

Kinetics of color degradation of chestnut kernel during thermal treatment and storage

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Abstract: Thermal treatments are commonly used for disinfesting chestnuts before storage. Color is a first and sensitive quality attribute of chestnuts after thermal treatments and storage. The purpose of this study was to investigate L^* and b^* values in chestnuts during thermal treatment at 50-70°C for periods varying from 0 to 160 min, and during storage at 3°C for 5 months using a computer vision system (CVS). Results showed that the L^* and b^* values decreased with increasing temperature, and heating or storage time. The first-order reaction model showed a better fit for the L^* and b^* values than zero-order reaction model with coefficients of determination (R^2) ranging from 0.923 to 0.977 during the thermal treatment and storage. The activation energies of chestnuts were 287.19 and 347.48 kJ/mol during thermal treatments and 89.18 and 78.47 kJ/mol during storage for L^* and b^* values, respectively, suggesting that the yellowness (b^*) was more sensitive to temperature changes than the lightness (L^*). According to requirements of $L^* > 50$, a potential disinfestation treatment protocol could be developed for chestnuts at 52-62°C.

Keywords: color degradation, kinetics, thermal treatment, storage, chestnuts

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1 Introduction

Chestnut is one of popular nuts in the world. In 2012, the production of chestnuts was 1 650 000, 70 000, 59 789, 57 000 and 52 000 MT in China, Korea, Turkey, Bolivia and Italy, respectively^[1]. During the harvesting, processing and long-time storage, insect infestations are the major problems of chestnuts. Losses caused by insects in chestnut include reduced nutrition, low germination rate, weight reduction, and reduced market price of chestnut^[2]. It is estimated that the annual losses

of chestnuts due to insects is about 20% in China^[3]. Therefore, developing an effective and efficient disinfestation method can reduce insect damages to chestnuts. Thermal treatments, such as hot air, hot water and radio frequency heating, are proposed to disinfest chestnuts and other nuts while maintaining the products quality^[4-8].

Except for flavor, texture and nutrition, color is one of the most important sensory quality attribute of chestnuts related to quality change^[9]. Color provides the basic quality information for human perception, and directly influences consumer's acceptance and preference^[10]. Color has been measured in the laboratory and food industry by visual inspection, colorimeter, and computer vision system. The visual inspection has been commonly used for color measurement in many fields since it is easy and cheap to apply^[11]. Because the color results depend on judgments of the various inspectors, the visual inspection is not suitable for large-scale color measurements^[12]. Colorimeter has been extensively used in the food industry due to its rapid and simple measurement of the

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food color^[13,14]. But the colorimeter is only suitable to measure uniform and small samples^[10]. Therefore, the computer vision system has been developed to replace the visual inspection and colorimeter and to provide automatic pixel-based color measurement with rapid, non-contact, objective and economic characteristics. In recent years, the computer vision system has been used for objectively measuring the color of almond^[4], bean^[5], beef^[15], Guava salmon muscle^[16] and wheat kernel^[17]. The computer vision could be an appropriate method to evaluate the chestnut color related to quality change.

Since the color of chestnuts is sensitive to heat^[3,18,19], it is critical to obtain the thermal kinetics of chestnut color changes during thermal treatment and storage, and determine the up limit of temperature-time range for developing an effective thermal treatment protocol. The important value of kinetic models is to predict quality changes inside and outside the test temperature range and optimize thermal processes under given non-isothermal conditions. Various kinetic models are used to describe the color changes in fruits and vegetables during thermal treatment and storage, such as apple^[20], carrot^[21], rice^[21], jackfruit^[22], hazelnuts^[23], pepper^[24], pineapple puree^[25], rocket puree^[26], tomato^[14] and wheat germ^[27]. Most kinetic models of color degradation follow zero- or first-order kinetic models^[14,28,29], except for some following second-order kinetic model^[30], and fractional conversion model or combined kinetic model^[13,29]. Knowledge of color kinetics is essential to define the chestnut quality change during thermal treatments.

