

RESEARCH PAPER

Magnetic Fe₃O₄/Montmorillonite Nanocomposite as an Efficient and Reusable Adsorbent for Crystal Violet Dye Removal from Aqueous Solutions: Kinetic, Isotherm, and Thermodynamic Studies

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ABSTRACT

A magnetically recoverable Fe₃O₄/montmorillonite (Fe₃O₄/MMT) nanocomposite was synthesized through a simple chemical co-precipitation route and examined as an adsorbent for the removal of crystal violet (CV), a widely used cationic triphenylmethane dye, from aqueous solution. The as-prepared material was characterized by Fourier-transform infrared spectroscopy (FTIR), X-ray diffraction (XRD), field-emission scanning electron microscopy (FESEM) coupled with energy-dispersive X-ray analysis (EDX), and vibrating-sample magnetometry (VSM); together these techniques confirmed the successful loading of Fe₃O₄ nanoparticles onto the clay surface and the superparamagnetic-like behaviour of the composite. Batch experiments were conducted to assess the influence of solution pH, adsorbent dosage, contact time, initial dye concentration and temperature. Under the optimum conditions (pH 8, adsorbent dose 0.6 g L⁻¹, contact time 60 min) the removal efficiency reached about 96%, with a maximum Langmuir monolayer capacity of 178.6 mg g⁻¹ at 298 K. The kinetic data were best represented by the pseudo-second-order model, while the equilibrium results fitted the Langmuir isotherm more closely than the Freundlich or Temkin models, implying monolayer uptake on a fairly homogeneous surface. Thermodynamic analysis showed that the process was spontaneous and endothermic and was accompanied by an increase in entropy at the solid-liquid interface. Importantly, the adsorbent could be separated within a few seconds with an ordinary hand-held magnet and retained roughly 85% of its initial performance after five consecutive adsorption-desorption cycles, which highlights its potential as a low-cost and reusable material for the treatment of dye-contaminated wastewater.

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INTRODUCTION

The discharge of coloured effluents from the textile, leather, paper, cosmetics and printing industries continues to be one of the most pressing environmental concerns of recent decades [1]. Synthetic dyes are, by design, chemically and photolytically stable, and as a consequence they resist conventional biological degradation once they reach natural water bodies [2]. Crystal violet (CV) is a cationic triphenylmethane dye that is extensively employed as a biological stain, in textile dyeing and as an additive in animal feed and veterinary medicine [3]. Despite its usefulness, CV is poorly biodegradable and has been associated with several adverse effects, including eye and skin irritation, respiratory difficulties and, in a number of reports, mutagenic and carcinogenic activity [4]. Even at fairly low concentrations the dye imparts an intense colour to water, lowering light penetration and interfering with the photosynthetic activity of aquatic organisms [5]. The removal of CV prior to discharge is therefore essential. From a broader public-health perspective, safeguarding the quality of water and the surrounding environment is essential for protecting human health, and particularly the health of vulnerable groups such as children and patients with chronic conditions [6–10].

Dye-laden wastewater has been treated by different procedures as coagulation-flocculation, membrane filtration, enhanced oxidation, electrochemical treatment and biological degradation [11]. More recently, advanced photocatalytic systems based on mixed transition-metal oxides and their graphene-oxide composites, such as ZnCo_2O_4 nanospheres and $\text{ZnCo}_2\text{O}_4/\text{GO}$ nanocomposites, have also been reported for the visible-light degradation of organic pollutants in water [12]. Among these, adsorption is still the most attractive choice because of its operational simplicity, low cost, ease of operation and the ability of renewing the used material [13]. Accordingly, a broad range of low-cost adsorbents, including activated nano-carbons derived from natural precursors, has been developed and reviewed for water-treatment applications [14]. Clay minerals, especially montmorillonite (MMT), are gaining much attention owing to their availability and low cost as adsorbents. Their layered structure, high surface area and persistent negative surface charge make them very suitable for the removal of cationic dyes [15]. However, a

practical disadvantage of fine clay powders is the difficulty of their recovery from treated water by common filtration or centrifugation [16].

To overcome this limitation, magnetite (Fe_3O_4) nanoparticles can be incorporated into the clay matrix so that the resulting composite is easily collected by applying an external magnetic field [17]. In this regard, the synthesis and characterization of new compounds and nanostructured composite materials, in which inorganic nanoparticles are dispersed within or anchored onto a host matrix, has emerged as a versatile strategy for tailoring the surface and functional properties of such systems [18–20]. Beyond environmental remediation, such engineered nanomaterials, including metallic nanoparticles such as gold, are also widely exploited in biomedical and therapeutic applications, which underlines their broad functional versatility [21]. In the present study, a $\text{Fe}_3\text{O}_4/\text{MMT}$ nanocomposite was prepared, characterized and evaluated for CV removal, with particular attention given to the kinetic, equilibrium and thermodynamic behaviour of the system.

