

Application of Silica-Coated Magnetic Nanoparticles in the Treatment of Dye-Contaminated Water

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Abstract:

The treatment of wastewater contaminated with industrial dyes remains a significant environmental challenge due to the high chemical stability and low biodegradability of these pollutants. This study aimed to synthesize and evaluate silica-coated magnetic nanoparticles functionalized with amino groups for the efficient removal of Chicago Sky Blue dye from aqueous media. The hybrid nanomaterial, Fe₃O₄@SiO₂-Epoxy-NH₂, was successfully prepared via the sol-gel method and characterized using Fourier Transform Infrared Spectroscopy (FTIR) and Field Emission Scanning Electron Microscopy (FESEM). The influence of key operational parameters, including solution pH, adsorbent dosage, contact time, and initial dye concentration, was systematically investigated. The highest removal efficiency (~90%) was achieved under optimized conditions of pH 5, adsorbent dosage of 40 mg, contact time of 40 min, and initial dye concentration of 40 mg/L. The enhanced adsorption performance is attributed to the synergistic effect of the magnetic core, silica shell, and surface amino-functionalized active sites, which promote strong electrostatic interactions and hydrogen bonding with dye molecules. These findings demonstrate that the synthesized hybrid material is a promising and reusable adsorbent for the efficient treatment of dye-contaminated industrial wastewater.

Keywords: Magnetic nanoparticles, silica, sol-gel, adsorption, dye removal, wastewater treatment.

المخلص

لا تزال معالجة مياه الصرف الصحي الملوثة بالأصبغ الصناعية تمثل تحديًا بيئيًا كبيرًا، وذلك بسبب الثبات الكيميائي العالي لهذه الملوثات وقابليتها المنخفضة للتحلل الحيوي. هدفت هذه الدراسة إلى تحضير وتقييم جسيمات نانوية مغناطيسية مغطاة بالسيليكا ومعدلة بمجموعات الأمينو، بهدف إزالة الفعالة لصبغة Chicago Sky Blue من الأوساط المائية. تم تحضير المادة النانوية الهجينة Fe₃O₄@SiO₂-Epoxy-NH₂ بنجاح باستخدام طريقة السول-جل (Sol-Gel)، كما تم توصيفها باستخدام مطيافية الأشعة تحت الحمراء بتحويل فورييه (FTIR) والمجهر الإلكتروني الماسح بانبعث المجال (FESEM). تمت دراسة تأثير مجموعة من العوامل التشغيلية الأساسية بصورة منهجية، شملت درجة حموضة المحلول (pH)، وجرعة المادة المازة (Adsorbent Dosage)، وزمن التلامس (Contact Time)، والتركيز الابتدائي للصبغة (Initial Dye Concentration). وقد تحققت أعلى كفاءة لإزالة الصبغة، والتي بلغت نحو 90%، تحت الظروف المثلى المتمثلة في pH = 5، وجرعة مادة مازة قدرها 40 mg، وزمن تلامس 40 دقيقة، وتركيز ابتدائي للصبغة قدره 40 mg/L. ويُعزى الأداء المحسّن لعملية الامتزاز إلى التأثير التآزري بين النواة المغناطيسية، وغلاف السيليكا، والمواقع الفعالة على السطح والمعدلة بمجموعات الأمينو، والتي تعمل على تعزيز التفعلات الكهروستاتيكية والروابط الهيدروجينية مع جزيئات الصبغة. وتُظهر هذه النتائج أن المادة الهجينة المُحضرة تُعد مادة مازة واعدة وقابلة لإعادة الاستخدام للمعالجة الفعالة لمياه الصرف الصناعي الملوثة بالأصبغ.

الكلمات المفتاحية: الجسيمات النانوية المغناطيسية، السيليكا، السول-جل، الامتزاز، إزالة الأصباغ، معالجة مياه الصرف الصحي.

