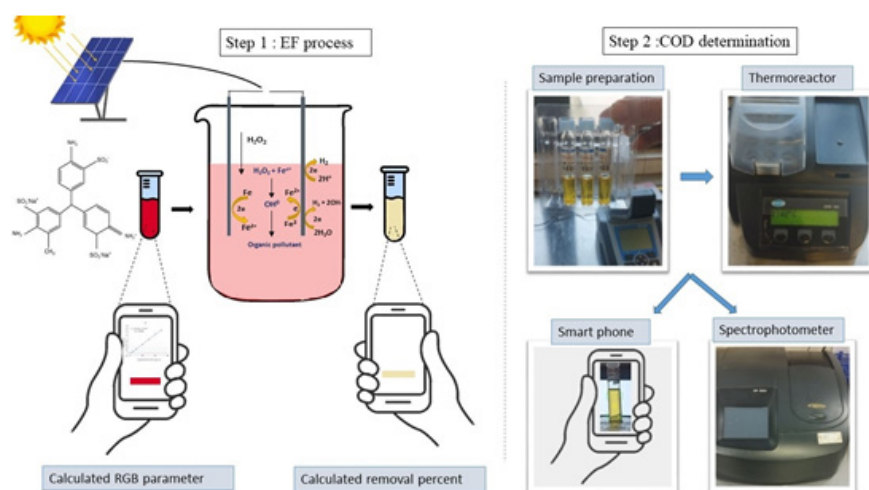


ARTICLE

Response Surface Optimization of Acid Fuch sine Removal using Electro-Fenton coupled with Smartphone-based Colorimetry and Chemical Oxygen Demand Measurements

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Today, smartphone-based digital colorimetry is used as an inexpensive and portable tool. In this study, electro-Fenton (EF) method was coupled with a smartphone for acid fuch sine (AF) dye removal. In this method, the colored sample was photographed before and after the EF process. Then, the Red, Green, and Blue (RGB) and Hue, Saturation, and Value (HSV) codes of the image were extracted using Color Grab software. Experimental parameters were optimized by the central

composite design (CCD). Optimized parameters were pH = 3, voltage = 20 V, time = 12 min, $C_{H_2O_2} = 10^{-5}$ mol L⁻¹. Under these conditions, 96.2% AF removal was attained. The chemical oxygen demands (COD) evaluated by the EF processes were in good agreement with each other. In this EF system, a 65% decrease in COD was observed after 12 min.

Keywords: acid fuch sine, central composite design, chemical oxygen demand, electro-Fenton, smartphone

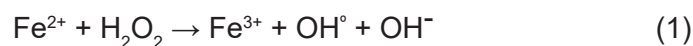
INTRODUCTION

The world's industrialization is causing an increase in water pollution every day. Many industries produce wastewater containing dyes, poisons, and non-degradable compounds and introduce them into the environment. Colored compounds in wastewater not only cause environmental pollution but also cause serious health risks to living organisms. The textile industries use approximately 10,000 tons of dyes and pigments annually. Therefore, it is necessary to treat these dye wastewaters before they are discharged and contaminate surface waters.¹⁻³

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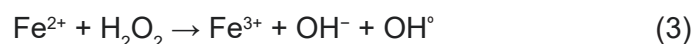
The production of strong radicals for the mineralization of pollutants leads to the widespread use of advanced oxidation methods (AOP).⁴ The production of hydroxyl radicals can be widely achieved through Fenton and Haber-Weiss reactions. The Fenton reaction-based oxidation methods are widely used because of their simplicity and low toxicity. Hydroxyl radical (OH°) is produced during the Fenton reaction (1) and is a strong oxidant that attacks pollutants.⁵



