

RESEARCH ARTICLE

Modeling of decolorization dyes by ozonation techniques using Levenberg-Marquardt neural network

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Article history Received 25 May 2017 Accepted 4 October 2017

Abstract

Acid Black 1 (AB1) and Acid Yellow 19 (AY19) are synthetic dyes widely used in industries such as textiles and cosmetics, which have complex structures. Special techniques are needed to degrade the dye before it goes out into the environment. One of the techniques is to Advance Oxidation Process (AOPs) using ozonation process. The process requires a combination of the values of four parameters to obtain the maximum decolorization percentage. The parameters are dye concentration (mg / L), ozone concentration (mg / L), pH, and temperature (°C). Laboratory experiments to optimise the parameters decolorization percentage is costly and takes a lot of times. This research proposes a simulation modelling study using a Levenberg-Marquardt neural network (LM-NN) to obtain the combination of parameters that can yield maximum decolorization percentage. The experimental results with the ozonation method obtained decolorization percentage values AB1 and AY19 were 93% and 98%. The percentage of decolorization was produced from a combination of four parameters, with values of 740 mg/L, 13 mg/L, 6.20, and 38°C, while for the AY19 was 830 mg/L, 15 mg/L, 6.20, and 39°C. The result of prediction of AB1 and AY19 by using LM-NN model in a sequence is 95,25% and 99,99%. The condition was generated from the combination of the four parameters, for AB1 the values of each parameter were 707.50 mg/L, 12.70 mg/L, 7.50 and 38°C, while the AY19 was 587.50 mg/L, 15.50 mg/L, 7,65, and 33°C. The result of experimental and predicted using the LM-NN model, referring to statistical test results with 95% confidence level, indicating that the percentage of decolorization did not occur of difference significantly (p-value> 0.05), both for AB1 and AY19. It shows that modelling with LM-NN can be used to predict the maximum decolorization percentage with predetermined parameters.

Keywords: Decolorization, ozonation, acid black 1, acid yellow 19, Levenberg-Marquardt, neural network

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INTRODUCTION

Waste generated from the textile industry process contains various dyes with high concentration. Dyes contained in the waste of this textile industry, among others, is the azo class dyes. The azo dyes has a high degree of stability and very complex structure. Some of the commonly used azo dyes include Acid Black 1 (AB 1) and Acid Yellow 19 (AY 19). Azo dyes contained in industrial waste are very difficult to handle using simple technique. Therefore, specials treatment is required [1]. Currently, various methods for handling textile waste are being developed so that the waste coming out of the industry is not harmful to the life of water biota.

Azo dye waste discharged into rivers or aquatic ecosystems are capable of influencing water transparency, which affects the penetration of sunlight from rivers, toxic, and mutagenic to organisms or aquatic biota [2]. Azo dyes easily dissolved in water, when discharged into the aquatic ecosystem will be mixed in the waters, accumulate and able to enter the body of water resulting bioaccumulation. Physically, the azo dye that enters the river makes the river water becomes coloured and block the light entering the body of water, thus affecting the process of photosynthesis of phytoplankton or water plants which then will also affect zooplankton and other aquatic organisms. Chemically, the substance is capable in reducing oxygen levels in the contaminated waters and can result in the death of water biota. In addition, on aquatic grounds, azo dyestuffs that have been overhauled by anaerobic microorganisms may produce aromatic amine compounds whose toxicity levels may be more dangerous than the azo dye itself [3]. One example of compounds formed in the anaerobic process is chloroaniline, which can interfere with human health because it is thought to affect the respiratory, urogenital, and neurological disorders [4].