The objectives of this study were (1) to determine color degradation in chestnuts in a temperature range of 50-70°C using a test cell and develop kinetic models for the color degradation during thermal treatment, (2) to determine color degradation in chestnuts during cold storage at 3°C for 5 months with kinetic analysis, (3) to compare the color kinetic changes between chestnuts and other fruits, grains or nuts after thermal treatments, and (4) to guide the thermal treatment protocol development of chestnuts for disinfestations.

2 Materials and methods

2.1 Materials

Fresh chestnuts (variety *Sanyang*) were harvested

from Jingshan County, Hubei, China in September, 2013 and used for this study thereafter. The initial moisture content and individual weight of chestnut kernels were 51.27%±1.19% in wet basis and (16.22±1.19) g, respectively. The chestnut samples were stored with mesh bags in a refrigerator (BD/BC-297KMQ, Midea Refrigeration Division, Hefei, China) at (3±1)°C. For each experiment, the required amount of chestnuts was taken out from the refrigerator and kept at ambient temperature (20±1)°C for 24 h until equilibrium. The chestnuts were shelled manually and kernels without defects were selected for further thermal treatments.

2.2 Thermal treatment

To minimize the influences of slow heat transfer on sample color during heating up time, a kernel sample was sliced into a piece with thickness of 4 mm and diameter in 18 mm, which was just fitted into the internal volume of a custom-designed test cell (Figure 1). The test cell was made by two pieces of high thermal conductivity aluminum alloy, which provided a high heating rate and quick cooling, and easily loading or unloading. A screwed-on bottom part with a rubber O-ring provided a hermetic seal to maintain the moisture contents of the tested sample. The detailed description of the test cell can be found elsewhere^[31]. The core temperature of the sample was measured using a thin Type-T thermocouple (TMQSS-020-6, Omega Engineering Ltd., CT, USA) and recorded every 10 s by a data logger (CR1000, Campbell Scientific Inc., Logan, Utah, USA). The similar test cells have been successfully used to study quality changes of cashew apples^[32], blackberry juice^[33], blue mussel^[34], salmon muscle^[16] and walnut^[35].

Based on disinfestation requirements from the literature^[8,36], and possible quality changes of chestnuts from preliminary tests, temperatures between 50°C and 70°C with intervals of 5°C were selected for thermal treatments. To determine the come-up time and cooling time, the test cells were sealed and heated in a water bath (SC-15, Scientz Biotechnology, Ningbo, China) at the selected five temperatures. When reaching the set point, the sample in the test cell was maintained for 0-160 min and then cooled the cell in a cold water (about 5-7°C) until the sample temperature was below 20°C. The tests were repeated two times.

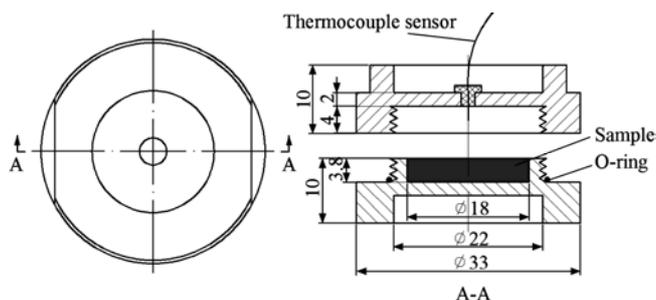


Figure 1 Schematic view of a test cell with a disk cavity of 18 mm diameter and 3.8 mm height (All dimensions are in mm)

For thermal treatments, 3 samples in the test cells were heated at each temperature and taken out after holding for the predetermined time indicated in Table 1. After cooling, the samples were removed from the cell for color measurements. All the thermal treatments were replicated three times.

