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Comparison of Sigmoid Logarithm and Hyperbolic Tangent Functions in Modeling the Oxidation Parameters of Soybean Oil Containing Extract of Black Plum Peels Natural Antioxidant

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Abstract

To predict the oxidation parameters of soybean oil (SBO), we utilized five levels of black plum peel extraction (BPPE) antioxidant concentration (0, 400, 800, 1200, and 2000 ppm) and four levels of oil storage time (0, 8, 16, and 24 days) under accelerated oxidation conditions (temperature 60 °C). We investigated the peroxide value (PV), thiobarbituric acid (TBA) value, acidity, conjugated diene (CD) content, and carbonyl value (CV). Artificial neural networks were employed using MATLAB software for prediction. Several feed-forward back-propagation networks with 2-6-5 topologies were examined, achieving correlation coefficients greater than 0.959 and mean square errors (MSE) < 0.009. The optimal model utilized a sigmoid logarithm activation function, a jumping learning pattern, and 1000 learning cycles. These models demonstrated high correlation coefficients (above 0.912) in predicting the oxidation process of SBO.

Keywords

Oxidation parameters, Soybean oil, Black plum peel extraction, Artificial neural network

Introduction

SBO is widely utilized in the food, pharmaceutical, and cosmetic industries [1-3] due to its high content of polyunsaturated fatty acids, notably α-linolenic acid (18:3 ω-3) [2, 4]. However, SBO is prone to oxidation due to these unsaturated fatty acids. This oxidation reduces its oxidative stability, especially during storage and food processing, resulting in undesirable aroma and taste, which limits its applicability in various formulations [1, 5].

There are various approaches to inhibit lipid oxidation, including enzyme deactivation, the use of chelating agents, implementation of appropriate packaging techniques, and the addition of antioxidants [1-4]. However, despite the affordability and stability of synthetic antioxidants in food technology, consumer awareness of their potential health risks, such as cancer and carcinogenesis, has led to a decrease in their usage [1, 3, 4]. Natural antioxidants are widely regarded as effective and preferable alternatives, due to concerns regarding the potential long-term toxicity of synthetic antioxidants containing phenolic compounds [3-5].

The plum tree, scientifically known as *Prunus domestica* L., is a deciduous plant belonging to the Rosaceae family [6, 7]. Plums hold significant agricultural

importance in Iran and are widely recognized as valuable fruits [5, 8]. Plums are renowned for their diverse range of flavors and textures, attributed to the cultivation of various plum varieties [7]. Plums are also notable for their low-calorie content and high nutritional value. They are rich in carbohydrates such as sucrose, glucose, fructose, and sorbitol. Additionally, plums contain organic acids like citric acid and malic acid, soluble fiber in the form of pectin, as well as tannins, volatile substances, and enzymes [7]. These compounds contribute to the distinctive taste, nutritional composition, and health benefits associated with plums. From a nutritional standpoint, plums are a valuable source of minerals such as iron, calcium, phosphorus, manganese, sulfur, magnesium, and potassium, as well as vitamins A, B, C, and PP. Plums are rich not only in dietary fiber, sorbitol, anthocyanins, carotenoids, and phenolics [7], but plums not only offer a range of health benefits but also have a significant by-product issue during processing, particularly in drying, where plum peel is often discarded due to its high moisture content and susceptibility to microbial spoilage, contributing to environmental pollution. However, recent studies have revealed that black plum peel puree contains a substantial concentration of antioxidants, with an impressive value of 88.59%. Additionally, the puree has been found to contain a total phenolic compound measurement of 105.91 mg/g GA (gallic acid equivalent). These findings underscore the remarkable antioxidant and phenolic content in black plum peel puree, suggesting its potential for delivering health benefits [7].

