Prediction of Adsorptive Capacity of Various Agricultural Wastes in the Removal of Heavy Metals, Dyes, and Antibiotic in Wastewater Using ANN

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***Abstract*—Artificial Neural Network model was proposed for the prediction of the adsorptive capacity of various agricultural wastes in the removal of heavy metals, dyes, and antibiotic in water. A total amount of 103 data sets were obtained from different literature and was split into training (70%), validation (15%) and testing (15%) data. After considering different architectures, an input layer that uses eight independent variables (molecular weight of the adsorbate, adsorbent, adsorbent pre-treatment preparation, average initial concentration of adsorbate in solution, mass of adsorbent, adsorbent dosage, pH, and temperature), one hidden layer with 18 neurons and one neuron in the output layer was found to give the best result. The overall mean square error was 3487, while the correlation coefficient for the test dataset is 0.91898.**

***Index Terms*—Adsorptive capacity, agricultural wastes, artificial neural network, correlation coefficient.**

I. Introduction

Aquatic pollution is now an increasing global problem due to continuous discharge of various chemicals (i.e. synthetic dyes, heavy metals and antibiotics) from different industries [1], [2]. Different technologies and techniques such as ion exchange, supercritical fluid extraction, adsorption, flocculation, coagulation, chemical precipitation, floatation, reverse osmosis, filtration, electrodialysis, precipitation, advanced oxidative process followed by membrane bioreactors, microbial system and electrochemical processes have been applied to remove these contaminants and improve the quality of water in accordance with local and national regulations. Among these technologies, adsorption is found to be the most effective method for wastewater treatment because of its efficiency, simplicity, and cost-effectiveness [3]. In search of highly effective, eco-friendly and cost-effective adsorbents, studies proved that over a wide variety of adsorbents such as zeolites, activated carbon, resins, silicate materials, among others, agricultural wastes are the most sustainable means of treating various contaminants in industrial effluents due to its abundance, cost, and minimal pre-treatment.

The most important parameter in assessing an adsorption process of a system is the adsorptive capacity of the adsorbent. This parameter is affected by different operating conditions such as pH, initial sorbate concentrations, adsorbent dosages, temperature, and other physical and chemical properties of the adsorbents like composition, pore size, surface area, and cationic exchange capacity. To determine the optimum operating conditions, experimentations must be conducted that would entail time and cost [3]. This concern is addressed with the application of several computer simulations like artificial neural networks (ANNs) to do prediction of properties [4]. ANN is a mathematical model based on biological nervous processing that utilizes interconnected nodes or neurons to model a response variable using several predictors through pattern recognition and information storage [3], [5]. ANN studies of this nature were used to predict properties of materials such as tensile strength and density [6] and bending strength and hardness [7]; in forecasting drying processes [8], energy systems [9], atmospheric systems [10], photovoltaic systems [11], and cooling systems [12]. Recently, different studies have shown that ANN could also be applied in adsorption systems to predict the process, kinetics, or removal efficiency [11]. Most ANN applications are being applied in the prediction of removal efficiency of dyes using nanoparticles or activated carbon. There have been very limited reports on applications of ANN in adsorption of pollutants using agricultural wastes [3].

The objective of this study is to produce a holistic model for predicting the adsorption capacity (mg/g) of a number of dyes, heavy metals, and antibiotics onto various types of agricultural wastes using ANN. Specifically, it aims to identify the most appropriate architecture, training algorithm, and transfer function for the dataset. The input parameters considered are the molecular weight of adsorbate, the agricultural waste used as adsorbent, the methods of pre-treating the adsorbent, the average initial concentration of the adsorbate in the solution (mg/L), mass of adsorbent used (g), adsorbent dosage (g/mL), pH of the solution, and the temperature (℃).

II. Methodology

A. Description of the Dataset

The data set is comprised of 103 observations obtained from various literature (Table I) that is described by 10 features, 3 of which are factor variables (adsorbent, adsorbate, and preparation) while the rest is either numerical variables or integers.

