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Structure–Dispersion–Performance Relationship in Fe-Functionalized Silicon Oxide for Enhanced Degradation of Recalcitrant Azo Dyes

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Abstract

Recalcitrant azo dyes in textile wastewater are a significant environmental concern due to their persistence and potential impacts on human and ecosystem health. Heterogeneous Fenton-like processes offer a promising approach for their removal, although catalyst performance depends strongly on the physicochemical characteristics of the active material. This study investigated Fe-functionalized silicon oxide (Fe-SiO) catalysts for sodium percarbonate (SPC)-activated removal of Reactive Orange 84 (RO84). The catalysts were prepared using silicon oxide supports with and without thermochemical pretreatment and different amounts of Fe during synthesis. Their structural, morphological, and elemental characteristics were evaluated by transmission electron microscopy (TEM), X-ray diffraction (XRD), and energy-dispersive X-ray spectroscopy (EDS), complemented by textural analysis. Increasing the amount of Fe used during synthesis shifted removal from adsorption-influenced to oxidation-associated, as reflected in the kinetic behavior. Thermochemical pretreatment promoted more homogeneous Fe dispersion and reduced aggregation in the S2 series, consistent with its higher catalytic response under several evaluated conditions. The PFO model empirically described the observed removal kinetics. Overall, the Fe-SiO materials showed potential for SPC-assisted RO84 removal under the evaluated conditions.

Keywords: heterogeneous catalysis; degradation; textile wastewater; catalytic oxidation; wastewater treatment



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1. Introduction

The global textile industry is a cornerstone of the modern economy; however, it remains one of the largest contributors to water pollution. It is estimated that 10–15% of the dyes used in textile processing are lost during the dyeing stage and subsequently discharged in wastewater [1]. Among these pollutants, azo dyes are of particular concern because they contain one or more azo linkages ($-N=N-$) and typically possess complex, highly sulfonated polycyclic structures. Their presence not only degrades aesthetic water quality by increasing turbidity and color but also poses significant environmental and human health risks because partial reductive cleavage can form carcinogenic aromatic amines. As a result, azo dyes are highly resistant to conventional wastewater treatment processes, particularly aerobic biological treatment [1,2].

To overcome the limitations of conventional water treatment methods, Advanced Oxidation Processes (AOPs) have emerged as effective alternatives for degrading refractory organic contaminants. Their effectiveness stems from the in situ generation of reactive oxygen species (ROS), which non-selectively oxidize complex organic molecules [3,4]. Among AOPs, Fenton-like reactions, which rely on transition-metal catalysis to decompose oxidants such as hydrogen peroxide and sodium percarbonate (SPC), particularly Fe-containing species, have attracted considerable attention because of their operational simplicity and the abundance of Fe-based catalysts [3–6].

Despite their effectiveness in contaminant degradation, conventional Fenton systems based on hydrogen peroxide (H_2O_2) present several operational limitations, including the instability of H_2O_2 during transport, safety concerns associated with storage, and the requirement for acidic conditions to prevent catalyst precipitation [7–9]. As an alternative, SPC has gained increasing attention as a solid and safer source of H_2O_2 , offering improved logistical stability and an intrinsic pH-buffering capacity resulting from the release of carbonate species [10,11]. Consequently, developing heterogeneous Fenton-like systems based on SPC activation not only simplifies catalyst recovery and reduces sludge generation but also broadens the operational pH range toward neutral and alkaline conditions, thereby enhancing the practical feasibility of these processes for water treatment applications [12].

Fe-supported silicate materials, including silica xerogels and SiO_2 -coated structures, have shown considerable potential as heterogeneous catalysts [13,14]. Their catalytic performance is commonly associated with stabilizing and dispersing Fe-containing species on the silicate framework, which enhances the accessibility of redox-active Fe-containing sites and facilitates interfacial electron-transfer processes during catalytic oxidation [3,13]. In particular, amorphous or highly dispersed FeO_x species stabilized within silicate frameworks have been reported to exhibit superior catalytic activity to their crystalline counterparts under near-neutral conditions [3,15]. However, the nature of the active species and the dominant degradation pathways remain subjects of ongoing debate.

Despite significant advances in this field, important knowledge gaps remain. Although dye degradation has been extensively investigated, detailed studies of degradation mechanisms using Fe-modified silicate-supported catalysts remain limited [1,3]. The combined influence of catalyst textural properties and Fe precursor concentration on the transition from adsorption-influenced removal to greater oxidation-associated removal is not yet fully understood [16,17]. Similarly, the role of the oxidant-to-catalyst ratio in controlling oxidant activation and catalytic efficiency in percarbonate-based systems requires further systematic investigation [5,6].

Therefore, this study aimed to investigate the removal of Reactive Orange 84 through a heterogeneous Fenton-like process using Fe-functionalized silicate materials prepared by controlled impregnation. Specifically, this work aimed to: (i) characterize the catalysts using TEM, XRD, and EDS to evaluate their structural, morphological, and elemental characteris-

tics under different Fe amounts used during synthesis; (ii) assess their catalytic performance for SPC activation; (iii) determine the influence of catalyst dosage and SPC concentration on the observed removal kinetics; and (iv) evaluate the transition from adsorption-influenced removal toward a greater contribution of oxidation-associated removal.

2. Materials and Methods

2.1. Reagents

Microparticulate silicon oxide (SiO_2 , Kieselgel GF254, Type 60; $\geq 99\%$ purity, 70–230 mesh), supplied by Merck (Darmstadt, Germany), was used as the inorganic support. Reactive Orange 84 (RO84; dye content $\geq 95\%$, $\text{C}_{58}\text{H}_{30}\text{Cl}_2\text{N}_{14}\text{Na}_8\text{O}_{26}\text{S}_8$, MW = $1850.28 \text{ g mol}^{-1}$) was obtained from Química-Mexifibras (León, Mexico).

All reagents were of analytical grade and used as received. Ferrous sulfate heptahydrate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$, $\geq 99\%$) and potassium permanganate (KMnO_4 , $\geq 99\%$) were supplied by Productos Químicos Monterrey S.A. Sodium SPC ($\text{Na}_2\text{CO}_3 \cdot 1.5\text{H}_2\text{O}_2$, $\geq 98\%$) and manganese (II) sulfate monohydrate ($\text{MnSO}_4 \cdot \text{H}_2\text{O}$, $\geq 98\%$) were obtained from Golden Bell. Sulfuric acid (H_2SO_4 , 98%) and nitric acid (HNO_3 , 70%) were provided by Jamek, while phosphoric acid (H_3PO_4 , 85.8%) was purchased from J.T. Baker. Oxalic acid ($\text{H}_2\text{C}_2\text{O}_4$) was used as a primary standard.

All aqueous solutions were prepared using ultrapure deionized water ($18.2 \text{ M}\Omega \cdot \text{cm}$, Milli-Q system). To minimize premature oxidation of Fe^{2+} , stock solutions were prepared freshly each day in an acidified medium ($0.01 \text{ M H}_2\text{SO}_4$). The Fe^{2+} concentration was verified by titration with standardized KMnO_4 before the impregnation procedure.

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2.2. Synthesis of Fe-Functionalized Catalysts

2.2.1. Preparation of Fe-SiO

Fe-functionalized materials were synthesized by impregnation under controlled acidic conditions to favor Fe^{2+} surface complexation and minimize hydrolysis. Briefly, 2.0 g of SiO_2 was dispersed in 50 mL of $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ aqueous solutions at precursor concentrations of 1.8, 7.2, 14, and 28 mM corresponding to 0.010, 0.040, 0.078, and 0.156 wt% Fe in the impregnation solutions, respectively. These concentrations refer to the Fe precursor used during synthesis.

