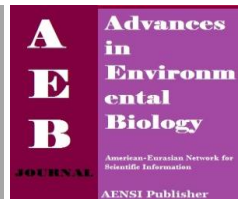




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Prediction of optimal adsorption of aqueous phenol removal with Oil Palm Empty Fruit Bunch Activated Carbon using Artificial Neural Network (ANN)

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ABSTRACT

Adsorption process has an edge in wastewater treatment over other techniques due to low initial cost, sludge free, ease of operation and insensitivity to toxic substance. It is a very essential part in the wastewater treatment process chain. It involves both physical and chemical phenomena and hence susceptible to high percentage of errors due to human factor, variation in the quality as well as chemical/physical characteristics of raw materials used. In order to reduce this percentage error and obtain optimal treatment efficiency, an intelligent method of predicting optimal adsorption capacity based on Artificial Neural Network (ANN) was proposed. Production of Powdered Activation Carbon PAC from processed oil palm empty fruit bunches, EFB was used as adsorbent. Since production of PAC is affected by many parameters, such as CO₂ gas flow rate, activation time and activation temperature. Adsorption design was carried out using all these parameters and production results were analyzed. ANN was used to forecast optimal adsorption capacity for aqueous phenol removal. Such ANN based system will be a useful method to address most errors common in wastewater treatment cause by human factors. Experimental results on real data show that the newly developed system is able to accurately predict the optimal adsorption capacity needed in wastewater treatment plant. The Regression and correlation between of optimal adsorption capacity for experimental result and ANN estimation model is 0.9999 of 1.000. This high Correlation of coefficient indicates that the ANN model is a perfect match.

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INTRODUCTION

The main purpose of wastewater treatment, a field belonging to environmental protection is to maintain a good quality in the receiving waters by removing organic substances and nutrients [1]. This is essential to life of both human and animal communities. Due to the rapidly growing of industrialization and urbanization, the risk of epidemics was growing, global warming, emissions of greenhouse gases and bad sanitary disposal were recognized as one of the most important factors in environment protection.

This was the driving force for creation of environmental agencies such as the Global Environment Facility (GEF) to address global environmental issues, the Environmental Protection Agency (EPA) that helps in monitoring and ensure that the urban wastewater - effluent from houses and businesses are treated before it is discharged to rivers, estuaries or the sea in order to prevent pollution. In this context, European laws envisage a series of specific orientations for treating and maintaining the water quality within legal limits (eg. surface water directives, 75/440/EEC and 79/869/EEC, drinking water directives 80/778/EEC/15July1980 and 98/83/EEC/3 November 1998, urban wastewater treatment directive 91/271/EEC), [2].

The wastewater treatment processes are very complex, it is a non-linear and characterized by many uncertainties such as the influent parameters, the structure and the coefficients of the model. Moreover, many wastewater treatment plants do not have measurement and control equipment. Therefore, there is a need in designing of intelligent method for better operation of the treatment plant.

Wastewater treatment is divided into three main processes.

1. Physical processes comprising screening or straining, sedimentation, flocculation and filtration.
2. Chemical treatment using adsorption, coagulation, ion exchange, and precipitation.
3. Biological treatment processes with dispersed growth system (activated sludge, stabilization ponds); fixed film reactors (biological filters such as tricking filter)

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Though wastewater treatment processes normally use physical processes initially and later chemical processes like precipitation or adsorption. However, if it not sufficiently treated by these two processes, the three processes can be applied. Figure 1 shows the complete wastewater treatment process chain.

