



Original research

Prediction of the changes in physicochemical properties of key lime juice during IR thermal processing by artificial neural networks

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ABSTRACT

Thermal processing of the key lime juice leads to the inactivation of pectin methylesterase (PME) and the degradation of ascorbic acid (AA). These changes affect directly the cloud stability and color of the juice. In this study, an artificial neural network (ANN) model was applied for designing and developing an intelligent system for prediction of the thermal processing effects on the physicochemical properties of key lime juice during conventional and infrared (IR) heating. The inputs of this network were time and temperature and the outputs were changes in PME activity, AA content, browning index (BI) and also cloud stability of the juice. The feed-forward neural network with a logarithmic transfer function, Levenberg–Marquardt training algorithm and eight neurons in the hidden layer (topology 2-8-4) was chosen as the best ANN model ($R^2 > 0.95$, RMSE=0.47 and SE=0.28). The predicted values using the optimal ANN model vs. experimental values represented a correlation coefficient higher than 0.95 and 0.90 during IR and conventional thermal processing, respectively. This model can therefore be applied in prediction of the effects of thermal processing on the physicochemical properties of the lime juice in pilot plants, processing factories and online monitoring.

Keywords: IR thermal processing; Physicochemical properties; Key lime juice; ANN; Modeling

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1. Introduction

Key lime is a fruit of citrus species; its juice is used as a food flavoring and also an acidifying agent (Ziena, 2000). Thermal treatment below 100°C is adequate to increase the safety and shelf life of key lime juice ($\text{pH} < 2.2$) by controlling the microbial and enzymatic activity (Chen et al., 1993). In high acid food products ($\text{pH} < 4.6$), pectin methylesterase (PME) has been introduced as the heat treatment index because of its higher thermal resistance than target microorganisms (Chen & Wu, 1998; Polydera, Galanou et al., 2004; Snir et al., 1996; Versteeg et al., 1980). PME also influences the cloud stability, appearance and total acceptance of the juice by de-esterifying the methoxylated pectin (Kimball, 1999). Thermal processing of the juice by hot water, a conventional thermal treatment method, needs long heating times that leads to loss of nutrients such as ascorbic acid (AA). Non-enzymatic browning occurs as a result of AA degradation which causes off-

taste and off-color of the juice (Burdurlu et al., 2006). Therefore, emerging food processing technologies such as infrared irradiation (IR) are introduced to reduce and/or eliminate the unwanted effects of conventional thermal treatments. IR treatment, as a radiation method, has advantages over the conventional thermal treatments such as direct heat penetration, high energy efficiency, faster and uniform heating, equipment compactness, lower degradation of nutritional components and physical properties (Aghajanzadeh et al., 2016; Rastogi, 2012).

Artificial neural networks (ANNs) as optimization algorithms, can mathematically model the learning processes. It doesn't require previous information about the relationships between process parameters (Sablani et al., 2002; Sablani & Rahman, 2003). The model, a simple approximation of complex process, is used in estimation, prediction and control of different food processing. ANN has been used in the study of thermal degradation of ascorbic acid in green asparagus (Zheng et al., 2011), prediction of juice

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viscosity as a function of concentration and temperature (Rai et al., 2005), the thermal conductivity of different foods (Sablani et al., 2002; Sablani & Rahman, 2003), lycopene extraction from tomato pulps (Dolatbadi et al., 2016), prediction of the quantity of lycopene and β -carotene content in food samples (Camara et al., 2009), dough rheological properties (Ruan et al., 1995), freeze drying behavior of strawberries (Menlik et al., 2009) and temperature variation of potato during solar drying (Tripathy & Kumar, 2009). Unlike the physicochemical experiments, ANN models are simple, fast with high accuracy. They have no destructive effects on the food products and also require less manpower and cost during online monitoring of process in labs and factories.

The objective of this study was to develop artificial neural network models for the prediction of PME activity, AA content, BI changes and cloud stability of key lime juice as a function of time and temperature during thermal processing using IR and conventional heating by hot water.

2. Material and Methods

2.1. Sample preparation

Fresh key lime (*Citrus aurantiifolia*) was obtained from a local market in Gorgan, Iran. It was stored at 4°C until used. The washed lime was then squeezed using a domestic juice extractor. The obtained juice was filtered (mesh size: 170) to remove large size particles and immediately heated.

