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Spent wash Decolorization using Granular and Powdered Activated Carbon: Taguchi's Orthogonal Array design and ANN approach

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Abstract: *The intense colour of the spent wash effluent results in the crucial ecological issue when released untreated into the natural water resources. The decolorization of distillery spent wash effluent is a very difficult task. In this study, the performance efficiencies of powdered activated carbon (PAC) and granular activated carbon (GAC) were compared towards the degradation of organic pollutants in terms of colour. The process parameters such as adsorbent dosage, pH, the initial concentration of the effluent and the operating temperature were optimized to attain the maximum decolorization efficiency. Optimization of the process parameters using Taguchi Orthogonal Array (OA) experimental design resulted in a maximum of 95% spent wash decolorization using PAC and 44% using GAC respectively. Using Artificial Neural Network (ANN), a two layered feedforward backpropagation model resulted as the best performance and predictive model for spent wash decolorization. The experimental data was found to be in excellent agreement with the predicted results from the ANN model.*

Keywords *- Adsorption, Color removal, Optimization, Validation, Wastewater treatment*

1. INTRODUCTION

The Distillery spent wash contains high levels of recalcitrant pollutants in the form of color and organic compounds. The dark color is due to the presence of dark brown acidic nitrogenous polymer – melanoidin [1], which is generated due to the non-enzymatic reaction between an amino acid and carbohydrate called Maillard reaction [2]. The major problem involved in treating spent wash is the color [3]. Moreover, the spent wash contains high levels of chemical oxygen demand (COD), biological oxygen demand (BOD), high total dissolved solids (TSS), low pH and intensely obnoxious odor. Even though the spent wash does not contain any toxic heavy metal pollutants, untreated disposal of the effluent into water streams may create potential problems such as turning the color of the receiving stream and the dissolved oxygen content gets depleted creating a potential threat to the aquatic organisms and microbiome. Conventional biological treatments involving anaerobic digestion (bio-methanation) are not much effective towards treating these highly recalcitrant pollutants [4]. Due to the antioxidant property of melanoidin, the microbial growth gets inhibited, which hinders degradation of color and toxicity [5]. Therefore, other potential alternatives have been tried for degrading the colour of the spent wash.

Physicochemical treatment methods involve adsorption, coagulation and flocculation, electro-coagulation, advanced oxidation, ozonation, membrane filtration and evaporation. Adsorption is one of the major physicochemical treatment methods employed for removing pollutants and color from the spent wash. In spite of the adsorbents employed, the decolorization process will be majorly influenced by the factors such as initial concentration of the effluent, molecular size, charge density and concentration of the other pollutant species that are present in the effluent.

Artificial Neural Networks (ANN) have been a great interest over the last decade and are being successfully applied across a range of problem-solving domains such as finance, medicine, engineering, geology, and physics. This computational tool can be employed where there are problems of prediction, classification, modeling, simulation or control [6, 7]. It is considered as a black-box device that receives input, transmits and produces output [8, 9]. Basically, for a network to work, a set of input and output data are necessary. Compared to the design of experiments, ANN requires a number of training data which is a major drawback and it can be rectified using the statistical experimental design, data comprising the minimum number of experiments as an input data for the ANN model. The network is trained with a set of experiments, consisting values of both input and output variables. Further, the network is tested with a series of experimental inputs to check whether the trained network is able to reproduce original experimental outputs [10, 11].

The neural network is a highly adaptable system with the capacity to learn relationships through repeated feeding of data and is capable of predicting new data. The principle of ANN is to obtain a number of outputs (response variables) from a number of inputs (measured data), using a series of layers of artificial neurons which are interconnected between them [12]. The input and output data correspond to the sensory nerves and the motor nerves of a typical biological system. There are hidden neurons, which play an internal role in the network. The input, the hidden and the output neurons must be connected together. The algorithm progresses iteratively through a number of epochs. On each epoch, the training cases are submitted to the network. The target and the actual outputs are compared to calculate the error. This error, together with the error surface gradient, is used to adjust the weights, and then the process repeats. The initial network configuration is random and the training stops when a given number of epochs elapse or when the error reaches an acceptable level or when the error stops improving [6].

The objective of this study is to investigate the efficiencies of PAC and GAC for adsorptive degradation of distillery spent wash in terms of colour removal. The major process parameters were optimized using Taguchi Orthogonal Array (OA) design of the experiment. The effect of the key operating parameters, viz. adsorbent dosage, pH, initial concentration and temperature for the decolourization of real spent wash was studied. The experimental data were also predicted and validated using ANN tool to check the performance of the experiment.

2. MATERIALS AND METHODS

2.1. Collection of distillery spent wash effluent

The spent wash effluent sample was collected from Trichy Distilleries and Chemicals Limited (TDCL), located near the city of Tiruchirappalli, Tamil Nadu, India. The collected effluent was immediately brought to the laboratory and stored in the refrigerator at 4 °C [13] until further use in order to avoid any deterioration in the physicochemical property of the spent wash. The major physicochemical characteristics of the raw spent wash at the time of collection were: pH: 4.3, COD: 98,400 mg/L, BOD: 7500 mg/L, TDS: 84,800 mg/L, TSS: 3500 mg/L, conductivity: 27 µS/cm, color: dark brown, odor: burnt sugar, absorbance at 475 nm: 2.01 (10% diluted).

