2022

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Recommended Citation

Sharma, S., Perera K.Y. & Dileswar, P. (2022). Active Packaging Film Based on Poly Lactide-Poly (Butylene Adipate-Co-Terephthalate) Blends Incorporated with Tannic Acid and Gallic Acid for the Prolonged Shelf Life of Cherry Tomato. *Coatings 2022, vol. 12, no. 12, pg. 1902. doi:10.3390/coatings12121902*

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Article

Active Packaging Film Based on Poly Lactide-Poly (Butylene Adipate-Co-Terephthalate) Blends Incorporated with Tannic Acid and Gallic Acid for the Prolonged Shelf Life of Cherry Tomato

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Abstract: The antimicrobial property is the key feature of active packaging. Biological macromolecules such as tannic and gallic acids are naturally found in plants such as tea, fruits, berries, and grapes. The incorporation of tannic acid (TA) and gallic acid (GA) in the biodegradable polymer blend Poly Lactide-Poly (Butylene Adipate-Co-Terephthalate) (PLA-PBAT) was used in this study to assess the potential of active packaging. TA and GA (10 wt%) composite films showed a 65%–66% increase in the UV barrier property. The tensile strength value increased after the incorporation of TA and GA (10 wt%), respectively. Overall, 1.67 and 2.2 log reductions in *E. coli* and *L. monocytogenes* growth were observed, respectively, in the presence of TA (10 wt%) composite film. In addition, TA composite film was able to maintain and enhance the quality of cherry tomatoes for up to 20 days of storage at room temperature. For cherry tomatoes packed in PLA-PBAT-TA10 (wt%), TSS decreased by 6.3%, pH was 4.3, and a microbial reduction of 2.70 log CFU/mL was observed. In conclusion, TA composite film had confirmed significant UV blocking properties, surface hydrophobicity, and antibacterial properties, which show its potential as an active packaging film.

Keywords: active packaging; antimicrobial; Poly Lactic Acid; Poly (butylene adipate-co-terephthalate); polyphenols; cherry tomato

Citation: Sharma, S.; Perera, K.Y.; Pradhan, D.; Duffy, B.; Jaiswal, A.K.; Jaiswal, S. Active Packaging Film Based on Poly Lactide-Poly (Butylene Adipate-Co-Terephthalate) Blends Incorporated with Tannic Acid and Gallic Acid for the Prolonged Shelf Life of Cherry Tomato. *Coatings* **2022**, *12*, 1902. https://doi.org/10.3390/coatings12121902

Academic Editor: Raquel Sendón

Received: 24 October 2022
Accepted: 30 November 2022
Published: 5 December 2022

1. Introduction

One of the innovative approaches to food packaging that has been developed in response to current shifts in market trends and consumer demands is active packaging. The term "active packaging" refers to a type of packaging that alters its physical state to improve its safety, sensory, or shelf-life characteristics while preserving the food's quality. Antioxidants and/or antimicrobial agents must be loaded and embedded into the polymer material for it to act as an active packaging material [1]. As a result, to formulate active packaging material, effective loading and functional retention of bioactive substances are prerequisites. The requirement for active packaging systems, which are made up of environmentally friendly polymers and natural active agents, is increasing rapidly. These systems minimize the detrimental effects of food packaging on the environment and enhance consumer safety [2].

In recent years, there has been interest in films made from sustainable, abundant, cheap, and biodegradable natural polymers. Biopolymer films are made from bio-based resources like protein, polysaccharides, and microbial polyesters that are extracted from plants and marine organisms or produced by the fermentation of microorganisms. Biodegradable polymers such as Poly Lactic Acid (PLA), Poly-hydroxy-butyrate (PHB), etc., are made from natural and fossil resources, break down, and return to nature in a short period through biodegradation by microorganisms [3]. The environment-friendly and antimicrobial activity of the packaging material are among the key features of active packaging. Various chemicals are used as antimicrobial agents in food packaging, but the leaching of chemicals from the polymer into the food is a big concern for health and safety. Therefore, the research is now shifting towards natural antimicrobial agents such as polyphenols, essential oils, etc., which will be able to meet all the safety parameters without affecting human health. Creating a film that is environment-friendly and carries macromolecules of natural origin that have known health benefits–such as antioxidant and antibacterial activities–could make a huge difference to the future of the food packaging industry [4].

There is an innovative approach in active packaging where phenolic compounds could be used to extend the shelf life of food and products. Polyphenols such as tannic acid, gallic acid, rutin, quercetin, catechin, synergic acid, resveratrol, ellagic acid, caffeic acid, and apigenin are naturally found in plants such as tea, fruits, wine, vegetables, etc. [5]. For example, gallic acid and its derivatives are present in almost every part of the plant, such as the bark, wood, leaf, fruit, root, and seed, and are widely distributed throughout the plant kingdom. Similarly, tannic acid is present in berries, grapes, persimmons, gallnuts, and pomegranates [6]. In addition, these phenolic molecules possess antimicrobial properties as phenolics can retard the invasion of microbes in some products and avoid the decomposition of fruits and vegetables [5,7]. Therefore, these molecules could be encouraged to be incorporated into the food packaging to extend the shelf life of some foods [8]. Halim et al. [9] incorporated tannic acid (TA) into chitosan, gelatin, and methylcellulose films for food packaging and discovered that the biopolymer-tannic acid films exhibit key characteristics as an environmentally friendly film for fruit preservation. Feng et al. [10] developed a starch film incorporating tea polyphenols. According to their results, starch-tea polyphenol film can be applied as an inner film to prolong the shelf life of food items.

This study aimed to formulate a composite film using PLA/PBAT (poly (lactide)/poly (butylene adipate-co-terephthalate) polymers in combination with tannic acid (TA) and gallic acid (GA). The effect of TA and GA incorporation on composite film was observed by studying its structural, morphological, optical, mechanical, and antibacterial properties. In addition, the effect of the developed composite film on the quality of cherry tomatoes was evaluated and the weight loss, color difference, pH, total soluble solids content (TSS), and total bacterial count were measured at regular intervals for up to 20 days. To the best of the author's knowledge, this is the first study to develop a composite film (PLA-PBAT) incorporating tannic acid and gallic acid and to study its effect on the shelf life of cherry tomatoes to investigate its potential as an active food packaging film.

2. Materials and Methods

2.1. Materials

The pure form of polyphenolic compounds (tannic acid, gallic acid) was purchased from Sigma Aldrich, Gillingham, UK. Chloroform was purchased from Sigma Aldrich (Wicklow, Ireland) and ethanol was obtained from Merck KGaA (Darmstadt, Germany). Polymer resins of Poly (lactide) (PLA, Synterra BF 2004, Belfeld, The Netherlands; average molecular weight of 200 kDa) and PBAT (Ecoworld PBAT003; m.p. 110–120 °C, the density of 1.26 g/cm^3) were obtained from Helian Polymer (Belfeld, The Netherlands). Tryptic soy broth (TSB) and agar powder were procured from Sigma Aldrich, Ireland.