2.2. Thermal processing of lime

2.2.1. Conventional thermal processing

Prepared lime juice (30 ml) was transferred into a clean 100 ml beaker and heated in a water bath (WNB-22, Memmert, Germany, 1800 W) at 60, 70, 80 and 90°C for 15, 10, 5, 2.5 min, respectively. Initial time was set as the juice reached to the desired temperature (end of come up time). Temperature variation of the juice was recorded using a data logger (TC-08, Pichotechnology Co, UK) equipped with a 1 mm diameter copper–constant thermocouple (T-type). The heated juice was finally cooled to 25°C using an ice-water bath (Aghajanzadeh et al., 2016).

2.2.2. Infrared heating system

A developed infrared heating system consisted of a radiant wall heating chamber with infrared modules (1500 W) was used. The outer diameter of infrared modules was 10 mm. The distance between the surface of the juice and infrared source was 8.5 cm (Aghajanzadeh et al., 2016). The temperature of the juice ($\pm 1^\circ\text{C}$) was controlled by connecting the temperature controller to the IR lamp. The juice was mixed every 15 s to ensure uniform heating. Other thermal processing conditions (sample volume, container, temperature and time) were similar to the conventional heating process.

2.3. Physicochemical analyses

2.3.1. Measurement of PME activity

Based on the Kimball method (1999), 5 ml of lime juice was mixed with 20 ml of 1% pectin-salt solution (10 g pectin and 15.3 g NaCl diluted in 1 L distilled water) at 30°C. Two different normalities of NaOH (0.05 and 2 N) were used to adjust the pH of the solution equal to 7.7. Finally, 0.1 ml of NaOH (0.05 N) was added and the time taken to regain pH to 7.7 was recorded to calculate the enzyme activity unit (PEU) using Eq. 1 (Kimball, 1999):

$$\text{PEU (unit/ml)} = \frac{N \times V}{V' \times t} \quad (1)$$

where N, V, V' and t are normality of the NaOH (0.05 N), the last added volume of the NaOH (0.1 ml), juice volume (5 ml) and the recorded time (min), respectively.

2.3.2. Ascorbic acid measurement

20 ml of the juice was diluted in distilled water (150 ml). The solution was titrated using iodine solution (5 g potassium iodide (KI), 0.268 g potassium iodate (KIO₃) and 30 mL of 3 M sulfuric acid (H₂SO₄)) and was diluted with distilled water until 500 mL in presence of 1% starch solution as an indicator. The consumed volume of iodine solution, until observing a fixed dark - blue color, was recorded to calculate the AA content using Eq. 2 (Kashyap & Gautam, 2012):

$$\text{mg ascorbic acid/100 mg sample} = 0.88 \times \text{ml iodine solution} \quad (2)$$

2.3.3. Browning index

The image processing method was used to study the BI changes. 15 ml of processed juice was filled in a plate (with 1 cm height and 6 cm diameter) and its surface image was taken by a scanner (Scanjet G2710, HP, USA) which was completely shielded by a black cover. The taken images were saved as JPEG format with 600 dpi resolution (RGB color). According to Eqs. 3 and 4, the browning index (BI) was calculated based on the analyzed L*, a* and b* parameters using Image J software (version 1.42e, Wayne Rasband, National Institutes of Health, USA) (Lee & Coates, 1999):

$$X = \frac{a^* + 1.75 \times L^*}{5.64 L^* + a^* - 3.012 b^*} \quad (3)$$

$$\text{BI} = \frac{100 (x - 0.31)}{0.17} \quad (4)$$

2.3.4. Cloud value measurement

Based on Versteeg et al. (1980) method, 5 ml of lime juice was centrifuged (3000 rpm, 10 min, 25°C). The cloud stability of the juice was determined by reading the absorbance of the supernatant at 660 nm (T-80, UV/VIS Double Beam Spectrophotometer, PG Instrument, USA). The absorbance of distilled water was considered as a blank.

Table 1. List of the used networks, transfer functions and back propagation training algorithms in ANN training.