MATERIALS AND METHODS

Chemicals

Natural montmorillonite clay was used as the supporting matrix. Iron(III) chloride hexahydrate ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$), iron(II) sulfate heptahydrate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$), ammonia solution (25%), sodium hydroxide, hydrochloric acid and crystal violet ($\text{C}_{25}\text{H}_{30}\text{ClN}_3$, $M = 407.98 \text{ g mol}^{-1}$) were of analytical grade and used as received without further purification. All solutions were prepared with deionized water. A 1000 mg L^{-1} stock CV solution was prepared and diluted to the desired working concentrations as required.

Preparation of the $\text{Fe}_3\text{O}_4/\text{MMT}$ nanocomposite

The $\text{Fe}_3\text{O}_4/\text{MMT}$ nanocomposite was synthesized by an in-situ co-precipitation procedure (Fig. 1) [22]. In brief, 2 g of montmorillonite was dispersed in 100 mL of deionized water and stirred vigorously for about 1 h to give a homogeneous suspension. $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ and $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ were then dissolved in the clay suspension at a $\text{Fe}^{3+}/\text{Fe}^{2+}$ molar ratio of 2:1, and the mixture was heated to 80°C under a nitrogen atmosphere with continuous stirring. Ammonia solution was added dropwise until the pH reached about 10–11, whereupon a black precipitate formed almost

at once, indicating the formation of magnetite on the clay surface. The stirring was continued for an additional hour to allow the particles to develop and to anchor to the silicate sheets. The black product was separated by magnetic separation, washed multiple times with deionised water and ethanol until the supernatant was neutral, and then dried at 60°C overnight. The dried solid was gently ground and stored in a desiccator until use.

Characterization

The functional groups present in MMT, Fe₃O₄ and the composite were identified from FTIR spectra recorded over the 400–4000 cm⁻¹ region. The crystalline phases were examined by XRD using Cu K α radiation, while the surface morphology and the elemental composition were studied by FESEM and EDX, respectively. The magnetic response of the composite was measured at room temperature with a VSM.

Batch adsorption experiments

All adsorption tests were carried out in batch mode using 100 mL conical flasks containing 50 mL of CV solution. A fixed mass of adsorbent was added and the flasks were agitated on a shaker at 200 rpm at room temperature unless stated otherwise. The effect of pH was studied over the range 2–10, the initial value being adjusted with 0.1 M HCl or 0.1 M NaOH. The adsorbent dosage was varied from 0.2 to 1.0 g L⁻¹, the contact time from 5 to 120 min, the initial dye concentration from 10 to 100 mg L⁻¹ and the temperature from 298 to 318 K. After the chosen contact time the adsorbent was withdrawn with a magnet and the residual dye concentration in the supernatant was determined spectrophotometrically at $\lambda_{\text{max}} = 590$ nm. The amount of dye adsorbed at equilibrium (q_e , mg g⁻¹) and the removal efficiency (R, %) were obtained from Eqs. 1 and 2:

$$q_e = (C_0 - C_e) V / m \quad (1)$$

$$R (\%) = [(C_0 - C_e) / C_0] \times 100 \quad (2)$$

where C_0 and C_e are the initial and equilibrium dye concentrations (mg L⁻¹), V is the solution volume (L) and m is the mass of adsorbent (g).

Adsorption modelling

To explore the rate-controlling step, the experimental data were analyzed with the pseudo-

first-order (PFO), pseudo-second-order (PSO) and intraparticle diffusion (IPD) models, given in their linear forms by Eqs. 3–5:

$$\ln (q_e - q_t) = \ln q_e - k_1 t \quad (3)$$

$$t / q_t = 1 / (k_2 q_e^2) + t / q_e \quad (4)$$

$$q_t = k_{ia} t^{0.5} + C \quad (5)$$

where q_t (mg g⁻¹) is the amount adsorbed at time t (min); k_1 (min⁻¹), k_2 (g mg⁻¹ min⁻¹) and k_{ia} (mg g⁻¹ min^{-0.5}) are the respective rate constants; and C is a constant related to the boundary-layer thickness. The equilibrium data were fitted to the Langmuir, Freundlich and Temkin isotherms (Eqs. 6–8):

$$C_e / q_e = 1 / (q_{\text{max}} K_L) + C_e / q_{\text{max}} \quad (6)$$

$$\ln q_e = \ln K_f + (1/n) \ln C_e \quad (7)$$

$$q_e = B \ln K_t + B \ln C_e \quad (8)$$

Here q_{max} (mg g⁻¹) is the maximum monolayer capacity, K_L (L mg⁻¹) is the Langmuir constant, K_f and n are the Freundlich constants, K_t (L g⁻¹) is the Temkin binding constant and B ($= RT/b_t$) is related to the heat of adsorption. The favourability of the Langmuir process was further judged from the dimensionless separation factor $R_L = 1/(1 + K_L C_0)$. Finally, the thermodynamic parameters ΔG° , ΔH° and ΔS° were evaluated from Eqs. 9–11:

$$\Delta G^\circ = -RT \ln K_c \quad (9)$$

$$\ln K_c = (\Delta S^\circ / R) - (\Delta H^\circ / RT) \quad (10)$$

$$K_c = q_e / C_e \quad (11)$$

where R is the universal gas constant (8.314 J mol⁻¹ K⁻¹), T the absolute temperature (K) and K_c the distribution coefficient. ΔH° and ΔS° were obtained from the slope and intercept of the van't Hoff plot of $\ln K_c$ versus $1/T$.

