REGRESSION ANALYSIS FOR THE ADSORPTION OF OIL USING HUMAN HAIR AS NATURAL ADSORBENT

Pavithira Sathinathan, Peck Loo Kiew*

Department of Chemical and Petroleum Engineering, Faculty of Engineering, UCSI University, 56000 Cheras, Kuala Lumpur, Malaysia.

Received: 22 December 2020, Accepted: 22 January 2021, Published: 30 April 2021, Publisher: UTP Press, Creative Commons: CC BY-NC-ND 4.0

ABSTRACT

The abundance of human hair waste (known as keratin biomass) in most parts of the world and its accumulation in waste streams are causing many environmental problems. Human hair's potential as a natural adsorbent to remove oil from wastewater was emphasised in this study. Mathematical and deep learning approaches were adopted to develop the regression models of oil adsorption using gents and ladies' hair wastes. The experimental results were obtained from literature review to perform regression analysis using Artificial Neural Network (ANN) in Matlab, Microsoft Excel, and Design Expert 6.0.6. The efficiency of these tools was compared in predicting the oil removal percentage within a specified range of adsorption parameters. This was done by comparing the R^2 value of the established adsorption models. In this study, the effect of different adsorption parameters, namely pH, contact time, and adsorbent dosage, on the oil removal percentage was included in the regression analysis. Subsequently, using the regression model with the highest R^2 value for both gents and ladies' hair adsorbent, a graphical user interface (GUI) was developed for the oil adsorption process to ease users in predicting the oil removal percentage within the specified range of adsorption parameters. The results showed that the adsorption model with the highest R^2 value of 0.9570 for gents hair and 0.9650 for ladies hair was developed using the ANN tool, implying its superiority in correlating the adsorption parameters to the oil removal percentage. However, in creating the GUI layout for the oil adsorption process, the regression model generated by the RSM regression tool (the second-highest R^2 at 0.8290 for gents hair and 0.6158 for ladies hair) was adopted to the limitations in retrieving the regression model from the ANN tool. The outcome of this study is expected to benefit users without prior knowledge of the oil adsorption process using human hair adsorbent to predict the removal percentage within a specified range of adsorption parameters, without the hassle of conducting experimental work.

Keywords: *Human hair waste, keratin biomass, regression, oil adsorption, graphical user interface*

INTRODUCTION

Human hair (50 – 100 mm in diameter) is a natural adsorbent consisting of the medulla, cortex, and dead cells made up of cuticle, water, lipids, and trace elements. It also contains 65–95% proteins, mainly polymers of amino acids such as keratin and cysteine [1]. Human hair is strongly hydrophobic because it is water repellent due to the cuticle content. Each cuticle cell is approximately 0.3 – 0.5 mm thick, and the visible length is about 5 – 10 mm [2]. Human hair has an abundance of peptide bonds, and CO- and NH-groups that form hydrogen bonds between neighboring molecules on the hair surface and have a highly porous cortex [3].

In most countries, human hair is considered waste material, and the accumulation of human hair in the waste stream causes environmental issues such as land pollution. Besides, it is an undesired material that is almost useless and can be abundantly found in the municipal waste stream of the cities and towns every place in the world. The population's density determines the amount of human hair waste accumulated in solid waste [4]. Human hair waste in the solid waste stream leads to choking of the drainage system and causes complicated problems [5]. Human hair waste may accumulate in the waste streams for an extended period, and it occupies huge space due to the slow degradation properties. This, in turn, promotes leachate formation [6] that causes the nitrogen level in the water to increase and leads to eutrophication. Eutrophication is a type of water pollution whereby water is rich with minerals and nutrients, which causes excessive algae growth. This is not desirable as it causes oxygen depletion in water bodies [7].

Human hair is also known as keratin biomass because approximately 80% of human hair is formed of keratin only [8]. Keratin is a structural protein that is highly stable and biopolymer that is biodegradable. Keratin protein is organised into filaments of hair cells, forming a polypeptide chain. The chain provides strength, flexibility, durability, and functionality to the hair in different conformations [9]. Generally, the natural keratin obtained from biomass such as human hair does not contain harmful chemicals and can be used directly as a natural adsorbent. Among all natural resources, keratinous materials can be used as biosorbent, either directly or after activation, due to their intricate networks characterised by high stability, insolubility in water, and the increased surface area containing many carboxyls, amido, and sulfur functional groups [10]. According to Zhang et al. [11],

*Corresponding author.
E-mail address : kiewpl@ucsiuniversity.edu.my

keratin wastes such as human hair from hairdressers amount globally to more than four million tons per year. In particular, the presence of carboxyl, amido, and disulfide groups in human hair suggests this waste product as suitable biosorbent of several chemicals, including oil and grease. However, the literature on this application is still limited up to now [11].

The presence of oil in water affects the environmental quality whereby humans' and animals' health is at risk, water sources are polluted, and various natural resources are destroyed. Certain components in oil and grease are poisonous to humans and other living organisms. It is worth noting that the wastewater stream that consists of oil and grease from different industries varies significantly in the pollution strength [12]. Oil and grease tend to accumulate at the inner surface of wastewater pipelines. Over time, these deposits become thicker as more grease and other solid materials build up. The pipelines' capacity then reduces, causing wastewater stream to overflow, unpleasant odour and unhealthy environment to humans and animals. The cleaning process of grease deposits on the wastewater pipeline is complicated, risky, and costly [13]. In this context, the adsorption process had been explored to remove or recover oil from oily wastewater and oil spill incidents. Anurangi [14] reported that it was possible to recover up to 70.5 % of oil from oily water, with 1.35 ml of oil being recovered using 1 g of hair. Also, a maximum adsorption capacity of 7470 mg/g for crude oil was proven possible in the work of Ukotije-Ikwut et al. [15] using African hair waste. The authors revealed that the oil experimental data were best fitted to the Freundlich isotherm model, demonstrated an excellent fit to the pseudo-second-order model, and thus governed by the chemisorption process [15].

Artificial Neural Network (ANN) is one of the non-traditional data-based tools for process modelling. ANN modelling has been successfully used for the adsorption process in the past decade [16]. According to Sutherland et al. [17], the ANN approach accurately simulates complex multi-layered adsorption processes of Cr(VI). Besides, ANN was also used in batch adsorption experiments to predict the amount of phenol being adsorbed onto activated carbon from aqueous solutions [18]. An empirical equation correlating the final phenol concentration with the weights of the network was developed. The authors compared the predicted results obtained from the optimised ANN model with the experimental data and other conventional equilibrium models. They reported that the accuracy of the expected results was as high as 99.98%. The developed ANN showed an accurate prediction of experimental data with mean square error, mean absolute error, root mean square error, and a correlation coefficient of 0.1540, 0.0565, 0.2378, and 0.9998, respectively [16].

In Response Surface Methodology (RSM), analysis of variance (ANOVA) has been used for graphical analyses of the data to define the interaction between the process variables and the responses to estimate the statistical parameters. The statistical significance can be checked by the F-test in the software. In regression analysis, the fitted polynomial model's accuracy is determined by the coefficient of correlation, R^2. On the other hand, the significant model terms are evaluated by the probability value (P-value) at a 95% confidence interval [18]. RSM is widely adopted for the regression modelling and optimisation of various processes such as adsorption and extraction. According to Saini et al. [19], RSM was used for optimising the adsorption conditions of cadmium ions by G-MCM-41. RSM analysed the interactions between the independent and response variables. The effects of different physiochemical parameters such as pH, heavy metal ion initial concentration, and contact time were examined using Box–Behnken design (BBD) in that study [20]. Besides, RSM was also adopted to identify the optimum conditions for the oil removal process using banana peels as biosorbent via the regression model developed [20]. Validation of the optimum conditions revealed that the developed model could predict the adsorption result successfully as the expected value from the model was 99.18%, and the obtained result was 98.86%.

Oil and grease removal from wastewater through an adsorption process using human hair as a natural adsorbent can be achieved in easy steps. However, the adsorption parameters affect the oil removal percentage significantly. To date, the existing studies are mainly focusing on how each adsorption parameter, namely the pH, contact time, and adsorbent dosage, affects the oil removal percentage individually, while there is limited study on how these adsorption parameters affect the oil removal percentage as a whole. A correlation can be established by performing regression analysis to predict the oil removal percentage by varying all or any of the adsorption parameters. Comparing the efficiency of different regression tools ranging from simple platforms such as Microsoft Excel to more comprehensive and sophisticated software such as Matlab and RSM in developing the regression model for the oil adsorption process is not yet reported in the literature. The paper aims to establish a correlation between different adsorption parameters, namely pH, contact time, and adsorbent dosage, on oil removal percentage using gents and ladies hair adsorbent. The regression analysis is performed using different regression tools. The developed model with the highest correlation coefficient is then adopted to design a graphical user interface (GUI) to ease users in predicting the oil removal percentage at different adsorption process parameters with a simple click.

