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Synergistic heterogeneous electro-Fenton process using iron-loaded bentonite for binary dyes degradation from wastewater

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Abstract

This study examines the impact of operational parameters on the performance of a heterogeneous electro-Fenton process for removing a binary mixture of malachite green (MG) and methylene blue (MB) using iron-loaded bentonite clay (Fe-Bent.) particles as a heterogeneous catalyst. The effects of current density (j), Fe-Bent. dosage (Bent. quantity variable) and Fe*-Bent. dosage (iron concentration variable), were investigated. Brunauer-Emmett-Teller analysis, X-ray diffraction, X-ray fluorescence and scanning electron microscopy were used to extensively evaluate the synthesized catalyst, confirming the effective incorporation and uniform distribution of iron within the bentonite clay. The results showed that increase of j enhanced dye removal efficiency to over 95 % for MG and 90 % for MB at 20 mA cm⁻². Furthermore, increasing the dose of Fe-Bent.* from 1 to 3 g improved the overall removal efficiency, although this was accompanied by a decrease in adsorption capacity and a deviation from ideal kinetic behaviour at higher loadings. Fe*-Bent. dosage with a variable iron concentration from (0.1 to 0.2 mM) exhibited the highest removal efficiency, while increasing to 0.3 mM resulted in decreased process performance. The comparison results showed that the electro-Fenton process alone achieved limited removal, whereas bentonite adsorption alone provided high but incomplete removal. In contrast, the heterogeneous electro-Fenton process achieved the highest efficiency (98.06 % for MG and 91.14 % for MB, with COD values of 84.53 % for MG and 79.82 % for MB), with the lowest iron leaching (<0.1 ppm), well below regulatory limits. These results demonstrate the clear advantage of the heterogeneous electro-Fenton process for efficient and environmentally friendly removal of mixed dye pollutants.*

Keywords

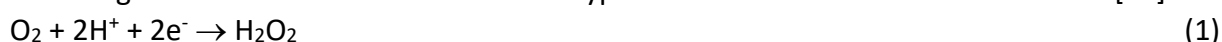
Natural clay; iron incorporation; heterogenous catalyst; mixed dyes: electrochemical removal; wastewater treatment

Introduction

Water is necessary for both ecology and growth for the survival of life [1]. The world's urbanization and population growth are placing a great demand on water resources and quality [2]. Since water is already a scarce resource, its per capita availability worldwide will certainly decline, leading to a water crisis [3]. This problem does not extend beyond developing countries; pollution from industrial activity exacerbates outstanding water problems [4]. According to the World Health Organization (WHO), 1.6 million individuals suffered from water pollution in 2018, highlighting the critical need for clean water management. [5,6]. Some industries that use the most water and produce significant volumes of wastewater contaminated with various substances include petroleum refining [7], textiles [8], pharmaceuticals [9], and pulp and paper production [10]. There are serious environmental problems resulting from the widespread use of synthetic dyes in many industrial sectors, especially in the processing of textiles, paper, leather, cosmetics, and food, of the approximately 100,000 dyes sold commercially. Synthetic dyes pose major risks to aquatic life and human health because of their complex aromatic structures, which also confer excellent chemical stability, resistance to biodegradation, and long environmental persistence. As a result, research in chemical and environmental engineering and wastewater treatment is now heavily focused on developing dye removal techniques that are economical, sustainable, and efficient [11,12].

Dye-containing wastewater can be effectively treated using various methods, including chemical treatment [13], absorption [14], filtration [15], biological treatment, coagulation [16] and adsorption [17]. Physical methods are effective at eliminating colouring compounds and suspended solids, but they are typically costly, non-selective, and frequently result in secondary pollution from membrane fouling or sludge production [18]. AOPs, or advanced oxidation processes, have drawn a lot of interest as viable alternatives for dye degradation. Highly reactive hydroxyl radicals $\text{OH}^\bullet$, which may attack and degrade a wide range of organic contaminants, including dyes, non-selectively, are the basis of AOPs. The electro-Fenton (EF) process has become a particularly appealing alternative among the several AOPs because of its high efficiency, capacity to function in ambient circumstances, and potential for *in situ* oxidant generation [19,20].

To produce hydroxyl radicals for the mineralization of organic contaminants, the electro-Fenton process combines electrochemical and Fenton's chemistry concepts. Hydrogen peroxide H_2O_2 is produced electrochemically at the cathode in a conventional EF system by reducing the dissolved oxygen, shown by Equation (1) [21]. At the same time, ferrous ions (Fe^{2+}) are either electrochemically regenerated from ferric ions Fe^{3+} by Equation (2), or externally brought into the system [22]. These ions catalyse the breakdown of H_2O_2 into hydroxyl radicals ($\text{OH}^\bullet$) as illustrated in Equation (3). Radicals are continuously produced *in situ* as a result of the net reaction, fiercely oxidizing and mineralizing colour molecules into innocuous byproducts like carbon dioxide and water [23].



Fenton-like oxidation reactions are inexpensive, easy to utilize, non-toxic and highly appealing [24]. Significant disadvantages of the homogeneous EF process include the production of iron-containing sludge, the reduction in catalytic activity due to iron precipitation, the catalyst's low reusability, and the need for extremely acidic pH conditions (often around pH 3) [25]. The heterogeneous electro-Fenton (HEF) process is a promising, reliable, and eco-friendly substitute for this method that produces a stable and reusable catalyst that promotes the Fenton reaction without dissolving into the aqueous phase by disabling iron species on a solid support material [26].

The selection of electrode materials is one of the most important aspects of the EF process, as it has a significant impact on the electrochemical production of H_2O_2 , energy consumption, and overall efficiency [27]. Noble metal-based electrodes, such as platinum as well as boron-doped diamond, have been used historically, but their high cost has hampered large-scale applications [28]. On the other hand, because of their excellent electrical conductivity, chemical stability, large surface area, and environmental friendliness, carbon-based electrodes, particularly graphite, have attracted attention in the EF process [29]. Additionally, graphite's stratified planes and porosity increase the surface area available for electrochemical reactions. It is the perfect material for large-scale wastewater treatment systems due to its abundance and affordability [30].