To overcome the problem of dye waste an effective and efficient technique is required. One of the techniques being developed today is the use of Advance Oxidizing Process. According to Kasiri et.al[5] the method of Advanced Oxidation Process (AOPs), has the main advantage of being able to degrade and completely disentangle nonbiodegradable hazardous compounds in waste through oxidation (oxidative degradation). This technique uses chemicals such as oxidising agents or reducing agents as a treatment based on the formation of hydroxyl radicals produced in the oxidation process. Some examples of this technique are the use of ozone[6], hydrogen peroxide fenton [2], photocatalyst[4], ultrasonic radiation [7], UV/H₂O₂ [8]. In the process of AOPs technology, the principle is a combination of several reducing agents to produce free radicals \bullet OH. These hydroxyl radicals are selective against pollutants so they are effectively used to handle textile industry waste. The use of ozone has a high oxidation

rate of 2.08 V so it can produce free radicals \bullet OH. This makes free radicals OH very easy to oxidise both organic and non-organic compounds [9].

Some of the variables that affect the degradation efficiency process by using ozonation include pH, an initial concentration of the dyestuff, ozone concentration, and temperature. The colours AB 1 and AY 19 have the different amount of azo bonds, AB 1 has two azo bonds while AY 19 has one azo bond. This difference in the number of azo groups will affect the degradation efficiency of the dyestuff under the same treatment conditions. In acidic conditions, the ozone molecule is highly selective in attacking the active group of the dyestuff, such as the presence of double bonds or aromatic groups present in the dyestuff. In the basic conditions of hydroxyl radicals are produced from the process of decomposition or decomposition of the dyestuff itself. Under alkaline pH conditions up to neutral pH conditions there may be reactions which can not produce radical reactions of hydroxyl formation. The pH value of the solution will decrease with the formation of the hydroxyl compound from the ozonation process. The reaction mechanism of the ozonation process as reported by Basturk et.al [3], that ozone reacts directly with the dyestuff through the ozonolysis process and the formation of a radical chain. The reaction between ozone and anion hydroxide to intermediate radicals and hydroxyl radicals is essential in the process of dye oxidation

The degradation efficiency of the ozonation process can be done systematically by testing each variable by making the other variable fixed. This is repeatedly done for several times the experiment to produce maximum efficiency. Repetition of course will require time and cost a lot. In order for this repetition process can be minimised, then one method is done by using Artificial Neural Network (ANN) modelling. By using ANN modelling it can reduce time and cost. In addition, using this ANN we can predict linearity, interaction, quadratic factor (\mathbb{R}^2), and provide a modelling of the experiment in detail.

 Table 1. Characteristics of AB1 and AY19.

Attributes	Acid Black 1	Acid Yellow 19				
Structure	O2N O H NH2 O H N NH2 O H N NaO35 SO3Na	$ \begin{array}{c} \underset{H}{\overset{SO_3Na}{\underset{H}{\overset{H}{\underset{H}{\overset{O}{\underset{H}{\underset{H}{\overset{O}{\underset{H}{\underset{H}{\overset{O}{\underset{H}{\underset{H}{\overset{O}{\underset{H}{\underset{H}{\overset{H}{\underset{H}{\underset{H}{\overset{O}{\underset{H}{\underset{H}{\overset{O}{\underset{H}{\underset{H}{\overset{O}{\underset{H}{\underset{H}{\overset{O}{\underset{H}{\underset{H}{\overset{O}{\underset{H}{\underset{H}{\overset{O}{\underset{H}{\underset{H}{\overset{O}{\underset{H}{\underset{H}{\overset{O}{\underset{H}{\underset{H}{\overset{O}{\underset{H}{\underset{H}{\overset{O}{\underset{H}{\underset{H}{\overset{O}{\underset{H}{\underset{H}{\overset{O}{\underset{H}{\underset{H}{\overset{O}{\underset{H}{\underset{H}{\underset{H}{\underset{H}{\underset{H}{\underset{H}{\underset{H}{\underset$				
Formula	$C_{22}H_{14}N_6Na_2O_9S_2$	$C_{20}H_{12}C_{12}N_4Na_2O_7S_2$				
Number of Azo	Double	Single				
Mr (g/mol)	616.49	551.29				
λ _{max} (nm)	618	401				

This neural network modelling has been widely used in the field of chemical engineering, for example for steady state and dynamic modelling, identification processes, and maximum yield. The advantages of this neural network modelling are that this model can be based solely on historical data of the input-output process, requiring no phenomenological-related knowledge for model development, and can simulate the relationship between multiple input-multiple outputs. Modeling using ANN is a modelling using the principle of artificial neural network, where a data processing system to obtain optimal operating conditions. The operating conditions are to create parameters of the input variables to produce maximum performance. The purpose of this optimisation process is to maximise the process performance simultaneously to obtain optimal process input.

