

Sulfur-Iron Modified Carboxylated Cellulose Nanofiber/Reduced Graphene Oxide Aerogel for Wastewater Remediation

(Aerogel Nanofiber Selulosa Terkarboksil/Grafena Oksida Terkurang Diubah Suai Sulfur-Besi untuk Pemulihan Air Sisa)

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ABSTRACT

Integrating adsorption with advanced oxidation processes (AOPs) is a promising but challenging strategy for wastewater remediation. Herein, a sulfur-iron modified carboxylated cellulose nanofiber/reduced graphene oxide (CCNF/rGO/FeS) aerogel was fabricated using CCNF as a fibrous skeleton to function as a dualfunctional adsorbent and catalyst for synergistic pollutant adsorption and catalytic degradation. The CCNF/rGO/FeS aerogel showed prominent selective adsorption toward cationic dyes including methylene blue (MB), rhodamine B (RhB) and crystal violet (CV), with adsorption capacities of 757.1, 911.0, and 1281.4 mg·g⁻¹, respectively, obviously higher than those of the iron modified carboxylated cellulose nanofiber/reduced graphene oxide (CCNF/rGO/Fe) aerogel. Its initial adsorption rates were over three times higher than those of the CCNF/rGO/Fe aerogel, and the composite maintained pollutant removal efficiency above 80% after five cycles, showing favorable reusability and structural stability. Theoretical analysis confirmed that $\pi\pi$ stacking and electrostatic interactions driven by CCNF carboxyl groups dominate pollutant adsorption. Differing from traditional photocatalytic CCNF aerogels, the material efficiently activates PMS without light irradiation to generate singlet oxygen (¹O₂), sulfate radicals (SO₄^{·-}) and hydroxyl radicals (·OH), realizing deep degradation and mineralization of adsorbed organics. CCNF carboxyl groups synergize with FeS sites to integrate adsorption and catalysis rather than merely acting as a skeleton. This study offers a feasible route to fabricate highefficiency fiberbased composites for practical complex wastewater system.

Keywords: Adsorption; cellulose nanofiber aerogel; degradation; sulfidated zero-valent iron; wastewater remediation

ABSTRAK

Penggabungan proses penjerapan dengan proses pengoksidaan lanjutan merupakan strategi yang berpotensi namun mencabar bagi pemulihan air sisa. Dalam kajian ini, aerogel nanofiber selulosa terkarboksil/grafena oksida terkurang yang diubah suai sulfur-besi telah dihasilkan dengan menggunakan nanofiber selulosa terkarboksil sebagai rangka gentian. Bahan ini berperanan sebagai penjerap dan pemangkin dwifungsi untuk melaksanakan penjerapan bahan pencemar dan penguraian pemangkin secara sinergistik. Aerogel nanofiber selulosa terkarboksil/grafena oksida terkurang yang diubah suai sulfur-besi menunjukkan keupayaan penjerapan selektif yang luar biasa terhadap pewarna kation, termasuk metilena biru, rodamin B dan kristal violet. Kapasiti penjerapannya masing-masing ialah 751.2, 911.0 dan 1281.4 mg·g⁻¹, yang jelas lebih tinggi berbanding aerogel nanofiber selulosa terkarboksil/grafena oksida terkurang yang diubah suai besi tulen. Kadar penjerapan awal bahan ini adalah lebih tiga kali ganda berbanding aerogel nanofiber selulosa terkarboksil/grafena oksida terkurang yang diubah suai besi. Komposit ini mengekalkan kecekapan penyingkiran bahan pencemar melebihi 80% selepas lima kitaran ujian, membuktikan kebolehgunaan semula dan kestabilan strukturnya yang baik. Analisis teori mengesahkan bahawa tindakan $\pi\pi$ dan interaksi elektrostatik yang dicetuskan oleh kumpulan karboksil nanofiber selulosa terkarboksil menjadi faktor dominan dalam proses penjerapan bahan pencemar. Berbeza dengan aerogel nanofiber selulosa terkarboksil fotopemangkin konvensional, bahan ini mampu mengaktifkan peroksimonosulfat dengan cekap tanpa keperluan sinaran cahaya, lalu menjana oksigen singlet, radikal sulfat dan radikal hidroksil. Hal ini membolehkan penguraian mendalam dan mineralisasi sebatian organik

yang telah terjepap. Kumpulan karboksil nanofiber selulosa terkarboksil bersinergi dengan tapak sulfur-besi untuk menyatukan fungsi penjerapan dan pemangkinan dan bukan sekadar bertindak sebagai rangka struktur sahaja. Kajian ini menyediakan pendekatan yang praktikal untuk menghasilkan komposit berasaskan gentian berkecekapan tinggi, yang sesuai digunakan dalam sistem rawatan air sisa kompleks sebenar.

Kata kunci: Aerogel selulosa nanofiber; besi sifar valen tersulfidasi; pemulihan air sisa; penguraian; penjerapan

INTRODUCTION

Industrial modernization has intensified anthropogenic water pollution globally, seriously endangering ecological balance and human public health (Bochynska et al. 2024; Edo et al. 2024; Wang et al. 2024). The textile industry, an essential basic industry for social development, also discharges massive amounts of highchroma, toxic and refractory organic wastewater, bringing great difficulties to conventional water treatment engineering (Al-Tohamy et al. 2022; Yan et al. 2024; Zhong et al. 2026). Common remediation techniques for organic wastewater mainly include adsorption, membrane separation, coagulation, biological treatment and advanced oxidation processes (Shabir et al. 2022; Worku et al. 2025; Zhao et al. 2021). Among them, adsorption is widely favored in practical engineering due to its low economic cost and easy operational management. Activated carbon and sludge are the most traditional and commonly used adsorbents with stable pollutant removal effects (Guo et al. 2025; Liu et al. 2025; Wen et al. 2024). Nevertheless, such commercial adsorbents generally suffer from low adsorption capacity, poor cyclic regeneration performance and potential secondary pollution risks, restricting their large-scale long-term application (Bagotia 2025; Liu et al. 2024). Therefore, developing high-efficiency, recyclable and eco-friendly novel adsorbents is urgently required for the advanced purification of complex textile wastewater.

Graphene possesses a distinctive two-dimensional honeycomb sp^2 -hybridized structure with a large specific surface area and excellent mechanical strength, rendering it an advanced adsorbent for water remediation (Liu et al. 2022; Zhang et al. 2025b). Nevertheless, severe interlayer aggregation inevitably decreases available active sites, greatly restricting its practical water treatment feasibility (Gumaste & Kulkarni 2023). Graphene aerogels have been fabricated to effectively alleviate sheet stacking and maximize surface utilization, exhibiting prominent advantages in dye adsorption, including rich reactive sites and convenient solid-liquid separation (Debili, Fares & Messaoudi 2025; Yang et al. 2025). However, pure graphene aerogels commonly suffer from poor structural rigidity and unavoidable restacking behaviour, weakening their cyclic stability (Wang et al. 2025b). The introduction of functional polymers is a proven modification strategy to reinforce mechanical performance and optimize structural integrity (Luo et al. 2025). Typical composites, such as cellulose/graphene

oxide and polyvinyl alcohol/graphene oxide hybrid aerogels, effectively inhibit nanosheet collapse and achieve enhanced adsorption capability (Fu et al. 2024; Ou & Xiang 2025; Zhang et al. 2025a). Even so, conventional graphene-based adsorbents only achieve simple physical enrichment of pollutants without *in-situ* degradation. The inevitable adsorbent regeneration procedure and residual waste sludge easily trigger secondary environmental risks. Therefore, it is imperative to develop novel multifunctional graphene composites that integrate efficient adsorption and *in-situ* catalytic degradation to achieve sustainable wastewater treatment.

