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ORIGINAL PAPER

Intelligent Estimation of the Canola Oil Stability Using Artificial Neural Networks

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Abstract In the present study, a multi-layer perceptron neural network and radial basis function (RBF) network were used to estimate the oxidative stability of canola oil during storage. Artificial neural networks (ANNs) were used to model oxidative stability of canola oil during storage, and comparison was also made with the results obtained from a regression analysis. The oxidative stability of canola oils was considered as dependent variable, and independent variables were selected as time (in week), variety, C14:0, C16:0, C18:0, C20:0, C18:1, C18:2, C18:3, and C22:1 fatty acid content. The results were compared with experimental data and it was found that the estimated oxidative stability by RBF neural network is more accurate than multi-layer perceptron network and regression model. It was also found that the oxidative stability of canola oil decreased with increase in storage time and C18:3 fatty acid content.

Keywords Canola oil · Oils stability · Artificial neural network · Radial basis function neural network · Multi-layer perceptron network

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Introduction

Lipid oxidation is a major cause of deterioration of fats and oils leading to losses of quality and nutritional value and the development of unpleasant flavors. Autoxidation is considered to be the main route of spoilage of edible oils and canola oil; with high proportion of unsaturated fatty acids, has the potential to develop undesirable odors and flavors during storage.

The stability of canola oil is limited mostly by the presence of unsaturated fatty acids specially linolenic acid and other factor, such as chlorophyll and its decomposition products and other minor components with high chemical reactivity, such as trace amounts of fatty acids with more than three double bonds. The flavors of oxidized oils are attributed to primary and secondary oxidation products (Gray 1978; Sherwin 1978; Wanasundara and Shahidi 1994; Rovellini et al. 1997; Shahidi and Wanasundara 1997).

The primary products of lipid oxidation are hydroperoxides, which are generally referred to as peroxide. Although the peroxide value is a common measurement of lipid oxidation, its use is limited to the initial stages of oxidation (Gray 1978). Oxidative stability is known as the resistance to oxidation under defined conditions and is expressed as the period of time required to reach an end point, usually corresponds to a sudden increase in oxidation rate. As oxidation normally proceeds very slowly until this point is reached, this time period is known as the induction period (IP). Numerous methods, using accelerated oxidation conditions, have been developed for the evaluation of oxidative stability (Rossell 1994; Wan 1995; Velasco et al. 2004). Elevated temperatures in the presence of oxygen or air, in excess, are applied to obtain results in reasonably short periods of time. The oil stability index method, also commonly known as the Rancimat method, allows oxidative stability to be determined automatically under standardized conditions (AOCS 1998). This method is widely used in the fats and oils industry and it can be applied by using two commercially available instruments: the Rancimat from Methrohm Ltd. (Herisau, Switzerland) and the Oxidative Stability Instrument from Omniom Inc. (Rockland, MA). The end point corresponds to a sudden rise of volatile acids generated from the oil samples heated at high temperature under constant aeration. These compounds are trapped in water and monitored by electro-conductivity.

Artificial neural networks (ANNs) are mathematical models whose architecture has been inspired by biological neural networks. ANNs are very appropriate for the modeling of non-linear processes, i.e., the case of oxidatative stability of vegetable oils. However, the application of an ANN with experimental data only is valid for the range of collected data in each study; if the data changes, a new ANN must be trained. Artificial neural networks have already been applied to simulate processes such as fermentation (Latrille et al. 1993), cross-flow microfiltration (Dornier et al. 1995), drying behavior of different food and agricultural materials such as carrot (Erenturk and Erenturk 2007; Kerdpiboon et al. 2006), tomato (Movagharnejad and Nikzad 2007), ginseng (Martynenko and Yang 2006), cassava and mango (Hernandez-Perez et al. 2004), osmotic dehydration (Trelea et al. 1997), wheat soaking (Kashaninejad et al. 2008), and osmotically dehydrated kiwi fruit (Fathi et al. 2009), but there is no information about application of artificial neural networks in simulation of oxidative stability of vegetable oils (in particular, canola oil). This study was carried out to examine and validate the efficiency of ANN for simulating the oxidative stability behavior of canola oil and to study the effect of fatty acids composition and time on the canola oil stability.

Materials and Methods

Materials

Three major canola varieties grown in Golestan Province of Iran namely Hyolla 401, Hyolla 420, and RGS003 were obtained from Cultivation, Research and Development Center of Oil Seeds of Golestan, Iran.