No.	Network	Transfer function	Training algorithms
1	Feed-forward back propagation network (newff)	Hyperbolic tangent sigmoid transfer function (tansig)	Levenberg-Marquardt back propagation (lm)
2	Cascade-forward back propagation network (newcf)	Log-sigmoid transfer function (logsig)	Scaled conjugate gradient back propagation (scg)
3	-	Linear transfer function (purelin)	Gradient descent back propagation (gd)
4	-	-	Resilient back propagation; Rprop (Rp)

2.4. Designing and selecting the optimal artificial neural network model

The optimal ANN was determined using the Neural Network tool of MATLAB software (R2013 a). Based on the dependent and independent factors of the process, the used ANN contains two inputs and four outputs. The inputs of the network were temperature and time of heating treatment while the outputs were included PME activity, AA content, BI and cloud stability of the juice. To achieve an optimal ANN, feed-forward and backward networks were considered with different numbers of neurons in the hidden layers, transfer functions and the training algorithms (Table 1). The selection of the optimal neural network was done based on the correlation coefficient (R^2), Standard Error (SE) and Root Mean Square Error (RMSE) (Bahmani et al., 2015).

The raw data inputs reduce the processing speed and also the accuracy of the network. Data normalization is known as a fundamental data preprocessing step for learning from data before starting the ANN model (Nayak et al., 2014). Without performing this step, minimizing bias within the neural network to guarantee the quality of the data could not be achieved. In this study, the Eq. 5 was used for normalizing the data; so, the inputs and outputs would be standardized and ranged between 0 and 1.

$$V_N = \frac{V_R - V_{\min}}{V_{\max} - V_{\min}} \quad (5)$$

where V_N and V_R represent the normalized and raw data, respectively. V_{\max} and V_{\min} are the maximum and minimum of raw data.

Finding the optimal network architecture is a big deal in the best ANN model developing. So, lots of trial, error and replications were used to reduce the errors in prediction the PME activity, AA content, BI changes and cloud stability of key lime juice during conventional and IR heating. Several network configurations with a combination of various model parameters (number of neurons in the hidden layer, transfer functions and feed-back training algorithm) were examined to achieve this goal (Table 1).

In this study, different networks including feed-forward backpropagation network (newff) and cascade-forward back propagation network (newcf) were used. Applied transfer functions were hyperbolic tangent sigmoid transfer function (tansig), log-sigmoid transfer function (logsig) and linear transfer function (purelin). Levenberg-Marquardt backpropagation (lm), scaled conjugate gradient backpropagation (scg), gradient descent backpropagation (gd) and resilient backpropagation; Rprop (Rp) was served as the training algorithm.

3. Results and Discussion

3.1. Changes in physicochemical properties of key lime juice

Key lime juice is a good source of antioxidants and vitamins that are thermal sensitive chemical compounds. The changes in chemicals will influence the physical properties of the juice. In this study, it was found that thermal treatment caused changes in the nutritional value, stability and color properties of the juice. During IR heating, come up time decreased due to quick rising in initial temperature of the juice to the target temperature in comparison to the conventional heating process. This resulted in less AA degradation, less development in browning reactions and lower color variation in the IR processed juice. The more PME inactivation was observed during thermal processing at higher temperature causing as increase in the juice cloud stability during using both thermal treatment.

Table 2. Results of error measurements in ANN model considering hyperbolic tangent sigmoid transfer function, resilient back propagation and four neurons for different network types.

Treatment	Network type	RMSE	SE	R^2
IR heating	newff	0.03	0.30	0.90
	newcf	0.16	0.51	0.77
Conventional heating	newff	0.33	0.33	0.92
	newcf	0.74	0.51	0.87

3.2. ANN modelling

3.2.1. Type of the ANN network

At first, selecting the type of network was based on the design of models with two common feed-forward and cascade-forward back networks with a resilient training algorithm, four neurons in the hidden layer and tangent sigmoid transfer function. The performance of this network was then evaluated based on the statistical calculated values (Table 2). Based on these obtained indices, the feed-forward network had the lowest errors and the highest correlation coefficients. It was selected as a preferable and optimized network in comparison to the cascade-forward back network.

3.2.2. The best transfer function in the ANN model

In the next step, the selected feed-forward network with four hidden layers, resilient learning algorithm and different transfer functions was tested to determine the optimal transfer function (Table 3). By considering the various statistical indices which are presented in Table 3, the ANN with log-sigmoid transfer function exhibited the highest satisfaction in the prediction of the experimental values.

