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Cu(II) ion removal from aqueous solution using different adsorbents

Garima Nagpal[a,*], A. Bhattacharya[b], N.B. Singh[a,*]

[a]*Research and Technology Development Centre, Sharda University, Greater Noida, India, Tel. +919911876222; email: gnagpal19fb@gmail.com (G. Nagpal), Tel. +917838500311; email: nbsingh43@gmail.com (N.B. Singh)*
[b]*Department of Chemistry, University of Petroleum and Energy Studies, Dehradun, India, Tel. +91997799473; email: amitabh_miet@yahoo.co.in*

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ABSTRACT

Removal of Cu(II) ions from aqueous solution by different adsorbents such as Khangar, bangal gram husk (BGH), and orange mesocarp (OMS) was studied. The equilibrium adsorption level was determined as a function of pH, adsorbent dose, metal ion concentration, and contact time. The working conditions were optimized by using a Taguchi L_{16} (4^4) experimental design. Statistical tools viz. signal-to-noise ratio (S/N) and analysis of variance have been used at 95% confidence level for all considered parameters. It was found that pH is the most important parameter for removal of Cu(II) from aqueous solution. The maximum adsorption capacities for Cu(II) on Khangar, BGH and OMS adsorbents were found to be 92.14, 90.34, and 85.73%, respectively. Langmuir and Freundlich adsorption isotherms were used to model the equilibrium adsorption data and it was found that for Khangar both isotherms fit the data. The study revealed that out of three adsorbents Khangar was found to be the most promising adsorbent for the removal of Cu(II) ions from aqueous solution.

Keywords: Adsorption; Langmuir isotherm; Freundlich isotherm; Modeling; Taguchi method

1. Introduction

Industrialization has resulted in release of larger amounts of heavy metals in the environment. These can be accumulated by living organisms throughout the food chain as a nonbiodegradable pollutant [1]. Among heavy metals, copper poses a significant effect on public health due to its toxicity [2,3]. To remove heavy metals from aqueous solution many physicochemical methods such as membrane filtration, coagulation, chemical precipitation, and ion exchange have been used [4–7]. The application of such processes is often limited because of technical and economical constraints. However, the adsorption technique is one of the preferred methods for removal of heavy metals because of its efficiency and low cost. The most common adsorbent materials are: alumina, silica, metal hydroxides, and activated carbon [8].

Considerable attempts have been made to prepare low-cost activated carbon adsorbents from cheaper and readily available materials. A number of lignocellulosic byproducts have been tested as precursors in the production of activated carbon, including pomegranate peel, coffee residue, tamarind wood, bagasse, fly ash, etc. [9–12]. In the present study, Khangar, bangal gram husk (BGH), and orange mesocarp (OMS) have been used as a substitute for activated carbon.

*Corresponding authors.

These are waste products. Adsorption studies have been made and optimum conditions for Cu(II) removal have been found out by batch adsorption method as well as Taguchi's method.

For optimization of process parameters, experiments have been designed using Taguchi's optimization technique and developed various possible interactions of identified process parameters. In the analysis of results powerful statistical tools; S/N ratio, ANOVA with F-test, have been used to obtain the results [13].

2. Material and methods

2.1. Materials

Carbon waste (Khangar) obtained from coal refining industry Ghaziabad, India, Black gram husk obtained from local pulse mill of Harduaganj, Aligarh, UP, India, and OMS obtained from local juice industry, Ghaziabad, India were used as adsorbents. The collected materials were washed three times with deionized water and then air dried for several days. It was oven dried at 110˚C for 2 h. The dried materials were crushed in a mechanical grinder and sieved through 350-μm mesh sieve to obtain fine powder. A stock solution of 1,000 mg/L concentration of copper sulfate was prepared in double distilled water.

2.2. Characterization of adsorbents

Proximate analysis and chemical analysis of the adsorbents were carried out as per ASTM standard [14]. Bulk densities of adsorbents were determined using bulk density meter. Moisture contents of the adsorbents were analyzed by using MB 50X moisture analyzer. The FTIR spectra of all the adsorbents before and after adsorption of Cu(II) ions were recorded in KBr phase in the wave length range 400–4,000 cm^{-1}. SEM photographs of gold coated adsorbents before and after Cu(II) ion adsorption were also recorded. Gold coating was done by sputtering technique.