2.2. Preparation of Composite Films with Efficient Polyphenols

The solvent casting method was used to create PLA-PBAT (Poly (lactide)/Poly (butylene adipate-co-terephthalate) composite films with the selected polyphenols tannic acid (TA) and gallic acid (GA). To form the composite film, polymer resins were mixed in a 98:2 ratio of PLA and PBAT [11]. Tannic acid (TA) (MW: 1701.20 g/mol, MP: >218 °C) and gallic acid (GA) (MW: 170.12 g/mol, MP: 251 °C) were dissolved in an absolute ethanol solution at a concentration of 100 mg/mL. Various concentrations of polyphenol solution (0.4 mL, 2.0 mL, and 4 mL which are equivalent to 1%, 5%, and 10% of the wt% of polymer resin) were added to 100 mL of chloroform [12]. To make up the equal volume, ethanol was added to the samples accordingly. In the prepared polyphenol (TA and GA) solution, PLA (3.92 g), PBAT (0.08 g), and glycerol (50 wt%) were added and dissolved by continuous stirring at room temperature for 18 h [13]. PLA-PBAT control composite solution was prepared by following the same procedure by adding 4.0 mL of ethanol (without polyphenol). The prepared solution was applied to a Teflon-coated glass plate (24 cm 30 cm) and formed a homogeneous thin film. After 24 h, the films were dried at room temperature and removed from the glass plate. Furthermore, the films were conditioned for at least 48 h at 50% relative humidity (RH) and 25 °C temperature [14]. The prepared films were labelled as PLA-PBAT, PLA-PBAT_TA1%, PLA-PBAT_TA5%, PLA-PBAT_TA10%, PLA-PBAT_GA1%, PLA-PBAT_GA5%, and PLA-PBAT_GA10%.

2.3. Film Characterization

2.3.1. Morphological Observation

The microstructural morphology of the film surface was analyzed using scanning electron microscopy (SEM) (Hitachi SU70, Hitachi Ltd., Tokyo, Japan) as well as the film's cross-sectional area. A small piece of prepared film was mounted on the sample holder of the SEM to examine the surface morphology. Analysis of the image was done with a 5-kV accelerating voltage.

2.3.2. Surface Colour and Optical Properties

The surface colour of the composite films was tested using an XE spectrophotometer (ColorQuest XE, Hunter Lab, Northants, UK) using a white colour plate as a standard background. The value of reflectance was measured for L (+lightness, −darkness), a (+red, −green) and b (+yellow, −blue) as the parameters of colour. The background values for the standard white plate were set as L = 97.75, a = −0.42, and b = 1.83. For each film sample, three readings of Hunter colour values (L, a, and b) were taken, and the averages were determined. The total colour difference (ΔE) was calculated using the following formula.

$$\Delta E = [(\Delta L)^2 + (\Delta a)^2 + (\Delta b)^2]^{1/2} \tag{1}$$

where ΔL, Δa, and Δb are the differences between the standard colour value of each colour plate and sample film [11,12].

The optical properties of the films were measured using the light transmittance spectra of a UV-Vis spectrophotometer (Agilent technologies, Cork, Ireland). Each sample was cut into a rectangular piece of film (3 cm × 7 cm) and clamped between the two magnetic cells of the spectrophotometer. For each film sample, three readings were observed. The UV barrier property of the film samples was evaluated by measuring the percent transmittance at 280 nm (T_{280}). To determine the transparency of the film samples, the percent transmittance was measured at 600 nm (T_{600}). This measured the amount of light transmittance in the film while investigating the transparency values using the UV -Vis spectrophotometer.

2.3.3. Thickness and Mechanical Properties

The thickness of each sample film was measured using a digital micrometer (Mitutoyo, Takatsu-ku, Japan) with a precision of 0.001 mm. The thickness of each sample film was evaluated at five random positions. Mechanical properties such as tensile strength (TS), elastic modulus (EM), and elongation at break (EB) for each sample film were measured using the standard ASTM D 882–88 method. Rectangular strips (3 cm × 15 cm) for each film sample were cut and mounted in the Instron Universal Testing (Model 5565, Instron Engineering Corporation, Canton, MA, USA). The intron instrument was operated at

50 mm of grip length and a 50 mm/min crosshead speed using a 500 N load cell. TS was expressed in MPa. The following formula was used to define the tensile strength:

$$TS = F/A \tag{2}$$

where F refers to the maximum force (N) that is needed to pull the film apart and A is the initial cross-sectional area of the film sample (m^2).

The tensile elongation at break (EB) is the measure of the ratio between the length increased at the breakage of film and the initial length of the film samples. The EB was calculated as follows:

$$EB\ (\%) = [(X_f - X_o)/X_o] \times 100 \tag{3}$$

where the initial grip separation (50 mm) is represented by X_o and the elongation of the film at the failure is represented by X_f [12].

The elastic modulus (EM) determines the resistance of the film to elastic deformation. The EM could be simply defined by the ratio between stress and strain. Five measurements were carried out for each film, and the mean values were considered.

2.3.4. Thermal Stability

The thermal stability of the film was determined using a thermogravimetric analyser (TGA) (Shimadzu, DTG60, Kyoto, Japan). Each film sample weighing around 9 mg was scanned at a temperature ranging from 30 to 500 °C under a nitrogen flow of 50 cm^3/min at a heating rate of 10 °C/min. The TGA curve was formed and the weight loss (%) and the maximum decomposition temperature (T_{max}) of films were measured.

2.3.5. Surface Hydrophobicity

The surface hydrophobicity was determined by the water contact angle (WCA) using the WCA analyser (FTA-200 systems, FTA Europe, Cambridge, UK). Each film sample was cut into rectangular strips (3 cm × 8 cm) and placed on a stainless-steel stage with the water contact angle analyzer attached. Using a microsyringe, a droplet of water (~10 µL) was placed on the surface of the film samples. The contact angle was then measured on each side of the droplet of water. A high-speed camera recorded the formation of the drop, and the image was processed by computer. Each sample was evaluated in triplicate and the average value was determined.

2.3.6. Water Vapour Permeability

The water vapor permeability (WVP) of the films was determined gravimetrically. First, 10 g of oven-dried calcium chloride ($CaCl_2$) was placed in a circular container (diameter of 30 mm). The top of the container was covered with the tested films (n = 3). The containers with calcium chloride were left without covers as control samples. Cups were placed in a container at a temperature of 25 °C and 97% relative humidity. The initial weight of the sample was determined, and its weight was measured at fixed intervals (12 h) for four days. The amount of water vapor that penetrated through the film was determined by the increased weight of the circular dish. The WVP was calculated as follows:

$$\mathrm{WVP}\ [\mathrm{g}/(\mathrm{m}{\cdot}\mathrm{h}{\cdot}\mathrm{Pa})] = \frac{m \times d}{A \times t \times P} \tag{4}$$

where m is the weight increment in grams, d is the thickness of the film in meters, A is the exposed area in meter square, t is the permeation time lag in hours, and P is the water vapour partial pressure difference across the film in pascal [15,16].