The suspension pH was carefully adjusted and maintained at 3.0 ± 0.05 using $1.0 \text{ M H}_2\text{SO}_4$. The suspension was stirred for 24 h at room temperature (600 rpm, $25 \pm 1^\circ \text{C}$, pH 3.0 ± 0.05), following established operational procedures used by our research group for the preparation and evaluation of functional materials [19,20], to provide sufficient contact time for interaction between the Fe^{2+} precursor and the SiO_2 surface before filtration and drying. The resulting solids were recovered by vacuum filtration (Whatman No. 42), washed with acidified deionized water (pH ≈ 3) to remove weakly bound species, and dried at 60°C for 48 h.

2.2.2. Preparation Fe-SiO (S2)

To modify the surface properties of the support and potentially improve Fe dispersion, a combined thermochemical activation was performed before Fe incorporation. First, the SiO_2 support was calcined at 550°C for 15 h (heating rate: $5^\circ \text{C min}^{-1}$) in a muffle furnace to remove residual organics and stabilize the porous structure. After calcination, 2.0 g of the calcined SiO_2 was treated with concentrated HNO_3 (13 M) under ultrasonic agitation (40 kHz) at 80°C for 4 h to modify the surface chemical and structural characteristics of the

SiO₂ support. The suspension was diluted, filtered, and repeatedly washed with deionized water until a neutral pH was achieved. The material was then dried overnight at 60 °C. Fe incorporation was subsequently carried out following the impregnation procedure described in Section 2.2.1.

2.3. Catalytic Degradation Experiments

Catalytic oxidation experiments were conducted in 250 mL jacketed borosilicate reactors containing 200 mL of RO84 aqueous solution (100 mg L⁻¹). The Fe-SiO catalyst was used at a dosage of 4 g L⁻¹. The selected concentration range was based on preliminary experiments and literature values to ensure measurable degradation within the experimental timeframe, as is commonly reported for dye degradation in Fenton-like systems [21,22]. The system temperature was maintained at 25 ± 1 °C using a thermostatic bath, and continuous stirring at 700 rpm was used to keep the catalyst suspended and promote mixing. External mass-transfer limitations were not independently evaluated by varying the stirring rate; therefore, their contribution cannot be quantitatively excluded. Internal diffusion effects were considered in relation to the catalyst's textural properties [13].

The Fe/percarbonate (Fe/SPC) system was evaluated at initial pH values of 3.0 and 6.0, adjusted using dilute H₂SO₄ or NaOH solutions. The pH increased during the reaction because SPC contributed alkalinity. In the experiments initiated at pH 3.0, the final pH reached approximately 11 at the end of the experimental period [6]. SPC (Na₂CO₃·1.5H₂O₂), used as the oxidant, serves as an in situ source of hydrogen peroxide, enabling Fenton-like reactions as previously described for percarbonate-based oxidation systems [5,23]. SPC concentrations of 20, 30, and 40 mM were investigated.

At predetermined time intervals (0–30 min), 3 mL aliquots were withdrawn and immediately treated with excess methanol to terminate further reaction before analysis [15]. The samples were filtered through 0.22 µm PTFE membranes to remove suspended catalyst particles before analysis. The filtered aqueous phase was also analyzed for dissolved Fe²⁺ by titration with standardized KMnO₄. All experiments were performed in sextuplicate (*n* = 6), and results are reported as mean ± standard deviation. Adsorption controls in the absence of SPC were conducted to evaluate the contribution of pollutant removal associated with the catalyst itself to decouple the contributions of adsorption and oxidative degradation, following standard practices in heterogeneous Fenton-like systems [24,25].

2.4. Characterization Techniques

2.4.1. Transmission Electron Microscopy (TEM) Analysis

The morphology, surface structure, and particle size distribution (PSD) of the Fe-functionalized materials (S1 and S2 series) were characterized using transmission electron microscopy. Powdered samples were prepared by dispersing the solids in high-purity ethanol through an ultrasonic process, after which a droplet of the resulting suspension was deposited onto carbon-coated copper grids. Micrographs were collected at magnifications of 4000×, 10,000×, and 20,000× using Digital Micrograph™ (Version 3.7.1. 1999) to evaluate particle size distribution, dispersion, and homogeneity of Fe-containing species, and potential aggregation effects within the silicate framework.

To ensure statistical reliability, PSD was determined by analyzing multiple representative images and fitting the experimental data to a log-normal distribution function Equation (1)

$$y = y_0 + \frac{A}{\sqrt{2\pi}wx} \exp\left(-\frac{\left[\ln\left(\frac{x}{x_c}\right)\right]^2}{2w^2}\right), \quad (1)$$

where y represents the frequency of particles with size x , y_0 is the baseline offset, A corresponds to the area under the curve, $x(c)$ represents the characteristic mean particle size, and w is the dispersion parameter. The goodness of fit was rigorously evaluated using the adjusted coefficient of determination (R^2). This structural difference may be associated with the thermochemical pretreatment of the support, which could influence Fe dispersion and surface accessibility [26,27].

2.4.2. EDS Analysis and Fe Content

Energy-dispersive X-ray spectroscopy (EDS) was performed to complement the TEM characterization and evaluate the elemental composition and spatial distribution of Fe within the catalyst particles. Multiple regions of the catalyst particles were analyzed for each sample. The Fe content was estimated as the average Fe weight percentage (wt%) obtained from the analyzed EDS regions for each catalyst.

2.4.3. XRD Analysis

X-ray diffraction (XRD) patterns were collected using $\text{CuK}\alpha$ radiation ($\lambda = 1.54056 \text{ \AA}$) over a 2θ range of $4\text{--}60^\circ$, step size 0.05° , and 10 s per step. Phase identification was performed using X'Pert HighScore Plus 3.0 [28]. RO84 concentration was determined using a Thermo Scientific Evolution 300 (Waltham, MA, USA) spectrophotometer at 492 nm; a calibration curve for RO84 was constructed over $5\text{--}120 \text{ mg L}^{-1}$, showing excellent linearity ($R^2 > 0.995$).

UV–Vis analysis was primarily used to assess chromophore degradation and decolorization; this study did not directly assess complete mineralization, as is common in dye degradation studies [21,22].

The relative concentration (C/C_0) and removal efficiency were calculated as in Equations (2) and (3):

$$\frac{C}{C_0} = \frac{A}{A_0}, \quad (2)$$

$$\text{Removal (\%)} = \left(1 - \frac{C}{C_0}\right) \times 100, \quad (3)$$

2.5. Kinetic Analysis

The degradation kinetics were analyzed using a pseudo-first-order (PFO) model to describe RO84 disappearance under the evaluated reaction conditions [3,21,22]. The resulting rate constants are therefore reported as pseudo-first-order rate constants (k_{obs}). Uncertainties in the kinetic parameters were derived from the standard error of the regression slope, and the uncertainty in $t_{1/2}$ was propagated accordingly according to standard statistical procedures.

The initial degradation rate (r_0) was calculated from the apparent pseudo-first-order rate constant and the initial RO84 concentration using $r_0 = k_{\text{obs}}C_0$, where C_0 was 100 mg L^{-1} . Initial rates are reported in $\text{mg L}^{-1} \text{ min}^{-1}$.

3. Results

3.1. XRD Characterization

The diffraction pattern of the silicate material (Figure 1) indicates that its crystalline structure is predominantly composed of silicon dioxide (SiO_2) in the α -cristobalite phase (PDF 01-082-0512) [29,30]. The most intense reflection at $2\theta \approx 21.6^\circ$ corresponds to the (101) plane of α -cristobalite, indicating a high degree of silicate-framework crystallinity. Additional peaks located at 28.0° , 31.0° , 35.8° , and 41.3° are consistent with secondary reflections of the same phase, indicating the absence of other polymorphs (e.g., quartz,

tridymite). The sharpness and relative intensity of these signals are consistent with a relatively well-ordered crystalline structure, typical of thermally stable silicate materials.