METHODOLOGY

The overall research flow chart of this research is illustrated in Figure 1. The process methodologies involved were of three different stages. The first stage was data collection, in which the experimental results (oil adsorption percentages at different adsorption parameters) used in this study were obtained from the literature review. The regression model's development and comparison of varying regression tools' efficiency were performed in stage two. This final stage of this study was to develop a GUI for the oil adsorption process using human hair adsorbent (gents and ladies hair wastes).

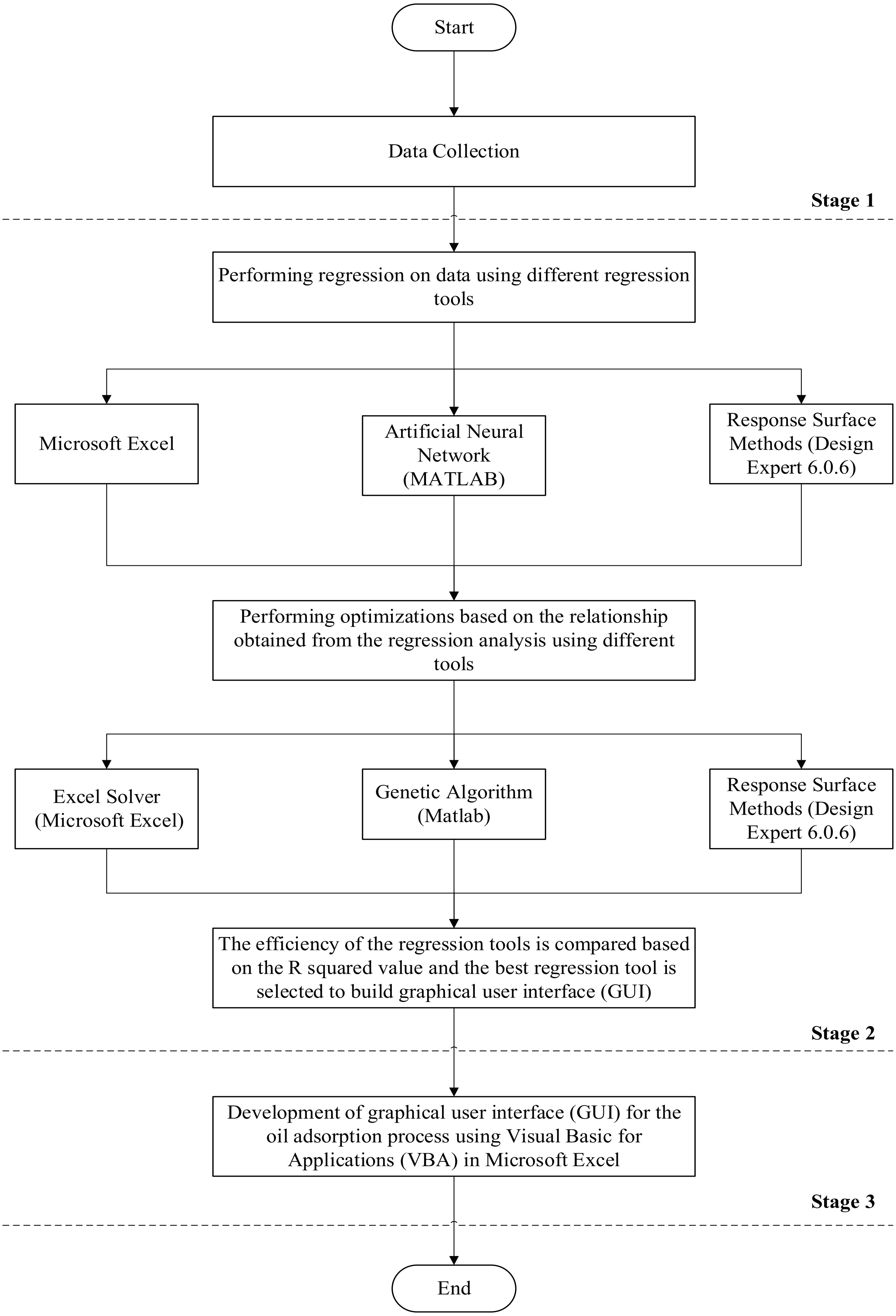


Figure 1 The process flow chart of this study

Data Collection

The experimental data were extracted from "Adsorption of Oil from Waste Water by Using Human Hair," reported by Nitin et al. [21]. This journal reported the oil removal percentage using gents and ladies' hair as natural adsorbents. Three parameters were investigated by the authors, which included pH, contact time, and adsorbent dosage. The results of the above-mentioned study were initially presented in graphical format. A web-based tool known as Web Plot Digitizer was used to extract numerical data from the graphs. Upon extracting numerical results from the graphical plots, the oil removal percentage using gents hair and ladies hair adsorbents at different adsorption parameters are tabulated in Table 1 and Table 2, respectively.

Development of Regression Model

In this section, three different regression tools were adopted to compare their respective efficiency in predicting the oil removal percentage (response variable) under the effects of varying adsorption parameters (manipulating variables). Based on the experimental results tabulated in Table 1 and Table 2, three adsorption parameters were designated as the manipulating variables with X1 was the pH, X2 was the contact time and X3 was the adsorbent dosage. The regression tools used were Microsoft Excel, Matlab, and Design Expert 6.0.6, respectively. For Microsoft Excel, the Data Analysis function was used to establish the regression correlation between the oil removal and the adsorption parameters. Subsequently, Excel Solver was adopted to optimise the objective function generated and predict the highest oil removal percentage within the specified range of adsorption parameters. As for Matlab, ANN was used to develop a regression model for the oil adsorption process using the built-in neural network toolbox and manual coding in M-file. Genetic Algorithm (GA) was then applied to identify the optimised oil removal percentage within the specified range of adsorption parameters with the developed regression model. The last regression tool investigated was Design Expert 6.0.6, whereby Box–Behnken design (BBD) approach in RSM was used to generate the oil adsorption models based on the experimental results. Regression analysis was also performed in RSM to retrieve the models' correlation coefficient (R^2) and their corresponding optimised adsorption conditions. All these regression tools' efficiency was compared based on the R^2 value

Table 1 Data Set for Gents Hair [21]

Input			Output
pH	Contact Time (min)	Adsorbent Dosage (mg)*	Oil Removal (%)**
1.0	30.0	500.0	77.4
3.0	30.0	500.0	70.2
5.0	30.0	500.0	59.1
7.0	30.0	500.0	51.2
1.0	9.9	500.0	52.6
1.0	19.8	500.0	61.7
1.0	29.9	500.0	78.5
1.0	39.7	500.0	67.1
1.0	49.8	500.0	75.5
1.0	59.8	500.0	91.8
1.0	69.9	500.0	94.2
1.0	80.0	500.0	85.8
1.0	90.0	500.0	80.7
1.0	99.7	500.0	74.9
1.0	110.1	500.0	72.8
1.0	119.8	500.0	71.0
1.0	70.0	99.1	59.4
1.0	70.0	248.2	71.2
1.0	70.0	498.2	94.8
1.0	70.0	752.6	65.3
1.0	70.0	1002.6	65.4
1.0	70.0	1248.2	59.7

* *With respect to 50 ml sample.*
** The percentage difference of oil concentration before and after adsorption with respect to initial oil concentration.

Table 2 Data Set for Ladies Hair [21]

Input			Output
pH	Contact Time (min)	Adsorbent Dosage (mg)*	Oil Removal (%)**
1.0	30.0	500.0	87.5
3.0	30.0	500.0	84.5
5.0	30.0	500.0	74.1
7.0	30.0	500.0	66.9
1.0	9.9	500.0	81.0
1.0	20.0	500.0	85.8
1.0	30.1	500.0	88.2
1.0	39.7	500.0	88.8
1.0	49.8	500.0	88.8
1.0	59.8	500.0	91.8
1.0	69.7	500.0	88.8
1.0	79.8	500.0	86.1
1.0	89.8	500.0	84.6
1.0	99.9	500.0	75.5
1.0	109.7	500.0	73.7
1.0	119.8	500.0	72.5
1.0	60.0	96.9	64.7
1.0	60.0	248.2	71.4
1.0	60.0	498.2	92.6
1.0	60.0	746.1	72.7
1.0	60.0	998.2	81.8
1.0	60.0	1248.2	83.2

* *With respect to 50 ml sample.*
** The percentage difference of oil concentration before and after adsorption with respect to initial oil concentration.

of all the developed regression equations. The regression model developed with the highest R^2 value was selected as the reference model used in GUI development to predict oil removal using human hair absorbent in the subsequent section.

