Synthesis and characterization of cupric oxide (CuO) nanoparticles and their application for the removal of dyes

Ghulam Mustafa¹, Hajira Tahir¹*, Muhammad Sultan² and Nasir Akhtar¹

¹Department of Chemistry, University of Karachi 75270, Pakistan.
²Federal Urdu University of Arts, Science and Technology Karachi- Pakistan.

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In the present work, cupric oxide (CuO) nanoparticles (NPs) were prepared by adopting aqueous precipitation method using copper sulphate 5-hydrate as a precursor and NaOH as a stabilizing agent. This gives a large scale production of CuO-NPs which are utilized for the removal of methylene blue (MB) dye. The CuO NPs were characterized for the studying of their structure and composition from XRD which reveals the single phase monoclinic structure. The surface morphology of these NPs was carried out by using scanning electron microscopy (SEM). Moreover, the effect of optimization parameters such as time, concentration and temperature was also examined. Spectrophotometric technique was used to evaluate the removal of MB in aqueous solution by NPs. The equilibrium adsorption of cationic dye (MB) was carried out at various temperatures ranging from 303 to 318 K ± 2K. The adsorption isotherm equations like Langmuir, Freundlich and Dubinin-Radushkevich were applied and the values of their respective constants were evaluated by adopting graphical method. Thermodynamic and kinetic studies were also performed to determine the feasibility of the process. The maximum MB removal was observed to be 88.93%. The pH of point zero charge (pH_{PZC}) of adsorbent was also estimated by pH drift method. The results indicate that aqueous precipitation method is a reliable and cheap method for the development of CuO-NPs which can be effectively used for the removal of dyes from effluents. This method is beneficial for the remediation of industrial waste.

Key words: CuO nanoparticles, XRD, SEM, methylene blue, adsorption, UV Spectrophotometer.

INTRODUCTION

There is a number of metal oxide in nature but some of the metal oxides are most useful in accordance with their applications of day to day life in science and technology. Physico-chemical properties of special relevance in Chemistry are mostly related to the industrial use of oxides as sensors, ceramics, adsorbents and catalysts. A nano-silica-Ag NPs composite material is proposed as a novel antifouling adsorbent for cost-effective and eco-friendly water purification (Das et al., 2013), novel magnetic Fe₃O₄@C nanoparticles have been synthesized and employed as high efficient adsorbent for removal of cationic dyes from polluted water (Zhang and Kong, 2011). The utilization of modified magnetite nanoparticles (Fe₃O₄ NPs) with a cationic surfactant (cetyl trimethyl ammonium bromide, CTAB) as an efficient adsorbent was successfully carried out to remove reactive black 5 (RBBA), reactive red 198 (RRR) and reactive blue 21 (RTB) dyes from aqueous solutions (Faraji et al., 2010).

The creation of environmental problems due to rapid development of technology causes a threat to our horizon. Dyes can be said to be colored, ionizing and aromatic organic compounds which shows an affinity
towards the substrate to which it is being applied. It is generally applied in a solution that is aqueous and required a mordant to improve the fastness of the dye on the material on which it is applied.

They have many different structural variations like acidic, basic, disperse, azo, anthra quinone based and metal complex dyes among others. The textile industry is the largest consumer of dye stuffs. During the coloration process, a large percentage of the synthetic dye does not bind and is lost to the waste stream (Weber and Adams, 1995). Approximately 10-15% dyes are released into the environment during dyeing process making the effluent highly colored and aesthetically unpleasant (Ratna and Padhi, 2012). Dyes have been applied in many industries such as textiles, printing, leather, pulp, food and plastics, etc. The effluent from these industries normally contains presence of these dyes. About 10,000 different commercial dyes and pigments exist and more than 7x10^5 tons are produced per year worldwide (Wanchanthuak and Nurunung, 2011; Shah et al., 2010). Approximately 10-15% of these dyes are released after dying process Al-Degs et al. (2008) since many organic dyestuffs are harmful to human being and toxic to animals and microorganisms. The dye removal has been in considerable attention over the past decades before release to natural stream. Various dye removal process have been used such as coagulation, chemical oxidation, membrane separation, electrochemical process, biological treatment and adsorption techniques Durai and Rajasimman (2011). Adsorption was recognized to be an effective process for the removal of dyes from waste water effluents which are the easiest in separation after the process and highly effective in dye removal (Orthman et al., 2003). Different kinds of adsorbents have been developed for various applications such as activated carbon (Iqbal et al., 2007), active carbon from pyrolysis of bagasse (Lori et al., 2008), spent activated clay (Weng et al., 2007), sand (Rauf et al., 2008), soil Cheng et al. (2008), kaolin (Nandi et al., 2009), biomass from calotropis procera leaf (Ali et al., 2008), natural resin (Clinoptilolite) (Moazed, 2008), and Mg-Al-CO_3 layers (Gaini, 2009). CuO NPs Aparna et al. (2012) and carbon-encapsulated super magnetic colloidal NPs Wang et al. (2013). In the present work, CuO nanoparticles were prepared and utilized as nano adsorbents for the removal of cationic dye. The XRD and morphological study of CuO-NPs was observed by scanning electron microscopy (SEM).