Metal-catalyzed peroxymonosulfate (PMS)-based advanced oxidation processes (AOPs) efficiently produce reactive oxygen species (ROS) for organic pollutant elimination, showing great potential in wastewater remediation (Wang et al. 2025a; Zhang et al. 2025c). Low-toxic, low-cost heterogeneous iron-based composites, including Fe^0 (Tang et al. 2023), $Fe-C_3N_5$ (Jiang, Du & Liu 2025), Fe-doped $ZnCo_2O_4$ (Lin et al. 2025), and Fe-MOFs-derived biochar composites (Xie et al. 2025), are widely applied to activate PMS with high catalytic efficiency. However, practical applications face key limitations. Complex water matrices and interfering ions easily consume ROS and weaken degradation efficiency. Fe^0 is also prone to oxidative deactivation during PMS reaction, further reducing catalytic performance. Moreover, conventional adsorbents require tedious regeneration procedures and easily cause secondary pollution. Most previous studies only optimize adsorption performance but ignore the recyclability and post-treatment of spent materials. Therefore, it is urgent to develop dual-functional composites with synchronous adsorption and catalytic degradation ability to achieve efficient, recyclable and eco-friendly wastewater treatment.

In this study, a sulfuriron modified carboxylated cellulose nanofiber/reduced graphene oxide aerogel (CCNF/rGO/Fe-S) was successfully fabricated via chemical reduction and freeze-drying to synergistically remove organic pollutants through combined adsorption and peroxymonosulfate (PMS) activation. Using graphene oxide as the basic framework and CCNF as the reinforcing skeleton, the optimized Fe-S doping significantly improved the mechanical stability and dual adsorption-catalytic performance of the composite. Electrostatic interaction and π - π stacking dominated the adsorption process, while $SO_4^{\cdot-}$ and $\cdot OH$ generated by PMS activation mainly contributed to pollutant

degradation. The prepared aerogel also exhibited reliable treatment efficiency in practical complex water environments, showing promising prospects for real wastewater remediation.

MATERIALS AND METHODS

Permanganate (KMnO_4), potassium peroxydisulfate (PMS), hydrochloric acid (HCl), sulfuric acid (H_2SO_4), hydrogen peroxide (H_2O_2), sodium borohydride (NaBH_4), phosphoric acid (H_3PO_4), methylene blue (MB), crystal violet (CV), rhodamine B (RhB), reactive red 195 (3BF), acid orange 7 (AO7), acid red1 (AR1), sodium dithionite ($\text{Na}_2\text{S}_2\text{O}_4$), potassium thiocyanate (KSCN), carboxylated cellulose nanofiber (CCNF) powder, cellulose nanofiber (CNF) powder, natural graphite flakes (100 mesh), tert-butyl alcohol (TBA), p-benzoquinone (p-BQ), and L-histidine were obtained from Sigma-Aldrich (Shanghai). Ferrous sulfate heptahydrate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$) was purchased from Macklin (Shanghai). The spin-trapping agent of 2,2,6,6-tetramethyl-4-piperidone (TEMP) and 5,5-dimethyl-pyrroline-N-oxide (DMPO) were provided by J&K Scientific Co., Ltd. All chemicals were analytical grade and used without further purification.

PREPARATION OF THE AEROGELS

Preparation of Graphene Oxide (GO)

GO was synthesized using a modified Hummers' method (Zaaba et al. 2017). Natural graphite flakes (1 g) and KMnO_4 (6 g) were combined in a 250 mL three-necked flask. H_3PO_4 (13 mL) and H_2SO_4 (120 mL) were slowly added to the flask with stirring in an ice bath. The flask was then transferred to an oil bath, heated to 50 °C, and allowed to react for 12 h with stirring. Subsequently, deionized water (380 mL) and H_2O_2 (10 mL) were sequentially added to terminate the reaction, followed by centrifugation to obtain a yellow precipitate. The precipitate was washed several times with 1 mol·L⁻¹ HCl and deionized water until the pH of the washing residue became neutral. Finally, the washed precipitate was ultrasonically dispersed in deionized water to obtain a GO dispersion.

Preparation of CCNF/rGO/Fe-S aerogel and CNF/rGO/Fe-S aerogel

CCNF powder (0.2 g) or CNF powder (0.2 g) was uniformly dispersed in deionized water (50 mL). The GO dispersion (5.7143 g, 1.75 wt%) and $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ solution (50 mL, 10 mg·mL⁻¹) were sequentially added to the CCNF dispersion and stirred for 30 min; next, a mixed solution of NaBH_4 and $\text{Na}_2\text{S}_2\text{O}_4$ (total volume, 80 mL; 0.225 mol·L⁻¹ NaBH_4 and 3.37 mmol·L⁻¹ $\text{Na}_2\text{S}_2\text{O}_4$) was added in a dropwise manner under a N_2 atmosphere. The mixture was allowed to react for 60 min, followed by centrifugation and washing multiple times with

deionized water to obtain a gel-like substance. The gel-like substance was placed in a mold, cooled in a refrigerator at -18 °C for 12 h, and freeze-dried for 48 h to obtain CCNF/rGO/Fe-S aerogel or CNF/rGO/Fe-S aerogel (S/Fe molar ratio, 0.3). To investigate the influence of the extent of sulfurization on the adsorption performance, CCNF/rGO/Fe-S aerogel with S/Fe molar ratios of 0.1-0.6 were prepared in a similar manner.

Preparation of the CCNF/rGO/Fe aerogel

CCNF powder (0.2 g) was uniformly dispersed in deionized water (50 mL). 5.7143 g of GO dispersion (1.75 wt%) and 50 mL of an $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ solution were sequentially added under a N_2 atmosphere. After stirring for 30 min, 80 mL of 0.225 mol·L⁻¹ NaBH_4 solution was added dropwise. The reaction was allowed to proceed for 60 min, after which the mixture was centrifuged and washed multiple times with deionized water to obtain a gel-like material. The gel was placed in a mold and frozen in a refrigerator for 12 h, followed by freeze-drying for 48 h to obtain the CCNF/rGO/Fe aerogel. CCNF aerogel, rGO aerogel, CCNF/rGO aerogel without the addition of $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ were prepared under identical conditions.

