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Optimization and Modelling of the Removal of Groundwater Turbidity by Nanomagnetic Adsorbent Composite

A Huda¹, B Jayanthi¹, A L Zul Ariff¹, TYS Leony¹, M A Siti Nuurul Huda¹ and SA Palsan¹

¹ Faculty of Agro-Based Industry, Universiti Malaysia Kelantan, Jeli Campus Kelantan, Malaysia.

Email: hudaawang@gmail.com

Abstract. Groundwater pollution with turbidity problem is a matter of concern because at least 50 % of global population consume groundwater. The presence of suspended particles comprise of clay and silts as well as organic and inorganic particles is the main cause for the water to be turbid. A nanomagnetic adsorption composite (NMAC) was applied in this study to purify turbid polluted groundwater. A 3^k full factorial design was used to investigate five factors; dosage of adsorbent (0.02, 0.04, and 0.06 g), time of agitation (15, 30, and 60 min), rate of adsorption (150, 200, and 250 rpm), size of adsorbent (<45 µm and >300 µm), and initial concentration of sample (<21.3 and < 48.8 NTU). The optimum parameters were found to be 0.02 g, 249 rpm, 46 min, <45 µm and <21.3 NTU with 94.13 % turbidity removal efficiency. The turbidity of purified groundwater complies with the Drinking Water Quality Standard.

1. Introduction

Groundwater is a vital natural source of life and means of livelihood. According to the United Nations, about 2.5 billion of the world's population use groundwater as a source for drinking [1]. Nevertheless, pollution of the groundwater is caused by rapid and unsystematic population growth, as well as wasteful industrial and irrigation use. The risk of waterborne disease is becoming a threat to approximately 884 million people who consumed contaminated water [1]. Therefore, it indicates that suspended sediment in groundwater contributes significant implications for human health [2].

Turbidity is known as the cloudiness that changes the aesthetic of water. This problem always occurs in groundwater due to the presence of suspended particles comprise of clay and silts as well as organic and inorganic particles [3]. The turbidity might occur due to heavy rainfall, excessive extraction of groundwater, fissures within the aquifer, and faecal pollution of shallow domestic well [3][4]. So, the presence of turbidity groundwater is inevitable not only to the domestic consumption but also to the operation of the drinking water industry [3].



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The significant turbid water contains natural organic matter (NOM), and the amount of the NOM depends on the turbidity of water. Conventional water treatment uses chlorine to disinfect microbes in the water, excluding NOM. Thus, it remained NOM in the water might react with chlorine and form disinfection by-products (DBP) [5]. A study revealed that DBP is carcinogenic and causes bladder and colon cancer [6]. So, extensive research to remove turbidity in water is essential to reduce after-effect during water treatment [5].

One of the Sustainable Development Goals (SDGs) is the availability of safe and affordable sustainable management and sanitation for all. Also, based on the Drinking Water Quality Standard, the turbidity of drinking water must not exceed 5 NTU (Nephelometric Turbidity Unit) [7]. Traditional practice, such as boiling water that cannot remove dissolved toxins [8]. Besides, another tradition by rural people is to store water extracted from groundwater in a bottle; later, the impurities are usually removed (i.e. Larger contaminants appear to float or fall in the water). Often coagulant is added to improve the absorption of contaminants in the container, and then the stream is purified. However, the process requires several steps and resources to produce clean water [9].

Apart from low-cost, natural abundance of precursors, high porosity, and physiochemical stability in water, the powdered biochar faces a problem-separating the adsorbent from water [10]. Thus, an agriculture waste-derived nanomagnetic adsorption composite (NMAC) was synthesized by adding magnetic properties. In this study, an iron oxide nanomaterial was known as eco-friendly, low cost, and suitable adsorbent was applied to improve the performance of biocarbon. The separation of NMAC from water is aided by an external magnetic field. According to the previous study, the maximum adsorption capacity by NMAC to remove copper in aqueous solution is 113.63 mg/g [11]. Thus, a study to evaluate the performance of NMAC in removing turbidity in groundwater will be carried out in this study.

