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Using Iron/Nickel Coated Sand Nanocomposites Prepared by Eucalyptus Leaf Extract for Copper Removal from Aqueous Solutions

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ABSTRACT

As heavy metals are commonly found in water bodies today, the need for an efficient method to remove these pollutants simultaneously has become increasingly important. This research involved coating sand with Fe/Ni nanoparticles created through an environmentally friendly method using eucalyptus leaf extract. Green synthesis of Fe/Ni coated sand was characterized using Fourier transform-infrared spectrometry (FT-IR) analysis. The resulting Fe/Ni nanocomposite coated sand was then used to copper (Cu(II)) removal from water in batch experiments. The study examined five factors namely pH of solution, Cu(II) concentration, contact time, Fe/Ni coated sand dosage, and agitation speed that affected on the Cu(II) removal efficiency. Optimal values of this factors were found to be 200 rpm, pH 6, 130 minutes, 50 mg/L for Cu(II), and 0.5 g/50 mL Fe/Ni coated sand. The findings revealed that over 80% of Cu(II) was removed until the fifth cycle. This work could open up new possibilities for treating water contaminated with copper ions using eco-friendly composites made from waste.

Keywords: green synthesis, nano-composite, Cu(II) removal, adsorption, environmentally friendly method.

INTRODUCTION

Heavy metal water pollution has emerged as a significant global concern due to their presence in drinking water and wastewater surpassing safety limits in many countries (Zamora-Ledezma, et al., 2021). The release of heavy metals into aquatic environments poses a severe toxicological risk to human health, ecosystems, and agriculture (Zaynab, et al., 2022; Priti and Paul, 2016). Activities like mining, smelting, and copper ore processing leave behind residues that contaminate the environment with copper, leading to various health issues and developmental abnormalities in humans (Abd Ali, 2021). The most prevalent type of environmental contamination globally involves a mix of organic compounds and heavy metals, underscoring the critical need for efficient methods to eliminate these pollutants and safeguard a clean environment for human well-being (Lin et al., 2019; Qin, et al., 2018).

Adsorption is a crucial technique to clean heavy metal pollutants from water systems. This method is favored for handling new types of pollutants because it's cost-effective, easy to use, can regenerate the sorbent, and has minimal environmental impact (Saleh and Ali, 2018). The use of nano zero-valent iron (nZVI) processes is widespread for removing various environmental contaminants, including heavy metals (Gopal, et al., 2022). However, the effectiveness of nZVI can diminish because of the improvement of an oxide layer on its surface when exposed to air or during reactions (Weng, et al., 2014). To tackle these issues, incorporating a secondary catalytic metal like Pd, Cu, Ni, or Pt can be beneficial. Bimetallic compounds centered on nanoscale iron have proven effective in decontaminating diverse substances such as phenol (Gautam, et al., 2015). Although less studied compared to Pd and Ni, Cu has been sporadically used as a eco-friendly and cost-effective metal for catalytic coating. In the present study, Ni was selected as the secondary catalytic metal to develop a bimetallic nZVI system. Recent research has shown that combining Fe-Ni can effectively break down a range of substances, including metals, as demonstrated by studies from Kaduu et al., (2017) and Dongi et al., (2018b). Scientists have used bimetallic iron/nickel nanoparticles to remove triclosan (TCS) and copper (Cu(II)), as reported by Lin, (2019) and Gopal et al., (2022). The efficiency of these hybrid bimetallic Fe/Ni nanoparticles was tested for extracting heavy metals simultaneously from water solutions by Xue et al., (2021). However, these experiments often involved the use of harmful chemicals like borohydride during nanoparticle production, which could increase environmental risks. One potential solution to this challenge might involve producing nanoparticles from waste materials that contain the necessary organic components. Eucalyptus leaves, known for their rich variety of bioactive compounds, could be a promising source for this purpose.

The main goal of this paper is to create a new adsorbent capable of extracting Cu(II) from water solutions. Previous literature lacks any mention of using Ni with nZVI supported on sand to create Fe/Ni coated sand through an environmentally friendly approach for Cu(II) removal. To achieve this, the study focused on: (1) developing the Fe/Ni coated sand nanocomposite using a green method with Eucalyptus leaf extract, aiming to reduce waste and eliminate heavy metal, (2) assessing the nanocomposite's effectiveness in Cu(II) removal, (3) conducting reusability tests for up to five cycles to validate its efficiency and practicality. The green synthesis of Iron-nickel Nanocomposites coated sand and their application as a sorbent in this study represents a significant step in the creation of new, efficient adsorbents, and cost-effective.