Research on modelling prediction of dyestuff decolorization percentage that has been done, among others, is Kasiri et.al [5]. The study conducted AB 1 and AY 19 modelling using Response Surface Methodology. The result of this research resulted in the efficiency of decolorization percentage in the sequence of 91,28% and 96,89%. A similar study was conducted by Kousha et.al [1] using the Box-Behnken Design Matrix model to model the efficiency of AB 1 by using S.glaucescens bacteria which resulted in a percentage of decolorization of 94.3%. The methyl orange dye modelling has also been performed by Agarwal [10] with the ANN-Partial Swam Optimization model, which resulted in a percentage of decolorization of 98%.

Subsequent research on modelling using ANN was performed by Fatimah et.al [11]. The research modelled for the prediction of Acid Orange 7 (AO7) dye decolorization percentage, with ANN algorithm used is One Step Secant (OSS). The predicted percentage of optimum decolorization is 80%. The results of predicted percentage of decolorization are greater than using surface methodology method by Kasiri et.al [6] that is equal to 77,91%. The conclusion in the research shows that ANN with OSS training algorithm can give prediction result which is not significantly different with experiment result.

Based on the analysis of some modelling on dye decolorization it still has not been able to produce optimal decolorization percentage modelling. Referring to this matter in this research do modelling of decolorization percentage for dyes AB1 and AY19, using ANN. ANN algorithm used is Levenberg-Marquardt neural network (LM-NN). The algorithm has advantages over the speed of the training process and provides relatively better prediction results [12], so with the LM-NN model the experimental process will be more effective and efficient.

EXPERIMENTAL

Materials

In this study, the data used are secondary data obtained from research that has been done by Kasiri et.al [13]. The parameters used in analysing this model consist of 4 parameters as input and one parameter as output. The input parameters consist of ozone concentration (mg / L), dyestuff concentration (AB1 and AY19) (mg / L), pH, temperature (0 C), and the output is the percentage value of decolorization.

Method

The steps in modelling the decolorization process AB1 and AY19 can be shown in Figure 1.

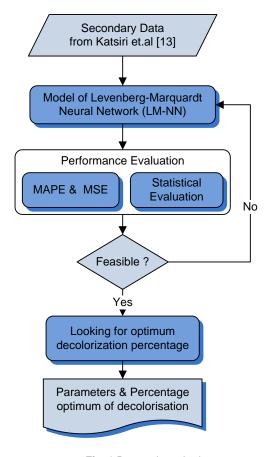


Fig. 1 Research methods.

For decolourization process, ozonation was carried out in a batch reactor, 500 mL in volume, equipped with magnetic stirrer and a

thermometer. Experimental setup for ozonation batch experiments is shown in Figure 2.

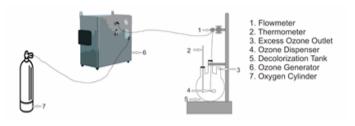


Fig. 2 Ozonation setup.