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1. Introduction

Water contaminated with industrial dyes has become an increasingly serious global environmental challenge due to its adverse impacts on aquatic ecosystems and human health (Robinson et al., 2001; Pearce et al., 2003). Water pollution is considered one of the most hazardous forms of environmental contamination compared to other types, owing to the widespread presence of organic pollutants and the difficulty of removing them using conventional treatment methods (Crini, 2006). Several studies have shown that a significant portion of industrial water pollution originates from textile industries and dyeing and chemical processing operations (Robinson et al., 2001; Pearce et al., 2003). Chemical industries generate large quantities of wastewater containing complex organic compounds, including synthetic dyes, phenols, dissolved salts, and non-biodegradable substances (Robinson et al., 2001). These effluents are characterized by their dark coloration and high concentrations of suspended and dissolved organic matter, which reduce light penetration in aquatic environments and consequently hinder the photosynthetic processes of aquatic organisms (Pearce et al., 2003). Even at low concentrations, these compounds may induce ecological toxicity and environmental hazards due to the accumulation of persistent organic pollutants (Crini, 2006). Therefore, the development of efficient technologies for treating dye-contaminated water has become an urgent necessity to protect aquatic environments and mitigate the adverse effects associated with the discharge of untreated effluents (Robinson et al., 2001). In this context, several treatment methods have been developed, including advanced oxidation processes, coagulation, precipitation, electrolysis, membrane filtration, and ion exchange (Crini, 2006). Despite the effectiveness of some of these methods, their high operational costs or limited efficiency in removing certain types of dyes have encouraged researchers to explore more efficient and cost-effective alternatives (Pearce et al., 2003). Adsorption is considered one of the most widely used techniques for dye removal from water due to its operational simplicity, high efficiency, and relatively low cost compared to other methods (Crini, 2006). Adsorption refers to the accumulation of pollutants on the surface of a solid material as a result of physical or chemical interactions between the surface and the adsorbate (Samiey et al., 2014). It is generally classified into physical adsorption and chemical adsorption depending on the nature of the interaction between adsorbate and adsorbent surface (Crini, 2006). In recent years, there has been increasing interest in the use of magnetic nanomaterials in separation and environmental remediation processes due to their unique properties, such as high surface area, ease of separation using an external magnetic field, and rapid attainment of adsorption equilibrium (Ahmad et al., 2019; Atia, 2023). Magnetic nanoparticles have demonstrated strong capabilities in removing both organic and inorganic pollutants from water, making them among the most promising materials in modern water treatment applications (Ahmad et al., 2019). Despite their efficiency, magnetic nanoparticles suffer from aggregation and chemical instability in aqueous media, which may reduce their adsorption performance (Atia, 2023). To overcome these limitations, these particles are coated with silica (SiO₂) layers using the sol-gel technique (Brinker & Scherer, 2013; Ahmad et al., 2019). This coating provides high chemical stability, increases effective surface area, and enables the incorporation of various functional groups that enhance adsorption efficiency (Walcarius & Collinson, 2009). Silicon dioxide is a low-cost, environmentally friendly material with high chemical stability (Brinker & Scherer, 2013). It also possesses abundant surface hydroxyl groups that facilitate chemical modification

and bonding with various organic compounds (Walcarius & Collinson, 2009). Therefore, silane derivatives are widely used to modify silica-coated magnetic nanoparticles to enhance porosity and improve active adsorption sites (Sitthiracha et al., 2015). Hybrid organic–inorganic materials synthesized via the sol–gel process have attracted considerable attention due to their excellent mechanical strength, tunable porosity, and versatile surface chemistry (Sanchez et al., 2005; Gómez-Romero & Sanchez, 2006). These materials combine the structural stability of inorganic silica networks with the functional flexibility of organic moieties, making them highly suitable for adsorption applications (Han et al., 2007; Hay & Raval, 2001). Based on the above, this study aims to synthesize and characterize silica-coated magnetic nanoparticles functionalized with amino groups ($\text{Fe}_3\text{O}_4@\text{SiO}_2\text{-Epoxy-NH}_2$) using the sol–gel technique, and to investigate their efficiency in removing Chicago Sky Blue dye from aqueous solutions (Ahmad et al., 2019; Atia, 2023). The study also aims to determine the optimal operational conditions for the removal process by examining the effects of pH, contact time, adsorbent dose, and initial dye concentration, as well as evaluating removal efficiency using UV–Visible spectrophotometric analysis (Crini, 2006).

2. Materials and Methods

2.1. Chemicals

In this study, Chicago Sky Blue dye with the molecular formula $\text{C}_{37}\text{H}_{24}\text{N}_6\text{Na}_2\text{O}_9\text{S}_3$ was used, along with ammonia solution (NH_3), deionized water, ferric chloride ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$), ferrous chloride ($\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$), tetraethyl orthosilicate (TEOS), 3-Glycidoxypyriltrimethoxysilane (GPTMS), 3-Aminopropyltriethoxysilane (APTES), ethanol ($\text{CH}_3\text{CH}_2\text{OH}$), sodium hydroxide (NaOH), and hydrochloric acid (HCl).

2.2. Instruments and Measurement Conditions

Fourier Transform Infrared Spectroscopy (FTIR) spectra were recorded using the Attenuated Total Reflectance (ATR) technique. Field Emission Scanning Electron Microscopy (FESEM) was used to examine surface morphology and particle size. The pH was measured using a HANNA HI 2213 pH meter. A magnetic stirrer was employed for continuous agitation during both synthesis and adsorption processes. Ultraviolet–Visible (UV–Vis) spectrophotometry was used to determine dye concentration at its maximum absorption wavelength.

2.3. Synthesis of Magnetic Nanoparticles

Fe_3O_4 magnetic nanoparticles were synthesized using the co-precipitation method. Appropriate amounts of $\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$ and $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ were dissolved in 20 mL of deionized water under continuous stirring. The solution was then heated to 50°C under strong magnetic stirring. Subsequently, 5.0 mL of ammonia solution (32% v/v) was added dropwise until a black precipitate was formed. The mixture was stirred for 3 hours, after which the precipitate was washed four times with deionized water and dried at 80°C for 24 hours.

