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# Evaluation of Degradation of Chromium from Wastewater by Using *Ricinus communis* Biochar along with ZnO and CuO Nanocomposites

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**Abstract** **Original Research Article**

Water contamination is a worldwide problem which leads to many diseases in animals, plants and in humans. It is highly documented that anthropogenic and industrial action are the key sources of surface and ground water contamination all over the world. Various industries like leather, textile, smelting and alloying directly discharges into water bodies which potentially affect water quality. This study illustrates the synthesis and application of *Ricinus communis* biochar along with ZnO/CuO NPs for the sequestration of chromium from natural water. To remove the Cr contaminated water, adsorption method was used-the most recommended, inexpensive, low energy and less time-consuming method. Adsorption experiments were conducted in batches with varying contact periods, pH, adsorbent dosage, initial metal ion concentration, and temperature. Findings shows that by increasing temperature, dosage and time, the removal percentage of Cr increases in a pattern RCB/CuO NCs>RCB/ZnO NCs>RCB. When compared to earlier investigations, the proposed materials are more efficient and have a higher adsorption capacity. The use of nanoparticle composites for Cr cleanup at the industrial level has been suggested.

**Keywords:** Chromium removal, Wastewater treatment, Zno/CuO nanocomposites, Adsorption, *Ricinus communis* biochar.



## INTRODUCTION

Natural water resources are becoming increasingly intimidating. Thus, the state of the environment has become one of the major problem due to its deterioration. This deterioration is mainly due to urbanization and industrial growth, which in most cases discharges the wastewater into the receiving environment without treatment (Shaukat *et al.*, 2022). These wastewater streams include toxic chemical elements, dyes and inorganic compounds including heavy metals. The accumulation of heavy metals in the environment is a major threat to public health and water quality (Mishra *et al.*, 2019). More generally, heavy metals come from a variety of sources such as industrial activities i.e., plating, mining, tanning, metallurgy and alloying Briffa (2020). The most commonly found toxic heavy metals in industrial wastewater are cadmium, chromium, zinc, lead and copper (Agoro *et al.*, 2020). Heavy metals are not biodegradable and may be carcinogenic (Rahman *et al.*, 2017). Therefore, the presence of these metals in inappropriate amounts in water can cause serious health problems for living organisms. Some of these heavy metals, such as nickel and manganese, are required for life. Cadmium, copper, and lead are classified as dangerous micro pollutants (Essaadaoui *et al.*, 2018), and their toxicity is believed to be severe even at trace levels.

Chromium is one of the most common and important heavy metals (Hussain *et al.*, 2023). It is not naturally found in water (Hameed *et al.*, 2021). However,

chromates and dichromate have been found in a number of industries such as metal plating, tanning, dyeing, printing, and oil and ink plating (Liu *et al.*, 2020). Chromium exists in two oxidation states which are $Cr^{3+}$ and $Cr^{6+}$ (Islam *et al.*, 2022). Hexavalent chromium is a highly toxic, carcinogenic and mutagenic substance that adversely affects human health, which is becoming a matter of public concern and interest to researchers (Toghaire *et al.*, 2020). Chromium is governed by international, national, and municipal legislation when its concentration exceeds legal limits and is deemed hazardous to the environment. A maximum of 100 µg/L has been imposed by the European Union (EU) for total chromium in drinking water under the Drinking Water Directive. The World Health Organization has not developed a distinct guideline value for hexavalent chromium (Munn *et al.*, 2005). Exceptionally, the European Union limits the maximum amount of hexavalent chromium that can be discharged into aquatic environments to 0.5 µg/L, while the WHO has established a provisional guideline value of 0.1 µg/L for the metal in drinking water. The maximum allowable concentration of total chromium in drinking water is 0.1 mg/L, as suggested by the United States Environmental Protection Agency (USEPA) in water quality criteria (2012). These requirements are dependent on chromium solubility, environmental receiver or human intake, and toxicity implications and toxicity implications. Nonetheless, no distinct threshold has been established for the detrimental hexavalent form of chromium. It is imperative to acknowledge that these principles are subject to modification and should be periodically examined and revised.

