

# Predictive machine learning of soybean oil epoxidizing reactions using artificial neural networks

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## Abstract

Experimental data on the epoxidation process of soybean oil with hydrogen peroxide/acetic anhydride were analyzed. This study utilizes experimental data to construct a training dataset for neural network training. Post-training, the neural network facilitates the optimization of epoxy curing reaction parameters, monitors its evolution, and refines the epoxy product synthesis process. Furthermore, a novel methodology has been devised to calculate the outcomes of epoxyation in unsaturated compound mixtures. This method empowers precise control over the epoxyation process at the synthesis phase, under specific reaction conditions, and elevates the technology involved in epoxy output production.

## Keywords

Neural network, Optimization, Soybean oil, Epoxidizing

## 1. Introduction

Modern information technologies enable the solution of a wide range of practical tasks that involve the procedure of deep machine learning and subsequent outcome testing [1]. At present, there are already many areas of human activity where diverse computational tasks have been successfully solved using artificial neural networks. For instance, by analyzing the electroencephalogram of a person expressing a particular phrase, using deep feedforward neural networks, it has been possible to build a human speech synthesizer [2]. Such a synthesizer is capable of guessing and articulating certain fixed expressions used by a person in the past but not recalled during the operation of the synthesizer. The use of neural networks in medicine is essentially a separate branch of science and technology: they can be used for interpreting X-ray images, making it easy to determine the degree of bone wear and tear in a person, as well as artificial implants, fasteners, and prosthetics injected into the patient's body [3].

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Chemists and biologists use neural networks to predict the outcomes of chemical and biological processes, select optimal conditions for such processes, and assess the quantities and types of catalysts [4] or inhibitors [5]. In the field of robotics and automation, neural networks are employed for the approximate calculation and prediction of motion trajectories for automated machines, mechanisms, and manipulators [6], as well as for optimizing the consumption of energy resources and materials. Among the natural sciences, including mathematics, mechanics, materials science, vibrations and waves, spectral analysis, the application of neural networks enables the solution of classical fundamental equations, including those involving Bessel and Neumann special functions [7], which describe real multi-dimensional systems. Numerical solutions to such equations were previously inaccessible.

Neural network structures are utilized in various branches and directions of physics. This includes mathematical modeling of the output of photovoltaic panels and approximating generated power [8], processing and analysis of sensor data and measurement devices, and substance identification.

Neural networks find extensive applications in physical problems related to the propagation, interference, diffraction, and absorption of electromagnetic waves [9, 10], as well as in ultrasound for biological research [11] and for the study of massive solid media [12].

The main goal and principle of green chemistry are the utilization of renewable, environmentally friendly raw materials, which will contribute to reducing biodegradation and the toxicity of industrial production [13]. The polymerization of soybean oil is essential for the creation of polymers used in the production of printing and textile inks. Epoxydized oils possess unique chemical properties and, therefore, have a wide range of applications. They are used to enhance the operational quality of rubber, as components for producing photographic films, packaging materials in medicine, and in the production of food products.

There is an urgent need for a fast and accurate method for identifying the true content of oil mixtures. In this study, Raman spectroscopy, combined with three deep learning models (CNN-LSTM, enhanced AlexNet, and ResNet), was used for the simultaneous determination of the quantities of extra virgin olive oil (EVOO), soybean oil, and sunflower oil in a blend of olive oils. The research demonstrated that all three deep learning models outperformed traditional chemometric methods in predicting the composition [14]. Currently, active research is being conducted on the synthesis of solid polymer materials using soybean oil with mechanical properties that can be utilized as construction and building materials. Epoxides belong to the group of cyclic ethers - metabolites that are often formed by cytochromes, acting on aromatic or double bonds. The specific site on a molecule that undergoes epoxidation is called the site of epoxidation (SOE). Thus, artificial intelligence methods significantly enhance the accuracy of SOE molecule identification and the selection of optimal parameters for controlling the epoxidation process. Indeed, this type of learning is facilitated by artificial neural networks (ANN), which are essentially a technical software implementation of the biological neural structure in the human brain. Such neural networks are also commonly referred to as connection systems because, for such ANNs, a fundamental component is the system of connections and links that enable one neuron to establish pathways for the propagation of signals to other neurons and, conversely, receive similar signals from them. Epoxy oils are widely used in the production of polyvinyl chloride (PVC) and polymers based on it because epoxy oils are among the best stabilizers and plasticizers for such polymers. Epoxy oils offer specific advantages

