruit samples had a

similar weight loss. The weight loss of fruit during storage can be attributed to respiration and water evaporation. Film CS-AV is believed to be effective in creating a protective layer on the fruit surface, thereby retaining the water inside the fruit and reducing rapid loss.

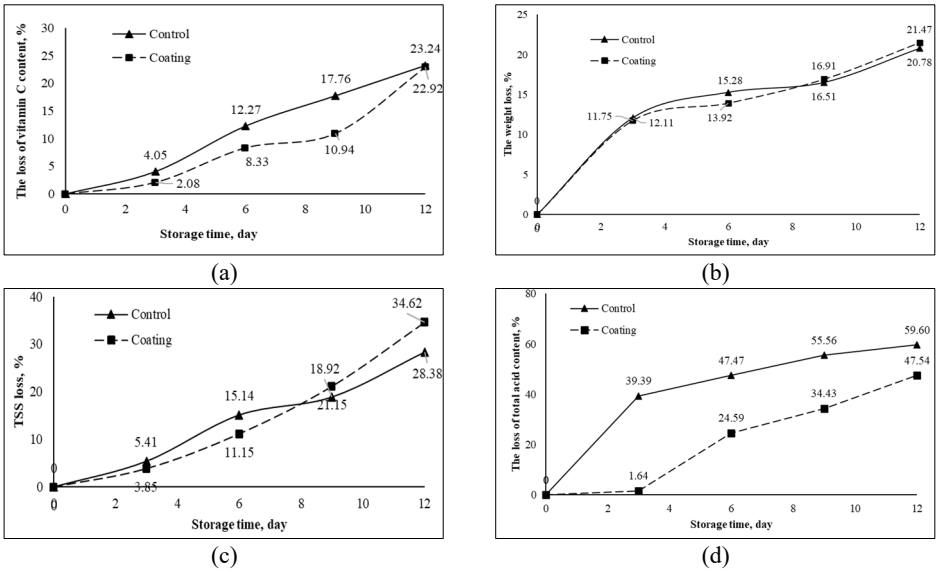


Fig. 9. The decrease in vitamin C content (a), the weight loss (b), the decrease in TSS (c), the loss of total acid content (d) of the fruit according to storage time.

3.3.4 TSS of fruit

Figure 9c shows the decline in TSS (total soluble solids) of tomatoes with storage time. The longer the tomatoes are stored, the greater their TSS loss. The study found that within nine days of storage, the TSS loss of unpreserved tomatoes was higher than that of tomatoes preserved with film CS-AV. However, when stored for more than nine days, this trend was reversed. This phenomenon explains the wrinkling of the tomato skin, which is detailed in Table 3. During storage, the dehydration process and the resulting change in TSS content caused the skin phenomenon. This was because the unpreserved tomatoes lacked an outer protective layer, making water evaporate and the fruit's respiration process faster. This led to the fruit losing moisture more quickly, with a large amount of dry matter lost due to respiration, causing the fruit to wrinkle. In contrast, the tomatoes preserved with film CS-AV were protected by an outer coating that hindered water evaporation and fruit respiration.

3.3.5 Total acide content of fruit

As per Figure 9d, ripe tomatoes preserved with the film CS-AV retain their total acid content better than unpreserved ones. The results showed that the film CS-AV helps maintain the fruit's colour and gloss and enables it to remain firm for longer periods. It also reduces the decrease in vitamin C and acid content over time during storage.

4 Conclusion

The films chitosan supplemented with AV at contents ranging from 0, 5, 10 and 15% have been studied for different properties. When adding AV to chitosan, the film formation ability

was relatively good. When observed with a normal probe, the film was more turbid than the original film. Different amounts of AV added to the film formulation affect some film properties. Among them, the film CS-10% AV had many superior properties compared to the remaining films, such as the high water uptake, swelling ratio, and solubility. Experimental application of the film CS-10% AV in preserving fresh tomatoes showed that the film CS-10% AV was effective in retaining the colour, gloss and hardness of the fruit for 12 days; the vitamin C and total acid content in the fruit has been significantly improved; the decrease in total soluble solids and weight loss of fruit was commensurate with control fruit.

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