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Research Article

Process Optimization of Zinc Removal Using Microwave Incinerated Sugarcane Bagasse Ash (MISCBA) Through Response Surface Methodology

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Abstract: Microwave Incinerated Sugarcane Bagasse Ash (MISCBA) was used in the exclusion of zinc from aqueous solution. Parameters of importance like initial metal concentration, adsorbent dosage and agitation time were examined to find their effect on the adsorption process. Respond surface methodology has been employed to enhance the process conditions based on Box-Behnken design. Response surface method suggest initial metal concentration of 50 mg/L, adsorbent dosage 1.0 g and contact time of 3 h with removal efficiency of 55.99% to be optimum conditions for zinc removal from aqueous solution. A high correlation coefficient of 0.9923 indicates the model is in agreement with the experimental values. The model indicates that adsorbent dosage is the major influencing factor among others responsible for the adsorption of zinc.

Keywords: Anova, adsorbent, adsorption, correlation coefficient, design expert, design matrix, metal concentration, press

INTRODUCTION

Industrialization is a key factor in measuring the growth and development of any Nation. It increases gross development product, upgrade social activities and create job opportunities to the country. However, with the aforementioned advantages of industrialization, it has some major set-back when related to Environmental Impact Assessment (EIA), more especially in deteriorating the natural environment.

Industries such as petrochemical, tannery, electroplating, pulp and paper etc. are known of releasing wastewater laden with toxic pollutants directly into the environment, with or without proper treatments. Heavy metals are among the major pollutants found in wastewater, this because of their known toxic effects on human and aquatic life, it tend to accumulate in human body over a period of time causing sudden death or other diseases such as diarrhea, vomiting, nausea, breathing problem, eye burns and gastritis (ALzaydien, 2009; Rafatullah *et al*., 2009).

In view of this, strict laws has been enacted and standards were set-up by regulatory authorities to control and monitor wastewater discharge by these industries are conform with the established limits of wastewater quality before they can be release to the receiving environment (Lim *et al*., 2002).

Conventional methods for removal of heavy metals in wastewater have been developed and being practiced for decades, such treatment methods includes physical and chemical processes such as chemical precipitation, ion-exchange, oxidation, electrodialysis, coagulation, sedimentation and flocculation (Kutty *et al*., 2012). These methods were found to have some limitations that render them not applicable so often, they require initial capital and operational cost, skilled man power, energy, creation of excess toxic sludge that requires special disposal method and above all does not removes the heavy metals completely especially at lower concentration (1-100 mg/L) (Li *et al*., 2013).

Adsorption process was found to be the most effective method for removal of heavy metals in wastewater owing to its inattentiveness to toxic constituents, easy to design and operation (Azargohar and Dalai, 2005). Commercial activated carbon is the most frequently and efficiently used adsorbent for adsorption process, but the finer it is the more expensive it becomes and also have high regeneration cost, this has limited its application in industries (Chatterjee *et al*., 2012).

The search for an alternative low cost adsorbent that is effective and efficient has become paramount important. Several researchers have look into agricultural by-products for the production of activated carbon, this includes, corncobs (Leyva-Ramos *et al*.,

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2005), sawdust (Baral *et al*., 2006), rice husk (Kutty *et al*., 2012), wheat bran (Bulut and Baysal, 2006) and peanut shell (Wilson *et al*., 2006).

In this study, Microwave Incinerated Sugarcane Bagasse Ash (MISCBA), produced from thermal treatment of sugarcane bagasse was used as a low cost adsorbent for removal of zinc from aqueous solution and its process optimization using response surface method.

MATERIALS AND METHODS

Materials and equipment's: Sugar-cane bagasse was obtained from a road side juice maker in Botah town, Malaysia. The bagasse was initially cut to an average size of 10 cm and washed with tap water to remove dirt's and lignin impurities, it was then washed several times with distilled and oven dried for 24 h at 105°C, the dried bagasse was grinded to a size of 6 mm using a laboratory grinder before it was incinerated using an industrial scale microwave incinerator at 500°C for 3 h. The resulting microwave incinerated sugarcane bagasse ash (MISCBA) was soaked in a weak acid (5% Nitric acid, pH 4.15) overnight; the mixture was filtered and oven dried at 70°C for 3 h. The final material was stored in a desiccator before its application.