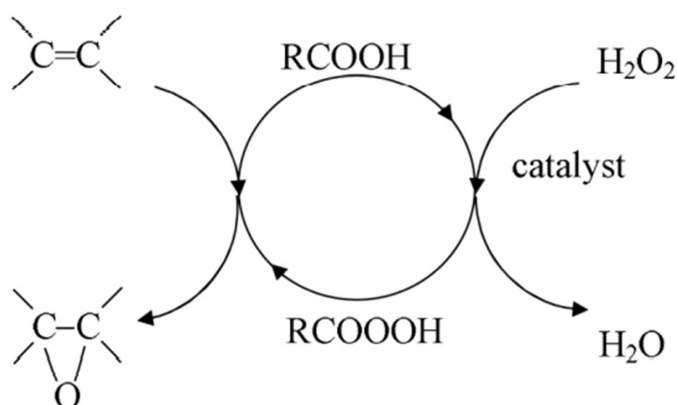
over stabilizers of other types. For example, adding epoxy stabilizers to a polymer significantly increases its thermal stability and provides resistance to ionizing radiation. Epoxy oils can be used as curing agents and binders (in compositions with various oligomers) as well as stabilizers (in PVC compositions). The production and use of epoxy oils continue to grow steadily as they are integral components of paint and varnish products based on epoxy, cellulose ether oligomers, PVC, and plasticizers for organodispersed coatings.

In this study, a deep learning algorithm is employed for the automatic detection of issues in pipelines containing epoxy oils [15]. In [16] the construction of an electromagnetic field shielding system using a multi-layer perceptron neural network-based system designed for predicting electromagnetic absorption by composite films based on polycarbonate and carbon nanotubes is examined. The proposed system includes 15 different multi-layer perceptron networks [17].

Organic peracids are utilized for the implementation of liquid-phase epoxidation of unsaturated organic substances [18].

## 2. Experimental

Nykulyshyn et al. [19] suggests the use of an epoxidation system consisting of  $H_2O_2$ /acetic acid/catalyst. In this system, the epoxidizing agent is also an organic peracid formed through the interaction of  $H_2O_2$  with organic acid in the presence of a catalyst (figure 1). In this case, the organic acid circulates within the system, with its molecule periodically gaining an additional oxygen atom, which is then transferred to another substance.



**Figure 1:** Epoxidizing of the substance using the  $H_2O_2$ /organic acid/catalyst system.

The authors propose using the anhydride of organic acid instead of organic acid, which will reduce the formation of water in the reaction mixture and accelerate the reaction of nadoctovoic acid formation.

The objective of this study is to improve and modify the technology for obtaining epoxy oils, establish optimal conditions for the economically viable production of epoxy oils that meet quality standards (figure 1).

**Table 1**

Quality indicators of epoxidized oils.

Physical-chemical indicator	Norm for brands (technical conditions)*		
	ST	SU	C
Epoxy number, % (oxyran oxygen content), not less than	6.5	6.4	6.0
Iodine number, g I <sub>2</sub> /100 g, no more than	1.5	2.0	8.0

\* As stabilizers and plasticizers for PVC-based polymers

The practical value of using the process model obtained through neural networks lies in its ability to monitor the quality of epoxy oil during the synthesis stage. The dependencies of the time parameters of the chemical epoxidation reaction on:

- Initial concentration of acetic anhydride
- Initial concentration of hydrogen peroxide
- Initial concentration of ion-exchange resin KU-2x8
- Process temperature.

The obtained dependencies allow researchers the assessment of the optimal concentration of epoxying mixture reagents, process duration, and temperature.

Experimental data indicates that the relationship between the epoxy number of epoxidized soybean oil and temperature at various concentrations of acetic anhydride, hydrogen peroxide, catalyst amount, and process duration exhibits a complex nature. In the initial stages, the reaction rate steadily increases with rising temperature and catalyst concentration. However, at excessively high temperatures (>348 K) and catalyst concentrations (>15 g per 100 cm<sup>3</sup>), there is a possibility of a decrease in the achieved epoxy number due to secondary reactions involving the opening of epoxy cycles.

Therefore, to determine the optimal conditions for the chemical process, it is advisable to construct a mathematical model and apply it to calculate the parameters of such processes. The following assumptions were made during the model creation:

- X1 – concentration of acetic anhydride, wt.%,  $2 < X1 < 9$
- X2 – concentration of hydrogen peroxide 46%, wt.%,  $25 < X2 < 40$
- X3 – amount of catalyst, wt.%,  $x3 < 15$
- X4 – temperature, K,  $333 < X4 < 353$
- X5 – duration of the process, min,  $X5 < 360$

### 3. Methods

For training a neural network using the results of experiments, a target dataset has been created. The neural network's inputs characterize the experimental conditions, while the outputs characterize the final results, specifically the concentrations of reaction products.