2.2. Preparation of adsorbents

Charcoal Activated, extra pure (granular about 1.5 mm) was procured from Loba Chemie. Charcoal Activated (powdered) was procured from Merck. Both the adsorbents were regenerated at 200 °C for 2 h to remove moisture content and stored in a desiccator until usage.

2.3. Experimental design for process optimization

A five-level-four-factor Taguchi Orthogonal Array (OA) design was employed using Minitab statistical software to generate experimental runs by considering four independent input variables viz. Adsorbent dosage (A), pH (B), concentration (C) and temperature (D). The percentage colour removed was the output response. Based on literature and preliminary experiments, the minimum and the maximum range levels of the process parameters were fixed. The range of controlling factors was in the range of 0.5–2.5 g/L for adsorbent dosage, pH 3–11, initial concentration 10–50% and temperature 25–45 °C.

2.4. Experimental

The batch adsorption process was conducted for 3 h in a thermostatic shaker. Aliquots of samples were withdrawn and centrifuged at 10,000 rpm to remove the particles. The decolorized supernatant was measured at a characteristic wavelength of 475 nm using UV-Visible spectrophotometer (Spectroquant, Pharo 300, Merck).

The percentage colour removal was calculated by:

$$\%\ \text{colour removed} = (C_0 - C_t)/\ C_0 \times 100 \qquad (1)$$

where, C_0 and C_t are the initial absorbance and absorbance at time t for the spent wash at a corresponding wavelength of 475 nm [14, 15].

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2.5. Validation using artificial neural network (ANN)

The experimental data on the photocatalytic degradation was trained and validated using the ANN tool. The four process parameters such as adsorbent dosage, pH, concentration and temperature were the input variables and the output response was the percentage colour removal. The experimental data was fed into the ANN tool by considering feedforward backpropagation, mean square error (MSE) method. The performance of the neural network depends on the number of hidden layers and the number of neurons per hidden layer. A few number of trials were made to select the best architecture for the neural network. For the iteration to be complete and to obtain the converged result, every time the loop had to be altered such that, the regression and the epoch reached the minimum value until its validation gets stopped. The initial time was set as normal so that the method took its own time to attain the minimum value. In this study, a neural network architecture was 4–10–10–1 (4 neurons in the input layer, 10 neurons in the first hidden layer, 10 neurons in the second hidden layer and 1 neuron in the output layer) was considered and shown in Fig. 1.

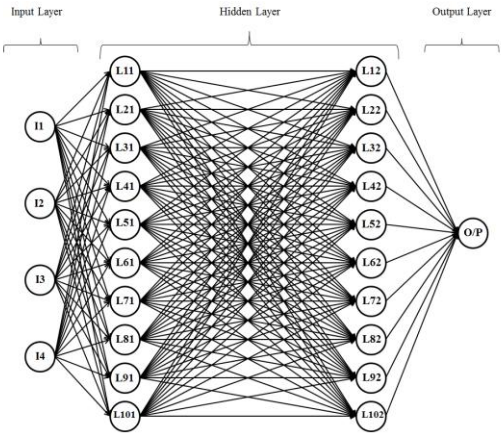

Fig. 1 ANN Architecture

The neurons are based on the purelin transfer function. The network was trained using the trainlm (Levenberg–Marquardt) algorithm and Mean Squared Error (MSE) was used as the performance function which was to be minimized to $1 \times 10-10$. All other ANN parameters were set as default values as in the ANN tool. The network was trained for 1000 iterations. When the iteration reached the minimal gradient value, the performance validation and the regression were tested.

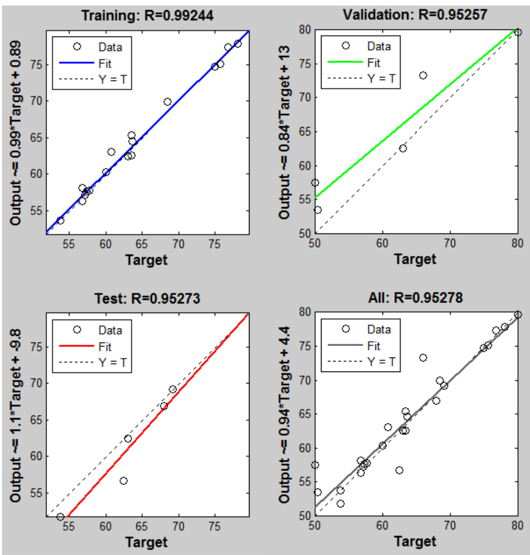

Fig. 2 ANN parity plots

From the regression analysis, the validation, the trial was dealt. From Fig. 2, it was noted that the fit data line and the cluster line almost coincided with each other with an R value of 0.99244, 0.95257 and 0.95273 for training, validation and test respectively. Since the R value approached almost 99%, it could be interpreted that the experimental error was minimized and the experimental data and model predictions were in excellent agreement.