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TABLE I: THE ARRANGEMENT OF CHANNELS

Adsorbate	MW *a*	Agricultural Waste	Ref *b*
Chromium (III)	51.9961	Spent tea dust, Coffee dust, Orange peel, Rice husk, Yellow passion fruit shell	[13], [14]
Chromium (VI)	51.9961	Groundnut husk	[15]
Tetracycline	444.435	Macadamia nut shells, Rice husk ash, Wheat straws ash, Apricot Nut Shells	[16], [17]
Methylene blue	319.85	Passion fruit peel, Rice husk, Brazil nut shells, Coconut coir pith, Unwanted weed, Banana peel, Orange peel, Ashoka leaf	[18], [19], [20] [21], [22]
Indigo carmine	466.36	Brazil nut shells	[20]
Amido black 10B	616.487	Banana peel, Orange peel	[22]
Methyl orange	327.33	Banana peel, Orange peel	[22]
Methyl violet	393.95	Banana peel, Orange peel, Sunflower seed hull	[22]
Rhodamine B	479.02	Banana peel, Orange peel	[22]
Congo red	696.665	Banana peel, Orange peel, Ground nut shells, Eichornia	[22], [23]
Brilliant Green	482.6	Ashoka leaf	[23]
Malachite green	364.9	Ashoka leaf	[23]
Rhodamine B	479	Ashoka leaf, Cypress cone chips	[23], [24]
Methylene blue	373.91	Cypress cone chips, Coconut shell, Factory-rejected tea, Karanj fruit hulls	[24]
Copper (II)	63.546	Ponkan Mandarin peels, Bagasse fly ash, Barley straw ash, Sugarcane bagasse, Orange peel, *Citrus maxima* peel, Passion fruit shell, Potato peel	[25], [26], [27], [28]
Nickel (II)	58.6934	Ponkan Mandarin peels, Bagasse fly ash, Barley straw ash, Sugarcane bagasse, *Citrus maxima* peel, Passion fruit shell	[25], [26], [27], [28]
Lead (II)	207.2	Sugarcane bagasse, Yellow passion fruit shell, *Citrus maxima* peel, Orange peel, Passion fruit shell, Pine cone, Banana peel	[28], [29] [30], [31]
Chromium (VI)	52.9961	*Melaleuca diosmifolia*leaf	[32]
ranitidine hydrochloride	350.87	Mung bean husk	[33]
Reactive blue 19	626.54	Pomegranate peel	[34]
Norfloxacin	319.331	Albizia lebbeck seed pods	[35]
Ciprofloxacin	331.346	Albizia lebbeck seed pods, Date palm leaflets	[35], [36]
Amoxicillin	365.4	Guava Seed	[37]

[a]MW = Molecular weight of the adsorbate in g/mol
[b]Ref = References used

B. Artificial Neural Network

To establish the neural network model, several key parameters were determined such as hidden layer nodes, learning algorithm, and transfer functions. A backpropagation (BP) neural network was used which includes the input layer, the hidden layer, and the output layer. The input layer has eight neurons given that there are eight input variables (molecular weight of the adsorbate, adsorbent, adsorbent preparation, average initial concentration of adsorbate in solution, mass of adsorbent, adsorbent dosage, pH, and temperature) and the output layer has one output variable which is the monolayer adsorptive capacity. The implementation of the modeling is conducted using the neural network toolbox in Matlab R2018b.

To determine the best number of hidden layer nodes, the formula below was used:

$$l = \sqrt{n + m} + a \quad (1)$$

$$l = 2n + 1 \quad (2)$$

where is l the number of hidden layer nodes, n is the input neurons, m is the output neurons and a is the regulating constant. Thus, based on (2), the best number of hidden layer nodes for this model is 17.

However, due to the complexity of network mapping and the uncertainty of the training process, the equations above may not always be true. Hidden layer nodes can still be best determined by experiments but (1) and (2) can be used as a basis for the starting point and be changed incrementally. Having the right number of hidden layer nodes is important as it affects the performance of the neural network. If the hidden layer node is too less, then the convergence rate of the whole network may be reduced. Conversely, if the hidden layer node is too much, then the network topology will be complicated, and the error may not be optimal .

There were three learning algorithms used in this paper: trainlm (Levenberg-Marquardt), trainbfg (BFGS Quasi-Newton) and traingd (gradient decent).

The number of hidden layer nodes were set to 18 to 24 in increments of two and the three transfer functions; hyperbolic tangent-sigmoid, log-sigmoid, and the linear transfer function were also used to determine the optimum prediction model based on the training result.

70% of the input data was used for training, 15% was used for testing and the other 15% was used for validation. The performance of the algorithm was evaluated using mean-squared error (MSE) and the correlation coefficient (R). The combination with the least training error and the maximum correlation coefficient was selected as the optimal neural network model.