Point of zero charge and reusability

The point of zero charge (pH_{pzc}) of the composite was determined by the salt-addition (pH-drift) method using 0.01 M NaCl, with the initial and final pH recorded after 24 h of equilibration. For the regeneration tests, the dye-

loaded adsorbent was recovered magnetically and stirred in an ethanol/0.1 M NaOH mixture to desorb the bound CV, then washed, dried and reused in five successive adsorption–desorption cycles under the optimum conditions.

RESULTS AND DISCUSSION

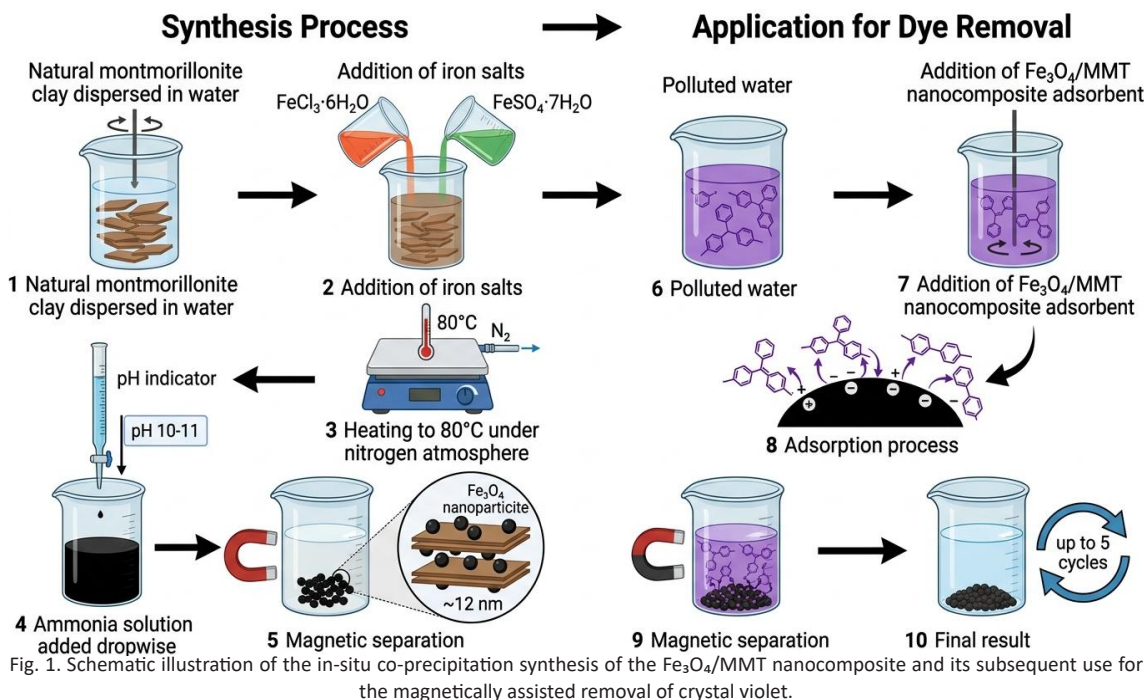
Characterization of the adsorbent

The FTIR spectra of pristine MMT, bare Fe_3O_4 and the Fe_3O_4 /MMT composite are presented in Fig. 2a. The spectrum of montmorillonite displayed a broad band near 3620 cm^{-1} assigned to the stretching of structural O–H groups, together with a band around 3430 cm^{-1} arising from hydrogen-bonded water and a bending vibration at about 1635 cm^{-1} [23]. The intense absorption centred at 1030 cm^{-1} is characteristic of Si–O–Si stretching of the silicate layers, whereas the peaks at 520 and 465 cm^{-1} correspond to Al–O–Si and Si–O–Si bending [24]. For Fe_3O_4 a strong band was observed at roughly 580 cm^{-1} , attributed to the Fe–O stretching of the spinel magnetite lattice [25]. The composite showed the presence of the Si–O band of the clay and the Fe–O band of magnetite with a small shift of the Si–O signal indicating that the iron oxide interacts with the clay surface and not a simple physical combination.

Fig. 2b exhibits XRD patterns. Montmorillonite

is characterised by low angle basal reflection (001) and reflections at around $2\theta = 19.8^\circ$ and 26.6° which are partly owing to related quartz [26]. The composite pattern kept the primary clay reflections and displayed additional peaks at $2\theta = 30.1^\circ, 35.5^\circ, 43.1^\circ, 53.4^\circ, 57.0^\circ$ and 62.6° which may be indexed to the (220), (311), (400), (422), (511) and (440) planes of the cubic inverse-spinel structure of Fe_3O_4 (JCPDS 19-0629) [27]. The presence of clay and magnetite reflections indicates that the structure of the montmorillonite crystal was mostly retained after the deposition of iron oxide. The average crystallite size of Fe_3O_4 phase was found to be around 12 nm determined from (311) peak using Scherrer equation.

