

Research Article

Hydroxide Sludge/Hydrochar-Fe Composite Catalysts for Photo-Fenton Degradation of Dyes

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The photo-Fenton oxidation process was employed to degrade methylene blue (MB) using a hydroxide sludge/hydrochar-Fe composite as a catalyst prepared by physical activation of raw hydroxide sludge from a drinking water treatment plant and hydrochar-Fe prepared by hydrothermal carbonization from two-phase olive mill waste. The prepared composite was characterized by XRD, SEM, EDS, ICP, and FT-IR. The effect of major parameters, including pH, H₂O₂ concentration, and a dose of composite on the removal of MB has been studied. The results indicated that the MB decolorization rate increased with the increase of H₂O₂ concentration and catalyst addition; however, further increase in H₂O₂ concentration and catalyst dosage could not result in an increase of MB removal efficiency. A high degradation of 95% was achieved within 150 min under UV light irradiation at natural pH (pH = 5), a catalyst loading of 2.5 g/L, a H₂O₂ dosage of 14.68 mol/L, and MB concentration of 50 mg/L. Recycling studies show a MB decolorization of 92% after three cycles and the use of the composite for the degradation of another dye (methyl orange) shows a degradation of 99%, demonstrating that this composite is a promising heterogeneous photo-Fenton catalyst for long-term removal of dyes from industrial wastewater.

1. Introduction

Hydroxides sludge (HS) is a byproduct from the coagulation-flocculation process using aluminum- or iron-based salts to precipitate clay, colloidal particles, algae, and humic substances from water resources [1]. The chemical composition of HS varies depending on the source of water under treatment as well as the type of coagulant used. Due to its high production rate and its environmentally unfavored disposal to landfill, several researchers paid a considerable attention for using this waste in different applications especially those of low cost. These applications include utilization of HS for removal of heavy metals [2], ceramic products [3], cement, and concrete production [4], as coagulant and sorbent for olive mill wastewater [5] and wastewater treatment [6, 7].

Wastewater management, in particular colored waste water, is a major occupation for textile, paper pulp, plastic,

and other industries. These effluents are difficult to be treated because of its high concentration on dyes, which have a synthetic origin and a complex molecular structure that makes them more stable and hardly biodegradable [8]. Several studies list the numerous impacts of dyes on the environment and human health [9–11] and the necessity of their removal from colored wastewater before its discharge [12–14]. Methylene blue (MB) is a good representative of cationic azo dyes, which does not decompose under ambient conditions and is usually resistant to classical biological treatment. Conventional physicochemical techniques such as adsorption, coagulation, and reverse osmosis are the most frequently used to remove MB from aqueous solutions [15]. However, these methods can merely accumulate MB and cannot convert the dye into being harmless.

Recent developments in the field of water treatment have led to an improvement of the oxidative degradation

processes of organic compounds in aqueous medium, thanks to the application of catalytic methods [9, 16–21]. New treatment processes have emerged over the last twenty years, including Advanced Oxidation Processes (AOPs), which have proven to be very interesting for the degradation of recalcitrant organic molecules [22–27]. The AOPs procedure is based on the production of extremely reactive radicals such as ($\bullet\text{OH}$) and ($\bullet\text{O}_2$) to eliminate the organic materials by oxidation and conversion into small and stable molecules like H_2O and CO_2 in the end [28]. These processes include a number of methods including ozonation, photo catalysis, electrochemical oxidation, and Fenton reaction. These treatment processes consist of the generation of highly reactive, nonselective hydroxyl radicals (OH^\bullet) and promote the complete mineralization of aqueous pollutants in carbon dioxide, water, and other mineral ions [29, 30].

Among the different AOPs, Fenton oxidation processes, i.e., reactions between iron salts and H_2O_2 , have been identified as an effective method for producing $\bullet\text{OH}$ radicals that could boost the conversion of organic pollutants to low-toxic or nontoxic species. [31–34]. The photo-Fenton process is similar to the Fenton process but offers much higher removal efficiency by employing irradiation to accelerate $\bullet\text{OH}$ radicals production. However, there are limitations on the conventional Fenton and photo-Fenton oxidation, such as narrow pH range, large amount of iron sludge produced, deactivation by the iron complexation reagents, and difficulty in recycling catalysts [31, 35, 36]. To overcome these drawbacks, the heterogeneous Fenton process was proposed for the degradation of dyes using various catalysts, as $\text{Fe}_2\text{O}_3\text{-SiO}_2$ [29], sewage sludge biochar [32], Fe-impregnated biochar catalyst [16], and magnetic Fe_2MO_4 (M: Fe, Mn) activated carbons [37], zinc oxide nanorods into the Surface of Activated Carbon Obtained from Agricultural Waste [38]. The performance of heterogeneous catalysts is strongly dependent on the distribution of active sites. In order to give a well-dispersed field of active sites, catalyst species are often immobilized onto a porous surface. Lately, considerable interest has been paid to apply biochar as an ecofriendly, low-cost, and porous support to the host catalyst species [16, 32]. Biochar has also demonstrated a positive impact on the acceleration of $\bullet\text{OH}$ production through H_2O_2 decomposition.

In this work, an extruded hydroxide sludge/hydrochar-Fe (HS/HC-Fe) composite was prepared and its photo-Fenton catalytic activities during the decomposition of MB and methyl orange (MO) were evaluated under UV light irradiation in the presence of hydrogen peroxide (H_2O_2). The influence of the different mass ratios between hydroxide sludge and HC-Fe, pH, H_2O_2 concentration and catalyst dosage is discussed. The stability of the catalyst is also investigated.