There is a need to treat chromium contaminated water for public health and environmental safety. Precipitation, adsorption with activated carbon, ion exchange, membrane processes, oxidation, and reduction are common procedures for treating chromium wastewater (Meunier *et al.*, 2006). However, most of these approaches resulted in incomplete metal ion removal, limited selectivity, high operational costs, reagent and energy consumption, and the formation of secondary pollutants (Wu *et al.*, 2004). The advantages of adsorption over conventional methods are numerous and include the capacity to reuse biomaterials, cheap operating costs, metal selectivity, quick operation times, and the absence of chemical sludge (Rahmani *et al.*, 2009).

Adsorption is one of these techniques that is seen to have the most promise because it can immobilize heavy metals even at low concentrations. Heavy metal ions in wastewater can be physically and chemically adsorbed using adsorption techniques that use porous materials or certain functional groups of adsorbents with a large specific surface area. Adsorbents are classified as polymeric, inorganic, or carbon-based and bio-adsorbents based on their chemical structures. In recent years, many synthetic materials have been successfully developed as adsorbents with excellent heavy metal removal performance (Zhao *et al.*, 2019).

The stable carbon-rich substance known as biochar exhibits remarkable potential in managing pollutants found in water and wastewater. Because of its comparatively low cost, relative availability, and comparable sportive characteristics, biochar has recently been suggested as a viable substitute for activated carbon in environmental remediation and water treatment. Because it is so widely available, biochar may be a desirable option for water treatment applications in isolated areas where it is not practical to install commercial adsorbents, such as in bio retention (Inyang and Dickenson 2015). Because of its high surface area and porosity, biochar is a promising adsorbent material with a great affinity for heavy metals (Xia *et al.*, 2021).

In recent study *Ricinus communis* (RC) will be used as biochar to remove Cr contaminated water. *Ricinus communis*, also known as castor, is an annual oilseed plant. It is often referred to as a castor bean, but it is not a real seed. Castor plants grow favorably between 20°C and 25°C while temperatures below 12°C or above 38°C affect germination and yield (Yeboah *et al.*, 2020). Recently, the use of *Ricinus Communis* seeds for the production of castor oil has increased due to their application in beauty products and biofuels (Hilioti *et al.*, 2017). Some recent studies show the adsorption capacity of Magnetite Corn Cob Silica (MCCS) for Cr, only 0.1g of adsorbent has shown effective adsorption of these ions with the maximum adsorption capacity ($q_m$) is 11.1mg $g^{-1}$(Kumari *et al.*, 2018).

Numerous countries use a variety of plants as a source of powerful and effective medicines for therapeutic purposes. There are many instances of R. communis being effective in treating a wide range of illnesses, which is why this scientific research of the plant is of interest (Kubmarawa *et al.*, 2007). To create extracts that will be used to check for bacterial and fungal activity, the Ricinus communis plant's seeds or leaves will be utilized. A batch-scale investigation will look into how several factors, including dosage of the adsorbent, contact time, solution pH, and starting concentration, affect the Cr concentration. Examining RCB/ZnO/CuO NPs' ability to inhibit microbial activity is the goal of this investigation.

## METHODOLOGY

### 3.1 Materials and Chemicals

From Merck, Faisalabad, we purchased Potassium dichromate ($K_2Cr_2O_7$), Sodium chloride (NaCl), Potassium hydroxide (KOH), Zinc sulphate heptahydrate ($ZnSO_4.7H_2O$) and Hydrogen chloride (HCl). Cr stock solution was made using prepared distilled water (Jameel *et al.*, 2024).

### 3.2 Preparation of Adsorbent (Biochar)

The biomass of *Ricinus Communis* (RC) was collected from the local areas. RC was cut into small

pieces by fodder making machine. The smaller RC flakes were washed with double distilled water to remove impurities adhering to the biomass surface. The air-dried material was placed in a biochar unit to prepare RC biochar at 600°C for 90 minutes. After 90 minutes, the biochar unit was turned off and the prepared biochar was allowed to cool (24h), then the biochar was crushed and sieved by fine sieve. The fine material was washed with DI water to remove the soluble components of the biochar and dried again in an oven at 85°C for 12 h. The dry RCB were kept to prepare the mixture, then for additional testing.