Table 1 Effect of heat treatment on the L^* and b^* values of chestnut kernels

Temperature/°C	Time/min	L^*^a	b^*^b
50	0	81.95±1.83	54.26±1.66
	5	77.09±2.67	53.90±1.75
	10	77.06±2.01	53.87±4.97
	20	76.86±4.27	53.78±1.52
	40	76.03±1.05	53.62±1.40
	80	75.71±4.35	53.44±4.35
	160	71.84±3.85	53.12±2.30
55	0	82.37±1.64	57.60±1.01
	5	78.13±4.31	53.61±3.15
	10	77.98±2.56	53.11±3.88
	20	75.60±4.93	51.27±2.31
	40	61.23±5.58	46.48±3.17
	80	57.53±6.63	43.66±2.21
	160	45.29±5.84	35.45±1.37
60	0	78.44±1.07	55.45±1.48
	5	65.90±3.82	47.27±1.80
	10	44.77±0.99	35.83±1.04
	15	39.53±3.07	31.31±2.24
	20	32.63±2.88	19.77±4.13
	30	26.46±4.28	13.77±1.47
65	0	70.47±1.37	56.14±3.04
	2	56.19±0.39	43.62±3.58
	4	45.76±5.13	39.17±3.51
	6	34.33±2.30	29.54±1.84
	8	28.99±2.54	25.75±2.99
	10	26.07±5.71	23.78±2.88
70	0	70.47±1.37	56.14±3.04
	1	51.31±1.84	41.59±0.22
	2	49.37±2.89	39.69±2.70
	3	40.99±0.34	31.87±2.23
	4	30.67±0.40	27.38±2.24
	5	26.53±3.70	21.85±0.33

Note: ^a L^* -Value: 0 = black and 100 = white; ^b b^* -Value: -128 = blue and 128 = yellow.

2.3 Storage test

The fresh chestnut samples (10 kg) were divided into sub samples of 150 g, packaged in woven bags, and kept in the refrigerator for 5 months at 3°C for storage experiments. Each sample was taken out each month and shelled to determine color parameters. Three replicates were made for each experiment.

2.4 Evaluation of color

Since the color of fresh chestnut kernels is yellow, and the lightness has been associated with browning in fruit^[29], L^* and b^* values were only considered as color degradation parameters in this study. Similar considerations are taken for color measurements in cashew apple^[32] and tomato^[14].

Surface color of chestnut kernels was measured with a similar computer vision system (CVS) (Figure 2) setup as described in elsewhere^[37]. The CVS included three parts: an illumination device, a Cannon EOS 600D Digital camera, with 18 to 55 mm zoom lens and 1800 megapixel resolution, and a Lenovo notebook computer with image-processing software.

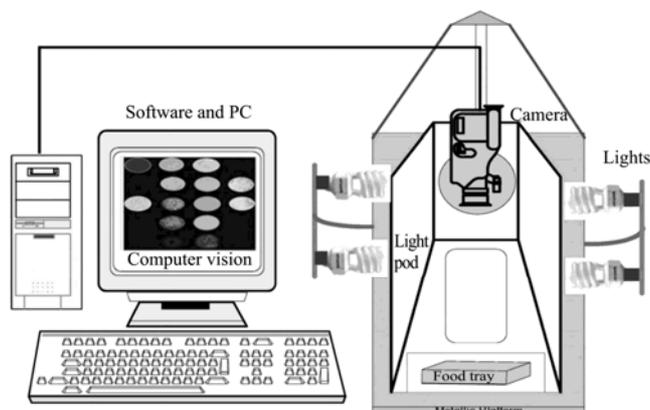


Figure 2 Schematic view of computer vision system^[33]

The illumination device consisted of a shooting tent (41 cm×30 cm×30 cm) and photography lamps (LS235 5500K) on the 2 top sides of the shooting tent as lighting sources. The lights were turned on at least 15 min before the pictures were taken. Samples were placed on the bottom of the shooting tent. The digital camera was fixed on a mount and downward on the top of the shooting tent. Based on the operational process suggested by others^[16], color images of chestnut kernels surface were obtained by the camera, stored in the computer, and analyzed by Adobe Photoshop CS3

