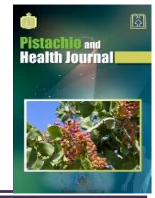

Optimization of the Pistachio Roasting Process with Hot Air Using the Response Surface Methodology

Ali Dini (PhD)[1*]

[1] *Pistachio safety Research Center, Rafsanjan University of Medical Sciences, Rafsanjan, Iran*

Information

Article Type:
Original Article

Article History:

Received: 08.08.2021
Accepted: 26.10.2021

Doi: 10.22123/PHJ.2021.298851.1107

Keywords:
Optimization
Hot air roasting process
Pistachio
Response Surface Methodology

Corresponding Author:
Ali Dini

Email: ali.dini2008@gmail.com

Tel: +98-9133920959

Abstract

Introduction: Roasting is a thermal process that increases the overall acceptance of edible nuts by physicochemical reactions during heating and reducing the moisture content. Optimizing and controlling the pistachio roasting process requires understanding the process scientifically. This study optimized the roasting process of pistachio nuts with hot air using the response surface methodology.

Materials and Methods: The response surface methodology was used to examine the effects of the roasting temperature, time, and hot airflow velocity on the roasted pistachios' physical (moisture content, and color parameteres), chemical (peroxide value, acidic value, and p-anisidine value), and sensory properties (overall acceptance, texture desirability, and crispiness). Twenty treatments with six replications were performed at the central point Based on the central composite design. the best quadratic model with the highest fit was determined using Design Expert 12 software.

Results: The temperature was the variable of the highest effect on physicochemical and sensory properties. Airflow velocity has a significant impact only on the parameters of moisture content, lightness (L-value), yellowness (b-value), and the peroxide value. The highest overall acceptance, texture desirability, and the lowest peroxide and p-anisidine value (p.AV) were considered optimal conditions for preparing the desired sample in the roasting process. The optimal sample production conditions included temperature 133.6 ºC, airflow velocity of 0.6 m/s, and roasting time of 23.6 minutes.

Conclusion: Comparison of predicted response values with laboratory ones showed that the model would be suitable for predicting response values in the pistachio roasting process and determining the optimal conditions.

►Please cite this article as follows:

Dini A. Optimization of the Pistachio Roasting Process with Hot Air Using the Response Surface Methodology. Pistachio and Health Journal. 2021; 4 (4): 4-27.

1. Introduction

Pistachios are cultivated in different parts of the world and countries, especially Iran, the USA, Turkey, and Syria, which amount to 500 to 600 thousand tons per year [1]. In addition to high popularity, it is of considerable nutritional and therapeutic value due to the presence of phytochemicals with antioxidant properties comparable to broccoli [2].

Roasting is one of the primary processes gone through in processing pistachios and edible nuts in the industry. This process improves these products' sensory and textural properties, thereby increasing marketability and the possibility of supplying them for consumption to the market [3]. This process increases the acceptance and delicacy of the nuts by physicochemical reactions occurring during heating in the edible nuts, which improves the taste, smell, color, and sensory properties [4]. The temperature and duration of the process are two critical factors that control the roasting process. When the temperature and duration of the process are not appropriately employed, the product's quality, shelf life, and flavor will be negatively affected. Therefore, it is necessary to predict the roasting conditions and study the changes in quality indicators such as color and moisture, chemical indicators, and sensory properties during the roasting process [5]. In addition to the variables mentioned above, the amounts of initial moisture and hot airflow velocity affect the rate of heat transfer, and physicochemical changes. Most physical and chemical changes during drying are affected by the amount of moisture and the rate of changes in this factor during the roasting process [6]. In the roasting process, when the temperature and roasting duration exceed a certain border, the heat transfer rate goes up accordingly, causing an excessive degradation of cell wall proteins and oleosomes. Subsequently, releasing fats inside oleosomal packages makes them vulnerable to oxidation during storage. Thus, increasing the temperature and process duration within a certain range initiates the destruction of oleosomes on a large scale. It decreases the product's shelf life by increasing the amount of peroxide value (PV) and decreasing sensory properties during storage [7].

The response surface methodology includes a set of statistical techniques used to optimize processes in which the desired response is affected by several variables. This technique enables the researcher to estimate the relationship between responses and independent variables. Besides, it is generally used to map the response (the dependent variable) in a three-dimensional manner within the specified range for independent variables, optimize the response, and decide the functional status of customer needs [8]. The graphical representation of the mathematical model has defined the term "response surface methodology." This statistical design reduces the number of experiments and makes it possible to estimate all coefficients of the quadratic regression model and interactions among factors. This analytical optimization tool is often used to improve product quality in food processes [8-10].

It is required to determine the optimal range of the temperature, process duration, and airflow velocity, i.e., the significant factors affecting the standard process of hot air roasting, to select a product with high chemical stability and durability, and maximum desirability in flavor, color, overall acceptance, as well as sensory and textural properties. Various studies have been conducted on mono-objective optimization of pistachios roasting with hot air by taking the two independent variables of temperature and process duration into account [11-14]. However, no study has been conducted on the multi-objective optimization process of roasting pistachios by considering the variables of temperature, process duration (roasting time), and airflow velocity. Against this backdrop, in this study, we intend to optimize the roasting process of pistachios with hot air by employing the response surface methodology and setting the variables of temperature at 120-170 °C, roasting time at 8-35 minutes, and airflow velocity at 0.6-2 m/s. This was realized by considering a sample with the lowest thermal degradation of oil (minimum PV and p.AV) and the highest overall acceptance, desirability of sensory factors, and color parameters.

2. Materials and Methods

Preparation

The Ahmad Aghaei variety of pistachios was prepared from Kashefan Kavir Rafsanjan Corporate (Doraj). Raw pistachio nuts were dried up to 4% then packed under vacuum and stored at 5 °C for further experiment. Before preparation, it was placed at the laboratory temperature for 24 hours, and after separating small and large seeds, pistachio seeds sized 24.5±0.5 per ounce, with an initial moisture content (MC) of 4% based on the dry matter, were salted using the Goktas Sihan method with some changes. The dried pistachios were primarily immersed in 17% saline for 8 minutes and drained for 4 minutes. Next, they were transferred to a laboratory roasting machine at 18.5-19.5% moisture content based on the dry matter [15].

Specifications of the Roasting System

The roasting system includes two air blowers, an anemometer for measuring hot air velocity, a heating section, a hot air flow conduction channel measuring (25 × 15 cm), and a steel cylinder with the speed drive rotating in hot air at 20 rpm (fig.1). A total of 250 g of the sample was placed inside the cylinder. The temperature and airflow velocity of the roasting system were adjusted using a Pt100 type heat sensor, with a 0.1 accuracy, and the external anemometer, respectively (Testo, Model400, UK). The average air humidity was 30%. The sample was tested at the airflow velocity of 0.6, 0.88, 1.3, 1.72 and 2 m/s and the temperatures of 120, 130, 145, 160, and 170 °C until it reached the specified time determined by the central composite design (CCD) technique. In addition, values of the independent variable were determined for each treatment by CCD based on Design-Expert software Ver.12.

Fig. 1. Scheme of the laboratory roasting machine and its components; (1) an air blower; (2) a damper; (3) the heating section and heating elements; (4) a steel duct; (5) a heat sensor; (6) a sample container; (7) a feed valve; (8) a control box; (9) a rotating motor

Moisture content Measurement

After turning on the device and stabilizing the hot air temperature for 15 minutes, the sample was transferred from the feed valve to the surface of the sample container in less than 4 seconds at the appropriate hot airflow velocity and temperature of each test. Besides, the lid of the feed valve was closed, and the moisture content changes were calculated according to Iran National Standard No. 672 [16].