The morphology of the samples was examined by FESEM, as shown in Fig. 2c. Montmorillonite appeared as stacked, plate-like sheets with relatively smooth surfaces, in keeping with its layered aluminosilicate nature [28]. After the co-precipitation treatment, numerous near-spherical magnetite nanoparticles were seen distributed over and between the clay platelets, giving a noticeably rougher and more porous surface. Some aggregation of the Fe_3O_4 particles was apparent, which is commonly observed for magnetic nanoparticles owing to their high surface energy and magnetic dipole interactions [29]. The EDX



spectrum (inset) confirmed the presence of Fe, O, Si, Al and Mg – the last three originating from the clay framework – providing further evidence for the formation of the composite.

The room-temperature magnetization curve of the composite (Fig. 2d) passed through the origin with negligible coercivity and remanence, indicating a superparamagnetic-like response, and gave a saturation magnetization of about 31 emu g⁻¹ [30]. Although this value is lower than that of pure magnetite, because of the diamagnetic clay content, it was more than enough to allow the adsorbent to be separated from solution within a few seconds with an ordinary magnet. The point of zero charge of the composite was found to be close to pH 6.3; the surface therefore carries a net negative charge above this value, which is favourable for the electrostatic attraction of cationic dyes such as CV.

Effect of solution pH

Solution pH is one of the most influential variables in dye adsorption, since it governs both

the surface charge of the adsorbent and the ionic form of the dye [31]. The effect of pH on CV removal is shown in Fig. 3a. The uptake was relatively low under strongly acidic conditions and increased markedly as the pH rose, reaching a plateau at around pH 8. At low pH the high concentration of H⁺ ions competes with the positively charged CV molecules for the available sites, and the composite surface, being below its pH_{pzc} (6.3), is positively charged, which causes electrostatic repulsion with the cationic dye [32]. As the pH increases beyond the pH_{pzc} the surface becomes progressively more negative, so the electrostatic attraction between the deprotonated sites and the CV cations is strengthened and removal improves substantially. Above pH 8 the increase levelled off; to avoid possible dye precipitation at very high pH, the remaining experiments were performed at pH 8, which was taken as the optimum value.

Effect of adsorbent dosage

The influence of adsorbent dosage was studied between 0.2 and 1.0 g L⁻¹ (Fig. 3b). The removal

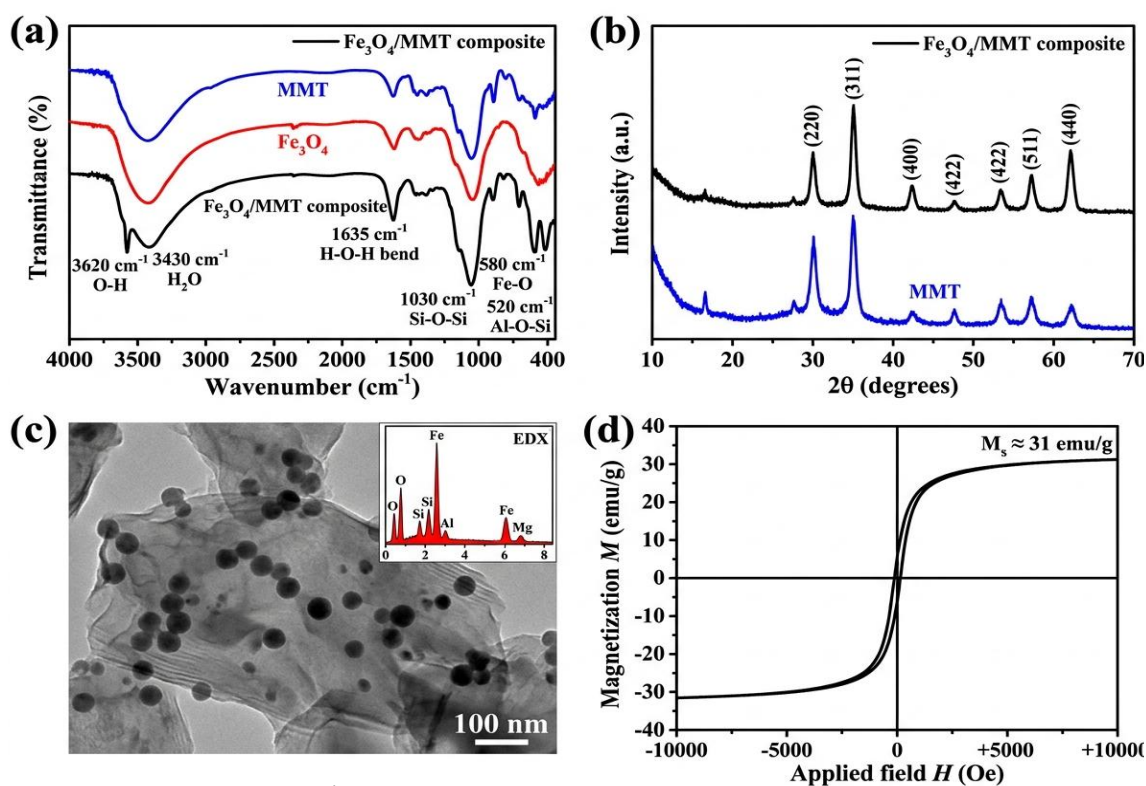


Fig. 2. Characterization of the Fe₃O₄/MMT nanocomposite: (a) FTIR spectra of MMT, Fe₃O₄ and the composite; (b) XRD patterns of MMT and the composite; (c) TEM micrograph with the corresponding EDX spectrum (inset); and (d) room-temperature VSM magnetization curve of the composite.

