Research Article

Optimization of Edible Coating Formulation Using Response Surface Methodology for Delaying the Ripening and Preserving Tomato (Solanum lycopersicum) Fruits

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Tomato is a climacteric fruit that has a short shelf life under natural conditions. However, some treatments can be applied in order to extend the shelf life and improve the quality of fruits. The present study aimed at formulating a coating solution based on coffee leaf extract, starch, and Gum Arabic in order to extend the postharvest shelf life of tomato. The response surface methodology (RSM) was used to determine the optimal concentrations of the different abovementioned components in the coating, for better delay the ripening process of tomatoes. The central composite design (CCD) was generated, and the effect of different factors on the shelf life and quality (percent ripening, firmness, physiological loss of mass, pH, chlorophylls $a$ and $b$ contents, lycopene, and $\beta$-carotene contents) of tomato fruits were determined 14 days after treatment and stored at room temperature. Chlorophyll $a$ and chlorophyll $b$ contents as well as firmness of treated fruits were higher than those of control fruits. In addition, treated fruits recorded low lycopene and $\beta$-carotene contents, physiological loss of mass, pH, and ripening percentage compared to control fruits. Coffee leaf extract had a significant effect ($p < 0.05$) on chl $a$ content, pH, and ripening percentage. On the other hand, starch and Gum Arabic did not significantly ($p > 0.05$) influence pH, physiological loss of mass, and percentage of ripening. The interaction between coffee leaf extract and starch $(X_1X_2)$ had the most significant effect ($p < 0.05$) on the studied parameters. The results showed that the experimental data could be adequately fitted into a second-order polynomial model with coefficients of determination greater than 80% for all variables studied. The optimal coating formulation consisted of 78.5 g/l coffee leaves extract, 56 g/l starch, and 9.5% Gum Arabic. There was no significant difference between the experimental and predicted data.

1. Introduction

Edible coatings are thin layers of edible component, which are applied to the surface of foods either by dipping or brushing, in addition to or as a replacement for the natural protective coating [1–4]. They are low-cost, derived from renewable sources, biodegradable, and have specific gas transmission properties. Edible coatings are mainly composed of polysaccharides such as starch and Gum Arabic which can be applied to provide a selective barrier to oxygen, carbon dioxide, and moisture, thus preserving fruits and vegetables from rapid spoiling [5].

Several studies have shown the effectiveness of these components in prolonging postharvest fruit shelf life and maintaining fruit quality [6–8]. Gum Arabic and starch are hydrocolloids and hydrophilic polymers with hydroxyl groups in their molecular structure. These hydroxyl groups could help scavenge free radicals during the browning process. To date, these hydrocolloids (Gum Arabic and starch) have been widely used in coating formulations for fruits preservation [9]. The effectiveness of these compounds for preservation could be further booster by combining them with bioactive substances from agricultural by-products [10].
Samuchaya et al. [11] reported that dried coffee leaves powder (*Coffea arabica*), as a by-product, has high polyphenol and procyanidins contents and high free radical scavenging capacity. Coffee leaves have been confirmed to be a food source for humans, owing to its richness in phenolic compound and its high antioxidant activity. Thus, coffee leaves could be used as a component of edible coating formulations, in addition to starch and Gum Arabic, to prolong the shelf life of fruits.

Tomato (*Solanum lycopersicum* L.) is a plant of the Solanaceae family. It is cultivated for its fruits commonly known as tomatoes which are consumed worldwide [12]. Tomatoes have a high nutritional value owing to their richness in vitamins, minerals, natural antioxidant compounds, and amino acids. Several other health-promoting substances have been found in tomatoes, such as carotenoids, folic acid, ascorbic acid, lycopene, and β-carotene, which have been correlated with reduced risk of cancer and some heart diseases in humans [14]. Tomato is a fruit with a short shelf life (ranging from one to two weeks) [15]. It is classified as a climacteric and highly perishable fruit with a high respiratory peak associated with a high rate of ethylene production at postharvest [16]. Low-temperature storage is inappropriate for tomatoes because this technique of conservation leads to the loss of some quality parameters such as color, firmness, flavor, and appearance, all of which affect the commercial interest of the fruit. Previous studies reported that at room temperature, the shelf life of tomato fruits can be extended from 19 to 30 days by applying different coating formulations notably, the edible coating based on Gum Arabic [8, 16], cassava starch [17], pectin and chitosan [18], and ethanol [19]. The experimental results suggested that coating formulations should be sought to further extend the shelf life of tomatoes. To this end, the combination of coffee leaf extract, starch, and Gum Arabic in a formulation could better preserve tomato fruits. The response surface methodology (RSM) has been proven to be a powerful tool to determine the effects of each factor and the interactions between them, which allow the optimization of the processes [20]. Therefore, the objective of this research study was to use the response surface methodology to determine the optimal formulation of edible coating based on coffee leaf extract, Gum Arabic, and starch, in order to increase the shelf life and preserve the quality of tomato fruits.