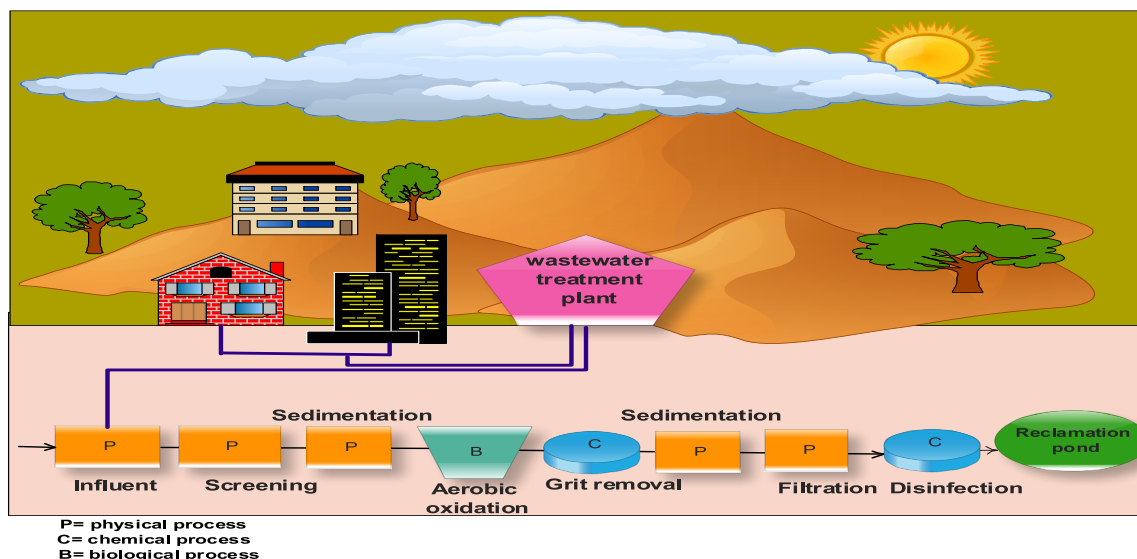


Fig. 1: Wastewater treatment process chain

Adsorption is a fundamental process in the physicochemical treatment of municipal wastewaters, a treatment which can economically meet today's higher effluent standards and water reuse requirements. It is the process through which a substance, originally present in one phase, is removed from that phase by accumulation at the interface between that phase and a separate (solid) phase [3].

Powdered activated carbon (PAC) for adsorption is one of the leading treatment technologies for reduction of organic contaminants in wastewater [4]. Adsorption on PAC finds growing use as an effective and economical process for purifying liquids by separating low concentrations of absorbable molecules from liquids. Example is the removal of dissolved organics from industrial and municipal waste streams. In PAC Adsorption, the activated carbon adsorbs impurities from liquids which are useful for optimizing process design, and for predicting the effect of changes in operating conditions. The process requires chemical knowledge of source wastewater characteristics to ensure that an effective adsorbent mix is employed. Inappropriate adsorbent makes these treatment methods ineffective. Hence an ANN based for prediction of optimal adsorption capacity is proposed in this paper.

Adsorption Process:

Adsorption practices are essential pretreatment for many wastewater purification systems, it is a technique widely used to remove certain classes of pollutants from waters, especially those that are not easily biodegradable [5]. Adsorption is the process in which some materials; (adsorbate) is concentrated from a bulk vapor or liquid phase on to the surface of a porous solid (adsorbent) that physical filtration processes can remove them easily. Particulate removal by these methods makes later filtering processes more effective. The driving force for adsorption is the reduction in interfacial (surface) tension between the fluid and the solid adsorbent. The surface tension, μ , is the change in free energy, K , resulting when the area between two phases, A , is increased.

μ is defined as:

$$\mu = \left(\frac{dK}{dA} \right) \quad (1)$$

Adsorption as a Physical Process:

In adsorption process, the material being adsorbed (e.g., a pollutant) is removed from one phase (e.g., a wastewater) and transferred to another phase such as activated carbon. This means that adsorption is a physical separation process in which the adsorbed material is not chemically altered. Hence, adsorption in wastewater treatment is associated with the removal of hazardous material(s) from the wastewater and its transfer to the activated carbon (P. M. Armenante). Therefore, activated carbon must be disposed properly. Figure 2 shows a typical adsorption process.