2. Materials and Methods

2.1. Materials. Hydroxide sludge (moisture 51%) was provided from the drinking water production plant in the city of Marrakech, Morocco. The HC-Fe was prepared by hydrothermal carbonization from two-phase olive mill waste (TPOMW) and FeCl_3 ; the protocol followed was already mentioned in another study [11].

Methylene blue (MB) ($\text{C}_{16}\text{H}_{18}\text{N}_3\text{S}$, purity $\geq 97\%$, cationic dye) was purchased from Sigma Aldrich and methyl orange (MO) ($\text{C}_{14}\text{H}_{14}\text{N}_3\text{NaO}_3\text{S}$, anionic dye) was used as a pollutants model. All other chemicals used like H_2O_2 , H_2SO_4 , and NaOH were of analytical reagent grade. All solutions were prepared with distilled water.

2.2. Preparation of Catalysts. The composites used in this study were prepared by extrusion and physical activation (Figure 1). For the extrusion, a mass of HC-Fe and a mass of wet hydroxide sludge are taken; the mass ratios of HS to HC were 1 : 0, 1 : 1, 2 : 1, and 3 : 1, and the mixture is well mixed manually with a spatula, until a malleable paste is obtained. The prepared dough is then shaped by extrusion. The dispositive used is a manual pressure extruder, and a stainless steel pelletizer that contains seventy cylindrical holes (0.4 mm in diameter and 0.6 mm in length). The extruded mixture was then transferred to a tubular furnace at 800°C for 1 hour. After cooling to room temperature, the products were stored for later use.

2.3. Characterization. The crystalline phase was analyzed by X-ray diffraction (XRD, PAN Analytical X'PERT diffractometer). The morphology and elemental composition was characterized by scanning electron microscopy (SEM-EDS, VEGA3 TESCAN microscope). The surface chemistry of catalysts was analyzed by Fourier transform infrared spectroscopy (FT-IR, Burker VERTEX 70). Inductively Coupled Plasma-Atomic Emission Spectroscopy (ICP-AES, ULTIMA 2c) was used for the determination of the concentration of inorganic elements in samples.

2.4. Experimental Procedures. Photo-Fenton experiments were performed on a photo-catalytic reactor equipped with three UVA lamps disposed at a diameter of 3 cm of a Pyrex; each lamp has a wavelength in the interval between 345 and 400 nm and a power of 15 W. Using a glass beaker containing 1 L solution of MB (50 mg/L) at room temperature and under natural pH (pH = 5), a given amount of H_2O_2 and catalyst were added into the beaker and the solution was mixed using a magnetic stirrer. The pH was adjusted with diluted H_2SO_4 or NaOH when investigating the effect of pH on MB removal. At preselected time intervals, a small quantity of solution was taken and the absorbance was measured immediately using a UV-Vis spectrophotometer (Secomam Uviline 9400 spectrophotometer) at the characteristic wavelength corresponding to maximum absorbance (660 nm). The values of the residual concentrations were obtained by interpolation using the calibration curve between absorbance and the concentration of the dye solution.

The MB removal was calculated by the following formula:

$$\text{MB removal (\%)} = \frac{C_0 - C_t}{C_0} \times 100, \quad (1)$$

where C_0 is the initial concentration of MB (mg/L); C_t is the concentration of MB in another given irradiation time (mg/L).

The recycling tests of catalyst were carried out by consecutive reuses. After the experiment, catalyst was separated and washed by distilled water, then dried at 100°C for 12 h. The catalyst was subsequently used in another oxidation cycle under optimal conditions. The content of Fe ions derived from the heterogeneous catalysts in the degradation solution after each cycle was detected using an Inductively Coupled Plasma-Atomic Emission Spectroscopy (ICP-AES, ULTIMA 2c).

The optimal composite in the removal of MB was also evaluated under the same conditions, to evaluate its ability to degrade methyl orange.

3. Results and Discussion

3.1. Effect of the Composite at Different HS/HC-Fe Ratios on the Degradation of MB by Photo-Fenton Oxidation. HS/HC-Fe composites with different mass ratios were applied for the catalytic degradation of MB under UV light irradiation in the presence of H₂O₂ under natural pH. The effect of mass ratio between HS and HC-Fe on the catalytic activity of the prepared composites is shown in Figure 2. The removal percentages of MB after 150 min of photo-Fenton reaction and by using HS/HC-Fe with 1:0, 1:1, 2:1, and 3:1 mass ratios were, respectively, 21%, 80%, 85%, and 75%. As can be shown, the MB degradation efficiency increased with the HS/HC-Fe mass ratio increase. However, a declining tendency was observed when increasing the HS/HC-Fe mass ratio more than 2:1. When we pass to 3:1, the effect of the reduction of the active sites of HC-Fe decreases the photo catalytic activity; the high accessibility of iron species to HC-Fe active sites could be facilitated when the ratio between HS and HC-Fe is lower [39]. Increasing the ratio will decrease the iron concentration because there is more iron in HC-Fe than in HS according to the ICP (Table 1). The high degradation efficiency of MB was obtained by using the composite with 2:1 HS/HC-Fe mass ratio. This composite was selected in the following experiments.