Color Measurement

The imaging system was created in a dark room to prevent light and reflections from the environment. The system included a digital camera (Canon PowerShot SX60) placed vertically at a distance of 250 mm from the sample and connected to the computer with Canon remote recording software V5.0. The lighting system consisted of three illuminating lights (Opple, 8W, model: M X396-Y82; length: 60 cm), placed at an angle of 45 ^{o} to the camera. The skin of the roasted pistachios was removed, the pistachios were crushed and powdered, and images were recorded with the maximum resolution (1080×1920×1920). In addition, image segmentation was performed using Adobe Photoshop V7.0 to isolate the actual prototype image from the background. Next, the images were converted to L *, a *, and b values. In the color space of CIE L * a * b, coordinate L-value indicates color lightness (L = 0 is black, and L = 100 is white), coordinate a-value indicates the red/green position (negative values indicate green, while positive values indicate red), and coordinate b-value indicates the position between yellow and blue (negative values indicate blue and positive values indicate yellow). RGB images were converted to the color space of CIE L * a * b * using the "Color Space Converter" plugin in Image J (1.45s).

Measurement of the Peroxide value (PV)

The amount of PV was determined using the spectrophotometry method introduced by Shanta *et al*.[17]. Next, 0.01-0.03 g of the sample was mixed with 9.8 ml of a mixture of methanol and chloroform (3: 7 v/v) based on the probable amount of peroxides. Besides, 50 µl of a saturated solution of 30% ammonium thiocyanate and 50 µl of a solution of 0.018 mol/L of iron II chloride were added. After 5 minutes of dark storage at room temperature, it was read at 500 nm. The standard curve was prepared using a tetrazole solution of iron chloride III in a 3.7% hydrogen chloride solution. The working standard solution was performed using a stock solution (1040 µg/ml) in 0-25 µg/ml dilutions. In addition, the slope of the curve was 40.39, with the amount of the peroxide calculated based on the following equation [18]:

$$PV\ (\text{meq}/\text{kg of oil}) = \frac{\text{sample absorption} \times \text{the slope of the calibration curve}}{55.85 * \text{sample weight} * 2}$$

Measurement of oil Acid value (AV)

The AV is the amount of potassium hydroxide required to neutralize free fatty acids per gram of the fat, measured according to the AOCS Cd 3d-63 standard [19].

A total of 50 ml of neutralized ethanol was poured on 10 g of the oil sample. After mixing and heating, it was titrated with a 0.1 N potassium hydroxide solution in the presence of the phenolphthalein reagent following initial boiling. To approximate the amount of free fatty acid based on grams of oleic acid per 100 grams of oil, the acid value was multiplied by 0.503.

Measurement of the Para-Anisidine value (p.AV)

At the beginning, 0.2-1.6 g of the sample (m) was weighed in a 10 ml volumetric flask with an accuracy of 0.001 g and brought up to a volume with an isooctane solution. After zeroing the absorbance of the spectrophotometer using an isooctane solvent, the absorption of the diluted oil was read at a wavelength of 350 nm (Ab).

Next, 5 ml of the sample diluted with isooctane was placed in a 10 ml glass flask with a lid, and 1 ml of the p-anisidin reagent (2.5 g/l reagent in glacial acetic acid) was added. To zero the spectrophotometer in another glass flask, 5 ml of the isooctane solution and 1 ml of the p-anisidin reagent were added. Roughly after 10 minutes of darkening, sample absorbance was read and recorded (As).

The p-anisidine value was calculated using the following equation ;

$$p - A.V = \frac{10 \times (1.2A_s - A_b)}{m}$$

Where m is the sample weight, Ab represents the adsorption of the oil sample diluted in isooctane at the 350 nm wavelength, and As is the adsorption of the sample after the addition of the p-anisidin reagent [20].

sensory quality measurement of roasted Pistachios

Twelve trained panelists were invited to perform the sensory test. Despite the individuals having been experienced in evaluating the roasted pistachio product, necessary training was provided in rancid and bitter tastes by presenting an undesirable product and a fresh one. In addition, concepts such as crispiness (getting cracked by incisors), stiffness and crunchiness (getting cracked by molars), and their differences were taught. Besides, a quantitative descriptive method was employed for analysis, with each sensory index set linearly at 15 cm in size. Next, a bowl of roasted pistachios, a glass of water, and a scoring form were given to every evaluator. The panelists evaluated all the samples randomly and individually. Besides, they rinsed their mouth with lukewarm water after each stage of the diagnosis and removed the contents of their mouth. Additionally, the score of each test was extracted by measuring the line with a ruler [21].

Experimental Design and Statistical Analysis

The response surface methodology was used to examine the effects of the roasting temperature and process duration (time) and hot airflow velocity on the roasted pistachios' physical, chemical, and sensory properties. Twenty treatments with six replications were performed at the central point Based on the central composite design (Table 1). Design Expert 12 software was used for analysis. The following quadratic polynomial regression model was used to express the dependent variables as a function of the independent variables. In addition, the best quadratic model with the highest fit was determined using the backward elimination process, and non-significant coefficients were removed from the equation.

$$Y = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \beta_3 X_3 + \beta_1 X_1^2 + \beta_2 X_2^2 + \beta_3 X_3^2 + \beta_{12} X_1 X_2 + \beta_{13} X_1 X_3 + \beta_{23} X_2 X_3$$

To determine the optimal conditions of the roasting process, different scenarios were used in which the goal of optimization for one or more sensory, chemical and physical dependent variables, shown in Table 7, with a moderate (+++) or High degree of importance (+++++) was considered. Evaluation of differences between samples in each sensory factor was determined using Minitab 17. And all diagrams were drawn using Excel 2013

.

Table 1. The variables and values used for the central composite design (CCD)

No.	Uncoded			Coded		
	Temperature (°C)	Airflow velocity (m/s)	Time (min)	Temperature (°C)	Airflow velocity (m/s)	Time (min)
1	170	1.30	21.5	1.68	0	0
2	145	0.60	21.5	0	-1.68	0
3	160	1.72	13.5	1	1	-1
4	130	0.88	13.5	-1	-1	-1
5	145	2.00	21.5	0	1.68	0
6	160	0.88	13.5	1	-1	-1
7	145	1.30	21.5	0	0	0
8	130	1.72	29.5	-1	1	1
9	145	1.30	8.0	0	0	-1.68
10	145	1.30	21.5	0	0	0
11	130	0.88	29.5	-1	-1	1
12	145	1.30	21.5	0	0	0
13	145	1.30	21.5	0	0	0
14	145	1.30	35.0	0	0	1.68
15	120	1.30	21.5	-1.68	0	0
16	145	1.30	21.5	0	0	0
17	160	1.72	29.5	1	1	1
18	145	1.30	21.5	0	0	0
19	130	1.72	13.5	-1	1	-1
20	160	0.88	29.5	1	-1	1

3. Results and Discussion

Effects of Roasting Conditions on Physical Properties (MC and Color parameters)

Moisture content was determined using the response surface methodology by considering three factors of hot air temperature (120, 130, 145, 160, and 170), airflow velocity (0.6, 0.88, 1.3, 1.72, and 2 m/s), and roasting time (8, 13.47, 21.5, 29.53, and 35 minutes), with the results presented in Table 2. In addition, Table 3 shows the results of the analysis of variance used to examine the effect of the independent variables on the moisture content.