### 2. Materials and Methods

#### 2.1. Plant Material.

Tomato fruits of Rio Grande variety were harvested at the “turning stage” in an experimental farm located in Dschang (5°10′–5°38′ N, 9°50′–10°20′ E, altitude 1400 m) and immediately transported to the laboratory of Applied Botany of the University of Dschang, Cameroon, for the experiments. The “turning stage” is defined as the developmental stage at which a yellowish ring is visible at the distal end of the fruit [21]. Only firm, disease-free, and uninjured fruits of the same size were selected for experimentation.

The coffee leaves used in this study were harvested from coffee plants in the locality of Foumbot, a district located in the western Cameroon (5°30′0″–5°30′53″ N, 10°37′60″–10°60′50″ E). These leaves were washed with water and air-dried in the shade to a constant weight. The Gum Arabic used was purchased from a local market in the city of Garoua (Cameroon). The starch used for the study was extracted from freshly harvested cassava tubers, following the method described by Belibi [22].

#### 2.2. Reagents.

All the chemicals which were used for the study were from Sigma-Aldrich (Germany), with purity > 98%.

#### 2.3. Preparation of the Coating Solution and Treatment of the Fruits

##### 2.3.1. Preparation of the Coffee Leaf Extract.

Using a stainless steel Wiley mill, the dried coffee leaves were ground into powder to pass through a 60-mesh screen and used to prepare the extracts for coatings, following the methodology described by Aghofack-Nguemezi et al. [23]. Different quantities of coffee leaves powder corresponding to the different concentrations tested were macerated in 1500 ml of water/ethanol mix at 1:2 (v/v) ratio. Each mixture was left to stand for 3 hours at room temperature and then filtered through a muslin cloth. The residue from the filtration was then macerated in 750 ml of the same solvent (water/ethanol at 1:2 v/v ratio) for one hour after which the mixture was filtered. The two filtrates were mixed and used as coffee leaf extract (CLE). The volume of each coffee leaf extract was thus 2250 ml.

##### 2.3.2. Coating Preparation.

The Gum Arabic coating was prepared following the method described by Khalig et al. [24]. Different weights of gum Arabic powder, corresponding to the different concentrations tested, were dissolved in 750 ml of distilled water for 60 minutes and then the mixtures were filtered. After filtration, different quantities of starch which were needed to have the different concentrations tested were added to the filtrate, and then the mixtures were heated to a temperature of 100°C while homogenizing, in order to have the Gum Arabic/starch solutions [20]. After these operations, different coffee leaf extracts were mixed with the corresponding Gum Arabic/starch solution to obtain the desired combinations of coffee leaf/Gum Arabic/starch concentrations. The total volume of each coating solution was thus 3000 ml consisting of a mixture water/ethanol (1:1, v/v). Bleach was added to each solution at the rate of 230 μl/l in order to disinfect the mixture.

##### 2.3.3. Treatment of the Fruits.

Tomato fruits were thoroughly washed with distilled water and soaked for 25 minutes in the different coating solutions T1 to T20. The control fruits were soaked in distilled water to which bleach
was added at the rate of 230 μl/l. Three replicates each of 8 fruits were used per treatment.

2.4. Experimental Plan. The response surface methodology (RSM) was performed using Minitab version 2018 software package. The central composite design (CCD) was used to determine the effects of three independent variables (coffee leaf extract concentration, starch concentration, and Gum Arabic concentration) on the dependent variables (responses) which were percentage of ripening (PR), firmness, pH, physiological loss of mass (PLM), and pigments (Chl b, Chl b, β-carotene and lycopene) contents.

The choice of factors and their levels of variation were made considering data from previous studies [8, 17, 25]. The different factors and levels (minimum, center, and maximum) are shown in Table 1. The experimental design generated 20 edible coating formulations from the central composite design in which there were 06 (six) replicates of the points at the center (Table 2) [26]. The six center points show the repeatability of the method [27].

2.4.1. Empirical Model for the Prediction of Ripening Parameters. Experimentally obtained data were subjected to multiple regression analysis. The empirical model was developed by fitting the experimental data obtained from the central composite design device into a second-order polynomial mathematical equation (1). In order to determine the relationship between the independent variables (factors) and the response variables from the mathematical model equation, a second-order polynomial function was developed for each response. The 3D curves were constructed with two independent variables while keeping the 3rd variable constant.