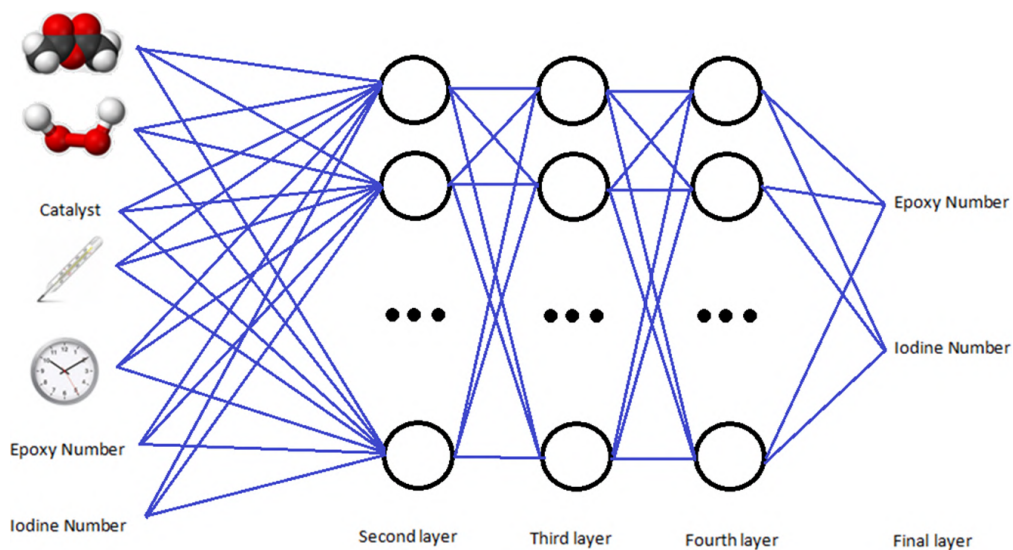
1. The input parameters of the neural network are as follows:

2. Concentration of Acetic Anhydride (Normalized)
3. Concentration of Hydrogen Peroxide (Normalized)
4. Catalyst KU-2×8 (Kat) (Normalized)
5. Normalized Temperature
6. Normalized Reaction Time
7. Initial Epoxy Number (Normalized)
8. Initial Iodine Number (Normalized)

The calculations were made with the assumption that the initial values of the Epoxy and Iodine numbers influence the course of the experiment. The experimental data sample did not exceed 400 minutes, so normalization was performed based on this time. The reactions were conducted in a temperature range of 423 to 443 K. The output parameters of the neural network are as follows:

1. Final Epoxy Number (Normalized)
2. Final Iodine Number (Normalized)

In figure 2, a five-layer neural network is depicted. It was trained using input experimental data obtained during the epoxidation of soybean oil. Seven input parameters were selected for the neural network, and the values of each input parameter were normalized to 1. The neural network's output layer consists of 2 neurons. This presented neural network includes three hidden layers, each of which contains 20 neurons.

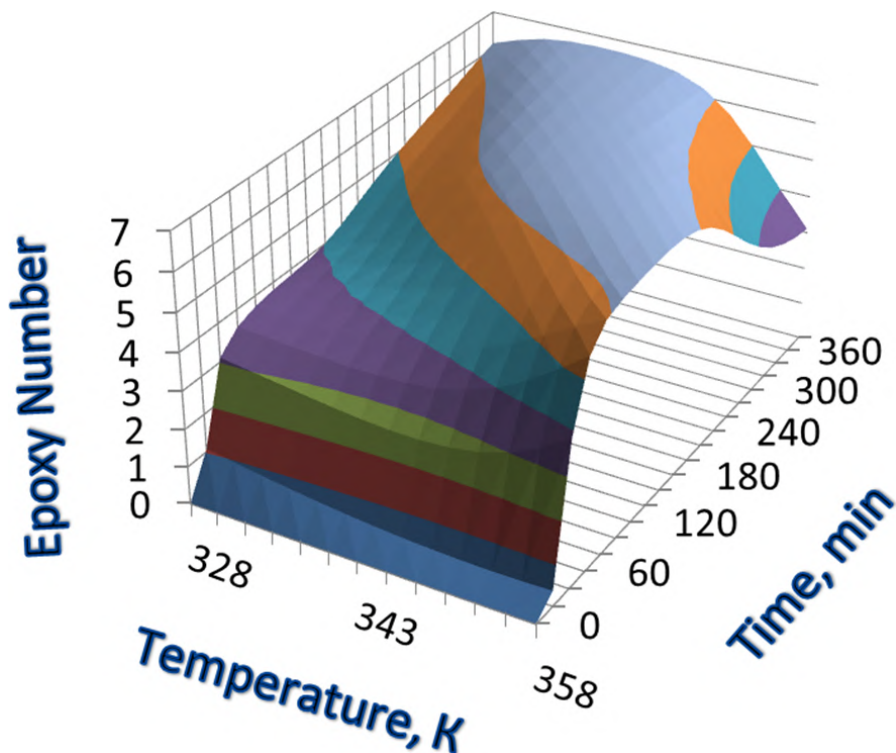


**Figure 2:** A five-layer neural network.

## 4. Results and discussion

As observed in figure 3, which was generated after training the neural network, an increase in reaction temperature leads to a rapid rise in the Epoxy Number within the first 100 minutes of

the reaction. Further progression of the reaction results in a plateau of Epoxy Number values, and continued reaction may even lead to a significant decrease.