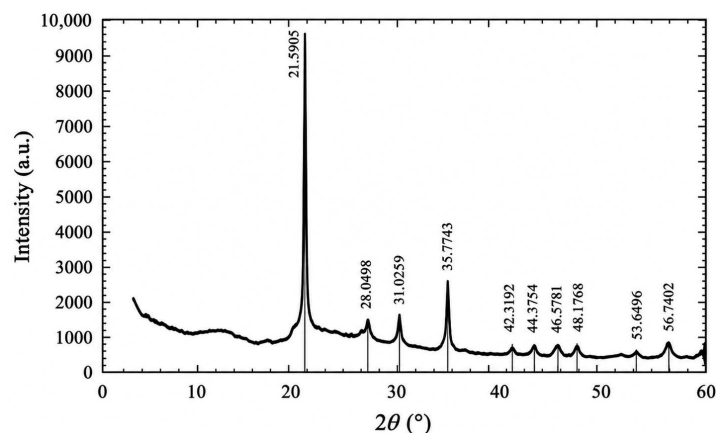


Figure 1. X-ray diffraction pattern of the pristine silicate material (DT), showing α -cristobalite (SiO_2) as the predominant crystalline phase.

The diffraction maxima were sharp and well-defined, indicating a relatively ordered crystalline structure, as reported in previous studies of thermally processed silicate materials where crystalline and disordered regions coexist [31]. Previous studies of thermally processed silica materials have reported the coexistence of crystalline and poorly ordered structural contributions, which may influence their mechanical and surface properties [32].

The X-ray diffraction (XRD) patterns in Figure 2 suggest progressive silicate matrix structural transformation after Fe incorporation for different synthesis conditions. The pristine sample (DT) exhibits sharp and intense reflection centered at $2\theta \approx 21.6^\circ$, characteristic of α -cristobalite, confirming the predominance of an ordered silica framework. After Fe addition, all modified samples retain a structurally stable silicate backbone even at the highest Fe concentrations. However, the gradual broadening and attenuation of peaks in S1_14 mM Fe, S1_28 mM Fe, S2_1.8 mM Fe, and S2_28 mM Fe indicate changes in the degree of structural order following Fe incorporation [33,34].

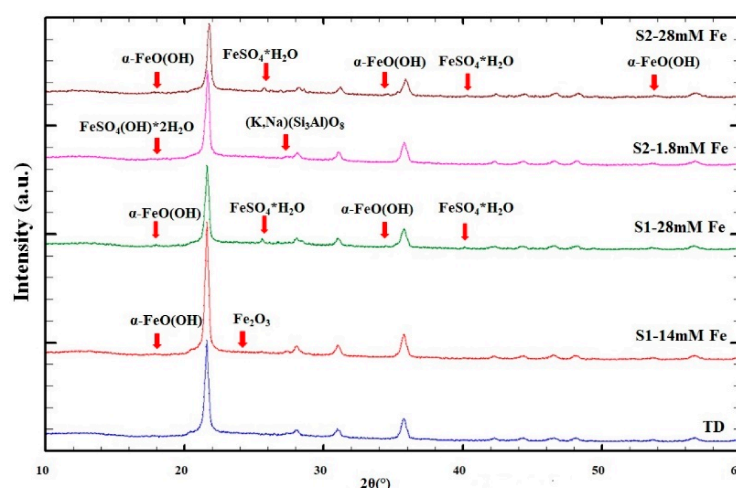


Figure 2. Superposition of X-ray diffraction patterns of Fe-functionalized materials, showing α -cristobalite (SiO_2) as the dominant phase and the emergence of additional Fe-containing crystalline phases upon Fe incorporation.

The observed changes in peak broadening and intensity indicate changes in the local structure following Fe incorporation [33,35]. Similar structural modifications have been

reported for transition-metal-modified silicate materials and have been associated with changes in local coordination environments [35].

After Fe incorporation, different crystalline phases were observed in the different materials. For example, Fe_2O_3 (PDF 00-024-0072) was detected in S1_14mMFe, while $\alpha\text{-FeO(OH)}$ (PDF 00-017-0536) and $\text{FeSO}_4\cdot\text{H}_2\text{O}$ (PDF 01-074-1332) were observed in S1_28mMFe and S2_28mMFe. $\text{FeSO}_4(\text{OH})\cdot 2\text{H}_2\text{O}$ (PDF 00-025-0409) and $(\text{K,Na})(\text{Si}_3\text{Al})\text{O}_8$ (PDF 00-019-1227) were also detected in S2_1.8mMFe. The S2 samples showed a greater diversity of Fe-containing phases, particularly at 28 mM Fe. The coexistence of Fe-containing crystalline phases with the silicate matrix suggests that Fe incorporation resulted in the formation of surface-associated and/or extra-framework Fe-containing species. However, XRD cannot quantitatively establish their specific distribution [27,35].

The identification of $\alpha\text{-FeO(OH)}$ is particularly relevant because iron oxyhydroxides have been widely investigated as active phases in heterogeneous Fenton and Fenton-like systems [36]. The presence of hydrated Fe sulfate phases further indicates the incorporation of Fe-containing species during the impregnation process. FeO_x species associated with silica have also been reported to contribute to interfacial oxidant activation in heterogeneous Fenton-like systems [36].

The structural changes observed after Fe incorporation, including peak broadening and attenuation, may reflect modifications in the local structural environment [33]. Such changes may favor the accessibility and distribution of surface-associated Fe-containing species, facilitating their interaction with RO84 and the oxidant and potentially contributing to the observed catalytic response. The retention of $\alpha\text{-cristobalite}$, along with the observed Fe-containing crystalline phases, indicates that the silicate support's main crystalline structure was preserved after Fe incorporation. In contrast, some synthesis conditions showed variations in structural order.

The differences between the S1 and S2 series highlight how synthesis conditions influence the Fe-containing phases identified by XRD. S2 samples exhibited a greater diversity of Fe-containing crystalline phases and more pronounced peak broadening, particularly at 28 mM Fe precursor concentration. These observations indicate differences in structural order and the distribution of Fe-containing crystalline phases between the two series. The XRD-observed structural differences are consistent with complementary TEM observations showing differences in Fe dispersion and aggregation [37].

The average crystallite size was estimated using the Scherrer equation [30]. The calculated values ranged from 24.0 to 33.5 nm (Table 1). The calculated crystallite sizes varied among the samples, indicating differences in the apparent size of the coherently diffracting crystalline domains. Changes in peak broadening may reflect these differences in the coherence of the crystalline domains. Because peak broadening can arise from crystallite size as well as other contributions, including instrumental broadening and microstructural effects, the Scherrer-derived values are considered comparative estimates rather than direct measurements of particle size or defect density [30].

Table 1. Average Crystallite Size of Fe-Functionalized Materials.

Sample/Treatment	S1	S2
DT	31.6 nm	
DT + 1.8 mM Fe	29.9 nm	28.1 nm
DT + 7.2 mM Fe	33.5 nm	29.9 nm
DT + 14 mM Fe	29.8 nm	23.9 nm
DT + 28 mM Fe	26.0 nm	31.2 nm

3.2. TEM Characterization

TEM images of S1 (non-pretreated support) are shown in Figure 3a,b. As shown, irregular morphology characteristic of siliceous supports, with regions of variable electron density, can be identified. Dark-contrast domains associated with Fe-containing species are clearly visible, indicating Fe deposition on the silicate surface, with a tendency toward agglomeration and the formation of clusters with heterogeneous size distributions, as reported in previous studies [38]. Without pretreatment, greater aggregation of Fe-containing species may restrict surface-site accessibility and contribute to the differences in catalytic performance observed between S1 and S2 (Figure S1).