2.4 Synthesis of $\text{Fe}_3\text{O}_4@\text{SiO}_2$

The sol–gel method was used to coat the magnetic nanoparticles with a silica layer. Briefly, 500 mg of the prepared nanoparticles were dispersed in 100 mL of a water/ethanol mixture (1:1, v/v), followed by the addition of 5.0 mL of ammonia solution and 0.5 mL of tetraethyl orthosilicate (TEOS). The mixture was stirred for 30 minutes and then left at room temperature for 24 hours. The resulting product was washed with a water/ethanol mixture and dried to obtain a dark brown $\text{Fe}_3\text{O}_4@\text{SiO}_2$ powder.

2.5 Synthesis of $Fe_3O_4@SiO_2$ -Epoxy

The surface of the silica-coated nanoparticles was functionalized by adding GPTMS under continuous stirring for 5 hours at room temperature. The product was then washed with ethanol and dried at 80°C for 24 hours to obtain an intermediate material containing active epoxy functional groups.

2.6 Synthesis of $Fe_3O_4@SiO_2$ -Epoxy- NH_2

The final adsorbent was prepared by adding APTES to the previously obtained intermediate material under continuous stirring for 5 hours at room temperature. The material was then separated using an external magnet, washed twice with deionized water, and dried at 80°C for 24 hours to obtain a dark black powder representing the final hybrid material.

2.7 Dye Removal Process

20 mg of the adsorbent was added to 20 mL of dye solution with an initial concentration of 20 mg/L. The pH was adjusted to 5 using 0.01 M HCl and 0.01 M NaOH solutions. The mixture was then shaken for 40 minutes using a rotary shaker at 150 rpm at room temperature. The adsorbent was separated using an external magnetic field, and the supernatant was finally analyzed using a UV-Vis spectrophotometer at the maximum absorption wavelength.

3. Adsorption Study Procedures

3.1 Effect of pH

The effect of pH was investigated in the range of 3–9 at room temperature. The initial dye concentration was fixed at 20 mg/L and the adsorbent dose at 20 mg. The solutions were shaken for 40 minutes, after which the magnetic adsorbent was separated and the supernatant was spectrophotometrically analyzed.

3.2 Effect of Adsorbent Dose

The effect of adsorbent dose was studied in the range of 5–60 mg while keeping the dye concentration constant at 20 mg/L and pH at 5. After shaking for 40 minutes, the adsorbent was separated using an external magnet and the remaining solution was analyzed.

3.3 Effect of Contact Time

The effect of contact time was examined over a period of 1–60 minutes at pH 5 and an initial dye concentration of 20 mg/L using 20 mg of adsorbent. The supernatant was then analyzed after magnetic separation.

3.4 Effect of Initial Concentration

The effect of initial dye concentration was studied in the range of 10–60 mg/L while keeping the adsorbent dose at 20 mg, pH at 5, and contact time at 40 minutes.

3.5 Analytical Methods

The concentrations of dye solutions were measured by monitoring the absorbance changes at a wavelength of the maximum absorbance. However, the amount of the dye sorbed at any time, q_t , was calculated from:

$$q_t = \frac{v(c_0 - c_t)}{w} \quad (1)$$

At equilibrium, $q_t = q_e$ and $c_0 = c_t$; therefore, the amount of sorbed dye q_e was calculated from:

$$q_e = \frac{v(c_0 - c_e)}{w} \quad (2)$$

Where, c_0 , c_t and c_e are the initial concentration at any time and equilibrium concentrations of the dye solution (mg/L), respectively, v is the volume of the solution (L), and w is the mass of adsorbent (g).

All in all, the dye removal percentage can be calculated as follows:

$$\text{Removal \%} = \frac{(c_0 - c_e)}{c_0} \times 100 \quad (3)$$

4. Results

4.1 Synthesis Mechanism of the Hybrid Sol-Gel Magnetic Material ($\text{Fe}_3\text{O}_4@\text{SiO}_2\text{-Epoxy-NH}_2$)

The magnetic hybrid sol-gel material ($\text{Fe}_3\text{O}_4@\text{SiO}_2\text{-Epoxy-NH}_2$) was engineered through a multistep functionalization pathway involving surface modification of magnetic nanoparticles using the silane precursors GPTMS and APTES (Fig. 1 A and B). This strategy aims to construct a stable hybrid silica-based network covalently anchored onto the SiO_2 -coated Fe_3O_4 core, thereby enhancing structural stability and introducing abundant active sites for adsorption processes.