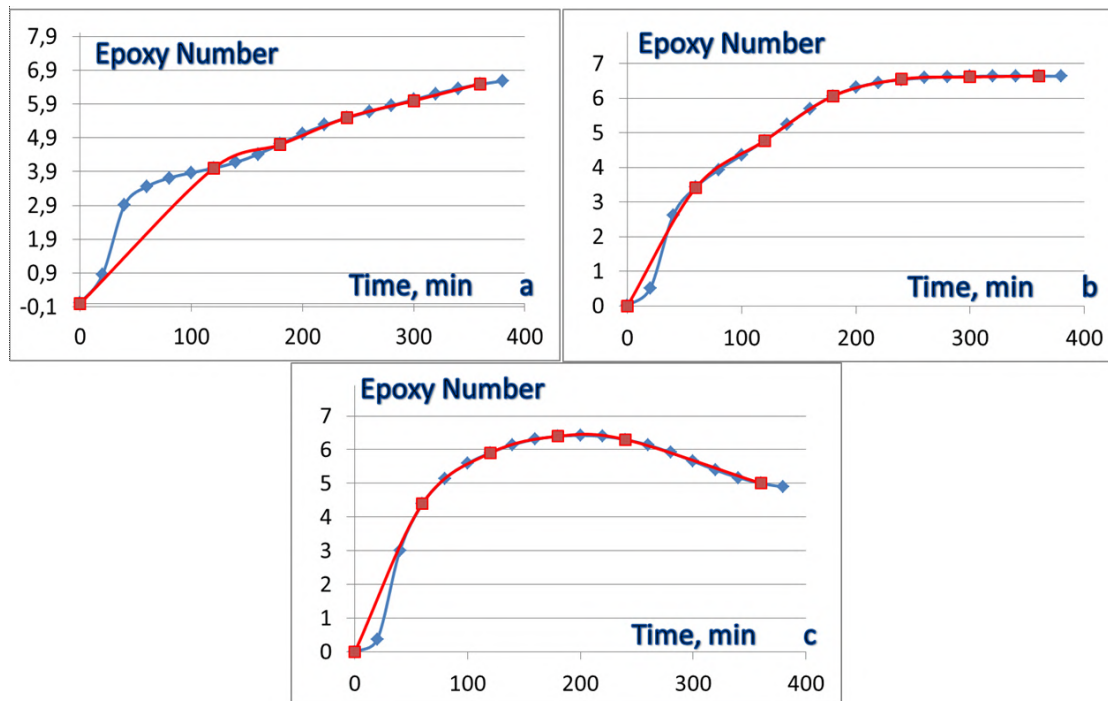
**Figure 3:** The relationship between the Epoxy Number and the reaction temperature and reaction time during the epoxidation of soybean oil. The calculations were conducted while interacting a soybean oil solution in toluene with the epoxidation system consisting of  $H_2O_2$ , acetic anhydride, and a catalyst. The concentrations used were as follows: Acetic anhydride concentration = 5.72 wt.%, Hydrogen peroxide concentration = 33.1 wt.%, and catalyst concentration = 5 wt.%.

The training of the neural network is accomplished using a training mechanism. To find the minimum of the error function, the backpropagation algorithm is applied, utilizing the stochastic gradient descent method.

At lower reaction temperatures, within the initial 60 minutes, the Epoxy Number increases rapidly to values of 3-4, with subsequent growth occurring more slowly. The data obtained during the experiment are limited in both quantity and the range of argument values. However, through neural network training, the system extrapolates the obtained values to create a more detailed graph by extending the range of argument values.

Figure 4 provides a comparison between experimental data and data obtained after training the neural network, which covers a wider range. Experimental data is represented by the red lines, while the predictions made by the neural network are shown in blue lines. The calculations were conducted during the interaction of a soybean oil solution in toluene with the epoxidation system involving  $H_2O_2$ , acetic anhydride, and a catalyst. The concentrations used were as follows: Acetic Anhydride concentration = 5.72 wt.%, Hydrogen Peroxide concentration = 33.1

wt.%, and catalyst concentration = 5 wt.%. The reaction temperatures are as follows: a – T=333 K, b – T=343 K, c – T=353 K. Through training, the neural network accurately processes the input data and predicts experimental outcomes beyond the scope of the available experimental data.