The aim of this analysis is to improve the removal efficiency of NMAC in the adsorption of turbidity from the actual sample (collected from well). The challenge, though, is the initial concentration of the sample is uncontrollable, except for two distinct turbidity ranges (high and low). This analysis is therefore performed to test multiple variables (adsorbent dosage, adsorbent size, agitation speed, agitation length, and initial turbidity) for optimal formulations for high turbidity in removal.

2. Materials and methods

2.1 Preparation and characterization of the adsorbents.

NMAC was obtained from Wannahari et al., [11] in this analysis. Then, the adsorbent was cleaned, neutralized, dried and sieved (range 45-300 nm).

2.2 Water sampling and preparation.

The water sample was collected from a well in Tanah Merah of Kelantan with the coordinate N 5 4856.8 E 102 ° 0757.1. In order to have a different range of turbidity (i.e. high turbidity and low turbidity), a low turbidity water sample was left for 24 hours to ensure the sedimentation of large impurities [12]. In the meantime, the high turbidity water sample was used fresh sample which is applied directly after collection. The initial turbidity of this raw water is tested (< 21.3 NTU) old sample and (<48.8 NTU) fresh sample.

2.3 Experimental design.

In this analysis, the experiment conducted a three-level full factorial design using Design Expert v.11 software. There are five factors involve in the experiment: the dosage of adsorbent, speed of agitation, time of agitation, size of adsorbent, and initial concentration of water sample. The responses of the combinations were evaluated and generated a second-order polynomial equation (Eq.1):

$$y = \beta_0 + \sum_{i=1}^k \beta_{ij}x_i + \sum_{i=1}^k \beta_{ij}x_i^2 + \sum_{i<j=2}^k \beta_{ij}x_ix_j + \varepsilon \quad (1)$$

Where y is the response, k is the number of factors, β_0 is constant, β_i is the i th linear coefficient, β_{ij} is the i th quadratic coefficient, β_{ij} is i th interaction coefficient, x_i is the independent variable, and ε is the error.

The design scheme of the experiments is shown in Table 1, where -1 is the lowest level, 0 is middle, and 1 is the highest level.

Table 1. The design scheme for three-level full factorial design.

Types of factors	Factors	Symbol	Unit	Levels		
				-1	0	1
Numerical	Dosage of adsorbent	g	A	0.02	0.04	0.06
	Time of agitation	min	B	15	30	60
	Rate of agitation	rpm	C	150	200	250
Categorical	Size of adsorbent	μm	D	< 45		>300
	Initial concentration of water sample	NTU	E	<21.30		>48.80

2.4 Batch adsorption studies

Batch adsorption was carried out with 10 % adsorbent in the working volume. Each batch was run for adsorption according to experimental design. The turbidity removal efficiency was calculated as in Eq.2:

$$\% \text{ Removal efficiency} = \frac{C_i - C_e}{C_i} * 100 \quad (2)$$

Where C_i is initial, and C_e is equilibrium concentration (NTU). The measurement of turbidity was performed by using turbidity meter with fast tracker (Hanna Instruments, Romania).

3. Results and discussions

3.1 Development of model.

Analysis of responses for the combination of parameters (Table 1) generates an equation. Thus, based on the given output, a quadratic model is suggested by considering R-squared (R^2) is 0.9904, adjusted R-squared is 0.9887, predicted R-squared is 0.9865 and predicted sum of squares (PRESS) is 17.32. Lack of fit test was carried out to determine the variation for the model inadequacy, so, the quadratic model is acceptable as the lack of fit for quadratic model (Table 2) is not significant (>0.05). The output of the analysis generated through Analysis of Variance (ANOVA) proposed quadratic model as in Equation 3:

$$\begin{aligned} \text{Turbidity Removal Efficiency [Y,\%]} = & 96.54 - 1.34 A + 2.02 B + 0.32C + 1.46 D + 0.98E + 0.44A^2 - \\ & 1.00B^2 - 3.08 C^2 + 0.50AB + 0.66AC + 0.19AD + 0.60AE + \\ & 0.71BC - 0.19BD - 0.21BE + 0.66CD - 1.27CE + 0.100DE \end{aligned} \quad (3)$$

Where, the positive sign indicated for synergistic effect while negative sign is for antagonistic effect.