(Adobe Systems Inc., USA). The color values acquired from Photoshop (L and b) were converted to CIE LAB (L^* and b^*) values using the following equations^[16]:

$$L^* = \frac{L}{2.5} \quad (1)$$

$$b^* = \frac{240}{255}b - 120 \quad (2)$$

2.5 Kinetic models

Since the zero- and first-order kinetic models have been used to evaluate the appearance of browning^[29,32,38], these two kinetic models were applied first to fit the experimental data using isothermal tests and obtain the coefficient of determination (R^2) using the following:

$$P = P_0 - k_0 t \quad (3)$$

$$\ln P = \ln P_0 - k_1 t \quad (4)$$

where, P_0 and P are the initial and the measured color values, respectively; t is the heating time (min) or storage time (month); k_0 and k_1 are the zero- and first-order kinetic constants (1/min), respectively.

In the second step, the Arrhenius equation was used to quantify the dependence of the reaction rate constant (k_T) on temperature (k - T curve) by the following^[25]:

$$k_T = A_0 \exp\left(-\frac{E_a}{RT}\right) \quad (5)$$

where, A_0 is the pre-exponential factor (1/min); E_a is activation energy (J/mol); R is the gas constant (8.314 J/mol·K), and T is absolute temperature (K). The E_a value can be calculated from the slope of the experimentally developed k - T curve by Equation (5).

The average coefficient of determination over the entire temperature range was used as the basis to select the better kinetic model. For the first order reaction, two key parameters (D and z values) were then determined from the values of color change curves. The D -value (min) represents a heating time to reduce the color values by 90% at a constant temperature and can be calculated by the rate constant k as follows:

$$D = \frac{2.303}{k_1} \quad (6)$$

By plotting log D -values against temperature often results in a linear relationship, commonly referred to as quality degradation time curve. A z -value ($^{\circ}\text{C}$) was

obtained as the temperature increase needed to achieve a reduction in D -values as follows:

$$z = \frac{T_2 - T_1}{\log D_1 - \log D_2} \quad (7)$$

where, D_1 and D_2 are D -values at temperatures T_1 and T_2 , respectively. The z -value was also obtained by the $-1/\text{slope}$ of the regression equation of the log D -values against temperature (D - T curve). If the z -value is determined over a temperature range (T_1 to T_2), the activation energy can be also calculated as follows^[39]:

$$E_a = \frac{2.303 R T_1 T_2}{z} \quad (8)$$

2.6 Statistical analysis

The experimental results were expressed as mean \pm standard deviation of three replicates and the linear regressions were established using the Microsoft Excel 2007.

3 Results and discussion

3.1 Temperatures-time histories of chestnut samples during heating and cooling

Figure 3 shows a measured temperature-time history of the chestnut sample during thermal treatment. The results indicate that the temperature at the center of the sample reached within 0.5°C of each set-point temperature after 120 s of heating and within 20°C after 30 s of cooling. The similar come-up time was observed by other researchers^[32,40]. The rapid heating and cooling using the test cell resulted in isothermal treatments and reduced possible quality degradation during come-up period and cooling processing.

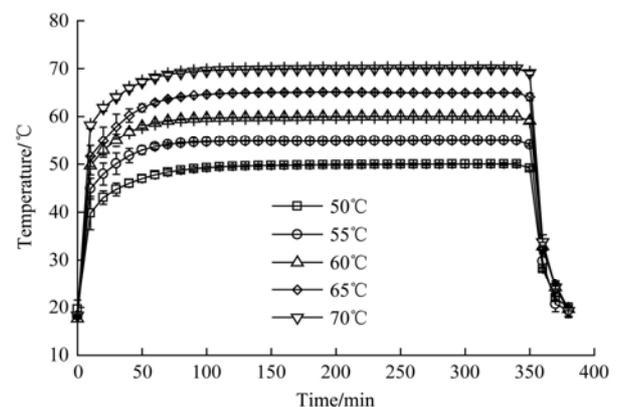


Figure 3 Typical temperature-time history at the core of a chestnut samples sealed in the test cell