efficiency rose sharply from about 58% to nearly 96% as the dose was increased from 0.2 to 0.6 g L⁻¹, after which further addition gave only a marginal improvement. The initial rise is readily explained by the greater number of active sites and the larger total surface area made available at higher dosage [33]. The plateau observed at higher loadings is usually ascribed to partial overlapping or aggregation of the adsorbent particles, which lowers the effective surface area, and to the fact that a large fraction of the sites stays unoccupied once most of the dye has already been removed [34]. In contrast, the adsorption capacity per unit mass (q_e) decreased with increasing dosage, a trend that is often reported and reflects the under-utilization of the sites at high adsorbent concentration. A dose of 0.6 g L⁻¹ was therefore chosen as the optimum.

contact time. Adsorption was fast during the first 20 min, when more than three-quarters of the total dye was removed, and thereafter it slowed considerably before reaching equilibrium at about 60 min. The rapid initial stage is attributed to the abundance of vacant and easily accessible sites on the external surface of the composite, whereas the slower second stage corresponds to the gradual occupation of the remaining sites and to diffusion of the dye into the interlayer and pore spaces, where transport is more restricted [35]. No appreciable change in uptake was detected beyond 60 min, so this time was adopted as the equilibrium contact time for the rest of the work. The relatively short equilibrium time is attractive from a practical standpoint, as it implies a high removal rate and a small required reactor residence time.

Effect of contact time

Fig. 3c presents the variation of CV uptake with

Effect of initial concentration and temperature

The effect of initial dye concentration was

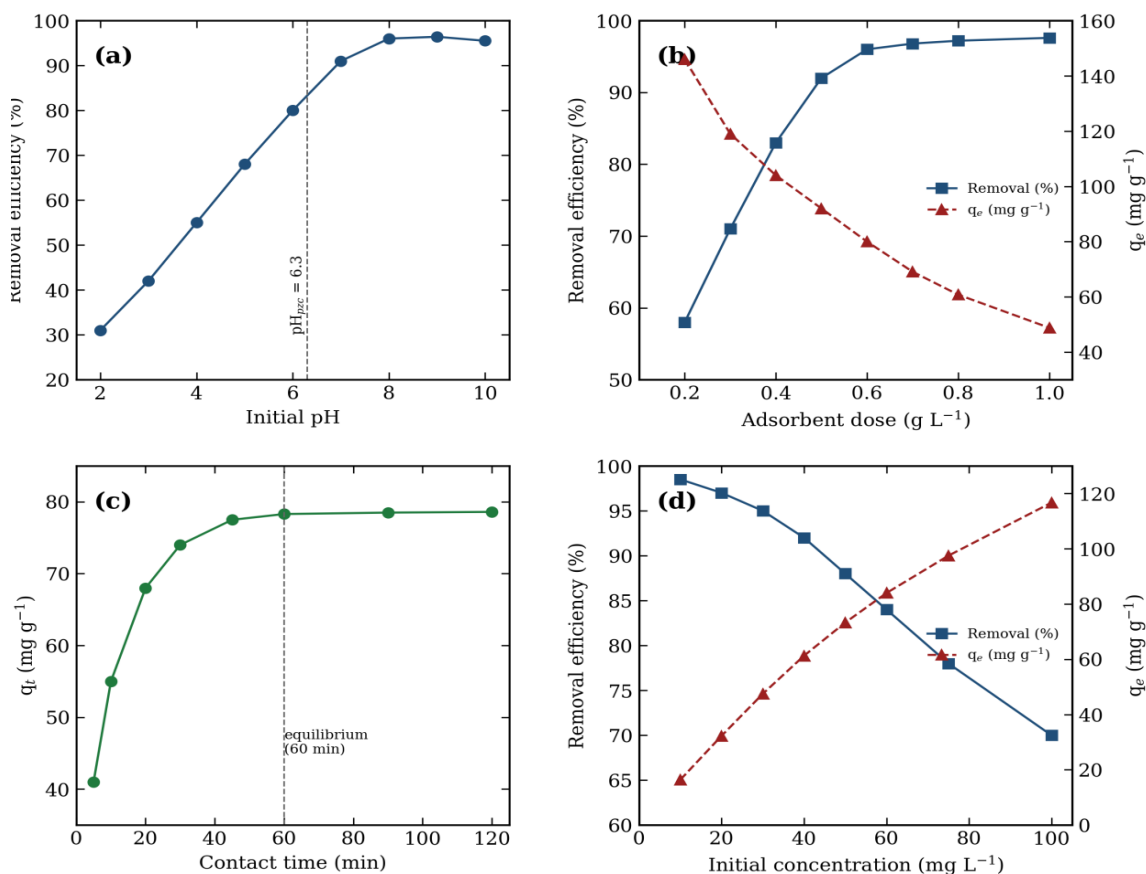


Fig. 3. Effect of the operating parameters on the removal of crystal violet by the Fe₃O₄/MMT nanocomposite: (a) solution pH; (b) adsorbent dosage; (c) contact time; and (d) initial dye concentration.