**MATERIALS AND METHODS**

**Synthesis of CuO nanoparticles**

CuO nanoparticles were synthesized by aqueous precipitation method using 55 g of copper sulfate 5-hydrate (Merck) mixed with 25 g of hydroxyl ammonium chloride (Sigma Aldrich) in 125 ml distilled water. The mixture was allowed to cool in a cold water bath with swirling well and solution of 40 g of sodium hydroxide in 750 ml distilled water was added. The precipitate was allowed to settle down and the supernatant liquid was poured off according to a modified method of Kannaki et al. (2012). The oxide was transferred to 250 ml flask and the volume was made up. The content was washed by repeated decantation until the rinsing was chloride-free, and then suction filtration was applied and the residue was washed with 95% alcohol and ether. The residue was dried at 200 to 250°C in air oxidized conditions to obtain CuO and then further the temperature was increased to 300°C which leads to the formation of CuO. For the formation of CuO NPs from CuO precipitant, the following reaction mechanism can be formulated as represented as:

\[
\begin{align*}
2\text{CuSO}_4 + 2\text{NH}_4\text{OH} + \text{6NaOH} & \rightarrow \text{CuO} + \text{N}_2 + 2\text{NaCl} + 2\text{Na}_2\text{SO}_4 + 7\text{H}_2\text{O} \\
\text{CuO} & \rightarrow \Delta \text{CuO}
\end{align*}
\]

**Characterization of nanoparticles**

The XRD patterns of nano CuO were recorded on a (BRUKER AXS) diffractometer in the scanning range of 20-70° (2θ) using CuKα as radiation source having a wavelength of 1.54060 Å at the scanning rate of 15.50 s with the instrument temperature of 25°C. The morphology of prepared NPs before and after adsorption was examined by SEM technique. SEM images were taken by Joel Quick Auto Coater and by using ion sputtering device.

**Treatment of methylene blue**

The removal of methylene blue from aqueous solution was carried out by adsorption method using CuO-NPs.

**Optimization of amount of adsorbent**

CuO-NPs of known amounts 0.01 - 0.1 g was added in shaking flasks and placed in electric shaker for a desired time period. The amount of dye adsorbed (mg/g) increased with increase in time and then reached equilibrium. The contents of the flasks were filtered to separate the NPs. Concentration of methylene blue was quantified in the filtrate by UV spectrophotometer. The initial dye concentration provides the necessary driving force to overcome the resistances to the mass transfer of MB between the aqueous and solid phases (Rauf et al., 2007). A similar phenomenon was observed for the adsorption of Methylene Blue (MB) dye onto banana stalk waste by Hameed et al. (2007), pomelo (C. grandis) peel (Wang et al., 2007) and castor shell seed (Ni et al., 2007).

**Optimization of shaking time**

The shaking time also varied from 2 to 24 min by keeping the optimum amount of adsorbent and concentration of methylene blue. After the interval of 2 min each, flask was ejected and the content filtered. The filtrate was analyzed by UV spectrophotometer to study the adsorption of MB on NPs as a function of contact time in order to find out the equilibrium time for maximum adsorption. Sureshkumar et al. (2008) reported an equilibrium adsorption time of 135 min for the adsorption of Methylene Blue onto wheat shells and 150 min for the adsorption of methylene blue on fallen phoenix tree’s leaves by Ofomaja et al. (2007)

**Optimization of temperature**

The shaking temperature also varied from 30 to 45°C by keeping
the optimum amount of adsorbent and time against known concentration of methylene blue (1x10^-5 to 1x10^-4 M). The adsorption sets were run at 30, 35, 40 and 45°C for 60 min. The contents of the flasks were filtered to separate the adsorbent. The filtrate was analyzed by UV spectrophotometer. Alkan et al. (2008) and Abd El-Latif et al. (2009) reported that the changing of the temperature will change the equilibrium capacity of the adsorbent for a particular adsorbate.