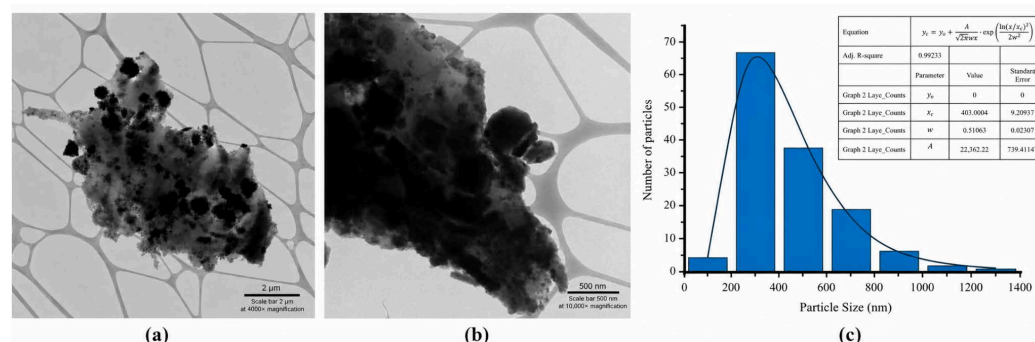


Figure 3. Characterization of sample S1: (a) TEM micrograph, scale bar 2 μm at 4000 $\times$ magnification; (b) TEM micrograph, scale bar 500 nm at 10,000 $\times$ magnification; (c) particle size distribution histogram fitted to a log-normal function.

XRD and TEM results highlight the association between Fe dispersion, silicate-framework structure, and catalytic performance, emphasizing the importance of synthesis strategies that minimize metal agglomeration and promote a more uniform distribution of Fe-containing species.

The observed non-uniform dispersion is consistent with surface-associated Fe-containing species, with the heterogeneous distribution suggesting limited spatial uniformity of Fe incorporation within the silicate matrix. The observed aggregation of Fe-containing species is consistent with the structural features identified by XRD (Figure 2), where partial segregation and increased crystallization of Fe-containing phases become evident at higher Fe precursor concentrations, indicating greater formation of Fe-containing crystalline phases [36].

Particle size distribution analysis for S1 was fitted to a log-normal function ($R^2 \approx 0.76$), yielding an average particle size of 1159 nm, with a broad distribution extending beyond 4000 nm (Figure 3c), confirming the highly polydisperse nature of the system, dominated by large aggregates. From a catalytic perspective, agglomeration may reduce the effective surface area and limit access to accessible Fe-containing sites, which could contribute to the lower catalytic efficiency observed for S1 [3]. Similar behavior has been reported in recent studies on Fe-based supported materials, where large aggregate formation was associated with reduced dispersion homogeneity, partial pore obstruction, and diminished interfacial accessibility of deposited metal species [36].

In contrast, TEM images of S2 (pretreated support, Figure 4a,b) show a markedly different morphology compared to S1. The S2 material exhibits a more homogeneous morphology, with Fe-containing species more uniformly distributed and a pronounced reduction in agglomeration and particle size compared with S1.

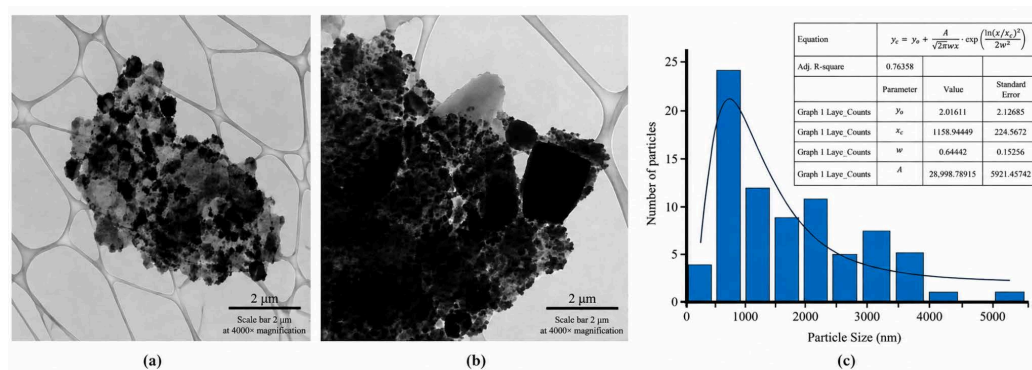


Figure 4. Characterization of sample S2: (a) TEM micrograph, scale bar 200 nm at 20,000 $\times$ magnification; (b) TEM micrograph, scale bar 500 nm at 10,000 $\times$ magnification; (c) particle size distribution histogram fitted to a log-normal function.

We hypothesize that the thermochemical pretreatment applied to the silicate support increased the availability of surface sites for Fe-containing species and facilitated their immobilization. The XRD patterns (Figure 2) show peak broadening and reduced diffraction intensity in S2, indicating increased structural disorder and smaller coherent crystallite domains after Fe incorporation. These structural changes are consistent with the more homogeneous distribution of Fe-containing species observed by TEM.

When fitted to a log-normal model, the S2 particle size distribution analysis (Figure 4c) showed an excellent fit ($R^2 \approx 0.99$). The average particle size was approximately 403 nm, representing a reduction of about 65% compared with S1. Additionally, the lower dispersion parameter ($w \approx 0.51$) indicates a narrower particle size distribution and reduced polydispersity [39].

3.3. Energy-Dispersive X-Ray Spectroscopy (EDS) Analysis

EDS analysis provided complementary information on the Fe content of the catalyst particles. Table 2 summarizes the average Fe contents obtained from the analyzed EDS regions. Fe content could not be reliably determined for the 7.2 mM samples. For the samples for which reliable EDS measurements were obtained, the Fe content increased with increasing Fe amount used during synthesis. However, the magnitude of this increase differed between the two support treatments, with S1 showing higher Fe contents than S2 at the corresponding Fe precursor concentrations. These differences are consistent with the heterogeneous Fe distribution observed in the TEM images.

Table 2. Average Fe content obtained from EDS analysis of the Fe-functionalized silicate catalysts.

Fe mM	Fe Content (wt%) S1
S1_1.8	0.41
S1_7.2	Not determined
S1_14	2.70
S1_28	21.07
S2_1.8	0.36
S1_7.2	Not determined
S2_14	1.49
S2_28	10.30

The observed differences in Fe content are consistent with the spatial heterogeneity observed in the TEM images (See Supplementary Information).

3.4. BET Characterization

N₂ adsorption–desorption measurements were performed to characterize the textural properties of the Fe-functionalized silicate (Fe-SiO) catalysts. The analyses were carried out using a static volumetric method with N₂ as the adsorbate at 77.35 K. Before analysis, the samples were degassed at 150 °C for 4 h. The BET specific surface area was calculated from the N₂ adsorption data, while total pore volume and average pore diameter were obtained from the corresponding analysis. The BET method was applied according to the Brunauer–Emmett–Teller model [40], and pore-size analysis was performed using the Barrett–Joyner–Halenda (BJH) approach [41].

Table 3 summarizes the BET surface area, total pore volume, and average pore diameter for the analyzed catalysts. The corresponding N₂ adsorption–desorption isotherms are presented in Figure 5, while the comparison of BET surface areas among the analyzed catalysts is presented in Figure S2.

Table 3. Textural properties of the Fe-SiO catalysts. N/A: BET characterization was not available for this sample in the present dataset.