Artificial neural networks are simplified models of real neural systems extensively employed in solving various scientific problems. Their applications span across classification, interpolation, estimation, detection, and more [9]. One of the advantages of artificial neural networks is their ability to simulate non-linear systems without prior assumptions about the process. They are also highly effective for prediction when input information is incomplete or ambiguous [9]. Some researchers have used these models to predict oil properties during extraction or oxidation [10, 11]. The aim of this study was to predict oxidation parameters of SBO containing BPPE using various artificial neural network functions, and to determine the optimal neuron arrangement for predicting these properties. To this purpose, the extract of black plum peel was prepared and added to SBO (0 - 200 ppm). The samples contained natural antioxidant stored at 60 °C for 24 days. Then, the oxidative parameters studied in the specific times.

Material and Methods

Preparation of BPPE

The plum peel underwent a thorough washing process to remove any impurities. Subsequently, the cleaned peel was crushed using an industrial crusher and stored in a freezer for the upcoming experiments. To create the BPPE, approximately 10 g of the crushed black plum peel was dissolved in 100 ml of 80% ethanol. This mixture was heated at 50 °C for 18 to 24 h. The resulting mixture was filtered using Whatman filter paper 1 and a Buchner funnel. The solvent evaporated, leaving the final BPPE, which was stored at -18 °C until the experiments commenced. This multi-step process ensured the plum peel was properly prepared, and the BPPE was ready for use in the study investigating its potential as a natural antioxidant.

Preparation of oil sample

SBO was obtained from the Shadgol Industry in Neyshabur, Iran. To evaluate its antioxidant properties, BPPE was added to SFO at four different concentrations (0 - 2000 ppm). The samples were then stored at 60 °C for 24 days. During this period, various quality characteristics of the SFO, including PV, acidity, TBA, CD, and CV, were measured at intervals of 0, 8, 16, and 24 days. Finally, the quality parameters of the SFO were compared with those of oils from the same brand fortified with commercial antioxidants.

PV determination

PV is a critical parameter used to assess the extent of oxidation in fats and oils. It measures the concentration of peroxides and hydroperoxides, which are primary oxidation products formed during the early stages of lipid oxidation. A higher PV indicates more extensive oxidation and potential rancidity, which impacts the quality and shelf life of the oil or fat [12, 13].

PV of the oil samples was determined using the method described by Tinello and Lante [4]. A 0.5 g oil sample was dissolved in 25 ml of a 3:2 (v/v) mixture of acetic acid and chloroform. Subsequently, 0.5 ml of saturated potassium iodide (KI) solution was added to the mixture, which was then shaken and kept at room temperature in the dark for 5 min. Afterward, 75 ml of distilled water and 1 ml of starch indicator were added to the solution. The resulting mixture was titrated with a 0.01 N solution until a colorless endpoint was achieved. This titration method enabled the quantification of the PV in the oil samples during storage.

TBA value determination

TBA value is a widely used measure for assessing lipid oxidation, particularly in fats and oils. It is commonly used to evaluate the extent of rancidity in edible oils, animal fats, and processed foods. The TBA test measures the concentration of malondialdehyde (MDA), a primary product of lipid oxidation. TBA value helps in assessing the oxidative stability of fats and oils. Higher values indicate more advanced oxidation and potential rancidity [13-16].

The analysis of TBA was conducted following the method described by Drinić et al. [17]. For each sample, 200 mg was dissolved in 25 ml of 1-butanol. After thorough mixing, 10 ml of 2% TBA solution was added to 5 ml of this mixture, and the resulting solution was incubated at 95 °C for 2 h. Subsequently, the mixture was cooled in a water bath until it reached 25 °C. The absorbance was then measured at 532 nm. TBA values were determined using equation 1.

$$TBA = \frac{5(A-B)}{m} \quad (1)$$

Where: A = absorbance; B = absorbance of the control sample; and m = mass of the oil sample.

Acidity

Oil acidity refers to the level of free fatty acids present in an oil. It is an important quality parameter that can affect the oil's flavor, stability, and overall quality. This measurement is usually expressed as a percentage or as milligrams of potassium hydroxide (KOH) required to neutralize the free fatty acids in 1 g of oil. The acidity of soybean and SBO was measured according to the AOCS 3-63 method [15, 18].