Development of Graphical User Interface (GUI)

A GUI layout was developed using the Visual Basic for Applications window in Microsoft Excel.

RESULTS AND DISCUSSION

Development of Regression Model

In this section, regression analysis and optimisations were performed using three different regression tools: data analysis and Excel Solver in Microsoft Excel, ANN, and GA in Matlab and BBD approach in Design Expert 6.0.6. The results obtained from each of the tools were reported accordingly.

Microsoft Excel

Regression analysis was performed using data analysis, and optimisation was done using Excel Solver. Table 3 shows the regression analysis performed on gents hair, and it was found that quadratic correlation was the best fit to the experimental results. The correlation generated is presented in Equation 1, with the R^2 value of 0.6828. Similarly, for ladies' hair, Table 4 presents the regression analysis results, and Equation 2 shows the adsorption model predicted by Microsoft Excel. The R^2 value of Equation (2) was 0.6399.

Equation 1:

$$\text{Oil Removal (\%)} = 18.19 + 0.43X_1 - 0.48X_1^2 + 1.04X_2 - 0.01X_1^2 + 0.10X_3 - 7.84 \times 10^{-5} X_3^2$$

Equation 2:

$$\text{Oil Removal (\%)} = 56.52 - 0.30X_1 - 0.38X_1^2 + 0.25X_2 - 0.01X_2^2 + 0.08X_3 - 4.98 \times 10^{-5} X_3^2$$

With Equations 1 and 2 as the objective functions, optimisation was performed using Excel Solver to identify the highest oil percentage removal within the specified range of adsorption parameters. The results are tabulated in Table 5. Based on the optimised results, the highest oil removal percentage of 84.75% can be achieved using gents hair as adsorbent at the adsorption conditions of pH 1, contact time 72.54 minutes, and adsorbent dosage 608.97 mg. On the other hand, the highest oil removal percentage of 90.18% can be achieved using ladies' hair as adsorbent at the adsorption conditions of pH 1, contact time 44.68 minutes, and adsorbent dosage 759.57 mg.

Generally, in adopting Microsoft Excel as the regression tool, the value obtained for both regression models developed were approximately 0.6 for both gents and ladies hair adsorbents.

Table 3 Regression analysis on gents hair

SUMMARY OUTPUT								
Regression Statistics								
Multiple R	0.827886595							
R Square	0.685396214							
Adjusted R Square	0.559554699							
Standard Error	8.282974659							
Observations	22							
ANOVA								
	df	*SS*	*MS*	*F*	*Significance F*			
Regression	6	2242.031348	373.6719	5.446503	0.003612			
Residual	15	1029.115038	68.60767					
Total	21	3271.146386						
	Coefficients	*Standard Error*	*t Stat*	*P-value*	*Lower 95%*	*Upper 95%*	*Lower 95.0%*	*Upper 95.0%*
Intercept	18.18971722	15.65287102	1.162069	0.263364	-15.1736	51.55302	-15.1736	51.55302
pH	0.433894771	6.772497189	0.064067	0.949763	-14.0013	14.86913	-14.0013	14.86913
pH^2	-0.479726862	0.911483415	-0.52631	0.606365	-2.42251	1.463054	-2.42251	1.463054
Contact Time (min)	1.035039481	0.271301728	3.815086	0.00169	0.456774	1.613305	0.456774	1.613305
Contact $Time^2$ (min)	-0.007134292	0.002049223	-3.48146	0.003349	-0.0115	-0.00277	-0.0115	-0.00277
Adsorbent dose (mg)	0.095481211	0.030614974	3.118775	0.007042	0.030227	0.160735	0.030227	0.160735
Adsorbent $dose^2$(mg)	-7.8396E-05	2.13009E-05	-3.68041	0.002227	-0.00012	-3.3E-05	-0.00012	-3.3E-05

Table 4 Regression analysis on ladies hair

SUMMARY OUTPUT								
Regression Statistics								
Multiple R	0.799936121							
R Square	0.639897798							
Adjusted R Square	0.495856917							
Standard Error	5.867161999							
Observations	22							
ANOVA								
	df	*SS*	*MS*	*F*	*Significance F*			
Regression	6	917.5553	152.9259	4.442474	0.008882			
Residual	15	516.3538	34.42359					
Total	21	1433.909						
	Coefficients	*Standard Error*	*t Stat*	*P-value*	*Lower 95%*	*Upper 95%*	*Lower 95.0%*	*Upper 95.0%*
Intercept	56.52481257	10.9763	5.149716	0.000119	33.12939	79.92024	33.12939	79.92024
pH	-0.303961073	4.809158	-0.0632	0.950438	-10.5544	9.946517	-10.5544	9.946517
pH^2	-0.379855581	0.649137	-0.58517	0.567133	-1.76346	1.003747	-1.76346	1.003747
Contact Time (min)	0.250220617	0.194762	1.284748	0.218366	-0.16491	0.665347	-0.16491	0.665347
Contact $Time^2$ (min)	-0.002786995	0.001453	-1.91829	0.074313	-0.00588	0.00031	-0.00588	0.00031
Adsorbent dose (mg)	0.075725707	0.02153	3.517291	0.003111	0.029837	0.121615	0.029837	0.121615
Adsorbent $dose^2$ (mg)	-4.98308E-05	1.5E-05	-3.32469	0.004619	-8.2E-05	-1.8E-05	-8.2E-05	-1.8E-05

Table 5 Optimisations using Excel Solver

Parameters	Lower Limit	Upper Limit	Optimised Value for Gent Hair Model	Optimised Value for Ladies Hair Model
pH	1	7	1	1
Contact Time (min)	9.9	119.8	72.54	44.68
Adsorbent Dosage (mg)	96.9	1248.2	608.97	759.57
Predicted Oil Removal (%)			84.76	90.18

Artificial Neural Network (ANN)

In this section, the regression models of the oil adsorption process using human hair adsorbents were developed using two different approaches: manual coding in M-file and the built-in neural network toolbox. Subsequently, the optimisation of the developed models was performed using the GA approach.

ANN using Manual Coding

To create the model, experimental results in Tables 1 and 2 were partitioned randomly into validation data or unseen data and training and testing sets to avoid over-training and over parameterisation. Before the model development, bootstrap re-sampling was applied to resample the collected data. The general idea of the network development is illustrated in Figure 2.

Since there were no theoretical principles in choosing the proper network topology, several structures were tested to obtain the best one. The number of neurons in the hidden layer was varied from 1 to 30, and the network was trained and tested after each addition of neurons. The lowest mean squared error (MSE) obtained for the trained network was 2.9237 with three neurons in the hidden layer for the gents' hair adsorbent. The configuration of ANN was therefore defined as 3-3-1. Evaluation of the established ANN model with the validation data (previously unseen) indicated that the model demonstrated an R^2 value of 0.3695 and an MSE value of 1.2953.