studied in the range of 10–100 mg L⁻¹ (Fig. 3d). Equilibrium adsorption capacity was enhanced with rise in concentration while the percentage removal was lowered gradually. At low concentrations the ratio of accessible sites to dye molecules is high so practically all the dye is collected, at higher concentrations the fixed number of sites becomes saturated and a greater fraction of the dye stays in solution [36]. The increase of q_e with concentration is due to the stronger driving force of the bigger concentration gradient, which helps overcome the mass-transfer resistance between the liquid and the solid phases [37]. The temperature was changed from 298 to 318 K and the uptake slightly increased with temperature suggesting an endothermic process. The enhancement at higher temperature could be due to a better diffusion of the dye and to the activation or formation of more adsorption sites, and to a probable expansion of the clay interlayers leading to the increased access to the interior surface [38].

Adsorption kinetics

The kinetic behaviour of adsorption was studied using PFO, PSO and IPD models and the associated parameters are presented in Table

1. The correlation coefficients of the PSO model ($R^2 \geq 0.996$) for all the initial concentrations studied were higher than those of the PFO model. Moreover, the equilibrium capacities calculated from the PSO equation ($q_{e,cal}$) were in good agreement with the measured values ($q_{e,exp}$) more satisfactorily. In contrast, the PFO model showed a constant underestimation of q_e . These observations suggested that the adsorption of CV on the Fe₃O₄/MMT composite is well described by pseudo-second-order kinetics, which is typically interpreted as chemisorption occurring in the rate-limiting step involving the electron sharing or exchange between the dye and the surface functional groups [39]. The plots of intraparticle diffusion were not linear and did not pass through the origin ($C \neq 0$) which indicated that the pore diffusion is not the only rate governing mechanism and film (boundary-layer) diffusion also contributes to the process. The multilinear feature of the IPD plot confirms the adsorption in numerous subsequent stages, i.e. exterior surface adsorption followed by progressive intraparticle diffusion.

Adsorption isotherms

The equilibrium data obtained at 298, 308 and

Table 1. Kinetic parameters for the adsorption of crystal violet onto the Fe₃O₄/MMT nanocomposite at different initial concentrations.

Parameter	C ₀ = 25 mg L ⁻¹	C ₀ = 50 mg L ⁻¹	C ₀ = 75 mg L ⁻¹
$q_{e,exp}$ (mg g ⁻¹)	39.8	78.5	112.4
PFO – k_1 (min ⁻¹)	0.052	0.041	0.034
PFO – $q_{e,CaL}$ (mg g ⁻¹)	24.1	49.7	71.2
PFO – R ²	0.951	0.963	0.958
PSO – k_2 (g mg ⁻¹ min ⁻¹)	0.0035	0.0014	0.00071
PSO – $q_{e,CaL}$ (mg g ⁻¹)	41.2	81.3	116.5
PSO – R ²	0.998	0.997	0.996
IPD – k_{ia} (mg g ⁻¹ min ^{-0.5})	2.10	3.80	5.60
IPD – C (mg g ⁻¹)	18.4	29.6	38.2
IPD – R ²	0.890	0.910	0.900

Table 2. Langmuir, Freundlich and Temkin isotherm parameters for the adsorption of crystal violet onto the Fe₃O₄/MMT nanocomposite at three temperatures.

Isotherm model / parameter	298 K	308 K	318 K
Langmuir – q_{max} (mg g ⁻¹)	178.6	192.3	205.8
Langmuir – K_L (L mg ⁻¹)	0.082	0.104	0.131
Langmuir – R ²	0.997	0.998	0.996
R _L range	0.109–0.549	0.088–0.494	0.071–0.433
Freundlich – K_F	30.4	36.8	43.2
Freundlich – n	2.74	2.91	3.08
Freundlich – R ²	0.962	0.957	0.951
Temkin – K_t (L g ⁻¹)	1.02	1.38	1.81
Temkin – b_t (J mol ⁻¹)	72.5	68.1	63.4
Temkin – R ²	0.945	0.939	0.933

318 K were analyzed using the Langmuir, Freundlich and Temkin models; the fitted constants are listed in Table 2. The Langmuir isotherm gave the highest correlation coefficients ($R^2 \approx 0.996$ – 0.998) at every temperature, indicating that CV is adsorbed as a monolayer on energetically equivalent sites with no significant interaction between the adsorbed molecules [40]. The maximum monolayer capacity calculated from the Langmuir model increased from 178.6 mg g^{-1} at 298 K to 205.8 mg g^{-1} at 318 K, once again pointing to an endothermic process. The dimensionless separation factor R_L lay between 0 and 1 over the whole concentration range, confirming that the adsorption is favourable. Although the Freundlich model also gave reasonable fits, with the heterogeneity factor n between 2 and 3 (i.e. $1 < n < 10$,

indicating favourable adsorption), its correlation coefficients were somewhat lower than those of the Langmuir model. The Temkin model produced positive values of the constant related to the heat of adsorption, which is consistent with an endothermic interaction, but it described the data least well of the three. Overall, the dominance of the Langmuir model supports a largely monolayer, site-specific uptake mechanism.