Although the electro-Fenton technique is quite effective at dissolving dye molecules, the overall effectiveness of pollution removal can be greatly increased by incorporating catalyst mechanisms [31]. Natural clay minerals such as bentonite have attracted considerable interest as solid supports for heterogeneous Fenton catalysts due to their high surface area, chemical stability, ion-exchange capacity, and environmental friendliness [32]. Bentonite, which is primarily made up of montmorillonite, offers a great medium for iron species dispersion and stabilization [33]. Encouraging interaction between the target chemicals and the reactive sites improves the effectiveness of catalytic reactions and makes it easier for contaminants to be adsorbed. Iraqi bentonite, in particular, is an inexpensive, locally available, and underutilized resource that may be used as a useful support material in heterogeneous electro-Fenton systems [34].

Catalytic particle electrodes can be prepared using metal oxides or metal oxide-loaded carriers. Granular active carbon (GAC), metal particles, carbon aerogel, clay, and other particles are often used as particle electrodes in 3D reactor systems [35].

This study examines the electro-Fenton degradation of synthetic mix dyes in water using Iraqi bentonite particles as a heterogeneous catalyst. The practical potential of the catalyst for wastewater treatment applications is assessed based on its efficiency, structural stability, reusability, and resistance to iron leaching. By turning a naturally occurring, locally accessible clay into a useful treatment technique, this research not only seeks to improve the electro-Fenton process's performance through material innovation, but also supports environmentally friendly management.

Experimental

Materials

Bentonite stones (Bent.) were obtained from the Western Desert of Iraq. Distilled water was used to prepare all aqueous solutions; no further purification was required because all the reagents were analytical grade. The chemicals included sodium sulphates (Na_2SO_4 , $\geq 99.0\%$, SDFCL), sulfuric acid (H_2SO_4 , 98 %, Sigma-Aldrich), methylene blue (MB) and malachite green (MG) purchased from Merck Company, Alpha Chemical Reagent Company, India, and ferrous sulphate heptahydrate $\text{FeSO}_4 \times 7\text{H}_2\text{O}$ from CDH Company, India. The graphite electrode was purchased from the local market and utilized as anode and cathode.

Preparation of Fe-Bent. catalyst

The catalyst Fe-loaded bentonite (Fe-Bent.) was prepared by the impregnation method [36,37]. In a beaker with 500 mL of distilled water, a certain amount of $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ was dissolved. Next, a specified amount of bentonite was added to the aqueous solution under continuous stirring and heated until the water evaporated. Following impregnation, the sample was dried for 12 hours at 105°C and then calcined in a furnace for 4 hours at 500°C .

Experimental methods

In a 1 L cylindrical beaker containing 0.5 L of aqueous solution, the electro-Fenton reaction was carried out. Both graphite electrodes (with dimensions of $15 \times 6 \times 0.6$ cm) were immersed in 0.5 L with a concentration of 25 mg L^{-1} (MB and MG) dyes solution that was contained in the batch reactor shown in Figure 1. A graphite plate (active surface area 24 cm^2) served as the anode and cathode. The space between the anode and cathode was 3 cm, while Fe-Bent. particles were packed between the anode and cathode. Aeration was used in the solution with (ACO-001 electromagnetic, China) for 20 minutes prior to the reaction and continued until the experiment was finished to increase the amount of dissolved oxygen. $0.1 \text{ M H}_2\text{SO}_4$ acid was added to the solution to bring the pH down to 3, and a HANNA (Romania) pH meter was used to test the pH. 0.05 M of Na_2SO_4 was added to the solution to raise the medium conductivity and maintain the ionic strength. Electrolytes boost the efficiency of electrodes and processes, including the conversion of oxygen to hydrogen peroxide at the cathode surface, by increasing the rate of ion exchange and the electrostatic resistance between electrodes and ions.

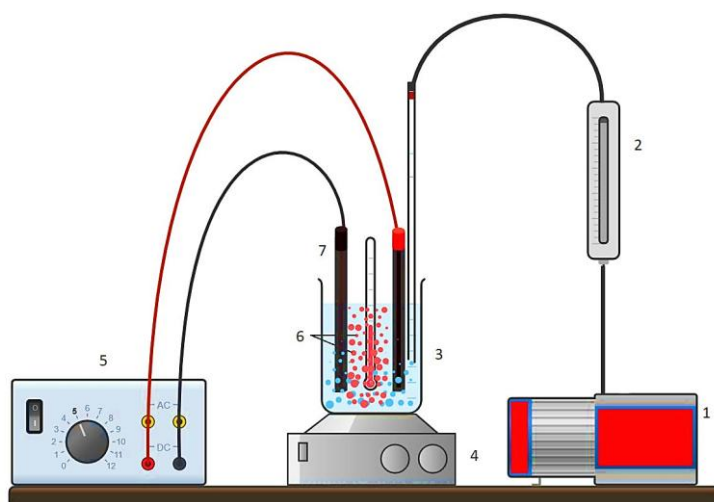


Figure 1. Schematic diagram of heterogeneous electro-Fenton oxidation system: 1) pump, 2) flow meter, 3) beaker, 4) magnetic stirrer, 5) power supply, 6) Fe-loaded bentonite particles, and 7) graphite electrodes

A DC power source (UNI-T UTP3315PE) operating at a steady current density (j) was connected to the electrodes. To measure voltage and current, a digital voltmeter (FLOWTECH, China) was employed. To remove suspended pollutants, periodic samples of the treated solution were centrifuged at 400 rpm for 15 minutes prior to analysis throughout the electro-Fenton process; the temperature was maintained at 25 ± 1 °C. Before calculating additional examination, samples were extracted from the reactor at certain times and centrifuged.

Advanced techniques were used to examine the physical and chemical properties of Bent and Fe-Bent particles, including X-ray diffraction (XRD) on a Shimadzu-6000 instrument using Cu-K α radiation ($\lambda = 0.1541874 \text{ nm}$) at 30 mA and 40 kV. The chemical composition was determined using X-ray fluorescence (XRF) (Spectro-XEPOS-Germany), the porosity and specific surface area were determined using Brunauer-Emmett-Teller analysis (BET) (Horibe, SA-9600 model, USA) and a scanning electron microscope (SEM) with Inspect S50 at 25 kV and 100 microamperes (FEI Company, Netherlands) was employed to assess the particle size and surface morphology.