CD determination

CDs are a type of organic compound containing two double bonds separated by a single bond, creating a conjugated system. In the context of oil and fat analysis, the determination of CDs is a crucial method for assessing the extent of lipid oxidation and the quality of the oil. CDs are formed early in the oxidation process of unsaturated fats and oils. Measuring their concentration helps assess the degree of oxidation and the shelf life of the product. The analysis of CD was carried out following the method outlined by Umeda and Jorge [19] in 2021.

CV determination

CV is a measure used to assess the level of oxidation in fats and oils. Carbonyl compounds, which include aldehydes and ketones, are formed as primary oxidation products when lipids undergo oxidative degradation. Determining the CV helps in evaluating the quality and shelf life of edible oils and fats. A higher CV indicates a higher degree of oxidation, which can affect the flavor, aroma, and safety of the oil [13, 20].

The determination of the CV followed the method described by Delfanian et al. [2]. A calibration curve of standard aldehyde (2,4-Decadienal) was plotted over a concentration range of 50 - 500 µM. A mixture of 50 mg of 2,4 D-nitrophenyl hydrazine (DNH) and 100 ml of 2-propanol was prepared. Approximately 0.15 g of the oil sample was placed in a volumetric flask and filled with a solvent containing 0.4 mg/ml of triphenylphosphine (TPP). Next, 1 ml of the DNH solution was added to 1 ml of the prepared sample solution and allowed to incubate at 40 °C for 20 min. The solution was then cooled, and 8 ml of 2% KOH was added before centrifuging for 5 min at 2000 rpm. The absorbance of the supernatant was measured at 420 nm, and the results were reported in micromoles of 2,4-Decadienal per gram of each sample.

Modeling using artificial neural networks

To determine the optimal neural network, the neural network tool in MATLAB R2013a software was utilized. The design of this network involved defining two inputs-antioxidant concentration and oil storage time-in a two-line matrix, and PV, TBA, acidity, CD, and CV in a 5-line matrix as targets. Various neural networks were tested, each employing different activation and learning functions, as well as varying numbers of neurons in the hidden layer. Their performance was evaluated using two criteria: correlation coefficient (R^2) and MSE, calculated by equation 2 and 3, respectively. Initially, the feedforward neural network with the highest efficiency was selected, with 1000 learning cycles established. Subsequently, different networks with a hidden layer were designed, allowing for varying numbers of neurons from 1 to 10. The input layer was connected to the hidden layer using hyperbolic tangent and logarithmic sigmoid activation functions, tested through iterative network trials. Additionally, a linear activation function was employed to connect the hidden layer to the output layer. Furthermore, two different learning models, Levenberg-Marquardt and resilient backpropagation (trainrp), were applied across different networks to assess their impact on network accuracy.

$$R^2 = 1 - \frac{\sum_{i=1}^{N}(Y_{pi} - Y_{ei})^2}{\sum_{i=1}^{N}(Y_{pi} - \overline{Y})^2} \quad (2)$$

$$MSE = \frac{1}{N}\sum_{i=1}^{N}(Y_{pi} - Y_{ei})^2 \quad (3)$$

Where: Y_{pi} = Ratio of features predicted by the network; Y_{ei} = Ratio of features obtained from experiments; $\overline{Y}$ = Average ratio of laboratory features; and N = Total number of observations.

Entering data in raw form reduces the speed and accuracy of the network. Therefore, the input data to the network should be normalized. If this step is not taken, the network will not converge during the training phase and the desired results will not be produced. In this study, equation 4 was used to normalize the data, which standardizes the inputs and outputs between 0 and 1.

$$V_N = \frac{V_R - V_{min}}{V_{max} - V_{min}} \quad (4)$$

Where: V_R = Initial raw data; V_{max} and V_{min} = Maximum and minimum values of the initial data; and V_N = Normalized data respectively [9].