3. RESULTS AND DISCUSSION

3.1. Colour removal efficiencies of GAC and PAC

The experimental results in terms of percentage colour removal were evaluated and tabulated in Table 1. From the results obtained it was very evident that the spent wash concentration played a very major role in the adsorption efficiencies of the adsorbent materials. As a well-known fact, that the surface area of the PAC is very higher in comparison with that of the granular carbon, a maximum of 94% colour removal was obtained using PAC whereas only 44% was obtained using GAC.

Table 1. Adsorption efficiencies of GAC and PAC

Spent wash conc. (% dilution)	% colour removed	
	GAC	PAC
10%	43.9	94.7
20%	33.5	88.9
30%	14.7	73.4
40%	4.5	57.1
50%	0.5	40.9

3.2. Taguchi design and optimization of process parameters

The key process parameters determining the spent wash decolorization were optimized by Taguchi orthogonal array design. The results of the ANOVA test showed that the p-values of the model terms were significant, i.e. $p<0.05$, for dosage which is the factor with major influence on decolorization. The p-value indicates the significance of a particular term in a model. The F-values (Fisher test) showed the level of significance for the terms in the model, without defining the positive or a negative effect of that particular term.

3.3. Effect of process parameters on decolorization efficiency

The mutual interaction between the key process parameters in terms of spent wash decolorization was plotted. Surface plots showing interactions between adsorbent dosage, concentration vs. % colour removed, adsorbent dosage, pH vs. % colour removed and concentration, pH vs. % colour removed are shown in Fig.3.

From the analysis of variance of the experimental data, the initial concentration of the spent wash had much impact on the decolorization efficiency followed by adsorbent dosage, pH and temperature. At increased spent wash concentration (50% dilution), the process resulted in minimal efficiency. Using PAC, a maximum efficiency of 94.7% colour removal was obtained at a minimal concentration of 10% dilution. As the initial concentration increased, the adsorption capacity of the adsorbents decreased. For all the interactions involving adsorbent dosage, % decolorization increased as the dosage concentration increased. The decolorization efficiency was found to be the highest at acidic conditions as shown in Fig. 3(b).

There was not much of an influence of temperature on the decolorization efficiency. As a result of this present study, optimization of the key process parameters viz. adsorbent dosage, pH, concentration and temperature by Taguchi method resulted 95% decolorization of the distillery spent wash effluent using PAC. In comparison with the GAC used, there was only 38% colour removal efficiency exhibited by it.

3.4. ANN analysis

A feedforward backpropagation model was developed with inputs of dimensionless variables such as adsorbent dosage, pH, concentration and temperature. The output was in terms of decolorization efficiency. The input data was trained, validated and tested for best fit using the ANN tool. A two-layered network was used. The performance of the degradation process was evaluated using the neural network and validated graphically.

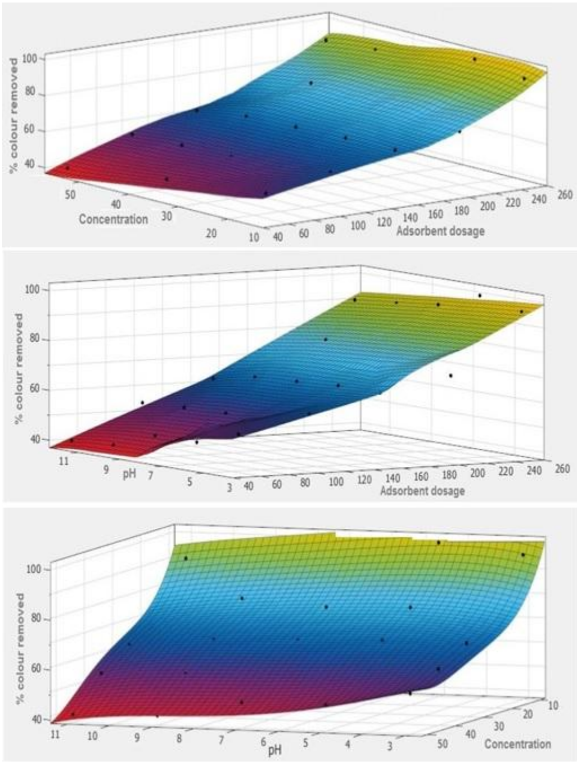

Fig.3 Surface plots showing mutual interactions

4. CONCLUSION

GAC and PAC were tested for their ability to adsorb organic pollutants in terms of colour from distillery spent wash effluent.

The major process parameters were optimized using Taguchi OA design.

Experiments were conducted following the Taguchi design.

The experimental data and the results were validated using ANN method. The experimental data was found to be in excellent agreement with the predicted results from the ANN model.

The mutual interaction between the key operating parameters was analysed from the surfaced plots plotted.

At optimized conditions of 2.5 g/L adsorbent dosage, pH 3 and 10% diluted spent wash concentration and at room temperature, 95% and 44% colour removal was exhibited by PAC and GAC respectively.

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