On the other hand, the lowest MSE obtained for ladies' hair adsorbent was 0.8819 with only one neuron in the hidden layer, leading to the ANN configuration of 3-1-1. Similarly, the developed ANN model was validated based on unseen data to determine the network's generalisation capacity, and the evaluation results revealed R^2 value of 0.5654 and MSE value of 0.1357. The higher R^2 value of the ladies hair adsorption network compared to the gents' hair showed that closer agreement between the adsorption experimental results and theoretical values predicted by ANN for ladies hair, indirectly indicating that the trained ANN model was capable of defining the actual behavior of the oil adsorption system better for ladies hair as compared to the gents' hair. The relatively lower values of MSE also supported this for the

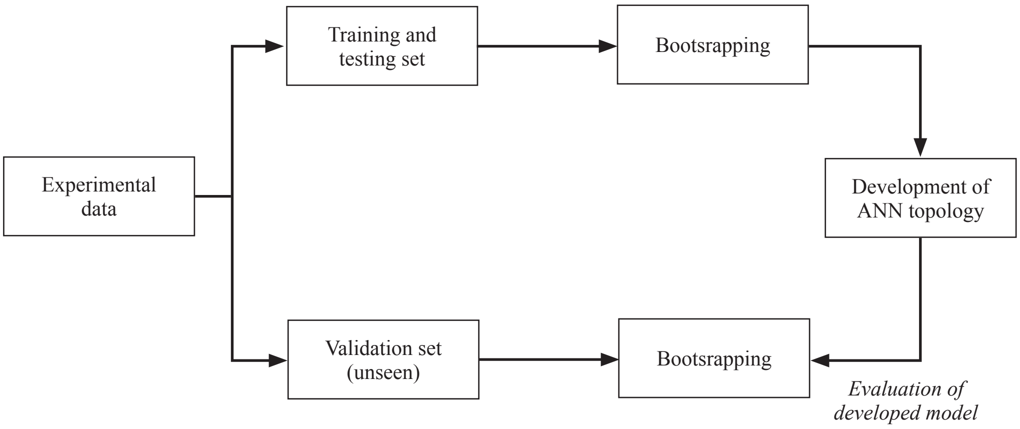


Figure 2 Illustrations of AAN network development

ladies' hair network. Generally, in adopting ANN using manual coding as the regression tool, the values obtained for both regression models developed were in the range of 0.37 – 0.57, which were relatively low. One of the most plausible explanations for this was probably due to the selection of data as training and validation sets could be easily manipulated and hence directly affected the R^2 value of the regression model generated. The flexibility to manually separate the experimental data into training and validation sets was beneficial for improving the network's generalisation capacity. Nevertheless, in this study, best network configurations of both models are still lowly correlated with the experimental results.

Following the establishment of ANN models of both adsorbents, optimisation via the GA approach was applied. The upper limits and lower limits of the parameters were pre-defined, and the GA optimisation results are presented in Table 6.

ANN using Matlab Built-In Neural Network Toolbox

Regression analysis was performed using Matlab's built-in neural network toolbox, and the predicted oil removal percentage based on the established network was extracted. However, using this method, the optimisation using GA in Matlab could not be performed as the toolbox generated regression models in the unretrievable network. Due to the limitation of retrieving the ANN network models from the toolbox, the values of both gents and ladies' hair adsorption models were calculated in Microsoft Excel using the ANN predicted results. Table 7 shows the regression analysis results for gents hair, and R^2 value as high as 0.9569 was attained. As for the ladies' hair, the results are tabulated in Table 8, and the R^2 value of the developed regression model was found to be 0.9650. The R^2 values of the ANN models were consistently higher than the results using manual coding for both gents' and ladies' hair.

Design Expert

All 17 experimental points were evaluated, and the oil removal percentage results based on the BBD experimental design are shown in Table 9 for gents hair and Table 10 for ladies' hair. Response surface optimisation is favorable over the conventional single parameter optimisation in that it saves time, space, and raw materials [23].

A maximum oil removal percentage of 88.97% was recorded for gents hair under the experimental conditions pf pH 1, contact time 71.75 minutes, and adsorbent dosage of 642.27 mg. By applying multiple regression analysis on the experimental data, the predicted response variable and the independent variables were related in terms of coded values by the quadratic Equation 3. For ladies' hair, a maximum oil removal percentage of 90.38% was obtained under the experimental conditions of pH 1, contact time 44.77 minutes, and adsorbent dosage of 796.22 mg. The predicted response variable and the independent variables were related to coded values by the quadratic Equation 4.

Table 6 Optimisations via GA approach

Parameters	Lower Limit	Upper Limit	Optimised Value for Gent Hair Model	Optimised Value for Ladies Hair Model
pH	1	7	1	1
Contact Time (min)	9.9	119.8	119.74	9.9
Adsorbent Dosage (mg)	96.9	1248.2	268.63	1196.9
Predicted Oil Removal (%)			90.59	86.35

Table 7 Regression analysis results for gents hair

Input			Output(Y)	Network Output (Y*)	Y - Y*	Y-Ymean
pH	**Contact Time (min)**	**Adsorbent dose (mg)**	**% of oil removal**	**% of oil removal**	**sqrd err w/line**	**sqrd from mean y**
1.0	30.0	500.0	77.4	77	1.96422E-05	30.84060646
3.0	30.0	500.0	70.2	70	0.000494739	2.743139942
5.0	30.0	500.0	59.1	59	0.000836186	161.3683208
7.0	30.0	500.0	51.2	51	0.000239063	424.7334225
1.0	9.9	500.0	52.6	52	0.051922607	369.3290131
1.0	19.8	500.0	61.7	62	0.002551113	103.4839991
1.0	29.9	500.0	78.5	77	1.43715397	45.04709902
1.0	39.7	500.0	67.1	71	13.74832118	22.52046181
1.0	49.8	500.0	75.5	76	0.000337156	13.66513391
1.0	59.8	500.0	91.8	92	0.006265478	399.1222911
1.0	69.9	500.0	94.2	92	4.485100253	501.3168255
1.0	80.0	500.0	85.8	88	3.218455833	194.5437602
1.0	90.0	500.0	80.7	80	0.593310222	77.83238323
1.0	99.7	500.0	74.9	75	0.001419548	9.570503981
1.0	110.1	500.0	72.8	73	0.093837853	0.966427317
1.0	119.8	500.0	71.0	71	0.029044723	0.682234493
1.0	70.0	99.1	59.4	52	53.17183851	155.5033378
1.0	70.0	248.2	71.2	71	0.001629458	0.42937425
1.0	70.0	498.2	94.8	92	7.299608843	527.0779752
1.0	70.0	752.6	65.3	73	56.85831406	42.72119428
1.0	70.0	1002.6	65.4	65	0.00143729	41.62974413
1.0	70.0	1248.2	59.7	60	0.003210585	146.0191403
			Mean Y=		Σsqrd err w/line=	Σsqrd from mean Y=
			71.831315		141.0053483	3271.146388

Σsqrd(sqrd error w/line-from mean y) =	R^2
R^2=	0.04310579
1- R^2=	0.95689421

Table 8 Regression analysis results for ladies hair

Input			Output(Y)	Network Output (Y*)	Y - Y*	Y-Ymean
pH	Contact Time (min)	Adsorbent dose (mg)	% of oil removal	% of oil removal	sqrd err w/line	sqrd from mean y
1.0	30.0	500.0	87.5	87.9	0.125814821	40.53354654
3.0	30.0	500.0	84.5	84.5	4.35834E-05	11.17967281
5.0	30.0	500.0	74.1	72.3	3.357058952	49.07804093
7.0	30.0	500.0	66.9	66.9	0.000200313	202.0732418
1.0	9.9	500.0	81.0	81.2	0.066268213	0.032122006
1.0	20.0	500.0	85.8	85.3	0.270810407	21.57504466
1.0	30.1	500.0	88.2	87.9	0.099827419	49.80061068
1.0	39.7	500.0	88.8	89.3	0.305621176	58.67513809
1.0	49.8	500.0	88.8	90.2	2.015917657	58.67513809
1.0	59.8	500.0	91.8	90.5	1.692198302	113.9565906
1.0	69.7	500.0	88.8	90.1	1.663403862	58.67513809
1.0	79.8	500.0	86.1	88.3	4.872605886	24.46689285
1.0	89.8	500.0	84.6	84.6	0.000183215	11.82578782
1.0	99.9	500.0	75.5	79.2	13.29230833	31.43129273
1.0	109.7	500.0	73.7	73.8	0.002756989	54.9882615
1.0	119.8	500.0	72.5	70.1	5.677558628	74.32917915
1.0	60.0	96.9	64.7	64.9	0.076307688	271.2786372
1.0	60.0	248.2	71.4	71.6	0.049137248	94.86310467
1.0	60.0	498.2	92.6	90.5	4.299210852	131.3809708
1.0	60.0	746.1	72.7	72.7	0.007989609	70.45277922
1.0	60.0	998.2	81.8	78.3	12.35327831	0.480142391
1.0	60.0	1248.2	83.2	83.1	0.004952911	4.157834698
			Mean Y=		Σsqrd err w/line=	Σsqrd from mean Y=
			81.13396582		50.23345437	1433.909167