### 3.3 Preparation of RCB/ZnONPs composite

To prepare a mixture of RCB with ZnO nanoparticles (RCB/ZnONPs), the precipitation method was used. 0.1M $ZnSO_4$ and 0.2M KOH solutions were prepared and used for RCB/ZnO synthesis. A solution of 1000 ml $ZnSO_4$ was mixed in 0.5g RCB for 20 min, then titrate the mixture with 1000 ml of KOH solution. This process was repeated until 1000 ml of each solution had been titrated according to the aforementioned approach. The RCB/ZnONPs combination was filtered using what man filter paper 42, and the RCB/ZnONPs mixture were oven dried for 24 hours at 85°C. (Imran *et al.*, 2021). Both RCB and RCB/ZnONPs was stored in a plastic container for additional testing to eliminate Cr from polluted water.

### 3.4 Preparation of RCB/CuONPs composites

To prepare a mixture of RCB with CuO nanoparticles (RCB/CuONPs), the precipitation method was used. 0.1M $CuSO_4$ and 0.15M KOH solutions was prepared and used for RCB/CuO synthesis. A solution of 1000 ml $CuSO_4$ were mixed in 0.5g RCB for 20 min, then titrate the mixture with 1000 ml of KOH solution. This process was repeated until 1000 ml of each solution is titrated according to the above procedure. The mixture of RCB/CuONPs was filtered using what man filter paper 42 and the RCB/CuONPs were oven dried for 24h at 85°C. Both RCB and RCB/CuONPs was stored in a plastic container, for further testing to remove Cr contaminated water.

### 3.5 Batch scale study

Cr was dissolved in distilled water in the lab to make a standard solution. Different Cr concentrations were obtained by diluting the standard solution. A variety of large-scale adsorption studies were conducted to remove Cr. The impact of several process factors, including initial metal concentration, adsorbent dosage, solution pH, contact time, and material type, was assessed. The metal solutions were swirled in a 250ml conical flask using a mechanical stirrer at 150 rpm for a set amount of time. The equilibrium time to remove Cr was calculated in the early trials, and 10mL of sample was drawn from each vial at various times, including samples at equilibrium. The sample was stored in a 20ml plastic vial to determine the residual Cr level in the water sample. To determine Cr's maximum absorption wavelength (λmax), a UV-visible spectrometer (PerkinElmer, series 25) was used to scan the solution. Standard Cr solutions were also used to create a calibration curve. The experiment was carried out in duplicate (n=2), and the calibration curve's relationship was employed to convert the sample absorbance to Cr concentration.

#### 3.5.1 Effect of Dosage and Initial Concentration

Effect of initial concentration of Cr was evaluated using different initial concentrations (25, 50, 75, and 100 mg/L) at constant dosage (25mg), pH (5), and contact time (180 min). To determine dosage effect, experiment was carried out at 12.5, 25, and 50mg/Keeping other factors constant.

#### 3.5.2 Effect of Contact time on removal rate

To find the impact of contact time, three flasks with 100ml Cr solution and 25mg dosage of three distinct adsorbents were used at pH 5 (optimum values). The samples of each material were taken after 15, 30, 60, 120, and 180 min of shaking time.

#### 3.5.3 Effect of pH on removal rate

The effect of pH was quantified utilizing Cr contaminated water with different pH. It's kind of is very essential to genuinely consider the kinetic conduct of the adsorbent to fundamentally put off contaminants from the aqueous solution in a subtle way. For all intent purposes find the impact of time, different concentrations of Cr were utilized at a fixed dosage and constant pH.

### 3.6 Data Analysis

The preliminary ($C_o$) and last ($C_f$) concentrations of Cr were used to calculate its elimination capacity (R %) for every adsorbent by the usage of the given equation; (Iqbal *et al.*, 2021)

$$q_e = \frac{(Co-Cf)}{Mb} \times V$$

The preliminary ($C_o$) and ultimate ($C_f$) concentrations of Cr were used to calculate its elimination (R %) through every adsorbent material using given equation;

$$R\% = \frac{Co-Cf}{Co} \times 100$$

## RESULTS AND DISCUSSION

### 4.1 Effect of pH on Cr removal

The pH effect of Cr from wastewater is determine by changing the pH from 3 to 9 while keeping Cr initial level at 100mg/L, dose 25mg, and ambient temperature at equilibrium state. The removal percentage of Cr by RCB is 50.2%, 64.7%, 40.8% and 32.5%. The Cr removal is maximum at 5 pH. The result shows that Cr removal decreases from 64.7 to 32.5% with the increase in pH. The removal percentage of Cr by RCB/ZnONPs is 62.5%, 77.6%, 57.3% and 38.9% on 3pH, 5pH, 7pH and 9pH. This shows that with increase of adsorbent pH, there is decrease in removal of Cr.