Characterization of the CCNF/rGO/Fe-S aerogel

The morphology and crystal structure of the CCNF/rGO/Fe-S aerogel was examined using scanning electron microscopy (SEM; Sigma 300, Zeiss, Germany), energy dispersive X-ray spectroscopy (EDS; Quantax Xflash 60 SDD, Bruker, Germany), and X-ray diffraction (XRD; D8 Advance, Bruker, Germany). The chemical composition and valence state were studied using X-ray photoelectron spectroscopy (XPS; K-Alpha, Thermo Scientific, USA). The mechanical properties were tested using a universal testing machine (Instron 5943, USA). Raman spectroscopy (LabRAM HR Evolution, Horiba, France) was employed to test the degree of graphitization and the defect level of CCNF/rGO/Fe-S aerogel before and after the adsorption of each dye at a laser wavelength of 785 nm. The zeta potential of the aerogel was determined using a zeta potential analyzer (Zetasizer Nano ZS, Malvern Panalytical, UK), and the conductivity was measured using an electrochemical workstation (CHI-760E, ChenHua, China).

Adsorption experiments

Dye powders were dissolved in deionized water to obtain simulated wastewater. The solution pH was adjusted to 2-12 using 0.1 mol·L⁻¹ HCl and 0.1 mol·L⁻¹ NaOH. For adsorption experiments, CCNF/rGO/Fe-S aerogel (40 mg) was added to dye solutions (40 mL) and held at 25-55 °C with stirring at 130 rpm for 30 min. Aliquots of supernatant were removed at certain intervals, and the absorbance was measured by ultraviolet-visible (UV-Vis) spectroscopy (UV-2550, Shimadzu, Japan). The

dye-adsorption capacity and dye-removal efficiency at time t were calculated as follows:

$$Q_t = (C_0 - C_t)V/m, \quad (1)$$

$$\text{Removal efficiency (\%)} = (C_0 - C_t)/C_0, \quad (2)$$

where Q_t ($\text{mg}\cdot\text{g}^{-1}$) is the adsorption capacity at time t (min); C_0 and C_t ($\text{mg}\cdot\text{L}^{-1}$) are the initial dye concentration and concentration at time t , respectively; V (L) is the volume of the dye solution; and m (g) is the mass of the aerogel.

Pseudo-first-order and pseudo-second-order kinetic models were used to investigate the adsorption kinetics, and their corresponding equations are expressed as follows:

$$Q_t = Q_e(1 - \exp(-k_1t)), \quad (3)$$

$$Q_t = Q_e(1 - 1/(1 + K_2Q_e')), \quad (4)$$

where Q_e ($\text{mg}\cdot\text{g}^{-1}$) is the adsorption capacity at equilibrium and K_1 and K_2 ($\text{g}\cdot\text{mg}^{-1}\cdot\text{min}^{-1}$) are the rate constants of the pseudo-first- and pseudo-second-order kinetic equations, respectively.

Commonly used adsorption isotherm models include the Langmuir and Freundlich models. The Langmuir model describes monolayer adsorption, in which the adsorbate is uniformly adsorbed on the adsorbent surface. The maximum adsorption capacity is reached when the monolayer adsorption sites are saturated. The equation is as follows:

$$Q_e = (Q_m K_L C_e)/(1 + K_L C_e), \quad (5)$$

where Q_m ($\text{mg}\cdot\text{g}^{-1}$) represents the maximum adsorption capacity; C_e ($\text{mg}\cdot\text{L}^{-1}$) is the equilibrium concentration; and K_L ($\text{L}\cdot\text{mg}^{-1}$) is the Langmuir constant related to the binding energy.

The Freundlich model describes multilayer adsorption on heterogeneous surfaces. The equation is as follows:

$$Q_e = K_F C_e^{1/n}. \quad (6)$$

where K_F and n are Freundlich constants representing the adsorption capacity and intensity, respectively. When $1/n = 0.1-0.5$, the adsorption process is favorable, whereas when $1/n > 2$, the adsorption process is unfavorable.

The measurement procedure for C_e is described as follows: Dye aqueous solutions (MB, RhB, CV) with various initial concentrations C_0 were prepared, followed by the addition of a fixed mass of CCNF/rGO/Fe-S aerogel to each solution at 25 °C. The resulting suspensions were magnetically stirred at 130 rpm until adsorption equilibrium was reached. Specifically, aliquots of supernatants were sampled

every 1 h for concentration analysis, and equilibrium was identified when two consecutive measurements exhibited nearly identical concentration values. After that, the supernatants were filtered through 0.22 μm aqueous filter membranes, and the absorbance of the filtrates was recorded using a UV-Vis spectrophotometer. The equilibrium liquid-phase concentration (C_e , $\text{mg}\cdot\text{L}^{-1}$) of MB, RhB and CV was quantified based on the pre-calibrated standard curves of the corresponding dyes.

DEGRADATION EXPERIMENTS

Adsorption was carried out using 40 mg of CCNF/rGO/Fe-S aerogel and a dye solution with a volume and concentration of 40 mL and 100 $\text{mg}\cdot\text{L}^{-1}$, respectively. The adsorption temperature, time, and stirring speed were 25 °C, 30 min, and 130 rpm, respectively. After the adsorption experiment, the aerogel was separated from the dye solution and subsequently immersed in PMS solution (40 mL, 1.5 $\text{g}\cdot\text{L}^{-1}$) for a 60 min degradation reaction. The reaction was carried out at room temperature (25 °C) with a stirring speed at 130 rpm and an initial pH of 2.73. The supernatant was removed after the reaction, and the absorbance was measured by UV-Vis spectroscopy (UV-2550, Shimadzu, Japan).

Cyclic adsorption experiments

After the degradation reaction, the aerogel was separated from the aqueous solution, rinsed with deionized water, and reused for the subsequent adsorption-degradation cycles. The regeneration efficiency of the CCNF/rGO/Fe-S aerogel was calculated as follows:

$$\text{Regeneration efficiency (\%)} = (Q_r/Q_0) \times 100\%, \quad (7)$$

where Q_0 and Q_r ($\text{mg}\cdot\text{g}^{-1}$) are the initial adsorption capacity and the adsorption capacity of the adsorbent after regeneration, respectively.

Adsorption and degradation mechanisms

FTIR and Raman spectroscopy were employed to investigate the adsorption mechanisms of the CCNF/rGO/Fe-S aerogel. Electron paramagnetic resonance (EPR) spectroscopy was conducted to study the reactive oxygen species (ROS). Spin-trapping was achieved using DMPO to capture $\bullet\text{OH}$ and $\text{SO}_4^{\bullet-}$ in aqueous solution; DMPO to capture superoxide anions ($\text{O}_2^{\bullet-}$) in methanol; and TEMP to $^1\text{O}_2$ in aqueous solution. XPS and XRD were used to study the CCNF/rGO/Fe-S aerogel following PMS activation.