3.2. HS/HC Composite Characterization. The FT-IR spectra in Figure 3 show the functional groups and chemical bonds on the surfaces of HS, HC-Fe, and 2:1 composite. As shown in Figure 3, both spectra for HS and 2:1 composite showed identical peaks, while their intensities were different. The bands at 3437 and 1636 cm⁻¹ are associated to the O-H stretching and bending vibrations, respectively [1]. In the spectra of HC-Fe, the band located at 1394 cm⁻¹ could be attributed to the methylene and methyl group and the other at 610 to the deformation of the bond (CH) in aromatic compounds. The broad high intensity band at 1038 cm⁻¹ related to the asymmetric Si-O stretching vibrations and a Si-O-Si symmetrical stretching was assigned at 800 cm⁻¹ [40]. The band at 471 cm⁻¹ was associated to the stretching vibration and bending vibration of the Si-O-Fe [9]. The band at 530 cm⁻¹ was assigned to an asymmetric Fe-O stretching vibration [41]. It was noticed that the bands intensity between 651 and 471 cm⁻¹ has been increased when HC-Fe was mixed with HS.

X-ray diffraction analysis was used to obtain structural information from our samples. The X-ray diffraction pattern in Figure 4 shows that the majority of the peaks detected in the composite diffractogram correspond to Fe₃O₄ (code: 01-076-0955) and SiO₂ (quartz, code: 01-085-0794). The structure of HS and HC-Fe was previously identified by Chahid et al. [5] and IZGHRI et al., respectively [11]. The results show that the HS contains mainly SiO₂ (quartz) and HC-Fe contains Fe₂O₃ (Maghemite).

The electron microscopy images presented in Figure 5 show the morphology and microstructure of HS, HC-Fe, and 2:1 composite. The images reveal the apparent changes between the HS, Fe-HC, and the composite, with many small particles appearing on the surface of the composite. The EDS spectra (Figures 5(d)–5(f)) of 2:1 composite clearly revealed a high iron content in the catalyst, indicating that the iron might load on the composite. The content of Fe in the samples was also determined by ICP (Table 1), the Fe content increased from 6.93% to 11% when the HC-Fe was mixed with the sludge.

3.3. Decolorization of MB under Different Systems. To investigate the removal of MB under different conditions, the experiments were carried out with light alone, H₂O₂ alone, light + H₂O₂, 2:1 composite + light, 2:1 composite + H₂O₂, and 2:1 composite + light + H₂O₂, respectively. The degradation curves of MB under different experimental conditions are presented in Figure 6. Light alone, H₂O₂ alone, and light + H₂O₂ were incapable of significantly degrading MB in 150 min, and the degradation efficiency was negligible. The decolorization efficiency of MB was 30% and 59%, respectively, within 150 min by adsorption and heterogeneous Fenton oxidation of 2:1 composite in the dark; nevertheless, the degradation efficiency greatly improved to be 85% under UV light irradiation within 150 min. Similar results were obtained by several authors in the presence of UV-light irradiation; the degradation of BM has been improved [42, 43]. Based on the literature, the photo-Fenton process is more efficient than Fenton alone. An et al. [44] studied the removal of methyl violet by Fenton and photo-Fenton processes. They observed 49.8% methyl violet removal within 120 min in the dark and increased degradation up to 92% within 120 min in presence of light. Another study by Shemer et al. [45] on the degradation of metronidazole indicated higher removal percentage with photo-Fenton process compared to Fenton process. Then better MB degradation could be obtained when using the composite, H₂O₂ and light.

3.4. Effects of H₂O₂ Concentration, Catalyst Dosage, and pH on MB Removal. The concentration of hydrogen peroxide is a primary factor in heterogeneous photo-Fenton system for the creation of hydroxyl radicals (OH), and the effect of the H₂O₂ concentration on the decolorization of MB was investigated by using 2.5 g/L of composite and 50 mg/L MB concentration under natural pH. In this experiment various concentrations of H₂O₂ were employed ranging from 6.52 mM to 18.58 mM. As shown in Figure 7(a), the MB