While, the removal percentage of Cr by RCB/CuO is 76.7%, 92.1%, 71.7%, 54.3%. Figure shows highest removal of Cr from wastewater on pH 5 with RCB/CuONPs than with RCB/ZnONPs and RCB alone. Numerous adsorbents and treatment materials (e.g., hydroxyl, carboxyl) are utilized to remove chromium from the environment. These functional groups may ionize in an acidic environment, forming sites that are more receptive to positively charged metal ions such as Cr.

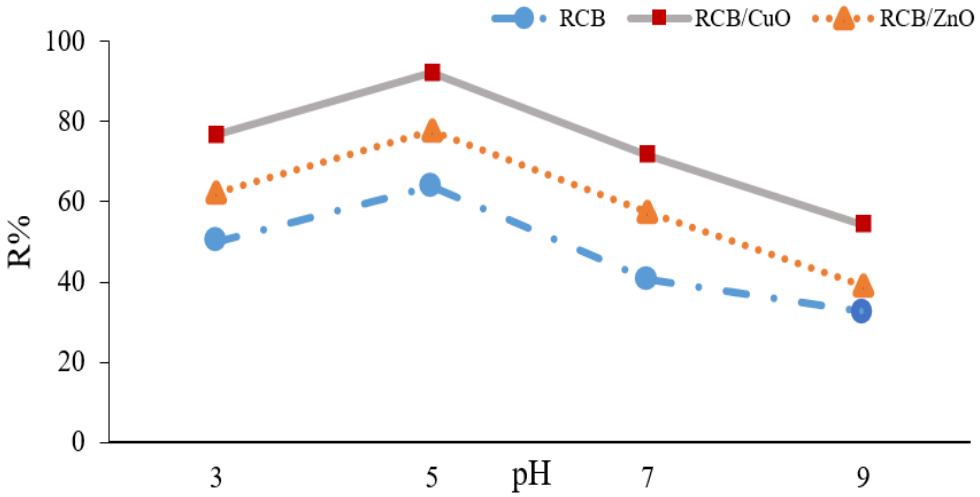


**Figure 4.1: Removal of Cr by RCB, RCB/CuO and RCB/ZnONPs at different pH and constant IC 100 mg/L, dose 25mg and at 180 min**

**4.2 Effect of dose on Cr removal**

The dose effect of Cr from wastewater is determine by changing the dose from 12.5 to 50 mg while keeping Cr initial level at 100mg, pH 5, and ambient temperature at equilibrium state. The removal percentage of Cr by RCB is 65.5%, 74.8% and 80.9% respectively. The Cr removal is maximum at 50mg dose. The result shows that Cr removal increases from 65.5 to 80.9% with the increase in adsorbent dose. The removal percentage of Cr by RCB/ZnONPs is 76.9%, 84.6% and 88.6% respectively on 12.5mg, 25mg, and 50mg dose. This shows that with increase of adsorbent dose, there is an increase in removal of Cr. While, the removal percentage of Cr by RCB/CuO is 91.2%, 94.4% and 95.7%. This is because there are more adsorption sites available for chromium ions to attach to when there is a higher adsorbent. This increase in accessible sites improves the adsorbent's ability to draw out and hold onto more chromium from the water.

Moreover, it is clearly seen that RCB/CuONPs is more effective than RCB/ZnONPS and RCB even at low dosage. The RCB/CuONPs material shows efficient increase in removal (91.2-95.7%) with the increase in adsorbent dose (12.5-50mg) and minor increase (91-94%) is observed with higher adsorbent dose (12.5-25mg). This minor increase may be due to particle interaction such as aggregation.