2.3. Adsorption studies

The pH of the copper solution was adjusted using 0.1 N NaOH and 0.1 N HCl. Sorption studies were carried out in batch experiments by adjusting pH of the solution between 1.0 and 7.0, the initial metal ion concentration 1.0–30 mg/L, adsorbent dose 0.1–0.7 mg/50 mL, agitation time 60–150 min were chosen and the experiment were conducted at room temperature (≈35˚C). Fifty milliliter of copper sulfate solution was taken in a 250 mL iodine flask and shaken on a mechanical shaker at 150 rpm. After certain contact time, iodine flask was removed; solution was allowed to settle down and then filtered with Whatman filter paper No.1. Filtrate was collected and Cu(II) ions were estimated with the help of atomic absorption spectrometer. The amount of copper removed by the adsorbents (%) was calculated using following equation:

$$\text{Percent removal} = \left[\frac{(C_0 - C_t)}{C_0}\right] \times 100 \quad (1)$$

where C_0 is the initial Cu(II) concentration in solution (mg/L) and C_t is the concentration at the end of adsorption process (mg/L) [15,16].

2.4. Taguchi method for optimization of copper removal

Taguchi's optimization method is a unique and powerful problem-solving technique that allows optimization with minimum number of experiments. It reduces cost, improves quality, and develops rules to carry out experiments [17]. In this study, L_{16} orthogonal array experimental design was followed to explore the effect of different variables on performance of Khangar, BGH, and OMS for the removal of Cu(II) ions from solution. The selection of array was based upon the number of process parameters and their levels [17]. Four process parameters, (1) pH of the solution, (2) adsorbent dose, (3) initial metal ion concentration, and (4) contact time were selected with four levels and are given in Table 1. The interactions of process parameters and their levels are given in Table 2. Experiment was conducted as per the combinations shown in Table 2. For each interaction, three sets of samples were prepared and percent removal was calculated using Eq. (1). In Taguchi method, the analysis of the mean response for each run in the inner array as well as the variation using an appropriately chosen signal-to-noise ratio (S/N) [18] are used and calculated using following equation:

$$\left(\frac{S}{N}\right)_{HB} = -10 \log\left[1/R \sum_{j=1}^{R} 1/Y_j^2\right] \quad (2)$$

where Y_j is the response variable and R is the replication number of the experiment. Taguchi method uses S/N ratio to study the variation of response i.e. percent removal. This analysis signifies the minimization

Table 1
Factors and levels in experimental design

Factors	Level 1	Level 2	Level 3	Level 4
pH	1	3	5	7
Adsorbent dose (g/50 mL)	0.1	0.3	0.5	0.7
Metal ion conc. (mg/L)	1	10	20	30
Interaction time (min)	60	90	120	150

of fluctuations of quality characteristics due to unfavorable factors [18]. The S/N ratios are different according to the type of characteristics used in the experimental design. Condition used was higher is better (HB), i.e. larger the characteristic property, the better the result [17,19].

An analysis of variance, ANOVA was applied to the data obtained from the statistical design in order to perform a systematic analysis of the relative importance of each factor onto the copper adsorption capacity of Khangar, BGH, and OMS [20].

3. Results and discussion

3.1. Characterization of adsorbent

The FTIR spectra of Khangar, OMS, and BGH before adsorption of Cu(II) are given in Fig. 1. The spectra of OMS and BGH show an intense band between 3,300 and 3,500 cm^{-1} indicating the presence of hydrogen bonded OH/NH groups. This band is of very low intensity in the case of Khangar. This indicates the presence of both free and hydrogen bonded OH/NH groups on the adsorbent surface. The band at around 2,900 cm^{-1} in all the cases is due to C–H stretching frequency. However, this is of much low intensity in the case of Khangar. Band at about 1,500 cm^{-1} is due to C=C stretching frequency. After adsorption of Cu(II), the bands due to OH/NH vibrations are broadened and diminished and other bands shifted to lower frequency. This indicates that Cu(II) ions interact on the surfaces of adsorbents with both physical and chemical forces (see Fig. 2).