**pH at the point of zero charge (pH\text{zpc})**

The point of zero charge (pH\text{zpc}) is the pH at which the total number of positive and negative charges on its surface becomes zero (Jia et al., 2002). The pH at the point of zero charge (pH\text{zpc}) of CuO-NPs was measured by using the pH drift method. The pH of 0.005 mol/dm³ NaCl solution was adjusted between 2-12 by adding either HCl or NaOH. The 0.03 g of adsorbent was added in 20 ml of the solution in Erlemeyer flask and left at room temperature for 24 and 48 h. After the pH stabilized, the final pH was recorded. The graph of pHs was drawn and used to determine the points at which the initial and final pH values were equal (Vijayakumar et al., 2012).

**RESULTS AND DISCUSSION**

Powder X-ray diffraction study was carried out on the prepared CuO NPs. These NPs retained its monoclinic structure with lattice parameters: \(a = 4.688\text{Å}, b = 3.422\text{Å}, c = 5.131\text{Å}, \beta = 99.506\) and \(V = 82.31\text{ Å}^3\). The crystalline CuO NPs were confirmed by the powder X-ray diffraction study and diffraction peaks are indexed in Figure 1.

The crystal structure parameters obtained show that all the diffraction peaks can be indexed with lattice planes and compared to the International center for diffraction data (ICDD) Card No:41-0254. The d-spacing values of the peaks were well matched with the reported values in the card. The grain size for different FWHM (\(\beta\)) values was calculated using Debye - Scherrer’s equation (Lisa et al., 2007).

\[
D = \frac{K\lambda}{\beta \cos \theta}
\]  

(3)

Where, \(K\) is a constant representing shape factor which is about 0.9, \(\lambda\) is the X-ray wavelength used which is 1.5406\(\text{Å}\), while \(\beta\) is the full width half maximum (FWHM) of the diffraction angle. No peaks of impurities were found in XRD pattern. The peaks were broad due to the nano size effect. The average crystalline size of CuO

**Figure 1. XRD pattern of CuO nanoparticles.**
NPs was found to be 20 - 28 nm.

**Surface morphological studies**

The images of Scanning Electron Microscope of CuO-NPs before and after adsorption of MB dye are shown in Figures 2 and 3 at different magnification. SEM image of CuO before adsorption of MB dye indicates that the material is composed of irregularities in shape from 0.02 to 1 µm which provides a large surface area for the adsorption experiment. After the adsorption of MB dye, the structures become saturated with MB molecules.

**Investigation of adsorption parameters**

The present work represents the removal of methylene blue dye onto CuO-NPs by adsorption method. The effect of point of zero charge, thermodynamic and kinetic studies were run by batch adsorption processes under the optimized conditions of amount of adsorbent, contact time and temperatures.

**Effect of amount of adsorbent dosage**

The adsorption of methylene blue (MB) dye onto CuO-NPs was carried out by varying the amount of adsorbent from 0.02 - 0.2 g. The values of % removal of MB dye were increased with the increase in amount of CuO-NPs and 0.03 g of adsorbent showed the optimum adsorption as shown in Figure 4. This may be due to the increase in adsorbent surface area and viability of more adsorption sites. The amount of adsorbent at time t and \( K_d \) (mol/g) were calculated as:

\[
K_d = \frac{(C_i - C_f)V}{W} \quad (4)
\]

Where \( C_i \) (mol/dm\(^3\)) is the concentration of dye after removal, \( C_f \) (mol/dm\(^3\)) is the initial concentration of dye in the solution, \( V \) is the volume of the solution (ml) and \( W \) is the mass of the adsorbent (g).

The amount of equilibrium adsorption, \( x/m \) (mol/g), was calculated as:

\[
x/m = \frac{(C_0 - C_e)V}{W} \quad (5)
\]

Where, \( C_0 \) and \( C_e \) (mol/dm\(^3\)) are the initial and equilibrium concentrations of the dye in the solution.