Figure 1 (a) shows normal probability plot for the studentized residual for turbidity removal efficiency by NMAC. The points fall along a straight line indicates that the data is normally distributed. Even though there some points scattered along the line, it is considered acceptable for normal distribution data [13].

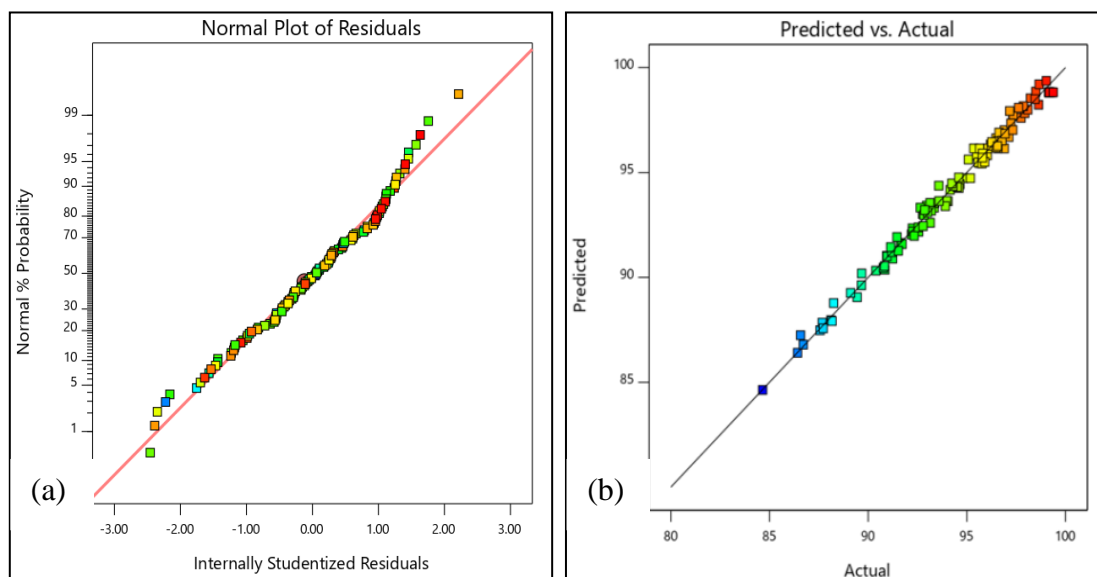


Figure 1. (a) Normal probability plots of residuals for turbidity removal efficiency by NMAC and (b) predicted turbidity removal efficiency versus the actual turbidity removal efficiency by NMAC.

Figure 1 (b) shows the plot of “Predicted vs Actual” to determine the reliability of the response for turbidity removal efficiency. The comparison between the plots for generated predicted response and actual response reveal that the quadratic model equation is accurate and reliable. These results are agreeable with earlier observations by Yu [14].

3.2 Analysis of the effects of parameters on turbidity removal efficiency.

Table 2 presents the ANOVA regression parameters for predicted quadratic surface response models of turbidity removal efficiency. Based on Table 2, all of the parameters (dosage of adsorbent, speed of agitation, time of agitation, size of adsorbents, and initial concentration of water sample), as well as, interactions between parameters are significant at a 5 % confidence level (p-value <0.05).

Table 2. Analysis of Variance (ANOVA) for turbidity removal efficiency by NMAC.

