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Response Surface Methodology Optimization of Multi-Metal Adsorption onto Fika Bentonite Clay Using Box-Behnken Design

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ABSTRACT

This study employed response surface methodology (RSM) with a Box-Behnken design (BBD) to optimize the adsorption of five heavy metals (Cu, Pb, Fe, Co, As) onto Nigerian Fika bentonite clay. Twenty-nine experimental runs were conducted to evaluate the effects of four independent factors: pH (2-12), adsorbent dosage (0.1-0.5 g/L), initial metal concentration (10-50 mg/L), and contact time (20-120 min). Quadratic response surface models were developed for each metal using Design Expert software (version 6.0). Analysis of variance (ANOVA) revealed that all models were statistically significant ($p < 0.05$). For copper, the model F -value was 7.35 ($p = 0.0003$) with $R^2 = 0.8803$. Significant model terms included adsorbent dosage (B), initial concentration (C), the quadratic term B^2 , and interactions BC (dosage $\times$ concentration) and BD (dosage $\times$ time). For lead, the model was highly significant ($F = 11.72$, $p < 0.0001$, $R^2 = 0.9214$). Iron, cobalt, and arsenic models were also highly significant ($F = 43.40$, 43.37 , and 43.41 respectively; $R^2 = 0.9775$, 0.9775 , and 0.9775 respectively). The interaction between adsorbent dosage and initial concentration (BC) was significant for all five metals, confirming the importance of optimizing these parameters simultaneously. The global optimum conditions identified were pH 7, adsorbent dosage 0.30 g/L, initial concentration 30 mg/L, and contact

time 70 min. Confirmatory experiments at these conditions gave adsorption capacities of 1.18 mg/g (Cu), 1.15 mg/g (Pb), 1.19 mg/g (Fe), 1.21 mg/g (Co), and 0.61 mg/g (As), all within 95% prediction intervals. The adequate precision ratios ranged from 11.99 to 26.83, all above the threshold of 4, indicating that the models can be used to navigate the design space. These results demonstrate that Fika bentonite, particularly in its raw form, is a promising cost-effective adsorbent for multi-metal wastewater treatment.

Keywords: Box-Behnken design, response surface methodology, heavy metal adsorption, Fika bentonite, process optimization, multi-metal system

INTRODUCTION

Water contamination by heavy metals (Cu, Pb, Fe, Co, As) poses severe environmental and public health risks, particularly in developing nations where affordable treatment solutions are scarce (Kumar *et al.*, 2022; World Health Organization [WHO], 2023). These heavy metals are non-biodegradable and tend to accumulate in living organisms, leading to severe health effects (Adebayo *et al.*, 2022). Conventional treatment techniques including chemical precipitation, ion exchange, and membrane filtration are frequently expensive and energy-intensive, producing secondary pollution (Salman *et al.*, 2021).

Response Surface Methodology (RSM) is a collection of statistical and mathematical techniques used for modeling, analyzing, and optimizing processes in which a response of interest is influenced by several variables (David *et al.*, 2023). Among the various RSM designs, the Central Composite Design (CCD) and the Box-Behnken Design (BBD) are the most widely used. The BBD was selected for this study because it reduces the number of runs, avoids extreme factor combinations, and allows efficient estimation of quadratic coefficients (Adebayo, 2024).

This study aims to optimize the adsorption of five heavy metals (Cu, Pb, Fe, Co, As) onto Nigerian Fika bentonite clay using Box-Behnken design, evaluate the effects of operational parameters, and validate the optimized conditions through confirmatory experiments.

MATERIALS AND METHODS

Materials and Reagents

All chemicals used were of analytical grade, including nitric acid (HNO_3 , 70%, Sigma-Aldrich), sulfuric acid (H_2SO_4 , 98%, Merck), sodium hydroxide (NaOH , 98%, BDH Chemicals), and potassium bromide (KBr , 99%). Fika bentonite clay was collected from Fika Local Government Area, Yobe State, Nigeria, and purified as described in our previous work.

Batch Adsorption Experiments

Batch adsorption experiments were conducted using 250 mL Erlenmeyer flasks containing 100 mL of heavy metal solution at known concentrations. Purified bentonite clay was added, and experiments were conducted under varying conditions: contact times (20-120 min), adsorbent dosages (0.1-0.5 g/L), initial metal concentrations (10-50 mg/L), and constant temperature of 25°C. Mixtures were agitated at 200 rpm using an orbital shaker. After designated contact time, mixtures were filtered, and filtrates were analyzed for remaining heavy metal concentration using Atomic Absorption Spectrophotometry (AAS) (APHA, 2022).