MATERIALS AND METHODS

Preparation of contaminated aqueous solutions

To create a Cu(II) synthetic solution with (1,000 mg/L) concentration, copper nitrate trihydrate (1 g) were dissolved in distilled water (1 L). Adjusting the contaminant concentration was achieved through dilution.

Green synthesis of Fe/Ni coated sand

Extracting Eucalyptus Leaves with Ethanol: After collecting the Eucalyptus leaves, they were dried for 40 hours in an oven at 60 °C. The resulting dried powder was then mixed with 100% ethanol (15%) in a shaking incubator for 24 hours. Following filtration, the liquid obtained was preserved at 20 °C for future applications. This extraction technique was based on research conducted by Hwang et al., (2011), Ravikumar (2019), Ravikumar et al., (2020), and Rocha et al., (2024).

Figure 1 illustrates the process of creating environmentally friendly Fe/Ni-coated sand. To start, 100 grams of pure sand were placed in a bottle with a screw cap, and 150 mL of $FeCl_3$ solution with 0.1 M was slowly added. The bottle was then placed on an orbital shaker for 30 minutes to ensure thorough mixing of the $FeCl_3$ solution

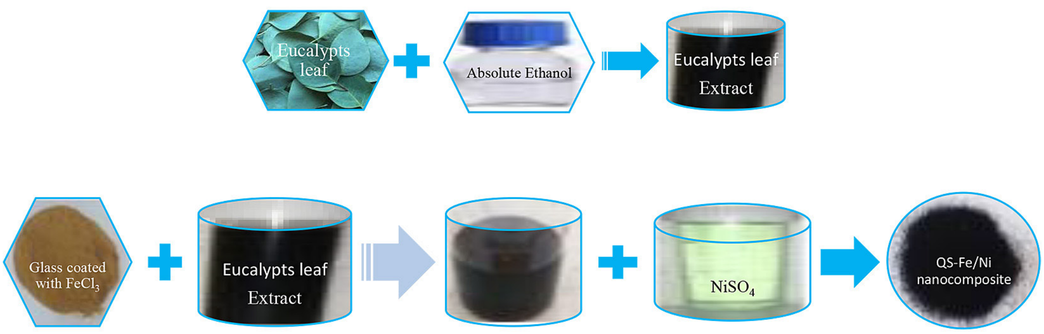

Figure 1. Preparation of Fe/Ni nanocomposite coated sand

with the sand. The Fe-coated sand was then dried overnight in a vacuum oven at 80 °C. Next, the dried $FeCl_3$-coated sand was mixed slowly with a 10% ethanolic extract of Eucalyptus leaf 29 °C. As the process advanced, the color of the mixture changed from golden yellow to black (Gopal, et al., 2020). Once the entire Eucalyptus leaf extract was added, the solution underwent another 30 minutes of shaking on the orbital shaker at room temperature to guarantee thorough mixing. CMC (0.1%) was introduced to stabilize NZVI, followed by an additional hour of shaking after the introduction of a 0.1 M $NiSO_4$ solution (150 mL). Finally, the FeNi-coated sand was filtered, washed three times with absolute ethanol, and dried in a vacuum oven at 80 °C.

Fe/Ni coated sand characterization

FTIR analysis

Fourier Transform Infrared Spectroscopy (FTIR) is seen as a direct method for studying how substances are absorbed, by pinpointing the specific functional groups that are involved in binding the solute (Chen et al., 2008). The infrared spectra were recorded in the range of (4000 – 400) 1/cm, provide the adsorption of copper by Fe/Ni Nanocomposite Coated Sand.

RESULT AND DISCUSSION

Impact of operational conditions

Contact time

In this test, the duration of interaction from 0 to 200 minutes under specific experimental conditions shown in Figure 2. The findings revealed that the peak removal rate (80%) was predominantly reached at 130 minutes for Cu(II). The notable rise in copper removal percentage as time progressed is attributed to the active composite sites designated for copper sorption. Nevertheless, as these locations ran out, the rate of absorption decreased, especially after 130 minutes for Cu(II) (Osman, 2020).