SEM studies help us to examine the surface morphology and porosity of the adsorbents. By the SEM images (Figs. 3–8), it is quite clear that the surfaces of the adsorbents are very suitable for adsorption. After adsorption the surfaces of adsorbents are drastically changed and have almost similar morphology.

3.2. Effect of process parameters on adsorption

Effect of pH on copper removal for all adsorbents is shown in Fig. 9. Adsorption increased with the increase in pH and was almost maximum at pH 7. This pH was chosen as optimum value as above this value copper precipitated as $Cu(OH)_2$ [9]. Percent removal of Cu(II) from aqueous solution in the presence of fixed dose of adsorbents (0.5 g/50 mL) at pH 7 was studied at different time intervals. It was found that the percent removal increased with time. However the removal was found to be maximum at 120 min. The time required to attain this value is termed as the equilibrium time [3].

Table 2
Experimentation data of orthogonal array L_{16}

S.No	pH of solution	Adsorbent dose (g/50 mL)	Initial metal ion conc. (mg/L)	Contact time (min)
1	1	0.1	1	60
2	1	0.3	10	90
3	1	0.5	20	120
4	1	0.7	30	150
5	3	0.1	10	120
6	3	0.3	1	150
7	3	0.5	30	60
8	3	0.7	20	90
9	5	0.1	20	150
10	5	0.3	30	120
11	5	0.5	1	90
12	5	0.7	10	60
13	7	0.1	30	90
14	7	0.3	20	60
15	7	0.5	10	150
16	7	0.7	1	120

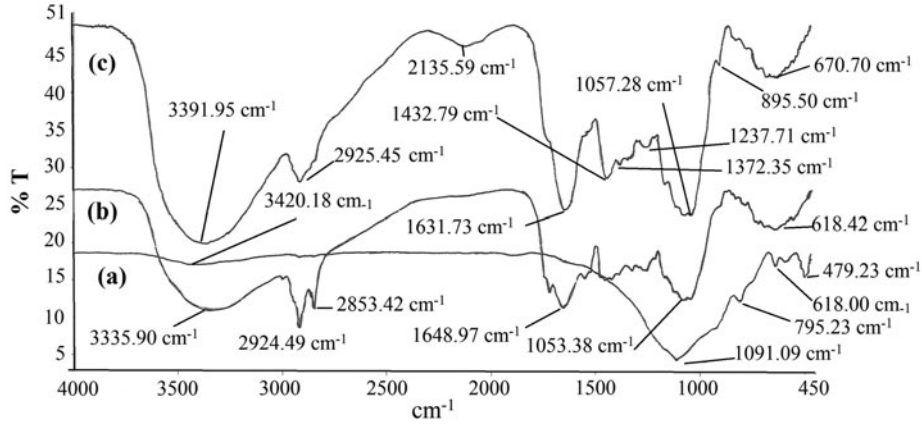

Fig. 1. FTIR image of Khangar (a), BGH (b), and OMS (c) before adsorption.

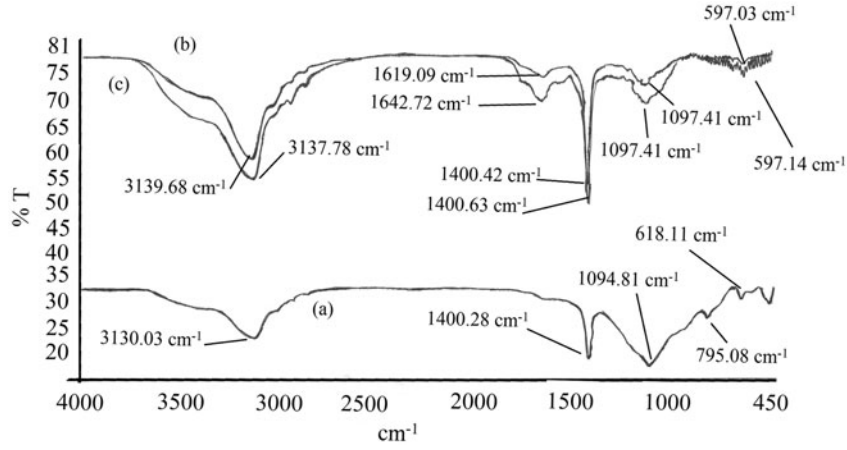

Fig. 2. FTIR image of Khangar (a), BGH (b), and OMS (c) after adsorption.