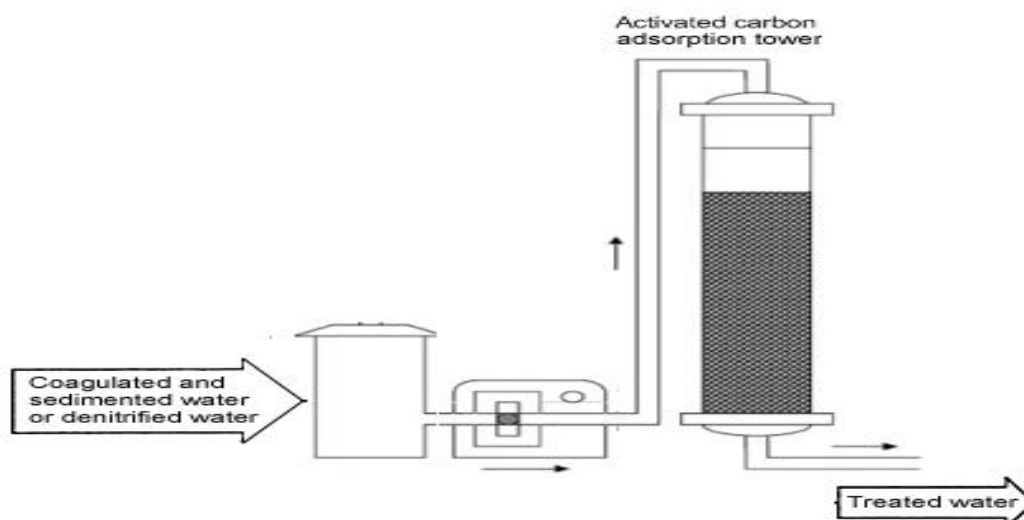


Fig. 2: Adsorption process

Factors affecting Adsorption:

The most important factors affecting adsorption are [3,6]:

- Surface area of adsorbent. Larger sizes imply a greater adsorption capacity.
- Temperature: adsorption reactions are usually exothermic, hence adsorption increases with decreasing temperature and vice versa.

$$\text{Adsorption} \propto \frac{1}{\text{Temperature}} \quad (3)$$

- Particle size of adsorbent. Smaller particle sizes reduce internal diffusional and mass transfer limitation to the penetration of the adsorbate inside the adsorbent.
- Pressure : At constant Temperature

$$\text{Adsorption} \propto \text{pressure} \quad (4)$$

- Solubility of solute (adsorbate) in liquid: Substances slightly soluble in water will be more easily removed from water (i.e., adsorbed) than substances with high solubility. Also, non-polar substances will be more easily removed than polar substances since the latter have a greater affinity for water.
- Size of the molecule with respect to size of the pores. Large molecules may be too large to enter small pores. This may reduce adsorption independently of other causes.
- Activation of Solid Adsorbent: When adsorbent is sub-divided or already adsorbed gases are removed, it becomes activated adsorbent and rate of adsorption increases.
- Degree of ionization of the adsorbate molecule. More highly ionized molecules are adsorbed to a smaller degree than neutral molecules.

The adsorption equilibrium:

Adsorption equilibrium is a function of the temperature. Hence, the adsorption equilibrium relationship at a given temperature is adsorption isotherm, that is;

$$q = f(C) \quad (5)$$

where:

q = mass of species adsorbed/mass of adsorbent

C = equilibrium concentration of adsorbable species in solution

Artificial Neural Network in Wastewater Treatment:

Related works:

Wastewater industries are now facing increased pressure to produce higher quality treated water before it can be discharged into streams. The efficiency of a treatment process closely relates to the design and operation of the plant. Most of the design and operations are still based on human experts. However decision making becomes very hard because the human experts, who have to make decisions, can hardly process the huge

