

Hybrid Ionic Liquid-assisted Biomagnetic Nanocomposite for Efficient Removal of a Cationic Dye

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Editor's note: Conventional methods for dye removal, such as adsorption, have several limitations, including low efficiency, high costs, and the risk of secondary pollution. This has led to significant research interest in developing sustainable and innovative materials for dye absorption. Thati et al. utilized coconut shells as a green resource to create biomagnetic nanocomposites that demonstrate high efficiency in removing cationic dyes.

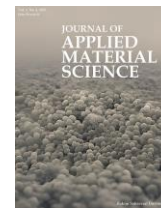
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Hybrid Ionic Liquid-assisted Biomagnetic Nanocomposite for Efficient Removal of a Cationic Dye

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Abstract

The presence of organic toxic dyes in the aqueous environment poses a potential threat to the ecosystems. Ionic liquids (ILs), typically considered green solvents, confer diverse benefits and versatility in applications. Magnetite biochar composites and ILs can be combined to form novel hybrid nanocomposites, providing collaborative advantages, preserving features of individual components, and attaining innovative performances. This study aims to fabricate an IL-assisted biomagnetic hybrid nanocomposite material, analyse its physiochemical characteristics, and evaluate its efficiency in the adsorptive removal of toxic rhodamine blue (RhB) dye in aqueous solution. The mean diameter of the IL-assisted biomagnetic nanocomposite material was found to be 45 ± 6 nm. The removal efficiency was optimised using various parameters such as pH, contact time, as well as adsorbent and adsorbate concentrations. The bionanocomposite demonstrated 97.5% of RhB removal percentage and 135 mg/g of adsorption capacity within 45 min and showed remarkable reusability of up to five cycles. The hybrid IL-assisted biomagnetic nanocomposite prepared in this work offers a viable and sustainable approach to removing industrial synthetic dyes in contaminated aquatic ecosystems.

Keywords: Ionic liquids; Biomagnetic nanocomposites; Adsorption; Rhodamine blue dye; Sustainability.

1. Introduction

The indiscriminate release of industrial dyes, such as Rhodamine blue (RhB), into water bodies has become a major environmental concern [1, 2]. RhB dye is not only

aesthetically pleasing but also toxic, carcinogenic, and mutagenic, posing severe threats to aquatic life and human health. Conventional dye removal methods, including adsorption, photocatalysis, and biodegradation, often suffer from limitations such as low efficiency, high cost, and secondary pollution [3-6].

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In recent years, magnetite iron oxide (Fe_3O_4) biochar nanocomposites have emerged as promising adsorbents for dye removal due to their high surface area, magnetic separability, and eco-friendly nature [7-9]. However, their adsorption capacity and selectivity can be further improved by incorporating hybrid ionic liquids (ILs), which can enhance the surface properties and functionality of the nanocomposites [10-13].

This study focuses on the design and synthesis of hybrid ionic liquid-assisted magnetite biochar nanocomposites for the efficient removal of Rhodamine blue dye from contaminated water.

2. Experimental

Coconut shells were cleaned, ground, oven dried, and calcined to 700°C . The calcined powder was then activated with KOH and neutralised with 0.5M HCl. Ferrous and ferric salts were dissolved in a small volume

of strong and dilute acid, and NaOH was added dropwise with sonication for 2h. The resulting precipitate was magnetically separated, washed, and dried. Magnetite nanoparticles and activated biochar were combined in a 1:2 ratio, followed by the addition of 0.5g of the ionic liquid 1-ethyl-3-methylimidazolium tetrafluoroborate. The mixture was then sonicated in diethyl ether at 50°C for 12 hours. The solid was washed, dried, and labelled as ILMBC.

ILMBC was subjected to different characterization techniques such as FESEM, FTIR, and XRD to assess its morphology, functional groups, and chemical surface character. Batch experiments were performed to investigate the adsorption of RhB dye on ILMBC. In these experiments, 50 mL of RhB dye solutions with concentrations of 50-200 mg/L were treated with ILMBC doses of 0.03 and 0.05 g at pH 8, with contact times ranging from 15-45 minutes. The initial and final concentration of RhB dye was determined using UV-Vis