These three varieties were used in this study because they are most important varieties that are grown and consumed in Golestan Province.

The seed samples were dried to 8% moisture at 30-35 °C in a laboratory incubator (Tebazma Co., Iran). These samples were kept in a cold place until use. All chemicals used were of analytical grade and obtained from Sigma (MO, USA) and Merck chemicals (Germany).

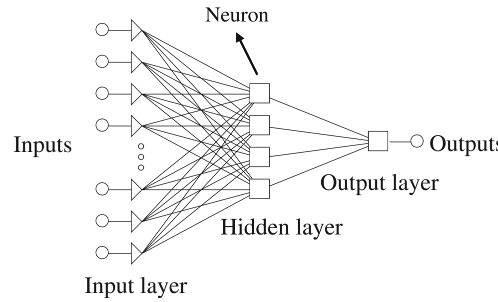

Fig. 1 Multi-layer perceptron neural network

Oil Extractions

The seeds were cold extracted using laboratory press (OEKOTECH, DD85—Berlin, Germany) at 30-45 °C and extracted oil was stored in dark at room temperature for 4 months.

Determination of Fatty Acids Composition

Fatty acid methyl esters (FAMEs) were prepared, according to the procedure described by the American Oil Chemists' Society-recommended method Ce 1b-89 (AOCS 1998). FAME were prepared by vigorous shaking of a solution of each oil sample in n-hexane (0.2 g in 8 mL) with 2 mL 2 N methanolic potassium hydroxide solution. The fatty acid composition of oils was determined by using a capillary gas chromatograph (Agilent, 6890 N Plus, Palo Alto, CA, USA) with a flame ionization detector and a BPX70 column (120 m × 0.25 mm i.d, 0.25 μm film thickness, Palo Alto, CA, USA). Temperature program was from 140 °C to 220 °C for 15 min with a 4 °C/min gradient. The injector temperature was fixed at 230 °C and detector temperature was fixed at 260 °C. Determination of each fatty acid contents was verified by comparison of retention times of test samples with those of reference standards.

Rancimat Test

Oxidative stability was evaluated by the Rancimat method. Stability was expressed as the oxidation induction period (h),

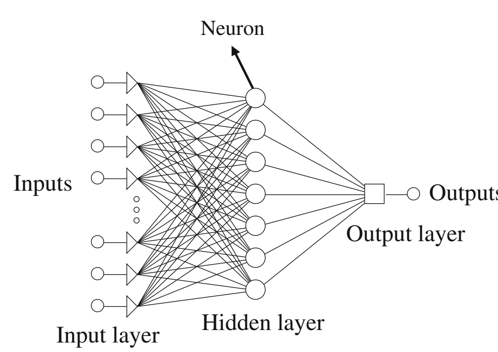

Fig. 2 General architecture of radial basis function networks

Table 1 The range of data (including train and test) for different variety

	Hyolla 401		Hyolla 420		RGS003	
	Train data	Test data	Train data	Test data	Train data	Test data
C14:0	0.076-0.083	0.076-0.096	0.081-0.095	0.084-0.092	0.063-0.069	0.066-0.068
C16:0	4.054-4.112	4.065-4.293	4.029-4.768	4.052-4.763	4.121-4.435	4.226-4.319
C18:0	2.223-2.302	2.207-2.301	2.275-2.896	2.867-2.886	2.229-2.452	2.229-2.452
C20:0	1.444-1.615	1.448-1.612	1.187-1.429	1.206-1.257	1.181-1.356	1.181-1.276
C18:1	63.2-63.7	63.2-63.34	61.09-62.8	62.12-62.52	63.94-64.09	63.94-64.01
C18:2	17.12-17.68	17.13-17.74	17.65-18.93	18.14-18.64	17.94-18.21	17.96-18.21
C18:3	10.24-10.58	10.3-10.57	7.255-7.717	7.273-7.626	8.116-8.468	8.329-8.406
C22:1	0.295-0.395	0.29-0.361	0.665-0.749	0.696-0.721	0.404-0.521	0.442-0.521
Rancimat	8.25-11.65	9.55-11.84	11.6-13.35	12.25-13.2	9.75-10.95	9.4-10.65

measured with the Rancimat (Model 679, Metrohm, Switzerland) using an oil sample of 2 g, heated up to 110°C, and a purified air flow rate of 20 ml/h. In the Rancimat method, the volatile degradation products were trapped in distilled water and measured conductmetrically. The IP was defined as the necessary time to reach the inflection point of the conductivity curve (AOCS 1998).

