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Eco-Friendly Synthesis of Iron Oxide Nanoparticles Using Mulberry Leaves Extract for Heavy Metal Removal from Water

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ABSTRACT. Various natural and anthropogenic factors, including inadequate water treatment, urbanisation, industrialisation, and the intensification of agricultural output, can result in the presence of heavy metals in water, the atmosphere, and sediments. Heavy metal pollution poses a severe threat to aquatic ecosystems and human health due to its persistence, bioaccumulation, and toxicity. Zinc ions (Zn^{2+}), though essential in trace amounts, becomes hazardous at elevated concentrations. This study reports the green synthesis of magnetite iron oxide nanoparticles (Fe_3O_4-NPs) using mulberry leaf (Morus alba) extract as a natural reducing and stabilizing agent. The nanoparticles were tested for Zn^{2+} removal efficiency under varying conditions of initial concentration (0.5–3.0 mg/L), pH (4–9), contact time (15–90 min), and adsorbent dosage (0.4–1.4 g). Optimal conditions were observed at 1.0 mg/L Zn^{2+}, pH 9, 75 minutes contact time, and 1.0 g adsorbent dosage, achieving a removal efficiency of 98%. Adsorption data fitted well to the Langmuir isotherm ($R^2 = 0.86$), indicating monolayer adsorption on a homogeneous surface, while kinetics followed the pseudo-second-order model ($R^2 = 0.95$), suggesting chemisorption. These findings highlight the potential of mulberry leaf-derived Fe_3O_4-NPs as an eco-friendly, cost-effective adsorbent for heavy metal remediation, supporting sustainable water treatment strategies.

Keywords: Green synthesis, Iron oxide nanoparticles, Mulberry leaf extract, Zinc removal, Adsorption isotherms and kinetics

1. INTRODUCTION

The sustainability of the environment and human health depend on having access to clean water. The occurrence of heavy metals in aquatic systems, the atmosphere, and sediments can be attributed to a combination of natural processes and human activities, such as insufficient water treatment, rapid urbanisation, industrial development, and intensified agricultural practices. A metal or metalloid is referred to as a "heavy metal" if its atomic density is greater than 4 g cm^{-3} (Rouibah et al., 2023).The existence of heavy metals in aquatic systems, including copper (Cu), zinc (Zn), cobalt (Co), nickel (Ni), chromium (Cr), and lead (Pb) have has grown to be a significant worldwide concern (Rouibah et al., 2023). Despite being a necessary trace element, excessive concentrations of Zn in water bodies, which are frequently brought on by industrial processes like mining, metal plating, and battery production, can have harmful effects on human health, including nausea, vomiting, and long-term organ damage. The World Health Organisation (WHO) states that 3 mg/L of zinc is the highest amount that can be present in drinking water.

Numerous low-cost biosorbents based on agricultural waste have been reported for the removal of heavy metals. These do not require any harsh physical or chemical treatment because they only need very basic pre-treatments like washing, drying, grinding, or light acid or alkali treatment (Imran-shaukat et al., 2022). There are several methods

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that have been developed to remove heavy metals from water, including chemical precipitation, ion exchange, membrane filtration, and electrochemical treatment (Nasir et al., 2024). Even though these methods work well, they frequently come with a high cost of operation, complicated infrastructure, and the potential to produce secondary waste like chemical sludge, which can be difficult to disposal. This has led to an increase in demand for economical, environmentally friendly, and sustainable alternatives.

Nanotechnology has introduced novel possibilities in environmental remediation, particularly via the application of nanoparticles for water purification. Due to their high surface area, magnetic properties, increased surface reactivity, adsorption capacity, catalytic efficiency, and lower toxicity compared to other metal nanoparticles (Revathy et al., 2023), iron oxide nanoparticles (Fe_3O_4-NPs) have garnered a lot of attention. With sizes ranging from 1 to 100 nm, nanoparticles are highly valuable due to their special qualities (Kaur & Sidhu, 2021; Sebastian et al., 2018). On the other hand, the traditional synthesis of Fe_3O_4-NPs usually requires high energy input or toxic reducing agents, which goes against the green synthesis principles.