Σsqrd(sqrd error w/line-from mean y) =	R^2
R^2=	0.035032522
1- R^2=	0.964967478

Table 9 Experimental design for gents hair

No.	pH	Contact Time (min)	Adsorbent dose (mg)	Y,% of oil removal
1	1.0	30.0	500.0	77.4
2	3.0	30.0	500.0	70.2
3	5.0	30.0	500.0	59.1
4	7.0	30.0	500.0	51.2
5	1.0	9.9	500.0	52.6
6	1.0	19.8	500.0	61.7
7	1.0	29.9	500.0	78.5
8	1.0	49.8	500.0	75.5
9	1.0	69.9	500.0	94.2
10	1.0	80.0	500.0	85.8
11	1.0	99.7	500.0	74.9
12	1.0	119.8	500.0	71.0
13	1.0	70.0	99.1	59.4
14	1.0	70.0	248.2	71.2
15	1.0	70.0	498.2	94.8
16	1.0	70.0	1002.6	65.4
17	1.0	70.0	1248.2	59.7

Table 10 Experimental design for ladies hair

No.	pH	Contact Time (min)	Adsorbent dose (mg)	Y,% of oil removal
1	1.0	30.0	500.0	87.5
2	3.0	30.0	500.0	84.5
3	5.0	30.0	500.0	74.1
4	7.0	30.0	500.0	66.9
5	1.0	9.9	500.0	81.0
6	1.0	20.0	500.0	85.8
7	1.0	39.7	500.0	88.8
8	1.0	59.8	500.0	91.8
9	1.0	79.8	500.0	86.1
10	1.0	89.8	500.0	84.6
11	1.0	99.9	500.0	75.5
12	1.0	119.8	500.0	72.5
13	1.0	60.0	96.9	64.7
14	1.0	60.0	248.2	71.4
15	1.0	60.0	498.2	92.6
16	1.0	60.0	746.1	72.7
17	1.0	60.0	1248.2	83.2

Equation 3:

$$\text{Oil Removal (\%)} = 18.19 + 0.43X_1 - 0.48X_1^2 + 1.04X_2 - 0.01X_2^2 + 0.10X_3 - 7.84\text{E-}05X_3^2$$

Equation 4:

$$\text{Oil Removal (\%)} = 56.52 - 0.30X_1 - 0.38X_1^2 + 0.25X_2 - 0.01X_2^2 + 0.08X_3 - 4.98\text{E-}05X_3^2$$

Table 11 Optimisations using RSM

Parameters	Lower Limit	Upper Limit	Optimised Value for Gent Hair Model	Optimised Value for Ladies Hair Model
pH	1	7	1	1
Contact Time (min)	9.9	119.8	71.70	44.77
Adsorbent Dosage (mg)	96.9	1248.2	642.13	796.22
Predicted Oil Removal (%)			88.97	90.38

where Y = percentage of oil removal; X_1 = pH; X_2 = contact time (minutes); and X_3 = adsorbent dosage (mg). The optimised adsorption conditions are tabulated in Table 11.

The coefficient with one factor represents the effect of that particular factor. In contrast, the coefficients with two factors and those with second-order terms represent the interaction between the two factors and the quadratic effect, respectively. The positive sign of the terms indicates the synergistic impact, while the negative sign indicates an antagonistic impact [24]. ANOVA was used to evaluate the significance of the coefficients of the model. The equation's regression coefficient values are listed in Table 12 and Table 13 for gents hair and ladies' hair, respectively.

Table 12 ANOVA results for parameters in gents hair model

Parameter	Regression coefficient	Standard error	F value	P value	Indication
Linear					
X_1	-11.03	3.56	9.61	0.0112	Significant
X_2	6.24	3.47	3.23	0.1024	Not Significant
X_3	-3.44	3.96	0.76	0.4047	Not Significant
Quadratic					
X_1^2	-3.42	6.77	0.26	0.6239	Not Significant
X_2^2	-25.01	5.80	18.62	0.0015	Significant
X_2^3	-31.49	6.35	24.61	0.0006	Significant

The P values were used as a tool to check the significance of each coefficient. Zhang et al. [23] stated that significant regression coefficient and small P value are implying more significant effect on the respective response variable(s) for any of the terms in a particular model. This indicates that the smaller is the value of P, the more significant is the corresponding variable. From Table 12, it can be seen that the linear coefficient (X_1) and the quadratic term coefficients (X_3^2, X_3^2) are significant while from Table 13, linear coefficient (X_1) and quadratic term coefficient (X_3^2) are significant, with significant P values (P < 0.05).

Table 13 ANOVA results for parameters ladies hair model

Parameter	Regression coefficient	Standard error	F value	P value	Indication
Linear					
X_1	-9.74	3.55	7.51	0.0208	Significant
X_2	-5.55	3.43	2.62	0.1368	Not Significant
X_3	6.67	4.30	2.41	0.1516	Not Significant
Quadratic					
X_1^2	-3.73	6.76	0.30	0.5932	Not Significant
X_2^2	-7.59	5.63	1.82	0.2076	Not Significant
X_3^2	-15.52	5.98	6.74	0.0267	Significant

According to Zhang et al. [23] and Wang et al. [25], exploration and optimisation of a fitted response surface might produce poor or misleading results unless the model exhibits a good fit, which makes the checking of the model adequacy essential. The P value of the model for gents hair was 0.0023 (Table 14), and the P value of the model for ladies' hair was 0.0818 (Table 15). Besides, a lower value of the variation (CV) coefficient at 9.62% for gents hair and 8.46% for ladies, which is lower than 10% indicated a better precision and reliability of the experiments [26]. CV was frequently used as an indication of the degree of accuracy with which the treatments were compared. The higher the value of CV, the lower the reliability of the experiment is [27].

Another critical analysis by ANOVA was checking the precision of a particular model by determining the correlation coefficient (R^2). It is defined as the ratio of the explained variation to the total variation and measuring the degree of fitness [18]. A regression model having R^2 value higher than 0.9 or approaches unity is considered to have a very high correlation, and the model can fit well with experimental data. Likewise, a small value of R^2 implies a poor relevance of the dependent variables in the model [25],[27]. Here, the value of R^2 is 0.8290 for gents hair (Equation 3), and the value of R^2 is 0.6158 for ladies hair (Equation 4) indicates a closer agreement between the experimental results and theoretical values predicted by the model equation for gents hair, as compared to the ladies hair.

Table 14 ANOVA analysis results for gents hair model

Source	Degree of freedom	Sum of squares	Mean square	F value	P value
Model	6	2245.04	374.17	8.08	0.0023
Residual	10	463.21	46.32	-	-

Table 15 ANOVA analysis results for ladies hair model

Source	Degree of freedom	Sum of squares	Mean square	F value	P value
Model	6	738.11	123.02	2.67	0.0818
Residual	10	460.53	46.05	-	-

Comparing the Efficiency of Different Regression Tools

The predictive capability of three regression tools, namely the Microsoft Excel, Matlab, and Design Expert, was evaluated and compared. The comparisons were made based on value of the regression models generated by all tools using similar experimental results. As summarised in Table 16, regression models developed using Matlab built-in neural toolbox demonstrated the highest value with approximately 0.96 for both gents' hair and ladies' hair. The results revealed a significantly higher generalisation capacity of ANN compared to the other regression tools. On the other hand, the accuracy of RSM in predicting the adsorption models was superior to Microsoft Excel. The higher predictive accuracy of the ANN could be attributed to its universal ability to approximate the nonlinearity of the system, whereas the RSM and excel were restricted [28]. In the work of Ram Talib et al. [29], similar observations were reported whereby the validation analysis showed that the predicted values by RSM and ANN were close to the experimental results for the reduction of chromium (VI) by newly isolated *Acinetobacter* radioresistance strain NS-MIE from agricultural soil. The authors reported that ANN showed the lowest deviation of 2.57 % in the validation result compared to the RSM though both the R^2 values were close to 1 (0.9974 for RSM and 0.9991 for ANN). This finding suggests that the ANN showed a better prediction and fitting ability than the RSM for nonlinear regression analysis [28]. Based on the current results, it is demonstrated that the ANN technique is more feasible than the RSM technique or Excel technique due to the requirement of a large number of data to develop an accurate regression model by the latters. At the same time, the neural network could recognise the associations with lesser information [30].