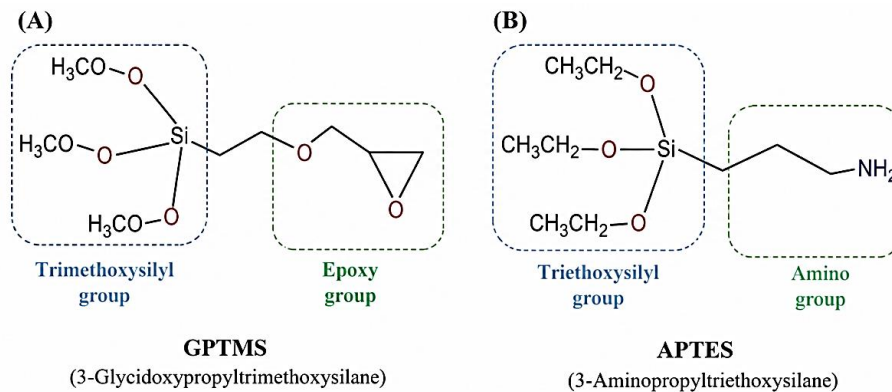


Fig. 1: (A) GPTMS and (B) APTES

Under alkaline conditions (high pH), the amino group ($-\text{NH}_2$) in APTES exhibits strong nucleophilic character due to the presence of a lone electron pair. This enables nucleophilic attack on the epoxy ring of GPTMS, resulting in ring opening and the formation of a stable covalent $-\text{C}-\text{NH}-\text{C}-$ linkage. This reaction represents a key step in the formation of the hybrid polymeric network, as illustrated in Fig. 2

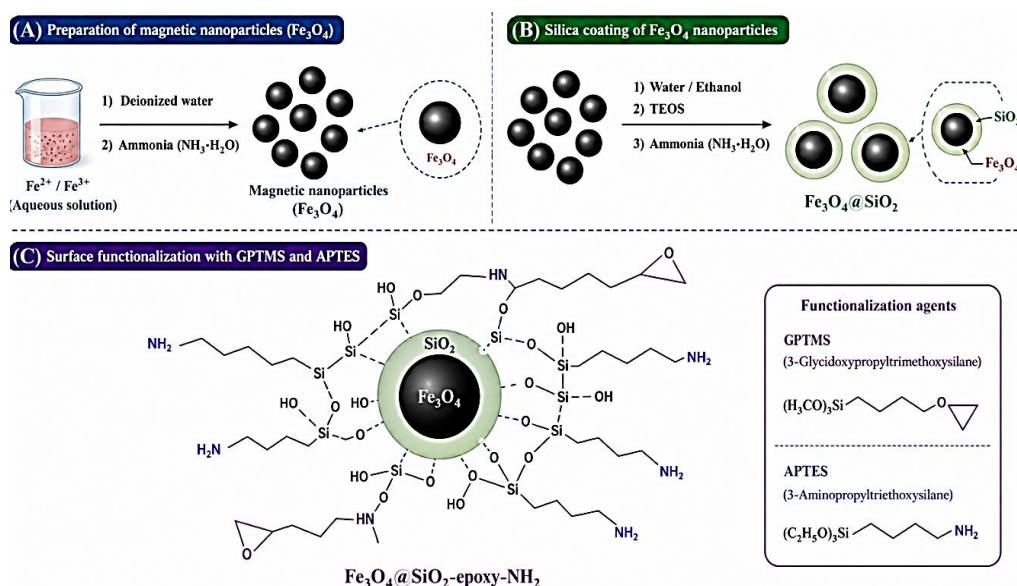


Fig. 2: Synthesis mechanism of (A) MNPs, (B) MNPs@SiO₂, and (C) magnetic hybrid material MNPs@SiO₂-Epoxy-NH₂

The synthesis route proceeds through three fundamental stages: formation of Fe₃O₄ magnetic cores, encapsulation with a SiO₂ shell, and subsequent surface functionalization using GPTMS/APTES to construct an organic–inorganic hybrid siloxane framework. The resulting material is characterized by the presence of three main functional groups: hydroxyl (–OH), amino (–NH₂), and amine/amide-linked bridges (–C–NH–C–). These functional moieties act as active binding sites that promote multiple interaction mechanisms, including electrostatic attraction, hydrogen bonding, and coordination interactions with dye molecules such as Chicago Sky Blue. Consequently, the integrated structure combining a magnetic core, silica shell, and functional organic network significantly enhances adsorption performance by increasing surface reactivity and the density of accessible active sites.

4.2 Characterization of the magnetic hybrid sol-gel MNPs@SiO₂-Epoxy-NH₂

4.2.1 FTIR Spectral Analysis

The FTIR spectra of the prepared materials are presented in the range of 4000–500 cm^{–1} as shown in the fig3. Curve (A) corresponds to bare Fe₃O₄ nanoparticles, exhibiting characteristic absorption bands associated with surface hydroxyl groups (O–H stretching around ~3400 cm^{–1} and O–H bending near ~1630 cm^{–1}) as well as a strong Fe–O vibration band in the low wavenumber region (~540–580 cm^{–1}), confirming the formation of magnetite nanoparticles.