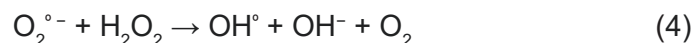
The Haber-Weiss reaction⁶ is a reaction between hydrogen peroxide and superoxide, which leads to the production of hydroxyl radicals. The reaction is kinetically slow, but is catalyzed by dissolved iron ions. The first step of the catalytic cycle involves the reduction of the ferric (Fe^{3+}) ion into the ferrous (Fe^{2+}) ion (2):



The second step is the Fenton reaction (3):



Net reaction (4):



In Fenton's method, Fe^{2+} and H_2O_2 reagents were manually added to the solution. To increase the efficiency, at least one of the two reactants or both are electrically produced on site (here, iron), which is called the electro-Fenton process. This method produces higher efficiency, faster speed, and less sludge than the Fenton method.^{3,7,8}

Colorimetry based on image processing with smartphones has been developed in recent years.⁹⁻¹² Due to their portability, fast response, easy use, and improved quality of smartphone cameras, smartphones are becoming increasingly popular. Manufacturing companies have increased efficiency by improving smartphone software capabilities. Software (such as Color grab, Color picker, Color detector & catcher) provides colorimetric data from the sample image. These data are red, green, blue codes (RGB) and hue, saturation, value codes (HSV). For example, the RGB scale provides values between 0 and 255 for each color, the values {0, 0, 0} represent absolute black and the values {255, 255, 255} represent true white.¹³

In COD, amount of oxygen for oxidization of chemicals in wastewaters is measured and expressed as mg L^{-1} .¹⁴ COD measurement is based on the reaction of sample with potassium dichromate and concentrated sulfuric acid as a strong oxidizer, and the expected response can be evaluated colorimetrically by a spectrophotometer. However, today, the replacement of the intended devices is increasing due to the expansion of smartphone technology and easy access.

Different methods such as biosorption^{15,16} and electro-Fenton¹⁷ have been used for AF removal. In this study, the electro-Fenton method was used to remove AF dye from samples and $\text{RGB}_{\text{parameter}}$ analytical curve from smartphone-based colorimetry was used for the determination of dye concentration. Process variables were optimized by experimental design and response surface methodology (RSM). The amount of AF removal under optimal conditions was compared for spectrophotometer and smartphone methods. In this end, the COD value of the solution under optimal conditions was measured using a smartphone and compared with reference methods.

MATERIALS AND METHODS

Materials

KCl, NaCl, Na_2SO_4 , NaOH, H_2SO_4 , H_2O_2 , acid fuchsine (AF) and potassium hydrogen phthalate (KHP) were obtained from Merck (Darmstadt, Germany). Metrohm pH meter (model 780, Swiss) was used for pH measurements. Two iron electrodes and a solar power supply (both made in Iran) were used to perform the electro-Fenton process. This power source uses a 10 W photovoltaic panel (39×34 cm, made of silicon semiconductors) that directly converts sunlight into electricity. Additionally, a 12 V (5 A) battery stores energy in this equipment. This panel is connected to the power source via a cable. The power source has positive and negative terminals connected to the anode and cathode electrodes immersed the solution. Xiaomi phone POCO X4 (storage: 256, ram: 8, camera: 108 mega pixel), Huawei Mate 10 Lite (source of light), commercial vials TNT821 (0-150 mg L^{-1} , HACH), thermal reactor (DR200, HACH), and spectrophotometer (DR 5000, HACH) were also used.

Electro-Fenton process

To remove the AF dye with the help of the electro-Fenton process, 42 mL AF dye (500 mg L^{-1}) were added to optimized amounts of Na_2SO_4 as an electrolyte and hydrogen peroxide. Final volume of mixture was 300 mL. Next, two iron electrodes (5×12 cm) were immersed into the solution by means of a non-conductive holder and connected to solar power source.