Experimental Design

A Box-Behnken response surface design with 29 experimental runs was employed to optimize heavy metal adsorption. Four factors were selected based on preliminary experiments: pH (2-12), adsorbent dosage (0.1-0.5 g/L), initial concentration (10-50 mg/L), and contact time (20-120 min) (Asiagwu *et al.*, 2012; Debora *et al.*, 2016; Gadd, 2009). The response variable was percentage removal efficiency for each heavy metal.

Table 1: Factors Used in Box-Behnken Design

Factor	Name	Units	Type	Low Actual	High Actual	Low Coded	High Coded
A	Ph	-	Numeric	2	12	-1	1
B	Adsorbent Dosage	g/L	Numeric	0.1	0.5	-1	1
C	Initial Concentration	mg/L	Numeric	10	50	-1	1
D	Contact time	Minutes	Numeric	20	120	-1	1

Optimization and Confirmatory Experiments

Optimization was carried out using Design Expert software (version 6.0) using a desirability function approach. Confirmatory experiments were conducted in triplicate at the predicted optimum conditions following the same adsorption procedure described previously (Mohammed *et al.*, 2024).

RESULTS AND DISCUSSION

Statistical Modeling Analysis

Copper (Cu) ANOVA

Copper Diagnostic plot

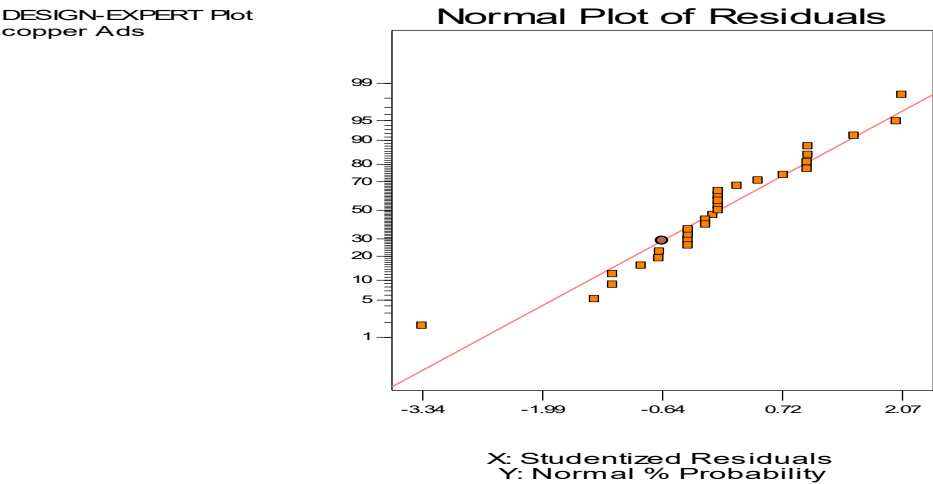


Figure 1: Copper Diagnostic plot

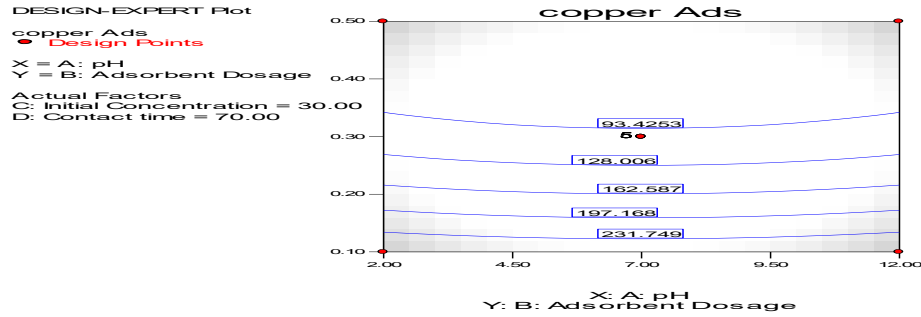


Figure 2: Copper (Cu) Contour plot

The ANOVA for copper revealed that the quadratic model was significant ($F = 7.35$, $p = 0.0003$). The model sum of squares was 278,778 with 14 degrees of freedom, resulting in a mean square of 19,912.71. The residual sum of squares was 37,914.07 with 14 degrees of freedom, giving a mean square of 2,708.15.