The general model of the polynomial equation is as follows:

\[ Y_1 = \alpha_0 + \alpha_1X_1 + \alpha_2X_2 + \alpha_3X_3 + \alpha_{11}X_1^2 + \alpha_{22}X_2^2 + \alpha_{33}X_3^2 + \alpha_{12}X_1X_2 + \alpha_{13}X_1X_3 + \alpha_{23}X_2X_3, \] (1)

where \( Y_1 \) is the predicted response; \( \alpha_0 \) is the intercept term; \( \alpha_1, \alpha_2, \text{ and } \alpha_3 \) are linear coefficients; \( \alpha_{12}, \alpha_{13}, \text{ and } \alpha_{23} \) are interaction coefficients; \( \alpha_{11}, \alpha_{22}, \text{ and } \alpha_{33} \) are quadratic coefficients, and \( X_1, X_2, \text{ and } X_3 \) are coded independent variables [28].

2.5. Evaluation of the Effect of Coating Solutions. The effect of the different coating solutions was evaluated on the 14th day after treatment (date at which the control fruits started their senescence). The parameters investigated included the percentage of ripening (PR), firmness, physiological loss of mass (PLM), pH, and pigments (chlorophyll a, chlorophyll b, β-carotene, and lycopene) contents.

2.5.1. Determination of the Percentage of Ripening. A fruit was considered ripe when it was fully red. The percentage of ripening at day 14 was calculated according to the following formula:

\[ \text{PR} = \left( \frac{\text{number of ripe fruits}}{\text{total number of fruits}} \right) \times 100, \] (2)

with PR = percentage of ripening.

2.5.2. Measurement of the Firmness. A penetrometer (GY-2, SAUTER GmbH, Germany) was used to measure the firmness of the tomato pulp. The tomato epicarp was removed from three locations on the fruit using a razor blade. The penetrometer was zeroed and the tip head placed on the pulp of the peeled area of the fruit. A cylindrical probe (diameter: 4 mm) of convex type was used to perforate the three previously peeled areas of the tomato. Continuous downward pressure was applied so that the tip sank into the fruit pulp to the depth marked (halfway up) on the tip. The tip was then removed and the value indicated on the dial of the penetrometer noted. The firmness values were expressed in Newton (N).

2.5.3. Measurement of the Physiological Loss of Mass. Tomatoes were weighed using balance (METTLER, PB603-S, Germany) on day 1 to obtain the initial mass and then on day 14 to obtain the final mass. The physiological loss of mass was calculated using the following formula by Athmaselvi et al. [29]:

\[ \text{PLM} \text{(%)} = \frac{\text{initial mass} - \text{final mass}}{\text{initial mass}} \times 100, \] (3)

with PLM = Physiological loss of mass.

2.5.4. pH Measurement. A pH meter (ATC, Lutron PH-221, Taiwan) was used to measure the pH of tomato juice. The tomato fruit juice was prepared as follows: 15 g of tomatoes were crushed and the crushed material was filtered through muslin cloth into a beaker. The pH meter probe was then inserted into the juice, and the pH value was directly read on the screen. Three measurements were made for each treatment.

2.5.5. Determination of Pigments’ Contents. The determination of pigments’ contents was carried out according to the method described by Nagata and Yamashita [30]. Pigments were extracted by grinding 5 g of fresh material in 10 ml of solvent (acetone/hexane in a 4/6 (v/v) ratio). The mixture was stored at 4°C for 24 hours. The mixture was filtered through a Whatman paper N’1. The absorbance of hexanolic extracts was measured at 663, 645, 505, and 453 nm wavelengths using a UV-visible spectrophotometer.

\[ Y_1 = \alpha_0 + \alpha_1X_1 + \alpha_2X_2 + \alpha_3X_3 + \alpha_{11}X_1^2 + \alpha_{22}X_2^2 + \alpha_{33}X_3^2 + \alpha_{12}X_1X_2 + \alpha_{13}X_1X_3 + \alpha_{23}X_2X_3, \] (1)
The concentrations of the different pigments were determined by the following equations:

(i) Chl \(a\) (mg/100 ml) = 0.0999A\(_{663}\) − 0.0989A\(_{445}\)

(ii) Chl \(b\) (mg/100 ml) = −0.328A\(_{663}\) + 1.77A\(_{445}\)

(iii) Lycopene (mg/100 ml) = −0.0458A\(_{663}\) + 0.204A\(_{445}\) + 0.372A\(_{505}\) − 0.0806A\(_{453}\)

(iv) \(\beta\)-Carotene (mg/100 ml) = 0.0216A\(_{663}\) − 1.22A\(_{445}\) − 0.304A\(_{505}\) + 0.452A\(_{453}\)

\(A_{663}\), \(A_{445}\), \(A_{505}\), and \(A_{453}\) are the absorbances at 663 nm, 445 nm, 505 nm, and 453 nm, respectively.