Source	Sum of Squares	DF	Mean Square	F Value	Prob > F
Model	1273.841	18	70.76894	579.0597	< 0.0001
A	113.1099	1	113.1099	925.5105	< 0.0001
B	275.5059	1	275.5059	2254.299	< 0.0001
C	6.967566	1	6.967566	57.01140	< 0.0001
D	241.4162	1	241.4162	1975.363	< 0.0001
E	108.3832	1	108.3832	886.8346	< 0.0001
A2	5.162859	1	5.162859	42.24457	< 0.0001
B2	26.05436	1	26.05436	213.1871	< 0.0001
C2	188.5878	1	188.5878	1543.100	< 0.0001
AB	11.09944	1	11.09944	90.82008	< 0.0001
AC	20.24599	1	20.24599	165.6608	< 0.0001
AD	2.372445	1	2.372445	19.41229	< 0.0001
AE	22.91633	1	22.91633	187.5106	< 0.0001
BC	24.40326	1	24.40326	199.6772	< 0.0001
BD	2.445216	1	2.445216	20.00773	< 0.0001
BE	3.109403	1	3.109403	25.44237	< 0.0001
CD	30.60861	1	30.60861	250.4518	< 0.0001
CE	112.6868	1	112.6868	922.0486	< 0.0001
DE	1.17241	1	1.172410	9.593129	0.0025
Residual	12.34357	101	0.122214		
Lack of Fit	10.43407	82	0.127245	1.266118	0.2877
Pure Error	1.909498	19	0.100500		
Cor Total	1286.184	119			

The effects of dosage of adsorbent and the speed of agitation are shown in Figure 2 (a). The significant positive quadratic effect ($p < 0.05$) speed of agitation and significant negative linear effect ($p < 0.05$) dosage of adsorbent have resulted in a curvilinear increment of turbidity removal efficiency. The positive effect for speed of agitation might occur because of the collision of the NMAC (adsorbent) increase rate of adsorption [15]. However, the number of active sites is higher at lower dosage concentration [16]. Thus, it shows that optimum synergistic interaction for the rate of agitation is 200 rpm and dosage of adsorbent is 0.02 g.

The effect of dosage of adsorbent and time is demonstrated in Figure 2(b). Based on the obtained result, the significant positive quadratic effect ($p < 0.05$) time of agitation and significant linear negative effect ($p < 0.05$) dosage of adsorbent are resulting curvilinear of turbidity removal efficiency for all agitation time. The decrease of adsorption as the dosage of adsorbent increase is likely due to overlapping or aggregation of the active sites, later resulting in a net reduction of total surface area available for contaminant [17].

Besides that, there are ten solutions to solve by Eq.3. Hence, one answer was selected with the desirability of 1.00 that provided dosage of adsorbent, rate of agitation, time of agitation, size of adsorbent, and initial concentration of water sample (turbidity) to be 0.02g, 249 rpm, 46 min, $< 45 \mu\text{m}$

and <21.3 NTU. The predicted turbidity removal efficiency is 94.84 % (1.25 NTU), with 0.75 % higher than the actual. Besides, the turbidity of groundwater after purification comply Drinking Water Quality Standard (>5NTU).