Catalyst	BET Surface Area (m ² g ^{−1})	Total Pore Volume (cm ³ g ^{−1})	Average Pore Diameter (nm)
S1_1.8	2.43	0.00672	11.06
S1_7.2	2.20	0.00749	13.59
S1_14	N/A	N/A	N/A
S1_28	2.92	0.00802	10.99
S2_1.8	116.60	0.19388	6.65
S2_7.2	2.21	0.00678	12.26
S2_14	3.45	0.01006	11.66
S2_28	12.00	0.03723	12.41

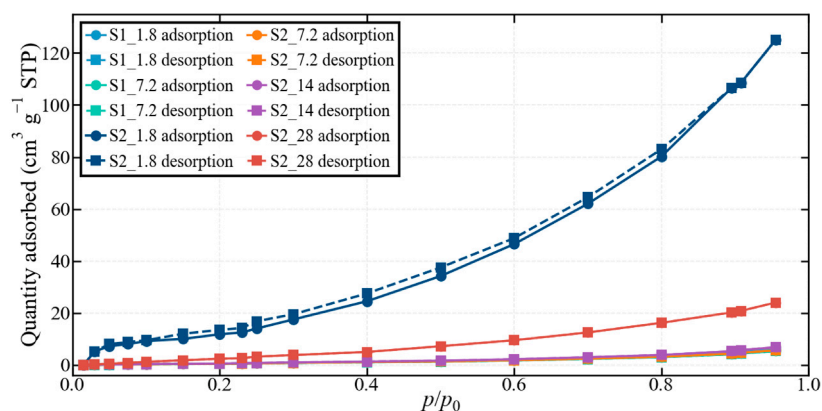


Figure 5. N₂ adsorption–desorption isotherms of the Fe-SiO catalysts measured at 77.35 K.

The measured BET surface areas ranged from 2.20 to 116.60 m² g^{−1} among the analyzed catalysts. S2_1.8 exhibited the highest BET surface area (116.60 m² g^{−1}), together with a total pore volume of 0.19388 cm³ g^{−1} and an average pore diameter of 6.65 nm. In comparison, S1_1.8 exhibited a BET surface area of 2.43 m² g^{−1} and a total pore volume of 0.00672 cm³ g^{−1}. At an Fe precursor concentration of 7.2 mM, the BET surface areas of S1_7.2 and S2_7.2 were very similar, with values of 2.20 and 2.21 m² g^{−1}, respectively. At 28 mM, the BET surface area of S2_28 (12.00 m² g^{−1}) was higher than that of S1_28 (2.92 m² g^{−1}). BET characterization was not available for S1_14 in the present dataset; therefore, Table S1 reports no textural parameters for this sample.

For the samples for which pore-size distribution data were available, the measured pore volume was predominantly associated with the mesoporous range. For example,

approximately 70.12% of the reported pore volume of S2_1.8 was located between 2 and 10 nm, with an average pore diameter of 6.65 nm. S1_28 and S2_28 exhibited average pore diameters of 10.99 and 12.41 nm, respectively. These results indicate predominantly mesoporous textural characteristics for the analyzed materials.

The t-plot analysis reported zero micropore area and zero micropore volume for the analyzed samples. Accordingly, the t-plot analysis detected no measurable microporosity. The BET results therefore provide complementary information regarding the surface area and pore characteristics of the catalysts. These results were used to assess whether differences in textural properties could contribute to the observed differences in catalytic performance, without assigning the catalytic activity exclusively to surface area.

Importantly, BET surface area does not directly quantify the density, oxidation state, or chemical nature of Fe-containing catalytic sites. Therefore, the differences in catalytic performance were not attributed to surface area alone but were considered together with the TEM and XRD characterization results.

3.5. Catalytic Degradation

The catalytic degradation of RO84 using Fe-functionalized silicate materials activated with SPC exhibited a clear dependence on the Fe precursor concentration used during synthesis and oxidant dosage. As shown in Figure 6a, the kinetic plots at the lowest Fe precursor concentrations (1.8 and 7.2 mM) showed shallow slopes and relatively low linearity, suggesting that adsorption strongly influenced dye removal under these conditions [13,42].

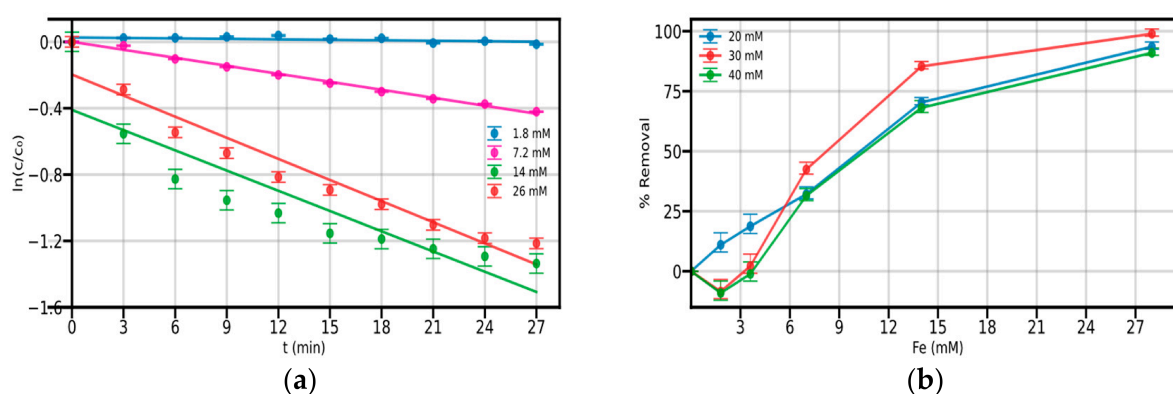


Figure 6. Catalytic degradation performance of RO84 using Fe-SiO catalysts under different Fe precursor and SPC concentrations. (a) Pseudo-first-order linearized plots ($\ln(C/C_0)$ vs. time) showing increasing contribution of reaction-associated removal relative to adsorption degradation with increasing Fe precursor concentration. (b) RO84 removal efficiency as a function of Fe precursor concentration at different SPC dosages (20, 30, and 40 mM). The error bars represent the standard deviation obtained from six independent measurements ($n = 6$). Their small magnitude relative to the plotted values makes them difficult to distinguish at some points.

Control experiments were also performed under the evaluated conditions, including no-catalyst, no-SPC, Fe-free SiO_2 , unloaded Fe materials, and homogeneous FeSO_4 /SPC conditions. None of these control conditions showed a significant catalytic contribution to RO84 removal.

Increasing the Fe precursor concentration from 7.2 to 14 mM improved the linearity of the pseudo-first-order plots ($R^2 > 0.95$), indicating a better fit of the kinetic model under these conditions (Figure 6a). At higher Fe precursor concentrations (14–28 mM), the system showed its highest apparent catalytic performance, with k_{obs} values up to 0.195 min^{-1} and RO84 removal based on UV-Vis measurements at 30 mM SPC (Figures 6b and 7b).

This behavior may reflect differences in the accessibility and distribution of Fe-containing species generated under the different synthesis conditions [15].

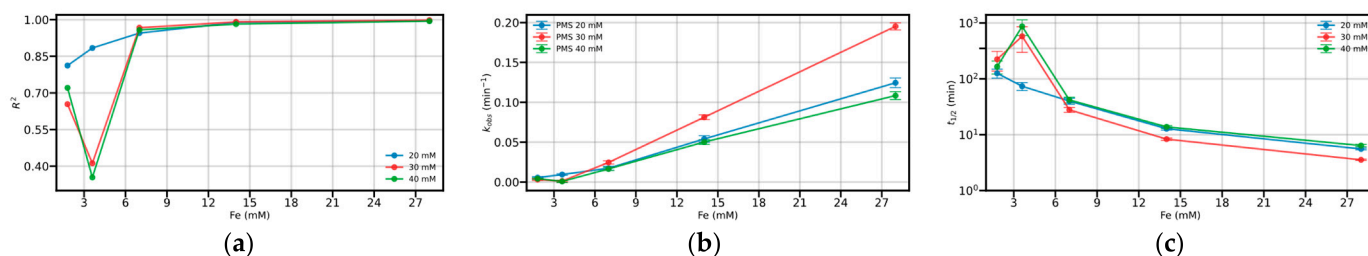


Figure 7. Effect of Fe precursor concentration on the pseudo-first-order kinetic parameters of RO84 degradation at different SPC concentrations (20, 30, and 40 mM). (a) Coefficient of determination (R^2) for pseudo-first-order model fitting as a function of Fe precursor concentration. (b) Apparent pseudo-first-order rate constant (k_{obs}) as a function of Fe precursor concentration. (c) Half-life time ($t_{1/2}$) as a function of Fe precursor concentration. The error bars represent the standard deviation obtained from six independent measurements ($n = 6$). Their small magnitude relative to the plotted values makes them difficult to distinguish at some points.