### 2.6. Analysis of the Data

Statistical analysis of data was carried out using Minitab version 18 software package. The response surface methodology (RSM) was used for the generation of response surface and for the optimization of process variables. Determination of the coefficients (\(a\)) of the different models was done by a matrix approach using multiple linear regression [31]. A total of twenty runs were provided by central composite design by RSM. Probability value (model significance) was used to assess the quality of the model on the one hand, and the calculation of coefficients of determination (\(R^2\)) that measure the fitness of the regression model on the other hand [32]. The regression analysis was used to determine the effects of variables in first-order, two-factor interactions, and second-order polynomial models [33]. Indeed, when the coefficient of determination (\(R^2\)) is close to 1 (\(R^2 \geq 0.8\)), the degree of correlation is high between the observed and predicted values, highlighting a reasonable agreement of the model with the experimental results [34]. Variability of terms in the regression equation for each response was determined using analysis of variance (ANOVA). The significance of the linear, quadratic, and interaction effects of the different factors; as well as that of each of these coefficients was determined by comparing the observed probability (\(p\) value) to a critical probability (\(p = 0.05\)).

### 2.7. Optimization and Validation

To determine the optimal values of the independent variables (coffee leaf extract, starch, and Gum Arabic concentrations), the graphical and numerical optimizations were performed [35]. The three-

| Table 1: Levels of independent variables used in the centered composite design. |
|---------------------------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| Independent variables           | Symbols         | Levels of coded variables |
| Coffee leaf powder (g/l)        | \(X_1\)         | 10.66           | -1.6817         | Center (0)      | Maximum +1      |
| Starch (g/l)                    | \(X_2\)         | 15              | 2               | 5               | 15             |
| Gum Arabic (%)                  | \(X_3\)         | 2               | 10              | 15              | 18             |

| Table 2: Central composite design in experimental design and the experimental data obtained for the responses of the studied variables. |
|---------------------------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| Order essay                     | Independent variables | Dependent variables |
| 1                               | CLE (g/l)        | Starch (g/l)   | GA (%) m/v      | Chl \(a\) (mg/ g) | Chl \(b\) (mg/ g) | Lycopene (mg/g) | \(\beta\)-carotene (mg/g) | pH | PLM (%) | Firmness (N) | PR (%) |
| 1                               | 66.66           | 40              | 10              | 0.036           | 0.033           | 0.045           | 0.054           | 4.300 | 6.78    | 39.16       | 44.44 |
| 2                               | 66.66           | 40              | 10              | 0.036           | 0.031           | 0.045           | 0.055           | 4.300 | 6.25    | 39.22       | 50.00 |
| 3                               | 33.33           | 55              | 5               | 0.034           | 0.012           | 0.123           | 0.082           | 4.300 | 5.53    | 36.67       | 57.89 |
| 4                               | 66.66           | 65.23           | 10              | 0.058           | 0.008           | 0.058           | 0.109           | 4.240 | 5.08    | 39.22       | 30.55 |
| 5                               | 100             | 25              | 5               | 0.020           | 0.012           | 0.145           | 0.073           | 4.310 | 6.98    | 34.84       | 83.33 |
| 6                               | 100             | 25              | 15              | 0.049           | 0.022           | 0.064           | 0.077           | 4.267 | 5.43    | 34.91       | 65.78 |
| 7                               | 100             | 25              | 15              | 0.058           | 0.012           | 0.023           | 0.095           | 4.233 | 5.71    | 38.24       | 55.55 |
| 8                               | 100             | 55              | 5               | 0.037           | 0.030           | 0.044           | 0.061           | 4.300 | 6.89    | 37.56       | 44.44 |
| 9                               | 100             | 55              | 15              | 0.047           | 0.015           | 0.057           | 0.063           | 4.210 | 5.49    | 34.71       | 55.55 |
| 10                              | 100             | 55              | 15              | 0.041           | 0.008           | 0.048           | 0.083           | 4.267 | 5.84    | 38.63       | 83.33 |
| 11                              | 100             | 55              | 15              | 0.053           | 0.017           | 0.052           | 0.091           | 4.333 | 6.97    | 39.22       | 66.66 |
| 12                              | 33.33           | 55              | 15              | 0.047           | 0.015           | 0.054           | 0.104           | 4.360 | 5.43    | 39.22       | 66.66 |
| 13                              | 66.66           | 40              | 10              | 0.036           | 0.033           | 0.045           | 0.054           | 4.300 | 6.25    | 39.22       | 50.00 |
| 14                              | 66.66           | 40              | 10              | 0.036           | 0.033           | 0.045           | 0.054           | 4.300 | 6.53    | 38.29       | 55.55 |
| 15                              | 122.72          | 40              | 10              | 0.041           | 0.008           | 0.048           | 0.083           | 4.267 | 5.84    | 38.63       | 83.33 |
| 16                              | 33.33           | 25              | 5               | 0.017           | 0.007           | 0.056           | 0.096           | 4.300 | 7.27    | 33.70       | 55.55 |
| 17                              | 100             | 55              | 5               | 0.046           | 0.004           | 0.056           | 0.081           | 4.200 | 5.37    | 38.89       | 55.55 |
| 18                              | 66.66           | 40              | 10              | 0.019           | 0.035           | 0.154           | 0.095           | 4.376 | 5.91    | 37.19       | 61.11 |
| 19                              | 100             | 55              | 15              | 0.058           | 0.023           | 0.054           | 0.091           | 4.233 | 5.71    | 39.22       | 66.66 |
| 20                              | 66.66           | 40              | 10              | 0.024           | 0.005           | 0.058           | 0.059           | 4.367 | 5.72    | 39.22       | 66.66 |
| Control                         | 0               | 0               | 0               | 0.022           | 0.000           | 0.189           | 0.091           | 4.26  | 7.47    | 25.95       | 100.00 |

