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Investigation of potato peel-based bio-sorbent efficiency in reactive dye removal: Artificial neural network modeling and genetic algorithms optimization

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Original Article

Abstract

Over the last few years, a number of investigations have been conducted to explore the low cost sorbents for the decontamination of toxic materials. Undoubtedly, agricultural waste mass is presently one of the most challenging topics, which has been gaining attention during the past several decades. Wastes are very cheap and easily available material in production of sorbent. Therefore, the Reactive Red 198 (RR198) removal efficiency from aqueous solutions by potato peel powder based sorbent (PP) was examined in this study. The Taguchi method was used in combination with full factorial methods to design the experiments. Based on the design of experiment outputs, 18 experimental sets were designed and the experiments were done in accordance with the experimental design. The sorption handmade batch reactor consists of a 200 ml beaker, 100 RPM magnetic stirrer, and a sampling port. Then, the experimental data were collected under desired conditions. In each sample sorbent was separated using a centrifuge (3000 rpm and 5 minutes). Then, dye concentrations were determined based on Beer's law and calibration plots using a UV-visible spectrophotometer. The wavelength resolution and the bandwidth were, respectively, 1 and 0.4 nm. The length of the optical path in glass cell was 1 cm. The maximum absorption wavelength was determined in each run to compensate the matrix effects. The results revealed that PP is effective for the sorption of RR198 from aqueous solutions. The maximum sorption of PP from RR198 solution was determined as 93 mg/g. Artificial neural network (ANN) model of dye removal efficiency (DR%) was developed based on the experimental data sets. The ANN model was strongly validated using statistical tests. The R^2 and RMSE of the test set were 0.98 and 4.3, respectively. The results demonstrate that PP can be successfully used as sorbent for RR198 removal from aqueous solutions. The results revealed that experimental parameters strongly influence the DR% and different experimental conditions cause different DR% (from 0 to 93).

KEYWORDS: Sorption, Potato Waste Powder, Design of Experiment, Artificial Neural Network, Genetic Algorithm, Dye Removal

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Introduction

Water and wastewater pollutions like toxic or colorful organic materials (such as dyes, pesticides, organic solvents, and etcetera), and toxic inorganic materials (especially heavy metals) have been a great concern in recent years. Conventional treatment techniques of water and wastewater include sorption, filtration, precipitation, flocculation, membrane technology, and advanced oxidation process. In

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developing countries, like Iran, industries cannot afford to use conventional wastewater treatment, because they are not cost-effective. Therefore, the search for new efficient technologies capable of treating contaminated waste water in a cost effective manner is underway.[1]

Among the numerous treatment technologies developed for the removal of pollutions from water, sorption is receiving increasing attention. The sorbent used in the sorption processes are various materials, including activated carbon, obtained from agricultural byproducts and commercial activated carbons. However, the high cost of the activation process limits its use in wastewater treatment, particularly for developing countries.[2]

Over the last few years, a number of investigations have been conducted to explore low cost sorbents for the decontamination of toxic materials. Undoubtedly, agricultural waste mass is presently one of the most challenging topics, which has been gaining attention during the past several decades. Wastes are very cheap and easily available materials in the production of sorbent. Production of sorbent from different wastes not only achieves the removal of pollutants in harmless forms but also has environmental effects of reducing waste. In previous studies, different substances such as fruit and vegetable wastes, cone, leaves, and etcetera have been employed.[3]

Potato waste is disposed as a zero value waste in many countries as part of the production of French fries, crisps, puree, instant potatoes, and similar products. The management problem of potato wastes causes considerable concern for potato industries; thus, implying the need to identify an integrated and environmentally-friendly solution. However, potato waste can be used as sorbent to remove most pollutants in aqueous solutions. The produced waste ratio and composition depends on the procedure applied; steam, abrasion, or lye peeling. Potato waste is almost 15% to 40% of influent potatoes and it consists of 55% potato skins, 33% starch, and 12% inert material.[4]

A high quality model can be applied to the process control. Applications of artificial neural network (ANN) and genetic algorithm have been successfully employed in environmental engineering for process modeling and optimization because of reliable, robust, and salient characteristics in capturing the non-linear relationships of variables in complex systems.[5]

ANN are parallel computational procedures consisting of highly interconnected processing element groups named neurons.[6] Owing to their inherent nature to model and learn complexities, ANNs have found wide applications in various areas of wastewater treatment.[7-10] ANN has been recently used for CR% (color removal) and energy consumption (EnC) modeling in electrocoagulation (EC).[11, 12]