RESULTS AND DISCUSSION

Synthesis and characteristics of CCNF/rGO/Fe-S aerogel

The CCNF/rGO/Fe-S aerogel was synthesized using CCNF, GO, and FeSO_4 with reducing agents, followed by freeze-drying (Figure 1(a)). The aerogel could steadily

stand on a flower petal without causing deformation, confirming its ultralight feature (Figure 1(b)). SEM results showed a typical layered porous structure with evenly loaded metal nanoparticles on the CCNF/rGO substrate (Figure 1(c)-1(d)). EDS elemental mapping further verified the uniform distribution of C, Fe, and S elements across the composite framework (Figure S(1) - data not shown). For comparison, the control CCNF/rGO/Fe aerogel displayed a similar porous layered morphology, indicating that sulfur doping barely changed the macroscopic structure (Figure S(2) - data not shown). Compressive mechanical tests demonstrated that the CCNF/rGO/Fe-S aerogel possessed much higher compressive stress than pure rGO aerogel under 70% strain (Figure 1(e)). The improved mechanical performance was mainly attributed to the rigid supporting effect of CCNF, which effectively reinforced the network structure and maintained structural integrity during compression. Figure S3(a) (data not shown) presents the N_2 adsorption-desorption isotherms of the CCNF/rGO/Fe-S aerogel recorded at relative pressures (P/P_0) ranging from 0 to 1. The Brunauer-Emmett-Teller (BET) specific surface area of the aerogel was calculated to be $496.66 \text{ m}^2 \cdot \text{g}^{-1}$ according to the isotherm data. The pore size distribution curve of the CCNF/rGO/Fe-S aerogel is shown in Figure S3(b)

(data not shown). It can be observed that the aerogel is dominated by mesopores, with a small amount of micropores and macropores coexisting, and the average pore diameter is 41.86 nm.

The crystal structures of the aerogels were characterized by XRD (Figure 1(f)). The CCNF/rGO/Fe-S aerogel showed distinct Fe^0 peaks at 44.5° and 64.5° (110, 200) (Zhang et al. 2025b), confirming Fe^0 formation. In contrast, the CCNF/rGO/Fe aerogel exhibited Fe_3O_4 peaks (220, 311, 400, 511, 440) without Fe^0 signals (Fe_3O_4 PDF#19-0629) (Liang et al. 2025), indicating the high oxidation resistance of Fe^0 in the sulfur-containing sample. XPS (Figure 1(g)) further showed that CCNF/rGO/Fe contained only Fe^{2+} and Fe^{3+} , while CCNF/rGO/Fe-S contained Fe^0 , Fe^{2+} , and Fe^{3+} . The Fe^{2+} peaks in the Fe-S sample shifted to higher binding energies due to the transition from Fe-O to Fe-S bonds, reducing ligand-to-metal charge transfer (Han et al. 2025). Thus, sulfur promotes Fe^0 formation, leading to a robust porous aerogel with homogeneously dispersed active phases.

Adsorption performance of CCNF/rGO/Fe-S aerogel

Methylene blue (MB), a typical toxic and high-chroma organic dye with severe ecological hazards, was selected as the model pollutant to evaluate the adsorption

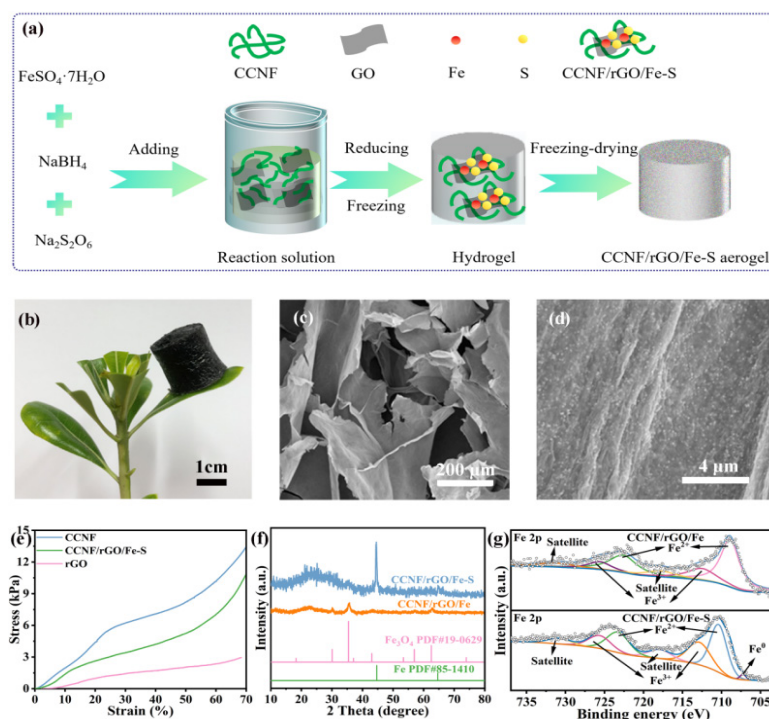


FIGURE 1. (a) Schematic illustration of the preparation process of CCNF/rGO/Fe-S aerogel; (b) digital photograph of CCNF/rGO/Fe-S aerogel; (c-d) SEM images of the CCNF/rGO/Fe-S aerogel; (e) compressive stress-strain curves of the CCNF, rGO, CCNF/rGO/Fe-S aerogels; (f) XRD patterns of the CCNF/rGO/Fe and CCNF/rGO/Fe-S aerogels; (g) high-resolution $\text{Fe} 2p$ spectrum of the CCNF/rGO/Fe and CCNF/rGO/Fe-S aerogels

capacity of CCNF/rGO/Fe-S aerogel. The effect of the S/Fe molar ratio on adsorption performance was first investigated to optimize synthesis conditions (Figure 2(a)). All groups with S/Fe ratios of 0.1-0.6 achieved over 98% MB removal within 30 min, and the optimal ratio of 0.3 realized the highest efficiency of 99.7%, which was adopted for all subsequent tests. A series of contrast materials, including pure rGO, CCNF, CCNF/rGO, CCNF/rGO/Fe and CNF/rGO/Fe-S, were further compared for MB adsorption (Figure 2(b)). The CCNF/rGO/Fe-S aerogel exhibited the best adsorption performance, far exceeding other counterparts, especially the CNF/rGO/Fe-S aerogel. The abundant carboxyl groups in CCNF effectively enhance electrostatic attraction to cationic MB molecules, greatly improving adsorption efficiency (Song et al. 2025). The porous structure of composite aerogels improves mass transfer compared with pure CCNF, while partial active site coverage slightly reduces performance relative to pure rGO. Fe loading slightly decreases

adsorption capacity due to site shielding, whereas sulfur doping enhances surface hydrophobicity and strengthens interfacial interaction with MB, thereby significantly boosting the overall adsorption performance of the optimized aerogel.