CLE: concentration of coffee leaf extract; GA: concentration of Gum Arabic; chl \(a\): chlorophyll \(a\); chl \(b\): chlorophyll \(b\); PLM: physiological loss of mass; PR: percentage of ripening.
dimension (3D) response surface plots were generated from the fitted model for each dependent variable in order to better observe the interaction effects of coffee leaf extract, starch, and Gum Arabic concentrations on the responses. The numerical optimization of the response(s) was carried out using the desirability function approach described by Derringer and Suich [36], that is, to have the maximum values of chlorophylls contents and fruit firmness and minimum values of percentage ripening, physiological loss of mass, pH, lycopene, and β-carotene contents.

Tukey’s comparison test was performed between the predicted and experimental response values to check the adequacy of the final model response.

3. Results and Discussion

3.1. Fitted of the Model. Table 3 shows that except the coffee leaf extract concentration, each of the factors had significant influence on responses. The interaction term between X1 and X2, X1 and X3 influenced the ripening parameters studied. Furthermore, the square (quadratic) terms of all these factors significantly influenced the different responses. In addition, the coefficients of determination $R^2$ showed values all above 80%. This would mean that the generated polynomial models are adequate to explain the effects of coffee leaf extract, starch, and Gum Arabic concentrations on the different measured parameters [37]. In other words, there is a good agreement between the data experimentally obtained and the data predicted by the software.

3.2. Response Surfaces

3.2.1. Effects of Edible Coating on Chlorophyll a and Chlorophyll b Contents. Chlorophyll a and chlorophyll b concentrations were higher in treated fruit than in control fruit. The highest chl a content (0.058 mg/g) was recorded with essays number 5 and 9, while essay number 3 resulted in the highest chl b content (0.034 mg/g) (Table 2). Figure 1 shows that tomatoes treated with 66.66 g/l leaf extract showed the highest chl a content. On the other hand, chl a content increased with increasing starch concentration.

Figure 2 indicates that chl b levels were highest in tomatoes treated with coffee leaf extract and Gum Arabic at the concentrations of 66.66 g/l and 10%, respectively. Each of the three factors significantly influenced chlorophyll a content, while chlorophyll b was only significantly influenced by starch and Gum Arabic concentrations (Table 3). However, the quadratic effect of each factor ($X_1^2$, $X_2^2$, and $X_3^2$) significantly influenced these two pigments contents. Meanwhile, $X_1X_2$ and $X_1X_3$ interactions significantly influenced the chl a and b contents. The coefficients of determination ($R^2$) were greater than 94% for the two pigments and the lack of fit were 0.148 and 0.02 for chl a and chl b, respectively, indicating a well fitted response models. Color change from green to red is an indicator of tomato ripening [38]. This change is associated with the degradation of chlorophylls, followed by biosynthesis of carotenoids such as xanthophylls and carotenes [39]. This degradation normally occurs during ripening of tomato fruit when stored under ordinary conditions (without treatment and at room temperature), but this degradation can be delayed when the fruit is stored at low temperature or when it is coated and stored. The coating may have slowed the activity of chlorophyllase, an enzyme that catalyzes the degradation of chlorophylls. Indeed, in fruits treated with cytokine (benzyl-aminopurin), Wang et al. [40] reported a slowdown of chlorophyll degradation as a response to a decrease in chlorophyllase activity. Takamiya et al. [41] showed that the degradation pathway of chlorophyll includes dephytylation, i.e., the removal of the magnesium atom from tetrapyrrrole macrocycle. The dephytylation involves chlorophyllase activity [42]. The chlorophyll a and b content of the control fruit was very low compared to that of fruit from all treated lots. This result is similar to that obtained by Aghofack et al. [23] who showed that coating with cocoa leaf extracts delays the degradation chlorophyll a in tomato fruit. Similar drop of chlorophyll a degradation were reported by Donjio et al. [10] with tomatoes coated with pineapple peel extract and Gum Arabic. During chlorophyll pigment degradation, chl b degrades faster than chl a [43]. The results of this study confirm this fact as it was observed that chl a was always higher than chl b. This shows that the degradation of chl a and chl b are carried out differently. Indeed, chl b is destroyed either directly by reductase [44] or by conversion to chl a before its degradation [43].