2.3.7. Antibacterial Activity

The viable colony count method was used to determine the antibacterial property of the composite film against foodborne pathogenic bacteria, *L. monocytogenes* (NCTC 7973) and *Escherichia coli* (NCTC 9001) [12,14]. Each sample film (100 mg) was taken and sterilised under UV light for 20 min. *L. monocytogenes* and *E. coli* were inoculated aseptically in tryptic soy broth (TSB) and incubated for 18 ± 2 h at 37 °C. The optical density of the overnight culture was adjusted to 0.5 McFarland turbidity standard (equivalent to 1.5×10^8 colony-forming units (CFU)/mL) using a Densimat photometer (BioMérieux, Craponne, France). To obtain a working bacterial concentration of 10^6 CFU/mL, 100 µL of diluted inoculum was transferred aseptically to 10 mL of TSB. In the inoculated broth, each film sample was added and incubated at 200 rpm at 37 °C for 15 h. From the broth culture, samples were taken every three hours, diluted appropriately, and plated on agar plates for viable cell count determination. For each film sample, a test was performed in three replicates.

2.4. Effect of Developed Composite Films on Cherry Tomato Quality

The cherry tomatoes were purchased from a local supermarket. For this investigation, tomatoes with a good appearance, physical integrity, and no visible mold were chosen. Cherry tomatoes were washed and dried with clean tissue paper after being washed with tap water [17]. Tomatoes were sealed in the composite films and kept at room temperature. A few small manual perforations have been made in the packaging film to match the market packaging of the cherry tomatoes and facilitate the smooth flow of gas inside the packaging. The packet from the market is also kept as the control, named 'control market'. The tomato visual appearance, weight loss, colour difference, pH, total soluble solids content (TSS), and total bacterial count were measured before and during storage at room temperature at regular intervals up to 20 days of storage (0, 3, 6, 9, 12, 15, and 20 days).

Weight loss during the storage was measured by taking the difference between the weight of the sample at a specific time (W_t) and the weight of the samples initially (W_o); it could be calculated as:

$$\text{Weight loss } \% = [(W_o - W_t)/W_o] \times 100 \quad (5)$$

Hydrolysis of insoluble polysaccharides results in simple sugars enhancing the TSS of fruits during ripening and senescence, a TSS value similar to that of the control samples indicates better shelf-life preservation. The Brix Index (°Brix) was used to determine the total soluble solids (TSS) content. Tomatoes were crushed, and a homogenized aliquot was measured directly with a refractometer (Hand-Held Refractometer, Atago Co. Ltd., Saitama, Japan) and expressed in °Brix scale. Tomatoes were crushed in a blender for the pH assays. The pH of the samples was evaluated by direct immersion of the electrode of a digital pH meter (Eutech pH 700 Meter, Lennox, Dublin, Ireland).

2.5. Total Bacterial Count

The total bacterial count on cherry tomatoes was evaluated according to ISO 6887-1: 1999 [18]. A sample of tomato was aseptically transferred from the packaging to a stomacher bag containing 20 mL of MRD and homogenized manually for 2 min. For the following assay, 10-fold serial dilutions were made and from each dilution, and 0.1 mL of sample was inoculated onto the plate count agar. The plates were incubated at 37 °C for 24 h and the colonies were counted. The total bacterial count was studied on 0, 3, 6, 9, 12, 15, and 20 days of storage. The result was analysed by calculating log values and determining the log reduction with respect to 0 h bacterial count. The analysis was performed in triplicates.

2.6. Statistical Analysis

All the studies on the film's samples were determined in triplicates. A study of significant difference by the analysis of variance (ANOVA) and multiple comparisons (Fischer's least significant difference test) were studied using STATGRAPHICS Centurion XV software (Stat Point Technologies Inc. Warrenton, VA, USA). For the significant value, $p < 0.05$ was considered. All results are expressed as mean ± standard deviation.

3. Results and Discussion

3.1. Characterisation of Blend Films

3.1.1. Morphology of the Surface

SEM was used to study the surface morphology of the films incorporated with the TA and GA. As shown in Figure 1, the control film showed a homogeneous structure. The PLA-PBAT film with TA had shown a small, agglomerated structure on the PLA-PBAT matrix. PLA/PBAT-TA1% had shown less agglomeration, whereas as the concentration of TA increased to 5 wt% and 10 wt%, the agglomerated structures also increased. Huang et al. [19] also incorporated tannic acid (>5%) in gelatin/zein films and observed that precipitation in the composite solution forms a non-uniform film. In addition, Halim et al. [9] observed that the chitosan biopolymer incorporated with TA reduced the unevenness of the film and exhibited a smooth surface texture.

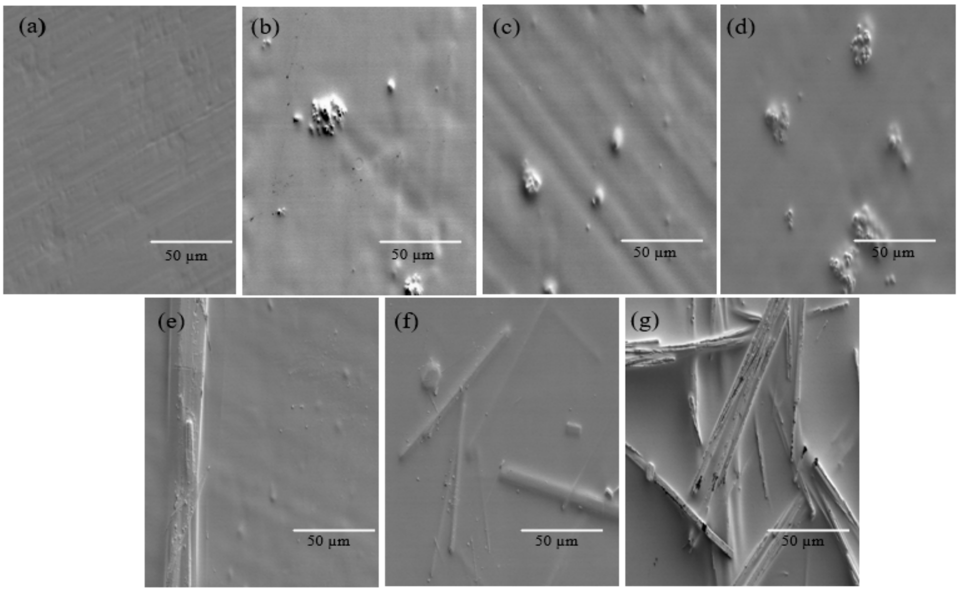

Figure 1. SEM micrographs of the composite film surface (**a**) PLA/PBAT (**b**) PLA/PBAT-TA1%, (**c**) PLA/PBAT-TA5%, (**d**) PLA/PBAT-TA10%, (**e**) PLA/PBAT-GA1%, (**f**) PLA/PBAT-GA5%, (**g**) PLA/PBAT-GA10%.