amounts of data. To improve the operating performance and decision making process, an artificial Intelligence tool is needed. Recent works have taken advantage of artificial intelligence in Neural Network to design wastewater treatment process. Owing to the inherent characteristic of ANN like learning and adaptive capabilities, pattern mapping and classification and ability to generalize, not only to reproduce previously seen data, but also provide correct predictions in similar situations give the trained networks ability to infer. This offers a convenient way to reduce the amount of data as well as to form an implicit model without having to form a traditional, physical model of the underlying phenomenon [7]. Raduly, et al. [8], used Neural networks to evaluate the performance of wastewater treatment plant, due the fact that the plant behavior is affected by a wide range of influent disturbances, NN was used to combine an influent disturbance generator with a mechanistic waste-water treatment process to model the influent time series. Likewise [9] have used neural network in monitoring water quality for wastewater treatment processes. Therein, a backpropagation network is used to construct a soft measurement approach to monitor the effluent Chemical Oxygen Demand (COD) and biochemical oxygen demand (BOD) in wastewater treatment processes. The estimated NN result was used in real-time monitoring of biochemical wastewater. Researchers like [10] have used ANN to develop a model for predicting and forecasting daily BOD needed in inlet of wastewater biochemical treatment plant. Similarly, a neural network was adaptively used to model the control of a continuous wastewater treatment process by [11]. Mahmoud et al [12], used ANN to predict the performance of EL-AGAMY waste water treatment plant in terms COD, BOD and Total Suspended Solids (TSSs). The approach provides an effective analyzing and diagnosing tool to understand and simulate the non-linear behavior of the plant. Similarly, a Back-propagation algorithm was applied to determine pH value of which was found to be one of the factors affecting the coagulation condition of the suspended particles during the waste water treatment process. Recently, Jami et al [13] used multiple inputs- single output ANN models to analyze wastewater parameters and how it affects each other. The developed system was found suitable for analyzes of complexity and non-linear nature of the parameters.

MATERIAL AND METHOD

Artificial Neural Network (ANN) as emerged as a powerful tool for computational model based on biological neural networks [14]. In most cases ANN is an adaptive system that changes its structure based on external or internal information that flows through the network during the learning phase. Figure 2 shows architecture of Feedforward NN model used in predicting adsorption capacity.

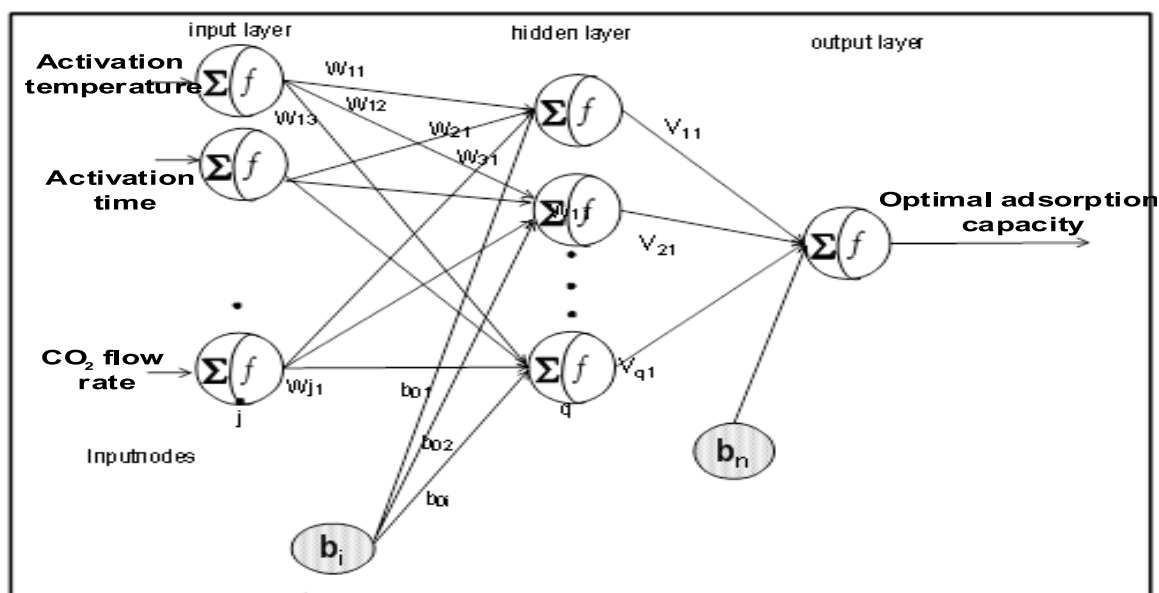


Fig. 2: Multilayer feedforward NN for prediction of Optimal Adsorption capacity

The operating parameters include activation temperatures, activation times and CO₂ gas flow rate. These three parameters serve as input neurons variables of the ANN at the input layer. Such parameters have the most influence on the determinant of the optimal condition for adsorption capacity of PAC produced from EFB. It has one output at the output layer which is the optimal adsorption capacity required for removal of phenol or pollutant in water treatment plant.