Colorimetric measurements

The amount of AF dye removal was determined by measuring one of the RGB, HSV, and HSL (here, RGB) values by using a smartphone. In colorimetry with the help of a spectrophotometer, the color intensity of the solution was calculated according to absorption at a maximum wavelength. The correlation between absorption and solution concentration can be obtained by drawing an analytical curve based on the Beer-Lambert law. Smartphones can record these color changes basing on RGB, HSV, and HSL values and processing image color by colorimetry analysis software (here, COLOR GRAB) to corresponding RGB values.^{18,19} Two smartphones were used for the experiment. The screen of one of the phones was used as a white light source, and the other phone was used to take pictures and record RGB values. The samples were photographed in a box with dimensions of ($25 \times 18 \times 12$ cm) to eliminate any disturbing lights (Figure 1).

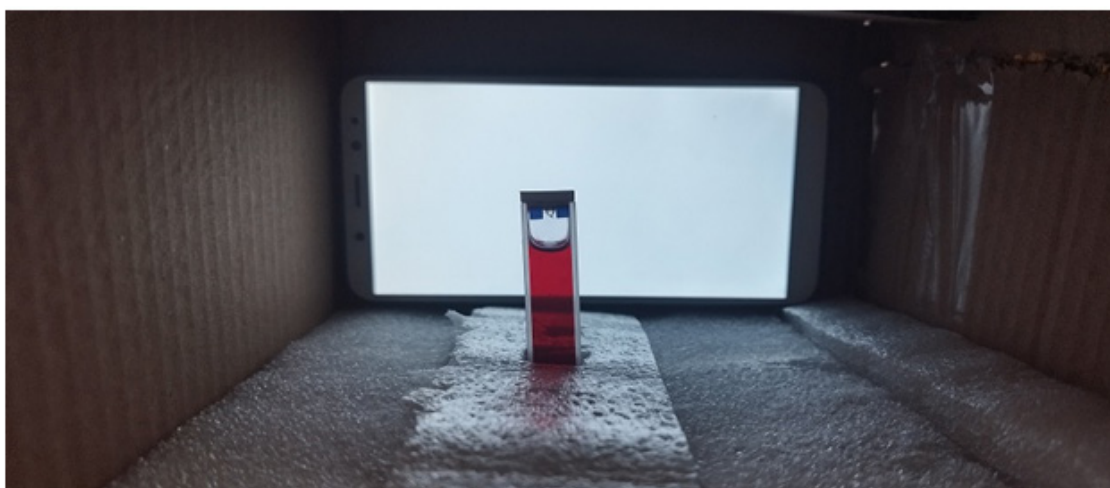


Figure 1. Home-made light box for taking sample photos.

At first, the blank solution (without dye) was photographed, and its R, G, and B parameters were obtained with the help of COLOR GRAB software. Thereafter, the R, G and B parameters were estimated according to Equation 5. Next, AF dye solutions at different concentrations were prepared, each one was photographed and the RGB values were obtained through Equation 6. Because the relationship between RGB and concentration is not linear,²⁰ two methods for drawing the analytical curve were used. In the first method, a logarithmic scale was used to facilitate the interpretation of the results.

Method 1

$$RGB_{\text{reference}} = (R+G+B)/3 \quad (5)$$

$$RGB_{\text{sample}} = (R+G+B)/3 \quad (6)$$

$$RGB_{\text{parameter}} = \log (RGB_{\text{reference}} / RGB_{\text{sample}}) \quad (7)$$

which is written as Equation 8:

$$(I_{\text{RGB}})_{\text{parameter}} = \log [(I_{\text{RGB}})_{\text{reference}} / (I_{\text{RGB}})_{\text{sample}}] \quad (8)$$

In the second method, the values of R, G, and B were calculated for each concentration (as before), and then the normalized value of R was obtained for each sample using Equation 9.

Method 2

$$R_{\text{normalized}} = R / R+G+B \quad (9)$$

AF dye removal was calculated using Equation 10:

$$\% \text{ Removal} = (C_0 - C_e) / C_0 \times 100 \quad (10)$$

where C_0 is the initial concentration and C_e is the equilibrium concentration of the dye solution.