3.2 Effect of temperature and time on color of chestnut kernels during thermal treatment

Table 1 shows the effect of temperature and holding time on the L^* and b^* values of chestnut kernel. The results indicated that there was a consistent decrease in L^* and b^* values with an increase both in processing time and heating temperature. For example, the color of chestnuts changed slowly during thermal treatment at 50°C (Figure 4), and clearly became dark (brown) since L^* value dropped down to about 26 at 60, 65 and 70°C

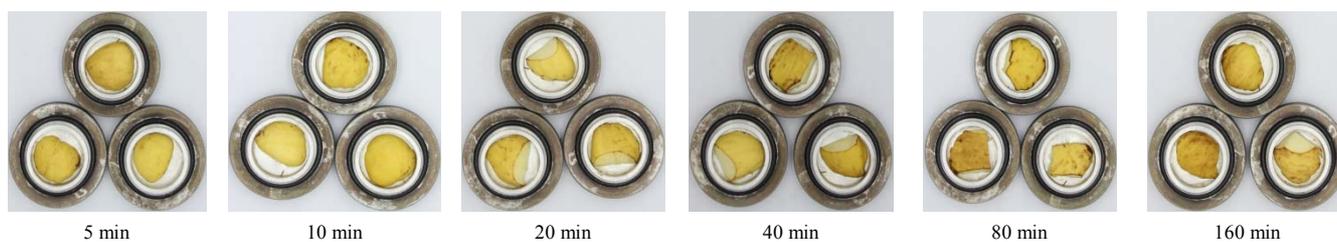


Figure 4 The change of chestnuts color during thermal treatment at 50°C

3.3 Color degradation kinetics of chestnut kernels during thermal treatment

Using the linear regression, color degradation data were analyzed by Equations (3) and (4) to obtain the reaction rate constant (k) and the coefficient of determination (R^2). As compared to zero order reaction model ($R^2=0.906$), the first order reaction kinetic model was better to fit the color data with the average $R^2=0.946$ for L^* and $R^2=0.972$ for b^* over the five temperatures. Table 2 shows the first order reaction rate constants, D -values and coefficients of determination for color degradation of L^* and b^* values of chestnuts at 50, 55, 60,

for 30, 10 and 5 min, respectively (Table 1). The same phenomenon was reported on air-dried chestnuts^[41]. The change in L^* and b^* values may be due to non-enzymatic browning or Millard reaction^[29]. The similar degradation was also found in the b^* values (Table 1). Pineapple puree and tomato puree showed the same color changes during thermal processing^[29]. High rate of color degradation suggested great heat sensitivity of chestnuts, especially at high temperatures.

65 and 70°C. The fitted model for b^* was slightly better than that for L^* . The reaction rate constants increased with increasing temperatures while the D -values decreased with increasing temperatures (Table 2), indicating that high temperatures accelerated the loss of lightness and yellowness^[29]. The D -value for b^* was larger than that for L^* , suggesting that L^* should be evaluated first in heat treated chestnuts. These results were in good agreement with those reported for cashew apples^[32], grapefruit juice^[42], peach puree^[29], pineapple puree^[25], rocket puree^[18], and tomato paste^[14, 43].

Table 2 The first order reaction rate constant k , correlation coefficient (R^2), D -values, and activation energy for color degradation in chestnut kernels after thermal treatment

Temperature/°C		50	55	60	65	70
L^*	k/min^{-1}	0.00044±0.00001	0.0036±0.0002	0.034±0.003	0.099±0.001	0.179±0.010
	R^2	0.949	0.923	0.927	0.975	0.951
	D/min	5234.09	767.67	67.74	23.26	12.87
	$E_a/\text{kJ}\cdot\text{mol}^{-1}$	D - T curve	277.00±7.99			
		k - T curve	287.19±7.39			
b^*	k/min^{-1}	0.00009±0.00001	0.0027±0.0012	0.051±0.006	0.081±0.008	0.165±0.084
	R^2	0.977	0.976	0.976	0.961	0.968
	D/min	25588.89	1151.50	46.06	28.43	13.96
	$E_a/(\text{kJ}\cdot\text{mol}^{-1})$	D - T curve	346.42±4.09			
		k - T curve	347.48±2.61			