The dye mixture concentration was measured using a UV spectrophotometer (9200, Biotech Engineering Management Co. Ltd., UK) and the COD reduction was estimated by a COD meter (MD 200 Lovibond, Germany). The efficiency of binary dyes colour removal and COD reduction was determined using Equation (4) [38]:

$$R = \frac{C_i - C_f}{C_i} 100 \quad (4)$$

where $R / \%$ is colour or COD removal efficiency, C_i and $C_f / \text{mg L}^{-1}$ are initial and final dye mixture concentrations of colour, or initial and final COD values for COD reduction.

The energy consumption (EC, kWh L^{-1}) was evaluated using Equation (5) [39]:

$$\text{EC} = \frac{EIt}{V} \quad (5)$$

where E / V is the average voltage, I / A is the applied current, t / h is time, and V / L is the treated volume of the dye solution. The adsorption capacity ($q / \text{mg g}^{-1}$) was computed using Equation (6) [40]:

$$q = \frac{(C_i - C_f)V}{m} \quad (6)$$

where m / g is Fe-Bent.* dosage.

Table 1 lists the operational parameters and their corresponding ranges used in the heterogeneous electro-Fenton process.

Table 1. Process variables and their ranges

Parameter	Ranges
Current density, mA cm^{-2}	8, 14, 20
Fe-Bent.* dosage, g	1, 2, 3
Iron concentration (Fe*-Bent.), mM	0.1, 0.2, 0.3

Results and discussion

Characterization of Bent. and Fe-Bent. particles

XRF analysis

Table 2 summarizes the chemical compositions of the Bent. and Fe-Bent. particles as measured by XRF. The results show a significant increase in Fe_2O_3 concentration from 5.24 wt.% in raw bentonite Bent. to 12.35 wt.% in Fe-Bent., indicating well-integrated iron loading into the clay layers. Furthermore, the relatively low concentrations of both SiO_2 and Al_2O_3 suggest that the iron particles are likely dispersed throughout or within the aluminosilicate structure of the bentonite, rather than deposited in large aggregates, indicating successful loading [41].

Table 2. Chemical composition of raw Bent. and Fe-Bent.

Oxides	Content, wt.%	
	Bent.	Fe-Bent.
SiO_2	53.42	44.36
Al_2O_3	8.99	5.56
CaO	6.18	4.89
Fe_2O_3	5.24	12.35
MgO	4.21	3.98
Na_2O	1.13	0.79
Others	12.83	19.07

X-ray diffraction analysis

To determine crystallinity and identify mineral phases, XRD was used. Figure 2 represents the XRD pattern for both raw Bent., Fe-Bent. and Fe_2O_3 reference pattern for comparison. The main crystalline phases identified in Bent. for SiO_2 at $2\theta = 21.9, 26.6$ and 36.1° , which were comparable with the reported literature [42]. The X-ray diffraction pattern of Fe-Bent particles, represented by

the red graph, shows peaks corresponding to Fe_2O_3 . During the preparation process, $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ oxidizes in the presence of air, leading to the formation of stable iron oxide phases, the most important of which is hematite ($\alpha\text{-Fe}_2\text{O}_3$). The characteristic peaks at $2\theta = 33, 35.6$ and 40.9° correspond to the Fe_2O_3 phase and are identical to the reference pattern (96-101-1268) shown in Figure 2 for comparison, indicating efficient iron deposition on the clay surface. The relatively low intensity and broadness of these peaks suggest nanometre-sized crystals and good dispersion of iron species on the bentonite surface, which is characteristic of catalysts produced by impregnation methods. The presence of Fe_2O_3 is very important for the electro-Fenton process, as it facilitates a continuous redox cycle between Fe^{2+} and Fe^{3+} ions, thereby promoting the generation of hydroxyl radicals responsible for pollutant degradation [43].

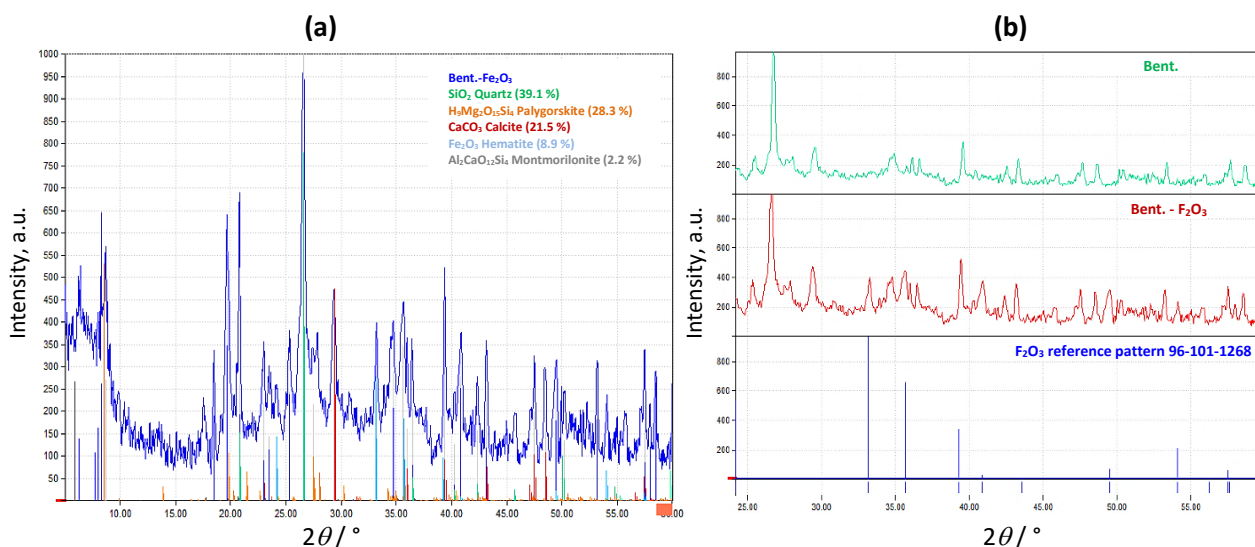


Figure 2. XRD spectra of (a) Fe-Bent. particles and (b) Bent., Fe-Bent. and reference pattern of Fe_2O_3 for comparison

To estimate quantitative analysis, the Scherrer equation was used to evaluate the crystallite size (D / nm), represented in Equation (7) [44]:

$$D = \frac{K\lambda}{\beta \cos \theta} \quad (7)$$

where K is the shape factor equal to 0.9, λ is the Cu X-ray wavelength ($\lambda = 0.1541874$ nm), β is the full width at half maximum FWHM, and θ is the Bragg angle. The calculated crystal size of 24.4 nm confirms the formation of finely dispersed iron oxide particles on the surface of the bentonite.