Effect of pH solution on Cu(II) removal

Considering the pH of the water environment is crucial for studying the Cu(II) removal. To do this, tests need to be conducted within specific pH ranges, typically between 3 and 7, using the parameters illustrated in Figure 3. Observations show that the removal percentage of copper conc. increased as the initial pH value rose from 3 to 6, reaching an optimal 80% removal rate. However, beyond pH 6, the removal percentage declined (Liu, et al., 2020).

Effect of agitation speed

Figure 4 shows that initially, around 10% of containant was removed without agitation. The removal efficiency increased with higher speeds, reaching 80% at 200 rpm. However, further increasing the speed to 250 rpm did not significantly improve removal, as shown in Figure 6C. Therefore, 200 rpm was determined as the most effective stirring speed for subsequent tests. This outcome can be explained by the fact that higher agitation speeds enhance the contaminants distribution on the sorbent surface (Fe/Fe coated sand), facilitating better contact between the active sites and sorbate solution, leading to successful sorbent sites (Abd Ali, 2021).

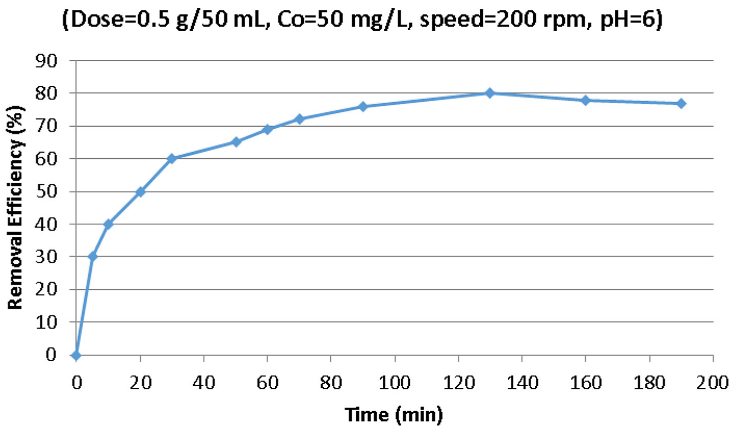

Figure 2. Removal percent of Cu(II) adsorption with contact time

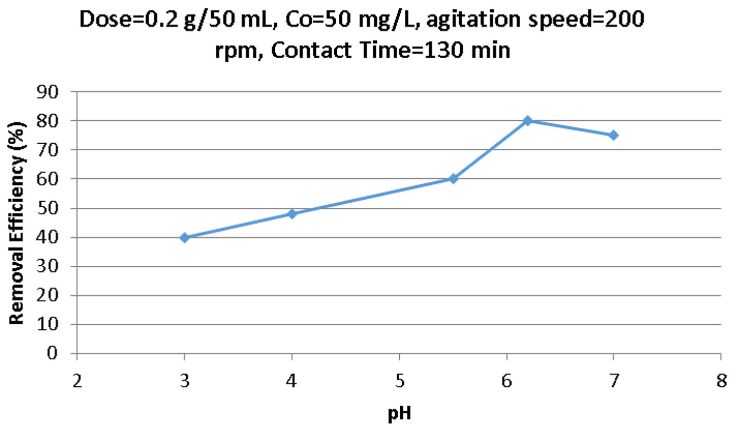

Figure 3. Removal percent of Cu(II) adsorption with pH

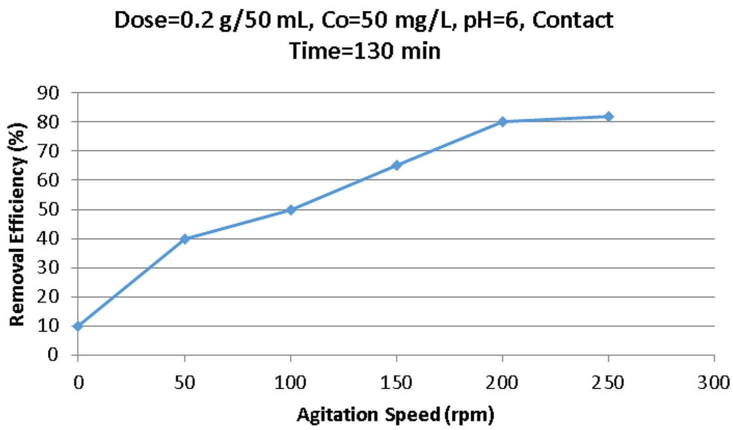

Figure 4. Removal percent of Cu(II) adsorption with agitation speed

Effect of initial concentration

In additional tests, different initial concentrations of C0 ranging (50–250) mg/L were examined under the conditions that show in Figure 5. The graph clearly illustrates a significant decline in the removal of copper with increasing initial concentration (C0). This decrease in removal percent was primarily caused by the saturation of the adsorbent's sites with the pollutants' molecules (Abd Ali, et al., 2020; Ezzat, et al., 2022). The study's findings revealed that the lowest dosage tested (50 mg/L) yielded the highest removal percentage.