Activation energy is important for developing effective heat treatments because it is useful in determining sensitivity of color degradation to

temperature changes. By plotting the $\log k$ value versus $1/T$ (Figure 5), the rate constant decreased proportionally with $1/T$ both for L^* and b^* . The Arrhenius plot (k - T

curve) showed that the regression line for L^* ($R^2=0.953$) was better fitted than that for b^* ($R^2=0.896$). Table 2 shows the activation energy for the chestnuts estimated both by the k - T curve (Equation (5)) and D - T curve (Equation (8)). The difference in activation energy obtained these two methods was insignificant both for color parameters of L^* and b^* . The activation energy of b^* was higher than that of L^* , indicating that the sensitivity to temperature changes of b^* was higher than that of L^* in chestnut kernels.

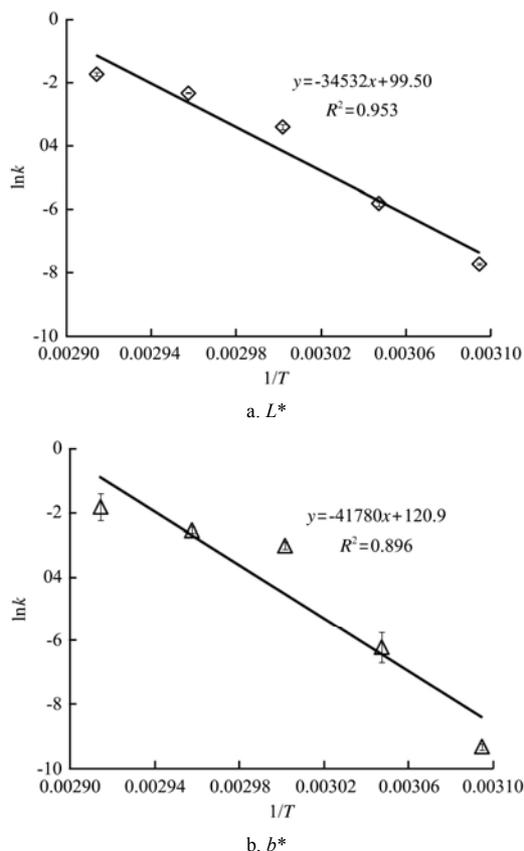


Figure 5 Arrhenius plot for color degradation (L^* and b^*) in chestnut kernels

3.4 Comparison of color kinetic changes between chestnuts and other fruits during thermal treatment

Table 3 shows the comparison of color kinetic parameters of chestnut kernels with those of various fruits reported in the literature. Most of color kinetics followed the first order reaction. Generally, quality changes are not sensitive to temperature changes with large z values ($>20^\circ\text{C}$) and low activation energy (<130 kJ/mol). The z value (6.1°C for b^*) of chestnuts color in this study was extremely low as compared to that of other fruits at relative low temperature (Table 3) but higher than that of insects, such as Indianmeal moth

(3.9°C). Similarly, activation energy (347.48 kJ/mol) of chestnuts color in this study was extremely higher than that of other fruits and nuts but smaller than that of insects, such as Indianmeal moth (513.7 kJ/mol). The comparison results suggested that the color of chestnuts could be more sensitive than that of the listed fruits but still tolerant than the insects when subjected to the same thermal treatments. More cares need to be taken for thermal disinfestations in chestnuts than other fruits and potential treatment protocol could be developed based on the color changes and control of insects.