The central composite design

Central composite design (CCD), a combination of full or fractional factorial and star design, is one of the most used optimization approaches. By using CCD, effect of the factors, factors interactions, experimental error and the optimum conditions can be determined. The five-level, four-factors CCD was used here, meaning 30 experiments, 16 full factorial points, 8 axial points, and 6 center points. Random error can be estimated from the six center points. For a 4-factor CCD, each factor has five levels (-1, 0, 1, + α , - α). The distance of each factor from the center point is ± 1 unit for factorial points and is $\pm \alpha$ unit for star points. The α is usually set at 2.0 for a four-factor design ($\alpha = 2^{n/4}$; $n = 4$, for 4 factors).²¹ Important experimental parameters in the electro-Fenton method were pH of the solution, applied voltage, the concentration of hydrogen peroxide $C_{\text{H}_2\text{O}_2}$, and process time t . These variables were chosen based on the results obtained from the preliminary study. This design was done with four factors ($X_1 = \text{pH}$, $X_2 = V$, $X_3 = C_{\text{H}_2\text{O}_2}$, $X_4 = t$) in 5 levels (-2, -1, 0, 1, and 2) and the results are shown in Table I. To avoid bias, 30 runs were carried out in a totally random order. In electro-Fenton process optimization, ranges of variables were pH (2 – 4), time (2 – 22 min), voltage (10 – 30 V), and concentration of H_2O_2 (10^{-6} – 10^{-4} mol L^{-1}). A second order polynomial equation was fitted to experimental data in response surface methodology by using Minitab 16 (Minitab Inc., USA).

Table I. Central composite experimental design for AF removal by electro-Fenton process

Run	X ₁	X ₂	X ₃	X ₄	pH	Voltage (V)	Time (min)	C _{H₂O₂} (mol L ⁻¹)	AF removal (%)
1	0	0	0	2	3	20	12	1×10 ⁻⁴	76.8
2	1	-1	-1	-1	3.5	15	7	5×10 ⁻⁶	45.7
3	-1	1	1	-1	2.5	25	17	5×10 ⁻⁶	69.6
4	0	2	0	0	3	30	12	1×10 ⁻⁵	84.9
5	-1	1	-1	-1	2.5	25	7	5×10 ⁻⁶	57.2
6	1	-1	-1	1	3.5	15	7	5×10 ⁻⁵	60.3
7	1	1	1	-1	3.5	25	17	5×10 ⁻⁶	76.2
8	-1	1	-1	1	2.5	25	7	5×10 ⁻⁵	64.7
9	1	-1	1	1	3.5	15	17	5×10 ⁻⁵	78.1
10	-2	0	0	0	2	20	12	1×10 ⁻⁵	40.2
11	0	0	0	-2	3	20	12	1×10 ⁻⁶	79.1
12	0	0	0	0	3	20	12	1×10 ⁻⁵	88.6
13	-1	1	1	1	2.5	25	17	5×10 ⁻⁵	81.8
14	-1	-1	-1	1	2.5	15	7	5×10 ⁻⁵	48.6
15	1	-1	1	-1	3.5	15	17	5×10 ⁻⁶	61.6
16	0	0	0	0	3	20	12	1×10 ⁻⁵	91.7
17	1	1	-1	1	3.5	25	7	5×10 ⁻⁵	70.8
18	0	-2	0	0	3	10	12	1×10 ⁻⁵	49.7
19	0	0	-2	0	3	20	2	1×10 ⁻⁵	45.8
20	0	0	0	0	3	20	12	1×10 ⁻⁵	86.2
21	1	1	1	1	3.5	25	17	5×10 ⁻⁵	84.7
22	0	0	2	0	3	20	22	5×10 ⁻⁵	87.9
23	-1	-1	1	-1	2.5	15	17	5×10 ⁻⁶	47.9
24	1	1	-1	-1	3.5	25	7	5×10 ⁻⁶	61.8
25	-1	-1	-1	-1	2.5	15	7	5×10 ⁻⁶	39.8
26	0	0	0	0	3	20	12	1×10 ⁻⁵	84.9
27	0	0	0	0	3	20	12	1×10 ⁻⁵	87.7
28	2	0	0	0	4	20	12	1×10 ⁻⁵	51.6
29	0	0	0	0	3	20	12	1×10 ⁻⁵	90.2
30	-1	-1	1	1	2.5	15	17	5×10 ⁻⁵	59.6