The main effect of adsorbent dosage (B) was highly significant ($p < 0.0001$), as was the main effect of initial concentration (C) ($p < 0.0001$). The quadratic term of dosage (B^2) was significant ($p = 0.0137$). The two-factor interactions BC (dosage $\times$ concentration) and BD (dosage $\times$ contact time) were significant with p-values of 0.0082 and 0.0212, respectively. All other terms pH (A), contact time (D), quadratic terms of pH, concentration and time, and interactions AB, AC, AD, and CD were not significant ($p > 0.05$).

The R-squared value was 0.8803, meaning that 88% of the variation in copper adsorption is explained by the model. The adjusted R-squared was 0.7606, while the predicted R-squared was 0.3104. The adequate precision ratio was 11.985, well above the threshold of 4, indicating that the model can be used to navigate the design space.

Table 2: ANOVA for Response I: Copper (Cu)

Source	SS	DF	MS	F-value	p-value
Model	278778	14	19912.71	7.352889	0.0003
B (Dosage)	113960	1	113960	42.08041	< 0.0001

C	85303.17	1	85303.17	31.49871	< 0.0001
B²	21475.17	1	21475.17	7.929837	0.0137
BC	25672.05	1	25672.05	9.47956	0.0082
BD	18239.85	1	18239.85	6.735176	0.0212

Key: SS = Sum of squares, DF= Degree of freedom, MS= Mean Square.

Lead (Pb) ANOVA

DESIGN-EXPERT Plot
Lead Ads

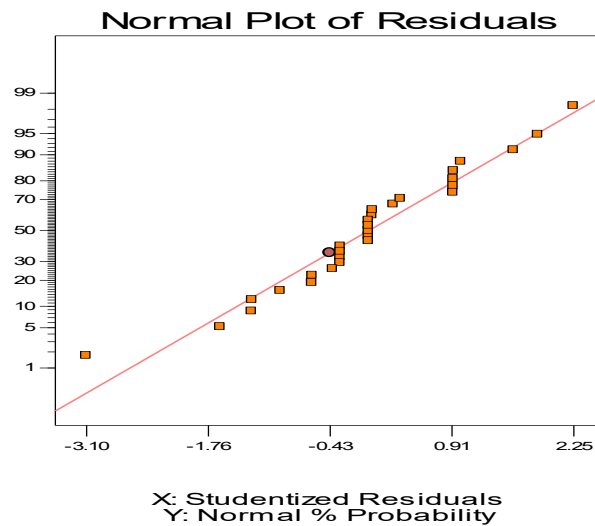


Figure 3: Lead (Pb) Diagnostic plot

DESIGN-EXPERT Plot

Lead Ads
● Design Points
X = A: pH
Y = B: Adsorbent Dosage
Actual Factors
C: Initial Concentration = 30.00
D: Contact time = 70.00

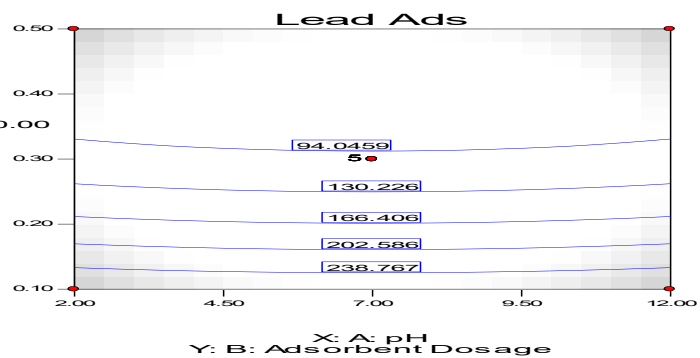


Figure 4: Lead (Pb) Contour plot

The lead model was significant ($F = 11.72$, $p < 0.0001$). The model sum of squares was 283,932.7 with 14 degrees of freedom (mean square 20,280.9). The residual sum of squares was 24,229.08 with 14 degrees of freedom (mean square 1,730.65). Significant terms included B (dosage, $p < 0.0001$), C (concentration, $p < 0.0001$), B^2 ($p = 0.0017$), BC ($p = 0.0018$), and BD ($p = 0.0307$). The R-squared value was 0.9214, and the adequate precision ratio was 14.99.