Statistical Analysis

The effect of fatty acid composition and storage time on oxidative stability of canola oils were determined using the analysis of variance method, and significant difference of means were compared using the Duncan's test at 95% significant level.

There are different models which might be adequate to describe different behavior of agricultural material. In the analysis of oxidation stability of canola oil, first, a regression model was developed to relate all independent variables to dependent variable (oxidative stability) as below:

$$\mathrm{OS} = K \times (\mathrm{Week})^a \times (C16:0)^b \times (C18:0)^c \times (C18:1)^d \times (C18:2)^e \times (C18:3)^f \tag{1}$$

After the calculation of the oxidation stability using the regression model, the equilibrium was fitted to the oxidative stability data and several important criteria such as coefficient of determination (R^2) and mean square error (MSE) were determined.

Artificial Neural Network

In this paper, multi-layer perceptron network (MLP) and radial basis function (RBF) based on back propagation learning rule were used.

Multi-layer Perceptron Network

The MLP network sometimes called back propagation network (Fig. 1) is probably the most popular ANN in engineering problems in the case of non-linear mapping and is called "Universal Approximator". It consists of an input layer, a hidden layer, and an output layer. The input nodes receive the data values and pass them on to the first hidden layer nodes. Each one of them collects the input from all input nodes after multiplying each input value by a weight, attaches a bias to this sum, and passes on the results through a non-linear transformation like the sigmoid transfer function. This forms the input either for the second hidden layer or the output layer that operates identically to the hidden layer. The resulting transformed output from each output node is the network output. The

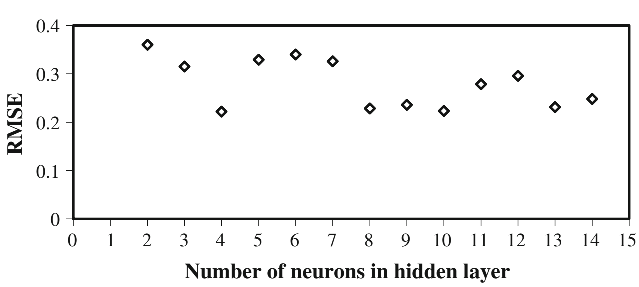

Fig. 3 Variation of RMSE of prediction against the number of neurons in hidden layer for MLP neural network

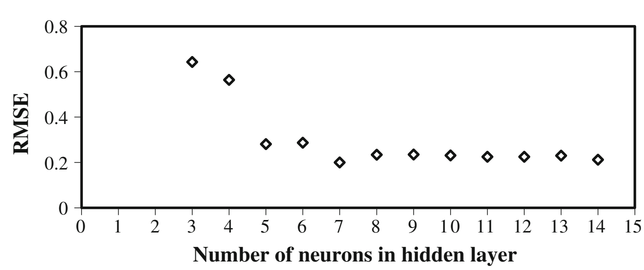

Fig. 4 Variation of RMSE of prediction against the number of neurons in hidden layer for RBF neural network

Table 2 Fatty acid compositions of oil extracted from three canola varieties used in this study (%)

Fatty acids	Hyolla401	Hyolla420	RGS003
C14:0	0.101±0.50c	0.117±0.15a	0.107±0.06b
C16:0	4.00±0.16c	4.38±0.29a	4.00±0.05b
C18:0	2.27±0.07c	2.860±0.07a	2.35±0.07b
C20:0	1.51±0.06a	1.26±0.09b	1.26±0.06b
SFA ∑	8.11±0.10b	8.68±0.17a	8.09±0.13b
C16:1	0.52±0.06b	0.52±0.11a	0.51±0.31c
C18:1(ω-9)	63.32±0.14b	61.35±0.34c	64.00±0.12a
C20:1	1.00±0.08b	1.11±0.10a	1.11±0.08a
C22:1	0.34±0.34c	0.61±0.13a	0.56±0.12b
MUFA ∑	65.27±0.16b	63.30±0.12c	66.19±0.23a
C18:2(ω-6)	17.28±0.20c	18.21 ± 0.32a	18.09±0.09b
C18:3(ω-3)	10.34±0.10a	8.48±0.18b	8.44±0.14b
PUFA ∑	27.62±0.16a	26.69±0.13b	26.54±0.10b
USFA ∑	92.90±0.39a	90.29±0.22b	92.73±0.33a