Prediction of adsorption capacity:

The correlation between production of powdered activated carbons from oil palm EFB and their adsorptive capacities on phenol removal is non-linear relationship. Therefore ANN based optimum adsorption capacity for removal of phenol involves 2 stages. The first stage involves determination of parameters affecting adsorption capacity of PAC prepared from oil palm empty fruit bunches as an adsorbent. These parameters are then considered as input variables to the neural network for training to determine the adsorptive capacities on phenol removal.

The parameters used are activation temperatures, activation times and CO₂ gas flow rate. The Fractional factorial design (FFD) experiments were conducted to obtain central point average of low and high rate for each parameter. Table 1 shows the FFD factors and their levels.

Table 1: Central point: average of high and low values for each parameter.

Factors	Temperature(°C)	Activation time (min)	Flow rate(L/min)
Average low	600	15	0.10
Average High	900	45	0.25

The oil palm empty fruit bunch (EFB) sample for production of powdered activation carbon was obtained from the Seri Ulu Langat Palm Oil Mill in Dengkil, Selangor, Malaysia. The Operating conditions of adsorption test by the PACs are as shown in Table 2.

Table 2: Operating conditions of adsorption test by PACs produced

PAC samples produced	Contact time(Hr)	Phenol aqueous solution			PAC dosage (g/L)	Agitation speed	Temperature
AC1, AC2, AC3, AC4, AC5, AC6, AC7, AC8, AC9 & AC10	0.2,0.5,0.75, 1, 2, 3, 4, 5, 6, 12, 18, 24, 48	Conc. 50mg/L	pH 6	Vol. 25ml	10g/L	150rpm	26±1°C, room temperature

The adsorption capacity (mg/g) was calculated using the following equations [4]:

$$\text{Adsorption capacity, } K_t = \frac{C_{P_i} - C_{P_f}}{N} V \quad (6)$$

where K_t is the adsorption capacity (mg/g) at time t ; C_{P_i} and C_{P_f} are the initial and final phenol concentrations (mg/L) respectively. N is the adsorbent dosage (g) and V is the volume of solution (L).

ANN Training and activation function:

The network is composed of a set of nodes/neurons and connections arranged in layers. Each neuron receives a signal from the neurons in the previous layer, and each of those signals is multiplied by a separate weight value. The weighted inputs are summed, and passed through an activation function which scales the output to a fixed range of values. The backpropagation (BP) algorithm [15,16,17] was used for the training. BP algorithm consists of four main steps as follows:

1. Initialization: set all weights and threshold level to random number, uniformly distributed in small range [14].
2. Activation: activate the forward phase of the BP algorithm by applying input and desired output
3. Weight training: update weights in the backward phase by propagating the error backwards
4. Iteration: increments iteration number p by one and repeat the cycle from step 2 until the overall error value drops below some predetermined threshold.

From Figure 2, for the training data input (Activation time, temperature and CO₂ flow rate) is represented by X and weights by W and Hidden layer by H .

For vector $X_m = (x_1, x_2, \dots, x_m)^T$ is applied to input of the network, the network input/output relationship is given by;

$$y(n) = f_i \left(\sum_{q=0} H_q V_{qi} + b_n \right) \text{ and}$$

$$H_q = f_n \left(\sum_{m=0} X_m W_{nm} + b_i \right) \quad (7)$$

Where W_{nm} is the weight between input and hidden neuron, V_{qi} weight between hidden neuron and output neuron, b_i is for the bias term of hidden neuron i while b_n is the bias of output neuron n .