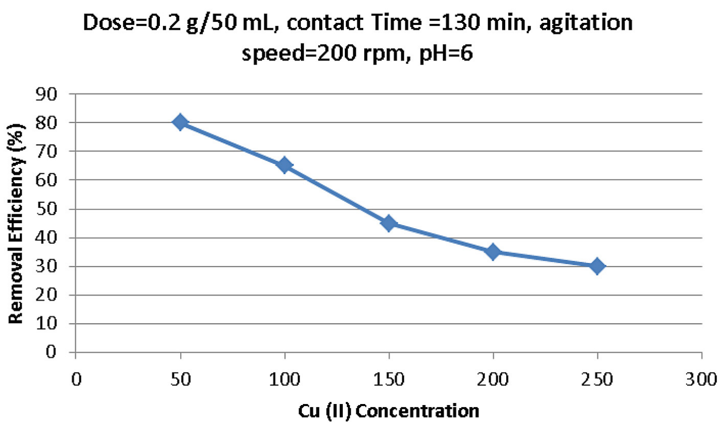

Figure 5. Removal efficiency of Cu(II) adsorption with copper concentration

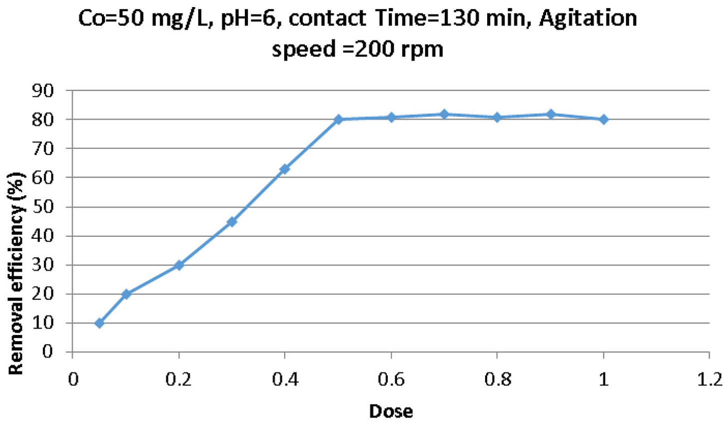

Figure 6. Removal percent of Cu(II) adsorption with Fe/Ni coated sand dosage

Effect of Fe/Ni coated sand dosage

This study examined how the amount of Fe/Ni coated sand dosage used affects efficiency of Cu(II) removing. Researchers utilized a composite material in amounts ranging (0.05 to 1) g per 50 ml. Each quantity was added and thoroughly mixed with a solution containing water-based pollutants, following the procedures outlined in Figure 6. The outcomes demonstrated that the use of 0.5 g per 50 mL of Fe/Ni coated sand lead to in the successful removal of 80% of Cu(II). Surprisingly, increasing the amount beyond 0.5 g per 50 mL did not notably affect the removal efficiency. This was due to the pollutant concentration in the water phase stabilizing. These results are consistent with prior research by Kango and Kumar, (2016), Jin, Yang, et al., (2019), and Ravikumar, et al., (2020).

CONCLUSION

A composite of Fe/Ni coated sand was created using an environmentally friendly method, avoiding any harmful chemicals that could impact the environment negatively during the Fe/Ni reduction process. This composite demonstrated the ability to remove copper through batch processes. The study focused on various factors affecting Cu(II) removal, such as agitation speed, initial pH, contact time, the concentration of copper, and the Fe/Ni coated sand dosage. Optimal values of this factors were found to be 200 rpm, pH 6, 130 minutes, 50 mg/L for Cu(II), and 0.5 g/50 mL Fe/Ni coated sand, resulting in an 80% removal rate. The research aimed at creating a Fe/Ni coated sand nanocomposite through an eco-friendly approach using Eucalyptus leaf extract. This process was designed to minimize waste and remove heavy metals. The eco-friendly production of Iron-nickel Nanocomposites coated sand as a sorbent marks a crucial advancement in developing novel, efficient, and affordable adsorbents.

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