Results and Discussion

Table 1 and table 2 illustrate the impact of the number of hidden layer neurons and the type of learning pattern on the prediction accuracy of feedforward backpropagation neural networks using hyperbolic and logarithmic sigmoid tangent

Table 1: Comparison of the effect of neurons number of hidden layer and the type of learning function and activation function of hyperbolic sigmoid tangent on predicting accuracy of various properties of oxidative parameter of SBO containing plum peel antioxidant.

Neurons number	trainlm		trainrp	
	R^2	MSE	R^2	MSE
2	0.69	0.033	0.591	0.074
3	0.81	0.03	0.809	0.032
4	0.869	0.028	0.816	0.028
5	0.882	0.016	0.865	0.025
6	0.871	0.022	0.867	0.016
7	0.9	0.014	0.887	0.026
8	0.901	0.024	0.899	0.017
9	0.757	0.032	0.894	0.018
10	0.838	0.07	0.867	0.02

Table 2: Comparison of the effect of neurons number of hidden layer and the type of learning function and activation function of sigmoid logarithm on predicting accuracy of various properties of oxidative parameter of SBO containing plum peel antioxidant.

Neurons number	trainlm		trainrp	
	R^2	MSE	R^2	MSE
2	0.437	0.132	0.015	0.101
3	0.817	0.025	0.818	0.027
4	0.746	0.046	0.855	0.024
5	0.876	0.012	0.853	0.025
6	0.839	0.032	0.959	0.009
7	0.898	0.023	0.854	0.024
8	0.864	0.031	0.86	0.03
9	0.912	0.018	0.899	0.017
10	0.915	0.019	0.851	0.036

transfer functions, with 1000 learning cycles. Based on the MSE and correlation coefficient values presented, the optimal neural network was identified as a feedforward neural network with a logarithmic sigmoid transfer function. Resilient backpropagation learning function, and a topology of 2-6-5 (2 neurons in the input layer, 6 neurons in the hidden layer, and 5 neurons in the output layer), achieving a correlation coefficient greater than 0.959 and a MSE of 0.009. Additionally, the high correlation coefficients depicted in figure 1, which compare values predicted by this optimal network against laboratory data for the 5 desired output variables, further support the accuracy of this model. As shown in figure 2, the proposed models demonstrated the highest and lowest accuracy in predicting acidity and CV, respectively. Huang et al. [10] employed an artificial neural network to predict the oxidation properties of various oils (cottonseed, rapeseed, sunflower, and palm) during heating, achieving a prediction accuracy of over 97% [10]. The modeling of the cooking stage in the industrial process of rapeseed oil extraction was investigated using the adaptive neural fuzzy inference system (ANFIS). Temperature and moisture content were considered as inputs, while insoluble fine matter in oil, oil and moisture content of the meal, and acidity of oil were considered as outputs. Three types of membership functions-gaussian, triangular, and trapezoidal were evaluated with 2-2 and 3-3 membership functions. The optimal models were selected based on the highest correlation coefficient and the lowest mean squared error: trapezoidal functions with 3-3 membership for insoluble fine matter in oil, acidity, and moisture content of the meal outputs, and triangular functions with 2-2 and 3-3 membership for oil and meal moisture content inputs, respectively. The high correlation coefficients observed between laboratory results and model outputs indicated acceptable accuracy, affirming the utility of these models in industrial process control [11]. Thakker et al. [21] employed an artificial neural network to model the oil extraction process from a type of herbal plant native to India. They used four parameters as test inputs: various ratios of solid matter, solvent volume, microwave powers, and extraction times. Through the evaluation of different neural networks, they identified the feedforward backpropagation network with a 4-7-3 topology, achieving a correlation coefficient greater than 0.9997 and MSE less than 0.0117. They found the sigmoid logarithm activation function to be optimal for this neural model [21]. Bakhshabadi et al. [9] modeled the process of extracting oil from sunflower seeds on an industrial scale using three levels of cooking temperature (70 °C, 80 °C, and 100 °C) and three levels of moisture content of seeds exiting from the cooking pot (7%, 7.5%, and 8%). They investigated the amounts of oil, moisture content and protein of the meal, insoluble fine matter in oil, and acidity of the oil. Through the evaluation of different neural networks, they identified the feedforward backpropagation network with a 2-5-10 topology, achieving a correlation coefficient greater than 0.999 and a MSE less than 0.003. They found the hyperbolic sigmoid tangent activation function, Levenberg-Marquardt learning model, and 1000 learning cycles to be optimal for this neural model [9].