Further batch experiments evaluated the adsorption stability of CCNF/rGO/Fe-S under varying temperatures, pH values and inorganic ions. As temperature increased, MB adsorption efficiency slightly declined, and thermodynamic calculations confirmed spontaneous but entropy-reduced adsorption behavior (Figure 2(c)-2(d), Table S1 - data not shown) (Alnajrani & Alsager 2020). pH-dependent tests showed satisfactory adsorption performance across neutral to alkaline conditions, with removal rates consistently above 99%, while only strong acidic conditions (pH 2, pH 4) slightly lowered adsorption capacity (Figure S(4) - data not shown). Zeta potential tests showed that the aerogel surface maintained negative charges and became more electronegative at higher pH, verifying that electrostatic interaction

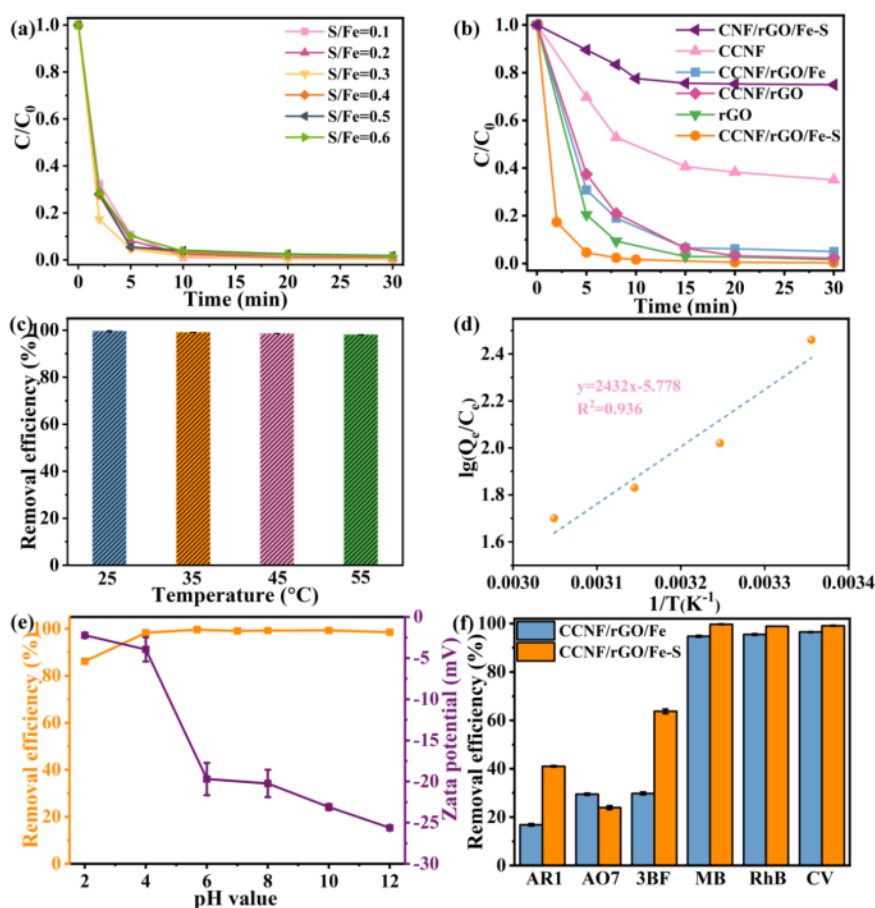


FIGURE 2. (a) The effect of S/Fe ratio on MB adsorption; (b) the adsorption performance of different adsorbents for MB (Cadsorbent = 1 g·L⁻¹, CMB = 200 mg·L⁻¹); (c) MB removal at different temperatures; (d) determination of the relationship between lg(Q_e/C_e) and 1/T in thermodynamic parameters; (e) effect of initial pH on adsorption of MB by the CCNF/rGO/Fe-S aerogel; (f) adsorption performance of CCNF/rGO/Fe and CCNF/rGO/Fe-S aerogels for different dyes (C_{dye} = 200 mg·L⁻¹)

dominates the adsorption process (Figure 2(e)). Common inorganic anions and cations barely interfered with MB adsorption; most ions maintained removal efficiency over 97% within 30 min. Only Ca^{2+} and Mg^{2+} slightly inhibited adsorption owing to competitive electrostatic attraction, yet the overall anti-interference ability remained excellent (Figure S(5)-S(6) - data not shown). Multiple dye adsorption tests demonstrated that sulfur modification significantly improved the removal performance toward cationic dyes including MB, RhB and CV. In contrast, both aerogels exhibited low removal efficiency for anionic dyes (AR1, AO7, 3BF), and the sulfidized sample showed no advantage for individual anionic dye (Figure 2(f)). The results confirm that the CCNF/rGO/Fe-S aerogel possesses obvious selective adsorption toward cationic organic dyes, showing great feasibility for practical complex dye wastewater treatment.

Binary mixed dye systems containing paired cationic and anionic dyes were employed to explore the selective adsorption performance of the CCNF/rGO/Fe-S aerogel. After 30 min of adsorption, UV-Vis spectra of different mixed dye solutions exhibited sharply weakened characteristic peaks of cationic dyes, while the absorption signals of anionic dyes remained almost unchanged (Figure S7(a)-7(c) - data not shown). The solution color also turned to the shade of residual anionic dyes, further verifying the preferential capture of cationic pollutants. These results confirm that the as-prepared aerogel possesses excellent selective adsorption ability toward cationic dyes, showing great potential for the targeted separation and purification of complex mixed dye wastewater.

Adsorption kinetic behaviors of MB, RhB and CV on the CCNF/rGO/Fe-S aerogel was investigated, and relevant adsorption curves showed rapid pollutant uptake within the initial 5-10 min before reaching adsorption equilibrium (Figure S8(a)-8(c) - data not shown). Kinetic fitting demonstrated that the pseudo-second-order model well matched experimental data, implying that the adsorption process was dominated by electron exchange and chemical adsorption mechanisms (Li et al. 2019; Manipadhy et al. 2020). Based on kinetic parameters, the initial adsorption rates of the aerogel toward MB, RhB, and CV were significantly higher than those of CCNF/rGO/Fe, which were 3.37, 3.98, and 3.30 times higher, respectively (Table S(2), Figure S(9) - data not shown). The faster adsorption kinetics benefited from abundant exposed active sites and strengthened interfacial affinity after sulfur doping. Adsorption isotherm results further showed that the Langmuir model exhibited higher correlation coefficients than the Freundlich model, confirming typical monolayer adsorption characteristics (Figure S8(d)-8(f), Table S(3) - data not shown). The maximum Langmuir adsorption capacities of CCNF/rGO/Fe-S for MB, RhB, and CV reached 757.1, 911.0, and 1281.4 $\text{mg}\cdot\text{g}^{-1}$, respectively,

which were 15.6%, 30.8%, and 33.2% higher than those of the original CCNF/rGO/Fe aerogel (Figure S(10) - data not shown). The CCNF/rGO/Fe-S aerogel exhibits superior adsorption capacities compared with most previously reported aerogel adsorbents (Table S4 - data not shown). The enhanced adsorption kinetics and equilibrium capacity further verify that sulfur-iron modification effectively optimizes the adsorption potential of composite aerogels for cationic dyes.