The XRD analysis was extended by assessing the degree of crystallinity (DOC), revealing a mixed crystalline structure with 41.59 % crystalline material and a significant proportion of amorphous material (58.41 %). This mixed structure is typical of the electro-Fenton process and the successful iron loading of bentonite, suggesting that the introduction of iron does not lead to complete crystallization but rather forms a hybrid system [45].

SEM analysis

SEM was employed to investigate the clay surfaces as shown in Figure 3a and b for the Bent., and Figure 3c and d for Fe-Bent. it is evident that the raw Bent. consists of irregular particles in the form of sheets. The original shape of Bent. is maintained, while iron particles appeared clearly on the surface in Figures 3c and 3d. The Fiji software was used to calculate the average particle size of approximately $58.455 \mu\text{m}$ from the analysis of 1207 particles. This relatively large size reflects the formation of aggregate structures rather than small particles, indicating a wide particle-size distribution. However, XRD analysis revealed a much smaller crystalline size of around 24 nm, confirming

that these micrometer-sized clumped particles are aggregates composed of nanocrystals. This property provided active catalytic sites. Therefore, the removal efficiency is mainly attributed to the synergistic effect between adsorption and the electro-Fenton process, where electrochemical reactions on the (Fe-Bent.) surface promote the generation of reactive oxygen species [46].

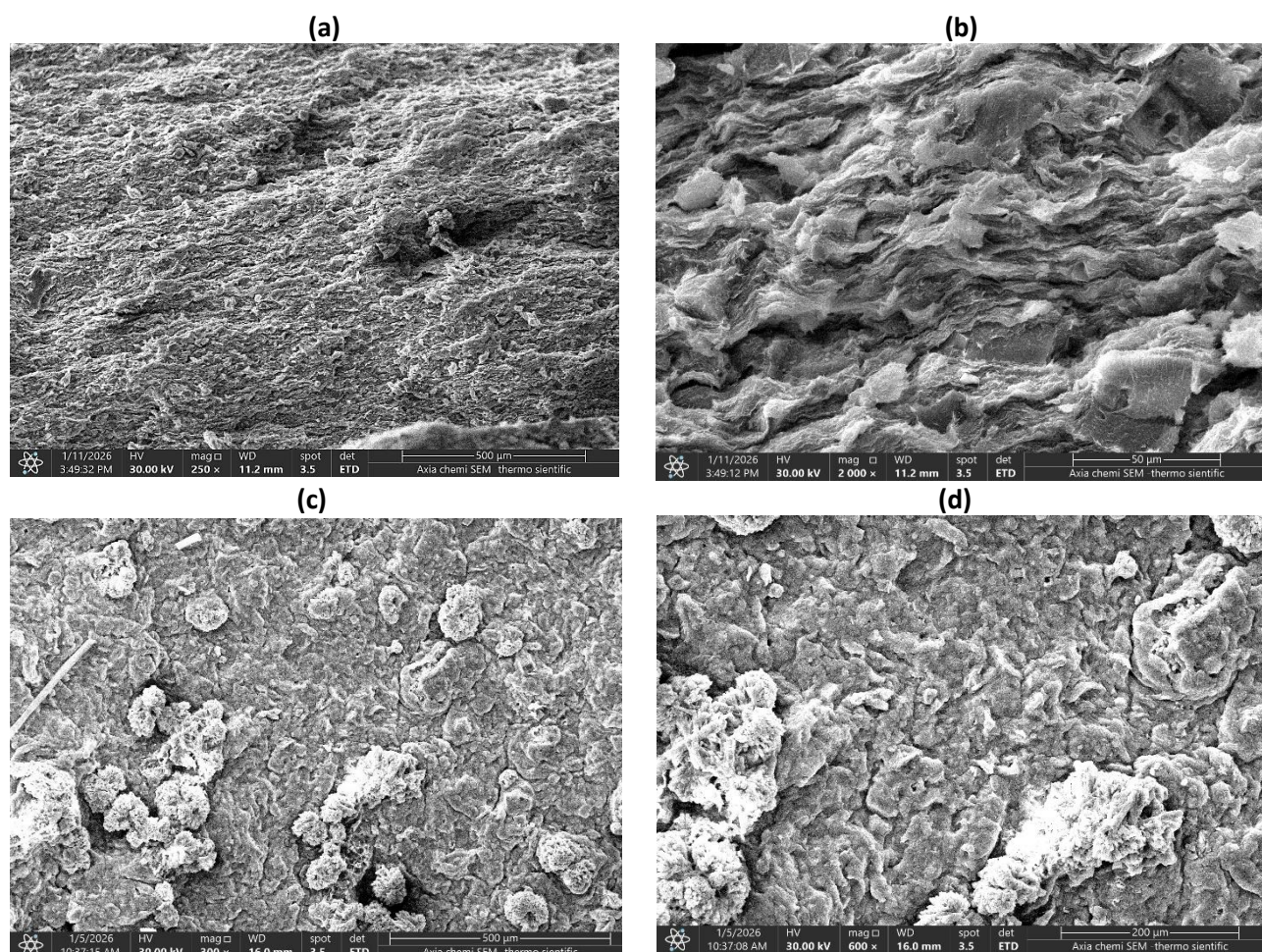


Figure 3. SEM images at different magnifications of: (a) and (b) Bent.; (c) and (d) Fe-Bent.

Brunauer-Emmett-Teller analysis

Based on nitrogen adsorption and desorption isotherms, the specific surface area and total pore volume were calculated using the BET method and summarized in Table 3.

Table 3. BET characterization for Bent. and Fe-Bent.