Genetic algorithms (GA) are adaptive heuristic search algorithms based on the evolutionary ideas of natural selection and genetic. They belong to the larger class of evolutionary algorithms, which generate solutions to optimize problems by carrying out stochastic transformations inspired by natural evolution, such as inheritance, mutation, selection, and crossover.[13-16]

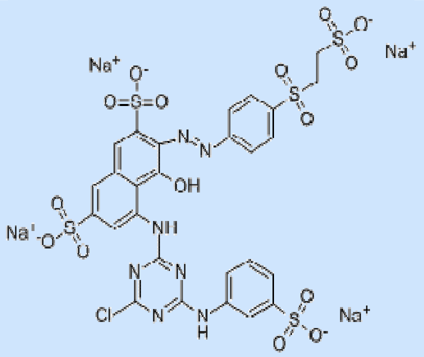
The present study had three objectives. First, to investigate the potential of PP as a sorbent in the removal of the model dye, Reactive Red 198 (RR198), from aqueous solutions. Second, DOE conducted studies to investigate the effects of five operational parameters: sorbent particle average size (S_z), initial pH (pH_0), dose of sorbent (D_s), initial dye concentration (C_0), and contact time (t_c) on sorption efficiency. The third objective was the application and assessment of ANN and GA for sorption system modeling and optimization.

Materials and Methods

RR198 was prepared from Alvan Sabet Co. (Iran). The chemical structure and some characteristics of this dye are shown in table 1. H_2SO_4 and NaOH were obtained from Merck. Distilled water was produced by a TKA Smart2Pure ultrapure water production system (Thermo Electron LED GmbH, Germany) for the preparation of dye solutions. UV-Vis spectrometer (T90$^+$ PG Instrument Ltd.)

was used for calibration curve and experimental measurements.

Table 1. The chemical properties of RR198

Molecular formula	C27H18ClN7Na4O15S5
Molecular structure	
Molecular weight	968.21
CAS number	145017-98-7

Potato wastes were collected from the garbage of local restaurants of Kurdistan University of Medical Sciences. Raw potato peal was washed with hot water to remove adhering dirt. Then, degradation was done by washing thoroughly with 1 M HCl solution and rinsing with distilled water several times. The degradation product was dried at 70ºC for 48 hours in order to gradually reduce the water content in the oven, crushed by a commercial mill, and sieved through two different sieve sizes with average size of 225 and 575 µm. The final product was stored in a sealed bottle for future application as sorbent.

The selected variables were S_z at two levels (225 and 575 µm), pH_0 at three levels (3, 7, and 11), C_0 at three levels (10, 50, and 100 mgl^{-1}), D_s at three levels (1, 5, and 10 gl^{-1}), and t_S at five levels (10, 30, 60, 90, and 150 min). A combined design of Taguchi for S_z, pH_0, C_0, D_s, and full factorial methods for t_c was used to design the experiments (DOE) using Minitab 14 (Minitab Inc., State College, PA, USA). The Taguchi was designed based on L_{18} orthogonal array with 4 factors. The order of experiments was made random in order to avoid noise sources, which could take place during an experiment. In these 18 experiments, the five levels of t_S were determined. All the selected experimental conditions can be seen in figure 1.[17]