Complementary COD measurements showed reduced chemically oxidizable matter after RO84 treatment under the evaluated conditions. The COD results provide additional evidence of changes in the treated solution beyond the decrease in RO84 absorbance observed by UV–Vis. However, these results were not interpreted as direct evidence of complete mineralization.

At 30 mM SPC, the highest k_{obs} value was obtained under the conditions evaluated, whereas increasing the SPC concentration to 40 mM did not further increase k_{obs} . This behavior suggests that increasing the oxidant concentration beyond 30 mM did not further improve the degradation kinetics under the conditions evaluated.

Structural characterization supports these catalytic trends. TEM analysis showed a more homogeneous distribution of Fe-containing species and reduced aggregation in the S2 catalysts. XRD patterns indicated changes in structural order following Fe incorporation. These structural features may favor greater accessibility of Fe-containing sites and are consistent with the improved catalytic response observed for S2 under several of the conditions evaluated [30].

The higher catalytic response observed for S2 is consistent with the more homogeneous Fe dispersion and reduced aggregation observed by TEM. Textural differences may also contribute under some conditions; however, the BET results do not show a consistent difference between S1 and S2 across the entire series. For example, the BET surface areas of S1_7.2 and S2_7.2 were very similar (2.20 and $2.21 \text{ m}^2 \text{ g}^{-1}$, respectively), whereas S2_28 exhibited a higher surface area than S1_28 (12.00 vs. $2.92 \text{ m}^2 \text{ g}^{-1}$). Therefore, the present dataset cannot quantitatively separate the contribution of surface area from that of Fe dispersion. The improved Fe dispersion and reduced aggregation observed for S2 provide the more direct structural evidence associated with its catalytic response, while a contribution from textural properties cannot be excluded.

3.6. Kinetic Analysis

RO84 kinetic degradation behavior was evaluated using the pseudo-first-order (PFO) model [22]. The PFO model was selected because the kinetic experiments followed the temporal decrease in RO84 concentration under fixed initial concentration conditions, allowing the degradation profiles to be described through the observed rate constant (k_{obs}) (Figure 7b). The regression coefficient values (R^2) progressively approached unity at intermediate and high Fe concentrations (Figure 7a), indicating that the PFO model

adequately described the experimental degradation profiles under these conditions. The observed rate constant (k_{obs}) increased with Fe concentration, reaching a maximum of 0.195 min^{-1} at 30 mM SPC. Using mg L^{-1} , the corresponding initial degradation rate was $19.5 \text{ mg L}^{-1} \text{ min}^{-1}$.

The increase in k_{obs} was accompanied by a decrease in $t_{1/2}$ (Figure 7c), confirming enhanced RO84 degradation kinetics with increasing Fe concentration. However, the reduction in $t_{1/2}$ became less pronounced at higher Fe concentrations, indicating that the kinetic improvement was not proportional across the entire concentration range. This trend is consistent with the relationship $t_{1/2} = \ln(2)/k_{\text{obs}}$, whereby the pronounced decrease in $t_{1/2}$ at lower Fe concentrations corresponds to the greater increase in k_{obs} observed in this range. In comparison, the subsequent plateau reflects the progressively smaller changes in k_{obs} at higher Fe concentrations. This behavior may be related to differences in the accessibility and distribution of Fe-containing species, which can influence oxidant activation under the conditions evaluated. Similar relationships between Fe dispersion, site accessibility, and degradation kinetics have been reported for Fe/SiO₂ and FeOx-supported catalysts [15].

The SPC experiments also showed the influence of oxidant dosage. The highest degradation performance was obtained at 30 mM SPC, whereas increasing the concentration to 40 mM did not further improve performance. This response may reflect changes in oxidant utilization at higher SPC concentrations, as reported for heterogeneous Fenton-like systems, where increased oxidant dosage can promote non-productive consumption [4,7].

The catalysts' structural features are consistent with these kinetic trends. The improved Fe dispersion and reduced particle aggregation observed by TEM, together with the structural changes identified by XRD, may increase the accessibility of Fe-containing surface sites and favor more effective interactions among RO84, the oxidant, and the catalyst surface.

The kinetic analysis indicates that the system evolves from a weakly active, adsorption-influenced regime at low Fe precursor concentrations toward a greater contribution of oxidation-associated removal at higher Fe precursor concentrations. The combination of higher k_{obs} , reduced half-life, and improved model fitting is consistent with an increased contribution of catalyst-associated processes under the conditions evaluated. This behavior may be associated with Fe-induced structural changes.

4. Conclusions

The Fe-functionalized silicon oxide materials exhibited different RO84 removal behaviors depending on the Fe precursor concentration, SPC dosage, and support pretreatment. Increasing the Fe precursor concentration shifted the regime from adsorption-influenced to more oxidation-associated removal, as reflected in the observed kinetic behavior. At higher Fe precursor concentrations, the improvement in the apparent removal kinetics became less pronounced.

Thermochemical pretreatment of the SiO₂ support produced differences in the structural and morphological characteristics of the catalysts. TEM showed that the S2 series had more homogeneous Fe dispersion and less aggregation, consistent with its higher catalytic response under several evaluated conditions. However, the present dataset could not quantitatively separate the contributions of Fe dispersion, surface area, and crystalline Fe-containing phases, and no specific Fe phase could be identified as the dominant active phase.

The PFO model provided an empirical description of the observed RO84 removal kinetics, with higher k_{obs} and lower half-life values under conditions exhibiting greater

removal rates. The Fe precursor concentrations investigated represent synthesis conditions, and the actual bulk Fe loading of the solid catalysts was not quantitatively determined.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/pr14182899/s1>, Figure S1: Additional TEM images of the Fe-SiO catalysts. Top row: S1 catalysts prepared using untreated SiO₂ support at Fe precursor concentrations of 1.8 mM (a, $\times 10,000$; b, $\times 50,000$) and 28 mM (c, $\times 10,000$; d, $\times 50,000$). Bottom row: S2 catalysts prepared using thermally-chemically pretreated SiO₂ support at Fe precursor concentrations of 1.8 mM (e, $\times 10,000$; f, $\times 50,000$) and 28 mM (g, $\times 10,000$; h, $\times 50,000$).; Figure S2: BET surface area of the Fe-SiO catalysts.; Table S1: Local elemental composition obtained by EDS (not representative of bulk Fe loading).