COD measurements with smartphone

This method was based on drawing the analytical curve of the standard solutions of potassium hydrogen phthalate (KHP) with the help of a smartphone and color processing using the COLOR GRAP software.²² Therefore, the HSV (hue, saturation, value) parameter was used. The COD measurement experiment was done in two steps:

Step 1: Drawing of KHP analytical curve: 2 mL of KHP solution (at different concentrations, Table II) was transferred to commercial vials for measuring COD. Vials were stirred well to be uniform, then heated at 150 °C for 2 h and placed in the particular reactor for COD. After the vials temperature reached the room temperature, with the help of a smartphone and the COLOR GRAP software, each sample's HSV (S) values were calculated, and the analytical curve was drawn.

Step 2: 2 mL of the wastewater sample with an unknown COD was added to the vial (if it becomes cloudy, it should be diluted). Then, it was heated at 150 °C for 2 h and the values of S calculated using a smartphone. In all stages, 2 mL distilled water was added to the commercial vial and use it as a blank solution in all stages.

Table II. KHP standard solutions

Standard	KHP concentration (mg L ⁻¹)	Theoretical COD (mg O ₂ L ⁻¹)
1	0	0
2	16	19
3	44	52
4	72	84
5	100	117
6	128	150

RESULTS AND DISCUSSION

Electro-Fenton with smartphone colorimetry

It was found that the RGB values decreased with the decrease in color concentration (thus AF concentration) (Figure 2). Then, two analytical curves based on RGB_{parameter} and R_{normalized} values and AF concentrations were drawn and compared with the analytical curve obtained from the spectrophotometer at the maximum wavelength of 540 nm (Figure 3). The correlation coefficients (r^2) were 0.9991 (n=7) for the spectrophotometer, 0.9986 (n=6) for RGB_{parameter} (method 1) and 0.9149 (n=6) for R_{normalized} (method 2) smartphone colorimetry. The spectrophotometer and RGB_{parameter} smartphone colorimetry methods had a good agreement.

Effects of initial dye concentration, electrodes distance, type and concentration of electrolyte on dye removal in electro-Fenton were studied by one-at-a time design (Figure 4). Maximal removal was observed at the initial dye concentration of 70 mg L⁻¹, electrodes distance of 1 cm and Na₂SO₄, 80 mg L⁻¹. Three electrolytes, Na₂SO₄, KCl, and NaCl, were investigated, and Na₂SO₄ was the most effective in AF removal. Different concentrations of Na₂SO₄ were used, and the highest efficiency was observed at a concentration of 80 mg L⁻¹. The distance between the electrodes (1 – 4 cm) was also examined. Increasing the distance between the electrodes decreased the removal efficiency. In fact, with the increase of the distance from the optimal value, the electrical resistance increases, resulting in a decrease in the conductivity of the solution and a reduction in current consumption. With the decline in the consumed current, the production of Fe²⁺ ions, followed by OH[•], decreases.²³ Next, the initial concentration of AF was investigated in the range of 30 – 80 mg L⁻¹. As the concentration increased from 30 to 70 mg L⁻¹, the amount of AF removal increased.