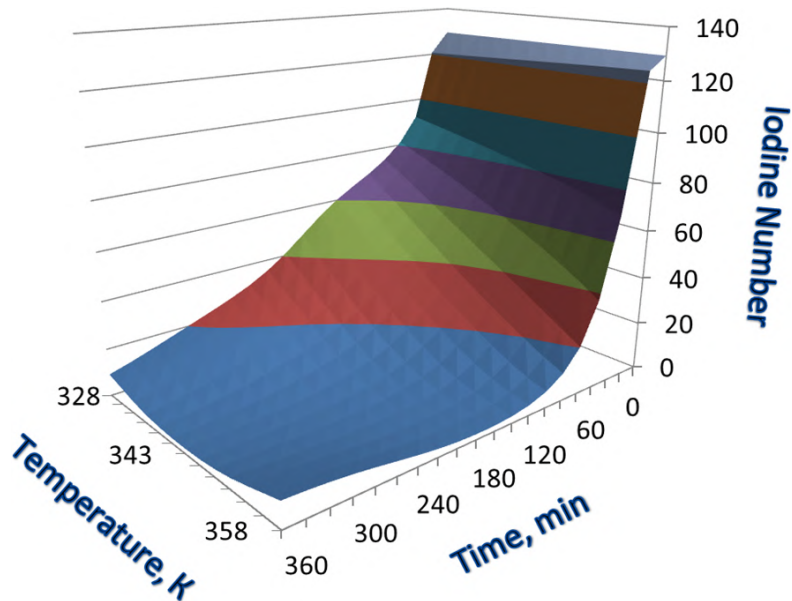


**Figure 4:** Comparison of Epoxy Number dependencies on reaction temperature and reaction time during the epoxidation of soybean oil. The reaction temperatures are as follows: a – T=333 K, b – T=343 K, c – T=353 K

The convexity observed in the dependency, as shown in figure 4a, at small reaction times is a result of the network being trained on intermediate experiment results. In other words, the network considered the entirety of the input data.

As seen in figure 5, which was generated after training the neural network, an increase in reaction temperature leads to a rapid decrease in the Iodine Number. The calculations were carried out during the interaction of a soybean oil solution in toluene with the epoxidation system comprising  $H_2O_2$ , acetic anhydride, and a catalyst. The concentrations used were as follows: Acetic anhydride concentration = 5.72 wt.%, Hydrogen peroxide concentration = 33.1 wt.%, and catalyst concentration = 5 wt.%. However, with a longer reaction time, there is a slight rebound in the Iodine Number towards higher values. At lower reaction temperatures in the mixture, the Iodine Number exhibits a much slower decreasing trend.

Similar to the data in figure 5, the data obtained during the experiment are limited both in quantity and the range of argument values. Nevertheless, through training, the neural network expands the range of argument values and interpolates the obtained values to create a more detailed graph.



**Figure 5:** Dependency of the Iodine Number on the reaction temperature and reaction time during the epoxidation of soybean oil.

In figure 6, a comparison between experimental data and data obtained after neural network training is presented. As a result of training, the neural network accurately reproduces the experimental data and predicts experiment outcomes beyond it.

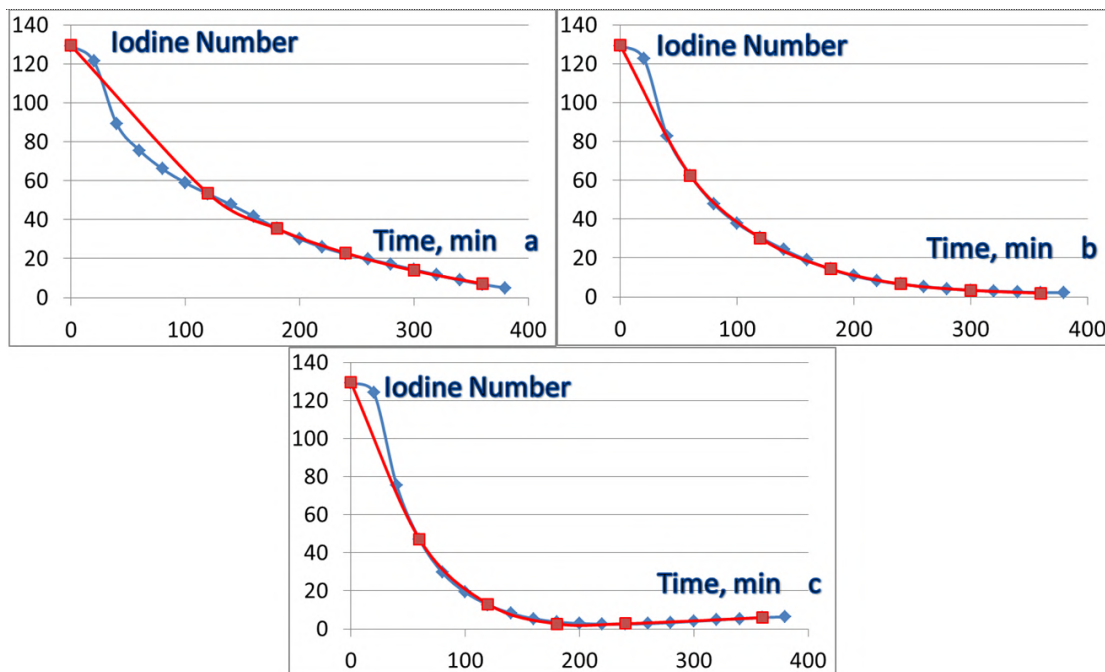
The wave-like dependencies observed in figure 5a at small reaction times and the convex shapes in dependencies figure 6(b,c) are predicted by the neural network. The first experimental data point in figure 5a was taken after 120 minutes, and the true dependence of the Iodine Number on time was unknown. However, based on the entire array of experimental data, the neural network predicts that the Iodine Number should vary according to this pattern.