In order to obtain best training iteration, training was terminated at interval of hundred iteration and the network error developed as follow:

$$E = y_n - d_n \quad (8)$$

where d_n is the desired optimum adsorption capacity and y_n is the output of network. The objective is to find the set of parameters that minimize the sum of the squared of the error function, where the average sum square error of the network is defined as;

$$E = \frac{1}{N} \sum_{n=1}^N (y_n - d_n)^2 \quad (9)$$

where N is the total number of neuron in the output layer. The network weight update is given by;

$$W_{nm}^{new} = W_{nm}^{old} + \Delta W_{nm} \quad (10)$$

Where

$$\Delta W_{nm} = -\eta \nabla E|_{W_{nm}} \quad (11)$$

η is the learning rate, $\nabla E|_{W_{nm}}$ is the gradient of the cost function. For the activation function, the Log sigmoid transfer function is used. Given by:

$$f(z) = \frac{1}{1 + \exp^{-z}} \quad (12)$$

where the derivatives of $f(z)$ is

$$f'(z) = z(1 - z) \quad (13)$$

RESULT AND DISCUSION

The model is assumed for central point between average high and average low values for each parameter (activation temperature, activation time and CO₂ gas flow rate). 40% of the data sets were used for training subsequent 40% for test and 20% for validation. The theoretical, experimental and ANN based adsorption capacity of activated carbon produced for its optimum production is shown in Table 3.

Table 3: Theoretical, experimental and ANN based results of adsorption capacity of activated carbon produced for its optimum production.

No of runs	Temperature (°C)	Activation time (min)	CO ₂ gas flow rate (L/min)	K_t Theory	K_t Experimental	K_t ANN based
1	600	45	0.25	0.9300	1.0280	1.0283
2	600	15	0.10	1.2700	1.3710	1.3715
3	900	45	0.10	4.7300	4.826	4.8224
4	900	15	0.10	4.9000	4.8040	4.8231
5	900	15	0.25	4.7100	4.8040	4.8040
6	750	30	0.175	4.8100	4.8130	4.8099
7	600	45	0.10	1.4700	1.3720	1.3694
8	900	45	0.25	3.6200	3.5190	3.5189
9	600	15	0.25	1.6500	1.5520	1.5522
10	750	30	0.175	4.8100	4.8100	4.8099

From Table 3, it can be seen that the ANN results closely resemble that of experimental result. It is also observed from the Regression plot; which determines the strength and the relationship between the ANN proposed and experimental data fit very well with yield of 0.9999 of 1.0000. This is an indicator that the ANN method predicted the data well as shown in Figure 3.

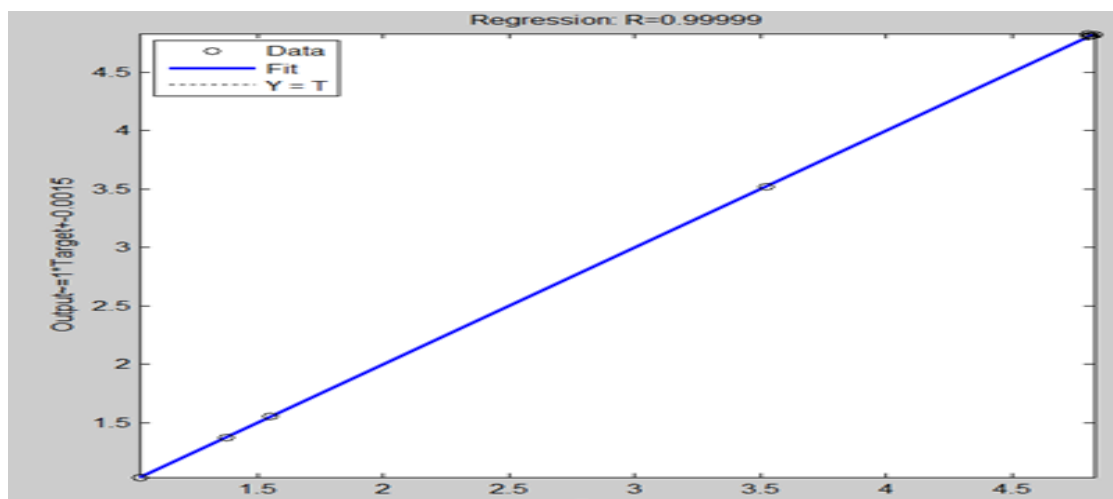


Fig. 3: regression line of Experimental and ANN model for adsorption capacity.