The percentage removal was calculated as:

\[
\% \text{ Removal} = \frac{C_0 - C_e}{C_0} \times 100 \quad (6)
\]

**Effect of contact time**

The rate of adsorption of dye decreased with the increase in contact time (02 to 24 min) and reached to an optimum value when the adsorption equilibrium was achieved. The rate of adsorption indicated that the removal of more than 86.28% of MB was observed in the first 2 min. The maximum adsorption capacity of MB on CuO-NPs was obtained at 06 min. The results show that the removal process was very fast and this is an important advantage of separation method as shown in Figure 5.

**Effect of temperature**

Adsorption isotherm is a graphical representation between the bulk activity of adsorbate and amount adsorbed at constant temperature. It characterizes the distribution of adsorbed solute between the adsorbate and solid phases at various equilibrium concentrations.

The effect of temperature on the removal tendency of dye was also studied at temperatures ranges from 303 to 318 K. The critical review of adsorption isotherm shows
Optimization of CuO amount

![Graph showing optimization of CuO amount for the removal of MB.]

Figure 4. Optimization of amount of CuO for the removal of MB.

![Graph showing optimization of time of CuO for the removal of MB.]

Figure 5. Optimization of time of CuO for the removal of MB.

an increase in the adsorption of dye with the rise in temperature. It shows that adsorption of dye on NPs is endothermic in nature.

**Freundlich adsorption isotherm**

Freundlich expressed an empirical equation for representing the isothermal variation of adsorption on the quantity of gas adsorbed by unit mass of solid adsorbent with concentration. It is expressed as:

\[ \log \frac{X}{m} = \log K + \frac{1}{n} \log C_e \]  

(7)

Where, \( \frac{X}{m} \) is the amount adsorbed per unit mass of the adsorbent (mol/g), \( C_e \) is the equilibrium concentration (mol/dm³) and the constant \( K \) relates to the degree of adsorption, while \( n \) provides the rough estimation of the
Freundlich Isotherm Plot of CuO NPs for the removal of MB dye.

Table 1. Freundlich parameters for methylene blue dye adsorption on CuO nanoparticles.

<table>
<thead>
<tr>
<th>Adsorbent</th>
<th>Temperature (K)</th>
<th>K</th>
<th>N (10^1)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CuO-NPs</td>
<td>303</td>
<td>2.03E+05</td>
<td>3.46</td>
</tr>
<tr>
<td></td>
<td>308</td>
<td>8.93E+03</td>
<td>1.73</td>
</tr>
<tr>
<td></td>
<td>313</td>
<td>8.83E+08</td>
<td>22.5</td>
</tr>
<tr>
<td></td>
<td>318</td>
<td>1.26E+07</td>
<td>8.63</td>
</tr>
</tbody>
</table>

The decrease in the values of K with the rise in temperature indicates that adsorption affinity of dye on CuO-NPs is less favorable at higher temperatures.

Langmuir adsorption isotherm

The Langmuir isotherm assumes the monolayer adsorption on a homogeneous surface with a finite number of adsorption sites. The Langmuir model assumes that the adsorptions occur at specific homogeneous sites on the adsorbent and is used successfully in many monolayer adsorption processes. The theory can be expressed by the following linear equation:

\[ \frac{C_e}{X/m} = \frac{1}{kV_m} + \frac{C_e}{V_m} \]  \hspace{1cm} (8)

Where, \( C_e \) is the equilibrium concentration (mol/dm^3), \( X/m \) is the amount adsorbed at equilibrium (mol/g) and \( V_m \) (mol/g) and \( k \) (dm^3/mol) are the Langmuir constants related to monolayer capacity and adsorption coefficient respectively (Hameed and Daud, 2008). The linear plot of \( C_e/X/m \) vs \( C_e \) is shown in Figure 7 and the values of respective constants are shown in Table 2.