Green synthesis is an eco-friendly method of generating nanoparticles that makes use of natural biological resources such as plant extracts. Plant extract-based nanoparticle synthesis has proven to be economical and has increased the range of non-toxic nanoparticle production options (Nasir et al., 2024; Selvaraj et al., 2022). Plants are "chemical factories," producing a vast array of secondary compounds that can be employed as redox mediators and nanoparticles stabilisers. It has been reported that nanoparticles produced using plant products or extracts are more stable and synthesised rapidly than conventional techniques because green approaches are environmentally friendly, cost-effective, simple , easy to carry out, and do not involve any toxic agents (Bibi et al., 2019). These plant-based methods eliminate the need for harmful substances and increase the sustainability of the process by employing phytochemicals like tannins, phenolics, and flavonoids (Revathy et al., 2023) as reducing and stabilising agents while the nanoparticles are being formed. For large-scale nanoparticle production, green synthesis is an economical, environmentally benign, non-toxic, pollution-free, and more sustainable method (Revathy et al., 2023).

The leaves of the mulberry plant (*Morus alba*) are abundant in bioactive substances that can facilitate the synthesis of nanoparticles. Mulberry leaves contain polyphenols. Polyphenol can act as both a reducing and capping agent in the synthesis of iron-based nanoparticles. Mulberry leaf polyphenols may aid in the reducing and stabilizing of iron salts green iron nanoparticles (Nie Lim et al., 2018). Using this natural resource to produce nanoparticles contributes to the goals of sustainable development and gives agricultural waste an alternative use. A few studies have specifically looked at the removal of Zn using iron oxide nanoparticles derived from mulberry leaf extract. By synthesising Fe_3O_4-NPs using an aqueous extract of mulberry leaves and assessing their effectiveness in extracting zinc ions from tainted water through batch adsorption experiments, this study is focused on closing this gap.

2. METHODOLOGY

2.1. Apparatus and Instruments

The following equipment was used in this research study which are Uv-Vis spectrometer, electronic balance, oven, beaker, test tube, magnetic stirrer, filter paper, measuring cylinder, mixing cylinder, pH-meter, spatula, Erlenmeyer flask, volumetric flask, fritted filter, separation funnel, pipette, micro pipette, sample cell, retort stand, dish plate, tray, mortar and pestle, microwave, dropper and bar magnet.

2.2. Chemicals and Reagents

The chemicals and reagents used include ferric chloride ($FeCl_3$), hydrochloric acid (HCl), sodium hydroxide (NaOH), sulfuric acid (H_2SO_4), zinc sulphate heptahydrate ($ZnSO_4 \cdot 7H_2O$), zinc reagent, and cyclohexane solution. Mulberry leaf was collected from the surrounding area.

2.3. Preparation of the Mulberry Leaf (ML) for Extraction

Mulberry leaf extraction (MLE) was carried out following a previously reported method with slight modifications. Fresh leaves from mature Morus species trees, which are widely grown in Malaysia under favourable climatic conditions, were collected and thoroughly washed with tap water to remove dust and surface impurities. The cleaned leaves were then oven-dried at 105°C for 9 hours. Once dried, the leaves were finely ground and sieved through a 150 µm mesh. Coarse particles that did not pass through the sieve were reground with a mortar and pestle until a uniform powder was obtained. For extraction, 10 g of ML powder was mixed with 500 mL of distilled water, based on the procedure of with modifications (Ghoohestani et al., 2024). The suspension was continuously stirred at 700 rpm and maintained at 70°C for 24 hours.