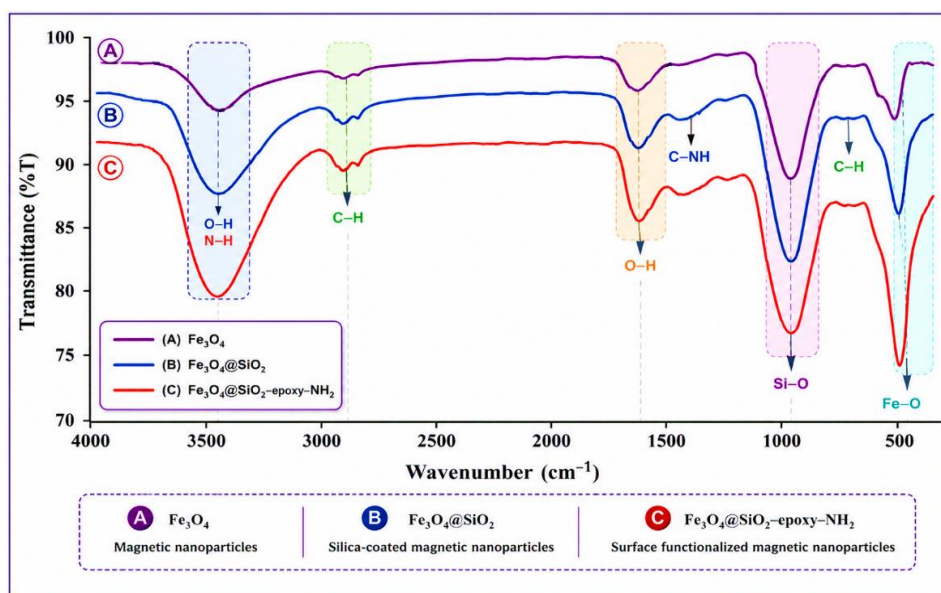


Figure 3. FTIR spectra of (A) MNPs, (B) MNPs@SiO₂, and (C) the magnetic hybrid sol-gel MNPs@SiO₂ Epoxy-NH₂

Curve (B) represents Fe₃O₄@SiO₂ core-shell nanoparticles, where the appearance of a pronounced band around ~1000–1100 cm⁻¹ is attributed to asymmetric stretching vibrations of Si–O–Si bonds, indicating successful formation of the silica coating. In addition, a slight shift and modification in the Fe–O band intensity is observed due to surface encapsulation by the silica layer. Curve (C) corresponds to the final functionalized hybrid material Fe₃O₄@SiO₂–Epoxy–NH₂, which shows multiple characteristic bands confirming successful surface modification, including broad O–H/N–H stretching (~3400 cm⁻¹), C–H stretching (~2920–2950 cm⁻¹), C–NH vibrations (~1470 cm⁻¹), and strong Si–O and Fe–O related bands in the fingerprint region. The coexistence of these functional groups confirms the successful grafting of epoxy-amine functionalities onto the silica-coated magnetic core, forming a stable organic–inorganic hybrid structure. Overall, the spectral evolution from (A) to (C) clearly demonstrates the stepwise synthesis and successful functionalization of the magnetic sol–gel hybrid system.

4.2.2 Scanning Electron Microscopy (SEM) Analysis

Scanning Electron Microscopy (SEM) is a high-resolution imaging technique based on the interaction of a focused electron beam with the sample surface, providing detailed information regarding surface morphology, particle size, and structural organization. In this study, SEM was employed to investigate the morphological features and size distribution of Fe₃O₄ nanoparticles, Fe₃O₄@SiO₂ core-shell structures, and the final hybrid material Fe₃O₄@SiO₂–Epoxy–NH₂.