III. RESULTS AND DISCUSSION

The results of the training and modeling showed that the optimum results can be achieved when the trainlm algorithm is used with hyperbolic tangent-sigmoid transfer function in the hidden layer. It was found out that the most appropriate

number of hidden layer neurons is 18. Table II below summarizes the best result per training algorithm used in this work.

TABLE II: BEST RESULTS PER TRAINING ALGORITHM USED OF THE ANN MODELING

Training Algorithm Used	Transfer Function Used	Number of Hidden Layers	Testing R	MSE
Traingd	tansig	18	0.27586	588068
Trainbfg	linear	24	0.81081	4037
Trainlm	tansig	18	0.91898	3487

The results showed that the values of R for the training (0.93257) and validation (0.90978) datasets are relatively close and the correlation between the output and the target is very high. It can also be inferred from the result that the R value for the test dataset (0.91898) is very close to the values of the training and validation datasets. A neural network could be more effective if a large amount of data is used [16]. Furthermore, it was found out that the best MSE is 3487 at the sixth epoch.

The neural architecture of the optimum model was 8-18-1 (input-hidden-output). Each of the eight input neurons receive one input (Xi, i = 1 to 8) and projects the result to each of the neurons in the hidden which eventually computes using the tansig transfer function and sends its result (Yj, j = 1 to 18) to the neuron in the output layer which finally produces the response of the network (Predicted Adsorptive capacity, Z). The output signal of each hidden neuron (Yj) is calculated as:

$$Y_j = tansig\left(\sum_{i=1}^{8} w_{i,j}\, X_i + b_j\right) = \frac{2}{(1 + exp(-2(\sum_{i=1}^{8} w_{i,j}\, X_i + b_j)))} - 1 \quad (3)$$

while the output was computed as:

$$Z = tansig\left(\sum_{i=1}^{18} w_{1,j}\, Y_j + b_1\right) = \frac{2}{(1 + exp(-2(\sum_{i=1}^{18} w_{1,j}\, Y_j + b_1)))} - 1 \quad (4)$$

where Z is the predicted adsorptive capacity, Y_j are neurons of the hidden layer, X_i are the input variables, $w_{i,j}$ and $w_{1,j}$ are the weights of the connections between the input and hidden neurons and between the hidden and output neuron, respectively and b_j and b_1 are the biases on the hidden neurons and output neuron, respectively. The values for the weights and biases between the input and hidden layers are summarized in Table III while the weights and biases between the hidden and output layers are summarized in Table IV.

The comparison of the predicted data with that of the actual data is shown in Fig. 1 while the error for all data is shown in Fig. 2.

The results show good agreement between the actual and predicted adsorptive capacity. With the optimal model determined, it was also validated with different input variables as shown in Table V.

TABLE III: WEIGHTS AND BIASES BETWEEN THE INPUT LAYER AND HIDDEN LAYER

j	$w_{i,1}$	$w_{i,2}$	$w_{i,3}$	$w_{i,4}$	$w_{i,5}$	$w_{i,6}$	$w_{i,7}$	$w_{i,8}$	b_j
1	2.13	-1.38	1.64	1.25	-0.44	-1.85	-0.40	0.64	-0.09
2	0.40	0.32	-0.47	1.01	-0.44	1.26	0.08	-0.62	-1.34
3	-2.31	5.35	-0.39	-0.62	0.70	-0.05	0.60	-0.10	1.09
4	-0.89	1.82	-1.24	0.80	-2.92	1.17	2.32	-0.48	-1.34
5	0.08	0.01	0.01	1.29	-1.63	-0.53	-0.65	0.99	0.69
6	-0.71	-0.09	0.51	1.03	1.12	-0.30	-0.17	0.97	-2.04
7	0.05	2.78	0.14	-1.23	-0.40	-0.59	-2.69	-1.15	-1.48
8	0.68	1.40	0.71	0.65	-0.06	0.39	0.29	1.96	1.16
9	-0.93	1.40	0.44	-1.26	-0.63	0.19	-0.32	-0.93	-0.83
10	0.65	0.05	-0.82	-0.43	0.30	-2.00	-1.34	-2.71	-2.23
11	0.31	-1.41	-1.01	-0.40	-0.33	0.24	-2.85	-1.54	-0.57
12	-0.72	0.64	1.96	0.91	-0.77	-0.55	0.07	-0.31	-0.66
13	-0.03	0.87	0.17	0.64	0.53	0.09	-1.95	0.88	-1.03
14	-0.16	-2.47	0.22	1.27	-1.27	-0.16	-0.57	0.40	2.62
15	-1.08	-0.18	0.45	-0.29	0.57	-0.61	-0.80	0.98	0.06
16	1.45	0.77	-0.49	-0.29	0.02	0.05	0.51	-0.17	-0.64
17	1.90	-3.17	-0.46	-0.57	0.26	-0.77	-3.22	0.51	-1.55
18	-2.65	-0.79	-1.00	-0.71	-1.31	-0.70	-1.26	0.01	-0.36