Table 2. Experimental data for the response of physical properties of pistachios concerning roasting conditions

No.	Temperature (°C)	Airflow velocity (m/s)	Time (min)	Moisture Content (%db)	Color Parameters		
					L*	b*	a*
1	120	1.3	21.5	1.89	54.46	44.25	-7.75
2	130	0.88	13.47	4.2	55.83	47.92	-9.96
3	130	1.72	13.47	2.85	55.27	45.62	-9.23
4	130	0.88	29.5	0.63	53.39	45.73	-7.36
5	130	1.72	29.5	0.218	52.83	44.33	-6.83
6	145	0.6	21.5	2.44	51.68	45.1	-0.76
7	145	2	21.5	0.32	49.59	43.83	0.50
8	145	1.3	8	5.46	56.87	48.00	-9.79
9	145	1.3	35	0.038	45.20	41.13	5.15
10	145	1.3	21.5	0.65	50.01	43.57	-0.05
11	145	1.3	21.5	0.30	48.95	44.75	-0.97
12	145	1.3	21.5	0.32	51.30	44.3	0.21
13	145	1.3	21.5	0.7	49.10	44.2	-0.79
14	145	1.3	21.5	0.9	50.64	43.57	-0.34
15	145	1.3	21.5	0.82	51.02	43.6	1.2
16	160	0.88	13.47	2.50	50.17	44.47	0.3
17	160	1.72	13.47	0.665	48.24	42.74	-0.27
18	160	0.88	29.5	0.085	38.72	37.20	10.6
19	160	1.72	29.5	0.001	36.79	34.92	11.31
20	170	1.3	21.5	0.041	36.21	32.25	13.8

According to the results of the analysis of variance of the regression model (non-coded values), changes in the moisture content and color parameters were obtained at a 5% significance level as follows:

MC = 29.29-0.1082X1-3.444X2-1.208X3 + 0.01018X32 + 0.00326X1X3 + 0.10074X2X3

L * = -111.11 + 2.343X1 - 1.495X2 + 2.303 X3 - 0.00794X12 - 0.0188X1X3

b * = = -141.243 + 2.63X1 - 1.73X2 + 1.489 X3 - 0.00892X12 - 0.01216X1X3

a * = 47.344 - 0.9788X1 - 1.3557 X3 + 0.00362X12- 0.0169X32 + 0.01764X1X3

Table 3. Analysis of variance (ANOVA) for the experimental variables as a linear (X_1, X_2, and X_3), quadratic ($X_{1,2,3}^2$), and interaction (X_1 $X_{2,3}$ and X_2X_3) terms of response variables (physical properties) and coefficients for the prediction models

Source (Coded)	Moisture Content %db		L-value		b-value		a-value	
	Coef.	p-value	Coef.	p-value	Coef.	p-value	Coef.	p-value
Intercept	0.8		50.51		44.42		-0.36	
Temperature (X_1)	-0.57	0.0002	-5.42	0.0108	-3.25	<0.0001	6.71	<0.0001
Airflow velocity (X_2)	-0.53	0.0003	-0.62	0.0068	-0.72	0.0023	-	-
Time (X_3)	-1.35	<0.0001	-3.47	<0.0001	-2.21	<0.0001	3.82	<0.0001
X_1 X_2	-	-	-	-	-	-	-	-
X_1 X_3	0.39	0.0185	-2.25	<0.0001	-1.45	<0.0001	2.10	0.0002
X_2 X_3	0.34	0.0366	-	-	-	-	-	-
$X_1{}^2$	-	-	-1.75	-	-1.97	<0.0001	0.8	0.0224
X_2^2	-	-	-	<0.0001	-	-	-	-
X_3^2	0.66	<0.0001	-	-	-	-	-1.09	0.0036
Model		<0.0001		<0.0001		<0.0001		<0.0001
Lack of Fit		0.0854		0.94		0.1490		0.1178
Mean		1.25		49.31		43.08		0.56-
Std. Dev.		0.41		0.73		0.71		1.19

*Coefficients were calculated to compare the effect of factors for coded variables.

Analysis of the experimental data showed that the effect of the roasting time, the temperature, and hot airflow velocity at $p < 0.01$ on reducing the moisture content was significant. The results of the response surface methodology were consistent with those of the kinetics of moisture changes in roasting pistachios in previous studies [3]. Fig. 2 shows the effect of each independent variable on the moisture content in the surface plates.

The roasted pistachios had a lower moisture content with increased hot air temperature and hot airflow velocity for the specified duration. The equation obtained from the central composite design for 8 to 35 minutes is a reasonable estimate of the values obtained from the kinetic equations of previous studies [3]; accordingly, the mean difference in the calculated moisture content was 0.39%, and the error range was minimally and maximally 0.0004 and 1.5%, respectively.

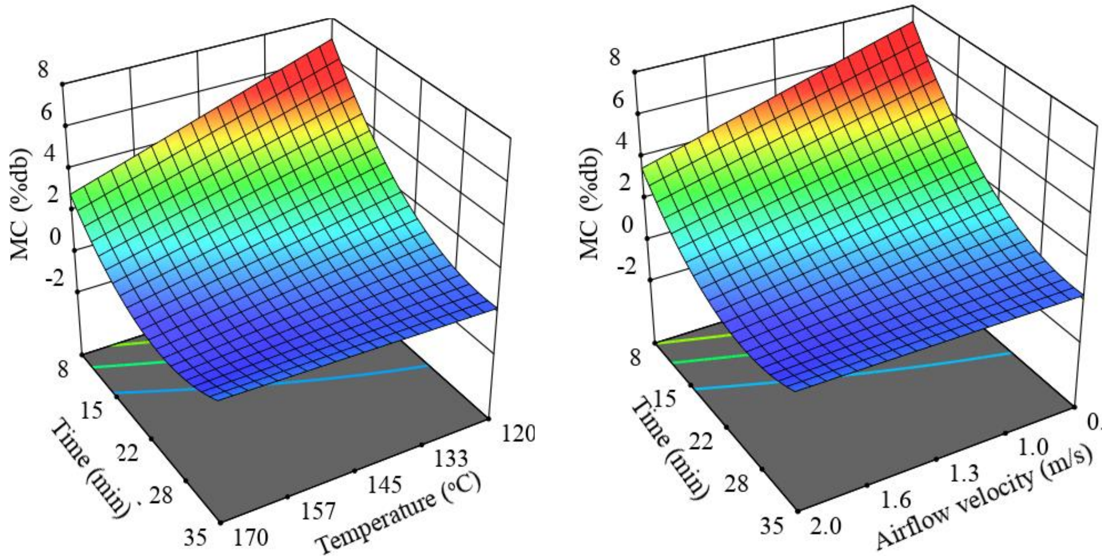

Fig. 2. Response surface plots of the effect of the temperature and airflow velocity on the MC during the roasting process

The effect of the roasting time and temperature (at $p < 0.0001$) and that of airflow velocity (at $p < 0.01$) were significant on the reduction in L and b values. However, the effect of airflow velocity was not significant on the a-value (Table 3). The results of the response surface methodology for L and b values were in line with those of the kinetic study of color changes in a previous study [22]. However, in terms of a-value, the effect of airflow velocity was insignificant, which could have been due to the different time range investigated compared with mentioned research [22], and negligible impacts of airflow velocity compared to other factors on changes in redness (a-value).

To examine the effect of each independent variable on color parameters, surface plots were drawn (Fig. 3). Accordingly, increasing the temperature and airflow velocity reduced the L and b values. Similarly, increasing the temperature increased the a-value. The equation obtained from the central composite design in 8 to 35 minutes is a reasonable estimate of L and b values similar to the kinetic equations used by Dini *et al.* (2019). So that the mean difference in the values obtained in the L and b values with the values calculated in the mentioned research were 0.3974 and 1.58, respectively, and the range of this difference in the L and b values were 0.0004-2.5 and 0.003-5.7, respectively.