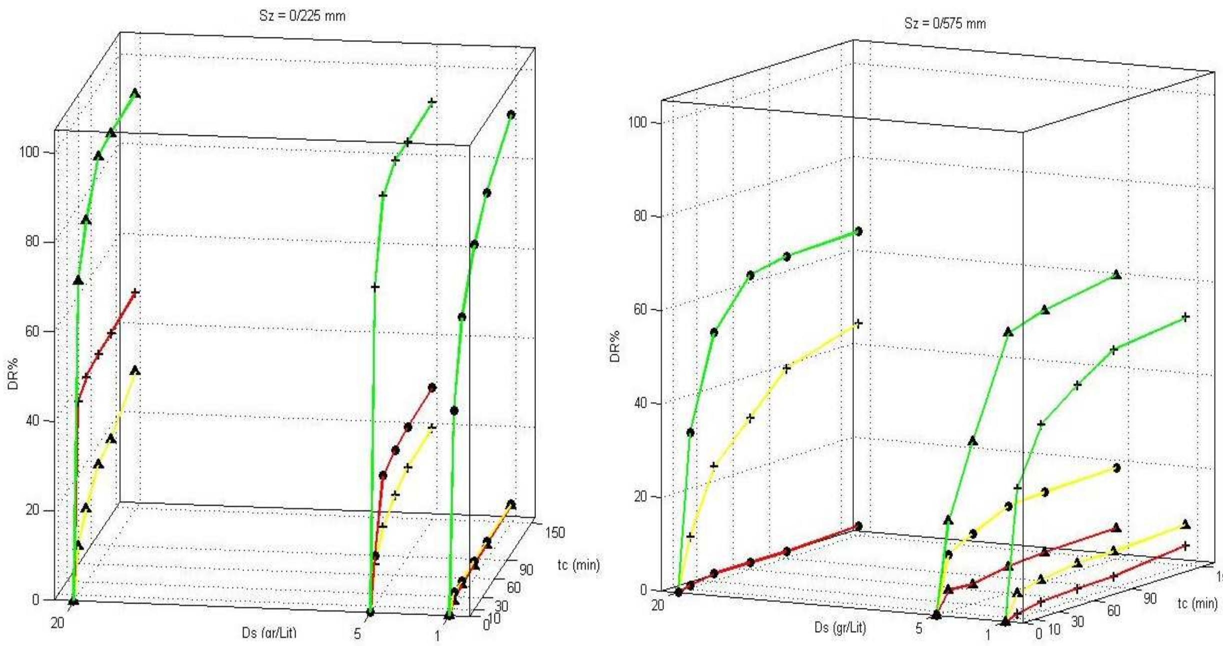


Figure 1. Ninety dye removal (DR%) of all 18 runs, green line (pH0 = 3), yellow line (pH0 = 7), red line (pH0 = 11), circle marker (C0 = 10 mg/l), plus sign marker (C0 = 50 mg/l), triangle marker (C0 = 100 mg/l)

To investigate the performance of synthetic wastewater containing DB41 using potato waste based sorbent in dye removal, DOE conducted sorption studies were carried out in batch experiments in a 200 ml beaker containing 100 ml DB41 and in room temperature as a function of S_z at two levels (225 and 575 µm), pH_0 at three levels (3, 7, and 11), C_0 at three levels (10, 50, and 100 mgl^{-1}), D_s at three levels (1, 5, and 10 gl^{-1}), and t_s at five levels (10, 30, 60, 90, and 150 min).

In each sample sorbent was separated using a centrifuge (3000 rpm and 5 minutes). Then, dye concentrations were determined based on Beer's law and calibration plots using a UV-visible spectrophotometer. The wavelength resolution and the bandwidth were, respectively, 1 and 0.4 nm. The length of the optical path in glass cell was 1 cm. The maximum absorption wavelength was determined in each run to compensate the matrix effects. The calibration plot was constructed in the range of each run concentration. The calibration plots usually provided a determination coefficient close to 99.9%. These data were used to calculate the sorption capacity of the sorbent. In most cases, an accurate dilution was necessary to obtain a measurable absorption. The percentage of dye uptake by the sorbent was computed using the equation:

$$\% \text{ Sorption} = (C_t - C_e)/C_t \times 100$$

where C_0 and C_t were the initial centrifuged sample and final concentration of RR198 in the solution, respectively.

The 90 data of dye removal (DR%) together with corresponding experimental conditions were used as a data set. The five operational parameters were considered as inputs of models whilst the DR% was considered as dependent variable. Data set was randomly divided into three parts; 60% as a training set, 20% as a validation set, and 20% as testing set. The ANN models were constructed based on the same datasets for both DR%. The multiple linear regression (MLR) and ANN models were constructed as two most popular linear and nonlinear models for comparison. For the ANN model, back propagation algorithm was used in this study, as it is very fast and can be employed quite easily. The number of hidden layers and nodes was determined via a trial and error procedure. GA toolbox in MATLAB (version 7) was used for generating the optimal solution for DR%. MATLAB functions using ANN model as the input were written for creating a fitness function for the optimization problem. The DR% component to be maximized was negated in the vector valued fitness function, since GA minimizes all the objectives. Experimental ranges were placed as bounds on the five inputs.[18-21]

Results and Discussion

Sorption process

Figure 2 shows all obtained data in 18 experiments. The effect of each operational parameter was illustrated in these figures, indicating that different levels of experimental parameters result in different DR%. In addition, it was found that PP is propitious for RR198 removal as sorbent.

SMLR models

The MLR models were developed for DR%. The model and related statistical characteristics are given in table 2.

Based on unbiased standardized coefficients presented in table 2, among linear parameters, D_s and t_S have a positive effect, but C_0, S_z, and pH_0 have negative effects on DR%. The most important parameters were pH_0 and D_s. Table 2 indicates that the MLR model does not have good predictability for DR% due to the complex mechanism of sorption process. It demonstrates new interest in using a more powerful modeling approach especially ANN model.