Table 16 Comparison of Regression Tools

		R^2 Value pH	Optimised Adsorption Parameters			
			Contact Time (min)	Adsorbent Dosage (mg)	Oil Removal (%)	
ANN (M-file)	*1*	0.3695	1	119.74	268.63	90.59
	2	0.5654	1	9.90	1196.90	86.35
ANN (NNTool)	*1*	0.9570	-	-	-	-
	2	0.9650	-	-	-	-
Microsoft Excel	*1*	0.6854	1	72.54	608.97	84.76
	2	0.6399	1	44.68	759.57	90.18
RSM – BBD Model	*1*	0.8290	1	71.70	642.13	88.97
	2	0.6158	1	44.77	796.22	90.38

1. Gents hair; 2. Ladies hair

The regression model with the highest R^2 value was selected to develop GUI for the prediction of oil removal within the specified range of process parameters. In this study, it was found that ANN using the built-in neural toolbox established the regression model with the highest R^2. The model however was not selected as the equation was generated using Microsoft Excel with the toolbox's predicted response. Though ANN is a powerful data-driven and flexible computational tool possessing the capability of capturing non-linear and complex data, the regression model was captured as a network whereby the relationship between the manipulating parameters and response variables was not accessible. Thus for the development of GUI in the subsequent section, the regression model generated using RSM was selected, which demonstrated the second-highest R^2 value among all models.

Development of Graphical User Interface (GUI)

Figure 3 displays the first layout that can be seen on the Microsoft Excel worksheet. This layout briefly explains human hair waste and its potential to be used as a natural adsorbent to remove oil. Besides, this layout gives a general idea of the user form as well. The user form is displayed by clicking on the 'Oil Adsorption Using Human Hair' button, as shown in Figure 4.

Figure 4 shows the GUI layout that was developed for the oil adsorption using human hair adsorbent. It comprises of two components. The first component (left panel) displays the relationship between the adsorption parameters with the oil removal percentage. The second component (right panel) is to display the oil removal percentage's prediction at the specified parameter value. The oil removal percentage at the specified

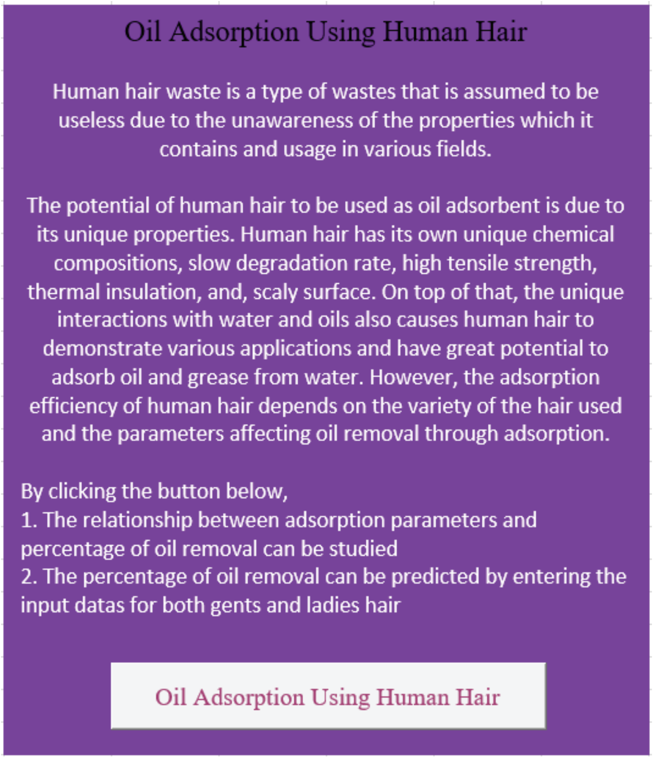


Figure 3 First layout of GUI

input parameters will be predicted based on the equation obtained using the RSM regression tool (Equation 3 for gents hair and Equation 4 for ladies hair). The user may select whether the adsorbent used is of gents or ladies' hair.