Material	Average surface area (BET), m ² g ⁻¹	Total pore volume, cm ³ g ⁻¹
Bent.	20.221	0.077
Fe-Bent.	23.559	0.098

From the results, the average surface area of Bent. was 20.221 m² g⁻¹ and increased relatively slightly to 23.559 m² g⁻¹ after being loaded with iron and calcination [47]. This increase in the surface area indicates a limited adsorption capacity, which is consistent with research on Iraqi clays that are improved by acid treatment [48]. Therefore, the high removal efficiency is not primarily attributed to adsorption alone, but rather to the high electro-Fenton oxidation that worked in conjunction with adsorption.

Comparative performance of electro-Fenton, bentonite clay adsorption and Fe-Bent. heterogeneous electro-Fenton processes

The comparison among these processes was conducted to establish each method's true performance under identical conditions. These systems differ fundamentally in their reaction mechanisms and their ability to handle mixed dyes, and no meaningful conclusion can be drawn without a direct benchmark that exposes their respective strengths and limitations. A comparative assessment was conducted to evaluate the performance of the hybrid configuration relative to the individual systems. The results were used to determine whether coupling the two methods leads to enhancing the removal efficiency of the dye mixture.

As presented in Figure 4, the removal behaviour of MG and MB differs depending on the treatment method. Under electro-Fenton conditions (current density of 14 mA cm^{-2} , concentration of Na_2SO_4 of 0.5 M, air flow rate of 0.4 L min^{-1} , Fe^{2+} concentration of 0.2 mM, and pH 3), the dye removal efficiency (Re, %; referring to colour removal efficiency) of MG was 56 % over 60 min, while MB achieved only 32 % during the same duration. This difference may be related to the structural characteristics of the two dyes. While MG has tertiary amine groups that facilitate oxidation by $\text{OH}^\bullet$, MB exhibits greater chemical stability [49].

Using bentonite alone, the adsorption process achieves dye removal efficiencies for MG and MB (over 60 min, bentonite dose of 2 g and temp. $25 \pm 1^\circ\text{C}$) of 87.78 and 80.83 %, respectively. These results highlight the role of surface adsorption due to the highly porous structure and negatively charged surfaces of bentonite that favour the fast interaction with these cationic dyes (MG and MB). However, the saturation of the available adsorption sites reduced the extent of further dye removal [50]. Moreover, decolorization alone does not necessarily refer to the actual degradation of the dye molecules. Therefore, COD analysis was performed, and the results showed that COD reduction (COD, %) for MG and MB was 30.84 and 23.07 %, respectively, which is lower than the corresponding removal efficiencies (87.78 and 80.83 %). This suggests that the pollutants are transferred onto the adsorbent surface rather than being chemically degraded.

Under the same conditions as in the electro-Fenton process, but using Fe-Bent. catalyst (with Bent. quantity of 2 g and iron concentration of 0.2 mM), the heterogeneous electro-Fenton Fe-loaded bentonite system exhibited the highest removal efficiencies and COD reductions, for MG (Re = 98.06 % and COD = 84.53 %) and for MB (Re = 91.14 % and COD = 79.82 %). This provides evidence of dye degradation, not only adsorption. This enhanced performance can be attributed to two synergistic effects. Adsorption of dye molecules onto bentonite surfaces concentrated the dyes near the reactive sites and increased interactions with $\text{OH}^\bullet$. The dispersion of bentonite particles stabilizes Fe^{2+} and supports H_2O_2 generation, promoting $\text{OH}^\bullet$ production. Additionally, the oxidative removal of the adsorbed dyes contributes to the regeneration of active sites, which maintains them available during the reaction and prevents early saturation. In other words, this behaviour does not show a simple additive effect between adsorption and oxidation. Adsorption alone provides high colour removal but limited COD reduction, while the hybrid system improved both parameters, indicating the interaction effect rather than parallel processing. As a result, the MG is almost completely removed, while MB also achieves high removal efficiency [51].

Based on the results and the above discussion, the electro-Fenton exhibited the lowest removal efficiency for both dyes (for MG Re = 56.08 % and COD = 34.07 % and for MB Re = 32.44 % and COD = 21.93 %), as it depends on free radical oxidation where the degradation rate is restricted by the limited generation of H_2O_2 and persistent nature of the dye molecules toward oxidative attack under the applied conditions. Adsorption alone provided higher colour removal efficiency, but surface

saturation with low COD reduction restricted its performance. In contrast, the heterogeneous electro-Fenton method achieved the highest decolorization and COD reduction rates compared with these methods, owing to the combination of free-radical oxidation and adsorption on the bentonite surface [31]. Because the heterogeneous electro-Fenton process is complex, COD analysis was used as a representative indicator of process performance. The variations among the dyes can be explained by their molecular structures and chemical properties, highlighting the importance of considering dye characteristics when designing hybrid systems for treating mixed-dye effluents [52].

Dissolved iron was measured using atomic absorption spectroscopy to verify the stability of each treatment system. The electro-Fenton process released about 3.15 ppm of iron, while the hybrid Fe-Bent. system maintained concentrations below 0.1 ppm, which is under both the Iraqi permissible limit (<3 ppm) and the limits generally applied under European Union discharge regulations (<2 ppm) [53]. This difference suggests that iron is effectively retained on the bentonite surface, thereby reducing iron leaching into treated solution [54]. These findings confirm that the hybrid system improves removal efficiency while avoiding secondary contamination, supporting its operational stability and suitability for real treatment applications.