ANN modelling and GA Optimization

The ANN model was constructed for DR%. One hidden layer with 7 neurons was applied in the model. The tansig transfer function was selected for input and hidden layer, and purelin for output.[18,19] Once the networks were trained by experimental train data set, the weights and bias

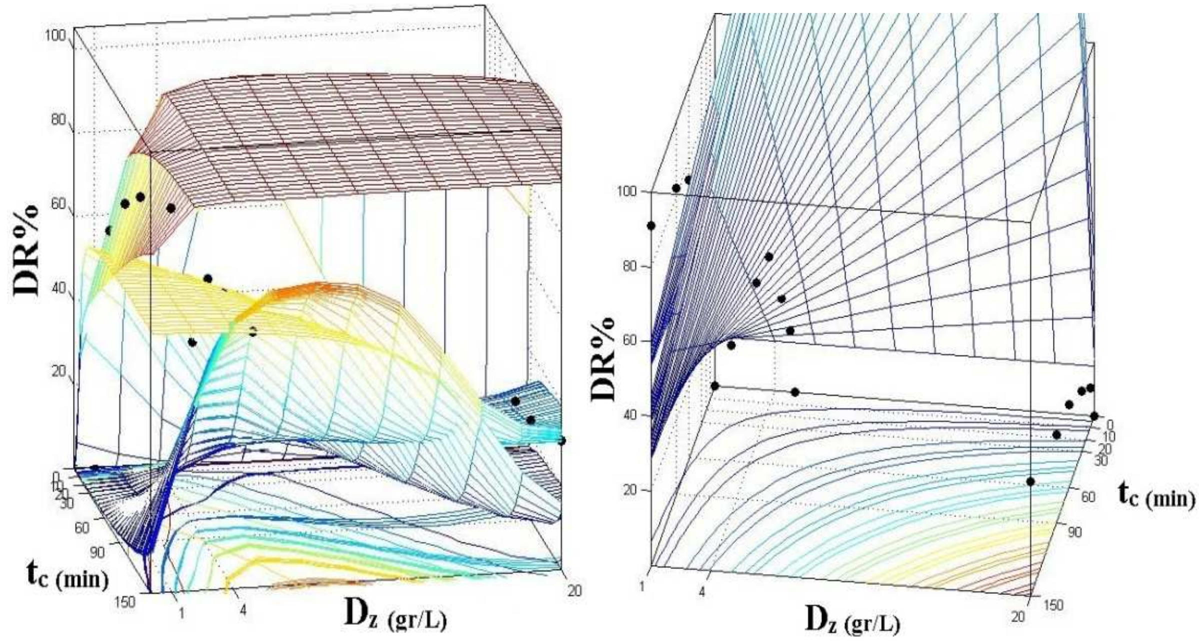


Figure 2. Sample plot of the artificial neural networks (ANN) (left) and multiple linear regression (MLR) (right) predicted values of dye removal (DR%) (colored surface) versus experimental data of DR% (black dot), for three sample conditions

Table 2. The MLR model and related statistical characteristics

	Coeff.	St. Coeff.	P
Constant	63.14	9.91	< 0.01
S_z	-0.04	0.01	0.02
pH_0	-5.52	0.76	< 0.01
C_0	-0.03	0.07	0.64
Ds	0.74	0.31	0.02
t_S	0.20	0.05	< 0.01
Data set	Train (64 data)		Test (44 data)
R^2	0.57		0.57
RMSE	19.3		18.6

Coeff.: Coefficients; St. Coeff.: Standardized coefficients; Sz: Sorbent particle average size; pH_0: Initial pH; Ds: Dose of sorbent; C_0: Initial dye concentration; RMSE: Root mean square error

of each neuron and layer were saved in the ANN model. Then, they were used to estimate the test set. The (5:7:1) ANN were trained using 64 data of the train set for DR% models by the back propagation algorithm. The parameters of ANN models are presented in table 3.

Moreover, figure 2 shows the samples of ANN and MLR model predictions and their corresponding experimental results together.