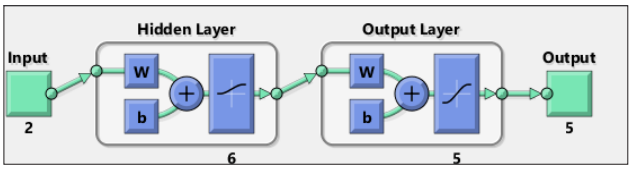

Figure 1: The schema of a selected optimized network containing 2 neurons in input layer, 6 neurons in hidden layer with activation function of hyperbolic sigmoid logarithm and 5 neurons in hidden layer with hyperbolic sigmoid logarithm activation function.

According to the selected neural network topology, which is 2-6-5, the weight matrix for the input layer to the hidden layer is a 2 × 6 Hessian matrix (connecting 2 neurons of the input layer to 6 neurons of the hidden layer) and for the hidden layer to the output layer of a 6 × 5 Hessian matrix (connecting 6 neurons of the hidden layer to 5 neurons of the output layer) will be in the form of matrices A and B, respectively:

$$A = \begin{pmatrix} 2.055 & 0.124 \\ 13.064 & -8.040 \\ -2.603 & -8.233 \\ 2.831 & -2.582 \\ -12.826 & 0.121 \\ -0.972 & -11.764 \end{pmatrix}$$

$$B = \begin{pmatrix} 0.516 & 0.831 & 0.534 & -12.513 & -8.363 & 3.660 \\ -1.806 & 0.139 & 0.075 & 0.841 & -0.595 & -3.224 \\ 5.732 & -0.327 & 0.190 & -0.144 & -0.138 & 2.658 \\ 3.355 & -1.351 & 1.395 & -3.142 & -1.605 & 3.372 \\ -0.944 & 0.752 & -0.842 & 2.261 & 0.905 & 3.499 \end{pmatrix}$$

In addition, the bias matrices for the hidden layer (matrix C) and the output layer (matrix D) will be 6 × 1 and 5 × 1 matrices, respectively.

$$C = \begin{pmatrix} -2.116 \\ -4.372 \\ 2.633 \\ 4.546 \\ -7.964 \\ -13.926 \end{pmatrix} \qquad D = \begin{pmatrix} 11.139 \\ -0.463 \\ -1.482 \\ 2.377 \\ -2.186 \end{pmatrix}$$