GA-incorporated PLA-PBAT film showed a rough surface. A long rough agglomeration was observed in the matrix which becomes denser as the concentration of GA in the film increases. In a study, Ahn et al. [20] incorporated gallic acid into low-density polyethylene film and observed an increase in irregularities. Their results also observed that gallic acid was unable to release during film formation [20]. Further, studies by Goudar et al. [21] on Poly (vinyl alcohol)/Tragacanth polymers incorporated with GA also agree with the results of the current study. The incorporation of GA causes major changes to the morphology of the films. In the lower concentrations of GA (5% to 15%), the films were continuous and homogenous. However, in higher concentrations of GA, the films showed heterogeneity and white spots. Due to the enhanced covalent and noncovalent interactions between matrix and dopant, surface unevenness increased with microscopic agglomerations of GA as the concentration increased [21].

3.1.2. Surface Colour

The control film, PLA/PBAT-TA1%, and PLA/PBAT-GA1%, were visually smooth, homogeneous, and transparent while as the concentration of TA and GA increased in the composite film, the film turned out to be rough and translucent. GA composite films were more translucent with a whitish colour than TA composite films. The surface colour and the optical properties of the TA and GA composite film are shown in Table 1. Lightness

(L-value) of the composite films had no significant difference ($p < 0.05$). The greenness (hunter a-value) and yellowness (hunter b-value) of the composite films incorporated with TA and GA had increased significantly.

Table 1. Surface colour, UV barrier, and transparency of composite films incorporated with tannic acid and gallic acid.

Film	L	A	B	ΔE	UV Barrier at $T_{(280)}$	Transparency at $T_{(600)}$
PLA/PBAT	93.06 ± 0.01^{a}	-0.31 ± 0.01^{d}	2.34 ± 0.01^{a}	4.38 ± 0.03^{c}	2.33 ± 0.03^{d}	73.38 ± 0.15^{c}
PLA/PBAT-TA1%	93.81 ± 0.02^{b}	-0.61 ± 0.01^{c}	3.41 ± 0.01^{b}	3.97 ± 0.01^{b}	1.71 ± 0.06^{a}	58.41 ± 0.22^{a}
PLA/PBAT-TA5%	94.07 ± 0.01^{c}	-0.69 ± 0.01^{b}	3.58 ± 0.01^{c}	3.87 ± 0.00^{a}	1.58 ± 0.07^{b}	55.63 ± 0.16^{b}
PLA/PBAT-TA10%	94.28 ± 0.03^{d}	-0.73 ± 0.01^{a}	3.73 ± 0.01^{d}	3.88 ± 0.01^{a}	1.45 ± 0.02^{c}	52.83 ± 0.05^{b}
PLA/PBAT-GA1%	93.68 ± 0.02^{b}	-0.56 ± 0.01^{c}	3.37 ± 0.01^{b}	3.83 ± 0.01^{a}	1.85 ± 0.07^{a}	52.16 ± 0.12^{b}
PLA/PBAT-GA5%	94.07 ± 0.05^{c}	-0.67 ± 0.00^{b}	3.56 ± 0.01^{c}	3.84 ± 0.00^{a}	1.69 ± 0.12^{b}	48.68 ± 0.06^{a}
PLA/PBAT-GA10%	94.25 ± 0.01^{d}	-0.86 ± 0.01^{a}	3.90 ± 0.01^{d}	3.92 ± 0.01^{b}	1.53 ± 0.03^{c}	45.17 ± 0.03^{d}

Note: The letters (a–d) at each concentration (in the same column) indicate groups that are significantly different ($p < 0.05$) by mean.

The greenness of PLA-PBAT-TA10% and PLA-PBAT-GA10% increased by 2.35 times and 2.58 times, respectively, with respect to the control. In addition, a linear increase in the yellowness of both polyphenols was observed, with the yellowish tint in the film samples increasing as the concentration increased. The yellowness of PLA-PBAT-TA10% increased 1.59 times over the control film, while PLA-PBAT-GA10% increased 1.66 times over the control (PLA-PBAT) film. These results align with the study of Luzi et al. [22] where the yellowness of the Poly (vinyl alcohol-co-ethylene) films increased from 0.12 ± 0.00 (control) to 0.33 ± 0.03 (15%GA). Further, studies by Ahn et al. [20] also agree with the same results where the yellowness of the films increased from 1.79 ± 0.06 (LDPE control film) to 4.05 ± 0.78 (20%GA incorporated film). Similar results were observed when TA was incorporated into a matrix of chitosan or gelatin, where an increase was observed in the yellowness and greenness of the films [9]. The colour change is attributed to the phenolic content and pigments of GA and TA [20,22]. A slight decrease in the total colour difference (ΔE values) was observed as the concentration of TA and GA increased in the composite film (Table 1).

3.1.3. Optical Properties

A packaging film must have a UV barrier property as the UV rays may spoil the food due to the photo-oxidation reaction. A significant ($p < 0.05$) increase in the UV barrier property was observed as the concentration of TA and GA increased (Table 1). The decline in the transmittance at 280 nm (T_{280}) signifies superior UV blocking property as the disruption in the light passage through film increases [23]. A higher concentration of both polyphenols (PLA/PBAT-TA10% and PLA/PBAT-GA10%) showed around 65%–66% UV barrier properties. The increase in the phenolic content in the composite film could block the transmission of UV light and avoid the spoiling of food due to photo-oxidation. Similar results have been observed in various studies. Zhang et al. [24] had functionalised chitosan film with GA and observed a high UV blocking property, which was due to the strong absorption of UV light by GA moieties. Halim et al. [9] had also observed that the addition of TA reduced the penetration of UV light through the films. The chitosan TA film was found to have the lowest UV transmission of 0.005%. Further, the studies of Yadav et al. [25] confirm with the above studies that the UV barrier properties of chitosan film have been enhanced with the addition of zinc oxide nanoparticles loaded with gallic-acid. The homogeneous dispersion of GA in the polymer matrix and the blocking of the light transmission results in the enhanced UV barrier properties of the packaging films.

The appearance and transparency of food packaging hold great market value. The transparency of composite film is measured by the transmittance at 600 nm (T_{600}) (Table 1).

A significant difference between the control film and the tannic acid composite film and the gallic acid composite film was observed. The GA composite film was observed to be less transparent than the TA composite film. T_{600} of the control film was observed to be 73.38%. On incorporation of tannic acid (PLA/PBAT-TA10%), the T_{600} value is observed to be 52.83%, whereas by the incorporation of gallic acid (PLA/PBAT-GA10%) transmittance value becomes 45.17%. Wang and Wang [26] had also observed that the addition of TA decreased the transparency of soy protein isolate film. As per the studies of Yadav et al. [25], the transparency of the films was reduced with the addition of GA, and the opacity of the films increased. Further, as per the studies by Halim et al. [9], the transparency values increased after the addition of TA to a chitosan matrix. In the present study, the TA composite film showed more transparency and UV barrier properties than the GA composite film.