D-R adsorption isotherm

Dubinin Radushkevich (D-R) isotherm was also applied for the adsorption of methylene blue (MB) on to CuO-NPs. The D-R equation can be expressed as:

\[ \ln \frac{X/m}{X_m} = \ln X_m - K \varepsilon^2 \]  \hspace{1cm} (9)

Where, \( X_m \) is the monolayer capacity of adsorbent, \( K \) is a constant related to adsorption energy, while \( \varepsilon \) is the adsorption potential which can be obtained as:

\[ \varepsilon = RT \ln (1 + 1/C_e) \]  \hspace{1cm} (10)

Where, \( C_e \) is the equilibrium concentration of dye (mol/dm^3), \( R \) is a gas constant and \( T \) is the absolute temperature. The D-R plot of \( \ln X/m \) versus \( \varepsilon^2 \) was
Figure 7. Langmuir Isotherm of CuO-NPs for the removal of MB Dye.

Table 2. Langmuir parameters for methylene blue dye adsorption on CuO nanoparticles.

<table>
<thead>
<tr>
<th>Adsorbent</th>
<th>Temperature (K)</th>
<th>Xm</th>
<th>Es (KJ/mol) $10^3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>CuO-NPs</td>
<td>303</td>
<td>4.11E+01</td>
<td>7.07</td>
</tr>
<tr>
<td></td>
<td>308</td>
<td>9.66E+00</td>
<td>7.45</td>
</tr>
<tr>
<td></td>
<td>313</td>
<td>2.34E+03</td>
<td>5.00</td>
</tr>
<tr>
<td></td>
<td>318</td>
<td>1.18E+01</td>
<td>10.0</td>
</tr>
</tbody>
</table>

obtained at various temperatures as shown in Figure 8 and in Table 3. The values of K and Xm were obtained from the slope intercept while the values of mean free energy of adsorption (Es) were estimated by using the value of K as expressed:

$$Es = (\frac{-2K}{1})^{1/2}$$  \hspace{1cm} (11)

Investigation of point of zero charge (pH<sub>pzc</sub>)

For the investigation of point of zero charge CuO-NPs, the values of the initial and final pH were plotted at 24 and 48 h time intervals as shown in Figure 9. From the graph, the values of pH<sub>pzc</sub> of CuO were determined from the points where the initial pH equals the final pH as shown in Table 4. The pH values implies that surface of CuO was basic in nature since the pH<sub>pzc</sub> values were increased from acidic pH to neutral and then moved from basic to neutral and finally toward the basic behavior again.

Investigation of adsorption kinetics

The time dependent behavior of adsorption was determined by varying the equilibrium time (2-24) min between adsorbate and adsorbent. The percentage removal of the MB dye against contact time plotted as shown in Figure 10 indicates that the equilibrium was established between MB dye and CuO NPs within 10 min. For the investigation of possible mechanism of adsorption, pseudo first order model was adopted by the
Table 3. D-R parameters for methylene blue dye adsorption on CuO nanoparticles.

<table>
<thead>
<tr>
<th>Adsorbent</th>
<th>Temperature (K)</th>
<th>( E_s ) (J/mol) (10^3)</th>
<th>( X_m ) (10)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CuO-NPs</td>
<td>303</td>
<td>7.07E+03</td>
<td>4.11E+01</td>
</tr>
<tr>
<td></td>
<td>308</td>
<td>7.45E+03</td>
<td>9.66E+00</td>
</tr>
<tr>
<td></td>
<td>313</td>
<td>5.00E+03</td>
<td>2.34E+03</td>
</tr>
<tr>
<td></td>
<td>318</td>
<td>1.00E+04</td>
<td>1.18E+01</td>
</tr>
</tbody>
</table>

Table 4. Point of zero charge (pH\(_{pzc}\)) of CuO nanoparticles.

<table>
<thead>
<tr>
<th>Adsorbent</th>
<th>Amount (g)</th>
<th>Initial pH</th>
<th>Final pH After 24 h</th>
<th>Final pH After 48 h</th>
</tr>
</thead>
<tbody>
<tr>
<td>CuO-NPs</td>
<td>0.03</td>
<td>2.20</td>
<td>5.55</td>
<td>5.62</td>
</tr>
<tr>
<td></td>
<td>0.03</td>
<td>4.10</td>
<td>7.00</td>
<td>7.11</td>
</tr>
<tr>
<td></td>
<td>0.03</td>
<td>6.04</td>
<td>7.25</td>
<td>7.31</td>
</tr>
<tr>
<td></td>
<td>0.03</td>
<td>7.98</td>
<td>7.39</td>
<td>7.38</td>
</tr>
<tr>
<td></td>
<td>0.03</td>
<td>10.00</td>
<td>7.88</td>
<td>7.96</td>
</tr>
<tr>
<td></td>
<td>0.03</td>
<td>12.01</td>
<td>12.87</td>
<td>12.80</td>
</tr>
</tbody>
</table>