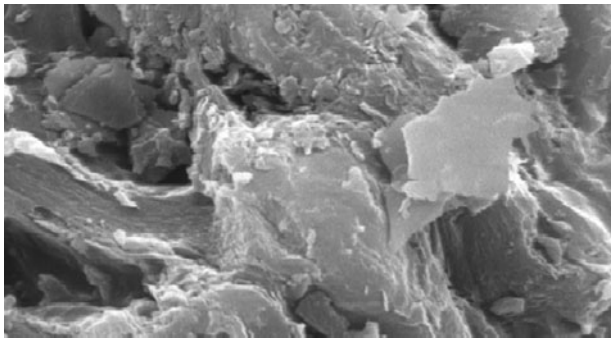

Fig. 3. SEM image of Khangar before adsorption.

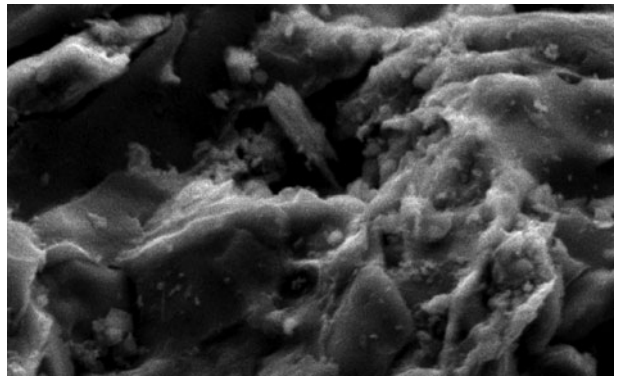

Fig. 4. SEM image of Khangar after adsorption.

The variation of Cu(II) removal from aqueous solution as a function of copper concentration at pH 7 and adsorbent dose 0.5 g/50 mL solution at 120 min is shown in Fig. 10. From the figure, it is evident that the percentage removal shows an increasing trend up to 20 mg/L and then became almost constant. The results show that the adsorption capacity of three

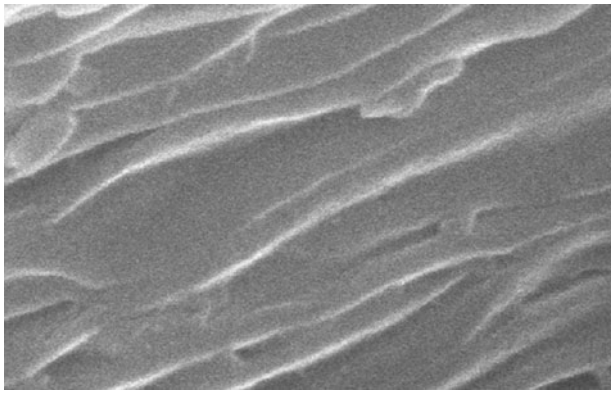

Fig. 5. SEM image of BGH before adsorption.

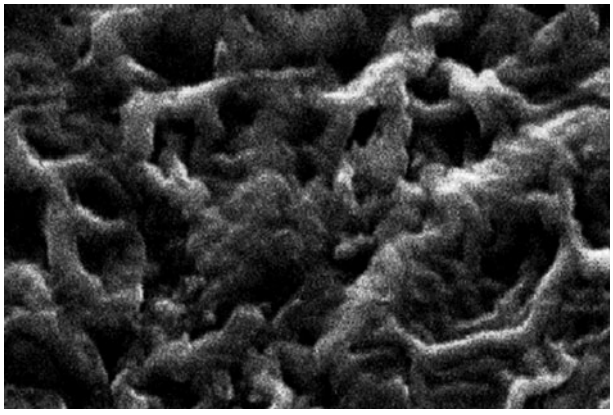

Fig. 6. SEM image of BGH after adsorption.