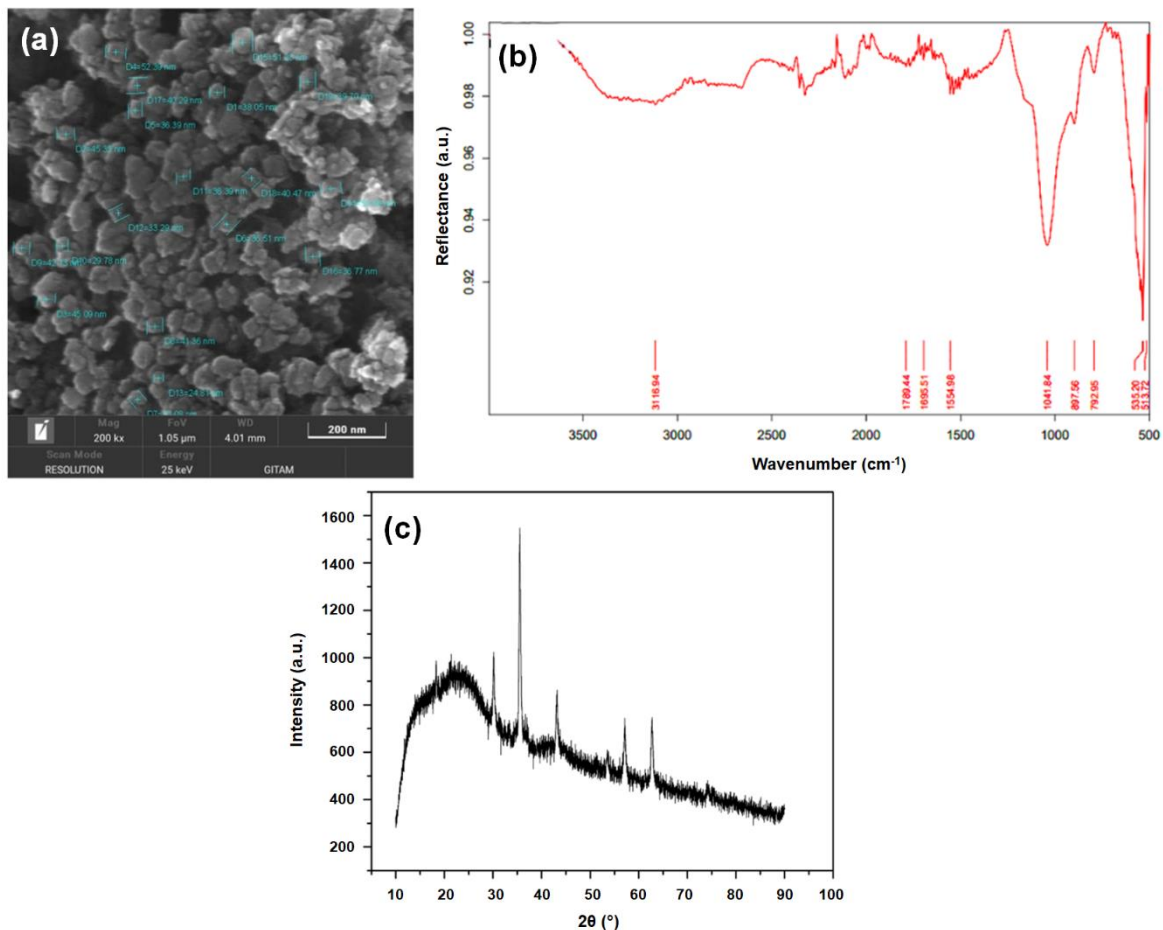


Figure 1. Representative analysis of ILMBC: (a) FESEM, (b) FTIR, and (c) XRD.

spectrophotometry at a wavelength of 554 nm. The adsorption efficiency of ILMBC for the removal of RhB dye was calculated using equation (1):

$$\% = \frac{C_0 - C_e}{C_0} \times 100 \quad (1)$$

where C_0 and C_e are the initial and equilibrium concentrations of RHB dye.