3.5 Color degradation profile and kinetics of chestnut kernels during storage

Figure 6 shows that the L^* and b^* values decreased with increasing storage time. The similar trend was reported for the L^* and b^* values of chestnuts during storage^[2]. The first order kinetic models were fitted well for the degradation of lightness (L^* value) and yellowness (b^* values) for chestnuts during storage since the coefficients of determination were 0.951 and 0.944 for L^* and b^* values, respectively. Based on the reaction rate constants, the D -values of L^* and b^* were determined to be 34.4 and 85.3 months, respectively. Besides, the activation energies of L^* and b^* values of stored chestnuts were 89.18 and 78.47 kJ/mol, respectively, which were higher than that of broccoli^[45] and raw meat^[46] during storage at 3°C . If requiring L^* to be large than 50, about 8 months could be stored at 3°C .

3.6 Applications of kinetic data

During the thermal treatment, chestnut kernels became dark (brown) and even unacceptable by the consumers when the darkness (brownness) reached some extent (e.g. $L^*>50$). This color limit is in the same magnitude as required by banana^[47], almond^[4] and walnut^[7]. According to the kinetic model of L^* listed in Table 2, the corresponding time and temperature to $L^*=50$ could be estimated to be 995, 144, 10, 3 and 2 min at 50, 55, 60, 65 and 70°C , respectively. By combining the temperature-time requirement for complete kill of the most serious insect in stored cereal grain (Indianmeal moth)^[39], the practical and effective heat treatment could be defined as shown in Figure 7. A similar method was

used for the development of thermal treatments to control codling moth in stone fruits and nuts^[48]. The possible operating range could be low temperature and long time or high temperature and short time. Due to high throughputs and acceptable large temperature variations for short holding time, a thermal treatment protocol by

combining 52-62°C with holding 5 min could be potentially developed for disinfecting chestnuts without damaging color. This protocol could be successfully achieved by radio frequency heating in chestnuts reported by Hou et al.^[49].

Table 3 Comparison of kinetic parameters for the thermal treatment of color degradation between chestnuts and various fruits reported in the literature

Product	Temp. range/°C	Color parameter	Reaction order	<i>k</i> /min ⁻¹	<i>D</i> /min ^a	<i>z</i> /°C ^b	<i>E_a</i> /(kJ·mol ⁻¹) ^c	Reference
Cashew apple	100-180	<i>L</i> *	1	0.0198 _{T=140°C}	116.3 _{T=140°C}	33.0	98.0	[32]
Hazelnuts	100-180	<i>L</i> *	1	—	—	—	64.5	[23]
Papaya puree	70-105	<i>a</i> * <i>b</i>	1	0.378 _{T=105°C}	6.1 _{T=105°C}	76.2	32.6	[44]
Peach puree	115-135	<i>L</i> *	1	0.0029 _{T=122.5°C}	794.1 _{T=122.5°C}	28.3	107.0	[29]
		<i>a</i> *	1 ^w	0.00003 _{T=122.5°C}	—	—	106.0	
		<i>b</i> *	1	0.004 _{T=122.5°C}	575.8 _{T=122.5°C}	27.4	109.0	
Pepper	60-80	<i>a</i> *	1	0.0089 _{T=60°C}	258.72 _{T=60°C}	27.08	20.16	[24]
	Pine apple puree	70-90	<i>L</i> *	1	0.00023 _{T=90°C}	10013.0 _{T=90°C}	47.6	65.3
<i>a</i> *			0	0.00067 _{T=90°C}	3437.3 _{T=90°C}	67.9	36.9	
<i>b</i> *		1	0.001 _{T=90°C}	2303.0 _{T=90°C}	38.1	62.2		
<i>L</i> *		1	0.0021 _{T=110°C}	1097.0 _{T=110°C}	27.1	129.4		
Seedless guava	80-95	<i>a</i> *	0	0.0043 _{T=110°C}	541.9 _{T=110°C}	30.1	94.9	[28]
		<i>b</i> *	1	0.0061 _{T=110°C}	378.0 _{T=110°C}	25.7	109.4	
		<i>L</i> *	1	0.0046 _{T=95°C}	500.7 _{T=95°C}	19.7	122.7	
Wheat germ	100-150	<i>a</i> *	1	0.0081 _{T=95°C}	284.3 _{T=95°C}	28.4	88.5	[27]
		<i>b</i> *	1	0.0083 _{T=95°C}	277.5 _{T=95°C}	23.4	104.9	
Chestnut	50-70	<i>L</i> *	1	0.179 _{T=70°C}	12.9 _{T=70°C}	7.7	277.0	This study
		<i>b</i> *	1	0.165 _{T=70°C}	13.9 _{T=70°C}	6.1	346.1	