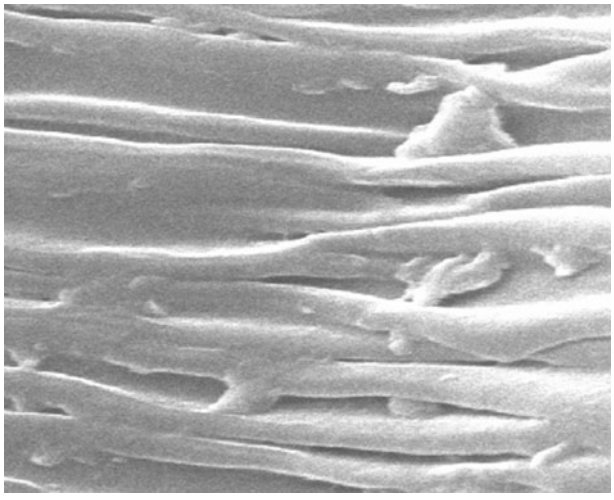

Fig. 7. SEM image of OMS before adsorption.

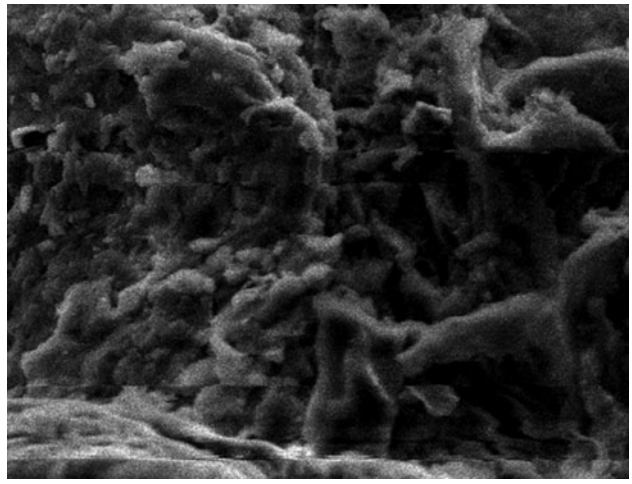

Fig. 8. SEM image of OMS after adsorption.

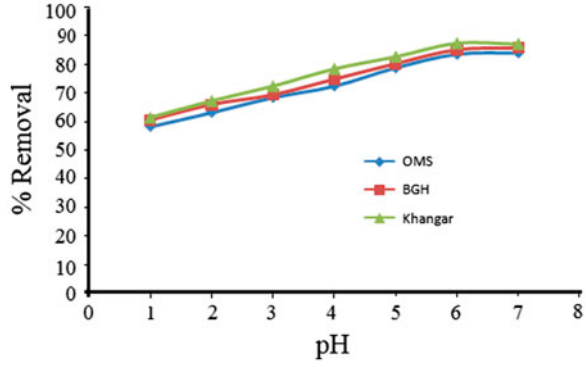

Fig. 9. Effect of pH on Cu(II) removal using Khangar, OMS, and BGH.

different adsorbents follow the sequence Khangar > BGH > OMS. This difference may be due to differences in surface area and morphology.

The effect of adsorbent dose on the percent removal of Cu(II) is shown in Fig. 11. A maximum removal of Cu(II) (%) is achieved at an adsorbent dose of 0.7 g/50 mL for Cu(II) concentration of 20 mg/L at pH 7, the adsorption capacity of the adsorbents follows the same sequence as given above. Increase in adsorption with adsorbent dosage can be attributed to the availability of more adsorption sites [21].

3.3. Adsorption isotherms

Both Langmuir (Eq. 3) and Freundlich (Eq. (4)) adsorption isotherms were allowed to fit the data (Figs. 12 and 13). Correlation coefficients (R^2) were calculated and the values are given in Table 3.

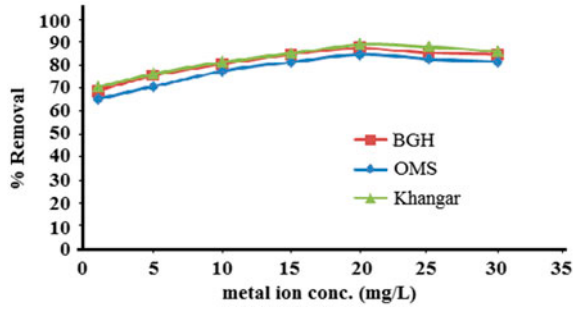

Fig. 10. Effect of initial metal ion concentration on Cu(II) removal using Khangar, OMS, and BGH.