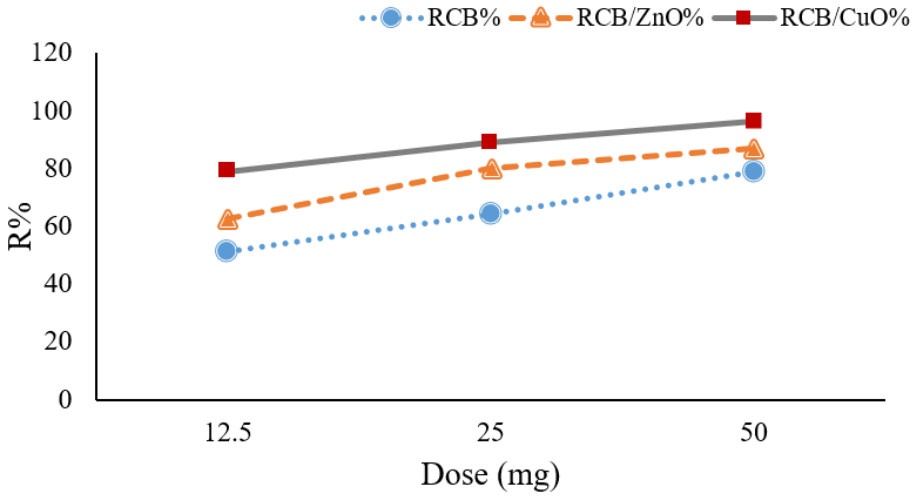

**Figure 4.2: Removal of Cr by RCB, RCB/CuO and RCB/ZnONPs at different dose at IC 100 mg/L, pH 5 and time 180 min**

### 4.3 Effect of contact time on Cr removal

For Chromium removal(Cr), samples were taken at the interval of 15, 30, 60, 120, and 180 minutes and the (R%) through use of RCB is 28.6%, 32.5%, 45.5%, 57.9%, and 68.9% that shows with the increase of time removal can be more effective. The removal percentage of Cr with RCB/ZnONPs is from 33.8 to 73.9%. While, the removal percentage of Cr by RCB/CuONPs is 46.7%, 55.5%, 68.9%, 80.3% and 94.4% respectively on 15min, 30min, 60min, 120min and 180min is observed. It is evident from the results that Cr removal is maximum at high time interval. The explanation for this is that longer contact times provide chromium ions greater chances to diffuse and absorb into the adsorbent material. Higher removal efficiency can be attained by the ions reaching internal active spots in the adsorbent through this diffusion process.

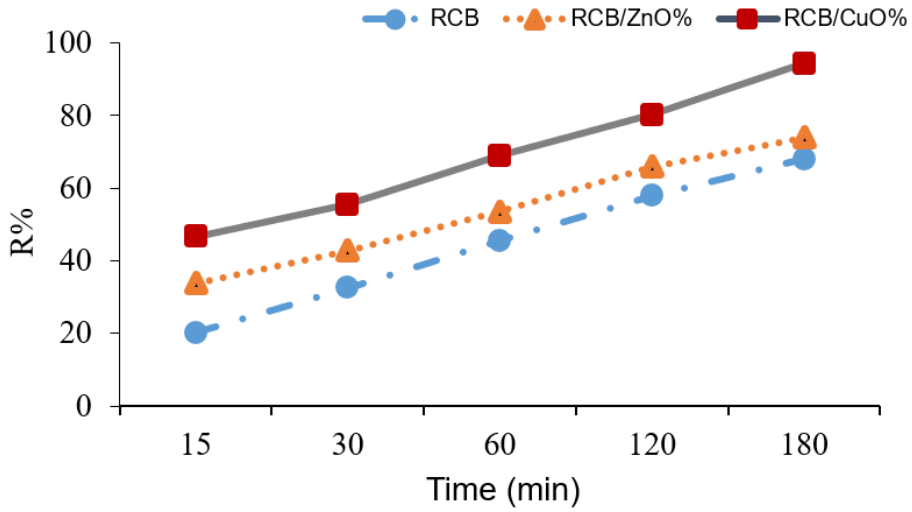


**Figure 4.3: Removal of Cr by RCB, RCB/CuO and RCB/ZnONPs at different contact times and constant IC 100 mg/L, dose 25mg and pH 5**

### 4.4 Effect of Initial concentration on Cr removal

The effect of IC was determined by varying solution IC from 25- 100 mg, while all other factors were kept constant such as pH (5), dose (25mg) and time (180 mins). The removal percentage of Cr by RCB was 64.2%, 72.7%, 50.8% and 38.7% respectively. The result showed that Cr removal is maximum at 50ppm concentration while the removal efficiency of RCB decreased with increasing IC. However, the removal percentage of Cr by RCB/CuONPs was 86.4%, 94.8%, 75.7%, and 66.8% respectively when IC was changed 25-100ppm of Cr.