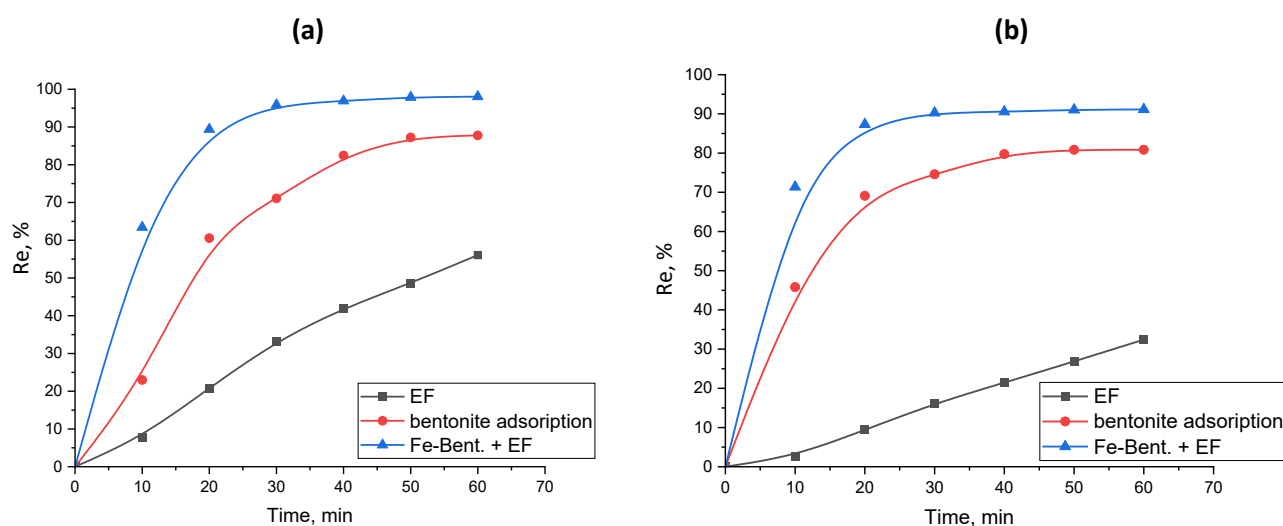


Figure 4. Removal efficiency of (a) MG and (b) MB by electro-Fenton, bentonite adsorption, and heterogeneous electro-Fenton using Fe-Bent. particles

Effect of operating parameters on performance of Fe-bentonite heterogeneous electro-Fenton process

Effect of current density

In this part of the research, Figures 5a and 5b illustrate how current density affects the rate of MB and MG elimination, respectively. When the current density increases from 8 to 20 mA cm⁻², the MB and MG elimination rate increases from 80.032 to 97.914 % for MG removal, and from 76.396 to 90.638 % for MB removal, respectively. These results can be explained by the higher rate of oxygen reduction on the cathode surface, leading to an increase in the electrical production of H₂O₂ as illustrated in Equation (1), and then by the Fenton reaction, when H₂O₂ reacts with the Fenton catalyst loaded onto bentonite particles, as shown by Equation (3). As a result, a larger amount of OH[•] was generated and attacked the dye molecules directly on the catalyst surface, reducing mass-transfer limitations and enhancing the degradation rate. Moreover, increasing j from 8 to 20 mA cm⁻² increased the energy consumption (EC) from 0.864 to 3.216 kWh L⁻¹, while 20 mA cm⁻² required more energy than lower j (8 mA cm⁻²), but it gave the greater improvement in dye removal. By increasing the j beyond this value, the degradation efficiency did not increase noticeably while EC continued to increase. This indicates that part of the applied current was consumed in parasitic reactions rather

than contributing to H_2O_2 generation. These side reactions include oxygen formation on the anode (Equation (8)), hydrogen evolution occurring at the cathode (Equation (9)) and the recombination of $\text{OH}^\bullet$ (Equation (10)) [55]. The H_2O_2 generation rate was not measured in this study; therefore, the proposed mechanism is based mainly on the degradation trend observed during the experiments and on previous electro-Fenton studies [39].

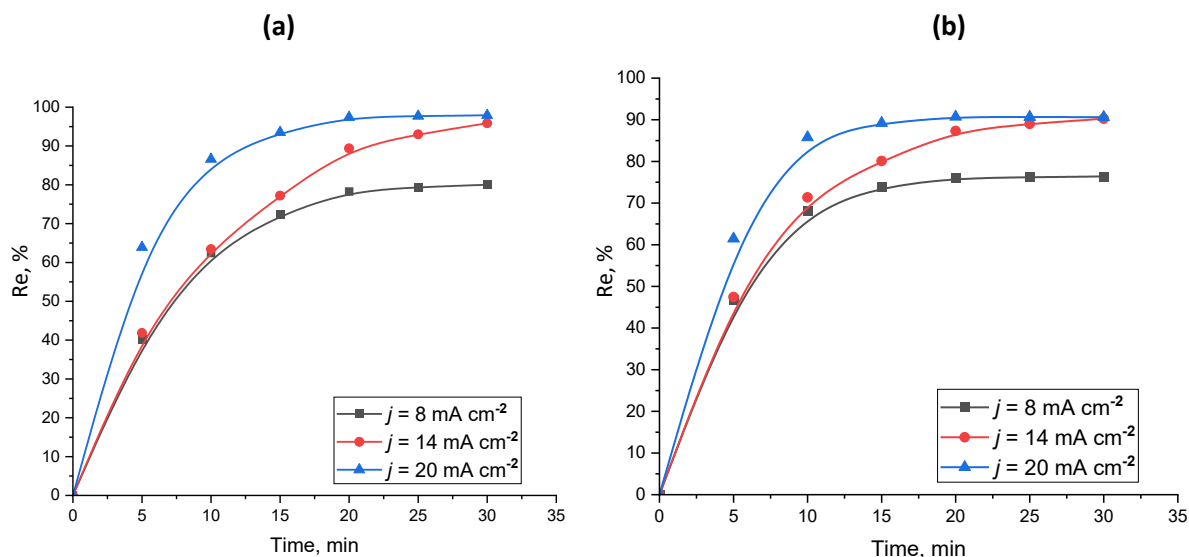


Figure 5. Effect of current density on the removal efficiency of (a) MG, and (b) MB, under the following operating conditions: Na_2SO_4 concentration = 0.5 M, air flow rate = 0.4 L min^{-1} , Fe*-Bent. dose = 0.2 mM, Fe-Bent.* dose = 2 g and pH 3