2.4. Synthesis of Iron Oxide Nanoparticles (Fe_3O_4-Nps)

A mixed solution of $FeCl_3 \cdot 6H_2O$ (0.004 mol) and $FeSO_4 \cdot 7H_2O$ (0.002 mol) in a 1:2 molar ratio was prepared in 200 mL of deionized water and heated to 80°C for 15 minutes under continuous stirring at 150 rpm. Afterward, 10 mL of mulberry leaf extract was introduced into the solution, followed by the gradual addition of 40 mL of 1 M sodium hydroxide (NaOH) under vigorous stirring at 700 rpm for 60 minutes at 80°C. Throughout the process, the solution pH was carefully maintained between 10 and 11 (Ghoohestani et al., 2024). After cooling, the mixture was washed several times with ethyl alcohol and then dried in an oven at 90°C for 12 hours

2.5. Preparation of Zinc (Zn) Solution

A stock solution of Zn (1000 mg/L) were prepared by dissolving predetermined amounts of $ZnSO_4 \cdot 7H_2O$ in distilled water (Tahiruddin et al., 2023). The stock solutions were thoroughly mixed using a stirrer and working solutions of varying concentrations were obtained by serial dilution. For each adsorption experiment, 200 mL of the required concentration (ranging from 0.5 to 3.0 mg/L in 0.5 mg/L increments) was prepared freshly in conical flasks, with the volumes measured precisely using a micropipette.

2.6. Batch adsorption experiment

Batch adsorption experiments were performed under agitation at 200 rpm to investigate the effects of initial concentration (0.5–3.0 mg/L Zn with 2000 mg adsorbent, 60 min contact time), solution pH (4–9 adjusted with HCl or NaOH), contact time (15–90 min at the optimal pH), and adsorbent dosage (2000–7000 mg/L Fe_3O_4-NPs at a fixed

Zn concentration). The removal efficiency (%) and adsorption capacity q_e (mg/g) of the adsorbent were determined using Eqs. (1) and (2):

$$Removal\ (\%) = \left(\frac{C_o - C_e}{C_o}\right) \times 100 \tag{1}$$

$$q_e = \left(\frac{C_o - C_e}{M}\right) \times V \tag{2}$$

where C_o (mg/L) is the initial concentration of the adsorbate, C_e (mg/L) is the equilibrium concentration (Tahiruddin et al., 2023).

3. RESULTS AND DISCUSSION

3.1. Effect of Zn^{2+} Concentration

The effect of initial Zn^{2+} concentration showed that maximum removal efficiency (85%) was obtained at 1.0 mg/L (Figure 1). This outcome was also supported by a prior study that found that at lower concentrations, around 80% of the zinc was removed (Ahmadi & Izanloo, 2023). The reason for this can be due to the abundance of available active sites on the Fe_3O_4-NPs allowed effective interaction with Zn^{2+} ions, resulting in high adsorption performance (Tahiruddin et al., 2023). However, as the concentration increased beyond 1.0 mg/L, the removal efficiency declined due to the progressive saturation of active sites and intensified competition among Zn^{2+} ions for limited binding positions on the nanoparticle surface (Tahiruddin et al., 2023). According to this result, retention is negatively affected at high concentrations since there are more Zn^{2+} ions than sorption sites on the support surface (Rouibah et al., 2023).

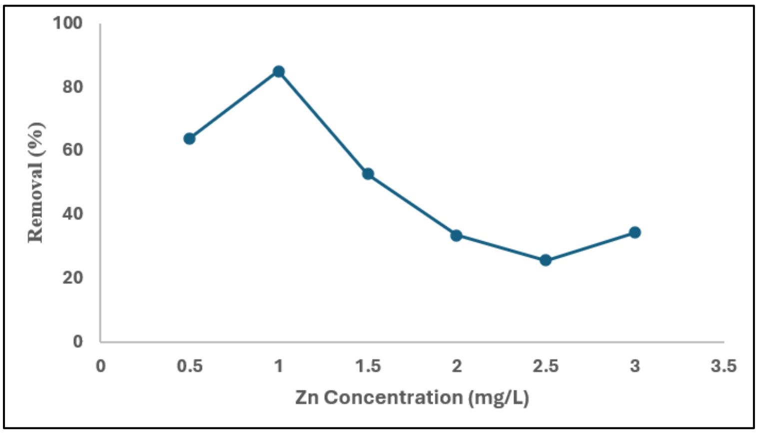

Figure 1. Variation of percentage removal with Zn^{2+} concentration.