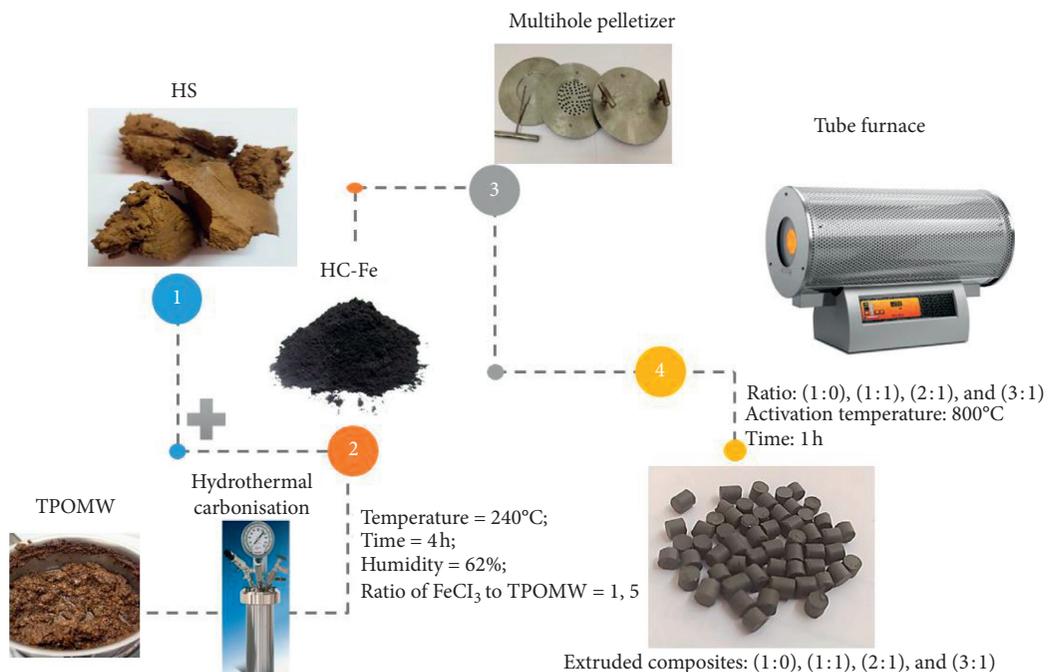


FIGURE 1: Protocol used for the preparation of extruded composites.

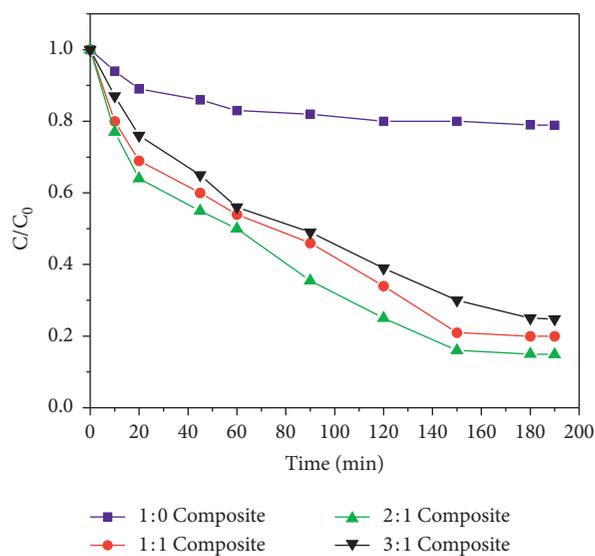


FIGURE 2: Photo-Fenton degradation of MB by different mass ratio HS/HC-Fe composite: [MB] = 50 mg/L, [H₂O₂] = 9.79 mM, and [catalyst] = 2.5 g/L under natural pH.

TABLE 1: Composition of the HS, HC-Fe, and 2:1 composite.

Element (wt %)	HS	HC-Fe	2:1 composite
Fe	6.93	13	11
Al	10.86	0.0027	9.5
Na	0.59	0.0031	0.79
Mg	2.2	<0.001	1.74
Ca	2.94	0.0038	1.71
Ti	0.91	<0.001	0.36
Mn	0.19	<0.001	0.06

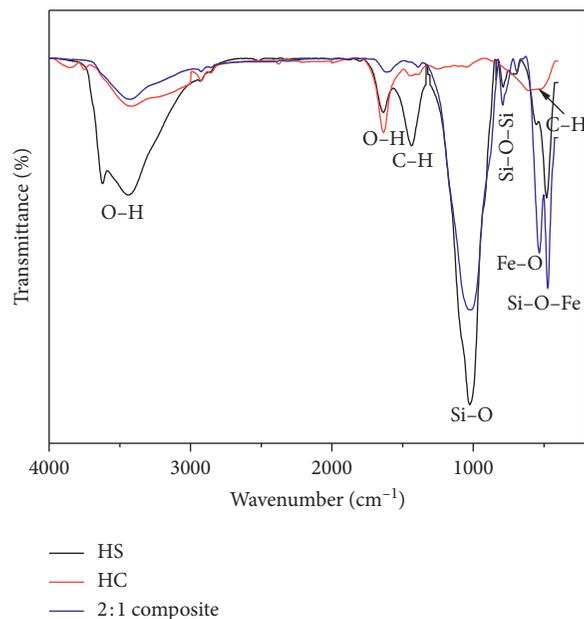


FIGURE 3: FT-IR spectrum of HS, HC-Fe, and 2:1 composite.

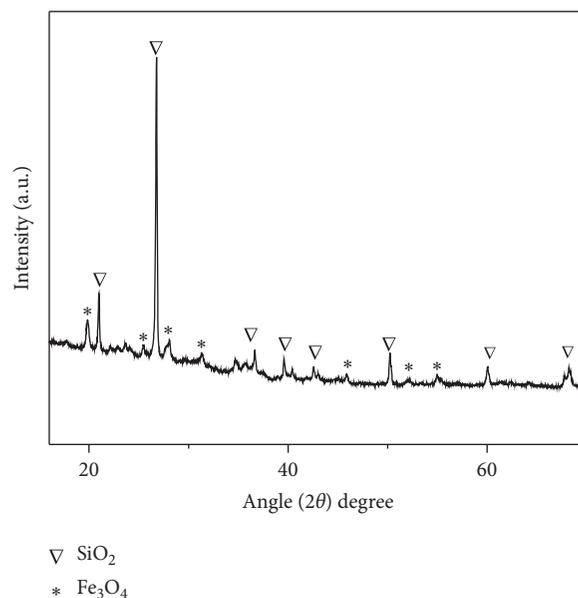


FIGURE 4: X-ray diffraction (XRD) spectrum of 2:1 composite.

removal efficiency increased with an increase in H_2O_2 concentration from 6.52 to 14.68 mM following the increase in the OH^\bullet radicals. However, the further increase in H_2O_2 concentration was not favorable for photo-Fenton reaction and induced a decrease in the removal efficiency. This is due to the generation of perhydroxyl radicals caused by the combination of extra H_2O_2 with hydroxyl radicals [10, 29]. For the upcoming experiments performed to degrade MB by photo-Fenton reaction, a concentration of 14.68 mM H_2O_2 was used.