Table 4 further shows the statistical parameters, indicating that the ANN model produced a reliable estimation for optimal adsorption capacity based on input temperature, activation time and flow rate. The Coefficient of efficiency 1.000 indicated that the model estimation is same as the observed value in experimental. Correlation coefficient shows the relationship between the NN prediction and the value recorded for the experimental results. Correlation coefficient of 1 indicated a perfect match of the model. Meanwhile the mean absolute error of 0.0008 implies that the error is highly insignificant which signifies that a very high accuracy is achieved by the model.

Table 4: The best result obtained from ANN

Adsorption capacity	Statistical Parameters					
Adsorption capacity	Coeff. of efficiency	Corr Coeff.	RMSE	Mean Absolute error	MSE	NMSE
	1.0000	1.0000	0.0063	0.0008	0.00003	0.00001

It can be deduced from Figure 4 that the estimated adsorption capacity and actual adsorption capacity needed for treatment of wastewater are closely related, as both results overlapped. This means that without carrying out FFD test, the ANN model can be used to predict the optimum adsorption capacity needed in wastewater treatment for removal of phenol. This can also be used as an automated prediction of adsorption capacity system without human intervention for real online system operation. It is pertinent to note that for high temperature of 750°C and above, the value of adsorption capacity significantly increased, this implies reduced cost.

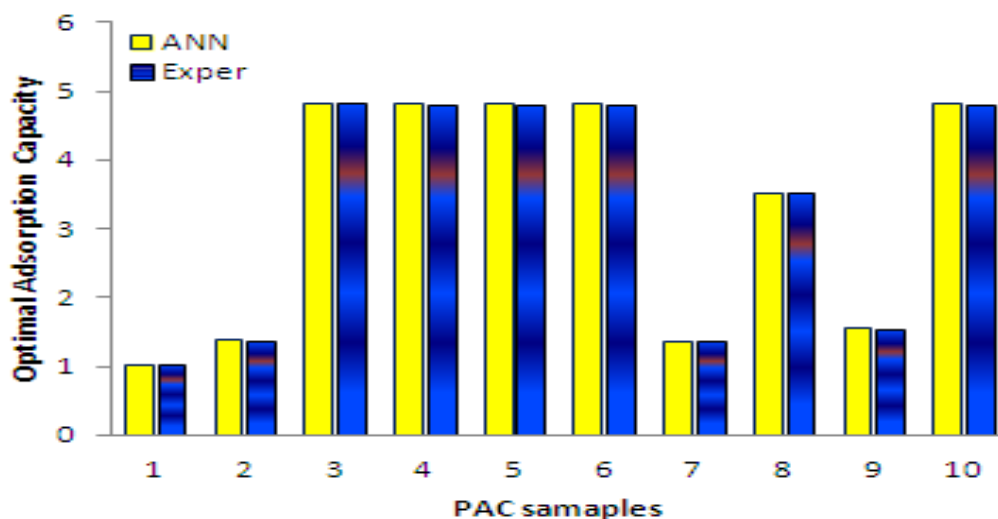


Fig. 4: Adsorption capacity for varying temperature, flow rate and time for different samples

Conclusion:

The ANN predictive model for optimal adsorption of aqueous phenol removal with Oil Palm EFB Activated Carbon can make the operation of wastewater treatment plant more effective and accurate. The insignificant error produced by ANN model indicated effective utilization of resources. The correlation between experimental and ANN predicted model is 0.999 of 1.00. This High Correlation of coefficient indicates that the ANN model is a perfect match. It is also pertinent to note that for average high temperature at either average low or average high time and flow rate, the adsorption capacity significantly increased which implied reduced in cost and resources. One of the advantages of the ANN prediction model is that there will be no need for FFD testing again, it also help in operational cost reduction, gives optimal adsorption values and ensuring quality of treatment process.

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