Experimentation: 1000 mg/L stock solution of zinc was prepared by dissolving its equivalent salt in 1 L of distilled water. Further required concentrations were made by diluting the stock solution using distilled water. 17 experiments were carried out in a batch system using varied time, adsorbent dosage and initial metal concentration. 250 mL conical flasks filled with 100 mL of aqueous solution and added with varied dosage of MISCBA are agitated in an orbital shaker operating at 150 rpm and are withdrawn at the design time intervals for each run. Adjustment of pH was done using 1.5N NaoH and 2N HCl and Solution pH was measured using pH meter (Model EW 53013, Hach). At the end of each run, content of the flask are filtered with 0.45 µm filter paper and the residual zinc content was analyzed using Atomic Adsorption Spectrophotometer (AAS). Removal Efficiency of zinc was calculated using:

$$\mathrm{R} = \frac{(Ci-Ce)}{Ci} \times 100\%\mathrm{mg/L} \qquad (1)$$

where, *Ci* and *Ce* are zinc concentration before and after adsorption

Design of experiment: Response Surface Methodology (RSM) is an important process mainly employed for design and process optimization of adsorption experiments (Kicsi *et al*., 2010). It eases to understand the various interactions between the parameters under study under less number of experimental runs. Metal concentration (K_1), adsorbent dosage (K_2) and contact time (K_3) are selected to be independent variables and

Table 1: Independent factors and their heights used for Box–Behnken design matrix

		Heights and range		
Factors	Notation	-1	0	1
Metal concentration (mg/L)	K_1	10	50	100
Adsorbent dosage (g)	K_2	0.5	1.0	1.5
Time (h)	K_3	1.0	3.0	9.0

their level of variations displayed in Table 1. The response factor removal efficiency Y (%) was determined by running experiment of the possible combinations of the actual design variables suggested by RSM shown in Table 2.

Coding of the selected independent variables was in accordance with Equation 2 below:

$$\frac{(k_i-k_0)}{\Delta K} \qquad (2)$$

where, K_i is the ith dimensionless oblique significance for free variable, K_0 is K_i value at middle point and ΔK stands the value at phase adjustment. Removal efficiency (R %) is the only response variable in the experimental design. Metal concentration was varied from 10 to 100 mg/L, contact time was varied from 1 to 9 hours and adsorbent dosage was varied from 0.5 to 1.5 g. As established by the software a total number of 17 experimentations were performed, consisting of six replicates at central points and twelve factorial points. Results of the experiment acquired from the Box-Behnken model were defined in the form of Eq. (3) below:

$$R = \beta_o + \sum_{i}^{n} \beta iki + \sum_{i}^{n} \beta i_1 ki^2 + \sum_{i<j} \beta ijkikj \qquad (3)$$

where,
R = The anticipated outcome
β_o = Central point fixed response value of the experiment
β_i = The undeviating quantities
β_{ii} = The quadratic quantities as well as
B_{ij} = Stands for cross (interaction) quantities

Design expert software (version 6.0.6) from stat-Ease Inc., USA has been employed for the analyses of variance (ANOVA) and the response surface as well. Numerical and graphical tools of the software were utilized for the response (removal efficiency) optimization.

RESULTS AND DISCUSSION

Analysis and interpretation of response values: Removal efficiency (R %) as a response factor of the design experiment obtained under dissimilar investigational settings created by Design Expert software are shown in Table 2.

Table 2: Three numeric factors Box-Behnken design matrix with experimental response for removal efficiency

Run	Metal concentration (mg/L)	Adsorbent dosage (g)	Time (h)	Removal efficiency, R (%)
1	100	1.0	9.0	47.0
2	10	1.5	3.0	60.0
3	50	1.5	9.0	56.0
4	50	1.0	3.0	51.4
5	50	1.0	3.0	51.4
6	50	1.0	3.0	51.4
7	100	0.5	3.0	29.6
8	10	1.0	9.0	63.0
9	100	1.5	3.0	42.9
10	10	0.5	3.0	52.0
11	50	1.0	3.0	51.4
12	50	0.5	1.0	35.0
13	50	1.0	3.0	51.4
14	10	1.0	1.0	54.0
15	10	1.0	1.0	54.0
16	50	1.5	1.0	49.8
17	50	0.5	9.0	49.0

From Table 2, it can be observed that each experimental condition has its distinct response outline in terms of zinc removal efficiency R%, percentage of zinc removal ranges from 35 to 60% in response under different permutations of operative factors as suggested by the software.