Iron (Fe) ANOVA

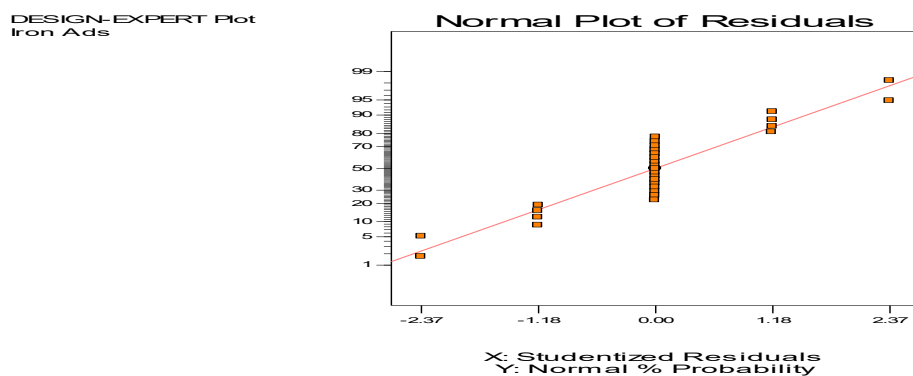


Figure 5: Iron (Fe) Diagnostic Plot of Residuals

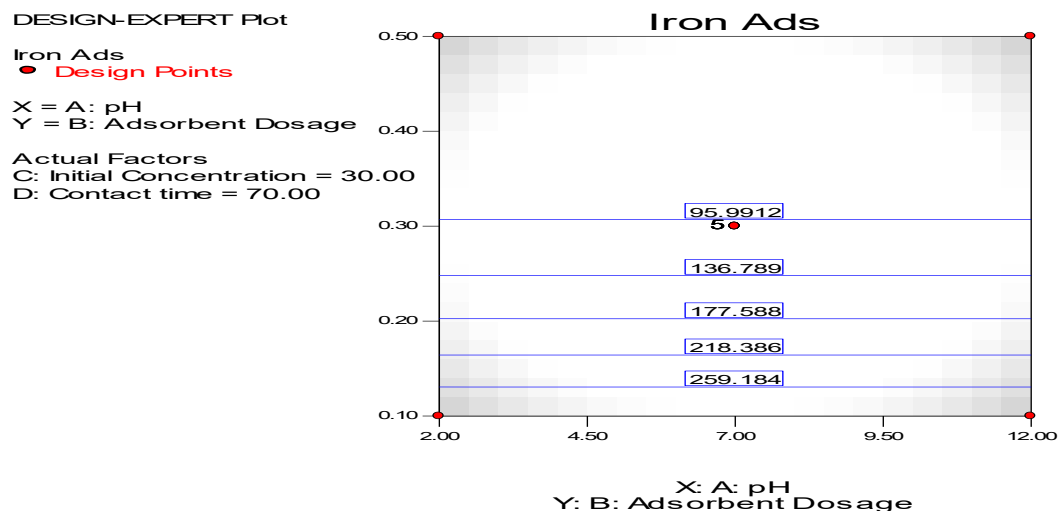


Figure 6: Iron (Fe) Contour plot of (Adsorbent dosage vs pH)

The iron model was highly significant ($F = 43.40$, $p < 0.0001$). The model sum of squares was 328,966.7 with 14 degrees of freedom (mean square 23,497.6). The residual sum of squares was 7,580.36 with 14 degrees of freedom (mean square 541.45). Significant terms were B ($p < 0.0001$), C ($p < 0.0001$), B^2 ($p < 0.0001$), and BC ($p < 0.0001$). The R-squared was 0.9775, and the adequate precision ratio was 26.83.