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Abbreviations

The following abbreviations are used in this manuscript:

AOPs	Advanced Oxidation Processes
Fe-SiO	Iron-functionalized Silicon Oxide
H ₂ O ₂	Hydrogen Peroxide
ROS	Reactive Oxygen Species
RO84	Reactive Orange 84
SPC	Sodium Percarbonate
TEM	Transmission Electron Microscopy
XRD	X-ray Diffraction
PSD	Particle Size Distribution
PFO	Pseudo-First-Order
PTFE	Polytetrafluoroethylene
UV-Vis	Ultraviolet-Visible Spectroscopy
SiO ₂	Silicon Dioxide
FeOX	Iron Oxides
α -FeO(OH)	Iron Oxyhydroxide (Goethite Phase)
Fe ₂ O ₃	Iron(III) Oxide
PDF	Powder Diffraction File (crystallographic reference database)
R ²	Coefficient of Determination
k _{obs}	Apparent Pseudo-First-Order Rate Constant
t _{1/2}	Half-Life Time

References

- Al-Ghouti, M.A.; Khraisheh, M.A.M.; Allen, S.J.; Ahmad, M.N. The Removal of Dyes from Textile Wastewater: A Study of the Physical Characteristics and Adsorption Mechanisms of Diatomaceous Earth. *J. Environ. Manag.* **2003**, *69*, 229–238. [\[CrossRef\]](#) [\[PubMed\]](#)
- Catrinescu, C.; Neamtu, M.; Yediler, A.; Macoveanu, M.; Kettrup, A. Catalytic wet peroxide oxidation of an azo dye, Reactive Yellow 84, over Fe-exchanged ultrastable Y zeolite. *Environ. Eng. Manag. J.* **2002**, *1*, 177–186. [\[CrossRef\]](#)
- Wang, J.; Tang, J. Fe-Based Fenton-like Catalysts for Water Treatment: Catalytic Mechanisms and Applications. *J. Mol. Liq.* **2021**, *332*, 115755. [\[CrossRef\]](#)
- Pignatello, J.J.; Oliveros, E.; MacKay, A. Advanced Oxidation Processes for Organic Contaminant Destruction Based on the Fenton Reaction and Related Chemistry. *Crit. Rev. Environ. Sci. Technol.* **2006**, *36*, 1–84. [\[CrossRef\]](#)
- Miao, Z.; Gu, X.; Lu, S.; Dionysiou, D.D.; Al-Abed, S.R.; Zang, X.; Wu, X.; Qiu, Z.; Sui, Q.; Danish, M. Mechanism of PCE Oxidation by Percarbonate in a Chelated Fe(II)-Based Catalyzed System. *Chem. Eng. J.* **2015**, *275*, 53–62. [\[CrossRef\]](#)
- Zhang, S.; Hu, X.; Li, L.; Huangfu, X.; Xu, Y.; Qin, Y. Activation of Sodium Percarbonate with Ferrous Ions for Degradation of Chlorobenzene in Aqueous Solution: Mechanism, Pathway and Comparison with Hydrogen Peroxide. *Environ. Chem.* **2018**, *14*, 486–494. [\[CrossRef\]](#)
- Li, L.; Niu, X.; Zhang, D.; Ye, X.; Zhang, Z.; Liu, Q.; Ding, L.; Chen, K.; Chen, Y.; Chen, K.; et al. A Systematic Review on Percarbonate-Based Advanced Oxidation Processes in Wastewater Remediation: From Theoretical Understandings to Practical Applications. *Water Res.* **2024**, *259*, 121842. [\[CrossRef\]](#) [\[PubMed\]](#)
- Han, Y.; Han, H.; Si, Y.; Xu, L.; Su, D.; Sun, C. WS₂@FeBC Assisted Fe(III)-Activated Percarbonate for Roxarsone and Tetracycline Simultaneous Removal: Interfacial Fenton-like Process Enhanced by Oxygen Vacancies and Tungsten Redox. *Sep. Purif. Technol.* **2025**, *354*, 129426. [\[CrossRef\]](#)
- Huang, J.; Li, N.; Zhou, Z.; Xu, Z.; Ali, M.; Zeng, G.; Yang, R.; Zhou, Z.; Danish, M.; Lyu, S. Comparative Studies on Trichloroethylene Degradation by Fe Foam Catalyzing Three Hydrogen Peroxide-Based Oxidants. *J. Environ. Chem. Eng.* **2022**, *10*, 107335. [\[CrossRef\]](#)
- Jin, X.; Wang, Y.; Huang, Y.; Huang, D.; Liu, X. Percarbonate Activation Catalyzed by Nanoblocks of Basic Copper Molybdate for Antibiotics Degradation: High Performance, Degradation Pathways and Mechanism. *Chin. Chem. Lett.* **2024**, *35*, 109499. [\[CrossRef\]](#)
- Chen, K.; Li, T.; Zhang, X.; Lei, B.; Li, Z.; Xu, Y. CuCoFe-LDHs Activated Sodium Percarbonate (SPC) for the Degradation of Ciprofloxacin. *J. Environ. Chem. Eng.* **2023**, *11*, 110651. [\[CrossRef\]](#)
- Xu, X.; Zhou, Y.; Li, S.; Lin, Y.; Zhang, S.; Guan, Q. Activation of Sodium Percarbonate with Fe₃O₄ @MXene Composite for the Efficient Removal of Bisphenol A. *J. Environ. Chem. Eng.* **2022**, *10*, 108702. [\[CrossRef\]](#)
- Le, A.; Pham, T. *Activation of Hydrogen Peroxide by Iron-Containing Minerals and Catalysts in Circumneutral PH Solutions: Implications for Ex Situ and In Situ Treatment of Contaminated Water and Soil*; University of California: Berkeley, CA, USA, 2012.
- Coelho, J.V.; Guedes, M.S.; Prado, R.G.; Tronto, J.; Ardisson, J.D.; Pereira, M.C.; Oliveira, L.C.A. Effect of Iron Precursor on the Fenton-like Activity of Fe₂O₃/Mesoporous Silica Catalysts Prepared under Mild Conditions. *Appl. Catal. B* **2014**, *144*, 792–799. [\[CrossRef\]](#)
- Pham, A.L.T.; Lee, C.; Doyle, F.M.; Sedlak, D.L. A Silica-Supported Iron Oxide Catalyst Capable of Activating Hydrogen Peroxide at Neutral PH Values. *Environ. Sci. Technol.* **2009**, *43*, 8930–8935. [\[CrossRef\]](#) [\[PubMed\]](#)
- Xiong, J.; Li, Y.; Pang, C.; Li, G.; Hu, C. Removal of Pollutants in Banknote Printing Wastewater by Mesoporous Fe/SiO₂ Prepared from Rice Husk Pyrolytic Residues. *Environ. Sci. Pollut. Res.* **2019**, *26*, 16000–16013. [\[CrossRef\]](#) [\[PubMed\]](#)
- Mahy, J.G.; Tasseroul, L.; Herlitschke, M.; Hermann, R.P.; Lambert, S.D. Fe³⁺/Iron Oxide/SiO₂ Xerogel Catalysts for p-Nitrophenol Degradation by Photo-Fenton Effects: Influence of Thermal Treatment on Catalysts Texture. *Mater. Today Proc.* **2016**, *3*, 464–469. [\[CrossRef\]](#)
- Michałowski, T.; Batorowicz, A.; Madej, A.; Kochana, J. Extended Gran Method and Its Applicability for Simultaneous Determination of Fe(II) and Fe(III). *Anal. Chim. Acta* **2001**, *442*, 287–293. [\[CrossRef\]](#)
- Valencia-Espinoza, N.; Abreu-Naranjo, R.; Medina-Ganem, L.F.; Medina-Ganem, R.E.; Méndez-Landin, F.G.; Vega-Rios, A.; Quevedo-Castro, A.; Picos-Benítez, A.R.; Bandala, E.; Rodríguez-Narvaez, O.M. Functionalized Agave Bagasse Hydrochar for Reactive Orange 84 Removal: Synthesis, Characterization, and ANN-GA Optimization. *Processes* **2025**, *14*, 10. [\[CrossRef\]](#)
- Medina-Ganem, L.F.; Valencia-Espinoza, N.; Bautista-Peñuelas, E.; Medina-Ganem, R.E.; Vega-Rios, A.; Peña-Cruz, M.I.; Bandala, E.R.; Quevedo-Castro, A.; Pacheco-Álvarez, M.; Rodríguez-Narvaez, O.M. Comparative Life Cycle Assessment of Acid-Activated Hydrochar Under Different Feedstocks. *Processes* **2026**, *14*, 1200. [\[CrossRef\]](#)
- Imene Aouni, S.; Benguerba, Y.; Ghodbane, H.; Boubli, A.; Lakikza, I.; Boualleg, L.; Abdel-Halim, K.S.; Albrahim, M.; Merouani, S. Degradation of Basic Fuchsin Using Iron Salt-Activated Sodium Percarbonate in a Homogeneous Fenton System. *Sep. Purif. Technol.* **2025**, *374*, 133628. [\[CrossRef\]](#)