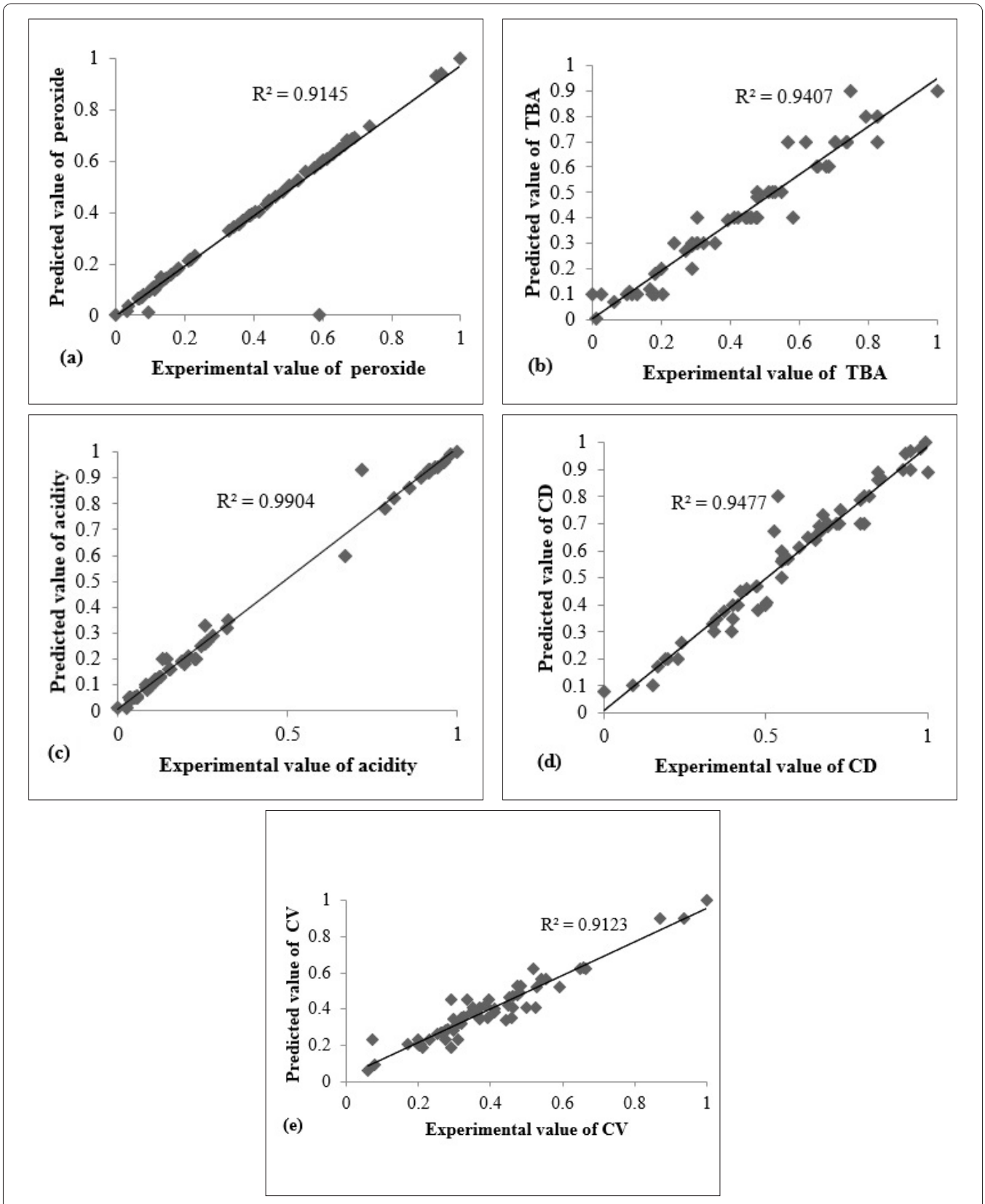

Figure 2: Diagram of predicted changes by the neural network for optimized topology (2-6-5) vs Laboratory's amount for: **(a)** peroxide, **(b)** TBA, **(c)** acidity, **(d)** CD, and **(e)** CV.

Conclusion

In this study, the modeling of the oxidation parameters of SBO containing black plum peel as a natural antioxidant during storage time was analyzed using an artificial neural network. The modeling approach included a single hidden layer, utilizing a feedforward backpropagation network. Different activation functions were explored to identify the most

effective function for estimating the oxidative properties of SBO. The findings indicated that the sigmoid logarithm activation function demonstrated optimal performance for the investigated parameters, achieving a 2-6-5 topology. Overall, comparisons among the functions revealed that the sigmoid logarithm activation function provided higher accuracy in predicting the oxidation parameters of SBO during storage, evidenced by lower relative error and higher coefficient compared to the hyperbolic tangent function. On the other hand, the results showed that with an increase in the storage time, oxidative parameters of SBO such as acidity, peroxide, etc., increased. However, with the increase in the concentration of antioxidants from plum skin in the oil, this increase was mitigated.

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None.

Conflict of Interest

None.

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