The data clearly show that the elimination of Cr reaches its maximum at low IC and diminishes with increasing starting concentration. There may be more competing ions in the solution as a result of high starting chromium concentrations. The removal material's binding sites may be contested by these ions, which would lessen the material's ability to remove chromium. Under the same testing conditions, RCB/CuONPs generally had a higher adsorption potential for Cr than RCB and RCB/ZnONPs. It could be because there are less active sites on the surface of RCBs than there are on RCB/CuONPs. There is a small change in Cr removal by RCB/CuONPs due to its higher potential when IC level varying from 25 to 50mg/L while at higher concentration (100mg/L) there is a clear decline in Cr removal.

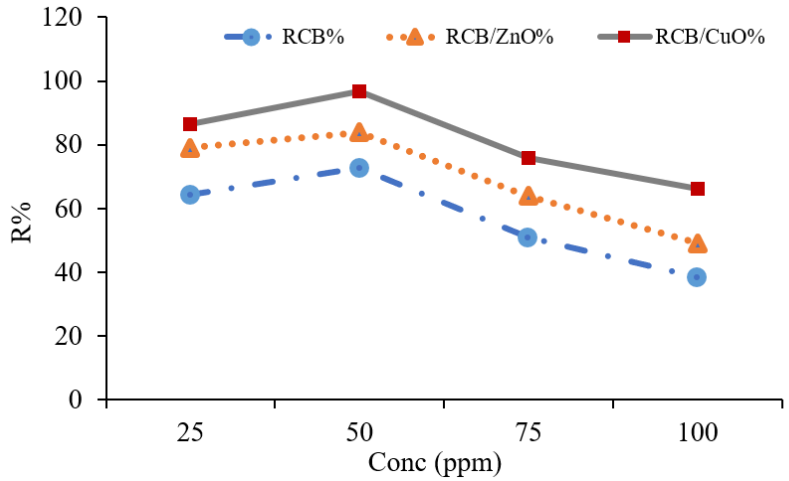


**Figure 4.4: Removal of Cr by RCB, RCB/CuO and RCB/ZnONPs at different ICs at constant pH 5, dose 25mg and 180 min**

**4.5 Effect of temperature on Cr removal**

By increasing temperature there is an increase in Cr removal. Highest removal of Cr was observed at 45℃ by using RCB/CuONPS than with RCB/ZnONPs and RCB alone. At 25℃ the RCB shows 40.1%, at 35 ℃ shows 52.8% and at 45℃ shows 68.9 5% Cr removal. RCB/ZnONPs shows Cr removal from 50.2% to 78.1% while changing temperature from 25 to 45℃. Moreover, RCB/CuONPs shows highest Cr removal from 67.8 to 95.8 among RCB and RCB/ZnONPs. The kinetic energy of molecules, particularly those engaged in the adsorption process, may generally be increased by greater temperatures. The adsorbent material may be able to absorb chromium ions more effectively due to faster adsorption kinetics brought on by this increased energy.