Catalytic degradation of the CCNF/rGO/Fe-S aerogel

CCNF/rGO/Fe-S aerogels saturated with MB were immersed in PMS solutions to explore the influence of diverse PMS dosages on the degradation of desorbed MB and the regeneration efficiency of aerogels, and the relevant results are displayed in Figure S11 - data not shown. As could be observed from the Figure S11(a) - data not shown, the aqueous MB concentration first increased and then decreased after immersing MB-saturated aerogels into PMS solution. The reason was that the acidic PMS medium (pH 2.73) induced partial desorption of MB through ion exchange, and the liberated MB was further removed via oxidative degradation. Furthermore, the degradation rates of MB were accelerated with increasing PMS dosage. At a PMS concentration of $1.5\text{g}\cdot\text{L}^{-1}$, nearly complete elimination of aqueous MB was achieved within 60 min, and the aerogel delivered a regeneration efficiency of 98.9% after oxidative regeneration (Figure S11(b) - data not shown). Accordingly, the optimal PMS dosage for catalytic degradation was determined to be $1.5\text{g}\cdot\text{L}^{-1}$.

Adsorption-regeneration cyclic experiments were conducted by alternating 30 min dye adsorption and 60 min PMS-based catalytic regeneration. After five consecutive cycles, the CCNF/rGO/Fe-S aerogel maintained high removal efficiencies of 87.1% (MB), 81.6% (RhB) and 89.6% (CV), with residual dye concentrations in the regenerated PMS solution consistently below $10\text{mg}\cdot\text{L}^{-1}$ (Figure 3(a)-3(b)). In contrast, the aerogel without PMS regeneration suffered severe performance degradation, retaining only extremely low adsorption capacity in the fifth cycle (Figure 3(c)). Such differences confirm that the composite can achieve *in-situ* regeneration by activating PMS to degrade adsorbed pollutants, efficiently recovering adsorption sites and avoiding secondary high-concentration wastewater discharge. Cyclic comparison further showed that the CCNF/rGO/Fe-S aerogel possessed much better reusability than the CCNF/rGO/Fe aerogel under identical experimental conditions (Figure 3(d)), highlighting its great practical potential for long-term stable wastewater remediation.

PMS consumption serves as an indicator of catalytic activity. Accordingly, the PMS consumption by the CCNF/rGO/Fe-S aerogel and the CCNF/rGO/Fe aerogel was investigated in the absence of dye molecules. As shown in Figure 4(a), after 60 min, only 27.6%

of PMS remained in the solution containing the CCNF/rGO/Fe-S aerogel, compared to 53.0% for the CCNF/rGO/Fe aerogel. This indicates the superior PMS activation capability of the CCNF/rGO/Fe-S aerogel. To further probe the electron transfer behavior, linear sweep voltammetry (LSV) was employed (Houam et al. 2023). Upon the addition of PMS to the electrolyte, a more pronounced reduction in current was observed for the CCNF/rGO/Fe-S aerogel than for the CCNF/rGO/Fe aerogel (Figure 4(b)-4(c)), suggesting more efficient electron transfer from the former to PMS. Electrochemical impedance spectroscopy (EIS) measurements were also conducted. In the Nyquist plot, the diameter of the semicircular arc corresponds to the charge-transfer resistance at the interface. Specifically, a larger diameter indicates higher resistance and lower electron transfer efficiency. As shown in Figure 4(d), the CCNF/rGO/Fe aerogel exhibited a larger arc diameter than the CCNF/rGO/Fe-S aerogel, confirming that the latter possesses lower charge-transfer resistance and superior electron transfer efficiency.

Adsorption and degradation mechanisms

Adsorption relies on dynamic interfacial equilibrium, where pollutants occupy the aerogel's porous active sites through physical or chemical forces, mainly including electrostatic attraction (Liu & Zhu 2026),

π - π stacking (Shi et al. 2025) and hydrogen bonding (Zhang et al. 2026). Combined with the negligible temperature effect on adsorption performance (Figure 2(c)), hydrogen bonding was excluded as a major driving force. Carboxyl groups on CCNF endow the aerogel with negative surface potential, strongly facilitating electrostatic adsorption toward cationic dyes, which was further verified by zeta potential analysis (You et al. 2023; Zhou et al. 2019). Raman spectra confirmed the existence of π - π interaction: after MB, RhB, and CV adsorption, the G band of rGO obviously shifted to higher wavenumbers, proving that dye molecules acted as electron acceptors and formed strong π - π stacking with the aerogel framework (Xiao et al. 2016) (Figure 5(a)). Overall, electrostatic interaction and π - π stacking jointly dominate the efficient dye adsorption of the CCNF/rGO/Fe-S aerogel. EPR tests further clarified the catalytic degradation mechanism by detecting reactive oxygen species (ROS) during PMS activation (Figure 5(b)-5(d)). The CCNF/rGO/Fe-S + PMS system exhibited prominent DMPO- $\cdot$ OH and DMPO-SO $_4^{\cdot-}$ adduct signals, much stronger than those of pure PMS, confirming effective generation of $\cdot$ OH and SO $_4^{\cdot-}$ radicals (Wang et al. 2017). No obvious difference in DMPO-O $_2^{\cdot-}$ intensity was observed between groups, indicating that O $_2^{\cdot-}$ radicals were negligible (Zhang & Zhang 2025). By contrast, significantly enhanced

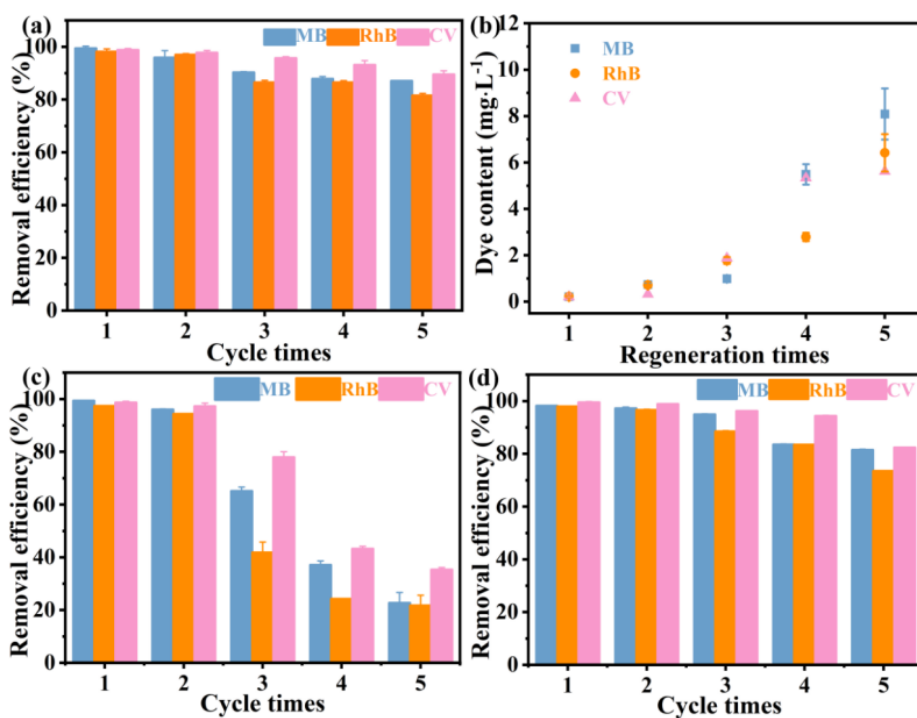


FIGURE 3. (a) Cyclic adsorption of MB, RhB, and CV by CCNF/rGO/Fe-S aerogel with catalytic degradation in PMS solution (1.5 g·L⁻¹); (b) residual concentrations of MB, RhB, and CV in PMS solution during cyclic degradation process; (c) cumulative adsorption of MB, RhB, and CV by CCNF/rGO/Fe-S aerogel without catalytic degradation; (d) cyclic adsorption of MB, RhB, and CV by CCNF/rGO/Fe aerogel with catalytic degradation in PMS solution (1.5 g·L⁻¹). In all experiments, the CCNF/rGO/Fe-S, CCNF/rGO/Fe, and dye concentration were 1 g·L⁻¹, 1 g·L⁻¹, and 100 mg·L⁻¹, respectively