Lack of fit test: Removal efficiency R% as a response factor was subjected to lack of fit test in a way to match its residual error with pure error from the replicated design points. The outcome of the test is shown in Table 3.

Both linear and 2FI models have been lined out, for the reason that there prob>F drops below 0.05 (Montgomery, 2008). The quadratic model, as previously suggested by the software was found to be the likely model for this experimental response, having shown no substantial lack of fit. The cubic model was initially aliased by the software and for that was not considered. Furthermore, model summary statistic in Table 4 was explored to confirm the reliability of the chosen model and also for model maximization, it has further indicated that the quadratic model is the best model for the response analysis; this is because it shows low Standard Deviation (SD), low press and high R^2 values (Montgomery, 2008). A projected R-squared of 0.9069 stands in sound agreement in relation to the Adjusted R-squared of 0.9929 (Montgomery, 2008).

ANOVA study for the quadratic model: To assess suitability of the selected model and the significances of the variable factors, ANOVA was employed. It simply classifies and cross- classifies statistical outcomes and tested using identified classification variance, which was approved by Fisher's statistical test (F-test). The proportion of mean square of regression (MRR) to error (MRe) defined the F-value. The worthiness of the corresponding coefficient is related to lower value of the F-value. Table 5 present the ANOVA quadratic model that proves the strength of the model.

Adsorbent dosage indicates to be the most significant factor among other variables having an F-value of 167.77 and Prob>F is less than 0.05, the model F-value of 248.61 having Prob>F of 0.0001 entails the significances of the model. Values of P > F less than 0.05 indicates model terms are significant (Montgomery, 2008), in this case K_1, K_2, K_3, K_2^2, K_3^2, K_1K_2 and K_2K_3 remain momentous. Values larger than 0.1 indicates terms in the model not momentous, in this

Table 3: Summary of lack of fit tests for different models

Source	Sum of squares	Degree of freedom	Mean square	F value	Prob>F
Linear	930.49	3	310.16	33.08	<0.0001
2FI	95.72	3	31.910	68.05	<0.0001
Quadratic	22.89	3	7.630	0.770	0.5363
Cubic	3.280	2	1.640	6.366E+007	<0.0001

Table 4: Model summary statics for different models

Source	S.D	R^2	Adjusted R-squared	Predicted R^2	PRESS
Linear	3.06	0.8842	0.8574	0.7978	212.81
2FI	3.35	0.9059	0.8495	0.6423	376.44
Quadratic	0.68	0.9969	0.9929	0.9069	98.010
Cubic	0.00	1.0000	1.0000	-	-

Table 5: Analysis of variance (ANOVA) for response surface quadratic model for zinc removal

Source	Sum of squares	Degree of freedom	Mean square	F value	Prob>F
Model	1049.10	9	116.57	248.61	<0.0001
K_1	50.200	1	50.200	107.07	<0.0001
K_2	78.660	1	78.660	167.77	<0.0001
K_3	64.090	1	64.090	136.69	<0.0001
K_1^2	1.5900	1	1.5900	3.4000	0.1078
K_2^2	47.17	1	47.170	100.60	< 0.0001
K_3^2	28.09	1	28.090	59.910	0.00010
K_1K_2	6.420	1	6.4200	13.700	0.00760
K_1K_3	4.170	1	4.1700	8.8900	0.02050
K_2K_3	12.61	1	12.610	26.890	0.00130
Residual	3.280	7	0.4700	-	-

case K_1^2 is not significant. Insignificant parameters were removed so as to improve the effect of the momentous terms. The model equation in relation to its coded factors is shown in Eq. (4):

$$R = 26.32–0.26K_1+42.23K_2+4.31K_3–15.53K_2^2–0.22K_3^2+0.056K_1K_2–0.84K_2K_3 \quad (4)$$

The correlation coefficient R^2 for Eq. (4) is 0.9372 almost close to unity, this exhibits that there is an excellent settlement amid the predicted and the experimental removal efficiency and also did not display any non-linear pattern (S-shaped curve) which demonstrating error terms abnormality and can only be modified by alteration (Körbahti and Rauf, 2009). There is no emblem of any problem in this numbers as depicted in Fig. 1 and also indicates that 93.72% of overall variation in the efficiency of the removal was credited to the variable factors studied. It is also clear that experimental removal efficiency follows the predicted outcome with a good accuracy as shown in Fig. 2.