Thermodynamic study

The thermodynamic parameters derived from the van't Hoff analysis are collected in Table 3. The negative values of ΔG° at all temperatures confirm that the adsorption of CV is thermodynamically spontaneous and feasible, and the fact that ΔG° becomes more negative as the temperature

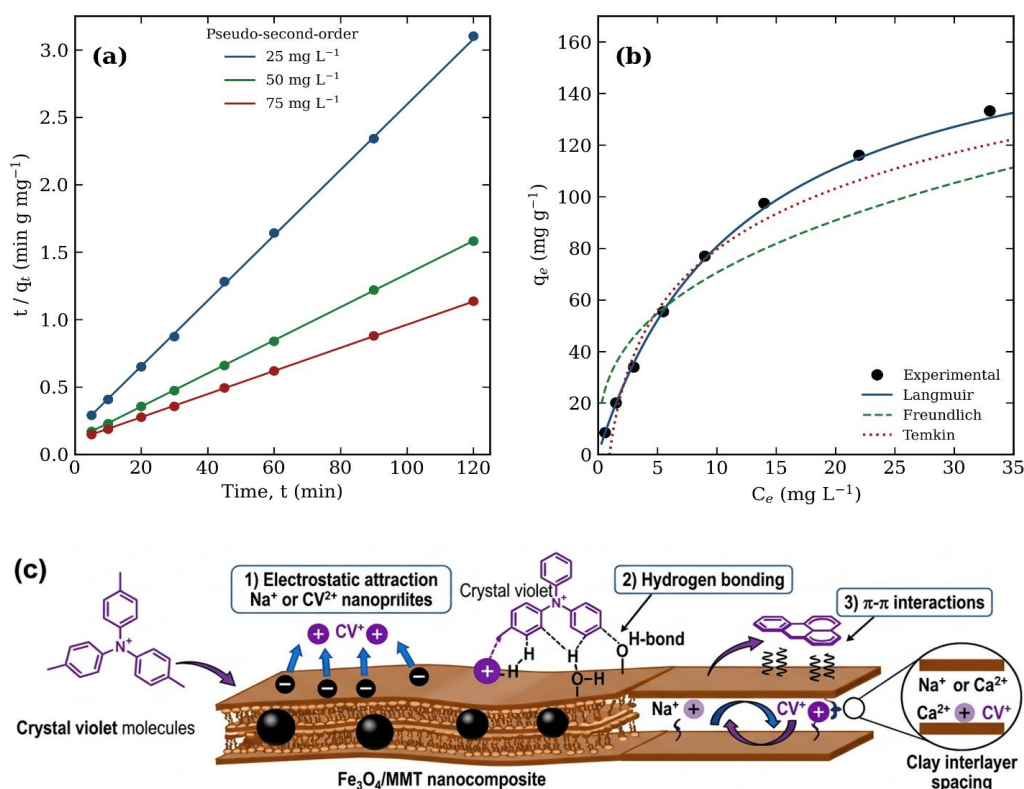


Fig. 4. (a) Pseudo-second-order kinetic plots; (b) Langmuir, Freundlich and Temkin isotherm fits; and (c) schematic of the proposed mechanism for the adsorption of crystal violet onto the Fe₃O₄/MMT nanocomposite.

Table 3. Thermodynamic parameters for the adsorption of crystal violet onto the Fe₃O₄/MMT nanocomposite.

T (K)	Kc	ΔG° (kJ mol ⁻¹)	ΔH° (kJ mol ⁻¹)	ΔS° (J mol ⁻¹ K ⁻¹)
298	4.09	-3.49	+22.7	+87.9
308	5.51	-4.37	—	—
318	7.29	-5.25	—	—

risers (from $-3.49 \text{ kJ mol}^{-1}$ at 298 K to $-5.25 \text{ kJ mol}^{-1}$ at 318 K) indicates that the process is more favourable at higher temperatures [41]. The positive value of ΔH° ($+22.7 \text{ kJ mol}^{-1}$) verifies the endothermic nature already inferred from the concentration and temperature experiments, while its magnitude is broadly consistent with a process having both physical and chemical contributions. The positive ΔS° ($+87.9 \text{ J mol}^{-1} \text{ K}^{-1}$) reflects an increase in randomness at the solid–solution interface during adsorption, which is often attributed to the release of ordered water molecules from the hydration shells of the dye and the surface as the dye becomes attached.