3.1.4. Mechanical Properties

Packed food must undergo several stress conditions during the processing, handling, and storage of food. The mechanical strength of the packaging film protects the food during these stress conditions. The ASTM D 882-88 method [27] was used to measure tensile strength (TS), elongation at break (EB), and elastic modulus (EM) (Table 2). Control film (PLA-PBAT) thickness was observed to be 55.53 µm. In the tannic acid incorporated composite film, the thickness has slightly increased to 56.43 µm (PLA/PBAT-TA10%), whereas on the incorporation of gallic acid, thickness has increased to 58.51 µm (PLA/PBAT-GA10%). For GA composite films a significant difference ($p < 0.05$) was observed between all the concentrations.

Table 2. Thickness, tensile properties, and hydrophobicity of the composite films with tannic acid and gallic acid.

Film	Thickness (µm)	Tensile Strength (TS) (MPa)	Elongation at Break (EB) (%)	Elastic Modulus (EM) (MPa)	Water Contact Angle (WCA)
PLA/PBAT	55.53 ± 0.03 ^{a}	4.80 ± 0.06 ^{a}	21.94 ± 11.42 ^{b}	199.80 ± 8.15 ^{c}	73.91° ± 2.79 ^{a}
PLA/PBAT-TA1%	55.52 ± 0.03 ^{a}	5.21 ± 0.85 ^{a}	21.98 ± 16.32 ^{c}	165.41 ± 6.32 ^{a}	73.15° ± 0.50 ^{ab}
PLA/PBAT-TA5%	56.02 ± 0.06 ^{b}	6.37± 0.08 ^{b}	22.38 ± 11.51 ^{a}	172.68 ± 5.13 ^{ab}	76.01° ± 3.48 ^{ab}
PLA/PBAT-TA10%	56.43 ± 0.02 ^{b}	8.63 ± 0.3 ^{c}	23.52 ± 9.18 ^{b}	181.56 ± 3.17 ^{b}	77.94° ± 0.78 ^{b}
PLA/PBAT-GA1%	56.19 ± 0.01 ^{b}	4.98 ± 0.51 ^{a}	23.10 ± 19.01 ^{c}	168.21 ± 6.64 ^{b}	72.51° ± 1.07 ^{b}
PLA/PBAT-GA5%	57.43 ± 0.02 ^{c}	5.97 ± 0.08 ^{b}	22.42 ± 16.43 ^{a}	177.46 ± 9.97 ^{a}	72.64° ± 1.07 ^{b}
PLA/PBAT-GA10%	58.51 ± 0.01 ^{d}	7.01 ± 0.95 ^{c}	22.09 ± 18.64 ^{a}	189.71± 16.04 ^{c}	68.34° ± 1.07 ^{a}

Note: The letters (a–d) at each concentration (in the same column) indicate groups that are significantly different ($p < 0.05$) by mean

Tensile strength is used to measure the strength of the film to resist the tension or outward pull. The TS value of the control film (PLA/PBAT) was observed as 4.80 MPa (Table 2). A significant increase ($p < 0.05$) in the TS value was observed as the concentration of tannic acid in the composite film increased. As the TA concentration increases from 1 wt%–10 wt%, the TS value increases by 1.08–1.80 times with respect to the control film, while as the GA concentration increases from 1 wt%–10 wt%, the TS value increases by 1.04–1.46 times that of the control film. For both TA and GA composite films, the tensile strength value was not significantly different ($p < 0.05$) between the control and 1 wt% concentration, though 5 wt% and 10 wt% were observed to be significantly different ($p < 0.05$).

The flexibility of the composite film is measured by the Elongation at Break (EB) or fracture strain. It shows the ability of the packaging film to resist shape deformation without fracturing in the processing, transportation, and storage conditions. The EB value of the control film (PLA/PBAT) was observed to be 21.94%. The incorporation of TA increased the flexibility of the composite film while decreasing the flexibility of the GA composite film. The EB value was observed to increase by 1.002–1.072 times as the concentration of

TA increased from 1 wt% to 10 wt%, whereas the EB value decreases by 1.052–1.007 times as the concentration of GA increased from 1 wt% to 10 wt%. However, the EB values of the control film and the TA and GA composite film were statistically insignificant ($p < 0.05$) (Table 2).

The stiffness of the composite film was measured by Elastic Modulus (EM). The EM value of the control film was observed as 199.80 MPa. The EM value was observed to increase from 165.41 MPa to 181.56 MPa as the concentration of TA increases from 1 wt% to 10 wt% in the composite film, while the EM value was observed to increase from 168.21 MPa to 189.71 MPa as the concentration of GA increases (1 wt% to 10 wt%). A significant ($p < 0.05$) difference was observed among all the (1 wt%, 5 wt%, and 10 wt%) GA composite films (Table 2).

Tannic acid incorporation is reported to increase the tensile strength of the packaging film in various studies. Kaczmarek et al. [15] observed that increasing the amount of tannic acid in the chitosan film resulted in the maximum increase in tensile strength. Rubentheren et al. [28] had also studied the influence of tannic acid in nanocrystalline cellulose film, where they observed an increase in tensile strength and stiffness due to the addition of tannic acid in the film. Moreover, Sun et al. [29] incorporated gallic acid in the chitosan film and observed an interruption in the inner structure of the film, reducing the tensile strength and elongation at the break by 33.6% and 66.1% [29]. The tensile strength and elastic modulus of the Poly (vinyl alcohol)/Tragacanth Gum films increased with the addition of GA. This enhancement in tensile strength and elastic modulus can be attributed to improved intermolecular interactions and cross-linking ability. Through several hydrogen bonds, the additional GA served as a crosslinker between the poly (vinyl alcohol) and tragacanth gum. In the present study, TS, EB, and EM have increased significantly ($p < 0.05$) with the addition of TA and GA; this may attribute to the enhancement of the intermolecular interactions and cross-linking ability.

3.1.5. Water Contact Angle (WCA)

The water contact angle (WCA) measures the hydrophobic or hydrophilic nature of the surface. It signifies the interactions of the film surface with the liquid interface [30], which is a necessity for film application in the food industry. The WCA of the control film was observed to be 73.91° (Table 2). Composite films are considered hydrophobic if the WCA is higher than 65° [12,31]. The tannic acid composite film's WCA rises from 73.15° to 77.94° as the tannic acid concentration rises from 1% to 10%, whereas the gallic acid composite film's WCA falls from 72.51° to 68.34° as the gallic acid concentration rises from 1% to 10%. Similar results have been observed in various studies, such as Huang et al. [19] who observed an increase in WCA on the addition of TA in the GA/zein composite film, resulting in a more hydrophobic film. Goudar et al. [21] used GA in a Poly (vinyl alcohol)/tragacanthin gum composite film and discovered that at lower GA weight percentages, the composite films formed hydrophobic films, whereas at higher GA weight percentages, the films became hydrophilic [21]. A cross-linked network was created by adding TA or GA to the matrix, which led to the production of a more compact structure than the control film. Composites' densely compacted structure minimized water molecules' ability to diffuse into the matrix, which raised the WCA of films.