After training, the neural network operates with output functions of multiple variables (in our case, 7 variables) and allows us to examine the projections of calculated (learned) functions onto selected parameter axes that are of interest to us. For example, in figure 7a, the relationship between the Epoxy Number after two hours of the epoxidation reaction of soybean oil and the concentration of Acetic Anhydride (OA) and temperature is shown. The reaction time, the concentration of  $H_2O_2$ , and the catalyst concentration are held constant.

Simultaneously reducing the concentration of OA and increasing the reaction temperature leads to the highest Epoxy Number value (figure 7a). Furthermore, at high OA concentrations and low reaction temperatures, a local maximum in the Epoxy Number dependence is observed within the selected range. The reaction performs the least efficiently at moderate OA concentrations and low reaction temperatures. It is important to note that in this case, we have considered a limited reaction time (time = 120 minutes). With a different reaction time, the dependencies may vary, and the optimal parameters (OA concentration and temperature) may fall within different ranges.

In figure 7b, we observe similar dependencies to those in figure 6, but for the Iodine Number.





**Figure 6:** Comparison of Iodine Number dependencies on reaction temperature and reaction time during the epoxidation of soybean oil. Experimental data is represented by the red lines, while the predictions made by the neural network are shown as blue lines. The calculations were conducted during the interaction of a soybean oil solution in toluene with the epoxidation system consisting of  $H_2O_2$ , acetic anhydride, and a catalyst. The concentrations used were as follows: Acetic Anhydride concentration = 5.72 wt.%, Hydrogen Peroxide concentration = 33.1 wt.%, and catalyst concentration = 5 wt.%. Reaction temperatures are denoted as follows: a –  $T=333$  K, b –  $T=343$  K, c –  $T=353$  K

As seen in the figure, even at the maximum reaction temperature (within the selected range) and the highest OA concentration, the Iodine Number does not reach its minimum value.

Optimal concentrations of OA and reaction temperature were calculated to achieve the maximum Epoxy Number and the minimum Iodine Number during the interaction of a soybean oil solution in toluene with the epoxidation system  $H_2O_2$ /acetic anhydride/catalyst (as shown in figure 7a and figure 7b). Conditions:

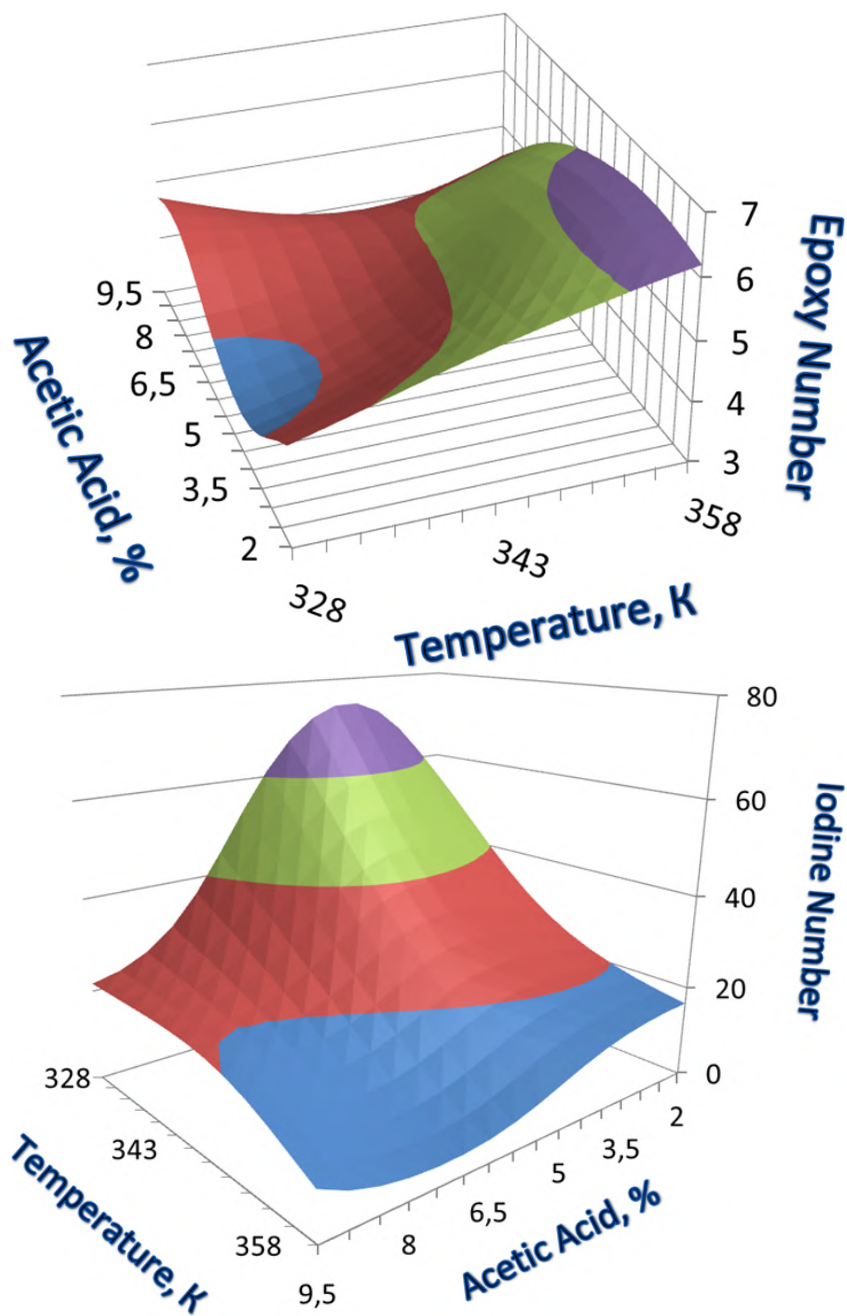
- $X_2 = 33.1$  wt.% (Hydrogen Peroxide concentration)
- $X_3 = 5$  wt.% (Catalyst concentration)
- $X_5 = 120$  minutes (Reaction time)

Optimal Epoxy Number values occur at:

- $X_1 = 3.5$  wt.% (Acetic Anhydride concentration)
- $X_4 = 358$  K (Reaction temperature)

Optimal Iodine Number values occur at:

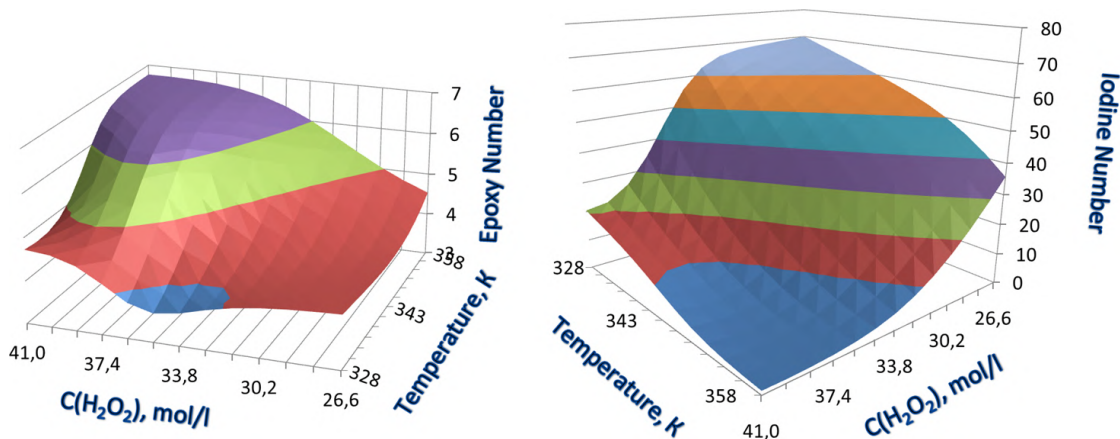
- $X_1 = 7.5$  wt.% (Acetic Anhydride concentration)



**Figure 7:** Dependency of Epoxy Number (a) Iodine Number (b) on the Acetic Anhydride concentration and temperature during the epoxidation of soybean oil. The calculations were carried out during the interaction of a soybean oil solution in toluene with the epoxidation system consisting of  $H_2O_2$ , acetic anhydride, and a catalyst. The catalyst concentration in the mixture was 5 wt.%, the Hydrogen Peroxide concentration was 33.1 wt%, and the reaction time was 120 minutes.