Levenberg-Marquardt Neural Network Algorithm

The modelling for dye degradation of AB1 and AY1 uses a multilayer architecture with training using the Levenberg-Marquard (LM) algorithm [14]. The LM algorithm is designed using a second derivative approach, with no need to compute the Hessian matrix. In the standard backpropagation algorithm[15], the process of weighting changes using negative gradient descent directly while LM algorithm using Hessian matrix approach (H), in making weight changes. The LM algorithm was developed using Newton's method which can be described as follows [16]:

$$\Delta w(x) = -[\nabla^2 V(x)]^{-1} \cdot \nabla V(x) \tag{1}$$

$$w(t+1) = w(t) + \Delta w(x) \tag{2}$$

where $\nabla^2 V(x)$ is a Hessian matrix. $\nabla V(x)$ is the gradient of V(x). $\Delta w(x)$ is a weight change. The value of V(x) is a sum square error function which can be expressed as follows

$$V(x) = \sum_{i=1}^{N} e_1^2(x)$$
(3)

Furthermore, the gradient and Hessian matrix can be represented in an equation by using the jacobian matrix, J(x), whose content is the first derivative of the network error, e(x).

$$\nabla V(x) = J^{T}(x)e(x) \tag{4}$$

$$\nabla^2 V(x) = J^T(x)J(x) + S(x) \tag{5}$$

where

$$S(x) = \sum_{i=1}^{N} e_i(x) \nabla^2 e_i(x)$$
(6)

Using the gauss-newton method, the equation (1) can be translated in the following equation (7)

$$\Delta w(x) = -[J^{T}(x)J(x)]^{-1}J^{T}(x)e(x)$$
(7)

So the modification of the LM algorithm in determining the weight change can be expressed in equation (8)

$$\Delta w(x) = -[J^T(x)J(x) + \mu I]^{-1}J^T(x)e(x)$$
(8)

The equation for the weight change in the LM algorithm is given in equation (9) below

$$w(t+1) = w(t) - [J^{T}(x)J(x) + \mu I]^{-1}J^{T}(x)e(x)$$
(9)

Evaluation Parameters

Evaluation of LM-NN model performance for decolorization using three parameters, Mean Mean Absolute Percent Error (MAPE), Mean Square Error (MSE) and statistical test. The MAPE parameter can be formulated in equation (10), this parameter to determine LM-NN's ability to make predictions. The MAPE value limit indicates an acceptable prediction result if the value is less than 20%.

$$MAPE = \frac{\sum_{i=1}^{N} \frac{\% Predicted_i - \% Experiment_i}{\% experiment_i}}{N} x100\%$$
(10)

The second parameter is MSE, this value is the sum of quadratic error difference, from the result of decolorization prediction with experiment. MSE can be formulated as shown in equation (11).

$$MSE = \frac{\sum_{i=1}^{N} (\%Predicted_i - \%Experiment_i)^2}{N}$$
(11)

Next, the third parameter is statistical testing using the t-test method. The test was done by using SPSS version 23 software, to find out the significance value of the difference of experiment and prediction result decolorization.

RESULTS AND DISCUSSION

Decolorization model of AB1 and AY19 with ozonation technique using LM-NN has been done well. The successful LM-NN architecture used to model the decolorization uses a multilayer architecture consisting of input layer, hidden layer and output layer. Input layer consists of 4 attributes that include ozone concentration, dye concentration, pH and temperature. The number of hidden layers used is two pieces. The first hidden layer consists of 20 neurons, while the second hidden layer consists of 15 neurons. The last layer is an output layer consisting of one neuron. The LM-NN architecture is as shown in Figure 3.

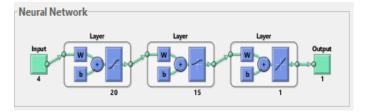


Fig. 3 Architecture of LM-NN.

Performance evaluation is performed using MAPE, MSE and statistical analysis. The results of the dye decolorization test of AB1 and AY19 were performed using experimental data performed by Kasiri et.al [13], as shown in Figure 4 and Figure 5. Referring to the test results shown in Figure 4 and Figure 5, Calculated MAPE and MSE values for each of AB1 and AY19 dye. MAPE is a value that shows the difference between the predicted results between experiments and predicted. If the value is less than 20% then this can still be used for decolorization modelling. MSE is the difference between the mean squared error value between the experiments and the predicted MAPE value results of AB 1 and AY 19 are 0.448% and 0.441%, respectively. The MSE values of decolorization model AB1 and AY19 were 0.756% and 0.700% respectively. Referring to the MAPE and MSE values indicates that LM-NN architecturally feasible to be used to model decolorization AB1 and AY19 is feasible.