3.1.6. Water Vapour Permeability

The water vapour permeability of packaging films is crucial in determining how much water is transmitted from the food to its environment. Figure 2 shows the water vapour permeability rate (WVPR) of control and PLA-PBAT-TA10%. The WVPR of the control was observed to be 81.62 ± 0.08 $g{\cdot}m^{-2}/h$, 78.48 ± 0.08 $g{\cdot}m^{-2}/h$ and 74.29 ± 0.14 $g{\cdot}m^{-2}/h$ after 24 h, 48 h and 72 h, respectively. Wherein, the WVPR of the PLA-PBAT-TA10% was observed to be 44.99 ± 0.06 $g{\cdot}m^{-2}/h$, 45.52 ± 0.09 $g{\cdot}m^{-2}/h$, 45.34 ± 0.1 $g{\cdot}m^{-2}/h$ after 24 h, 48 h and 72 h, respectively. When compared to the control sample, both the films had significantly lower WVPR. Similar results were observed in the study of Thiyagu et al. [32].

The samples containing cardanol oil displayed decreased WVP values when compared to the PLA/PBAT control film. This is because cardanol oil has longer side chains, which maintain its good hydrophobicity, and its aliphatic side chains lessen the polar effect of phenolic resins [32]. Similar results were reported by Roy et al. [33] where the WVP of the chitosan film reduced from $0.59 \pm 0.03 \times 10^{-9}$ g/m^2·Pa·s to $0.51 \pm 0.03 \times 10^{-9}$ g/m^2·Pa·s with the addition of TA. The decrease in WVP results in an increase in water vapour barrier properties; this is most likely due to a decrease in the film's free OH groups, which creates a convoluted path for vapour diffusion through the layer [33]. Thus, the PLA-PBAT-TA10% films had better water vapour barrier properties than the control film.

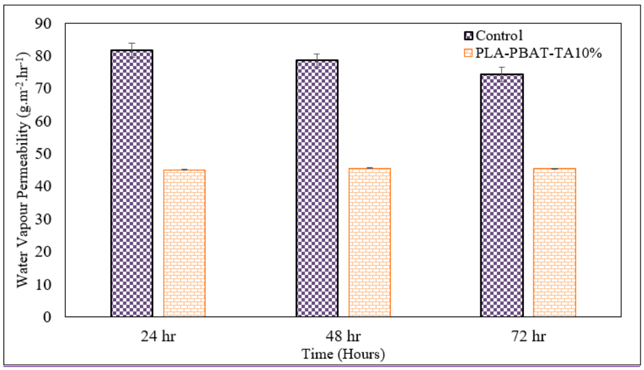

Figure 2. Water Vapour Permeability of PLA-PBAT-TA10%.

3.1.7. Thermogravimetric Analysis

The thermal stability of the composite film plays a significant role in the application of packaging film for the food industry. The potential use of films as a packaging material is supported by a TGA study that indicates adequate thermal stability. The weight loss pattern of the polyphenol (TA and GA)-incorporated composite film and the control film was observed by the TGA curve (Figure 3). All the composite films had shown two-stage thermal degradation. The first weight loss of around 6%–15% occurs due to the evaporation of solvent from the composite film and was observed around 30 to 70 °C. Similar results were observed where the polyphenol incorporated composite film had shown a stage weight loss of 5%–12% at 70 to 90 °C [12,14].

Second stage weight loss for the composite film was observed to range in between 300–360 °C. The second stage weight loss for the control film was observed to be at 310–320 °C. PLA/PBAT-TA1% was observed to degrade at 320–330 °C whereas PLA/PBAT-TA5% and PLA/PBAT-TA10% had shown weight loss at 350–360 °C (Figure 3a). The thermal stability of the tannic acid composite film increased significantly. PLA/PBAT-GA1% had degraded completely around 340–360 °C, which had declined to 340–350 °C as the concentration of gallic acid increased to 5 wt% and 10 wt% in the composite film (Figure 3b). Similar results have been found by Sharma et al. [12] where a two-stage thermal degradation was observed.

3.1.8. Antibacterial Activity

The antimicrobial efficiency of TA composite film and GA composite film against *E. coli* (NCTC 9001) and *L. monocytogenes* (NCTC 11994), as shown in Figure 4a–d; the control (PLA-PBAT) film had not shown antibacterial activity against *E. coli* (8.15 log CFU/mL) and *L. monocytogenes* (8.41 log CFU/mL) after 12 h. It was also observed (Figure 4a,b) that as the concentration of TA increased in the composite film, the antimicrobial activity against *E. coli* and *L. monocytogenes* also increased. TA composite film (1 wt%) had shown less antimicrobial activity (0.2 log CFU/mL) up to 9 h, after that *E. coli* growth was observed to increase. However, a slight growth reduction (0.4 log CFU/mL) in *L. monocytogenes* was observed after 12 h. In the presence of tannic acid composite film (5 wt%), *E. coli* growth was reduced by 1.3 log CFU/mL and *L. monocytogenes* was reduced by 1.8 log CFU/mL with respect to the control. While the 10 wt% TA composite film showed a significant growth reduction of 1.67 log CFU/mL for E. coli and 2.22 log CFU/mL for *L. monocytogenes* compared to the control, this could be attributed to an increase in phenolic content.

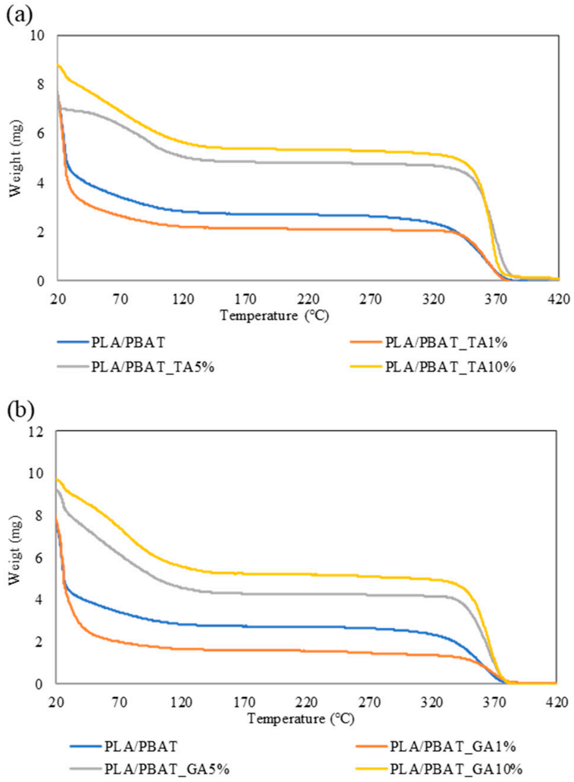

Figure 3. TGA of (**a**) Tannic acid (TA) incorporated PLA/PBAT film, (**b**) Gallic acid (GA) incorporated PLA/PBAT film.