22. Rache, M.L.; García, A.R.; Zea, H.R.; Silva, A.M.T.; Madeira, L.M.; Ramírez, J.H. Azo-Dye Orange II Degradation by the Heterogeneous Fenton-like Process Using a Zeolite Y-Fe Catalyst-Kinetics with a Model Based on the Fermi's Equation. *Appl. Catal. B* **2014**, *146*, 192–200. [\[CrossRef\]](#)
23. Danish, M.; Gu, X.; Lu, S.; Brusseau, M.L.; Ahmad, A.; Naqvi, M.; Farooq, U.; Zaman, W.Q.; Fu, X.; Miao, Z. An Efficient Catalytic Degradation of Trichloroethene in a Percarbonate System Catalyzed by Ultra-Fine Heterogeneous Zeolite Supported Zero Valent Iron-Nickel Bimetallic Composite. *Appl. Catal. A Gen.* **2017**, *531*, 177–186. [\[CrossRef\]](#) [\[PubMed\]](#)
24. Taheri, E.; Fatehizadeh, A.; Khiadani, M.; Ghasemian, M.; Bedia, J. Application of Natural Hematite as Catalyst in Heterogeneous Photo-Fenton like Process for 2,4-Dichlorophenol Degradation. *J. Photochem. Photobiol. A Chem.* **2025**, *459*, 116057. [\[CrossRef\]](#)
25. Wang, Z.; Zhu, S.; Deng, J.; Li, H.; Wang, L.; Luo, H.; Tang, Z.; Li, X. Facile Preparation of Fe₃O₄@SiO₂ Derived from Iron-Rich Sludge as Magnetic Catalyst for the Degradation of Organic Contaminants by Peroxymonosulfate Activation. *Sustainability* **2022**, *14*, 16419. [\[CrossRef\]](#)
26. Bahloul, A.; Benayahoum, A.; Bouakkaz, S.; Bordjiba, T.; Boudjahem, A.-G.; Amirouche, S.; Zoukel, A.; Fodil Cherif, N.; Boudriche, L.; Bachari, K. Microwave-Assisted One-Pot Synthesis of Cu/Cu₂O Nanoparticles for Enhanced Catalytic Degradation of Organic Dyes. *J. Mol. Liq.* **2026**, *444*, 129191. [\[CrossRef\]](#)
27. Wu, Y.; Wang, C.; Wang, L.; Hou, C. Recent Advances in Iron Oxide-Based Heterojunction Photo-Fenton Catalysts for the Elimination of Organic Pollutants. *Catalysts* **2025**, *15*, 391. [\[CrossRef\]](#)
28. Degen, T.; Sadki, M.; Bron, E.; König, U.; Nénert, G. The HighScore Suite. *Powder Diffr.* **2014**, *29*, S13–S18. [\[CrossRef\]](#)
29. Kabekkodu, S.N.; Dosen, A.; Blanton, T.N. PDF-5+: A Comprehensive Powder Diffraction File™ for Materials Characterization. *Powder Diffr.* **2024**, *39*, 47–59. [\[CrossRef\]](#)
30. Scherrer, P. Nachr Ges Wiss Goettingen. *Math. Phys.* **1918**, *2*, 98–100.
31. Masai, H.; Kohara, S.; Wakihara, T.; Shibazaki, Y.; Onodera, Y.; Masuno, A.; Sukenaga, S.; Ohara, K.; Sakai, Y.; Haines, J.; et al. Author Correction: Siliceous Zeolite-Derived Topology of Amorphous Silica. *Commun. Chem.* **2025**, *8*, 23. [\[CrossRef\]](#) [\[PubMed\]](#)
32. Gupta, A.; Pandey, V.; Singh, S.K.; Yadav, M.K.; Majhi, M.R. Microstructural Characterization and Hydrothermal Ageing Resistance of Rice Husk Silica-Doped Alumina Toughened Zirconia Biocomposite. *Silicon* **2025**, *17*, 2457–2467. [\[CrossRef\]](#)
33. Lan, H.; Wang, J.; Cheng, L.; Yu, D.; Wang, H.; Guo, L. The Synthesis and Application of Crystalline–Amorphous Hybrid Materials. *Chem. Soc. Rev.* **2024**, *53*, 684–713. [\[CrossRef\]](#) [\[PubMed\]](#)
34. Liu, Y.; Luo, Y.; Ye, J.; Zhao, L.; Chen, Y.; Dai, Y.; Xie, Y.; Wang, Y.; Zhang, Y. Photocatalytic Hydrogen Production Performance of Anatase and Brookite TiO₂ Based on Oxygen Vacancies and Noble Metal Pt Modification. *J. Photochem. Photobiol. A Chem.* **2026**, *479*, 117242. [\[CrossRef\]](#)
35. Li, Y.; Dong, H.; Xiao, J.; Li, L.; Hou, Y.; Chu, D.; Hou, X.; Xiang, S.; Dong, Q. Ascorbic Acid-Assisted Iron Silicate Composite Activated Peroxydisulfate for Enhanced Degradation of Aqueous Contaminants: Accelerated Fe(III)/Fe(II) Cycle and the Interaction between Iron and Silicate. *Chem. Eng. J.* **2023**, *455*, 140773. [\[CrossRef\]](#)
36. Ren, G.; Zhang, J.; Jing, H. FeOx(OH)_y Domains Chemically Anchored on Silica via Fe-O-Si Bonds: Interfacial Structure, Heterogeneous Fenton-like Catalysis, and Tungsten CMP Performance. *Colloids Surf. A Physicochem. Eng. Asp.* **2026**, *744*, 140650. [\[CrossRef\]](#)
37. Park, H.; Na, G.-J.; Kim, J.-C.; Ryoo, R. Assessment of Acid Catalytic Properties of Ferrosilicate MFI Zeolite by Methanol-to-Hydrocarbon Conversion. *RSC Adv.* **2024**, *14*, 29006–29013. [\[CrossRef\]](#) [\[PubMed\]](#)
38. Barberia-Roque, L.; Viera, M.; Bellotti, N. Hygienic Coatings with Nano-Functionalized Diatomaceous Earth by Equisetum Giganteum—Mediated Green Synthesis. *Nano-Struct. Nano-Objects* **2023**, *36*, 101055. [\[CrossRef\]](#)
39. Mota dos Santos, A.A.; Cordeiro, G.C. Investigation of Particle Characteristics and Enhancing the Pozzolan Activity of Diatomite by Grinding. *Mater. Chem. Phys.* **2021**, *270*, 124799. [\[CrossRef\]](#)
40. Brunauer, S.; Emmett, P.H.; Teller, E. Adsorption of Gases in Multimolecular Layers. *J. Am. Chem. Soc.* **1938**, *60*, 309–319. [\[CrossRef\]](#)
41. Barrett, E.P.; Joyner, L.G.; Halenda, P.P. The Determination of Pore Volume and Area Distributions in Porous Substances. I. Computations from Nitrogen Isotherms. *J. Am. Chem. Soc.* **1951**, *73*, 373–380. [\[CrossRef\]](#)
42. Nidheesh, P.V. Heterogeneous Fenton Catalysts for the Abatement of Organic Pollutants from Aqueous Solution: A Review. *RSC Adv.* **2015**, *5*, 40552–40577. [\[CrossRef\]](#)

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