3.2. Effect of pH

The adsorption efficiency was highly dependent on pH, increasing steadily as the solution became more alkaline and peaking at 98% at pH 9 as seen in Figure 2. Adsorption was suppressed at lower pH levels because excess hydrogen ions (H^+) competed with zinc ions for active binding sites on the surface of Fe_3O_4-NPs. In addition, ions of hydrogen and hydroxide may compete with metal cations for adsorption. A slight reduction in efficiency was observed between pH 6 and pH 7, which can be attributed to competition between hydroxide ions (OH^-) and Zn^{2+} ions for active adsorption sites. (Ahmadi & Izanloo, 2023) demonstrated that the optimal condition for Zn^{2+} ion removal was achieved under alkaline conditions at pH 7. In line with a study by (Dolgormaa et al., 2018), super-paramagnetic iron oxide nanoparticles (SPIONs) were found to be effective at removing zinc at this pH of 6. Consistent with previous findings, this study confirmed that alkaline conditions favour Zn^{2+} removal. As the pH increased, the surface functional groups' deprotonation increased the nanoparticles' negative charge, which strengthened their electrostatic attraction to Zn^{2+} ions and improved their removal efficiency. Therefore, the adsorbent surface valency is therefore influenced by pH (Ahmadi & Izanloo, 2023).

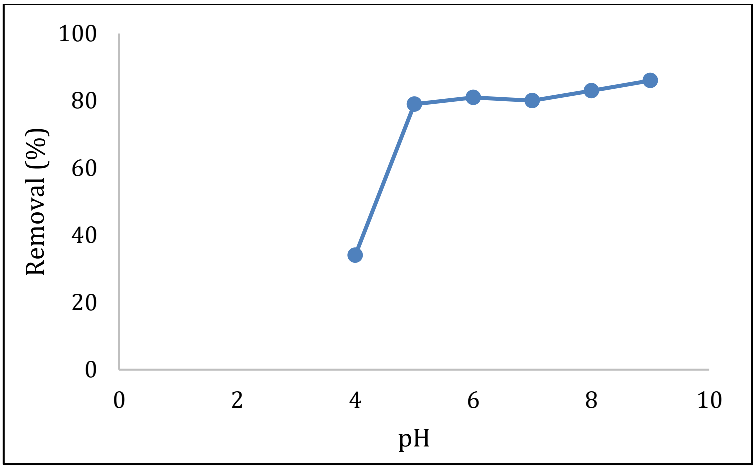

Figure 2. Variation of percentage Zn^{2+} removal with pH of solution.

3.3. Effect of contact time

Equilibrium contacts time plays a crucial role in wastewater treatment as it determines the adsorption rate and ensures both effectiveness and cost efficiency. In this study, the effect of contact time on Zn^{2+} removal by Fe_3O_4-NPs was investigated over a range of 15–90 minutes, using 0.4 g of adsorbent at an initial concentration of 1.0 mg/L and pH 9. Results indicated that approximately 50% of Zn^{2+} ions were removed within the first 15 minutes, increasing to 59% at 30 minutes and 78% at 45 minutes, before approaching equilibrium (Figure 3).