3.2.2. Effects of Edible Coating on Lycopene and β-Carotene Contents. Lycopene levels ranged from 0.044 to 0.144 mg/g for all coated fruits after 14 days of storage. In the control fruits, the lycopene content was 0.189 mg/g. For β-carotene content, values ranged from 0.043 to 0.109 mg/g in coated fruit, while the value in uncoated fruits was 0.091 mg/g. Lycopene and β-carotene levels were higher in control fruit compared to treated fruit, except the fruits treated with formulations 5, 9, and 12, in which β-carotene levels were slightly higher than in control fruits (Table 2). Only the starch and Gum Arabic concentrations had significant linear effect on these two responses during ripening (Table 3). The quadratic terms of the three factors significantly influenced the β-carotene content. Figure 3 shows that lycopene content decreases with increasing starch concentration. The lowest lycopene content was obtained at 10% Gum Arabic. Figure 4 shows that β-carotene content was lowest when coffee leaf extract and starch concentrations were around 66.66 g/l and 40 g/l, respectively. The lack of fit was significant ($p < 0.01$) for lycopene and not significant ($p = 0.098$) for β-carotene, but the coefficients of determination ($R^2$) were well enough for well-fitted models (Table 3). Lycopene is a carotenoid associated to color change during ripening [45, 46]. Previous studies have shown that in ripening fruits chlorophyll degradation and lycopene accumulation are positively correlated [47]. The present study shows that lycopene and β-carotene contents were lower in treated fruits compared to control fruits, indicating that the coating solutions formulated with coffee leaf extract, starch, and Gum Arabic slowed down the degradation of chlorophyll, and the synthesis of carotenoids as well. This is in agreement with Daisy et al. [45].
who reported that Gum Arabic-based coating has good ability to delay carotenoid synthesis during fruit ripening.

3.2.3. Effect of Edible Coating on pH. There was very little variation in pH among treatments. Indeed, pH ranged from 4.2 to 4.36 for treated fruit and was 4.26 for untreated fruit. The results of this work are consistent with those of Adjouman et al. [17] who recorded pH values ranging between 4.29 and 4.31 at 21 days after coating tomato fruits.

Table 3: Regression coefficients, coefficients of determination \( (R^2) \), and \( p \) value of lack of fit of predicted equations.

| Sources | Constant | \( X_1 \) | \( X_2 \) | \( X_3 \) | \( X_1X_2 \) | \( X_1X_3 \) | \( X_2X_3 \) | \( X_1^2 \) | \( X_2^2 \) | \( X_3^2 \) | \( R^2 \) (%) | Lack of fit |
|---------|---------|---------|---------|---------|-----------|-----------|-----------|-------|-------|-------|-------|---------|-----------|
| Chl a   | 0.036** | 0.0043**| 0.007***| 0.009** | 0.002     | 0.0001    | –0.004    | –0.0013*| 0.034**| 0.0010*| 98.99  | 0.148     |
| Chl b   | 0.032***| 0.0007  | –0.004**| 0.002*  | –0.003*  | 0.0001    | –0.0005   | –0.0092**| –0.004**| –0.0060**| 94.46  | 0.02*     |
| Lycopene| 0.045***| –0.004  | –0.0070*| –0.027**| –0.02*** | 0.0008    | 0.0032    | 0.0041  | 0.0056 | 0.0187***| 93.82  | 0.00**    |
| \( \beta \)-carotene| 0.052***| 0.002   | 0.01**  | 0.01**  | –0.0007  | –0.00003  | 0.008     | 0.0077**| 0.0128**| 0.0075**| 89.44  | 0.098     |
| pH      | 4.29*** | –0.035**| –0.007  | 0.018** | –0.017*  | 0.0087    | 0.0085    | 0.023** | 0.0026 | 0.00003  | 89.57  | 0.00**    |
| PLM     | 6.67*** | –0.033  | –0.310* | –0.15   | 0.111    | 0.032     | 0.395*    | –0.306*| –0.48**| 0.051   | 84.5   | 0.093     |
| Firmness| 3.92*** | –0.006  | 0.149** | 0.064*  | 0.030    | –0.084*   | –0.021    | –0.08** | –0.035 | 0.00003  | 85.4   | 0.22      |
| PR      | 49.06***| 3.44    | –5.28** | –0.24   | –5.74**  | 3.99**    | 2.60      | 9.24** | –2.05 | 6.29**   | 92.1   | 0.690     |