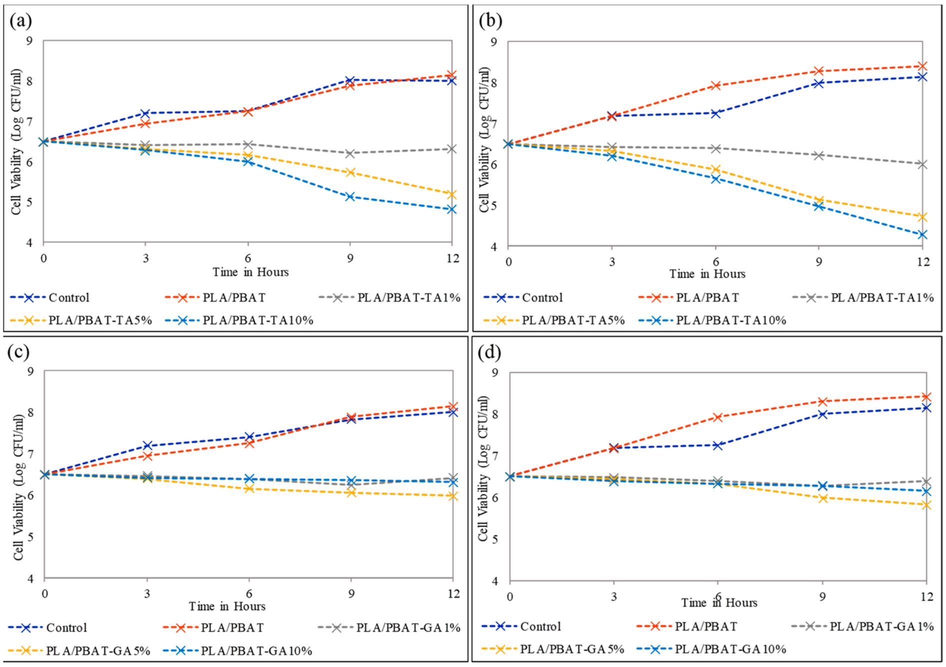

Figure 4. Antibacterial property of composite film: (**a**) effect of tannic acid on *E. coli*, (**b**) effect of tannic acid on *L. monocytogenes*, (**c**) effect of gallic acid on *E. coli*, and (**d**) effect of gallic acid on *L. monocytogenes*.

As shown in Figure 4c,d, the gallic acid composite film had depicted very low antibacterial efficiency against *E. coli* and *L. monocytogenes*. As the concentration of gallic acid in the composite film increased, the antibacterial activity was observed to be negligible. Overall, 5 wt% and 10 wt% gallic acid had shown less than 0.70 log CFU/mL reduction of both *E. coli* and *L. monocytogenes* bacterial growth. The low antimicrobial activity of 10 wt% gallic acid could be due to its denser rough structure, meaning the GA is unable to release from the film. The observed results indicate that TA composite film had shown higher antimicrobial activity than GA composite film; many studies had shown similar results. Pyla et al. [34] observed an enhancement in the antimicrobial and antioxidant activities of the packaging film on the incorporation of TA. Their results suggested that the use of tannic acid as an antimicrobial in a packaging system could results in extended shelf life for foods and improve food safety [34]. Widsten et al. [35] confirmed that tannic acid active packaging shows antibacterial properties against a broad spectrum of antibacterial activity.

3.2. Effects of Composite Films on the Cherry Tomato Quality

3.2.1. Weight Loss

Water loss in the respiration process and transpiration are the major cause of weight loss in tomatoes. Various studies have suggested the tendency of weight loss during ripening is due to transpiration [36,37]. The weight of the cherry tomatoes on average is taken to be 9.23 g. Figure 5 shows the incremental trend in the weight loss of cherry tomatoes during storage when packed in composite films. The use of GA composite film resulted in higher weight loss (39.67%) compared to the use of TA composite film (32.41%).

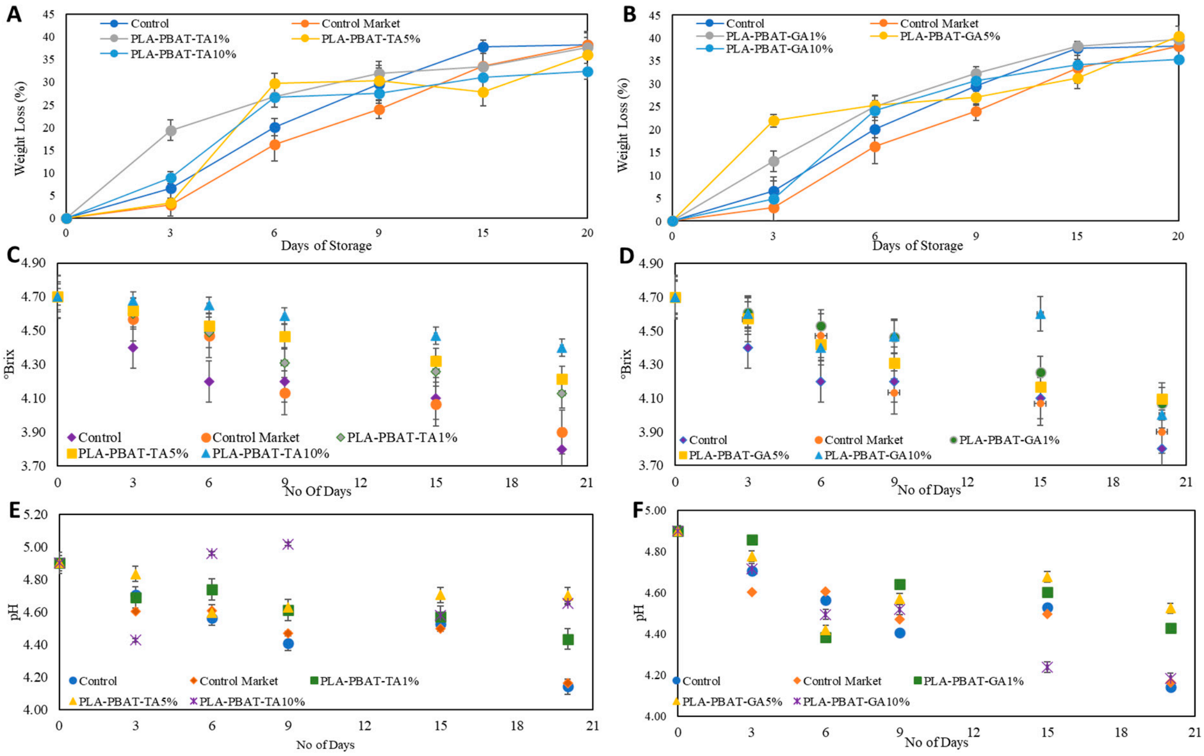

Figure 5. (**A**) Weight Loss of tomatoes during storage in TA composite film, (**B**) Weight Loss of tomatoes during storage in the GA composite film, (**C**) Total Soluble Solid (°Brix) of tomatoes in TA composite film, (**D**) Total Soluble Solid (°Brix) of tomatoes in GA composite film, (**E**) Change in pH of tomatoes in TA composite film, (**F**) Change in pH of tomatoes in GA composite film.