Table 3. Results of error measurements in feed-forward back propagation ANN model considering four neurons and resilient back propagation for different transfer functions.

Treatment	Transfer functions	RMSE	SE	R ²
IR heating	tansig	0.52	0.31	0.96
	logsig	0.38	0.23	0.97
	purelin	2.15	0.58	0.91
Conventional heating	tansig	0.67	0.18	0.93
	logsig	0.54	0.12	0.96
	purelin	0.98	0.49	0.90

Table 4. Results of measures of error in feed-forward back propagation ANN model considering log-sigmoid transfer function and resilient back propagation for different number of neurons.

Treatment	Number of neurons	RMSE	SE	R ²
IR heating	2	0.61	0.45	0.84
	4	0.38	0.32	0.89
	6	0.44	0.21	0.95
	8	0.04	0.13	0.98
	10	1.38	0.56	0.93
Conventional heating	2	0.32	0.65	0.87
	4	0.25	0.52	0.89
	6	0.48	0.35	0.91
	8	0.12	0.38	0.98
	10	0.68	0.81	0.89

Table 5. Results of error measurements in feed-forward back propagation ANN model considering eight neurons and log-sigmoid transfer function for different training algorithms.

Treatment	Training algorithms	RMSE	SE	R ²
IR heating	trainlm	0.43	0.24	0.96
	trainscg	0.82	0.32	0.93
	traingd	0.94	0.71	0.88
	traingd	1.26	0.79	0.91
Conventional heating	Trainlm	0.47	0.28	0.95
	Trainscg	0.93	0.82	0.91
	traingd	0.68	0.58	0.92
	traingd	0.52	0.41	0.93

3.2.3. The number of neurons in the hidden layer of the ANN model

To optimize the network and select the optimal number of neurons in the hidden layer of an optimum network, the feed-forward network with a hyperbolic tangent sigmoid transfer function, resilient training algorithm with different neurons in the

hidden layer was created and degree of the defined performance indices was calculated for each topology. As shown in Table 4, the ANN with eight neurons in the hidden layer was recommended as the optimal network. So as shown in Fig. 1, based on the inputs (time and temperature of hot water and IR heating process) and outputs (PME activity, AA, BI and cloud stability of the juice), the chosen topology of the optimal ANN was 2-8-4 (two neurons in the input layer-eight neurons in the hidden layer- four neurons in the output layer).

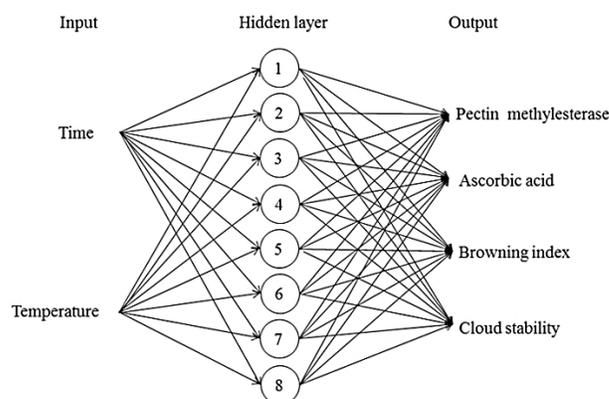


Fig 1. Topology of the optimal artificial neural networks.

3.2.4. The training algorithm

According to the aim of this study, the ANN with the highest accuracy and the most optimal results was ascertainable by selecting the best algorithm with an investigation of the different training ones. Thus, all of the selected optimal parameters (feed-forward network, logarithmic transfer function and six neurons in the hidden layer), different training algorithms, statistical analysis by comparing the RMSE, SE and R² were performed; based on the obtained results, the Levenberg-Marquardt training algorithm was finally selected as the algorithm to create a network with the lowest errors (Table 5).