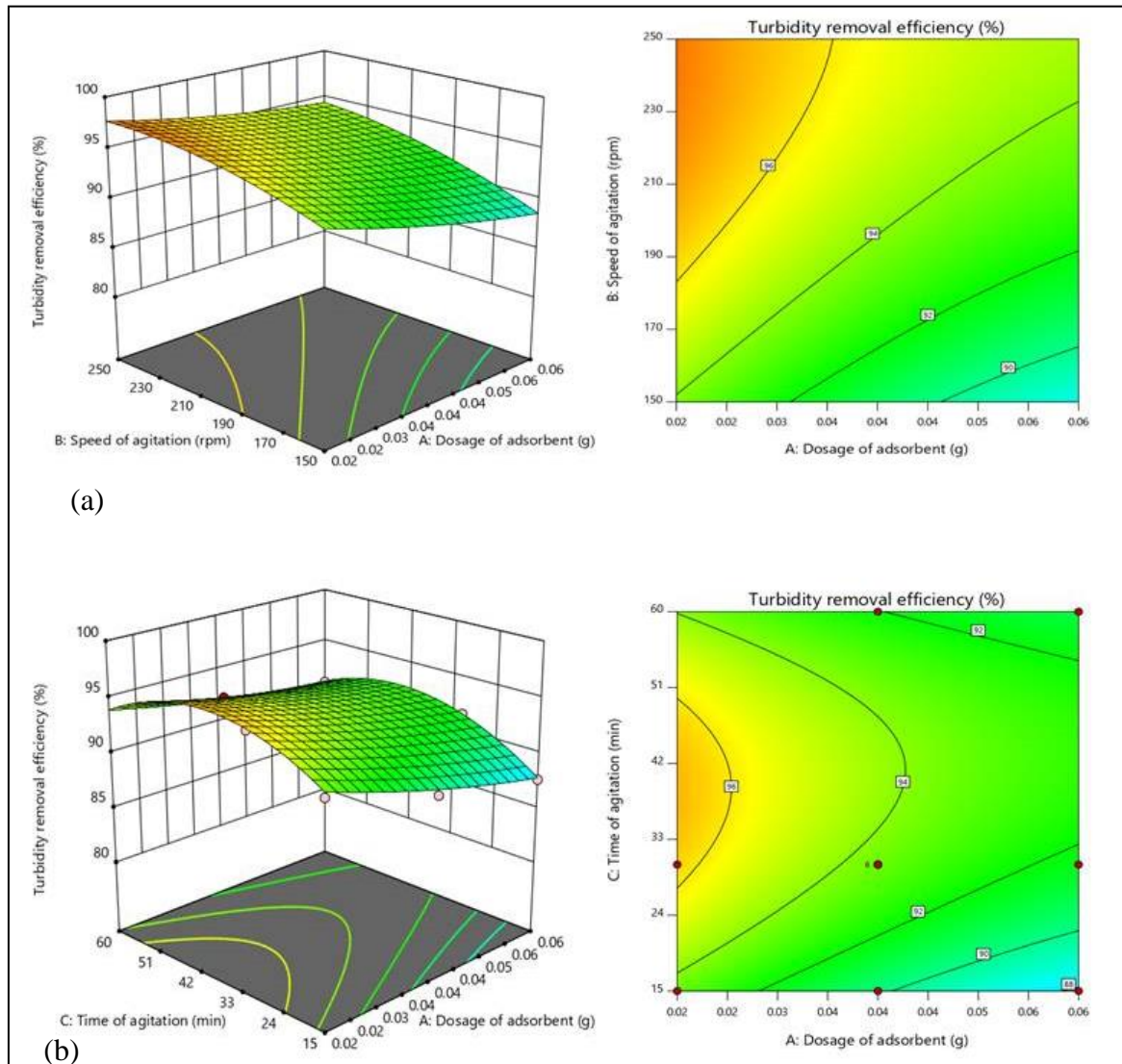


Figure 2. Contour plot showing the effects of variables for (a) speed of agitation and dosage of adsorbent and (b) time of agitation and dosage of adsorbent for turbidity removal efficiency.

4. Conclusion

The study demonstrated that the optimum parameter for turbidity removal efficiency is 0.02 g (dosage of adsorbent), 249 rpm (rate of agitation), 46 min (time of agitation), <45 μ m (size of adsorbent) and 21.3 NTU (initial concentration of sample) with 94.13 % efficiency. The study revealed that the individual parameters are significant for obtaining optimum adsorption. Thus, NMAC is a potential adsorbent for drinking water purification as the purified groundwater comply with the Drinking Water Quality Standard.

Acknowledgement

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