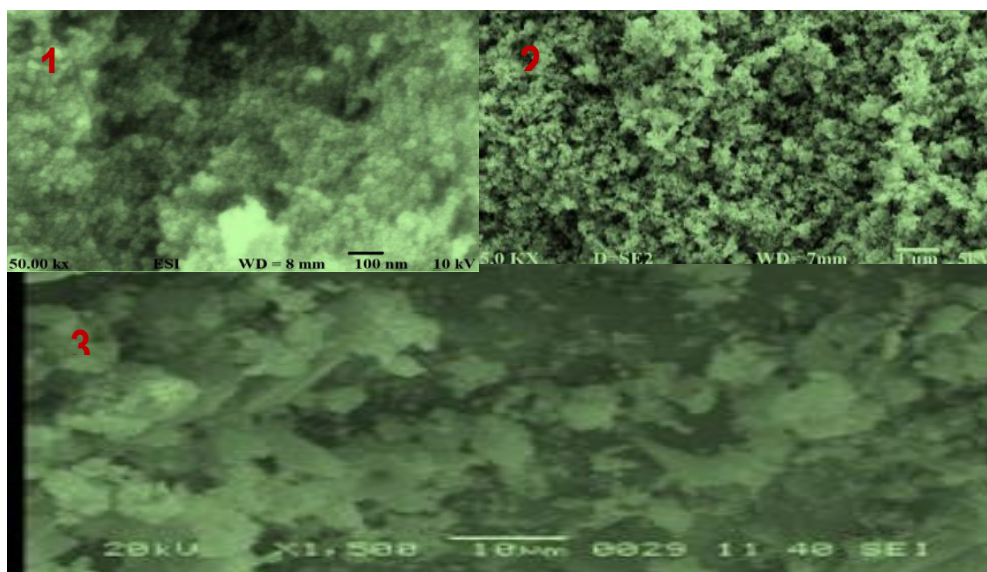


Figure 4: FESEM micrographs of 1 (Fe_3O_4), 2 ($\text{Fe}_3\text{O}_4@\text{SiO}_2$) and 3 ($\text{Fe}_3\text{O}_4@\text{SiO}_2\text{-Epoxy-NH}_2$)

The obtained results reveal that Fe_3O_4 nanoparticles exhibit a nanoscale size distribution in the range of approximately 50–100 nm, accompanied by noticeable particle agglomeration due to strong magnetic dipole dipole interactions. Following silica coating, the $\text{Fe}_3\text{O}_4@\text{SiO}_2$ core shell structures show an apparent increase in particle size together with a significant reduction in aggregation. This behavior is attributed to the formation of a uniform SiO_2 layer that acts as a steric barrier, enhancing particle dispersion and stability.

In contrast, the final hybrid material $\text{Fe}_3\text{O}_4@\text{SiO}_2\text{-Epoxy-NH}_2$ exhibits a well-developed porous morphology. This structure is primarily ascribed to the extensive cross-linking reactions between GPTMS, APTES, and TEOS through Si–O–Si bonding, leading to the formation of an interconnected organic–inorganic siloxane network. The resulting porous architecture is expected to provide a higher surface area and a greater number of accessible active sites, which are favorable for adsorption applications.

4.3 Effect of Various Parameters on Removal of Chicago Sky Blue Dye

4.3.1 Effect of Ph

The adsorption efficiency of Chicago Sky Blue (CSB) dye is strongly influenced by solution pH due to its effect on both dye ionization and adsorbent surface charge. The effect of pH was investigated over the range of 3–9. As shown in Fig 5(A), the removal efficiency increased significantly within the pH range of 5–9, with reduced adsorption observed at lower pH values. At acidic pH, protonation of both the adsorbent surface and dye molecules caused electrostatic repulsion, reducing adsorption efficiency. Under highly basic conditions, deprotonation led to similar repulsive interactions. In contrast, near-neutral pH favored electrostatic attraction and hydrogen bonding between functional groups of the dye and adsorbent, enhancing adsorption performance.

4.3.2 Effect of Adsorbent Dosage

Adsorbent dosage is a critical factor affecting dye removal efficiency, as it determines the number of available active adsorption sites. The effect of dosage was evaluated at different adsorbent amounts, as presented in Fig 5(B). The results showed that removal efficiency increased sharply with increasing dosage, reaching an optimum value at 40 mg. This improvement is attributed to the increase in available adsorption sites. Beyond this dosage, only slight enhancement was observed due to saturation of active sites and attainment of adsorption equilibrium.

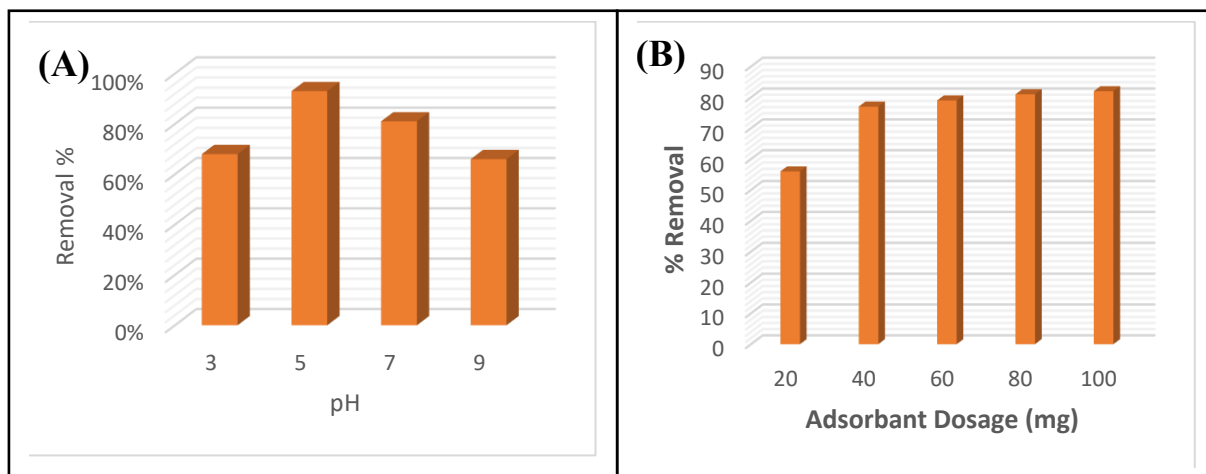


Fig 5: Effect of pH (A) and adsorbent dosage (B) on the percent adsorption of the elimination of CSB

4.3.3 Effect of Initial Concentration

The CSB initial concentration was also fixed at 40 mg/L with dosage of 40 mg. Later on, the magnetic hybrid sol-gel was used in order to collect the material that adsorbed the dye (CSB).