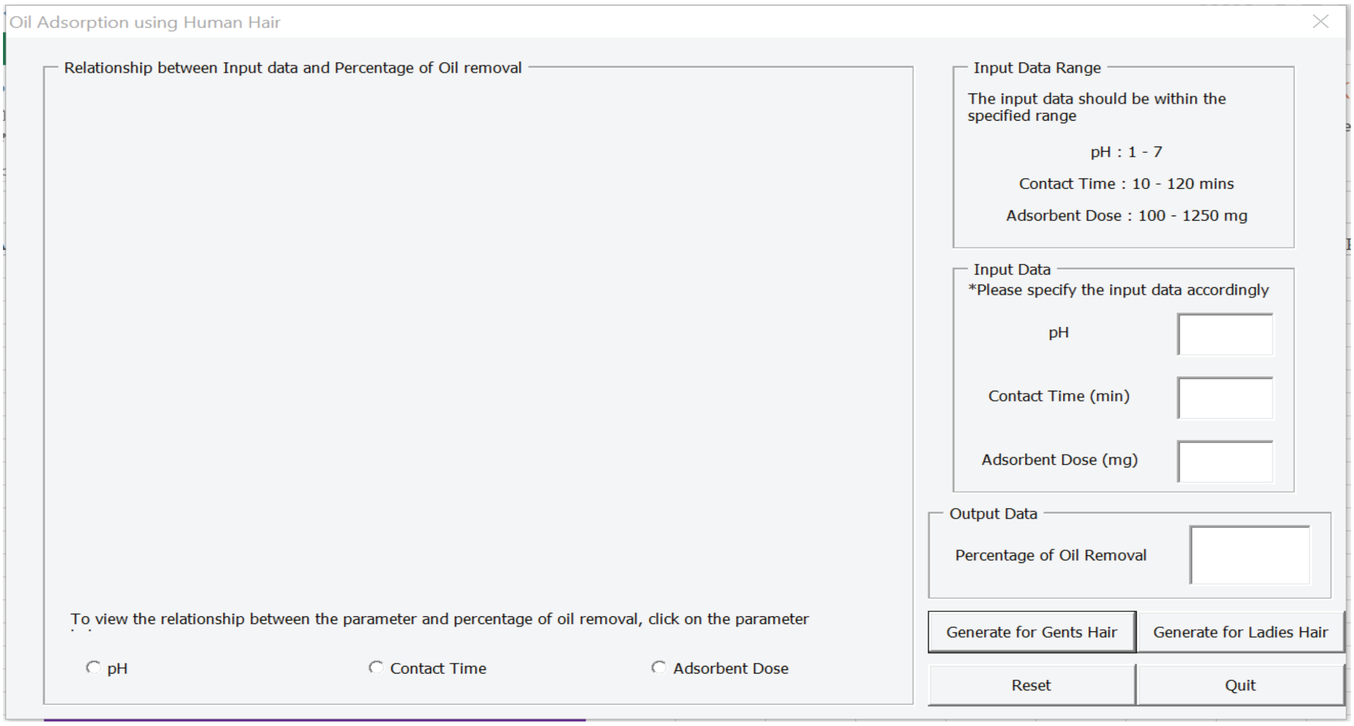
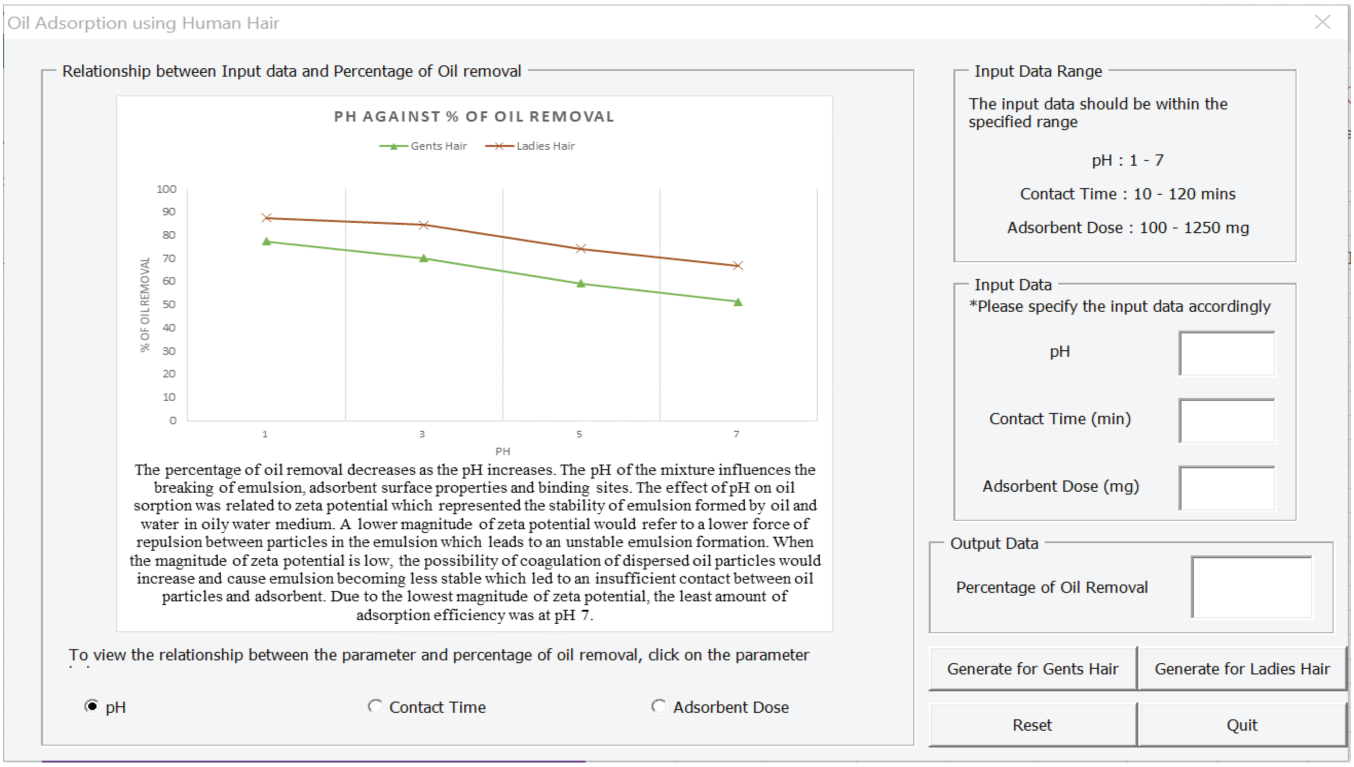
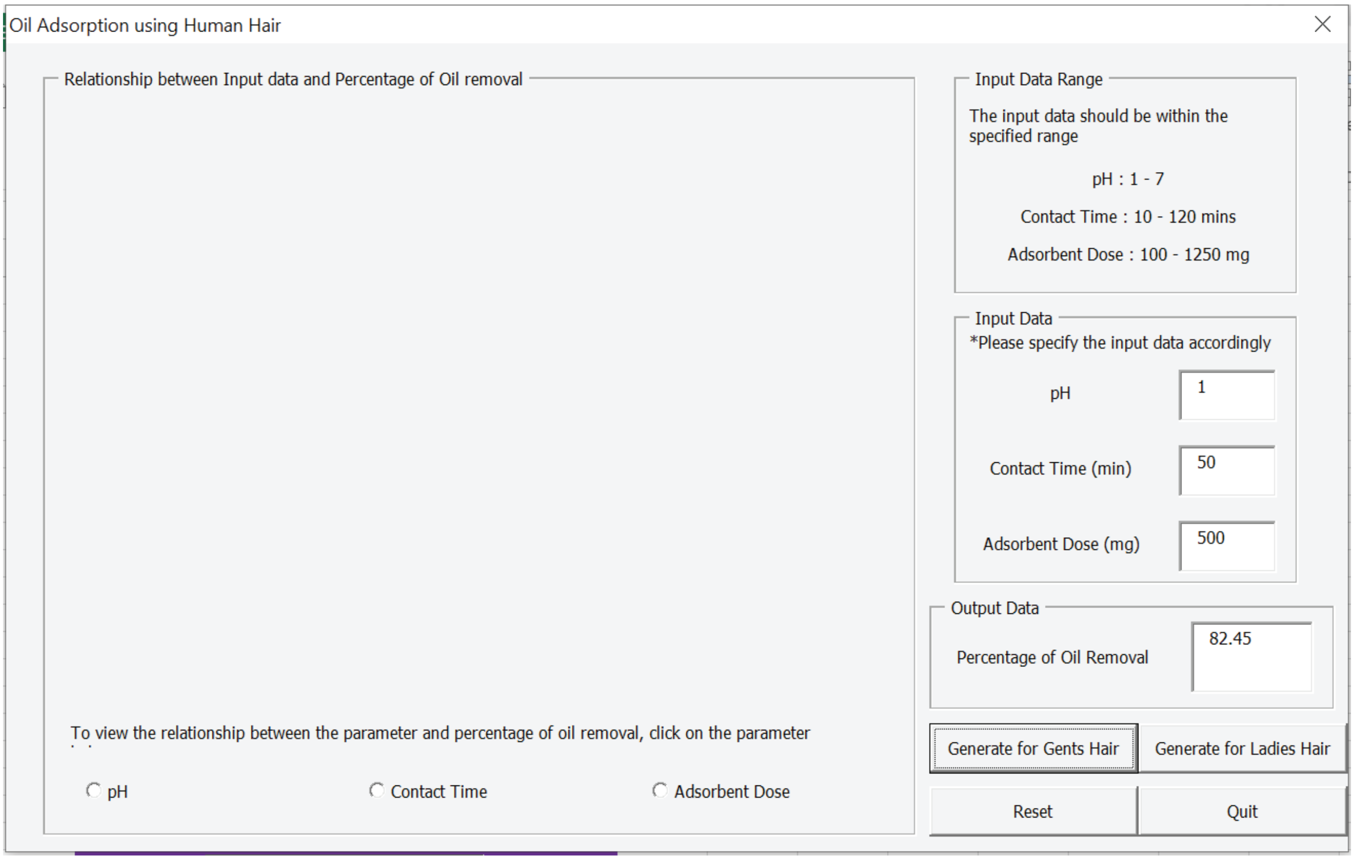


Figure 4 The created GUI layout for the adsorption of oil using human hair adsorbent



(a)



(b)

Figure 5 The results illustrated in the GUI layout showing (a) the relationship between the parameters and oil removal percentage and (b) predicted oil adsorption removal at adsorption parameters defined by users.

Figure 5a shows the results illustrated in the GUI layout for the relationship between pH (in the range of 1 – 7) and the oil removal percentage, while Figure 5b is the example of how the percentage of oil removal is revealed when the users define all adsorption parameters within the specific range.

CONCLUSION

In conclusion, the current study compared the efficiency of different regression tools in predicting oil removal percentage using human hair as an adsorbent and developed a user-friendly GUI for the oil adsorption process. Based on the comparison of regression tools, the ANN approach via Matlab built-in neural toolbox showed superiority over the manual ANN M-file coding, RSM and Microsoft Excel. Its capability to correlate the adsorption parameters to the oil removal percentage was evidenced through the highest R^2 values of the regression model for both gents and ladies hair adsorbents, among all other techniques. In creating the GUI layout, the correlation models generated using RSM (the second-highest R^2 values) were used as the reference equations instead, owing to the limitation in retrieving the regression models from the ANN toolbox. With the GUI development, users can predict the oil removal percentage by manipulating the adsorption process parameters within the specified range of process parameters. To conclude, there is ample room for further research in utilising human hair waste as a natural adsorbent to remove oil and grease from wastewater, requiring a little processing to result in a higher oil removal percentage. Future studies can also be extended to incorporate the effects of temperature, the particle size of hair adsorbent, and different pre-treatment types on human hair towards the oil removal efficiency. The accuracy of all models generated need to be evaluated using validation of the predicted optimised conditions to fortify all the tools' prediction capability further.

REFERENCES

[1] M.F. Dario, R. Pahl, J.R. de Castro, F.S. de Lima, T.M. Kaneko, C.A. Pinto, A.R. Baby, & M.V.R. Velasco, "Efficacy of Punica granatum L. hydroalcoholic extract on properties of dyed hair exposed to UVA radiation," *Journal of Photochemistry and Photobiology B,* 120, pp. 142–147, 2013. [Online]. Retrieved from http://dx.doi.org/10.1016/j.jphotobiol.2012.12.011. 364 23380541.

[2] C. LaTorre, & B. Bhushan, "Investigation of scale effects and directionality dependence on friction and adhesion of human hair using AFM and macroscale friction test apparatus," *Ultramicroscopy,* 106, 8–9, pp. 720-734, 2006. [Online]. Retrieved from https://doi.org/10.1016/j.ultramic.2005.11.010.

[3] R. Vargiolu, C. Pailler-Mattei, M. Coudert, Y. Lintz, & H. Zahouani, "Hair surface and mechanical properties of Copt mummies from Antinopolis," *Journal of Archaeological Science;* 40, 10, pp. 3686–3692, 2013. [Online]. Retrieved from http://dx.doi.org/10.1016/j.jas.2013.04.007.

[4] R. Chandrappa, & D.B. Das, "Solid waste management: Principles and practice," *Springer Science & Business Media*, 2012.

[5] A. Gupta, "Human hair "waste" and its utilization: Gaps and possibilities," *Journal of Waste Management,* pp. 1–17, 2014. [Online]. Retrieved from https://doi.org/10.1155/2014/498018.