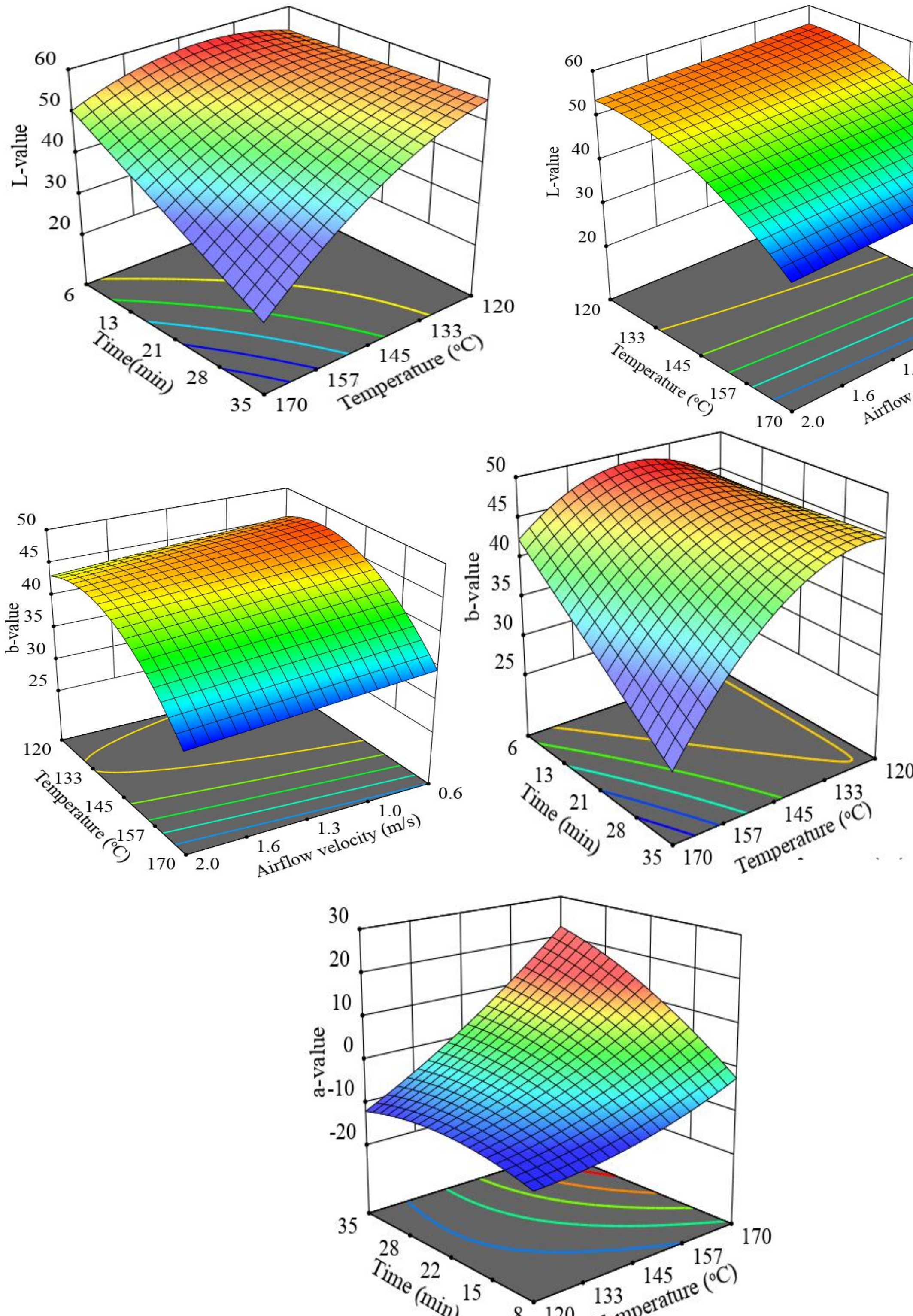

Fig. 3. Surface plots of the effect of the temperature and airflow velocity on changes in L, b, and a-value during the roasting process

Examination of the Effect of Roasting Conditions on Chemical Properties of Pistachio

Taste changes caused by the oxidation of pistachio oil are among the significant causes of pistachio spoilage during storage. Accordingly, the effects of roasting conditions on the PV, free fatty acids, and secondary oxidation compounds (investigation of the formation of compounds affecting the taste, especially aldehydes using the p.AV) were investigated under different roasting conditions using a central composite design.

The Effect of Roasting on the Peroxide value

Table 4 shows the analysis of the test results. Accordingly, the effects of the roasting time and temperature (at $p < 0.01$) as well as airflow velocity (at $p < 0.01$) were significant on the PV. Besides, increasing the roasting temperature and time increased the PV. Past research showed fluctuations in the PV during the roasting process, which has been indicative of oil degradation during the roasting process [23]. Azdmir *et al.* (2001) reported that roasting hazelnuts at high temperatures caused a significant increase in the PV [24]. Similarly, Kashani (1983) reported significant effects on the PV when roasting pistachios at 145ºC [25]. Increasing the temperature, time, and airflow velocity maximized the PV to 4.3 meq/kg. Accordingly, the PV increased rapidly at the beginning of the roasting process and decreased steadily after reaching the maximum value. Similar results were observed in the research by Tilury and Mille on heating linoleic acid at 60 °C [26]. In general, at temperatures above 100°C, peroxides are the primary oxidation compounds decomposing rapidly, getting converted into several volatile and non-volatile secondary compounds [27]. However, at higher temperatures, due to the faster decomposition of peroxides into secondary oxidation compounds, such as alcohols, saturated aldehydes, α,β-unsaturated aldehydes, and carbonyl components, the PV does not increase. Due to the transient nature of peroxide formation in the roasting process, the amount of this compound produced by oxidation in the final product did not exceed 4.3 meq/kg. This was similar to the results of the research by Yaacoub *et al.* on roasting edible nuts [28].

Table 4. Analysis of variance (ANOVA) for the experimental variables as a linear (X_1, X_2, and X_3), quadratic ($X_{1,2,3}^2$), and interaction (X_1 $X_{2,3}$ and X_2X_3) terms of response variables (chemical properties) and coefficients for the prediction models

Variables	AV		PV		p.AV	
	Coef.	p-value	Coef.	p-value	Coef.	p-value
Intercept	0.19		3.66		1.44	
Temperature (X_1)	0.00318	0.538	0.42	<0.0001	0.92	<0.0001
Airflow velocity (X_2)	0.00255	0.62	0.34	0.0003	-	0.09
Time (X_3)	0.00331	0.52	0.55	<0.0001	0.68	<0.0001
X_1 X_2	-	-	-	-	-	-
X_1 X_3	-		-0.18	0.065	0.41	0.0068
X_2 X_3	-	-	-	-	-	-
X_1^2	-0.013	0.019	-0.45	<0.0001	0.33	0.0040
X_2^2	-0.017	0.0049	-0.23	0.0046	-	-
X_3^2	-0.018	0.0028	-0.18	0.0193	-	-
Model		0.0095		<0.0001		<0.0001
Lack of fit		0.111		0.0534		0.068
Mean		0.15		3.07		1.67
Std. Dev.		0.019		0.25		0.37

Effects of the Roasting Process on the Acid value (AV)

The AV indicates the degree of hydrolytic intensity caused by enzymatic hydrolysis and spontaneity of triglycerides. Accordingly, the AV of the roasted pistachios was evaluated to be within the range of 0.11-0.19% in terms of oleic acid, being lower than the allowable value in the Iranian standard (<2.5%) [29]. Comparison of the means of the AV for the raw pistachios and roasted samples showed a significant difference between the AV of raw pistachios (0.8) and that of roasted pistachio. The AV was evaluated to be lower in roasted pistachios. It could be due to the conversion of free fatty acids into their oxidized forms, such as peroxides, in the oxidation reaction, with similar results reported for hazelnuts [24]. However, the results showed no significant relationship between roasting factors and the AV (Table 4).

The Effect of Roasting Factors on the p.AV

The experimental results showed that the roasting temperature and time (at $p < 0.01$) significantly changed the p.AV. Yet, airflow velocity had no significant effect on it. To examine the impact of each independent variable on the p.AV, a surface plot was drawn. As Fig. 4 shows, with an increase in the roasting temperature and time, the p.AV increased nonlinearly. The coefficient of the double effect of the temperature became significant. Besides, increasing the temperature and time accelerated the oxidation reaction and the production of oxidation by-products in roasted pistachios. The temperature increase on the p.AV was more than that of the time (Table 4). Increasing the temperature accelerates oxidation. It increases oxidation by-products in edible nuts due to the cumulative nature of high molecular weight aldehydes, observed by the p.AV, and over time, the amount of these secondary compounds increases [28]. In the end, the effect of airflow velocity on the amount of secondary compounds was not significant (p = 0.09). According to the above-described results of the changes in the chemical indices of oxidation of oil (the PV, AV, and p.AV) in edible nuts in high-temperature processes, the factors indicating secondary oxidation compounds (the p.AV) were better indices for evaluating the intensity of oxidation in the final products due to its cumulative nature.