Weight loss in TA composite film was observed to be steady. The composite film incorporated with TA had shown the least weight loss among all the other films. After 20 days of storage at room temperature, the weight loss of tomatoes in PLA-PBAT-TA1 (wt%) was 37.61%, PLA-PBAT-TA5 (wt%) was 36.10%, and PLA-PBAT-TA10% was 32.41% (Figure 5A). It was noticed that the weight loss in TA composite film was 5.77% less than the control composite film and 5.83% less than the market control packaging. However, GA-incorporated films had shown weight loss similar to that of control film and market control films. Weight loss in PLA-PBAT-GA1 (wt%) was observed to be 39.67%, PLA-PBAT-GA5 (wt%) was 40.44%, and PLA-PBAT-GA10 (wt%) was 35.39% after 20 days of storage (Figure 5B). Aragüez et al. [17] had also observed a weight loss of 15%–17% in an active box and suggests that the senescence of tomatoes led to the uniform increase in the weight loss of the plastic bagged tomatoes.

3.2.2. Total Soluble Solids and pH

The total soluble solids (TSS), indicated as a percentage of fresh matter mass, has a significant positive association with sugar content and are thus widely recognized as an important fruit quality feature. A TSS value that is near to that of a fresh sample indicates that the sample has better shelf-life retention. Figure 5C shows °Brix of the tomatoes during storage at room temperature. The °Brix of tomatoes on day 0 was observed to be 4.7. As expected, the TSS of the tomatoes was reduced during the storage. On day 20, it has slight decrease of 6.3% in the tannic acid composite film packed tomato (PLA-PBAT-TA10%) (Figure 5C), whereas for gallic acid packed tomato (PLA-PBAT-GA10%), the °Brix value decreased by 14.9% (Figure 5D), which is similar to the control film (19.14%) and control market film (17%). Similar results have been observed by Buendía–Moreno et al. [38] where the TSS value had reduced during storage of 20 days.

The pH is used to determine the quality of tomatoes. Due to maturity and the use of organic acids as a respiration substrate, the pH of tomatoes rises during storage [17]. The market requirement of tomatoes requires a °Brix between 5.5–9.5 and pH between 4.3–4.9. Figure 6E shows the effect of cherry tomatoes on pH. The pH of tomatoes at 0 days was observed to be 4.9. On the 20th day, the pH of cherry tomatoes in tannic acid composite film (10%) was observed to be 4.6, whereas it was was 4.1 for 10% gallic acid film–which is similar to control and control market film (Figure 5E,F). The cherry tomatoes in tannic acid composite films were in the market requirement range even on the 20th day.

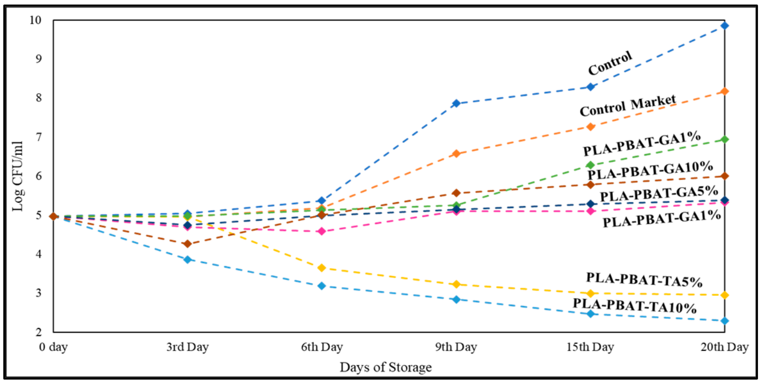

Figure 6. Effect of different packages on the total bacterial count of tomatoes stored for 20 days.

3.2.3. Antimicrobial Activity

The total bacterial count was used as a basic assessment of the fruits' total hygienic conditions. Figure 6 depicts the total bacterial count of samples during the course of the storage time up to 20 days. The bacterial growth reduced in the presence of tannic acid. At the 20th day, the bacterial count on PLA-PBAT-TA5% had reduced by 2.05 log CFU/mL and on PLA-PBAT-TA10% had reduced by 2.70 log CFU/mL. However, for PLA-PBAT-GA10% the bacterial growth had increased by 1 log CFU/mL. The control market and the control film has shown an increase in the bacterial count by 3.25 and 4.85 log CFU/mL, respectively, at the 20th day.

4. Conclusions

The potential of active food packaging was studied by incorporating tannic acid and gallic acid in the polymer blend (PLA-PBAT). The influence of TA and GA in the polymer blend on various physical, chemical, thermal, mechanical, and antimicrobial properties was investigated. In addition, the shelf life of cherry tomatoes packed in the polymer blend films were studied. Overall, 10 wt% TA and GA composite film had shown around a 65%–66% increase in the UV barrier property as the increase in the phenolic content in the composite film blocked the transmission of UV light and could avoid the spoiling of food due to photo-oxidation. The tensile strength value after the incorporation of 10 wt% of TA and GA had increased by 1.80 times and 1.46 times, respectively, in the composite film with respect to the control film. Furthermore, the tannic acid-incorporated composite film had demonstrated significantly more antibacterial efficacy than the gallic acid-incorporated composite films. Tannic acid composite film (10 wt%) showed a 1.7 log reduction in *E. coli* growth and 2.2 log reduction in *L. monocytogenes.* The PLA-PBAT-10%TA film had higher water vapour barrier properties when compared to the control. In addition, TA composite film (10%) was able to maintain the quality of cherry tomatoes for up to 20 days of storage at room temperature. For cherry tomatoes packed in PLA-PBAT-TA10 (wt%), the TSS has decreased by 6.3%, pH was 4.3, and a microbial reduction of 2.70 log CFU/mL was observed. Therefore, tannic acid composite film demonstrated a high UV blocking property, surface hydrophobicity, and antibacterial property, which shows its potential for active packaging film.

Author Contributions: Conceptualization, S.S., K.Y.P., D.P., A.K.J. and S.J.; Data curation, S.S., K.Y.P. and D.P.; Formal analysis, S.S., K.Y.P., D.P. and S.J.; Funding acquisition, A.K.J.; Investigation, S.S., K.Y.P. and D.P.; Methodology, S.S., K.Y.P., D.P. and S.J.; Project administration, A.K.J.; Resources, B.D., A.K.J. and S.J.; Supervision, B.D., A.K.J. and S.J.; Validation, S.J.; Writing—original draft, S.S., K.Y.P. and D.P.; Writing—review & editing, S.S., K.Y.P., D.P., A.K.J. and S.J. All authors have read and agreed to the published version of the manuscript.

Funding: The present work was supported by Technological University Dublin-City Campus, Ireland under the Fiosraigh Scholarship programme, 2017, and TU Dublin Researcher Award 2020, TU Dublin Researcher Award 2021.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Acknowledgments: Authors would like to acknowledge the funding from Technological University Dublin-City Campus, Ireland under the Fiosraigh Scholarship programme, 2017.

Conflicts of Interest: The authors declare no conflict of interest.

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