3.2.5. Optimum ANN structure and accuracy

A feed-forward neural network with a logarithmic transfer function, Levenberg–Marquardt training algorithm and eight neurons in the hidden layer with the correlation coefficients 0.96 and 0.95 was therefore selected for IR heating and hot water thermal processing of the juice, respectively. Also, RMSE and SE of these networks were 0.43 and 0.24 using IR thermal processing and 0.47 and 0.28 during conventional heating in hot water. These statistical indices showed the best-predicted results and the lowest obtained errors. The high accuracy of this model was also approved by the high correlation coefficients (Table 6) which represented the predicted values using the optimal ANN vs. experimental values in terms of four output changes. For the overall evaluation of the model, the data were divided into 70% for training and 30% for validation. Without considering the inputs, the values of training (> 0.96), validation (> 0.91) and RMSE (< 0.52) were estimated as shown in Table 6.

Weight and bias matrices were determined for both thermal processing methods in the selected optimal ANN. Table 7 presents

the weight values for connecting the input layer to the hidden layer and also linking the hidden layer to the output one. In addition, the bias values of the hidden and output layers for IR and hot water thermal treatment are shown in Table 8.

4. Conclusion

PME activity is considered as the index of thermal processing of high acid juices. During thermal processing, the loss in vitamins such as ascorbic acid and undesirable changes in physicochemical properties are undeniable. Experimental studies are sometimes time-consuming, high cost, complicated and have destructive effects on the sample. In this study, a neural network-based model was developed for the prediction of changes in PME activity, AA content, BI and also cloud stability of the key lime juice during thermal processing by hot water and IR heating. The feed-forward neural network with a logarithmic transfer function, Levenberg–Marquardt training algorithm and eight neurons in the hidden layer

was chosen ($R^2 > 0.95$) as the best structure. The chosen topology of the optimal ANN was 2-8-4 that could predict the thermal effects of the heating on some physicochemical properties of the juice with RMSE and SE less than 0.47 and 0.28, respectively.

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Conflict of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Table 6. Correlation coefficients of the predicted outputs changes in contrast to the experimental values and overall evaluation of the model for the optimal topology (2-8-4).

	Parameters	IR heating	Conventional heating
Output correlation evaluation	Pectin methylesterase	0.97	0.95
	Ascorbic acid	0.98	0.92
	Browning index	0.97	0.90
	Cloud value	0.95	0.90
Model overall evaluation	Training	0.97	0.96
	Validation	0.93	0.91
	RMSE	0.52	0.25

Table 7. Weight values for connecting the input layer to the hidden layer and hidden layer to the output layer in optimum network.

	INPUT TO HIDDEN				HIDDEN TO OUTPUT							
	IR heating		Water bath		IR heating				Conventional heating			
	INP 1	INP 2	INP 1	INP 2	OUT 1	OUT 2	OUT 3	OUT 4	OUT 1	OUT 2	OUT 3	OUT 4
HID 1	-2.22	9.02	-2.55	4.95	-1.07	-1.31	-0.28	0.25	-1.07	-3.75	0.90	-0.30
HID 2	6.56	2.81	-7.52	-2.82	-2.02	-1.45	-0.14	2.18	-2.02	-0.92	1.69	2.75
HID 3	4.56	6.53	-6.30	-7.28	2.83	-0.01	2.58	2.31	2.83	2.11	-3.31	-4.14
HID 4	2.04	4.66	-6.07	-4.05	1.01	-0.69	2.05	1.58	1.01	1.61	1.41	-0.19
HID 5	-0.02	-8.32	-2.73	6.93	-0.63	-0.17	1.20	0.54	-0.63	-2.00	0.49	0.45
HID 6	-3.04	-0.24	-6.64	5.34	-0.62	4.05	-1.18	-3.00	-0.62	0.57	0.42	0.42
HID 7	-0.08	-7.32	5.35	1.54	-1.70	-0.50	-2.44	1.86	-1.70	-1.63	1.19	1.61
HID 8	4.69	-7.27	-7.89	-0.23	1.86	0.25	2.60	-0.41	1.86	1.61	-1.58	-1.52

Table 8. Bias values for hidden and output layer in optimum network.

	HID 1	HID 2	HID 3	HID 4	HID 5	HID 6	HID 7	HID 8	OUT 1	OUT 2	OUT 3	OUT 4
IR heating	5.78	-5.65	-3.07	0.69	0.98	-3.32	6.30	7.31	1.35	0.18	-1.77	-2.50
Water bath	9.86	6.05	3.94	5.44	1.09	-2.30	1.74	-7.80	0.58	1.55	-1.82	0.37

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