[6] U. Mishra, "Human hair waste, uses & sources: A review," *International Journal of Scientific Research and Development,* 5, 02, pp. 914-917, 2017.

[7] Y. Zhang, C. Song, L. Ji, Y. Liu, J. Xiao, X. Cao, & Y. Zhou, "Cause and effect of N/P ratio decline with eutrophication aggravation in shallow lakes," *Science of the Total Environment,* 627, pp. 1294-1302, 2018.

[8] C. Thieulin, R. Vargiolu, & H. Zahouani, "Effects of cosmetic treatments on the morphology, biotribology, and sensorial properties of a single human hair fiber," *Wear 426*, pp. 186–194, 2019. [Online]. Retrieved from https://doi.org/10.1016/j.wear.2019.01.065.

[9] M.V.R. Velasco, T.C.D. S. Dias, A.Z.D. Freitas, N.D.V. Júnior, C.A.S.D.O. Pinto, T.M. Kaneko, & A.R. Baby, "Hair fiber characteristics and methods to evaluate hair physical and mechanical properties," *Brazilian Journal of pharmaceutical sciences,* 45, 1, pp. 153-162, 2019. [Online]. Retrieved from https://doi.org/10.1590/s1984-82502009000100019.

[10] S. Saha, M. Zubair, M.A.Khosa, S. Song, & A. Ullah, "Keratin and chitosan biosorbents for wastewater treatment: a review," *Journal of Polymers and the Environment,* 27, 7, pp. 1389-1403, 2019. [Online]. Retrieved from https://doi.org/10.1007/s10924-019-01439-6.

[11] H. Zhang, F. Carrillo, M. López-Mesas, & C. Palet, "Valorization of keratin biofibers for removing heavy metals from aqueous solutions," *Textile research journal,* 89, 7, pp. 1153-1165, 2019. Retrieved from https://doi.org/10.1177/0040517518764008.

[12] H.S. Abd El-Gawad, *Oil and Grease Removal from Industrial Wastewater Using New Utility Approach*. *Advances in Environmental Chemistry,* Article ID, 916878, 2014. [Online]. Retrieved from https://doi.org/10.1155/2014/916878.

[13] T. Wallace, D. Gibbons, M. O'Dwyer, & T.P. Curran, "International evolution of fat, oil and grease (FOG) waste management–A review," *Journal of environmental management,* 187, pp. 424-435, 2017. [Online]. Retrieved from https://doi.org/10.1016/j.jenvman.2016.11.003.

[14] W.A.J. Anurangi, *Natural and Waste Materials as Adsorbents in Oil Pollution Management,* (Master's Thesis, University of Moratuwa, Sri Lanka), 2011. [Online]. Retrieved from http://dl.lib.mrt.ac.lk/handle/123/2036.

[15] P.R. Ukotije-Ikwut, A.K. Idogun, C.T. Iriakuma, A. Aseminaso, & T. Obomanu, "A novel method for adsorption using human hair as a natural oil spill sorbent," *International Journal of Scientific & Engineering Research,* 7, 8, pp. 1754–1764, 2016.

[16] Z. Shahryari, A. Sharifi, & A. Mohebbi, "Artificial neural network (ANN) approach for modeling and formulation of phenol adsorption onto activated carbon," *Journal of Engineering Thermophysics,* 22, 4, pp. 322-336, 2013. [Online]. Retrieved from https://doi.org/10.1134/s181023281304005x.

[17] C. Sutherland, B. Chittoo, & C. Venkobachar, "Application of ANN predictive model for the design of batch adsorbers - Equilibrium simulation of Cr(VI) adsorption onto activated carbon," *The Open Civil Engineering Journal;* 13, 1, pp. 69–81, 2019. https://doi.org/10.2174/1874149501913010069.

[18] O.D. Samuel, & M.O. Okwu, "Comparison of Response Surface Methodology (RSM) and Artificial Neural Network (ANN) in modelling of waste coconut oil ethyl esters production," *Energy Sources, Part A: Recovery, Utilization, and Environmental Effects,* 41, 9, pp. 1049-1061, 2019. [Online]. Retrieved from https://doi.org/10.1080/15567036.2018.1539138.

[19] S. Saini, J. Chawla, R. Kumar, & I. Kaur, "Response surface methodology (RSM) for optimisation of cadmium ions adsorption using C16-6-16 incorporated mesoporous MCM-41," *SN Applied Sciences,* 1, 8, 2019. [Online]. Retrieved from https://doi.org/10.1007/s42452-019-0922-5.

[20] S. Najaa, M. G. Rozidaini, & I. Norhisyam, "Response surface methodology optimisation of oil removal using banana peel as biosorbent," *Malaysian Journal of Analytical Sciences,* 21, 5, pp. 1101-1110, 2017. [Online]. Retrieved from https://doi.org/10.17576/mjas-2017-2105-12.

[21] N.W. Ingole, S.S. Vinchurkar, & S.V. Dharpal, "Adsorption of oil from waste water by using human hair," *Journal of Environmental Science, Computer Science and Engineering & Technology,* 3, 1, pp. 207–217, 2014.

[22] A.O. Ifelebuegu, T.V.A. Nguyen, P. Ukotije-Ikwut, & Z. Momoh, "Liquid-phase sorption characteristics of human hair as a natural oil spill sorbent," *Journal of Environmental Chemical Engineering,* 3, 2, pp. 938-943, 2015. [Online]. Retrieved from https://doi.org/10.1016/j.jece.2015.02.015.

[23] L.L. Zhang, M. Xu, Y.M. Wang, D.M. Wu, & J.H. Chen, "Optimizing ultrasonic ellagic acid extraction conditions from infructescence of Platycarya strobilacea using response surface methodology," *Molecules,* 15, 11, pp. 7923-7932, 2010. [Online]. Retrieved from https://doi.org/10.3390/molecules15117923.

[24] L. Rodrigues, J. Teixeira, R. Oliveira, & H.C. Van Der Mei, "Response surface optimisation of the medium components for the production of biosurfactants by probiotic bacteria," *Process biochemistry,* 41, 1, pp. 1-10, 2006. [Online]. Retrieved from https://doi.org/10.1016/j.procbio.2005.01.030.

[25] L. Wang, B. Yang, X. Du, Y. Yang, J. Liu, "Optimisation of conditions for extraction of acid-soluble collagen from grass carp (*Ctenopharyngodon idella*) by response surface methodology," *Innovative Food Science & Emerging Technologies,* 9, 4 pp. 604–607, 2008. [Online]. Retrieved from https://doi.org/10.1016/j.ifset.2008.03.001.

[26] A. Nath, & P.K. Chattopadhyay, "Optimisation of oven toasting for improving crispness and other quality attributes of ready to eat potato-soy snack using response surface methodology," *Journal of Food Engineering,* 80, 4, pp. 1282-1292, 2007. [Online]. Retrieved from https://doi.org/10.1016/j.jfoodeng.2006.09.023.

[27] B. Zhang, Y. Chen, X. Wei, M. Li, & M. Wang, "Optimisation of conditions for collagen extraction from the swim bladders of grass carp (*Ctenopharyngodon idella*) by response surface methodology," *International Journal of Food Engineering,* 6, 3, 2010. [Online]. Retrieved from https://doi.org/10.2202/1556-3758.1772.

[28] S. Youssefi, Z. Emam-Djomeh, S.M. & Mousavi, "Comparison of artificial neural network (ANN) and response surface methodology (RSM) in the prediction of quality parameters of spray-dried pomegranate juice," *Drying Technology,* 27, (7-8), pp. 910-917, 2009. [Online]. Retrieved from https://doi.org/10.1080/07373930902988247.

[29] N.S. Ram Talib, M.I.E. Halmi, S.S. Abd Ghani, U.H. Zaidan, & M.Y.A. Shukor, "Artificial neural networks (ANNs) and response surface methodology (RSM) approach for modelling the optimisation of chromium (VI) reduction by newly isolated acinetobacter radioresistens strain NS-MIE from agricultural soil," *BioMed research international,* pp. 1–14, 2019. [Online]. Retrieved from https://doi.org/10.1155/2019/5785387.

[30] K.A. Patel, & P.K.A. Brahmbhatt, "A comparative study of the RSM and ANN models for predicting surface roughness in roller burnishing," *Procedia Technology,* 23, pp. 391-397, 2016. [Online]. Retrieved from https://doi.org/10.1016/j.protcy.2016.03.042.