Values are Means±SD of three determinations; values followed by different letters for each row are significantly different ($p<0.05$)

SFA saturated fatty acids, *MUFA* monounsaturated fatty acids, *PUFA* polyunsaturated fatty acids, *USFA* unsaturated fatty acids

network needs to be trained using a training algorithm such as back propagation, cascade correlation, and conjugate gradient. Basically, the objective of training patterns is to reduce the global error, defined below (Rumelhart et al. 1986):

$$\mathrm{SSE} = \sum_{i=1}^{n_p} \sum_{j=1}^{n_o} \left(T_{\mathrm{pj}} - O_{\mathrm{pj}}\right)^2 \tag{2}$$

Where T_{pj} is the jth element of the target output related to the pth pattern, O_{pj} is the computed output of jth neuron related to the pth pattern, n_p is the number of patterns, and n_o is the number of neurons in the output layer.

The goal of every training algorithm is to reduce this global error by adjusting the weights and biases.

Table 3 Coefficients of oxidative stability of oil extracted from three canola varieties

	K	*a*	*b*	*c*	*d*	*e*	*f*
Hyolla 401	0.595	−0.101	1.094	3.541	−1.481	4.119	−2.990
Hyolla 420	1.956	−0.044	0.030	0.01	−0.128	0.840	−0.010
RGS003	1.965	−0.038	0.012	0.016	−0.017	0.672	−0.080

Radial Basis Function Networks

Radial basis function networks have a very strong mathematical foundation rooted in regularization theory for solving ill-conditioned problems. Such networks, almost invariably, consist of three layers: a transparent input layer, a hidden layer with sufficiently large number of nodes, and an output layer (Fig. 2). As its name implies, radially symmetric basis function is used as activation functions of hidden nodes. The transformation from the input nodes to the hidden nodes is a nonlinear one, and training of this portion of the network is generally accomplished in an unsupervised fashion. The training of the network parameters (weight) between the hidden and output layers occurs in a supervised fashion based on target outputs.

The general mathematical form of the output nodes in an RBF network is as follows:

$$c_j(x) = \sum_{i=1}^{k} w_{\mathrm{ji}} \phi(\|(x - \mu_i\|; \sigma_i) \tag{3}$$

Where $c_j(x)$ is the function corresponding to the jth output unit and is a linear combination of k radial basis functions (ϕ) with center μ_i and bandwidth σ_i. Also, w_{ji} is the weight corresponding to the jth class and ith center. The functions $\phi(\|(x - \mu_i)\|$ are called as radial basis functions and $\|\cdot\|$ denotes Euclidean distance (Rumelhart et al. 1986).

The most widely used method of estimating the centers and bandwidths is to use an unsupervised technique called

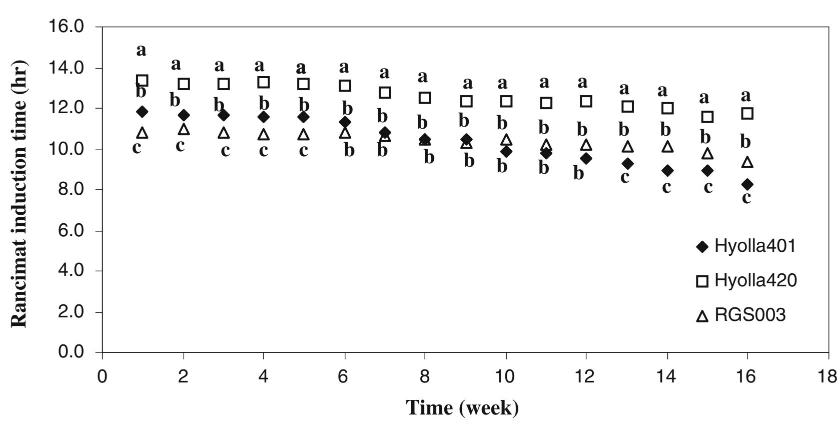

Fig. 5 Effect of storage time on oxidative stability of different canola oils with Rancimat tests