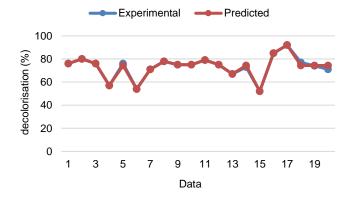


Fig. 4 Percentage of decolorization AB1 experiments and predictions.

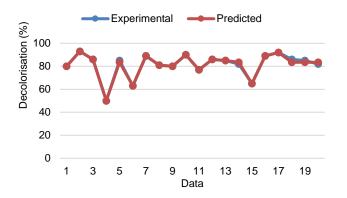


Fig. 5 Percentage of decolorization AY 19 experiments and predictions.

Further evaluation to assess this decolorization model is done by the statistical test. The statistical test of the feasibility of modelling was done using SPSS version 23. Based on the statistical test shown in Table 2. It was found that the p-value significance value of the LM-NN architecture modelling in the dye decolorization of AB1 and AY19 were 1,000 and 0.999, respectively. This suggests that there is no significant difference between prediction and experiment, so that the LM-NN architectural model designed can be used to model the decolorization of the dyestuffs AB1 and AY19. Referring to the result of performance evaluation with the statistical test, then modelling by using LM-NN for dye decolorization is feasible for use.

Based on the obtained LM-NN modelling results, it is used to predict the maximum decolorization percentage by using parameters by the experimental experiments. Parameters used are ozone concentration, dyestuff concentration, pH and temperature. The prediction is done by generating the values of 4 parameters randomly to obtain the combination of parameter values which yield the maximum decolorization percentage. The combination that built of these four parameters amounted to 14656, which was then used for the LM-NN model test. The results of the tests performed are shown in Table 3. The LM-NN model is capable of generating almost 100% decolorization percentages for AY19 of 99.99%, with the following parameters being 587 mg/L, 14.50 mg/L, 7.65, and 33^oC. For AB1 an increase in the percentage of decolorization with successive parameter conditions was 707.50 mg/L, 12.70 mg/L, 7.5 and 38^oC.

The AB1 and AY19 colours are the azo dye. The difference between the two is the amount of bond or azo group that AB1 has more than AY19. The AB1 colour has two azo group bonds, while AY19 has one azo group bond. Based on the experiments done, the AB1 dye has a maximum percentage of decolorization of 93%. This condition occurs when the AB1 concentration of 13 mg / L requires an ozone concentration of 740 mg / L with a pH of 6.20 at 38° C. The successful modelling result with LM-NN architecture has resulted 95.25% decolorization percentage. This prediction produces a better result than the prediction that has been done by Kasiri [13] which uses Response Surface Methodology model that produces a percentage of 91.28%. The increase in the percentage of decolorization occurring in this model was accompanied by an increase in pH of the solution from 6.20 to 7.50 and the decrease of ozone concentration from 740 mg / L to 707.50 mg/L. The basic conditions of hydroxyl radical availability resulting from the process of ozone decomposition are not specific to the AB1 compound so that the presence of ozone concentration decreases [17]. Lu et.al [17] explains that when decolorization takes place in an alkaline atmosphere, the process of forming a hydroxyl radical will increase so that the dyestuff oxidation process will increase. This is by the resulting modelling.

Table 2. Significance test prediction of LM-NN.