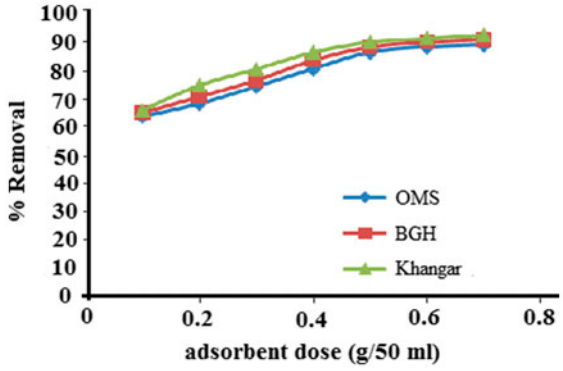

Fig. 11. Effect of adsorbent dose concentration on Cu(II) removal using Khangar, OMS, and BGH.

$$1/Q_e = 1/Q_0 + 1/Q_0 \cdot b \cdot C_e \quad (3)$$

$$\log Q_e = \log K + 1/n \log C_e \quad (4)$$

where C_e is the equilibrium concentration (mg/L), Q_e the amount of Cu(II) ions adsorbed at equilibrium (mg/g), Q_0 is the maximum adsorption capacity (mg/g), b is constant (L mg^{-1}), K (L/mg) and $1/n$ in Eq. (4) are Freundlich constants [22].

To investigate the favorability of a process, the dimensionless separation factor R_L from the Langmuir model was also calculated from Eq. (5) [23].

$$R_L = 1/(1 + bC_0) \quad (5)$$

Q_0 values given in Table 3 clearly indicate that the adsorption capacity of Khangar is maximum and minimum for OMS. This was also supported by R_L values (0.52, 0.45, and 0.34 for Khangar, BGH, and OMS, respectively).

3.4. Statistical analysis of Taguchi method

Experiments were conducted as per the design of the orthogonal array L_{16} and percentage removal of Cu(II) ion determined by Eq. (1) is given in Table 4. MINITAB-15 software was used to calculate S/N ratio and mean response for percent removal of Cu(II) from the aqueous solution. The minimum and maximum response values (response variable is removal capacity) of response were found to be 70.53 and 91.22%, respectively, for Khangar, 70.64 and 86.70%, respectively, for BGH and 62.6 and 83.25%, respectively, for OMS. The ratio between maximum and minimum value was calculated and found to be 1.29, 1.22, and 1.32 for Khangar, BGH, and OMS, respectively. As this ratio is less than 10 no transformation is required for further calculation for Taguchi design [24]. The results were analyzed using coefficient of determination (R^2) and analysis of variance (ANOVA). ANOVA was performed to see which process factor significantly affect the process response. Tables 5–7 give details of ANOVA for response by Taguchi design. Probability value (p) is lower than 0.05 which shows that model is statistically significant for all the adsorbents. Coefficient of determination (R^2) and adjusted R^2 values of all the adsorbents are given in Table 8. These values show good correlation for the proposed model.

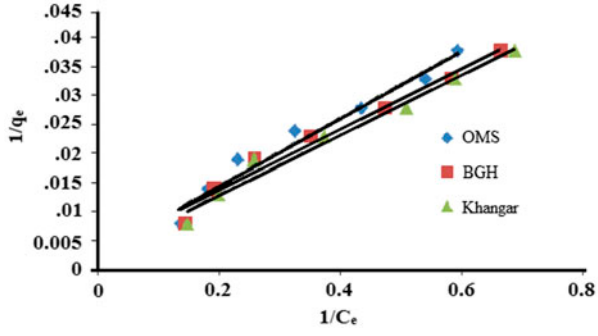

Fig. 12. Langmuir isotherm for Cu(II) adsorption onto Khangar, BGH, and OMS.