The percentage removal of CSB was found to increase with increase of the initial dye concentration Fig 6(A). The percentages of CSB removal increase probably from 3 to 12 ppm was constant. Because at low concentration not all dyes (about 80 %) can adsorb by adsorbent. From 20 to 40 ppm the efficiency increased slightly probably due to the multilayer adsorption mechanism. 40 mg was chosen as the optimum condition.

4.3.4 Effect of Time

In this experiment, the solutions were mixed slowly with a rotary shaker at a rate of 150 rpm 40 minutes at room temperature. Later on, the magnetic was used in order to collect the material that adsorbed the dye (CSB).

Similarly, the adsorption efficiency was then determined by spectrophotometer. As it is illustrated in Fig6(B) above, the percentage adsorption was calculated and the result revealed that 40 minutes were enough to eliminate CSB with 90.08%.

This is because, by increasing time the adsorption efficiency increased due to adsorption of more CSB dyes onto the active sites of adsorbent. However, after 40 minutes adsorption of all dyes by adsorbent in the system reached equilibrium, hence there is no significant increase of % removal.

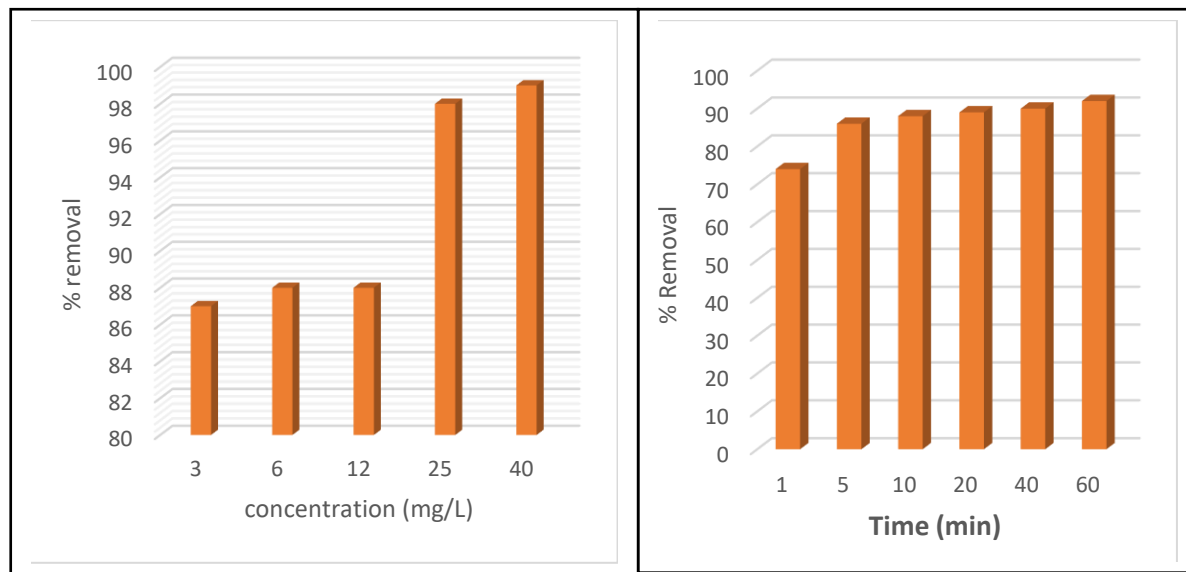


Fig 6: The effect of the amount of initial CSB dye concentration (A) on the studied time (B) on the removal of CSB