However, beyond 70 mg L^{-1} , color removal decreased. At constant H_2O_2 concentration, a certain amount of OH° is produced; therefore, by increasing the concentration of AF beyond the optimal value, the amount of OH° becomes insufficient to remove the dye.²⁴ Analytical curve of $\text{RGB}_{\text{parameter}}$ values vs. AF concentration were used for determinations.

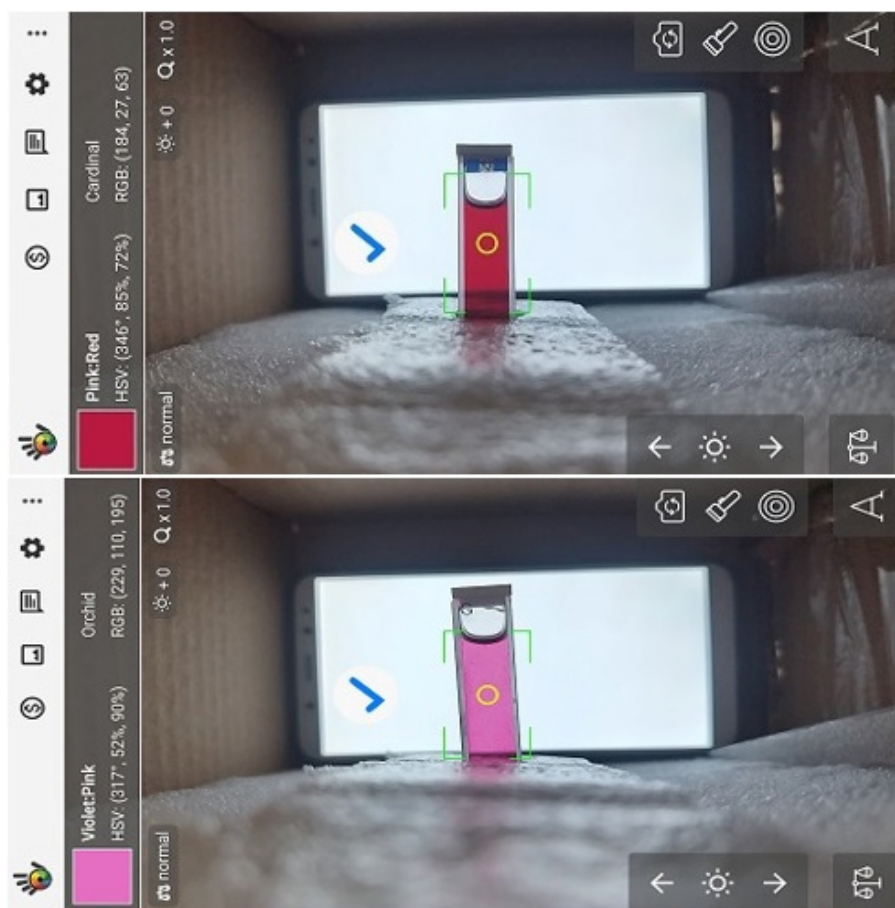


Figure 2. Initial tests for different AF standard solutions.

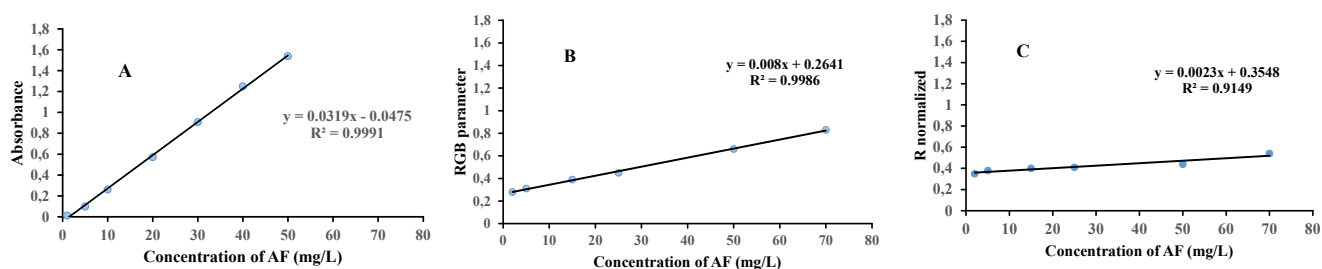


Figure 3. Analytical curves for AF concentration based on (A) UV-Vis spectrophotometry, (B) $\text{RGB}_{\text{parameter}}$, (C) $R_{\text{normalized}}$.