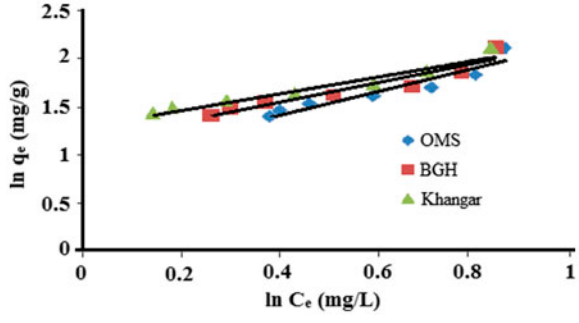

Fig. 13. Freundlich isotherm for Cu(II) adsorption onto Khangar, BGH, and OMS.

Table 3
Freundlich and Langmuir parameters for adsorption of Cu(II) on Khangar, BGH, and OMS

	Freundlich isotherm			Langmuir isotherm		
Adsorbents	$1/n$	K	R^2	Q_o	b	R^2
Khangar	0.878	1.294	0.941	0.004	0.046	0.957
BGH	0.991	1.150	0.897	0.003	0.061	0.942
OMS	1.191	0.948	0.895	0.001	0.093	0.944

Table 4
Percent removal of Cu(II) ions for various combinations of Table 2 for Khangar, BGH, and OMS

S. No.	Khangar	BGH	OMS
1	65.9	65.2	58.2
2	69.4	69.4	61.6
3	73.1	73.5	64.6
4	73.7	73.7	64.0
5	76.1	73.2	66.3
6	76.6	74.3	67.2
7	78.2	76.8	69.4
8	80.7	79.1	71.3
9	83.6	78.6	74.1
10	84.8	80.8	75.4
11	84.9	81.0	75.9
12	86.3	81.8	76.1
13	88.7	84.7	80.6
14	90.3	85.9	82.7
15	92.5	87.8	84.5
16	93.4	88.4	85.1

After optimization, the optimum values were found to be: pH 7.0, adsorbent dose = 0.7 g/50 mL, initial metal ion concentration = 20 mg/L, and contact time = 120 min.

3.5. Optimum performance characteristics for removal of Cu(II) ions

The Fischer ratio (F) of the ANOVA was used to determine significant process factors. The F value for each process factor is only a ratio of the mean of the squared deviations to the mean of the squared error. The estimated mean of percent removal characteristic can be computed from Eq. (6) [25,26].

$$\mu_{\%\text{Removal}} = \bar{T} + (\bar{A} - \bar{T}) + (\bar{B} - \bar{T}) + (\bar{C} - \bar{T}) + (\bar{D} - \bar{T}) \tag{6}$$

where $\bar{T}$ is the overall mean of the response, $\bar{A}$ is the average removal of metal ion concentration at level four of parameter pH of the solution, $\bar{B}$ is the average removal of metal ion concentration at level four of parameter adsorbent dose, $\bar{C}$ is the average removal of metal ion concentration at level three of parameter Initial metal ion concentration, and $\bar{D}$ is the average removal of metal ion concentration at level three of parameter contact time. $\mu\%$ removal by all the adsorbents is given in Table 8.

The confidence interval for the predicted mean for the confirmation experiment can be calculated by Eqs. (7) and (8).

$$\text{CI}_{\text{POP}} = \sqrt{F_\alpha(1, f_e) V_e \left[\frac{1}{\eta_{\text{eff}}}\right]} \tag{7}$$

$$\text{CI}_{\text{CE}} = \sqrt{F_\alpha(1, f_e) V_e \left[\frac{1}{\eta_{\text{eff}}} + \frac{1}{R}\right]} \tag{8}$$

where F_α (1, f_e) is variance ratio at the level of significance α (where $\alpha = 95\%$) and the confidence level is $(1-\alpha)$ against degree of freedom (DF) 1& error DF (f_e), V_e = error variance (from ANOVA); and η_{eff} is calculated from Eq. (9)

Table 5
ANOVA for response model of Taguchi design for OMS

Source	DF	Sum of squares	Mean square	F value	P
pH of the solution	3	984.54	328.18	2,969.73	0
Adsorbent dose (g/50 mL)	3	45.95	15.31	138.61	0.001
Initial metal ion conc. (mg/L)	3	5.25	1.75	15.85	0.024
Contact time	3	3.33	1.11	10.04	0.045
Residual	3	0.33	0.11		
Total	15	1,039.41			

Notes: F value—fisher's value; P—probability value.