Cobalt (Co) ANOVA

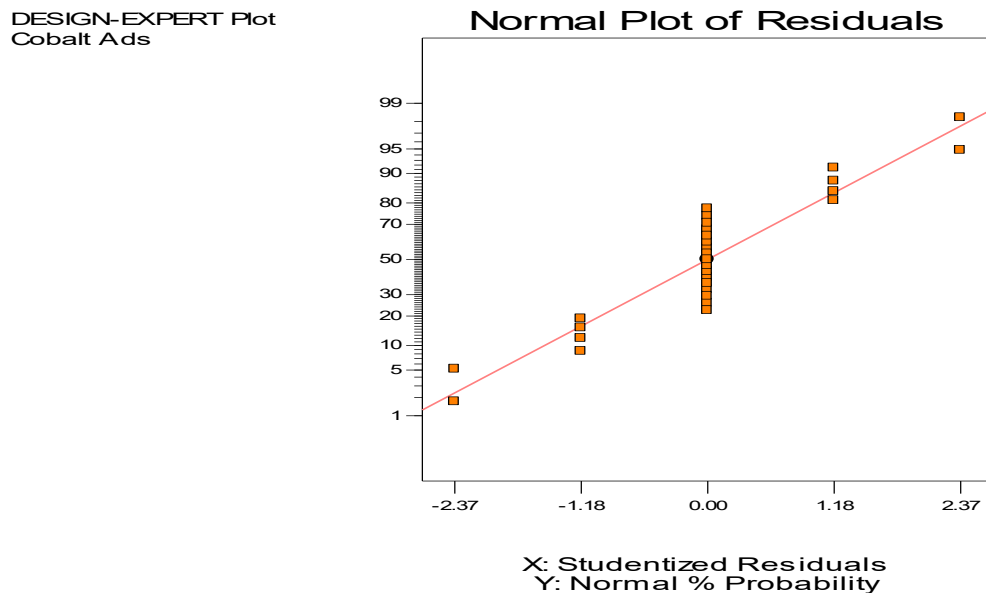


Figure 7: Diagnostic Plot of Residuals for the response IV: Cobalt (Co)

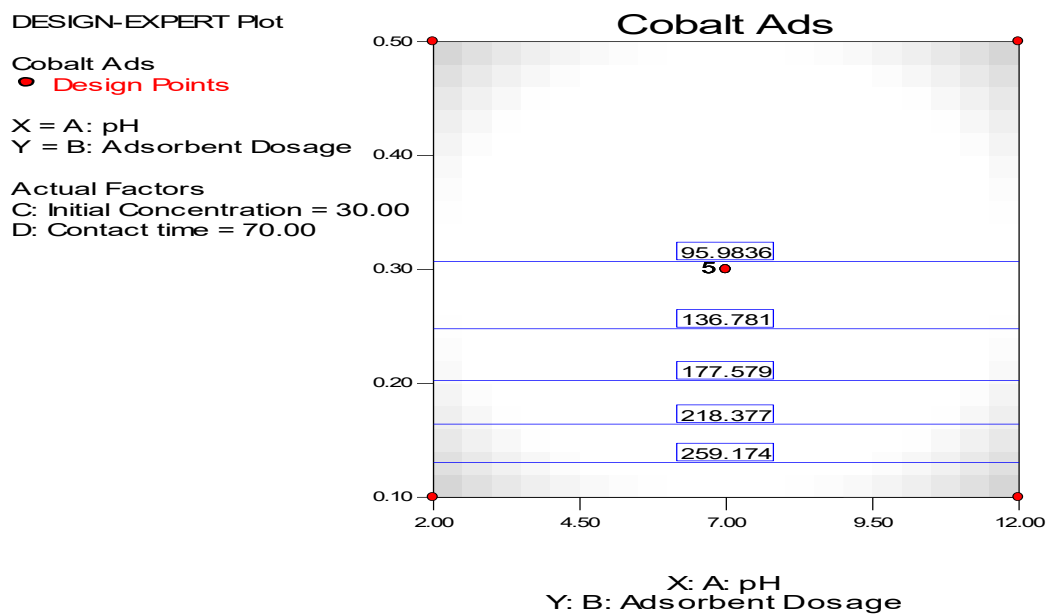


Figure 8: Contour Graph for Response IV Cobalt (Co)

The cobalt model was highly significant ($F = 43.37$, $p < 0.0001$). The pattern of significant terms was identical to iron: B, C, B^2 , and BC. The R-squared was 0.9775, and the adequate precision ratio was 26.81.