3. Results and discussion

FESEM analysis, presented in Figure 1a, revealed a porous surface topology, characterized by spherical particles with an average diameter of 45 nm. Upon high-magnification imaging, the presence of voids and pores within the magnetite biochar structure became evident, suggesting a high surface area. FTIR spectroscopy results in Figure 1b revealed the functional groups present in the IL-based magnetite coconut shell biochar composite. The spectrum showed distinct bands, including hydroxyl (3116 cm^{-1}) and carboxyl (1789 cm^{-1}) groups, C=O stretching vibrations (1695 cm^{-1}), and C=C

stretching vibrations (1554 cm^{-1}). The presence of IL was confirmed by the observed peaks for B-F stretching vibrations (1041 cm^{-1}) and aromatic C-H out-of-plane bending vibrations (792 cm^{-1}). Fe-O stretching vibrations ($535\text{-}513 \text{ cm}^{-1}$) indicated the magnetized nature of the material, confirming the effective immobilization of IL onto the surface of Fe_3O_4 and biochar. The X-ray diffraction (XRD) pattern of the ILMBC hybrid nanocomposite in Figure 1c exhibited a characteristic diffraction peak at a 2θ value of 35.52° corresponding to the (311) planes of magnetite. The absence of sharp IL peaks and iron oxide diffraction peaks indicates the IL's amorphous nature and its effective capping of magnetite, preventing oxidation. The average particle size was approximately 45 nm, as calculated by Debye-Scherrer's equation.

The adsorption efficiency of ILMBC was evaluated using the surface plot in Figure 2a, the contour plot in Figure 2b, and the linear form of the Langmuir adsorption isotherm (equation 2) in Figure 2c.

$$\frac{C_e}{q_e} = \frac{1}{Q_0} C_e + \frac{1}{Q_0 b} \quad (2)$$

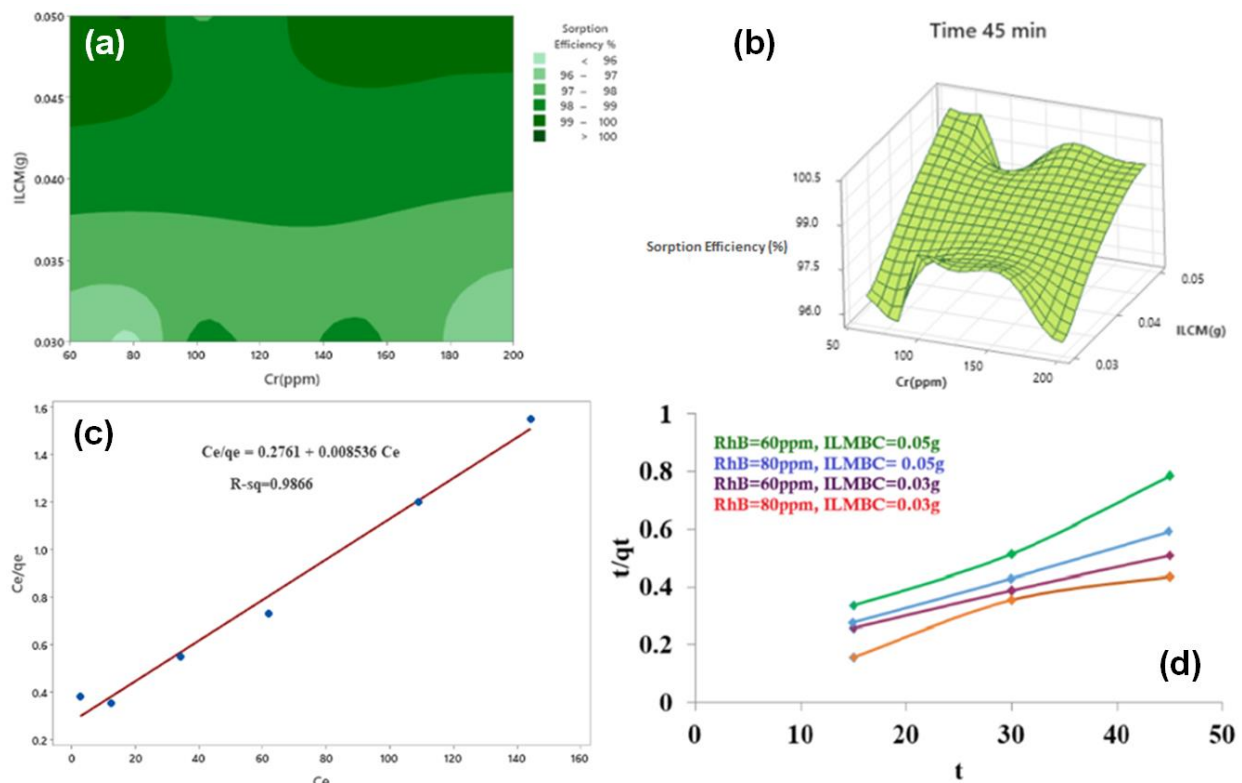


Figure 2. (a) Surface plot and (b) Contour plot of ILMBC for sorption efficiency at 45 min, (c) Langmuir adsorption isotherm, as well as (d) Pseudo second order kinetics results.