Lagergen (Lagergren, 1898) and pseudo second order models were adopted by the Ho and McKay (Ho and McKay, 1999). They are expressed as:

\[
\log (q_e - q_t) = \log q_e - k_1 t / 2.303 \tag{15}
\]

\[
t/q_t = \frac{1}{2} k_2 q_e^2 + \frac{1}{t} q_e \tag{16}
\]

Where, \( q_e \) and \( q_t \) are the amount of the dye adsorbed on the adsorbent (mol/g) at equilibrium and time \( t \), \( k_1 \) is the rate constant (min\(^{-1}\)) representing the pseudo first order kinetics and \( k_2 \) is the adsorption of pseudo second order rate constant (mol/g.min). The pseudo second-order adsorption kinetic plot is shown in Figure 10, which shows that it follows a pseudo-second order reaction.
Figure 9. Effect of Point of Zero Charge on CuO-N

Table 5. Thermodynamic parameters for Methylene Blue dye adsorption on CuO nanoparticles.

<table>
<thead>
<tr>
<th>Adsorbent</th>
<th>Conc. of dye (M) (10⁻⁵)</th>
<th>ΔH (KJ/mol) (10)</th>
<th>ΔS (J/mol)</th>
<th>ΔG (J/mol) x 10³</th>
</tr>
</thead>
<tbody>
<tr>
<td>CuO-NPs</td>
<td>1.00E-05</td>
<td>88.19</td>
<td>1.16</td>
<td>-263.29</td>
</tr>
<tr>
<td></td>
<td>2.00E-05</td>
<td>92.37</td>
<td>1.16</td>
<td>-259.11</td>
</tr>
<tr>
<td></td>
<td>3.00E-05</td>
<td>78.78</td>
<td>1.12</td>
<td>-260.58</td>
</tr>
<tr>
<td></td>
<td>4.00E-05</td>
<td>-0.60</td>
<td>0.86</td>
<td>-261.18</td>
</tr>
<tr>
<td></td>
<td>5.00E-05</td>
<td>65.91</td>
<td>0.83</td>
<td>-258.30</td>
</tr>
<tr>
<td></td>
<td>6.00E-05</td>
<td>21.66</td>
<td>0.93</td>
<td>-260.13</td>
</tr>
<tr>
<td></td>
<td>7.00E-05</td>
<td>-7.49</td>
<td>0.83</td>
<td>-258.98</td>
</tr>
<tr>
<td></td>
<td>8.00E-05</td>
<td>61.44</td>
<td>1.05</td>
<td>-256.71</td>
</tr>
<tr>
<td></td>
<td>9.00E-05</td>
<td>-3.89</td>
<td>0.83</td>
<td>-255.38</td>
</tr>
<tr>
<td></td>
<td>10.00E-05</td>
<td>-20.78</td>
<td>0.91</td>
<td>-296.51</td>
</tr>
</tbody>
</table>

Conclusion

CuO-NPs were prepared successfully with monoclinic structure and confirmed by XRD analysis. The fabricated NPs are composed of irregular shapes ranges from 0.02 to 1 µm which provide a large surface area for the adsorption as determined by SEM results.

These NPs used as adsorbent for the removal of MB dye. Adsorption, desorption, point of zero charge pHpzc, thermodynamic and kinetics studies were proceeded to determine the validity of process. The adsorption experiments were run under the optimized conditions of amount of adsorbent, stay time, initial concentration at different temperatures. The adsorption models like: Freundlich, D-R and Langmuir adsorption isotherm models were applied to determine the adsorption equilibrium data. Thermodynamic parameters, free energy change (ΔG°), enthalpy change (ΔH°) and entropy change (ΔS°) show that the adsorption of MB dye onto CuO-NPs was spontaneous and endothermic process.

The kinetic data showed that the adsorption process followed the pseudo second order kinetics.

The feasibility of adsorption / removal process was examined in the present study. It was estimated that inexpensive and cost effective materials can be prepared as adsorbents for the removal of dyes and metals. Undoubtedly low-cost adsorbents offer a lot of promising benefits for commercial purposes with respect to utili-
zation and minimization of waste.

REFERENCES


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