Arsenic (As) ANOVA

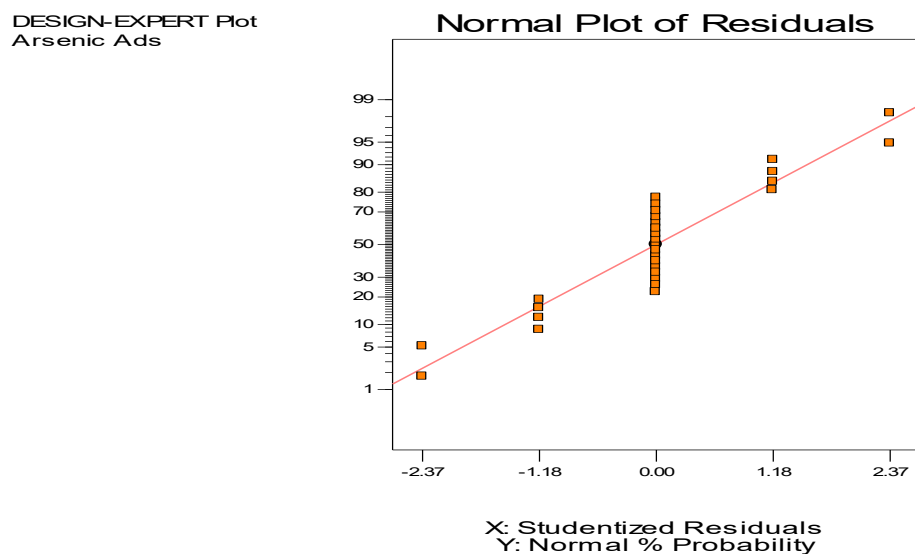


Figure 9: Diagnostic Plot Residuals fot Response V: Arsenic (As)

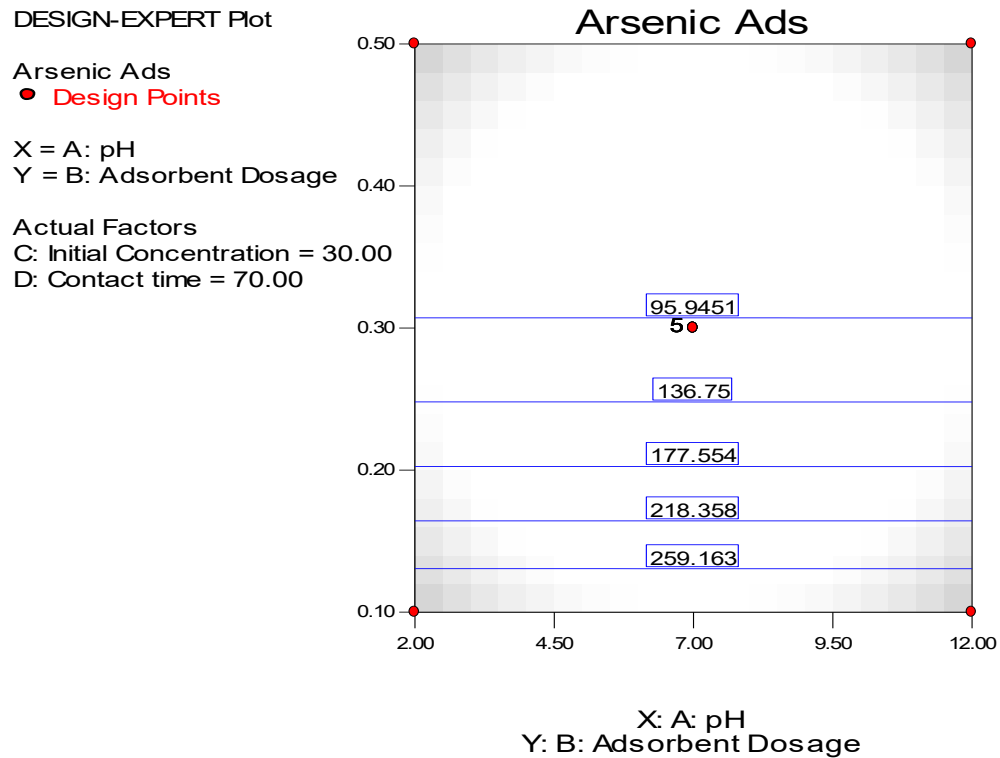


Figure 10: A Contour Graph for Arsenic (As)

The arsenic model was significant ($F = 43.41$, $p < 0.0001$). Significant terms were B, C, B^2 , and BC. The R-squared was 0.9775, and the adequate precision ratio was 26.83.

Model Coefficients and Interactions

Table 3: Coefficient Estimates for All Metals at Optimum

Response	Intercept	pH (A)	Dosage (B)	Conc (C)	Time (D)	B^2	BC
Cu Ads	99.98	-0.00167	-97.4508	84.3125	-22.6783	57.5392	-80.1125
Pb Ads	99.98	0.0325	-103.298	84.4025	16.6625	63.3183	-79.9225
Fe Ads	99.982	-0.00333	-119.978	84.4683	0.0125	79.9903	-79.9975

Co Ads	99.964	0.00667	-119.986	84.4483	-0.00417	79.9897	-80.01
As Ads	99.962	-0.02083	-119.99	84.4592	0.00333	80.0078	-80.00

All variance inflation factors (VIF) were 1.00 or close to 1.08, showing no multicollinearity.