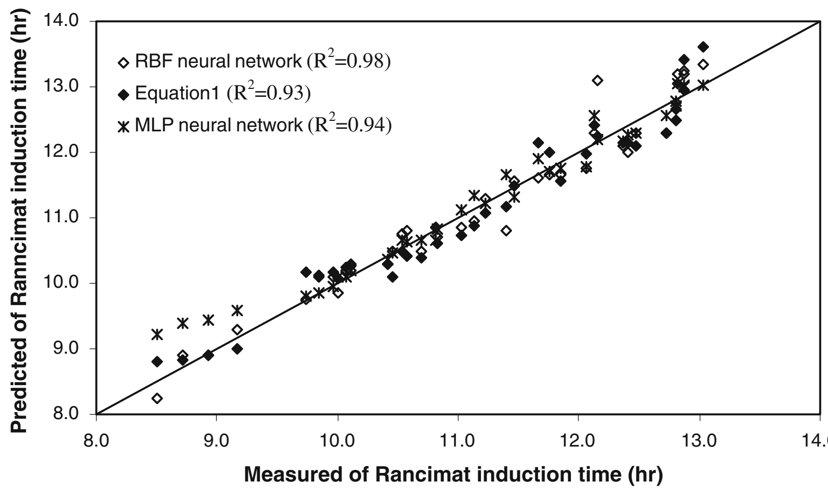

Fig. 6 Comparison between measured and Predicted values of oxidative stability in canola oil by Eq. 1, MLP, and RBF neural networks for training data

the k-nearest-neighbor rule. The error-correction learning described in the multi-layer perceptron section is normally used, but this problem is easier because the output unit is normally linear, so convergence is faster.

Training the Neural Networks

Experimental data from this study were used to train and test two artificial neural network models (MLP and RBF) for prediction of oxidative stability of oil extracted from three canola varieties. Three canola varieties were used in this study, and each variety was analyzed during 16 weeks of storage. For getting accurate results, five replicated tests were done for each variety. Therefore, 240 samples were analyzed in this study. For convergence and avoiding the over-learning of the ANNs, the available training data were split in two subsets: 80% of the patterns for training and 20% for cross-validation. Thus, the data were split into three subsets: training (64%), cross-validation (16%), and test data (20%). The range of train data (including train and cross-validation sets) and test data are presented in Table 1.

The data set was shuffled, 80% of which was used for the learning process and the 20% sets were used for validation, respectively.

The number of neurons in input and output layers depends on independent and dependent variables, respectively. The oxidative stability of canola oils was considered as dependent variable, and independent variables were selected as time (in week), variety, C14:0, C16:0, C18:0, C20:0, C18:1, C18:2, C18:3, and C22:1 fatty acid content. Therefore, one and ten neurons were devoted to output and input layers, respectively. The number of neurons in the hidden layer and the parameter α (momentum coefficient) were determined by calibration through several run tests. At the first step, the number of hidden layer was fixed to 1, because neural network with one hidden layer and sufficient neurons can model every complex non-linear problem (Rumelhart et al. 1986). Then, by choosing various number of nurons in hidden layer, the root mean square error (RMSE) of predicted values were computed and the variation of RMSE of prediction against the number of neurons in hidden layer were plotted. Figures 3 and 4

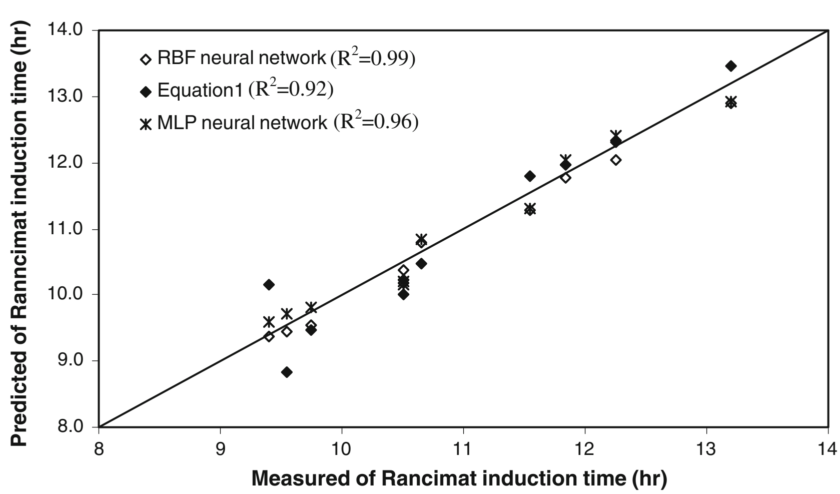

Fig. 7 Comparison between measured and Predicted values of oxidative stability in canola oil by Eq. 1, MLP, and RBF neural networks for validation data