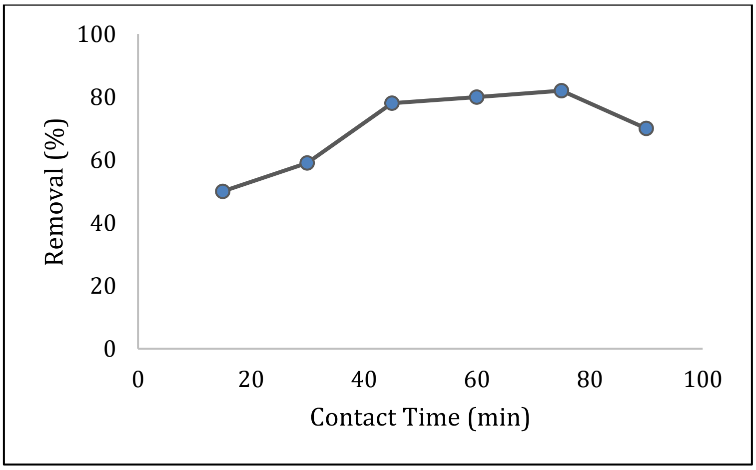

Figure 3. Variation of percentage Zn^{2+} removal with contact time.

The maximum removal efficiency of 82% was achieved at 75 minutes, after which a slight decline was observed. During the initial rapid phase, the abundance of vacant binding sites creates a steep concentration gradient between the adsorbate in solution and the adsorbent surface, driving fast adsorption (Narwal et al., 2025). As adsorption progresses, these sites gradually become occupied, reducing the concentration gradient and leading to a slower adsorption rate (Ahmadi & Izanloo, 2023; Tahiruddin et al., 2023).

3.4. Effect of Fe_3O_4-NPs dosage

In this study, the effects of adsorbent dosages ranging from 2000 mg/L to 7000 mg/L on the removal of Zn^{2+} ions were assessed while maintaining constant conditions. Although it slightly decreased to 95%, a high removal efficiency of 96% was noted at the lowest dosage of 2000 mg/L. With removal performance improving with increasing dosage, efficiencies reached 97% at 4000 mg/L and up to 98% at 5000 mg/L. The removal rate rose as the quantity of adsorbent dosage grew because there were more adsorption sites available, which gave metal ions more surface area to react with and be collected by Fe_3O_4-NPs (Narwal et al., 2025). Nevertheless, efficiency dropped to 96% and then 95% after increasing above 5000 mg/L as seen in Figure 4. This was probably caused by unsaturated adsorption sites and particle aggregation, which reduce the effective surface area for adsorption. A notable increase in adsorption occurs when additional active sites are available for Zn^{2+} ions binding. Similar to the findings of (Ahmadi & Izanloo, 2023), when the adsorbent concentration exceeds 0.3 g/L, the adsorption efficiency does not increase further and instead begins to decline. However, once a substantial portion of Zn^{2+} ions has been removed and the active sites approach saturation, further increases in adsorbent dosage exert minimal influence on the overall adsorption process (Imran-shaukat et al., 2022; Nasir et al., 2024).

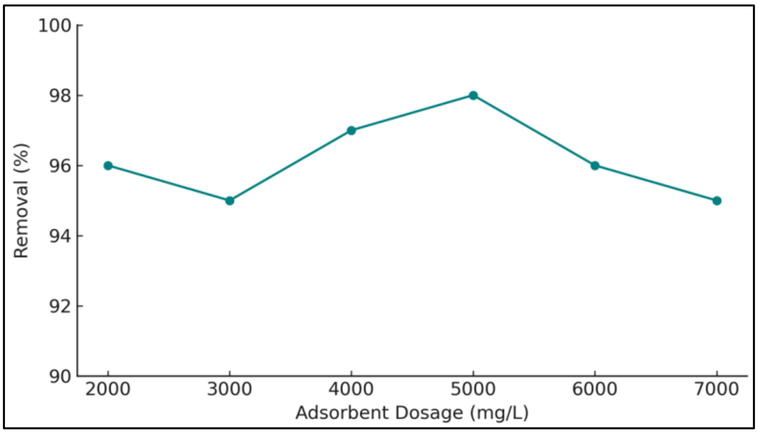

Figure 4. Effect of adsorbent dosage on Zn^{2+} removal.