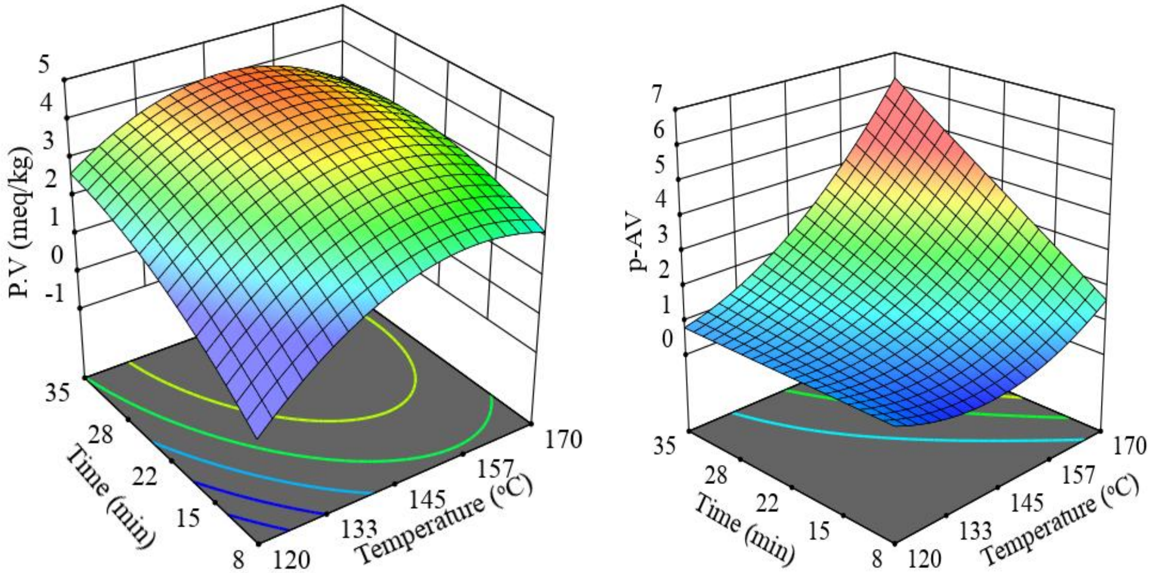

Fig. 4. Surface plots of the effect of the temperature on PV and p.AV during the pistachio roasting process with hot air

Examination of the Effect of Roasting Conditions on Sensory Properties

Table 5 presents the results of the scores given by the evaluators to the sensory properties of the samples. The panelists noticed a significant difference in the desirability of the color of the samples. As Fig. 5 shows, a meaningful relationship was observed between the scores given to the desirability of color and the parameters of burning, taste, and overall acceptance with a coefficient of determination (R^2) greater than 0.82. In the roasting process, an increase in the time and temperature increased the rate of changes in lightness (L-value) and yellowness (b-value). In addition, the development of the Maillard reactions led to the development of color changes [30, 31], while a decrease in the desirability of the color had an adverse effect on the taste and overall acceptance, according to panelists, with more intense burning having been reported.

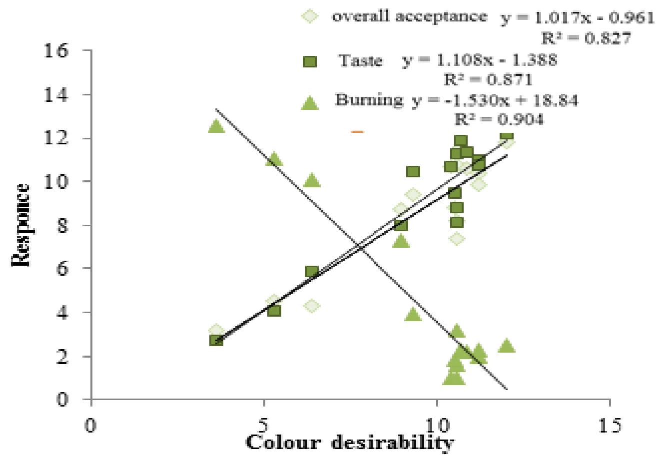

Fig. 5. The relationship between the color desirability factor with burning, taste, and overall acceptance in roasted pistachios, according to sensory evaluators

As Fig. 6, data analysis showed a significant relationship between taste and overall acceptance with a coefficient of determination of 0.945. During the roasting process, the temperature improved the textural properties of the nuts, with the aroma and flavor compounds intensified [32]. However, the continuation of the process caused extensive color changes, oil degradation, and color changes due to the formation of melanoidin pigments in pistachios, thereby reducing sugar and expanding the Maillard reaction [33]. Thus, the sweetness of samples decreased, and a positive correlation was observed between the desirability of the color and sweetness with a coefficient of determination of 0.7. Upon increasing melanoidins and creating a bitter taste, a reduction was observed in the desirability of taste, color, and overall acceptance in sensory evaluations.

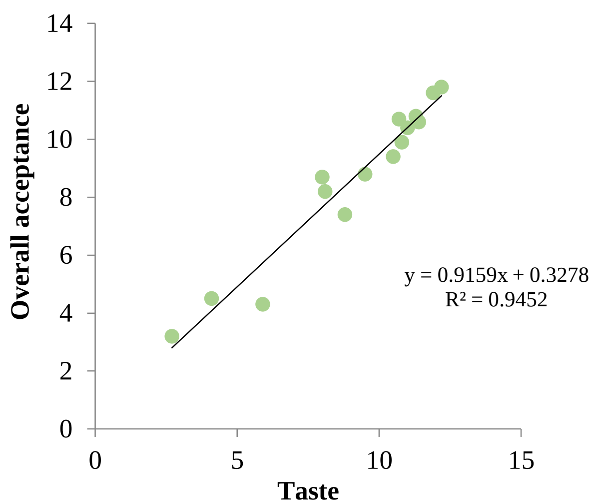

Fig. 6. The correlation between taste and overall acceptance in the evaluation of roasted pistachios

The sensory evaluation of the roasted samples showed no significant correlation between overall acceptance and crispiness. Yet, a correlation was observed between overall acceptance and texture desirability with a coefficient determination of 0.73. One of the reasons for the lack of correlation between crispiness and overall acceptance could be the fact that with increasing the roasting temperature and time due to reduced product moisture as well as the increased fragility and brittleness of the product [11, 34], the crispiness of the product increased; however, at high temperatures, the Maillard reaction and caramelization were intensified by expanding the process duration. Besides, despite the increase in the crispiness of the product, the taste of the product and the overall acceptance scores were less reported by the panelists.

In sensory evaluations, an inverse correlation was reported between hardness and crispiness with a coefficient of 0.9 (Fig. 7). The decrease in the initial hardness of the raw pistachios was observed parallel with the increase in their crispiness in the roasting process.

Table 5. Analysis of variance of the difference between the prepared samples based on the central composite design in the sensory properties