Table 4 Statistical result obtained for Eq. 1, MLP, and RBF networks

Model	R^2	MSE
Eq. 1	0.92	0.165
MLP	0.96	0.05
RBF	0.99	0.04

shows the variation of RMSE of prediction against the number of neurons in hidden layer of MLP and RBF neural networks, respectively. It was found that the minimum error occures by choosing four and seven neurons for MLP and RBF neural networks, respectively. Also, the selection of the ANNs architecures was based on the employment of selection algorithm integreated in the intelligent problem solver module of Statistica Neural Networks software. It automatically determines the network complexity using a variety of algorithms for different network types.

Various activation functions were tested for MLP neural networks and the sigmoid function presented the best results. The number of epoch for converging the RBF is 4,800, while in MLP neural networks is 7,800. So convergence is faster in RBF neural networks than MLP neural networks.

Result and Discussion

Fatty Acids Composition

The fatty acids profile of the oil extracted from different canola varieties is presented in Table 2.The main composition of canola oil are saturated fatty acids (C14:0, C16:0, C18:0, and C20:0), mono unsaturated fatty acids (C16:1, C18:1, C20:1, and C22:1) and poly unsaturated fatty acids (C18:2 and C18:3). The result showed that palmetic acid was the major saturated fatty acid in all three varieties and the highest level was found in Hyolla420 variety. The Hyolla420 showed the highest level of total saturated fatty acids compared to other varieties but there was no significant difference between saturated fatty acid in other two varieties ($p>0.05$). It has been confirmed that the unsaturated fatty acids were predominant in all varieties. However, Oleic acid (ω-9) was the major fatty acid in all three varieties and its content ranged between 61.35% and 64%. The highest level of oleic acid was found in RGS003 variety. The range of oleic acid in canola varieties has been reported between 51% and70% (Codex Alimentarius Commission 2001). Among these varieties, RGS003 showed the highest level of total monounsaturated fatty acid (66.19%), whereas the highest level of total polyunsaturated fatty acids was observed in Hyolla401 variety (27.62%). The linoliec acid in canola ranged between 15.0% and 30.0% (Codex Alimentarius Commission 2001). Based on our study, the seeds of all three varieties had similar (but not identical) fatty acid compositions and contained low amounts of saturated fatty acids.

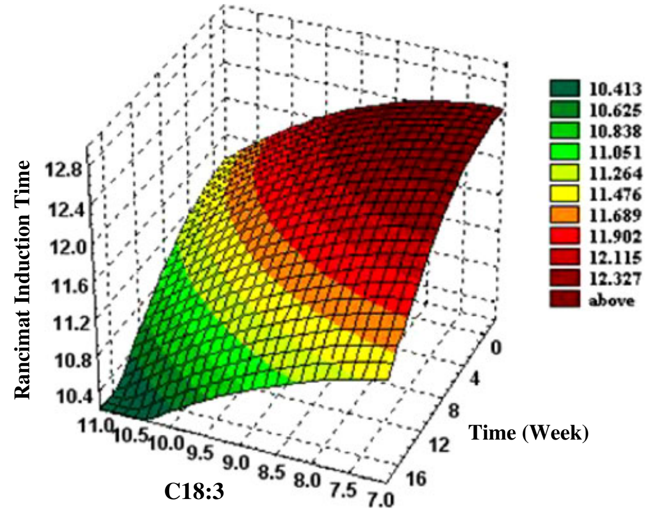

Fig. 8 Response surface of ANN for oxidative stability of canola oil against C18:3 and time

Oxidative Stability

Figure 5 shows the results of Rancimat induction time in the three canola varities during 16 weeks of storage. The Rancimat instruments measure the increase in the conductivity of deionized water resulting from trapped volatile oxidation products produced when the oil product is heated under a flow of air. Hyolla420 showed the highest induction time during 16-week storage; that means, this variety has more stability under accelerated condition of Rancimat test. There were significant differences between