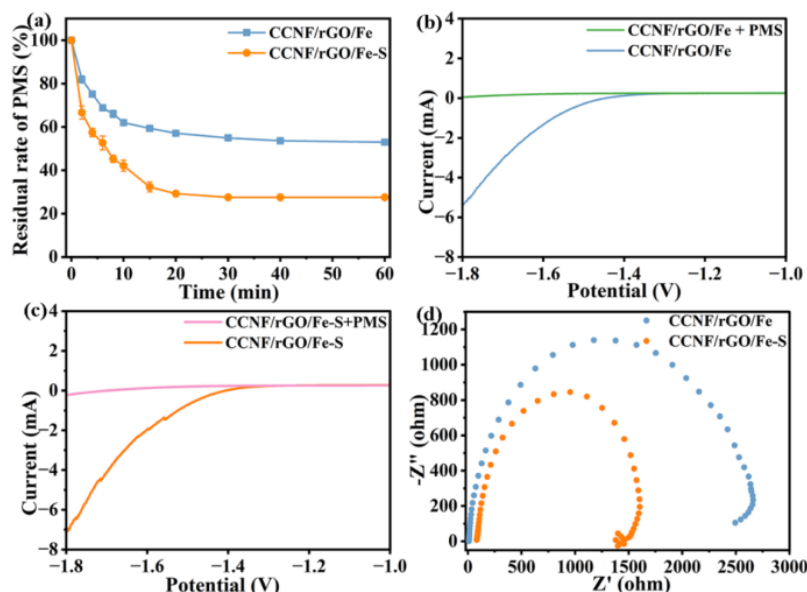
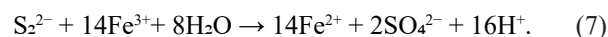
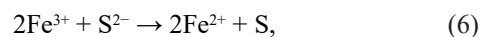
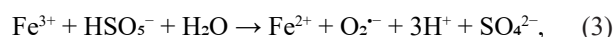
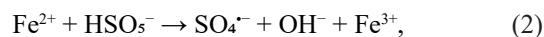
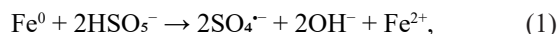


FIGURE 4. (a) Residual PMS in the CCNF/rGO/Fe and CCNF/rGO/Fe-S systems over time; (b) LSV curves of CCNF/rGO/Fe systems with and without PMS; (c) LSV curves of CCNF/rGO/Fe-S systems with and without PMS; (d) Nyquist plots of CCNF/rGO/Fe and CCNF/rGO/Fe-S

TEMP-102 signals verified the massive production of $^1\text{O}_2$ (Li et al. 2021). Radical quenching experiments further quantified ROS contributions (Figure S12 – data not shown). TBA was used to trap $\bullet\text{OH}$ and $\text{SO}_4^{\bullet-}$, *p*-BQ was used to trap $\text{O}_2^{\bullet-}$, L-histidine was used to trap $^1\text{O}_2$ (Di et al. 2025; Zhu et al. 2019). The addition of TBA sharply inhibited dye removal, proving that $\bullet\text{OH}$ and $\text{SO}_4^{\bullet-}$ served as major active radicals. *p*-BQ showed almost no inhibitory effect, while L-histidine only moderately reduced degradation efficiency. In conclusion, the aerogel can efficiently activate PMS to produce $\bullet\text{OH}$, $\text{SO}_4^{\bullet-}$ and $^1\text{O}_2$, synergistically degrading adsorbed organic dyes, in which $\bullet\text{OH}$ and $\text{SO}_4^{\bullet-}$ radicals play the decisive role.

XPS characterization was conducted to analyze the valence evolution of Fe and S on MB-adsorbed CCNF/rGO/Fe-S before and after PMS treatment (Figure 6). No Fe^0 signal was detected on the dye-loaded aerogel (Figure 6(a)), as surface-covered MB molecules shielded underlying active sites, which was consistent with XRD results (Figure S13 - data not shown). After PMS catalytic reaction, the relative proportion of Fe^{2+} decreased from 66.5% to 60.0% (Figure 6(b)), while Fe^{3+} content increased correspondingly, verifying that Fe^{2+} was oxidized by PMS. Meanwhile, low-valence sulfur species underwent significant transformation: S^{2-} signals completely disappeared, S^{2-} proportion decreased sharply, and oxidized SO_4^{2-} content increased remarkably (Figure 6(c)-6(d)). These results demonstrate that reductive S^{2-} and S_2^{2-} served as electron donors, efficiently promoting the cyclic conversion of Fe^{3+} to

Fe^{2+} and sustaining continuous PMS activation. Combined with electrochemical and radical evidence, a feasible synergistic catalytic mechanism was proposed. Electron-rich Fe^0 firstly triggered single-electron PMS activation to generate $\text{SO}_4^{\bullet-}$ and Fe^{2+} (Equation 1). The *in-situ* formed Fe^{2+} continuously activated PMS to produce additional radicals and regenerate Fe^{3+} (Equation 2). Reductive sulfur species synchronously reduced Fe^{3+} back to Fe^{2+} , realizing stable iron redox cycling. Synchronously, $\text{SO}_4^{\bullet-}$ hydrolyzed to produce $\bullet\text{OH}$, and the interaction between multiple radicals further generated abundant $^1\text{O}_2$ (Equations 3-5). Low-valence sulfur was partially oxidized and mineralized while driving iron circulation (Equations 6-7). Overall, the aerogel achieved efficient wastewater remediation via a two-step synergistic pathway: dyes were rapidly captured by electrostatic attraction and π - π stacking, and then *in-situ* generated ROS thoroughly degraded pollutants under the stable Fe-S synergistic catalytic system (Figure 6(e)).