Proposed adsorption mechanism

On the basis of the characterization data and

the influence of the operating variables, the removal of CV by the $\text{Fe}_3\text{O}_4/\text{MMT}$ composite can be rationalized in terms of several cooperative interactions, as illustrated in Fig. 4. The principal contribution appears to be electrostatic attraction between the cationic CV molecules and the negatively charged silicate surface, which becomes increasingly favourable above the pH_{pzc} . In addition, the partially negative oxygen atoms of the surface hydroxyl and Si–O groups can engage in hydrogen bonding and dipole interactions with the dye, while the aromatic rings of CV may take part in $n-\pi$ and $\pi-\pi$ interactions with the surface [42]. Ion exchange between the exchangeable interlayer cations of the clay and the dye cations is also expected to contribute. The combination of these mechanisms accounts for the comparatively

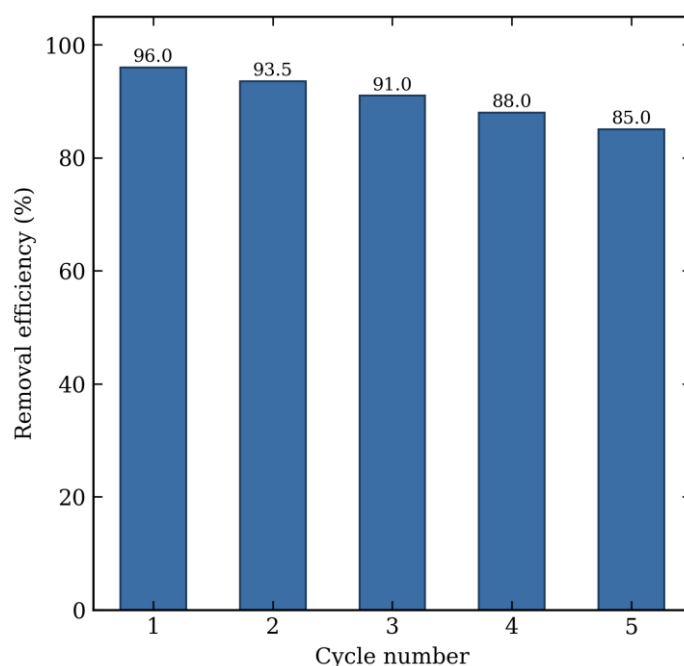


Fig. 5. Removal efficiency of the $\text{Fe}_3\text{O}_4/\text{MMT}$ nanocomposite for crystal violet over five consecutive adsorption–desorption cycles.

Table 4. Comparison of the maximum monolayer adsorption capacity of the $\text{Fe}_3\text{O}_4/\text{MMT}$ nanocomposite with that of other adsorbents reported for crystal violet removal.

Adsorbent	q_{max} (mg g^{-1})	Reference
Pine bark powder	32.78	[4]
NaOH-modified rice husk	44.87	[45]
Mesoporous MCM-41	35.0	[46]
Coconut male-flower activated carbon	85.84	[47]
Functionalized MWCNTs	90.6	[48]
$\text{Fe}_3\text{O}_4/\text{MMT}$ nanocomposite	178.6	This study

high capacity that was observed.

Regeneration and reusability

From an economic and environmental point of view, the reusability of an adsorbent is just as important as its capacity. The regeneration performance of the composite over five consecutive cycles is shown in Fig. 5. The removal efficiency declined only modestly, from about 96% in the first cycle to roughly 85% in the fifth, corresponding to a loss of around 11% after five uses. This gradual decrease is most likely caused by the incomplete desorption of strongly bound dye molecules and a small loss of material during the recovery and washing steps [43]. The retention of a high level of activity, together with the convenience of magnetic separation, makes the material attractive for repeated use in practical water-treatment applications.

Comparison with reported adsorbents

To place the present results in context, the maximum adsorption capacity of the $\text{Fe}_3\text{O}_4/\text{MMT}$ composite is compared with that of various adsorbents reported in the literature for CV removal in Table 4. The capacity obtained here is comparable to, and in several cases higher than, those of many previously studied materials. Taking into account, in addition, the simplicity and low cost of the preparation, the use of an abundant natural clay and the easy magnetic recovery of the spent adsorbent, the $\text{Fe}_3\text{O}_4/\text{MMT}$ nanocomposite can be regarded as a competitive candidate for the treatment of dye-bearing wastewater [44].

CONCLUSION

A magnetically separable $\text{Fe}_3\text{O}_4/\text{montmorillonite}$ nanocomposite was successfully synthesized by a simple co-precipitation method and applied to the removal of crystal violet from aqueous solution. The combined FTIR, XRD, FESEM–EDX and VSM analyses confirmed that magnetite nanoparticles with a crystallite size of about 12 nm were anchored on the clay sheets and that the composite was magnetically responsive. Batch adsorption experiments showed that CV removal was strongly dependent on pH, adsorbent dose, contact time and initial concentration, with a maximum removal of around 96% achieved at pH 8, a dose of 0.6 g L^{-1} and a contact time of 60 min. The kinetic data followed the pseudo-second-order model, and the equilibrium

results were best described by the Langmuir isotherm, with a maximum monolayer capacity of 178.6 mg g^{-1} at 298 K. The thermodynamic study indicated a spontaneous and endothermic process accompanied by an increase in entropy. The composite could be readily recovered with a magnet and reused for at least five cycles with only a small loss of efficiency, which confirms its promise as a low-cost and reusable adsorbent for the remediation of dye-contaminated water.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this manuscript.

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