The effect of composite dosage on the degradation efficiency of MB was also studied by varying the concentration

of composite from 0.5 to 3 g/L. As we can see in Figure 7(b), the removal efficiency increased rapidly from 70% to 95% with the increasing dosage of catalyst from 0.5 to 2.5 g/L. However, a further increase in the composite dosage to 3 g resulted in a decrease in MB decolorization rate. Indeed, increasing the catalyst quantity up to 2.5 g accelerated the removal of MB from aqueous solution, since more HO^\bullet radicals are formed from higher loaded composite. At a catalyst dosage >2.5 g, the scavenging of hydroxyl radicals dominated and reduced the degradation of MB in the presence of excessive metal species, producing HO^\bullet radicals [46]. It is believed that the generation of HO^\bullet radicals is

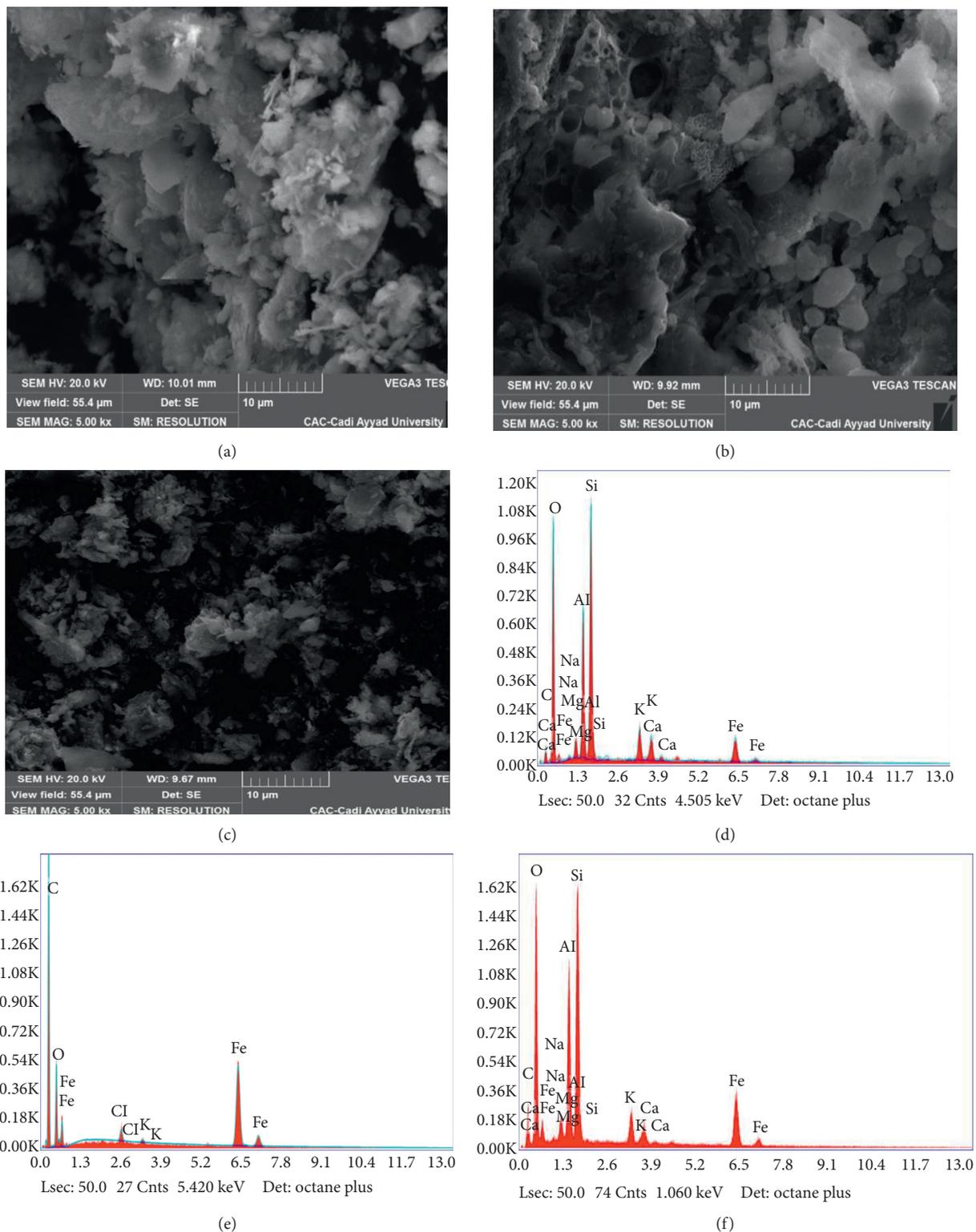


FIGURE 5: (a) SEM micrographs of HS, (b) HC-Fe and (c) 2:1 composite, (d) EDS of HS, (e) HC-Fe, and (f) 2:1 composite.

favored at high catalyst loadings until a certain limit beyond this limit apart of the $\text{HO}\cdot$ radicals consumed by self-scavenging of $\text{HO}\cdot$ radicals by iron species, which reduces the MB degradation rate [11, 47]. Moreover, excessive catalyst weakens the penetration of light and reduces the

available radiation [33]. In light of the above, a catalyst dosage of 2.5 g was chosen as optimal for the rest of the study.