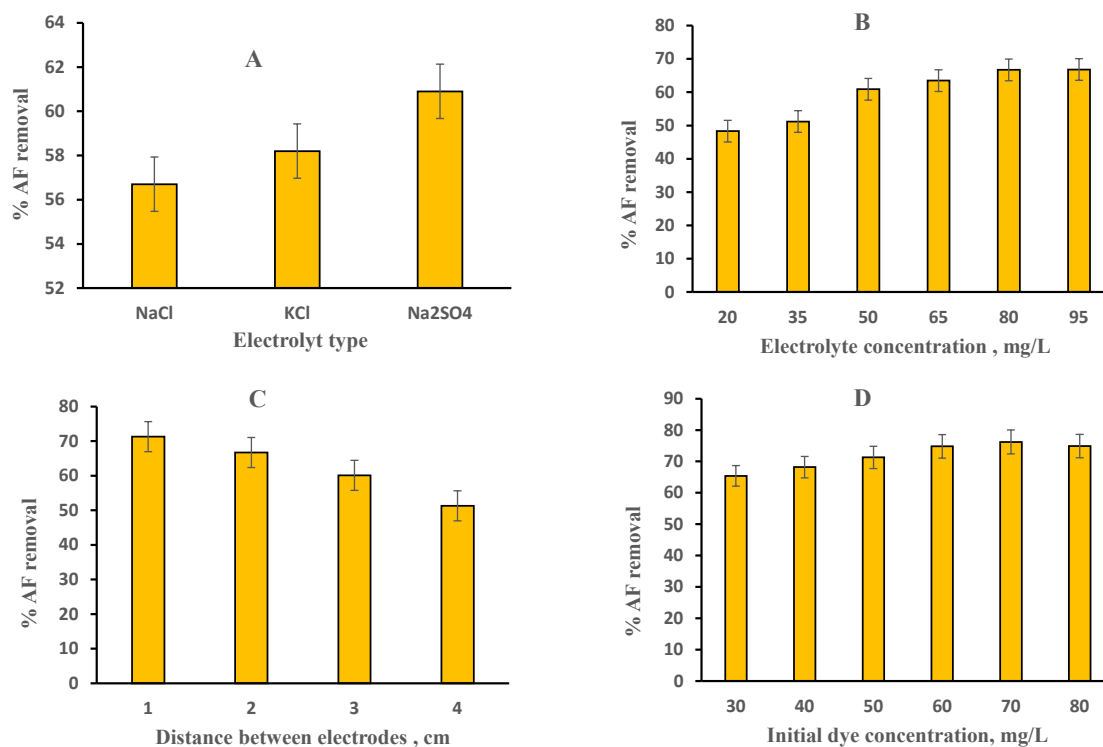


Figure 4. Effect of (A) electrolyte type, (B) electrolyte concentration, (C) electrodes distance and (D) initial dye concentration on the AF dye removal (%) in the electro-Fenton process (voltage = 25 V, pH = 3.5, time = 20 min, and $[H_2O_2] = 5 \times 10^{-6} \text{ mol L}^{-1}$).