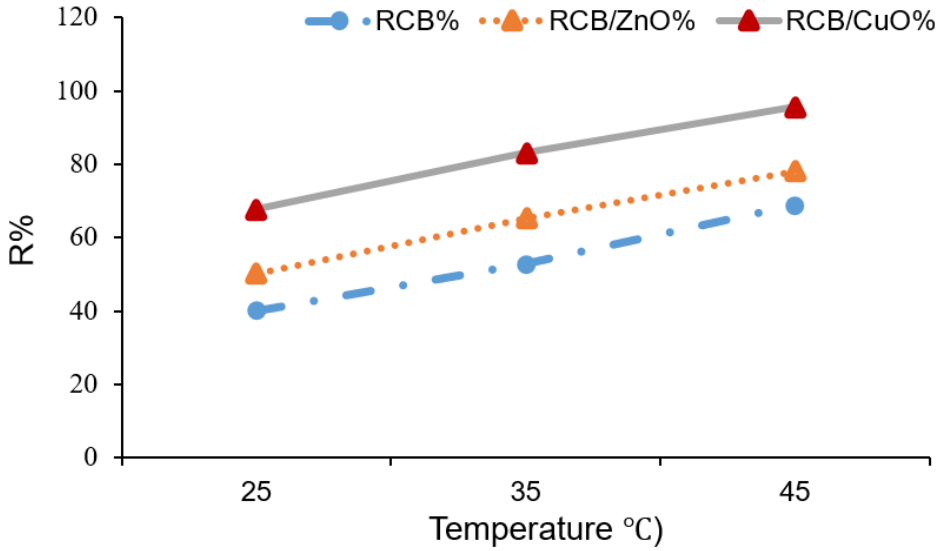


**Figure 4.5: Removal of Cr by RCB, RCB/CuO and RCB/ZnONPs at different temperatures, at constant pH 5, dose 25mg and 180 min**

**4.6 Comparative % Removal of Cr by different biochar at constant parameters**

The % removal by the sorbents is an important parameter to conclude the efficiency of a sorbent. The comparative % removal of different sorbents is presented in fig. 4.6, keeping all parameters constant. The biochar, ZnONPs, CuONPs, RCB/ZnO and RCB/CuO shows a % removal of 45. 68, 76, 82, and 94%, respectively. Highest removal is observed by using CuO nanocomposites with *Ricinus Communis* biochar. This is because variations in their surface characteristics, such as surface charge and functional groups, may give CuO NPs a greater affinity for chromium ions. Furthermore, CuO NPs' unique crystal structure and makeup might offer additional active sites for chromium adsorption. Because copper's electrical structure may promote a stronger interaction with chromium ions, it may also contribute to the enhanced removal.

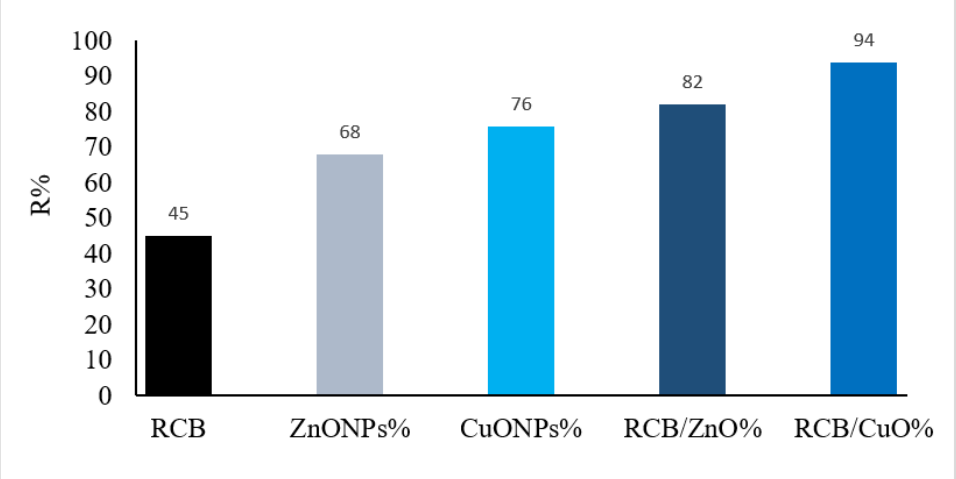


**Figure 4.6: Comparative Cr removal by using BC, ZnONPs, CuONPs, RCB/ZnONCs and RCB/CuONCs**

## 4.12 DISCUSSION

Water contamination is a worldwide problem which leads to many diseases in animals, plants and in humans. It is highly documented that anthropogenic and industrial action are the key sources of surface and ground water contamination all over the world. From industries like leather, textile, smelting etc. chemicals are directly discharges into water bodies which potentially affect water quality. In this study it was noticed that the removal percentage of Cr was significantly decreased with the increasing concentration of IC by keeping other factors constant. At low IC there are abundance of active sites on adsorbent surface, as IC increases, active sites decrease in number which cause reduction in removal of Cr from wastewater. This finding matches with the study of Wenling Yang (2022). According to his research, the Cr(VI) removal efficiency by biochar tends to gradually increase and then decrease, while the adsorption amount continues to increase. Indeed, when the Cr(VI) concentration is low, the adsorption sites of biochar are relatively complete and have stronger adsorption capacity. As the Cr(VI) concentration increased, the amount of Cr(VI) exposed to the biochar surface increased, leading to higher adsorption efficiency and adsorption amount. Because the adsorbent has a limited number of adsorption sites and the concentration of Cr(VI) in the aqueous solution increases, the competition for available adsorption sites of Cr(VI) becomes increasingly fierce. Therefore, the removal efficiency decreases (Nasseh *et al.*, 2021).