Independent Variable			Dependent Variable									
Duration (Time) (min)	Air Velocity (m/s)	Temperature (°C)	Color	Taste	Roasting flavor	Staleness	Burning	Crispiness	Sweetness	Hardness	Texture desirability	overall Acceptance
21.5	0.88	120	10.4^{AB}	7.10^{ABCD}	6.1^{FG}	1.6^{E}	1^{E}	5.7^{BC}	5.3^{A}	8.5^{ABC}	10.6^{ABCD}	10.7^{ABC}
13.5	0.88	130	10.6^{AB}	8.1^{CDE}	4.7^{G}	1.8^{DE}	1.6^{DE}	4.1^{C}	3.9^{ABCD}	10^{AB}	7.9^{CDE}	8.2^{CD}
29.5	0.88	130	11.2^{AB}	11^{AB}	8.4^{DEF}	2.1^{DE}	2^{CDE}	8.7^{AB}	4.1^{ABCD}	7.4^{BCD}	11.1^{ABC}	10.4^{ABCD}
13.5	1.72	130	10.5^{AB}	9.5^{ABCD}	6.9^{EFG}	2.5^{BCDE}	1.8^{CDE}	4.0^{C}	5^{AB}	8.8^{ABC}	8.7^{BCDE}	8.8^{ABCD}
29.5	1.72	130	11.2^{AB}	10.8^{ABC}	8.8^{CDE}	2^{DE}	2.3^{CDE}	6.7^{ABC}	4.2^{ABC}	7.4^{BCD}	9.4^{ABCDE}	9.9^{ABCD}
21.5	0.6	145	10.7^{AB}	11.9^{A}	10^{BCD}	1.4^{E}	2.3^{CDE}	9.6^{AB}	3.3^{ABCDE}	4.9^{DE}	11.8^{AB}	11.6^{A}
8	1.3	145	10.6^{AB}	8.8^{BCD}	6.2^{FG}	1.8^{DE}	1^{E}	3.6^{C}	4.9^{ABC}	10.4^{A}	6.8^{E}	7.4^{D}
21.5	1.3	145	12^{A}	12.2^{A}	10^{BCD}	2.3^{CDE}	2.5^{CDE}	9.9^{AB}	3.3^{ABCDE}	5.6^{DE}	12^{A}	11.8^{A}
35	1.3	145	9^{BC}	8^{DE}	10.3^{BCD}	4.7^{ABC}	7.3^{B}	8.3^{AB}	2.3^{CDEF}	5.6^{DE}	10.5^{ABCD}	8.7^{BCD}
21.5	2	145	9.3^{B}	10.5^{ABCD}	9.3^{CDE}	2.8^{BCDE}	3.9^{CD}	8.8^{AB}	2.6^{BCDEF}	4.9^{DE}	10.2^{ABCD}	9.4^{ABCD}
13.5	0.88	160	10.6^{A}	11.3^{AB}	8.3^{DEF}	2.6^{BCDE}	3.2^{CDE}	8.1^{AB}	3.2^{ABCDE}	6.9^{CD}	10.8^{ABCD}	10.8^{AB}
29.5	0.88	160	6.4^{CD}	5.9^{EF}	11.2^{ABC}	5.1^{AB}	10.1^{A}	10.3^{A}	1.4^{DEF}	3.9^{E}	7.7^{DE}	4.3^{E}
13.5	1.72	160	10.9^{A}	11.4^{AB}	10.1^{BCD}	2.5^{BCDE}	2.2^{CDE}	7.9^{AB}	4.7^{ABC}	7^{CD}	10.8^{ABCD}	10.6^{ABC}
29.5	1.72	160	5.3^{DE}	4.1^{FG}	13^{A}	5.9^{A}	11.1^{A}	10.3^{A}	1^{EF}	4^{E}	8.3^{CDE}	4.5^{E}
21.5	1.3	170	3.6^{E}	2.7^{G}	12^{AB}	4.3^{ABCD}	12.6^{A}	10.9^{A}	0.7^{F}	4^{E}	6.7^{E}	3.2^{E}
P-value			0.002	0.000	0.000	0.007	0.000	0.000	0.006	0.000	0.007	0.000

*Highlighted Latin numbers indicate a significant difference between the samples in a column.

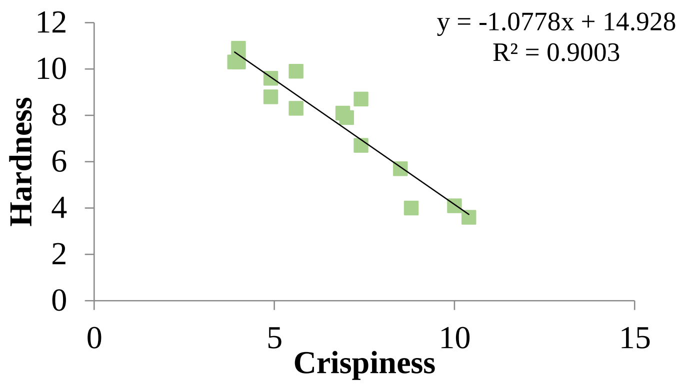

Fig. 7. The correlation between hardness and crispiness in the sensory evaluation of roasted pistachio samples

Data analysis showed that despite differences in the samples in terms of sensory properties, a statistical relationship was observed just between the parameters of the roasting process (i.e., temperature, roasting time, and airflow velocity) and sensory properties of overall acceptance, crispiness, and texture desirability.

Table 6 shows the statistical analysis and significance of the roasting process parameters in terms of crispiness, texture desirability, and overall acceptance. The roasting temperature and time had significant effects on sensory parameters (fig. 8). In contrast, airflow velocity had no considerable impact on sensory properties as evaluated by panelists.

Table 6. Statistical analysis and analysis of variance of changes in sensory properties for sensory evaluations of the roasted pistachios

Variable (Coded)	Texture desirability		Crispiness		Overall acceptance	
	Coef.	**p-value**	**Coef.**	**p-value**	**Coef.**	**p-value**
Intercept	11.73		10.03		10.96	
Temperature(X_1)	-0.44	0.1921	1.6	<0.0001	-1.45	0.0005
Airflow velocity (X_2)	-	-	-	-	-	-
Time (X_3)	0.33	0.3236	1.45	<0.0001	-0.51	0.013
X_1 X_2	-	-	-	-	-	-
X_1 X_3	-1.19	0.0140	-	-	-1.99	0.0003
X_2 X_3	-	-	-	-	-	-
X_1^{2}	-1.14	0.0026	-0.71	0.0023	-1.42	0.0004
X_2^2	-	-	-	-	-	-
X_3^2	-1.14	0.0026	-1.54	<0.0001	-1.05	0.0047
Model		0.0018		<0.0001		<0.0001
Lack of Fit		0.2358		0.4097		0.0595
Mean		10.17		8.49		9.28
Std. Dev.		1.2		0.74		1.19

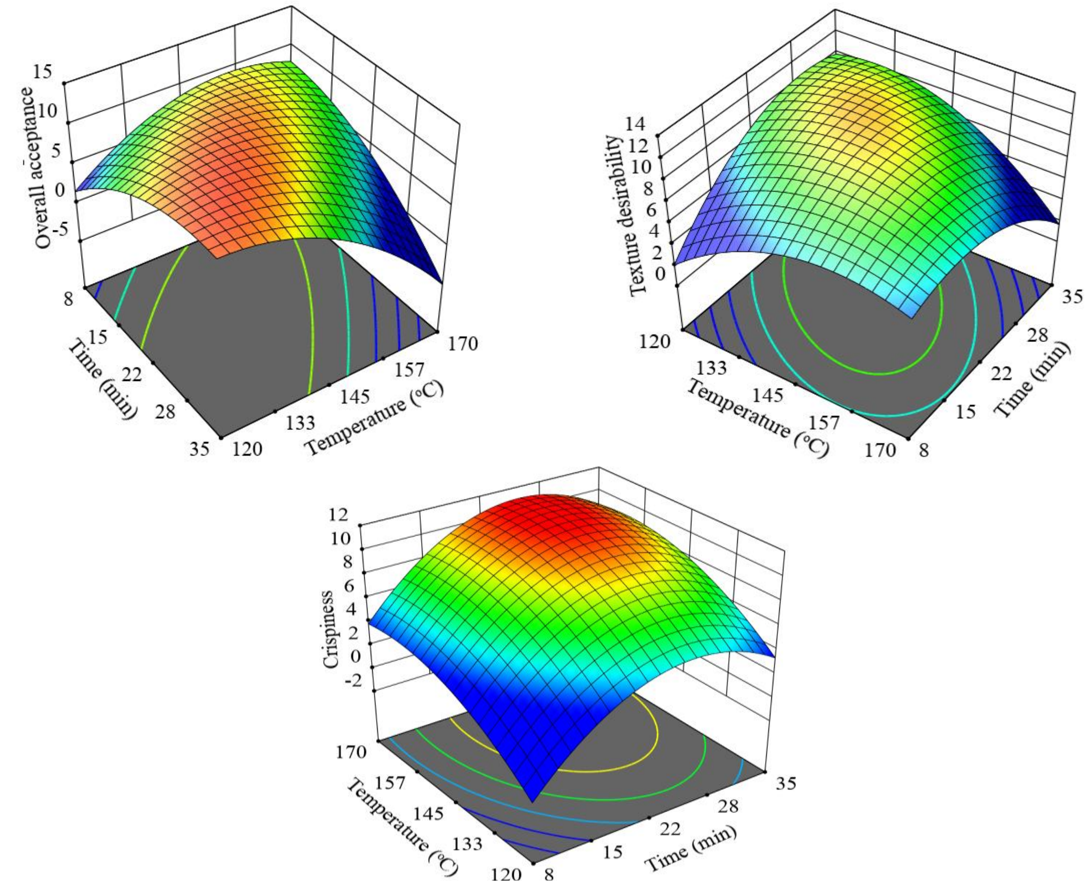

Fig. 8. Surface plots related to the effect of the temperature on the properties of A) sensory and overall acceptance, B) texture desirability, and C) crispiness during roasting pistachios with hot air