Model validation was an essential portion in the process of data analysis, as a poor model could end up to ambiguous results (Körbahti and Rauf, 2009). Appropriate accuracy quantifies the signal to noise

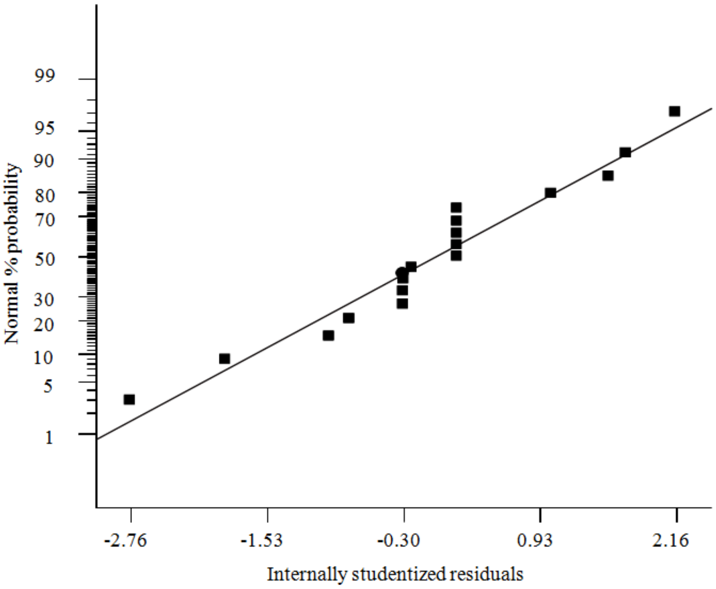

Fig. 1: Normal % probability against internally Studentized residuals

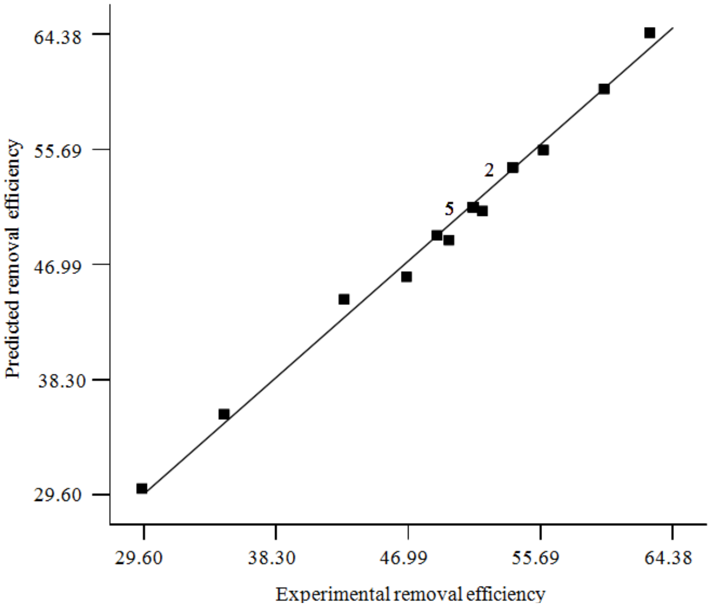

Fig. 2: Model comparison between predicted and experimental efficiency

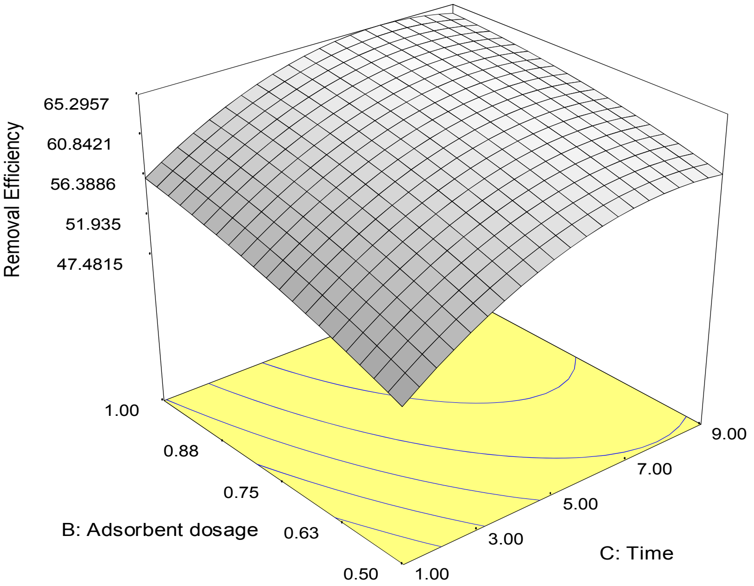

Fig. 3: Surface plot for zinc removal efficiency against Time (hrs) and adsorbent dosage (g)

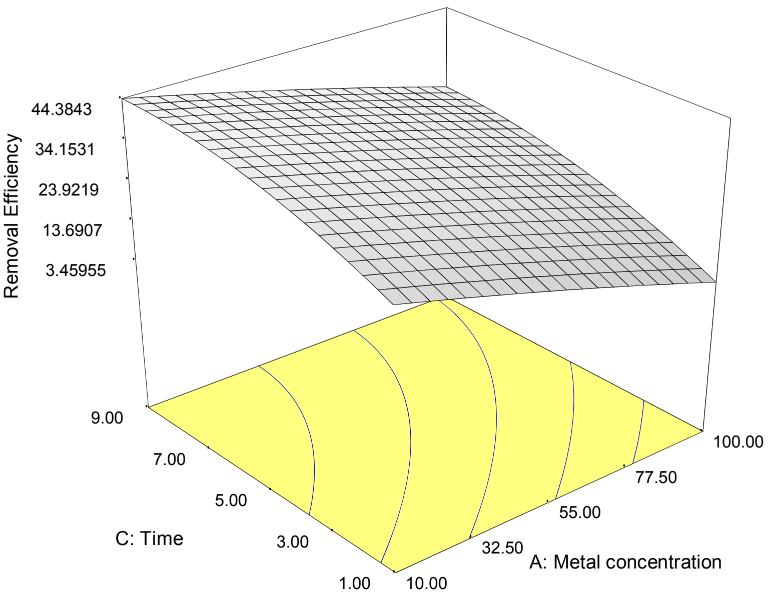

Fig. 4: Surface plot for zinc removal efficiency against metal concentration (mg/L) and contact time (hrs)

ratio. As a requirement a ration higher than 4 is acceptable. A ratio of 52.855 indicates an adequate signal. The chosen model can be used to navigate the design space.

Surface plots for response: For detail analysis and understanding of the relations, it has been recommended that three-dimensional views for regression model should be employed (Aktaş, 2005). The experimental design system behavior can be understood better with the use of three-dimensional surface plots which offers vital information within the experiment, furthermore, the plots will help to aid in inspecting the influence of variable factors considered in the experimentation on the responses and the contour between the variable factors (Ahmad and Hameed, 2010; Panesar, 2008).

Effects of the interactive variables, adsorbent dosage, initial zinc concentration and contact time on the removal of zinc from aqueous solution was determine using response surface plot (Fig. 3 to 5). Figure 3 indicates that for a fixed zinc concentration, as adsorbent dosage increases, zinc removal efficiency also increases from 47.5 to 56.4%, this can be linked to the availability of more surface area due to an increase in adsorbent concentration, this increment in the removal efficiency is directly related to the contact time allowed between