		Paired Differences							
		Mean	Std. Deviation	Std. Error Mean	95% Confidence Interval of the Difference		t	df	Sig. (2- tailed)
					Lower	Upper	_		-
AB1	Eksperimental- Predicted	00006	.88372	.15872	-,32421	.32408	.000	30	1.000
AY19	Eksperimental-Predicted	00010	.85077	.15280	31216	.31197	001	30	.999

In AY19 dye based on the experiment result that the maximum percentage of decolorization that happened was 98%. This condition requires 830 mg / L of ozone to decolourise AY19 as much as 15 mg / L at pH 6.20 and temperature 39ºC. The prediction model using LM-NN is capable of producing a percentage of decolorization that is close to 100%. This value is much better than the percentage of decolorization that has been done by Kasiri et.al [13] was 96.89% with the prediction model using Response Surface Methodology. According to Kasiri et.al [13] that at a relatively small concentration of ozone at the alkaline conditions will trigger the addition of hydroxyl formation reaction. This hydroxyl radical may increase the oxidation activity of the ozone molecule against the AY19 molecule. This will be accompanied by an increase in the efficiency of the percentage of decolorization. The AY19 dye only has one azo group, so when compared with the percentage of decolorization value at AB1 of 95.25%, the value is greater AY19. This is due to the presence of azo bond or a group of 2 pieces, will cause the existence of clouds of electrons in the cluster can interfere with the process of formation of hydroxyl radicals. The difference between the experiment and the apparent prediction is caused by several factors. The first factor is the pH value, at experiment the pH value is 6.20 while the prediction pH value is 7.65. This difference is accompanied by a decrease in the concentration of ozone and concentration AY19. Lu et.al [17] explained that when decolorization takes place at an alkaline atmosphere the process of forming the hydroxyl radical increases so that the dye oxidation process will increase. It is also explained by

Behind et al [18] that the hydroxyl free radicals generated from the oxidation process will break the double bond optimally when under basic pH conditions. From that reason, a maximum decolorization percentage will be generated. This is indicated by the percentage of decolorization value of 99.99%. The azo groups present in the AY19 dye as amount one so that the formation of the hydroxyl radical is made easier and causes the dye oxidation process of AY19 to increase.

Variable		iction lue	Experimental Value		
	AB1	AY19	AB1	AY19	
Ozonase concentration (mg/L)	707.5	587.50	740.0	830.00	
Initial Dye concentration (mg/L)	12.70	14.50	13.00	15.00	
Initial PH	7.50	7.65	6.20	6.20	
Temperatur (°C)	38.00	33.00	38.00	39.00	
Decolorization efficiency (%)	95.25	99.99	93.00	98.00	

The result of modelling that has been successfully done is tested the level of confidence by using the statistical test. Testing is done by comparing the percentage of decolorization experiments and predictions. Based on the experimental results obtained value percentage decolorization AB1 of 93% while the prediction of 95.25%. The consecutive decolorization percentage of AY19 between experiment and prediction is 98% and 99.99%. The results of statistical tests show that the percentage value of decolorization between experiments and predictions is not significantly different. This is indicated by the value of p-value obtained in the sequence of dyestuffs AB1 and AY19 is 0.430 and 0.367, as shown in Table 4. Based on this analysis then with LM-NN modelling used can provide better treatment solution to get the results the maximum percentage of decolorization.

Table 4. Significance test percentage of decolorization AB1 and AY19 between experiments and prediction

		Paired Differences							
		Mean	Std.	Std. Error		Confidence Interval f the Difference		df	Sig. (2- tailed)
		Deviation	Mean	Lower	Upper	_			
AB1	Predicted - Experimental	-5.85000	14.93302	6.67825	-24.39179	12.69179	876	4	.430
AY19	Predicted - Experimental	-49.11200	108.15339	48.36766	-183.40217	85.17817	-1.015	4	.367

CONCLUSION

The process of decolourising AB1 and AY19 dyes using ozonation process, successfully modelled using Levenberg-Marquardt Neural Network (LM-NN) model. Modeling using this architecture is effective and significant in making a prediction model of dye decolorization percentage of AB1 and AY19. This is proven by statistical tests obtained p-value values are sequential of 0.430 and 0.367.

ACKNOWLEDGEMENT

This research was held on cooperation between the Muhammadiyah University of Surakarta and Sebelas Maret University. Acknowledgments to Sebelas Maret University, especially the Informatics department that has collaborated on this research with the Muhammadiyah University of Surakarta.

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