TABLE IV: WEIGHTS AND BIASES BETWEEN THE HIDDEN LAYER AND OUTPUT LAYER

Weight Number	Values
$w_{1,1}$	1.10
$w_{1,2}$	0.53
$w_{1,3}$	1.30
$w_{1,4}$	0.89
$w_{1,5}$	-0.48
$w_{1,6}$	-1.45
$w_{1,7}$	-0.93
$w_{1,8}$	0.36
$w_{1,9}$	0.67
$w_{1,10}$	-0.28
$w_{1,11}$	0.50
$w_{1,12}$	-0.64
$w_{1,13}$	-0.40
$w_{1,14}$	0.60
$w_{1,15}$	0.22
$w_{1,16}$	0.67
$w_{1,17}$	-1.08
$w_{1,18}$	0.23
b_1	0.48

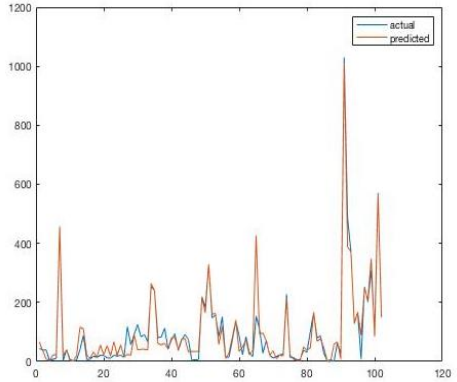

Fig. 1. Comparison of actual and predicted adsorptive capacity.

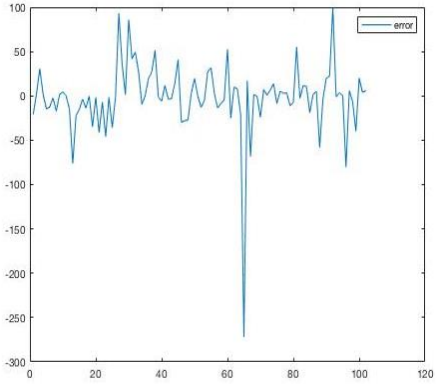

Fig. 2. Residual errors between the actual and predicted adsorptive capacity.

TABLE V: RESULTS OF THE TRAINING USING THE OPTIMAL MODEL WITH VARIATIONS IN INPUT PARAMETERS

Architecture Input	Test R	MSE
MW, AC, C_0, Dose, Temp	0.67369	14299
MW, AC, C_0, Mass, Dose, Temp	0.90711	2017
MW, AC, C_0, Mass, Dose, pH, Temp	0.9721	20307

It shows that the proposed ANN is sufficient and can be successfully used to predict the adsorption capacity of any contaminant adsorbed using any agricultural waste.

IV. CONCLUSION

The adsorptive capacity of any pollutant i.e. heavy metals, dyes, and antibiotics in water onto any agricultural waste can be investigated through ANNs without doing costly and time-consuming experimentations. The prediced values of the adsorptive capacity of the different agricultural wastes were affected by the following parameters: 1) the molecular weight of adsorbate, 2) the type of agricultural waste used as adsorbent, 3) the methods of pre-treating the adsorbents, 4) the average initial concentration of the adsorbate in the solution (mg/L), 5) mass of adsorbent used (g), 6) adsorbent dosage (g/mL), 7) pH of the solution and (8) the temperature (℃). It was found out the most appropriate neural architecture was 8-18-1. The correlation coefficient and mean squared error using the optimum ANN model for the testing dataset is 0.91898 and 3487, respectively. The results showed that the optimal model can be applied to predict the adsorptive capacity of various agricultural waste in adsorbing heavy metals, dyes, and antibiotics in response to any change in the input variables.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

AUTHOR CONTRIBUTIONS

A conducted the research and experiments, analyzed the data, and wrote the paper; *B*, C and *E* contributed to paper contents and edited and proofread the final paper, *D* facilitated and conceptualized the whole research process and write-ups construction; all authors approved the final revision of the paper.

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