- X4 = 358 K (Reaction temperature)

As seen in figure 8a, the Epoxy Number reaches its maximum values at the highest concentration of H<sub>2</sub>O<sub>2</sub> in the solution and the maximum reaction temperature (within the specified range of input values).



**Figure 8:** Dependency of the Epoxy Number (a) Iodine Number (b) on the concentration of H<sub>2</sub>O<sub>2</sub> and reaction temperature during the epoxidation of soybean oil. The calculations were performed during the interaction of a soybean oil solution in toluene with the epoxidation system consisting of H<sub>2</sub>O<sub>2</sub>, acetic anhydride, and a catalyst. The mixture contained a catalyst concentration of 5 wt.%, Acetic Anhydride concentration of 5.72 wt.%, and a reaction time of 120 minutes

As observed in figure 8b, the Iodine Number reaches its minimum values at the highest concentration of H<sub>2</sub>O<sub>2</sub> in the solution and the maximum reaction temperature (within the specified range of input values).

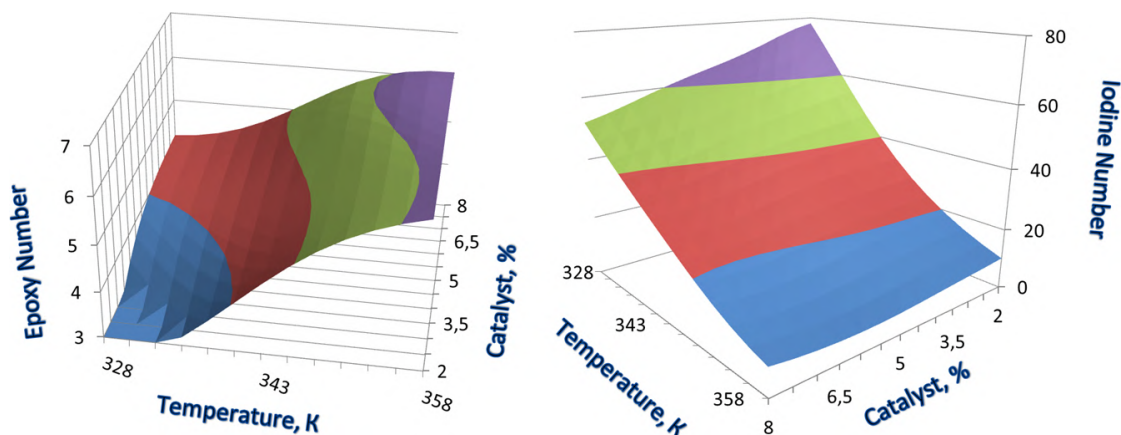
As observed in figure 9a, at the maximum reaction temperature, the maximum Epoxy Number is achieved after 2 hours of reaction across the entire specified range of catalyst concentration values.

As observed in figure 9b, at the maximum reaction temperature, the minimum Iodine Number is achieved after 2 hours of reaction across the entire specified range of catalyst concentration values.

In conclusion, it is important to emphasize that following training on a limited range of input experimental data, the neural network enables the expansion (prediction) of the range of experimental data. The addition of new data to the experimental database, which is used to train the neural network, offers the opportunity to refine the dependencies further.

## 5. Conclusions

The method presented in this study for determining the parameters of the soybean oil epoxidation process has demonstrated the nature of the dependence of these chemical reaction parameters on the concentration of OA and H<sub>2</sub>O<sub>2</sub>, the amount of catalyst, reaction temperature, and reaction time. The use of the neural network clearly illustrates the ability to quantitatively



**Figure 9:** Dependency of the Epoxy Number (a) Iodine Number (b) on the catalyst concentration and reaction temperature during the epoxidation of soybean oil. The calculations were conducted during the interaction of a soybean oil solution in toluene with the epoxidation system involving  $H_2O_2$ , acetic anhydride, and a catalyst. The mixture had a Hydrogen Peroxide concentration of 33.1 wt.%, Acetic Anhydride concentration of 5.72 wt.%, and a reaction time of 120 minutes

predict the outcome of soybean oil epoxidation with a limited sample of experimental data. The prediction of results includes determining the epoxy number, iodine number, and additional information about the progress of the reaction.

The application of the trained neural network allows for the determination of optimal conditions for conducting the epoxidation process of oil using the epoxidation system, which consists of acetic anhydride, hydrogen peroxide, and the catalyst KU-2x8, with specific values for duration and temperature.

While experimental studies were conducted on soybean oil, the dependencies obtained in this work are also applicable to other vegetable oils such as castor oil, rapeseed oil, flaxseed oil, sunflower oil, and olive oil. Experiments on these vegetable oils are already underway.

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