\( X_1, X_2, \) and \( X_3 \) are the linear effect terms; \( X_1X_2, X_1X_3, \) and \( X_2X_3 \) are the interaction effect terms; and \( X_1^2, X_2^2, \) and \( X_3^2 \) are the quadratic effect terms; significance of model terms at 5%, 1%, and 0.1% are, respectively, indicated by "\*", "\*\*", and "\*\*\*".

![Image 1](image1.png)

Figure 1: Effect of coffee leaf extract and starch concentrations on chlorophyll a content.

![Image 2](image2.png)

Figure 2: Effect of coffee leaf extract and Gum Arabic concentration on chlorophyll b content.

![Image 3](image3.png)

Figure 3: Effect of starch and Gum Arabic concentrations on lycopene content of tomato fruits.

![Image 4](image4.png)

Figure 4: Effect of coffee leaf extract and starch concentrations on \( \beta \)-carotene content of tomato fruit.
3.2.4. Effects of Edible Coating on Physiological Loss of Mass. The control fruits had a higher mass loss (7.47%) compared to all treated fruit lots, whose physiological loss of mass ranged between 5.08 and 7.27% (Table 2). The lowest mass loss (5.08%) was recorded with the coating solution containing 66.66 g/l coffee leaf extract, 65.23 g/l starch and 10% GA. The physiological loss of mass was significantly influenced by the linear term of starch concentration ($p < 0.05$). On the other hand, the interaction between starch and Gum Arabic concentrations, as well as the quadratic terms of coffee leaf extract and starch concentrations significantly influenced this response ($p < 0.05$ and $p < 0.01$, respectively) (Table 3). The lack of fit was not significant ($p = 0.093$) and the coefficient of determination ($R^2$) was more than 84%, indicating a well fitted response model. Figure 6 shows that physiological loss of mass decreased with increasing starch concentration while it increased with increasing Gum Arabic. Compared to the results obtained with control fruits, coating with a combination of coffee leaf extract, starch, and Gum Arabic resulted in a decrease of the physiological loss of mass of tomato fruits. Similar increase of physiological loss of mass in response to increasing the concentration of Gum Arabic in coating solution has previously been reported by Sanchita et Hari [25]. Ali et al. [8] showed that edible coating formulation based on high concentration of Gum Arabic (15%) results in excessive mass loss of tomatoes. It follows from the results obtained that the effectiveness of Gum Arabic in controlling physiological weight loss depends on its concentration. Because at high concentrations, the physiological loss of mass increases. Evapotranspiration and respiration are the physiological phenomena that best explain the mass loss of fresh fruits during their ripening process [51]. Previous studies have shown that starch used as a coating material acted as a semipermeable barrier against gas ($CO_2$ and $O_2$) and moisture exchange in tomatoes, thus reducing respiration rate, mass loss, and oxidation [52, 53]. Water exchange between the indoor and outdoor environments is considered the main cause of postharvest mass loss of fruits. Gum Arabic behaves as a hydrophilic polysaccharide with the polar -OH groups and C-O covalent bond in their structure. However, the covalent bonds between starch and Gum Arabic could play an important role in forming a stabilizing coating macromolecule to improve their barrier property. Therefore, the coating solution containing medium concentrations would have decreased gas exchange thus slowing down the physiological loss of mass of tomato fruits.