3.5. Adsorption isotherm studies

3.5.1 Langmuir Isotherm

The adsorption data for Zn were evaluated using the Langmuir isotherm model, which assumes a uniform adsorbent surface with energetically identical active sites. The Langmuir plot, describing the relationship between the equilibrium concentration of Zn^{2+} ions (C_e) and the ratio of C_e to the adsorption capacity (q_e), yielded a linear equation with an R^2 value of 0.86.

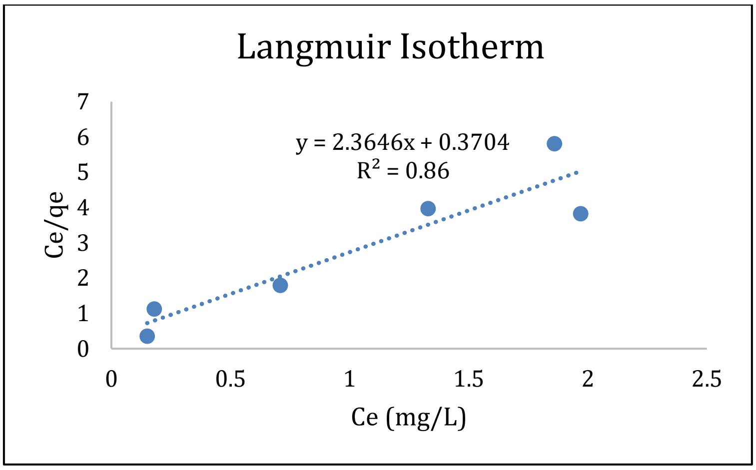

Figure 5. Adsorption Langmuir isothermal curves for Zn adsorption.

This strong correlation (Figure 5) indicates that the adsorption process followed the Langmuir model, confirming monolayer adsorption on a homogeneous surface. The calculated separation factor R_L was 0.86, suggesting that the adsorption was favourable and that Fe_3O_4-NPs possessed a high affinity for Zn^{2+} ions. These results demonstrate the strong potential of Fe_3O_4-NPs as an efficient adsorbent for Zn removal from aqueous solutions. The Langmuir model states that monolayer adsorption occurs on a uniform surface with few adsorption sites and little interaction between the molecules that are adsorbed (Tahiruddin et al., 2023).

3.5.2 Adsorption Kinetic Model

Form Figure 6, the pseudo-second-order adsorption model was a better equation to represent the adsorption kinetics of Zn^{2+} ions onto the Fe_3O_4-NPs. The adsorption mechanism of Zn^{2+} ions onto Fe_3O_4-NPs was evaluated using pseudo-first-order and pseudo-second-order kinetic models.

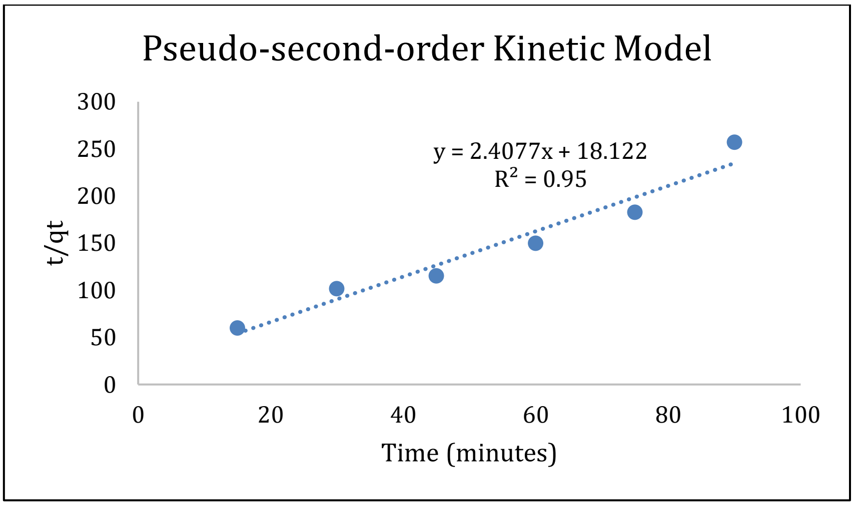

Figure 6. Adsorption Langmuir isothermal curves for Zn^{2+} adsorption.