As it was largely reported, solution pH can remarkably influence the catalytic effect during photo-Fenton reaction

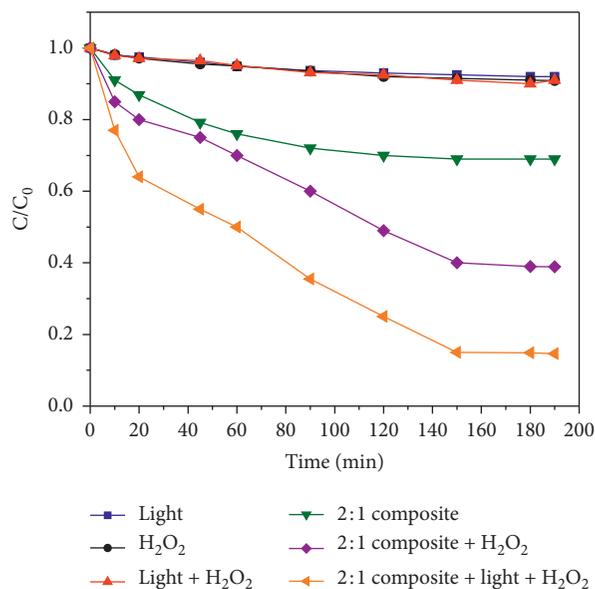


FIGURE 6: Degradation of MB in different operation systems. Reaction conditions: [MB] = 50 mg/L, [H₂O₂] = 9.79 mM, and [catalyst] = 2.5 g/L under natural pH.

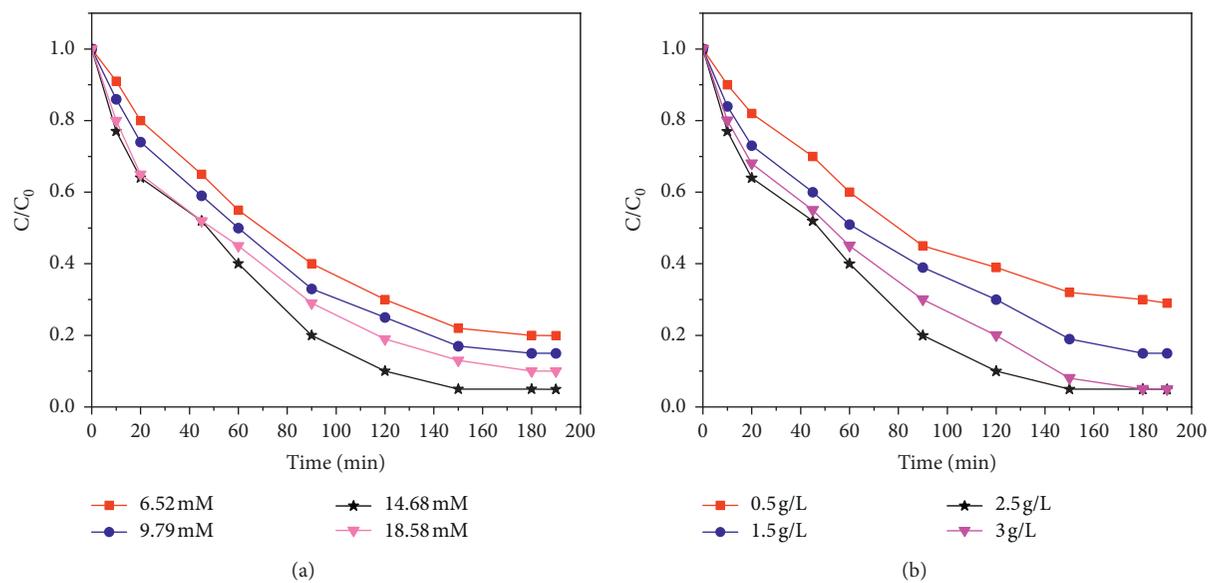


FIGURE 7: Continued.

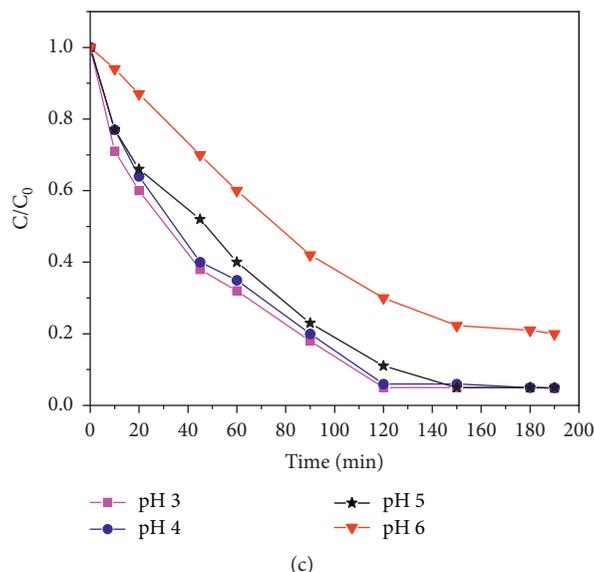


FIGURE 7: Photo-Fenton degradation of MB in different H_2O_2 concentrations (a), catalyst dosages (b), and initial pH conditions (c). Reaction conditions: $[\text{MB}] = 50 \text{ mg/L}$, $[\text{catalyst}] = 2.5 \text{ g/L}$, $[\text{H}_2\text{O}_2] = 14.68$, and natural pH, except for tested condition.