Optimization of the Process of Roasting Pistachios with Hot Air

One of the effective optimization factors is the determination of the appropriate range for dependent variables to optimize the process. These variables include physical and chemical factors as well as sensory properties and limitations specified in national and international standards (the maximum moisture content of roasted pistachios is 3%) [35]. The optimal conditions are determined based on independent variables. Organoleptic indices in roasted pistachios are of great importance. In this study, overall acceptance, texture desirability, and crispiness were taken into account. Chemical variables of PV and p.AV (with a significant effect on the product shelf life), as well as physical dependent variables, such as lightness (L-value), yellowness (b-value), and MC (the moisture content less than 3%), were considered crucial in optimization scenarios of the roasting process (Table 7). As illustrated in row 1, upon considering overall acceptance as the only sensory parameter for optimizing the roasting process, the following results were achieved: temperature 130 ^{o}C, air flow velocity 1.79, and the exposure time of 27.3 minutes. In row 2, optimization was performed taking into account all sensory factors. The optimization estimate showed that in this scenario, the highest temperature (138 ° C) was obtained compared to other scenarios. In rows 3 and 4, the chemical properties such as PV (with moderate importance) and p.AV (with higher importance) were also considered in estimating the optimization conditions. The least estimate roasting temperature was found to be for this scenario. These findings indicate that oxidation begins even in lower temperature during roasting process. In the optimization, lightness and yellowing were replaced by chemical factors due to the high correlation (<0.9) between this factor and p.AV (results not shown). In the scenarios shown in rows 8 and 9, the color parameters (lightness and yellowness) have been replaced by the chemical factors in estimating the optimal conditions of the roasting process. The results showed that the estimated optimal roasting temperature was higher than the previous scenarios. As reported by Dini et al. 2019 and 2017, the activation energy of color (L and b values) degradation is more than the activation energy of pistachio oil oxidation [22, 36]. As a result, the temperature dependence of color degradation has been higher than the oxidation of pistachio oil, so the onset of color changes provides the energy needed for oil oxidation and the roasting temperature was higher than the previous scenarios. Finally, in row 12, optimization scenarios were performed by considering all sensory, chemical, and physical factors as optimal samples. The optimal conditions for the roasting process were predicted 133.6, 0.6 m/s, 23.6 minutes for roasting temperature, airflow velocity, and roasting time, respectively. The optimal conditions obtained in roasting pistachios were used to produce laboratory samples, and the difference between the obtained responses and the values predicted in the model was evaluated. The difference in responses (dependent variables) in the produced laboratory sample compared to the predicted values in the coefficient of variation (CV) range was 1.3-24%, with the highest difference in PV and the lowest difference in L-value. The model is designed to predict the optimal roasting conditions except for the PV.

Table 7. Different scenarios to determine the optimal conditions of temperature, exposure time, and airflow velocity in optimizing the pistachio roasting process by considering the goal of optimization for one or more responses (dependent variable)

No. Scenarios	Independent Variables			Dependent Variables (Responses)																prediction of desirability
				Overall acceptance		L-value		b-value		p.aV		PV		Crispiness		Texture desirability		Moisture		
	Temperature (°C)	Air velocity (m/s)	Time (min)	Predicted value	Importance	Predicted value	Importance	Predicted value	Importance	Predicted value	Importance	Predicted value	Importance	Predicted value	Importance	Predicted value	Importance	Predicted value	Importance	
1	129.9	1.79	27.3	11.51	+++++	52.6	-	44.3	-	1.034	-	3.3	-	7.9	-	11.5	-	0.12	≤3%	0.914
2	137.8	0.9	24.1	11.36	+++++	52.5	-	45.7	-	1.23	-	3	-	9.4	+++	11.8	+++	1	≤3%	0.836
3	125.2	1.48	29.2	11.46	+++++	53.9	-	44.67	-	0.92	+++++	3	-	6.59	-	11.07	-	0.28	≤3%	0.869
4	121.5	0.6	31.1	11.35	+++++	55.8	-	45.9	-	0.8	+++++	1.4	+++	5.2	-	10.54	-	0.5	≤3%	0.88
5	130.3	0.6	25.7	11.48	+++++	54.5	-	46.5	-	0.997	+++++	1.91	+++	8.1	-	11.53	+++	1.23	≤3%	0.829
6	132.5	0.6	23.9	11.43	+++++	54.4	-	46.67	-	1	+++++	1.96	+++	8.48	+++	11.59	-	1.54	≤3%	0.818
7	134.6	0.6	23.8	11.45	+++++	53.9	-	46.6	-	1.07	+++++	2.1	+++	8.86	+++	11.72	+++	1.5	≤3%	0.81
8	136.3	0.6	24.1	11.42	+++++	53.4	+++	46.4	-	1.16	-	2.22	-	9.16	+++	11.81	+++	1.4	≤3%	0.833
9	136	0.6	23.8	11.43	+++++	53.5	+++	46.48	+++	1.13	-	2.19	-	9.11	+++	11.79	+++	1.46	≤3%	0.844
10	134.9	0.6	23.6	11.44	+++++	53.8	+++	46.6	+++	1.07	+++++	2.1	-	8.9	+++	11.7	+++	1.54	≤3%	0.834
11	134.6	0.6	23.8	11.44	+++++	53.9	+++	46.6	+++	1.07	-	2.1	+++	8.8	+++	11.7	+++	1.51	≤3%	0.833
12	133.6	0.6	23.6	11.43	+++++	54.2	+++	46.6	+++	1	+++++	2	+++	8.7	+++	11.65	+++	1.6	≤3%	0.828
Experimental value				12.4±0.52		53.5±0.35		47.8±0.5		0.92±0.1		2.66±0.15		9.5±0.45		11.1±0.4		1.8±0.2		
% Difference (CV)				7.82		1.3		2.5		8		24		8.4		4.7		11		

4. Conclusion

In this study, the effects of independent variables of temperature (120-170 °C), airflow velocity (0.6 -2 m/s), and roasting time (8-35 minutes) were examined on physical properties (MC and color parameters), chemical properties (PV, AV, and p.AV) and sensory properties (overall acceptance, crispiness, and texture desirability) of Ahmad Aghaei pistachios in the roasting process. The results showed that the hot air temperature, exposure time, and airflow velocity significantly affected MC and color parameters of lightness (L-value), yellowness (b-value), and the PV. However, airflow velocity had no considerable effect on redness (a-value), the p.AV, and sensory factors of crispiness, texture desirability, as well as overall acceptance. The independent variables had no significant effects on the AV. Besides, the temperature was evaluated as the most critical variable affecting the responses. Additionally, the roasting process was optimized by considering the highest overall acceptance, crispiness, and desirability of the texture as well as the minimum amount of PV and p.AV, and the highest amount of lightness and yellowness with a moisture content of less than 3% of the final product using the response surface methodology. The results showed that the best roasting conditions were a temperature of 133.6 °C, airflow velocity of 0.6, and process duration of 23.6 minutes. In addition, the comparison of the predicted values of the responses with the laboratory values showed that the model was effective in predicting the values of the responses in the process of roasting pistachios and determining the optimal conditions.

Acknowledgments

The author gratefully acknowledge Kashefane Kavir Agro Industries Co., and Rafsanjan University of Medical Sciences for providing raw pistachio nuts and the necessary equipment for this work, respectively.

Conflict of interest

The author declare that there is no conflict of interest.

Funding: This study is financially supported by Rafsanjan University of Medical Sciences (RUMS) under I.R.'s ethical code.RUMS.REC.1400.162 at RUMS.