Effect of bentonite quantity variable

The analysis of Figure 6 of the Re of MG and MB, respectively, at different Fe-Bent.* doses (1, 2 and 3 g), while maintaining a constant iron loading (0.2 mM), reveals that increasing the Fe-Bent.* dose leads to an observable change in system behaviour, which is shown by differences in removal efficiency. At a Fe-Bent.* dosage of 1 g, the removal curves exhibit a gradual increase with relatively low slopes, particularly during the early stages of treatment. This pattern indicates that the available catalytic surface area does not provide enough active sites to sustain rapid $\text{OH}^\bullet$ generation. As a result, removal still occurs but at a slower rate, as shown by the moderate curve slopes. This trend can be attributed to surface reactions and mass-transfer effects, especially in systems treating dye mixtures. Increasing the Fe-Bent.* dosage to 2 g leads to a pronounced change in curve shape. As shown in Figures 6(a) and 6 (b), the curves have a high slope, approaching high removal levels more quickly. This behaviour suggests an improvement in catalytic site density and iron distribution on the bentonite surface. Under these conditions, electro-generated H_2O_2 rises, and the production of $\text{OH}^\bullet$ is more effective. So, the surface area limitation observed at a 1 g dose of Fe-Bent.* is reduced. When 3 g of Fe-Bent.* was applied, the removal curves rose sharply at the beginning and then stabilized at high removal percentages, particularly for MG. This may initially suggest improved performance due to the increase in effective surface area and a higher number of heterogeneous catalytic sites. The early stabilization of the curves indicates that the process becomes limited by mass transfer and the amount of pollutant rather than by catalytic activity [54]. Kinetic analysis based on a pseudo-first-order model was performed to quantitatively support the obtained behaviour as shown in Figure 7. The obtained

plots indicate that MG removal shows an excellent fit with the pseudo-first-order model ($R^2 = 0.92$ to 0.99). For MB, there was an acceptable fit at lower dosages ($R^2 = 0.964$), whereas a significant deviation was noted at 3 g of Fe-Bent.* ($R^2 = 0.77$), which shows a departure from the ideal pseudo-first-order behaviour, especially at conditions of high bentonite loading. This behaviour can be attributed to possible mass transfer limitations, due to particles that tend to aggregate, or competitive interactions between dye molecules at elevated Fe-Bent.* dosage.

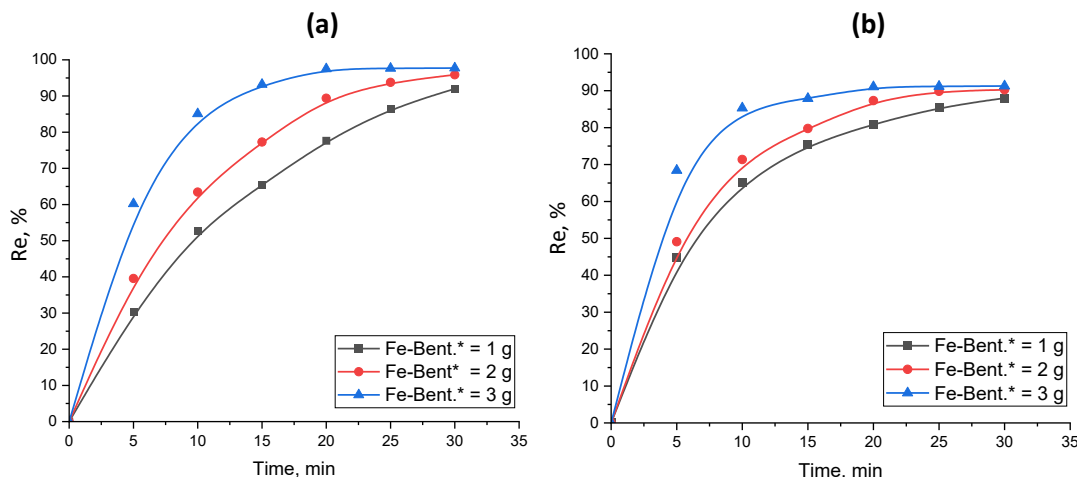


Figure 6. Effect of Fe-Bent.* dose on the removal efficiency of (a) MG and (b) MB under the following operating conditions: $j = 14 \text{ mA cm}^{-2}$, Na_2SO_4 concentration = 0.5 M , air flow rate = 0.4 L min^{-1} , $\text{Fe}^*\text{-Bent. dose} = 0.2 \text{ mM}$ and $\text{pH } 3$

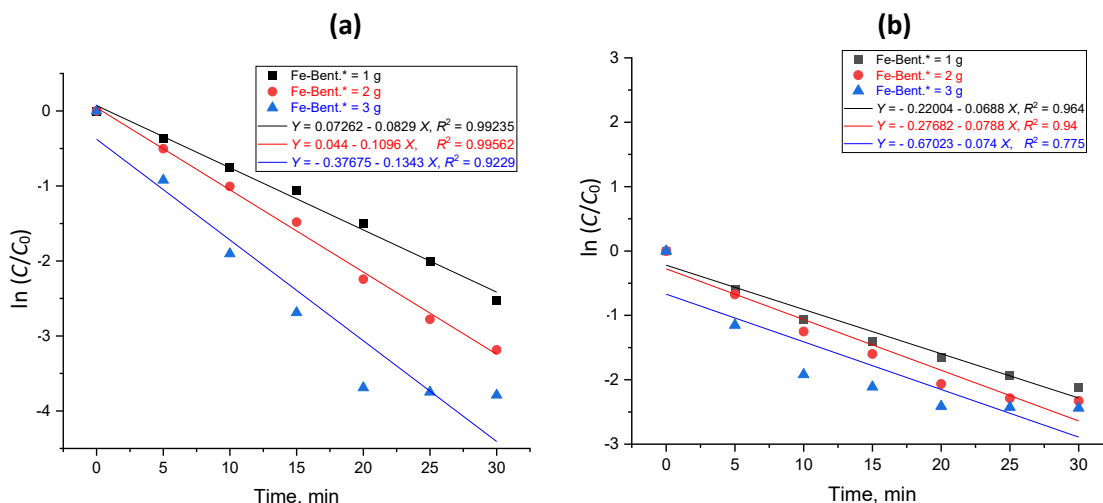


Figure 7. Pseudo-first-order kinetic plots of (a) MG and (b) MB removal under the following operating conditions: $j = 14 \text{ mA cm}^{-2}$, Na_2SO_4 concentration = 0.5 M , air flow rate = 0.4 L min^{-1} , $\text{Fe}^*\text{-Bent. dose} = 0.2 \text{ mM}$ and $\text{pH } 3$

For more explanation of this behaviour, the values of adsorption capacities (q_{MG} for MG and q_{MB} for MB) were estimated. As presented in Table 4, for all Fe-Bent.* dosages, the dye uptake per unit mass of Fe-Bent.* decreases with increasing Fe-Bent.* dosage. The decline in adsorption capacities means that many active sites remain unsaturated at higher doses, so the enhancement in the colour removal may come from the increase in adsorbent mass rather than enhanced per-site efficiency [54].