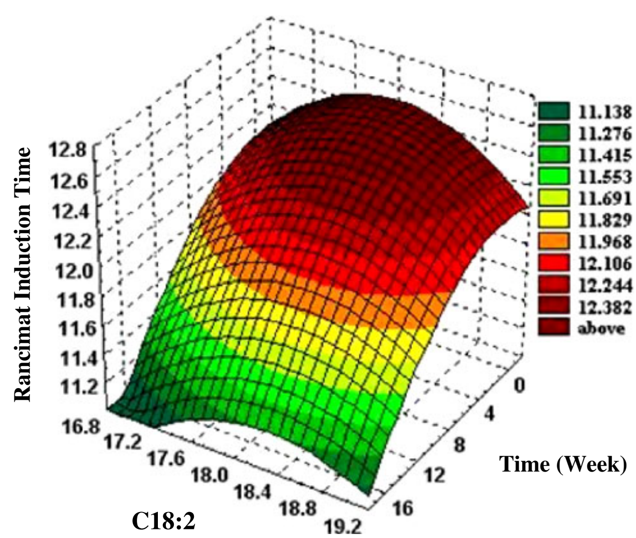

Fig. 9 Response surface of ANN for oxidative stability of canola oils against C18:2 and time

oil stability of different variety during 16-week storage ($p<0.05$).The di- and triunsaturated fatty acid chains contain the most reactive sites for initiation of the autoxidation chain reaction sequence. Oxidation stability does not correlate with the total number of double bonds, but with the total number of bis-allylic sites (the methylene CH directly adjacent to the two double bonds). These sites react with oxygen via the autoxidation mechanism with the classical radical chain reaction steps of initiation, propagation, chain branching, and termination (Gunstone 2002).

Modeling of Oxidative Stability of Canola Oils

The coefficient and exponents of Eq. 1 (K, a, b, c, d, e, and f) for each oxidative stability of canola oils were calculated by regression analysis and presented in Table 3.

The oxidative stability of canola oils predicted with the Eq. 1, MLP and RBF networks are compared to observed oxidative stability of canola oils in Figs. 6 and 7 for training and validation data, respectively.

These models (Eq. 1, MLP and RBF networks) were compared based on the R^2) and MSE and the results are shown in Table 4. These results demonstrate that the agreement is very good in RBF neural network, and this model tracks the observed oxidative stability of canola oils well throughout the various conditions.

It is assumed that the model with lowest MSE and highest R^2 is best to describe oxidative stability of canola oils behavior. Therefore, the suitable model to describe stability of canola oil was found to be RBF neural network. It should be kept in mind that these models are valid for the range of data presented in Table 2.

In order to study the behavior of the neural network, the response surface of canola oil stability aginst two important input variable (C18:3 and C18:2 content with time) was plotted and results are presented in Figs. 8 and 9. This gives an idea of how the network output alters in response to the two input variables. It is clear that oxidative stability of canola oils decreases with increase of time and C18:3 content. However, stability of canola oil increased with increasing C18:2 content up to 18.4% and decreased afterward. It has been reported that oxidizability is the primary factor explaining differences in induction time for biodiesel derived from soy and canola oil samples. Oxidizability (McCormick et al. 2007) defined as:

$$Oxidizability = [0.02(\%oleic) + linoleic + 2(\%linolenic)]/100 \qquad (2)$$

This parameter applies only to biodiesel or fat containing predominantly 18 carbon fatty acid chains. The coefficients for oleic (C18:1), linoleic (C18:2), and linolenic (C18:3) fatty esters are proportional to the relative rates of oxidation of these compounds (Cosgrove et al. 1987). It can be concluded that the linolenic acid content in canola oil had great influence on oil stability. Figure 8 shows response surface of oil stability against C18:3 content and time. The results showed that as the C18:3 content was increased, the stability decreased over time.

Conclusions

The oxidatative stability of vegetable oils is a complex non-linear phenomenon which depends on several factors. ANNs are mathematical models whose architecture has been inspired by biological neural networks. ANNs are very appropriate for the modeling of non-linear processes. The advantage of ANNs over conventional method like Rancimat test is time and cost saving. Also, the ANNs can consider more input parameters and the performance is better than conventional methods. The result of this study showed that the suitable model to describe stability of canola oil was RBF neural network with the R^2 0.99 and MSE of 0.04. Moreover, the result showed that the fatty acid composition especially C18:3 content and time had great effect on oxidative stability of canola oil.Hyolla420 showed the highest induction time during 16-week storage; that means, this variety has more stability under accelerated condition of Rancimat test.

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