Figure 2 and comparison of table 2 with table 4 clearly show that ANN model outperformed the MLR model. Therefore, ANN model was used as optimization function in GA. GA optimization

Table 3. Network weights and biases of the artificial neural networks (ANN) model

Neuron	Input layer to hidden layer weights						Hidden layer to output layer weights	
	S_z	pH_0	C_0	D_s	t_S	Bias	Output	Bias
n_1	-1.445	1.395	1.167	-1.162	0.456	2.486	1.07	
n_2	-2.281	4.191	1.730	-0.976	-0.554	1.163	-0.84	
n_3	2.176	-0.673	1.859	-0.006	0.233	-0.323	1.84	
n_4	-1.644	-1.707	2.882	3.060	-2.604	-2.863	-0.5	1.07
n_5	0.684	1.240	2.427	-1.179	0.435	1.162	-1	
n_6	-3.698	-0.161	0.391	2.762	0.375	-1.135	2.23	
n_7	-0.124	-0.236	0.278	0.133	-9.259	-9.778	-4.77	

Sz: Sorbent particle average size; pH_0: Initial pH; Ds: Dose of sorbent; C_0: Initial dye concentration; n: neuron or processing elements

Table 4. Statistical characteristics of artificial neural networks (ANN) model of DR%

Data set	Train	Validation	Test
R^2	1	0.98	0.98
RMSE	0.8	3.8	4.3

RMSE: Root mean square error

process results in an optimal solution set with a set of decision variables. The input decision variables of the optimal solution (93.3% for DR%) were 396, 3.1, 66, 19.7, and 126 for S_z, pH_0, C_0, D_s, and t_S, respectively. Interpretation of table 2 and optimum values was used to interpret and to discriminate effect of each parameter.

Effect of experimental parameters

The influence of S_z on DR% was investigated at two levels of 225 and 575 µm. The results show that S_z clearly influences the DR% (Figure 1). Furthermore, as presented in optimal values of empirical parameters, the optimum value of S_z is 396 µm. On the one hand, DR% further decreased S_z from 575 to 396 µg, because of a more effective adsorption area for small sorbents. On the other hand, a decreasing in size from 396 µg to 225 µg of S_z caused the decreasing of DR%. DR% might increase due to a more effective volume of absorption in small particles.

The pH_0 of a suspension is an important factor that can affect the DR% of RR198 by PP. Sorption efficiency increases, as the pH_0 of the solution is decreased from 11 to 3.1. As the pH_0 of the solution decrease from 11 to 3.1, RR198 will get a natural or positive charge that might be more favorable as sorbent for PP. PP contains functional groups such as hydroxyl and carboxyl. These functional groups have a variety of structurally related pH dependent properties of generating appropriate atmosphere (positively and/or negatively charged sites) for attracting the cationic, natural, and anionic species of RR198.

The sorption behavior of RR198 on PP was investigated in the range of C_0 (10-100 mg/l) at three levels. As presented in optimal values of empirical parameters, the optimum value of C_0 was 66 mg/l. This means that sorption of RR198 on PP increased with the increasing of C_0 until C_0 reached an optimal level (66 mg/l). Later, an increase in concentration decreased the percentage binding. These observations can be explained by the fact that in medium concentrations, the ratio of sorptive surface area to the molecules available is high, thus, there is a greater chance of dye removal. When dye concentrations are increased, binding sites become more quickly saturated as the amount of mass concentration remains constant. However, it is important to see that the corresponding P-value of C_0 presented in table 2 statistically reveals that C_0 is not effective on DR%.

The D_s influence was investigated in the range of 1-20 mg/l at three levels. As presented in optimal values of empirical parameters, the optimum value of D_s was 19.7 g/l. DR% increased with the increasing of mass dosage from 1 to 19.7 g (near the maximum investigated range of D_s). This is due to the fact that more sorbent has more capacity and binding sites for sorption.

The effect of t_c on RR198 sorption on PP was studied in 0-150 minutes at five levels. DR% increases with increasing t_c then becomes stable (Figure 1). The optimum value obtained by GA algorithm was 126 minutes that is in accordance with figure 1. This level off is due to the attainment of equilibrium between sorbate and sorbent at optimum t_c or saturation of binding sites at that time.

Conclusion

The potential of PP for the sorption of RR198 from aqueous solution was investigated in the present study. Taguchi was used in combination with full factorial methods for the design of experiments, 18 experimental sets were designed, and the experiments were done in accordance with the experimental design. The handmade batch sorption was constructed as sorption reactor. The experimental data were collected under desired conditions. The effects of five experimental parameters on dye sorption were studied. The results demonstrate that PP can successfully be used as sorbent for RR198 removal from aqueous solutions. The results revealed that experimental parameters strongly influence the DR% and different experimental conditions cause different DR% (from 0 to 93). The ANN modeling technique was successfully applied to model the process, and a reliable model was constructed and tested. The optimization of the process over the ANN model was done by GA algorithm. The obtained optimum values and experimental parameters effects were in accordance with previous studies and famous reported scientific theories.

Conflict of Interests

Authors have no conflict of interests.

Acknowledgements

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