Table 1. Comparison of maximum RhB dye adsorption capacities (mg/g) of various adsorbents

Adsorbent	Adsorption capacity (mg/g)	Reference
Furfural residue	37.93	[14]
Coconut coir	14.9	[15]
Sodium montmorillonite	38.27	[16]
Coffee grounds	7.51	[17]
Natural zeolite Clinoptilolite	2.81	[18]
Sulphuric Acid-modified fly ash	36.36	[19]
Casuarina equisetifolia needle	82.34	[20]
Artocarpus heterophyllus	103.92	[21]
ILMBC	135	This study

In this equation, C_e , q_e , Q_0 , and b are the equilibrium concentration (mg/l), adsorption capacity of adsorbent (mg/g), maximum adsorption capacity, and Langmuir adsorption constant (L/mg), respectively. The maximum removal and adsorption capacities of ILMBC were found to be 97.5% and 135 mg/g, respectively.

The adsorption rate was determined by pseudo-second order kinetics to assess the adsorption ability of ILMBC according to equation 3 (Figure 2d). The correlation coefficient was greater than 0.9, suggesting good adsorption fits.

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (3)$$

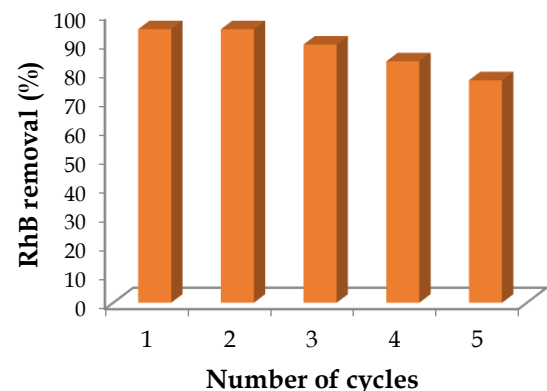
Table 1 provides a comparison between the maximum adsorption capacity (mg/g) of ILMBC nanocomposite in removing RhB dye with different reported adsorbents in the literature. In particular, the ILMBC nanocomposite shows a higher adsorption capacity than most polymeric and bioadsorbents, surpassing natural and synthetic ones.

The magnetic property of ILMBC allows easy regeneration and reusability of the adsorbent. After adsorption, recovery of ILMBC can be easily achieved by employing an external magnet, elution with methanol, and reuse for the next RhB adsorption experiment. Importantly, the adsorbent showed great reusability and retained its performance up to 5 cycles in 50 mL solution containing 100 mg/L RhB solution, as shown in Figure 3

The ILMBC nanocomposite is very promising for the treatment of wastewater because it possesses superior adsorption capability for RhB dye removal. Nonetheless, there are apprehensions about the environmental and biological hazards that the application of ionic liquids

(ILs) and Fe_3O_4 nanoparticles may pose. These include persistence, toxicity, and bioaccumulation in aquatic organisms and microorganisms. In order to combat these risks, future studies will aim to study biodegradability, toxicity, and disposal practices, surface modification, and extensive toxicity testing.

There are various challenges that must be overcome, ranging from technical ones like scaling up synthesis and use, stability, and regeneration of the nanocomposite. There is also the need for regulatory compliance and risk evaluation, as well as economic factors such as cost-effectiveness and industrial scale-up. Through recognition and solution of these challenges, we can ensure safe development and utilization of ILMBC nanocomposite for wastewater treatment and ultimately enhance sustainable use and reduce possible risks.

**Figure 3.** Regeneration efficiency of ILMBC against removal percentage up to 5 cycles.

4. Conclusions

In summary, this research effectively proved the synthesis and application of a new magnetic hybrid nanocomposite, ILMBC, for effective removal of RhB dye from water solutions. The ILMBC showed great adsorption capacity, fast kinetics, and outstanding reusability for several cycles. The magnetic nature of the composite facilitated easy regeneration and separation, and it is a promising candidate for wastewater treatment processes. These results underscore the promise of ILMBC as a cost-effective and environmentally friendly dye removal method and open new avenues for further development and research.

Conflict of Interest


The authors declare no conflict of interest.

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Graphical Abstract