[48]. In the present study, the influence of pH was also considered. In general, there is a large accord in the literature about the optimal pH value, close to 3. The pH of 3 is the pH for which the Fenton and photo-Fenton processes are most effective [49]. We studied the reaction with 50 mg/L of MB, 14.68 mM of H_2O_2 , and 2.5 g/L of 2:1 composite under UV light irradiation at different pH values, between 3 and 6. The data shown in Figure 7(c) indicate that the degradation rate was almost the same at pH 3, 4, and 5. This result indicated that 2:1 composite as photo-Fenton catalyst can effectively work in a wide range of pH value. The MB degradation efficiency slightly decreased for pH=6. The reduction tendency of the MB removal efficiency could be explained by the decrease in concentration of hydroxyl radicals in solution with increasing pH [50].

3.5. UV-Vis Spectra Study. Figure 8 illustrates the UV-Vis absorption spectra of MB in aqueous solution during the degradation process via H_2O_2 catalyzed with 2:1 composite under UV light irradiation conditions. The main absorption band for the MB was seen at 665 nm, the intensity of which reflected its concentration in the solution. The MB degradation was clearly evidenced by the change in the band intensity of the UV-Vis absorption spectra, which almost disappeared absolutely within 150 min. The chromophores responsible for characteristic color of the MB were broken down and MB had been degraded.

3.6. Reusability of Composite and Application to Methyl Orange Degradation. In order to evaluate the reusability of catalyst, the MB removal efficiency was evaluated by 5 consecutive photo-Fenton reactions with optimal composite under the optimal conditions ($[\text{MB}] = 50 \text{ mg/L}$, $[\text{H}_2\text{O}_2] = 14.68 \text{ mM}$, $[\text{composite}] = 2.5 \text{ g/L}$ and under natural pH).

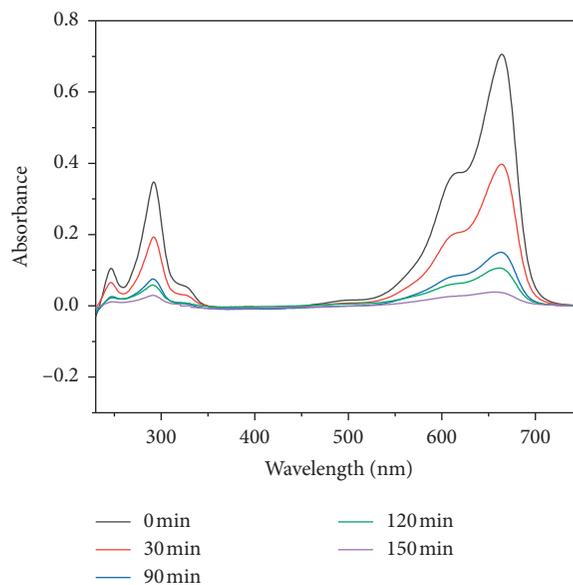


FIGURE 8: UV-Vis spectral changes with reaction time ($[\text{MB}] = 50 \text{ mg/L}$, $[\text{H}_2\text{O}_2] = 14.68 \text{ mM}$, natural pH, and $[\text{composite}] = 2.5 \text{ g/L}$).

After the first reaction, the composite was separated easily from the reacted aqueous solution rinsed with distilled water after each repeated experiment and then dried around 100°C . As shown in Figure 9, the degradation performance of 2:1 composite was well maintained with a slight reduction of removal efficiency from 95% to 92% after four reusing cycles. In the 5th cycle, the catalytic activity of the composite began to decrease, possibly due to the deactivation of the catalyst via the deposition of small molecular contaminants on its surface additionally, and the leaching test indicated that the leached iron in the five cycles was less than 0.005%. The poor

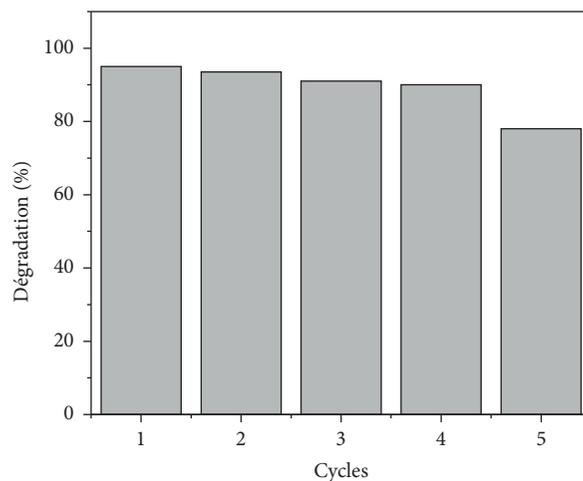


FIGURE 9: Reusability of heterogeneous Fenton composite. Reaction conditions: $[MB] = 50 \text{ mg/L}$, $[H_2O_2] = 14.68 \text{ mM}$, natural pH, and catalyst dosage = 2.5 g/L.