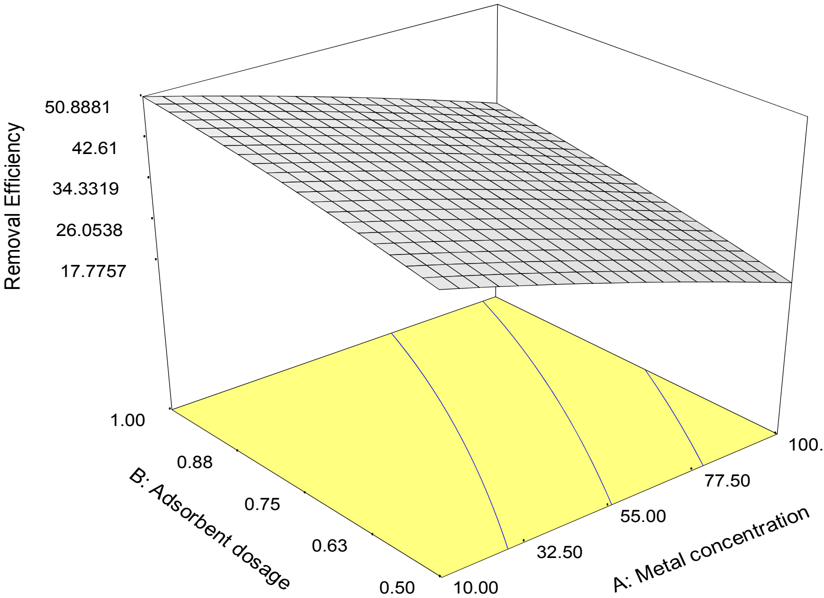

Fig. 5: Response surface plot for zinc removal efficiency against metal concentration (mg/L) and adsorbent dosage (g)

Table 6: Optimum conditions for zinc removal efficiency using model validation

Metal concentration (mg/L)	Adsorbent dosage (g)	Contact time (hours)	Removal efficiency (%) Predicted	Removal efficiency (%) Experimental	Desirability
50	1	3	56.34	55.99	0.316

the two variables (dosage and time), it reaches a maximum value at about 7 hours with corresponding removal efficiency of 65.3% and continue in an equilibrium condition with no further significant removal being noticed.

Figure 4 shows the shared effect of initial metal concentration and contact time at fixed adsorbent dosage on the removal efficiency of zinc. It shows that as metal concentration increases, removal efficiency decreases, this is because the mass transport resistance between solution and adsorbent is mainly control by the concentration of the pollutant presence in the solution, at lower concentration the movement resistance tends to be less compared to that at a higher concentration level (Rafatullah *et al*., 2009). It also shows that as the time increases the efficiency of removal increases from 3.5 to 44.4, it can be observed that removal efficiency was noticed even at the initial time and kept on increasing until it reaches a point where further removal becomes insignificant, the increase at initial time is due to the presence of surface area of the adsorbent but as the time increase the adsorbent surface tends to worn out and at this stage the removal is measured by the rate of movement from the outer to the inner side of adsorbent material (Rafatullah *et al*., 2010).

Figure 5 also shows the relationship between initial metal concentration and adsorbent dosage on the removal efficiency of zinc. It indicate that removal efficiency decreases with an increase of initial metal concentration from 17.8 to 50.9 mg/L. RSM result shows that the interactive sign between the two variables (K_1K_2) is positive and which indicates that they are significant in the removal efficiency but the first order effect of metals concentration (K_1) shows a negative response which suggest that metal concentration on its own have no contribution on the removal efficiency. Decrease in removal efficiency at higher concentration might be related to the competitive nature of the metal ion for a fewer available sites of the adsorbent and also due to binding sites saturation (Kiran and Thanasekaran, 2011).

The main objective is to optimize zinc removal efficiency under varied adsorbent dosage, time and initial metal concentration, optimum removal efficiency were recognized using Design Expert software. It suggest the optimum conditions to be metal concentration 50 mg/L, adsorbent dosage 1.0 g and contact time of 3 hours which gives a response value of 55.99% in terms of removal efficiency (Table 6).

CONCLUSION

The study was carried out to investigate the effect of initial metal concentration, adsorbent dosage and contact time on the removal efficiency of zinc from aqueous solution and also to optimize the process situations employing response surface method. The experimental data obtained in agreement with the suggested quadratic model, having correlation coefficients of 0.9969. Adsorbent dosage as variable factor is the most significant, having a positive coefficient value of 42.23 in the model equation, compared to other variables that have less coefficient values. According to "Design-Expert" software, optimum conditions for zinc removal efficiency were found to be a zinc concentration of 1 mg/L, adsorbent dosage of 1 g and contact time of 0.99 h, with the zinc removal efficiency of 55.99%.

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