Adsorption of Zn^{2+} ions by Fe_3O_4-NPs was a multi-step process that involved sorption on the external surface and diffusion into the interior of the adsorbent, according to the best correlation for the system given by the pseudo-second-order model. Furthermore, the excellent fit of the pseudo-second-order model indicated that the Fe_3O_4-NPs and Zn^{2+} ions would share or exchange electrons, resulting in chemical sorption involving valency forces (Sharma & Srivastava, 2010). According to previous study, the "pseudo-second order" kinetic model and the "Langmuir" isotherm model both suit the adsorption process well for the removal of Zn^{2+} ions (Mohamed et al., 2023).

4. CONCLUSION

Mulberry leaf extract was successfully used to synthesize magnetic Fe_3O_4-NPs via a green route. The nanoparticles showed excellent potential for Zn^{2+} ions removal, achieving 98% efficiency at optimal conditions (1.0 mg/L zinc concentration, pH 9, 75 min, 5000 mg/L dose). Adsorption followed the Langmuir isotherm and pseudo-second-order kinetics, confirming monolayer chemisorption. This eco-friendly approach provides a promising pathway for sustainable wastewater treatment. Future research should focus on nanoparticle regeneration, large-scale application, and extending the method to other heavy metals and mixed-contaminant systems.

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AUTHOR CONTRIBUTIONS

Nur Shafieza Aizzan: conceptualisation, investigation, methodology, data analysis and writing; **Nur Husnina Athirah Baharudin**: methodology and data analysis. All authors have read and agreed to the published version of the manuscript.

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COMPETING INTEREST

The authors declare that there are no competing interests.

COMPLIANCE OF ETHICAL STANDARDS

Not applicable

SUPPLEMENTARY MATERIAL

Not applicable.

REFERENCES

Ahmadi, S., & Izanloo, C. (2023a). Biosynthesis of iron oxide nanoparticles at different temperatures and its application for the removal of Zinc by plant mediated nanoparticle, as an eco-friendly nanoadsorbent. *Results in*

Chemistry, *6*, 101192. https://doi.org/10.1016/j.rechem.2023.101192

Bibi, I., Nazar, N., Ata, S., Sultan, M., Ali, A., Abbas, A., Jilani, K., Kamal, S., Sarim, F. M., Khan, M. I., Jalal, F., & Iqbal, M. (2019). Green synthesis of iron oxide nanoparticles using pomegranate seeds extract and photocatalytic activity evaluation for the degradation of textile dye. *Journal of Materials Research and Technology*, *8*(6), 6115–6124. https://doi.org/10.1016/j.jmrt.2019.10.006

Dolgormaa, A., Lv, C. jiang, Li, Y., Yang, J., Yang, J. xing, Chen, P., Wang, H. peng, & Huang, J. (2018). Adsorption of Cu(II) and Zn(II) ions from aqueous solution by gel/PVA-modified super-paramagnetic iron oxide nanoparticles. *Molecules*, *23*(11), 2982. https://doi.org/10.3390/molecules23112982

Ghoohestani, E., Samari, F., Homaei, A., & Yosuefinejad, S. (2024). A facile strategy for preparation of Fe_3O_4 magnetic nanoparticles using *Cordia myxa* leaf extract and investigating its adsorption activity in dye removal. *Scientific Reports*, *14*(1), 84. https://doi.org/10.1038/s41598-023-50550-1

Imran-shaukat, M., Wahi, R., & Ngaini, Z. (2022). Bioresource Technology Reports The application of agricultural wastes for heavy metals adsorption : A meta-analysis of recent studies. *Bioresource Technology Reports*, *17*, 100902. https://doi.org/10.1016/j.biteb.2021.100902