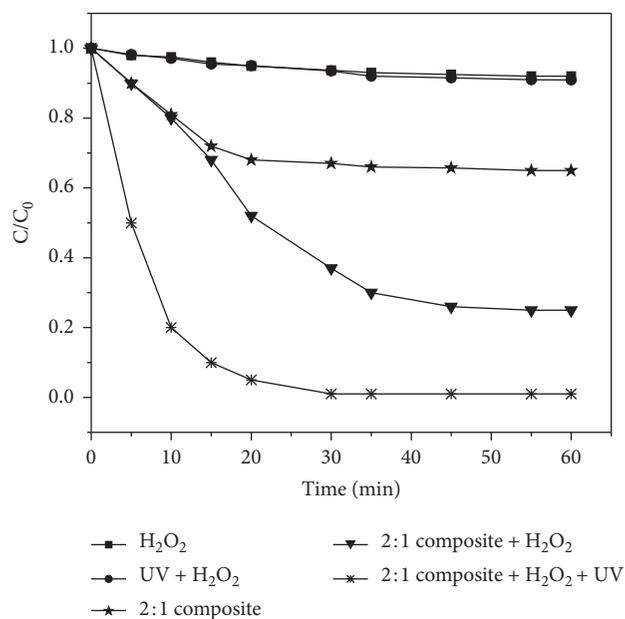


FIGURE 10: MO removal over 2:1 composite as a function of time. Reaction conditions: $[MB] = 50 \text{ mg/L}$, $[H_2O_2] = 14.68 \text{ mM}$, natural pH, and catalyst dosage = 2.5 g/L.

dissolution of iron means that heterogeneous catalysis was mainly responsible for the degradation of BM.

The most effective composite for removing MB was also evaluated for removing MO under the same optimal conditions. Figure 10 shows the MO decolorization efficiency as a function of time in different systems: MO + H_2O_2 , MO + composite, MO + composite + H_2O_2 , and MO + composite + H_2O_2 + UV. The removal rate in the presence of MO and H_2O_2 only is negligible, the maximum conversion of MO reaches a steady state of 7% after 140 minutes of reaction time. When the composite was used alone, 35% discoloration of MO was obtained due to the adsorption of MO by this material. In the case where the

composite and H_2O_2 coexisted in the reaction mixture, there is a removal of MO attaining 60% after 45 min, which could be defined by two processes. One results from the adsorption of MO to the surface of the composite, which mainly causes the discoloration of MO in the first 20 minutes. Another is attributed to the oxidation of this organic dye by OH radicals resulting from the Fenton reaction. The combination of the composite and UV irradiation results in complete removal of the color, indicating the improvement in fading efficiency which exceeds 99%. This result verifies the role of UV light as a promoter of good dispersion of iron oxides. Comparing these results with those reported in other studies, a remarkably high

TABLE 2: Comparison of MB degradation for various methods and catalysts.

Method/catalyst	C (dye) (mg/L)	pH	Concentration H ₂ O ₂ (mmol/L)	Reaction time (min)	Decolorization efficiency (%)	Ref
Hetero-Fenton/magnetic porous carbon	10	4	23.58	90	100	[52]
Fenton-like/iron ore tailing from tailing dam	20	6	635.25	180	80	[53]
Photo-Fenton/2:1 composite	50	5	14.68	150	95	This work
Fenton-like/Ferrocene	10	4	23.58	90	100	[54]
Photo-Fenton/Fe(II)Fe(III)-LDHs	10	3	0.01	60	100	[55]
Heterogeneous Fenton/iron oxide supported on activated carbon	25	—	489.5	180	98	[56]
Heterogeneous Fenton-like/magnetic nanocomposite	50	—	97.9	180	46	[57]

photo-Fenton performance for the removal of MO was achieved using low concentration of H₂O₂ [23, 29, 51]. These results showed the ability of the composite for oxidizing dyes.

3.7. Comparison with Other Methods. Table 2 compares the results obtained in terms of decolorization efficiency and reaction time, in the presence of the 2:1 composite presented in this work, to other previously published results, considering different treatment approaches, such as adsorption, Fenton-like, photo-Fenton, and heterogeneous Fenton oxidation. It can be seen from Table 2 that the H₂O₂ concentration required in this work is much shorter except for Photo-Fenton/Fe(II)Fe(III)-LDHs. Meanwhile the decolorization efficiency for MB is super. In addition, a comparison with other reported catalyst [52, 54, 55] shows the wider range of pH for 2:1 composite, suggesting that 2:1 composite is a good candidate in photo-Fenton reaction. These results show that each catalytic system behaves differently, and the optimal conditions for achieving maximal removal of MB cannot be easily extrapolated from one system to another, which is similar to the previous reported [41].

4. Conclusion

In the present study, a series of extruded HS/HC-Fe composite catalysts were synthesized by activation for the degradation of MB and MO. For photo catalytic oxidation experiments, 2:1 composite exhibited the highest activity allowing for a decolorization efficiency of 95%, indicating that appropriate addition of HC could promote the photo catalytic process. Extrusion allows the composite to be easily separated and the reusability study showed that the 2:1 composite exhibited rather high activity for the degradation of MB after 3 repeated uses, where the decolorization efficiency of MB achieved 91%. The use of 2:1 composite to methyl orange degradation allowed for a decolorization efficiency up to 99%. Most important of all, the composite can work effectively under near neutral pH conditions. Therefore, the composite catalyst would be considered as a new economical and environmentally friendly photo catalyst for removing organic contaminants from the aqueous

solution due to its low cost and no-toxic properties, as well as high photo catalytic performance.

Data Availability

The data can be sourced upon request to the authors.

Conflicts of Interest

The authors declare no conflicts of interest.

Acknowledgments

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