Table 6
ANOVA for response model of Taguchi design for BGH

Source	DF	Sum of squares	Mean square	*F* value	*P*
*p*H of the solution	3	561.84	187.28	3,581.49	0.000
Adsorbent dose (g/50 mL)	3	63.55	21.18	405.14	0.000
Initial metal ion conc. (mg/L)	3	8.97	2.99	57.22	0.004
Contact time	3	4.11	1.37	26.24	0.012
Residual	3	0.15	0.052		
Total	15	638.65			

Notes: *F* value—fisher's value; *P*—probability value.

Table 7
ANOVA for response model of Taguchi design for Khangar

Source	DF	Sum of squares	Mean square	*F* value	*P*
*p*H of the solution	3	956.08	318.694	5,883.58	0.000
Adsorbent dose (g/50 mL)	3	56.35	18.783	346.75	0.000
Initial metal ion conc. (mg/L)	3	6.19	2.064	38.11	0.007
Contact time	3	6.77	2.257	41.68	0.006
Residual	3	0.16	0.054		
Total	15	1,025.56			

Notes: *F* value—fisher's value; *P*—probability value.

Table 8
Coffecient of determination and R^2 adjusted value for Khangar, BGH, and OMS

	Khangar	BGH	OMS
R^2	0.999	1	1
R^2 adjusted	0.996	0.999	0.998
$\mu_{\%\ \text{Removal}}$	94.74	90.40	86.43

$$\eta_{\text{eff}} = \frac{N}{1 + \text{Total DF involved in estimation of mean}} \quad (9)$$

where N = total number of experiments performed, R = sample size for the confirmation experiment, i.e. number of times an experiment repeated, which is in the present case is 03.

Table 9
Responses at optimum levels of process parameters for Khangar, BGH, and OMS

Responses	Predicted mean value	Experimental value (average)	Confidence interval
% Removal of metal ions using Khangar	92.75	92.14	$92.16 < \mu_{\%\ \text{Removal}} < 93.31$ (CI_{CE}) $92.35 < \mu_{\%\ \text{Removal}} < 93.12$ (CI_{POP})
% Removal of metal ions using OMS	86.43	85.94	$85.61 < \mu_{\%\ \text{Removal}} < 87.26$ (CI_{CE}) $85.88 < \mu_{\%\ \text{Removal}} < 86.98$ (CI_{POP})
% Removal of metal ions using BGH	89.93	90.34	$89.84 < \mu_{\%\ \text{Removal}} < 90.96$ (CI_{CE}) $90.02 < \mu_{\%\ \text{Removal}} < 90.78$ (CI_{POP})

By substituting values N: total no of results = 16 × 3 = 48; Total DF = 12, f_e = 3, and V_e (Error variance i.e. residual from Tables 5–7); $F_{0.05}(1,3)$ = 10.1 (Tabulated F-value) in Eqs. (8)–(10). The 95% confidence intervals (Cl_{Pop} and Cl_{CE}) of the predicted ranges for adsorption of Cu(II) ions onto Khangar, BGH, and OMS were compared with the predicted values (Table 9). Experimental results have shown that the maximum removal of Cu(II) ions from an aqueous solution by the three adsorbents (Khangar, BGH, and OMS) has been achieved at pH–7, adsorbent dose—0.7 g/50 mL, Cu(II) ion concentration—20 mg/L, and contact time—120 min. The same conditions were used in Taguchi optimization process. The results obtained by the Taguchi process and the experimental values are in good agreement (Table 9). Thus, the results very clearly indicate that the optimized conditions are the most appropriate conditions. When the removal efficiencies of the three adsorbents were compared it was found that Khangar is the best adsorbent followed by BGH and OMS (Khangar > BGH > OMS).

4. Conclusion

Khangar, BGH, and OMS were used as adsorbents to find out optimum conditions for Cu(II) removal from aqueous solution using batch adsorption method and Taguchi method. The optimum conditions for removal of Cu(II) are pH—7, initial metal ion conc.—20 mg/L, adsorbent dose—0.7 g/50 mL, and contact time—120 min. Based on the experimental results Khangar was found very effective.

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