Koul, B., Bhat, N., Abubakar, M., Mishra, M., & Arukha, A. P. (2022). Application of natural coagulants in water treatment :A sustainable alternative to chemicals. *Water*, *14*, 3751. https://doi.org/10.3390/w14223751

Mohamed, A., Atta, R. R., Kotp, A. A., Abo El-Ela, F. I., Abd El-Raheem, H., Farghali, A., Alkhalifah, D. H. M., Hozzein, W. N., & Mahmoud, R. (2023). Green synthesis and characterization of iron oxide nanoparticles for the removal of heavy metals (Cd^{2+} and Ni^{2+}) from aqueous solutions with Antimicrobial Investigation. *Scientific Reports*, *13*, 7227. https://doi.org/10.1038/s41598-023-31704-7

Narwal, N., Katyal, D., & Bathi, J. R. (2025). Green synthesis of iron oxide nanoparticles using *Momordica charantia*: Kinetics of removal of heavy metal and microplastic pollutants. *Regional Studies in Marine Science*, *86*, 104189. https://doi.org/10.1016/j.rsma.2025.104189

Nasir, G., Batool, F., Noreen, S., Gondal, H. Y., Mustaqeem, M., Saeed, Z., Gul, Y., Ur Rehman, F., & Ali, H. M. (2024). Biosynthesis of calcium oxide nanoparticles by employing Mulberry (*Morus nigra*) leaf extract as an efficient source for Rhodamine B remediation. *Scientific Reports*, *14*, 23744. https://doi.org/10.1038/s41598-024-71172-1

Nie Lim, S., Ming Ng, W., Kang Lim, J., & Xin Che, H. (2018). Performance of mulberry leaves mediated green synthesis zero-valent iron nanoparticles in dye removal. *International Journal of Engineering & Technology*, *7*(3.36), 113. https://doi.org/10.14419/ijet.v7i3.36.29089

Revathy, R., Sajini, T., Augustine, C., & Joseph, N. (2023). Iron-based magnetic nanomaterials: Sustainable approaches of synthesis and applications. *Results in Engineering*, *18*, 101114. https://doi.org/10.1016/j.rineng.2023.101114

Rouibah, K., Ferkous, H., Delimi, A., Himeur, T., Benamira, M., Zighed, M., Darwish, A. S., Lemaoui, T., Yadav, K. K., Bhutto, J. K., Ahmad, A., Chaiprapat, S., & Benguerba, Y. (2023). Biosorption of zinc (II) from synthetic wastewater by using Inula Viscosa leaves as a low-cost biosorbent: Experimental and molecular modeling studies. *Journal of Environmental Management*, *326*, 116742. https://doi.org/10.1016/j.jenvman.2022.116742

Selvaraj, R., Pai, S., Vinayagam, R., Varadavenkatesan, T., Kumar, P. S., Duc, P. A., & Rangasamy, G. (2022). A recent update on green synthesized iron and iron oxide nanoparticles for environmental applications. *Chemosphere*, *308*, 136331. https://doi.org/10.1016/j.chemosphere.2022.136331

Sharma, Y. C., & Srivastava, V. (2010). Separation of Ni(II) ions from aqueous solutions by magnetic nanoparticles. *Journal of Chemical and Engineering Data*, *55*(3), 1441–1442. https://doi.org/10.1021/je900619d

Tahiruddin, N. S. M., Aziz, R. A., Ali, R., & Taib, N. I. (2023). Potential of using jackfruit peel (*Artocarpus heterophyllus*) as green solution for removal of copper (II) and zinc (II) from aqueous solution: Adsorption kinetics, isotherm and thermodynamic studies. *Journal of Environmental Chemical Engineering*, *11*(3), 109953. https://doi.org/10.1016/j.jece.2023.109953