References:

1- Panahi B, Khezri M. Effect of harvesting time on nut quality of pistachio (Pistacia vera L.) cultivars. Scientia horticulturae. **2011**;129(4):*730-4*.

2- Haytowitz DB, Bhagwat S. USDA database for the oxygen radical absorbance capacity (ORAC) of selected foods, Release 2. US Department of Agriculture. **2010**.

3- Dini A, Sedaghat N, Razavi S, Koocheki A, Malaekeh-Nikouei B. The Effect of Temperature and Air Velocity on Drying Kinetics of Pistachio Nuts during Roasting by using Hot Air Flow. Journal of Agricultural Machinery. **2017**;7(2):*401-14*. "Farsi"

4- Pittia P, Dalla Rosa M, Lerici C. Textural changes of coffee beans as affected by roasting conditions. LWT-Food Science and Technology. **2001**;34(3):*168-75*.

5- Bolek S, Ozdemir M. Optimization of roasting conditions of Pistacia terebinthus in a fluidized bed roaster. LWT-Food Science and Technology. **2017**;80:*67-75*.

6- Vega-Gálvez A, Di Scala K, Rodríguez K, Lemus-Mondaca R, Miranda M, López J, Perez-Won M. Effect of air-drying temperature on physico-chemical properties, antioxidant capacity, colour and total phenolic content of red pepper (Capsicum annuum, L. var. Hungarian). Food Chemistry. **2009**;117(4):*647-53*.

7- Dini A. The Effect of Tragacanth Edible Coating Containing α-Tocopherol on the Chemical and Sensory Properties of Roasted Pistachio Nuts During Storage. Pistachio and Health Journal. **2020**;3(2):*5-25*.

8- Chayjan RA, Kaveh M, Dibagar N, Nejad MZ. Optimization of pistachio nut drying in a fluidized bed dryer with microwave pretreatment applying response surface methodology. Chemical Product and Process Modeling. **2017**;12(3).:*1-15*.

9- Bagheri H, Kashainejad M, Aalami M, Ziaiifar A. Optimization of Hot Air Roasting of Peanut Kernels Using Response Surface Methodology. Journal of Food Technology and Nutrition. **2019**;16(2):*33-44*.

10- Rostami A, Sadrnia H, Khojastehpour M. Modeling and determination of optimized conditions of microwave pistachio (Ahmadaghaee cv.) thermal process using the response surface Methodology. Innovative Food Technologies. **2019**;6(2):*277-92*.

11- Shakerardekani A, Karim R, Ghazali HM, Chin N. Effect of roasting conditions on hardness, moisture content and colour of pistachio kernels. International Food Research Journal. **2011**;18:*704-10*.

12- Pumilia G, Cichon MJ, Cooperstone JL, Giuffrida D, Dugo G, Schwartz SJ. Changes in chlorophylls, chlorophyll degradation products and lutein in pistachio kernels (Pistacia vera L.) during roasting. Food Research International. **2014**;65:*193-8*.

13- Hojjati M, Calín-Sánchez Á, Razavi SH, Carbonell-Barrachina ÁA. Effect of roasting on colour and volatile composition of pistachios (Pistacia vera L.). International Journal of Food Science & Technology. **2013**;48(2):*437-43*.

14- Kahyaoglu T. Optimization of the pistachio nut roasting process using response surface methodology and gene expression programming. LWT-Food Science and Technology. **2008**;41(1):*26-33*.

15- Goktas Seyhan F. Effect of soaking on salting and moisture uptake of pistachio nuts (Pistachia vera L.) from Turkiye. GIDA. **2003**;28(4):*395-400*.

16- Institute of Standard and Industrial Research of Iran (ISIRI): Dry fruits –Determination of the moisture content- test methods, Standard No.672 [cited 2015]. Available from: http://standard.isiri.gov.ir/StandardView.aspx?Id=46679 "Farsi"

17- Shantha NC, Decker EA. Rapid, sensitive, iron-based spectrophotometric methods for determination of peroxide values of food lipids. Journal of AOAC International. **1994**(77):*421-4*.

18- IUPAC. IUPAC Standard Methods Determination of the peroxide value, method 2.501. IUPAC standard methods for the analysis of oils, fats and derivatives (7th ed.). . Oxford, UK, Alden Press. **1987a**:*199*.

19- AOCS. Official methods and recommended practices of the American Oil Chemists’ Society AOCS Press, Champaign; **1993**.

20- IUPAC. IUPAC Standard Methods, Determination of the p-anisidine value (p-A .V.) method Number2.504. IUPAC standard methods for the analysis of oils, fats andderivatives (7th ed.). . Oxford, UK, Alden Press. **1987b**:*143-4*.

21- Meilgaard MC, Carr BT, Civille GV. Sensory evaluation techniques: CRC press; **2006**.

22- Dini A, Khanamani Falahati-Pour S, Behmaram K, Sedaghat N. The kinetics of colour degradation, chlorophylls and xanthophylls loss in pistachio nuts during roasting process. Food Quality and Safety. **2019**;3(4):*251-63*.

23- Gardner H. Lipid hydroperoxide reactivity with proteins and amino acids: a review. Journal of Agricultural and Food Chemistry. **1979**;27(2):*220-9*.

24- Özdemir M, Açkurt F, Yildiz M, Biringen G, Gürcan T, Löker M. Effect of roasting on some nutrients of hazelnuts (Corylus Avellena L.). Food Chemistry. **2001**;73(2):*185-90*.

25- Kashani GG, Valadon L. Effect of salting and roasting on the lipids of Iranian pistachio kernels. International Journal of Food Science & Technology. **1983**;18(4):*461-7*.

26- Tilury K, Mille H. Chemiluminescence from the oxidation of model lipid systems. School of Chemical and Physical Sciences Victoria University of Wellington, PO Box. **2000**;600.

27- Frankel EN. Lipid oxidation: Elsevier; **2014**.

28- Yaacoub R, Saliba R, Nsouli B, Khalaf G, Birlouez-Aragon Is. Formation of lipid oxidation and isomerization products during processing of nuts and sesame seeds. Journal of agricultural and food chemistry. **2008**;56(16):*7082-90*.

29- Institute of Standard and Industrial Research of Iran (ISIRI): Edible fats and oils – Virgin pistachio oil. Standard No.6655 [cited 2002]. Available from: http://www.isiri.gov.ir/portal/files/std/6655.doc "Farsi"

30- Adhvaryu A, Erhan S, Liu Z, Perez J. Oxidation kinetic studies of oils derived from unmodified and genetically modified vegetables using pressurized differential scanning calorimetry and nuclear magnetic resonance spectroscopy. Thermochimica Acta. **2000**;364(1-2):*87-97*.

31- Wang X, Lim L-T. A kinetics and modeling study of coffee roasting under isothermal conditions. Food and bioprocess technology. **2014**;7(3):*621-32*.

32- Bagheri H. Application of infrared heating for roasting nuts. Journal of Food Quality. **2020**;2020.

33- Severini C, Gomes T, De Pilli T, Romani S, Massini R. Autoxidation of packed almonds as affected by Maillard reaction volatile compounds derived from roasting. Journal of agricultural and food chemistry. **2000**;48(10):*4635-40*.

34- Ahmed ZB, Yousfi M, Viaene J, Dejaegher B, Demeyer K, Vander Heyden Y. Four Pistacia atlantica subspecies (atlantica, cabulica, kurdica and mutica): A review of their botany, ethnobotany, phytochemistry and pharmacology. Journal of Ethnopharmacology. **2020**; 265: *113329*.

35- Malekjani N, Jafari SM. Food process modeling and optimization by response surface methodology (RSM). Mathematical and statistical applications in food engineering: CRC Press; **2020**. *p. 181-203*.

36- Dini A, Farrokhi H, Sedaghat N, Bagheri M, Mohammadkhani N. Evolution of Oxidative stability and Shelf-life prediction of pistachio oil using Rancimat. Journal of Rafsanjan University of Medical Sciences. **2016**;15(5): *399-412*. "Farsi"