Overall, increasing Fe-Bent.* dosage improves the total removal efficiency; however, it is accompanied by a decrease in adsorption capacity and a clear deviation from pseudo-first-order kinetics. This behaviour suggests that the system gradually shifts from a kinetic control to a more diffusion-limited one at higher Fe-Bent.* dosages.

Table 4. Adsorption capacity for MG and MB at different Fe-Bent.* dosages

Fe-Bent.* dosage, g	$q_{MG}, \text{mg g}^{-1}$	$q_{MB}, \text{mg g}^{-1}$
1	12.28	10.42
2	6.32	5.18
3	4.19	4.06

Effect of iron concentration variable

Figure 8 presents the removal of MG and MB at different Fe*-Bent. dosages using 2 g of bentonite. The results show that the removal efficiency does not increase linearly with increasing iron concentration. An optimal iron loading can be identified from the curves at which the highest removal efficiency is achieved. At 0.1 mM Fe*-Bent. dosage, the removal curves exhibited a rapid increase at the beginning, followed by a slower approach to equilibrium. This suggests effective OH• generation from the early stages of the process because of the effect of the production of H₂O₂ on the bentonite surface, without noticeable radical inhibition. In addition, the difference in removal efficiency between MG and MB confirms that MG undergoes oxidation more readily than MB. When the Fe*-Bent. dosage is increased to 0.2 mM, the removal curves show a lower slope, particularly at the end of the treatment period. While removal still increases with time, the slower rate suggests that the decrease in efficiency is due to the radical utilization. This behaviour can be explained by the presence of excess iron effects, where Fe²⁺/Fe³⁺ species compete with dye molecules for OH•, slowing the overall oxidation rate. At 0.3 mM of Fe*-Bent. dosage, the curves show a limited initial increase followed by rapid stabilization. These results may suggest a scavenging effect of excess iron species, in which OH• is rapidly consumed by excess iron species rather than reacting with dye molecules, as shown in Equations (11) and (12) [29].

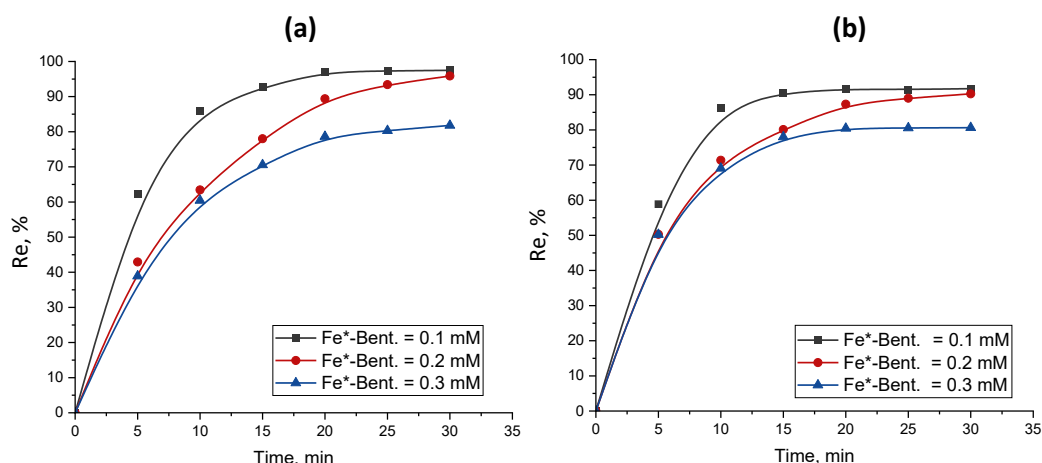


Figure 8. The influence of Fe*-Bent. dosage on the removal efficiency of (a) MG, and (b) MB, under fixed operating conditions: $j = 14 \text{ mA cm}^{-2}$, Na_2SO_4 concentration = 0.5 M, air flow rate = 0.4 L min⁻¹, Fe-Bent.* dose = 2 g and pH 3

Similar behaviour has been reported in previous studies, where increasing the concentration of Fenton catalyst beyond the ideal, decreases the availability of OH• [54]. Additionally, the accumulation of inactive iron complexes on the bentonite surface may limit access to catalytic sites, which affects mass transfer and surface reactions [54]. Similar behaviour was observed for both dyes, suggesting that the performance of the system is mainly controlled by the radical availability rather than the type of dye. However, under the studied conditions, MG always shows higher removal than MB, which agrees with their different oxidative tendencies.

Finally, the results suggest that the best performance of heterogeneous electro-Fenton is not achieved at the highest Fe^{*}-Bent. dosage. Instead, optimal performance was seen at a concentration that allows radicals to oxidize dye efficiently while avoiding excessive radical scavenging.

Conclusions

The heterogeneous electro-Fenton process with iron-bentonite loaded particles for mixed dyes (methylene blue and malachite green) removal was successfully investigated and strongly governed by operational parameters. The current density was identified as the most effective parameter, which is followed by bentonite (Fe-Bent.*) and iron (Fe^{*}-Bent) dosages. It was found that the removal efficiency increased with increasing j up to 20 mA cm⁻², higher j promotes the electrogeneration of H₂O₂ and then the formation of hydroxyl radicals (OH^{*}), while the decrease in j shows incomplete dye removal. Increasing Fe-Bent.* dosage up to 3 g significantly improved removal efficiency by increasing surface area and the interplay between oxidation and adsorption. The best elimination efficiency was demonstrated by a dosage with a varied iron concentration between 0.1 and 0.2 mM, while increasing of Fe^{*}-Bent up to 0.3 mM negatively affected the process performance due to OH^{*} scavenging. The effect of current density and Fe^{*}-Bent. dosage on the removal efficiency, along with the considerable COD reduction, suggested that the degradation process mainly depends on hydroxyl radical formation during the electro-Fenton reaction. Comparison between treatment methods showed that the electro-Fenton process alone is limited by iron dissolution and by the oxidation efficiency of the medium, whereas bentonite adsorption achieves high decolorization but incomplete degradation. The heterogeneous electro-Fenton process combines the advantages of both processes, delivering high dye removal while enhancing catalyst stability and minimizing iron release. Finally, the results confirmed that the heterogeneous electro-Fenton process is a more effective and environmentally friendly strategy for removing mixed dye contaminants than traditional methods.

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