Note: ^a*D* value estimated by Equation (6); ^b*z* value calculated by Equation (7); ^c*E_a* value estimated by Equation (8). 1^w: reversible first order.

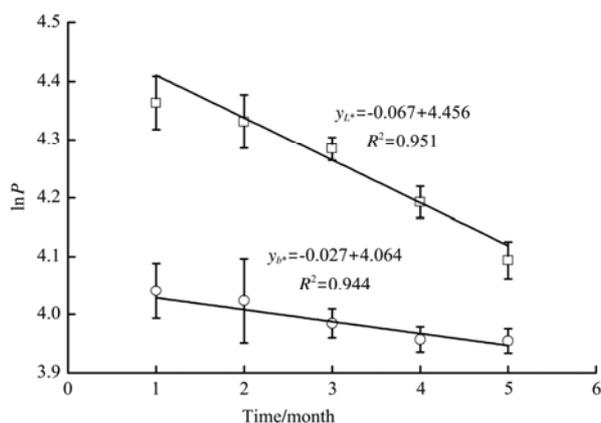


Figure 6 First order plots for color degradation (*P* means *L** or *b**) in chestnuts during storage

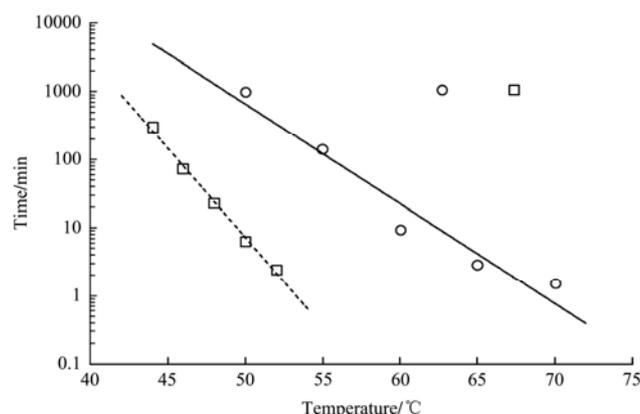


Figure 7 Product quality (*L**) curves of chestnuts from this study and typical insect (Indianmeal moth) thermal-death-time curves^[35] to define effective thermal disinfection regions

If the first order reaction model would be inadequate to describe quality changes, some other order model or more sophisticated models could be used to improve

kinetic model accuracy together with fractional conversion instead of using an absolute measurement value to indicate quality changes^[13,25,26,50]. Except for

color, other quality attributes of chestnuts must be validated in developing successful thermal treatment protocol. In industrial heat treatment of chestnuts, however, an actual process that includes heating, holding and cooling, is non-isothermal. The general solution is to calculate the accumulated chestnuts quality loss over the entire process time based on the developed kinetic model.

4 Conclusions

During thermal treatment and storage, the L^* and b^* values of chestnut kernels decreased with increasing temperature, heating time and storage time. The color degradation in chestnuts followed first order kinetics during thermal treatment and storage, with coefficients of determination (R^2) ranging from 0.923 to 0.977. The z values of L^* and b^* were 7.7 and 6.1°C, and the activation energies of the L^* and b^* value were 287.19 and 347.48 kJ/mol, respectively, suggesting b^* was more sensitive to temperature changes than L^* . During cold storage, the activation energies were 89.18 and 78.47 kJ/mol for L^* and b^* , respectively. By combining the temperature time requirement for complete kill of the target storage insects, a thermal treatment protocol with holding 5 min at 52-62°C could be potentially developed for disinfesting chestnuts without damaging color.

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