Regarding the ability of different metal ions to be biosynthesized, the pH value of the solution plays a major role in the biosorption process. The transport, surface charge, and binding characteristics of metal ions and metals after creation are all impacted by pH changes. By adjusting the pH, the charge on the biosorbent surface can be preserved. As the charge on the surface of the biosorbent changes, the properties of the metal ions and metalloids also change. Therefore, changes in pH have an important impact on chromium adsorption and removal. In this study we observe that at acidic pH, the chromium removal from synthetic water is maximum. As we increase pH from 7-9 the removal efficiency of RCB/CuO and RCB/ZnO decreases. At pH 5, the maximum adsorption value was recorded. Neutralization was the primary cause of this maximal adsorption. The surplus of hydrogen ions (H+) neutralizes the hydroxyl ions (OH−), facilitating the passage of dichromate ions and their adsorption on the surface of the biochar. Because the chromium ions are negatively charged and the surface of the biochar adsorbent is positively charged at acidic pH, a pH of 5 is sufficient to interact with the chromium ions. However, as the pH rises over 5, extra $OH^-$ ions cause the concentration of hydroxyl ions to increase, and chromium ions in solution and the surface of biochar will reject one another as a result (Khalil *et al.*, 2020).

The impact of dose on Cr removal is clearly observed. Both have a direct relation. As we increase dose from 12.5-50mg, the removal percentage of Cr is increases, matches with the studies of Imran *et al.*, 2023. According to his research, the removal of Cr was assessed at various adsorbent doses (0.5–2 g) while maintaining a Cr concentration of 50 mg/L at pH 3.0. The findings showed that the dosage concentration significantly influences the removal of Cr, and for all adsorbents, increasing the adsorbent dosage (0.5–2.0 g) increased Cr removal linearly. As we see in this research that by increasing contact time the removal efficiency of RCB, RCB/ZnO and RCB/CuONPs increases. At initial time of the Cr removal was not evident but at 120-180min the above three used material shows maximum removal of 66-94.4%. These findings match with the literature review. It was discovered that the contact time affected the adsorption of Cr(VI) from solutions (20 to 120 mg/L) onto NWP, AWP, and CWP. Similar results were also reported by (Bansal *et al.*, 2022) for Cr(VI) adsorption as a function of time in their study18. This may be due to depletion of active sites for a constant biosorbent dose at equilibrium.

Temperature has a clear effect on the amount of chromium adsorbed on biochar. To study the effect of temperature on Cr removal rate, experiments were performed at different temperatures, i.e. 25, 35 and 45°C using the optimal adsorbent dose (25 mg) with the initial concentration was 100 ppm and the pH was 5. The above results show that the ability to remove Cr increases as the temperature increases. These results are consistent with those of Gorzin *et al.*, (2018), the removal rate of Cr(VI) from aqueous solution was 65.08% at 25ºC, increasing to 76.73% at 45ºC. The removal efficiency increased with increasing temperature, showing that the Cr(VI) adsorption process is endothermic.

## CONCLUSION

In conclusion, the complete examination of biosorption, by Ricinus communis and its composites with ZnO and CuO revealed excellent prospects for tackling chromium contamination. The combination of bio-based materials and metal oxides shows significant promise for successful water treatment. This work not only improves our understanding of sustainable ways to water remediation, but it also emphasizes the importance of continued research and practical use of these results to reduce the health concerns associated with chromium exposure. Overall, the results reported herein provide useful insights to the profession and argue for the continuous development and application of environmentally friendly solutions in water treatment procedures. Both biochar and its composites with CuO and ZnO nanoparticles are efficient for sequestration of Cr from waste water.

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