3.2.5. Effects of Edible Coating on Firmness. The firmness of the control fruits was significantly lower (25.95 N) compared to that of the treated fruits, whose values ranged from 33.70 to 39.22 N. Table 3 shows that two linear terms (starch concentration and Gum Arabic concentration) significantly ($p < 0.01$) influenced firmness, as did the interaction between coffee leaf extract concentration and Gum Arabic concentration. The quadratic term of starch concentration also influenced firmness. Figure 7 indicates that fruit firmness increased as the concentration of coffee leaf extract increased and as the concentration of Gum Arabic decreased. Thus, the highest firmness was recorded when leaf extract concentration above 66.66 g/l was associated with low Gum Arabic concentrations. The lack of fit was not significant ($p = 0.22$) and the coefficient of determination ($R^2$) was more than 85%, indicating a well fitted response model. Compared to the control treatment, the coating formulations based on a combination of coffee leaf extract, starch, and Gum Arabic, slowed down the decrease in firmness of the tomato fruits. Similar results of reduction in firmness loss were reported by Hernandez-Guerrero et al. [54] with mangoes coated with 2% (w/v) starch. Firmness is
an important indicator to determine the degree of ripeness of the fruits. The ripening of the fruits leads to their softening. Degradation of cell structure, cell wall composition, and intracellular compounds lead to the softening of the fruit [55]. Since the firmness of treated fruits was high compared to control fruits, this indicates that the coating slowed down the degradation of cell wall compounds through hydrolysis of pectin. Hydrolysis also leads to softening by converting starch into sugar [56]. This biochemical process is due to the action of hydrolases, which are the enzymes that hydrolyze pectin and starch [8].

3.2.6. Effects of Edible Coating on the Percentage of Ripening.

The fruits of the control lot had ripened at 100% after 6 days of storage. The percentage of ripening of treated fruits ranged from 30.33 to 83.55% at day 14. The combined coating solution comprising 66.66 g/l coffee leaf extract, 65.23 g/l starch, and 10% Gum Arabic recorded the lowest percentage of ripening. Table 3 indicates that the linear terms of coffee leaf extract and Gum Arabic concentration significantly ($p < 0.05$ and $p < 0.01$, respectively) influenced the ripening rate. Similarly, coffee leaf extract and starch, as well as coffee leaf extract and Gum Arabic interactions also influenced this response. Only the quadratic term of starch concentration did not have a significant ($p > 0.05$) influence on the response. The percentage of ripening was low when the coffee leaf extract concentration was between 33.33 and 66.66 g/l. It decreased with increasing starch concentration (Figure 8). The lack of fit was not significant ($p = 0.690$) and the coefficient of determination ($R^2$) was more than 92%, indicating a well-fitted response model. The coating solutions improved the shelf life of tomato fruits compared to uncoated fruits. Generally, the ripening process of tomatoes harvested at the turning stage lasts 5 to 6 days. This short green life severely limits its long distance marketing [57]. The shelf life of the fruit is related to its ripening rate. Ripening encompasses a set of irreversible and unavoidable physiological processes that change the composition of the fruit [57]. The edible coating has barrier characteristics that reduce the permeability of the fruit surface to oxygen and carbon dioxide, leading to a change in the internal gas composition that in turn reduces oxidative metabolism and increases the shelf life of the fruit [57]. Several works have shown that coating fruits with Gum Arabic would result in significantly slower ripening rate and increased shelf life [6]. In addition, previous investigations by Donjio et al. [10] showed a significant effect of a combination of coatings to delaying the percentage of ripening of tomato fruits.

3.3. Optimization.

Chlorophyll $a$ content significantly varied with coffee leaf extract, starch, and Gum Arabic concentrations. The other parameters were mostly significantly influenced by starch and gum concentrations (Table 3).
Optimization was carried out by maximizing chlorophylls ($a$ and $b$) contents and firmness and minimizing lycopene and $\beta$-carotene contents, pH, physiological loss of mass, and percentage of ripening. The resulting optimal concentrations obtained for coffee leaf extract, starch, and Gum Arabic were 78.5 g/l, 56 g/l, and 9.5%, respectively, with a desirability of 0.71. A new coating formulation based on 78.5 g/l, 56 g/l, and 9.5% coffee leaf extract, starch, and Gum Arabic, respectively, was applied to tomatoes under the same conditions. All parameters were measured on the 14th day after storage. The experimental data obtained as a result of coating fruits with the optimal formulation as well as the predicted data are shown in Table 4. The results of the comparative analysis between the experimental and predicted data show that there was no significant difference at 5% probability level ($p < 0.05$).

### 4. Conclusions

The response surface methodology (RSM) has been successfully used to optimize an edible coating formulation based on coffee leaf extract, starch, and Gum Arabic to extend the shelf life or delay the ripening process of tomato fruits at ambient conditions. The optimum concentration of coffee leaf extract, starch, and Gum Arabic was predicted to be 78.5 g/l, 56 g/l, and 9.5%, respectively, which had satisfactory thickness and was effective in slowing down ripening and associated parameter progressions. The present study reports an edible coating formulation which is effective in extending shelf life, slowing the ripening process beyond 14 days compared to control fruits which ripened at 100% after 6 days of storage, with the onset of senescence at the 14th day.

### Data Availability

All data used to support the findings of this study are from previously reported studies and datasets, which have been cited in this manuscript. Furthermore, the processed data will be provided upon request.

### Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

References