5. Discussion

The obtained results confirmed the successful synthesis of the $\text{Fe}_3\text{O}_4@\text{SiO}_2\text{-Epoxy-NH}_2$ hybrid material and its high efficiency for the removal of Chicago Sky Blue dye from aqueous solutions. FTIR and FESEM analyses verified the formation of a silica-coated magnetic core functionalized with amino groups, resulting in a porous organic-inorganic hybrid structure rich in active adsorption sites. These structural properties significantly enhanced adsorption efficiency through electrostatic interactions and hydrogen bonding between the adsorbent surface and dye molecules. The adsorption performance was strongly influenced by operational parameters including pH, adsorbent dosage, contact time, and initial dye concentration. Maximum removal efficiency (~90%) was achieved at pH 5, adsorbent dosage of 40 mg, contact time of 40 min, and dye concentration of 40 mg/L. The enhanced adsorption under moderately acidic conditions may be attributed to favorable surface charge interactions between the amino-functionalized adsorbent and dye molecules. Increasing adsorbent dosage improved dye removal due to the greater availability of active sites, whereas adsorption equilibrium was reached after approximately 40 min, indicating rapid adsorption kinetics. In addition, higher dye concentrations enhanced adsorption performance because of the increased mass transfer driving force between the solution and adsorbent surface. The excellent adsorption behavior of the prepared material is mainly attributed to the synergistic effect of the Fe_3O_4 magnetic core, silica shell, and GPTMS/APTES functional groups, which collectively improved surface reactivity, adsorption affinity, and magnetic separability. These findings demonstrate the promising potential of the synthesized hybrid material for wastewater treatment applications.

5.1 Comparison with Previous Studies

To evaluate the effectiveness of the synthesized $\text{Fe}_3\text{O}_4@\text{SiO}_2\text{-Epoxy-NH}_2$ hybrid adsorbent, the obtained results were compared with several previously reported hybrid and magnetic nanomaterials used for adsorption and wastewater treatment applications. Table X summarizes the comparison between the present study and selected related studies.

Table 1. Comparison between the Present Study and Previous Related Studies

Adsorbent Material	Target Pollutant/Dye	Removal Efficiency (%)	Main Feature	Reference
Graphene oxide-based silica coated magnetic nanoparticles	Methylene Blue dye	~88%	Magnetic silica-coated nanocomposite with easy magnetic separation	Atia (2023)
Magnetic silica-based hybrid organic–inorganic nanocomposite	Pb(II) and Ni(II) ions	>85%	Hybrid magnetic adsorbent with high surface activity	Ahmad et al. (2019)
Hybrid silica materials for dye encapsulation	Organic dyes	High encapsulation efficiency	Silica hybrid structure for dye interaction and stabilization	Wirnsberger et al. (2001)
Mesoporous silica-based organic–inorganic hybrid materials	Organic pollutant adsorption	High adsorption performance	High porosity and tunable surface chemistry	Hoffmann et al. (2006)
$\text{Fe}_3\text{O}_4@\text{SiO}_2\text{-Epoxy-NH}_2$ (Present study)	Chicago Sky Blue dye	~90%	Amino-functionalized magnetic hybrid adsorbent with rapid adsorption and magnetic recovery	Present study

The comparison demonstrates that the synthesized $\text{Fe}_3\text{O}_4@\text{SiO}_2\text{-Epoxy-NH}_2$ hybrid material exhibited competitive adsorption performance toward Chicago Sky Blue dye compared with previously reported magnetic and hybrid adsorbents. The high removal efficiency obtained in the present study may be attributed to the synergistic combination of the Fe_3O_4 magnetic core, silica shell, and amino-functionalized surface groups introduced through GPTMS/APTES modification. These functional groups enhanced electrostatic interactions and hydrogen bonding between the adsorbent surface and dye molecules, resulting in improved adsorption efficiency. In addition, the magnetic nature of the material enabled rapid and simple separation from aqueous media using an external magnetic field, which represents an important advantage for wastewater treatment applications.

Furthermore, compared with conventional silica-based hybrid materials, the prepared adsorbent showed improved surface reactivity and adsorption capability due to the formation of a porous organic–inorganic siloxane network rich in active binding sites. Therefore, the present hybrid material can be considered a promising candidate for efficient dye-contaminated wastewater treatment applications.

6. Conclusion

This study successfully synthesized and characterized a novel magnetic hybrid adsorbent, $\text{Fe}_3\text{O}_4@\text{SiO}_2\text{-Epoxy-NH}_2$, and demonstrated its high efficiency for the removal of Chicago Sky Blue dye from aqueous solutions. Characterization results (FTIR and SEM) confirmed the successful stepwise synthesis and the formation of a functional hybrid structure with improved surface morphology and a porous network rich in active sites, which significantly enhanced the interaction with dye molecules. Adsorption experiments revealed high removal performance under optimized conditions (pH = 5, adsorbent dosage of 40 mg, contact time of 40 min, and initial dye concentration of 40 mg/L). This excellent performance is attributed to the synergistic effect of the Fe_3O_4 magnetic core, the SiO_2 shell, and the functional groups introduced via APTES/GPTMS modification, which collectively provided abundant active sites for electrostatic interactions and hydrogen bonding, in addition to enabling easy magnetic separation and reusability. Accordingly, the synthesized material represents a promising candidate for wastewater treatment applications contaminated with dye pollutants. Future work is recommended to extend reusability studies over more cycles, evaluate performance in complex multi-contaminant systems, investigate the adsorption mechanism using advanced techniques such as XPS and BET analysis, and further explore pilot-scale applications, as well as enhance material performance through additional nanostructural modifications to improve adsorption capacity and selectivity.

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