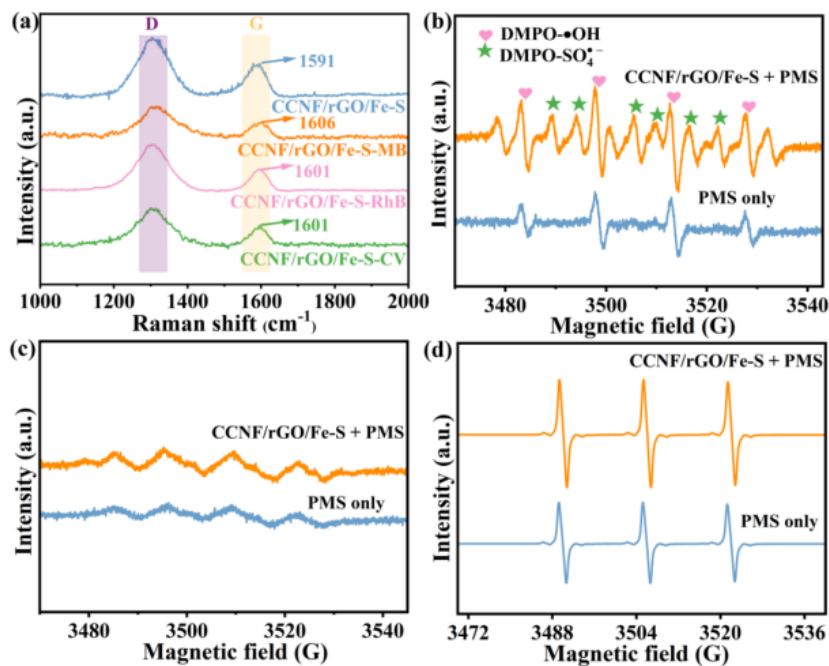


FIGURE 5. (a) Raman spectra of CCNF/rGO/Fe-S aerogel before and after the adsorption of MB, RhB, and CV; EPR spectra of (b) DMPO-•OH and DMPO-SO₄•⁻, (c) DMPO-O₂•⁻, and (d) TEMP-1O₂ adducts during the activation of PMS by CCNF/rGO/Fe-S aerogel

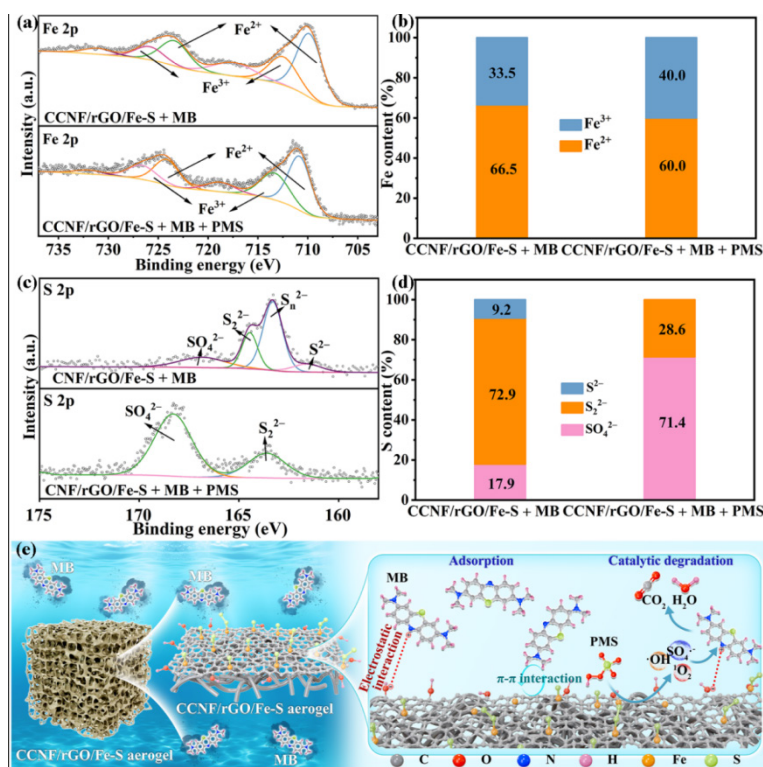


FIGURE 6. (a) High-resolution Fe 2p XPS spectra, (b) ratio of Fe²⁺ and Fe³⁺ species, (c) high-resolution S 2p XPS spectra, and (d) ratio of S₂²⁻, S₂²⁻, and SO₄²⁻ species for CCNF/rGO/Fe-S+MB system before and after PMS treatment, (e) schematic diagram the possible adsorption-degradation mechanism of dye onto CCNF/rGO/Fe-S aerogel

Treatment of simulated dye-containing wastewater

River water was employed as a substitute for deionized water to formulate a complex simulated dye-containing wastewater. The simulated wastewater was prepared by adding 100 mg·L⁻¹ MB, 500 mg·L⁻¹ glucose, 1000 mg·L⁻¹ sodium carbonate, 1000 mg·L⁻¹ sodium bicarbonate, 1500 mg·L⁻¹ sodium chloride, 500 mg·L⁻¹ sodium hydroxide, 200 mg·L⁻¹ ammonium chloride, and 100 mg·L⁻¹ sodium dodecyl sulfate to the river water sample. CCNF/rGO/Fe-S aerogel was then used to remove MB from the simulated wastewater. The results showed an initial MB-removal efficiency of 99.1%, which remained above 80% after six adsorption-regeneration cycles (Figure S14(a) - data not shown). The residual MB concentration in the PMS solution remained below 10 mg·L⁻¹ after five degradation processes (Figure S14(b) - data not shown). These results illustrate that the CCNF/rGO/Fe-S aerogel can effectively remove organic pollutants via the adsorption-regeneration process even in complex, real water matrices.

CONCLUSION

In summary, a dual-functional CCNF/rGO/Fe-S aerogel (with CCNF as the core skeleton) was successfully fabricated via chemical reduction and freeze-drying processes. This aerogel integrates synergistic dye adsorption and PMS-activated catalytic degradation, addressing key limitations of conventional adsorbents and single-function catalysts in wastewater treatment. Benefiting from CCNF's core skeleton role (providing carboxyl active sites and mechanical stability) and sulfur-iron modification, the CCNF/rGO/Fe-S aerogel selectively adsorbs cationic dyes. Its adsorption capacities for MB, RhB, and CV reach 757.1, 911.0, and 1281.4 mg·g⁻¹, which are 15.6%, 30.8%, and 33.2% higher, respectively, than those of the unmodified CCNF/rGO/Fe aerogel. Notably, its initial adsorption rates for the three dyes are over three times those of the unmodified aerogel, showing improved adsorption kinetics. After five adsorption-degradation cycles, the CCNF/rGO/Fe-S aerogel still maintains a pollutant removal rate exceeding 80%, outperforming the CCNF/rGO/Fe aerogel and indicating excellent reusability, stability, and practical application potential. Mechanistic investigations showed that the adsorption of pollutants is primarily governed by π - π stacking and electrostatic interactions (dominated by CCNF's carboxyl groups). Furthermore, the CCNF/rGO/Fe-S aerogel activates PMS to generate various reactive oxygen species (IO₂, SO₄⁻, and •OH), which effectively degrade the adsorbed organic pollutants and facilitate the self-regeneration of the aerogel. These findings highlight CCNF's critical role as the core skeleton in adsorption, and provide insights for developing high-efficiency, reusable fiber-based materials for treating organic pollutants in complex aquatic environments.

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