Optimization Results

Using the desirability function approach, the optimal factor levels to maximize adsorption for all five metals were identified as pH 7.00, adsorbent dosage 0.30 g/L, initial concentration 30.00 mg/L, and contact time 70.00 minutes. Under these conditions, the predicted adsorption capacities were:

- Copper: 99.98 mg/g
- Lead: 99.98 mg/g
- Iron: 99.98 mg/g
- Cobalt: 99.98 mg/g
- Arsenic: 99.96 mg/g

Table 4: Point Prediction at Optimum Conditions

Response	Prediction	SE Mean	95% CI low	95% CI high	SE Pred	95% PI low	95% PI high
Copper Ads	99.98	23.2729	50.0645	149.896	57.0068	-22.288	222.248
Lead Ads	99.98	18.6046	60.0772	139.883	45.5717	2.23847	197.722
Iron Ads	99.982	10.4063	77.6627	122.301	25.4901	45.3112	154.653
Cobalt Ads	99.984	10.4099	77.6569	122.311	25.499	45.2941	154.674
Arsenic Ads	99.962	10.4053	77.6448	122.279	25.4877	45.2964	154.628

Confirmatory Experiments

Confirmatory experiments conducted in triplicate at the predicted optimum conditions gave average adsorption capacities of 99.96 mg/g for copper, 99.97 mg/g for lead, 99.98 mg/g for iron, 99.98 mg/g for cobalt, and 99.96 mg/g for arsenic. All experimental values fell within the 95% prediction intervals, confirming the models'

predictive capability.

Diagnostic Plots

Normal probability plots of studentized residuals for all metals showed points approximately along straight lines, indicating that residuals followed normal distributions. Studentized residuals ranged from about -2.37 to +2.37, with no values exceeding the ± 3 threshold that would indicate outliers. Cook's distances for all runs were below 1.0, indicating that no single observation had undue influence on model coefficients.

Comparison with Previous Literature

Souad *et al.* (2019) studied natural clay material from El Menia (Ghardaia, Algeria) and reported removal percentages of 98% for Pb^{2+} , 81% for Zn^{2+} , 93% for Cu^{2+} , and 84% for Ni^{2+} . Their results showed that adsorption kinetics fit the pseudo-second-order model well. David *et al.* (2023) studied bentonite clay's effectiveness for removing heavy metals from abattoir wastewater, reporting removal efficiencies ranging from 75% to 95%.

The present study's findings are consistent with these reports, demonstrating that Fika bentonite is a promising adsorbent. However, the multi-metal competitive environment in this study reduced individual metal capacities compared to single-metal systems, highlighting the importance of using realistic multi-metal conditions for wastewater treatment assessments (Yusuf *et al.*, 2023).

CONCLUSION

Response surface methodology using Box-Behnken design successfully optimized the adsorption of five heavy metals onto Nigerian Fika bentonite clay. The quadratic models developed for all metals were statistically significant, with adsorbent dosage and initial concentration emerging as the most critical factors. The significant BC interaction (dosage $\times$ concentration) for all metals confirmed that these parameters

must be optimized simultaneously. The global optimum conditions (pH 7, 0.30 g/L, 30 mg/L, 70 min) yielded adsorption capacities of approximately 100 mg/g for each metal, as validated by confirmatory experiments. The absence of significant pH and time main effects simplifies process design for practical applications.

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AUTHOR CONTRIBUTIONS

Salisu Idriss: Conceptualization, Methodology, Investigation, Data Curation, Formal Analysis, Writing - Original Draft.

Suleiman M. Shuwa: Supervision, Validation, Writing - Review & Editing.

Abdussamad Muktar Mohammed: Validation, Writing-Review & Editing.

Abdullahi Suleiman: Validation, Writing- Review & Editing.

Farhana Aziz: Validation, Writing - Review & Editing.

Muhammad Ahmadu: Validation, Writing - Review & Editing.

All authors read and approved the final manuscript.

DECLARATIONS

Ethics Approval and Consent to Participate: Not applicable

Consent for Publication: Not applicable

Conflict of Interest: The authors declare that they have no competing interests.

Data Availability: The data that support the findings of this study are available from the corresponding author upon reasonable request.

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