Other important parameters such as pH, applied voltage, H_2O_2 concentration and time were optimized by experimental design. Five levels were used for each variable and the % AF removal was the response (Table I). Second order polynomial RSM model for % AF removal was as Equation 11.

$$\begin{aligned} \% \text{ AF removal} = & \mathbf{-553} + \mathbf{284 \times pH} + \mathbf{7 \times time} + \mathbf{13 \times Voltage} + \mathbf{3.87 \times 10^6 \times [H_2O_2]} \\ & - \mathbf{45 \times (pH)^2} - \mathbf{1 \times 10^{-4} \times (time)^2} - \mathbf{1 \times 10^{-4} \times (Voltage)^2} - \mathbf{6.36 \times 10^9 \times [H_2O_2]^2} - \\ & \mathbf{1 \times pH \times Voltage} + \mathbf{1 \times 10^{-4} \times pH \times time} + \mathbf{6.81 \times 10^4 \times pH \times [H_2O_2]} + \mathbf{1 \times 10^{-4} \times Voltage \times time} - \\ & \mathbf{8.53 \times 10^3 \times Voltage \times [H_2O_2]} + \mathbf{1.29 \times 10^4 \times time \times [H_2O_2]} \end{aligned} \quad (11)$$

$$R^2 = 0.968; \text{Lack of fit} = 0.082 \text{ } (p > 0.05).$$

The first and second order terms (in bold) were significant, ANOVA results are presented in Table III, and the RSM contour plots are shown in Figures 5 and 6. Under the optimized conditions (pH = 3, t = 12 min, $[H_2O_2] = 10^{-5} \text{ mol L}^{-1}$ and voltage = 20 V), the AF removal was 96.2%.

The optimum pH in this electro-Fenton system is consistent with previous reports.^{5,8} Deactivation of catalysts occurs at pH < 3 and precipitation of iron species at pH > 4. Thus, the best efficiency in H_2O_2 and Fe^{2+} reaction occurs in pH range of 3-4. The removal efficiency increased with increasing voltage until the 20 V and then decreased. More Fe^{2+} production led to increased removal efficiency. At higher voltages, increasing the Fe^{2+} concentration reduces the efficiency, as this ion reacts with hydroxyl radical. Dye degradation gradually increased with time and reached to maximum at 12 min.

Table III. Estimated regression coefficients, T-value and *P*-value (upper) and ANOVA results (lower)

Term	Coefficient	SE Coefficient	T	P
Constant	-553	47	-11.7	0.000
pH	284	22	13.0	0.000
V	13	2	6.9	0.000
time	7	2	4.1	0.001
C _{H₂O₂}	387661	338148	1.1	0.270
pH×pH	-45	3	-13.7	0.000
V×V	0.000	0.000	-7.1	0.000
time×time	0.000	0.000	-8.0	0.000
C _{H₂O₂} ×C _{H₂O₂}	-6.4×10 ⁺⁹	1.21×10 ⁺⁹	-5.2	0.000
pH×V	-1	0.000	-1.8	0.097
pH×time	0.000	0.000	0.8	0.436
pH×C _{H₂O₂}	68082	84853	0.8	0.435
V×time	0.000	0.000	0.3	0.769
V×C _{H₂O₂}	-8527	8485	-1.0	0.331
time×C _{H₂O₂}	12963	8966	1.4	0.169

Standard deviation (S) = 4.186; R square (R²) = 96.8%; Adjusted R square (R²_{adj}) = 93.9%.

ANOVA results

Source	DF	Seq SS	Adj SS	Adj MS	F	P
Regression	14	8025.2	8025.2	573.2	32.7	0.000
Linear	4	3626.7	3070.7	767.7	43.8	0.000
Square	4	4265.4	4301.5	1075.4	61.4	0.000
Interaction	6	133.1	133.1	22.2	1.3	0.329
Residual Error	15	262.8	262.8	17.5		
Lack-of-Fit	10	231.2	231.2	23.1	3.7	0.082
Pure Error	5	31.6	31.6	6.3		
Total	29	8288				

DF: Degree of freedom; Seq SS: Sequential (or extra) sum of squares; Adj SS: adjusted sum of squares; Adj MS: adjusted mean squares.

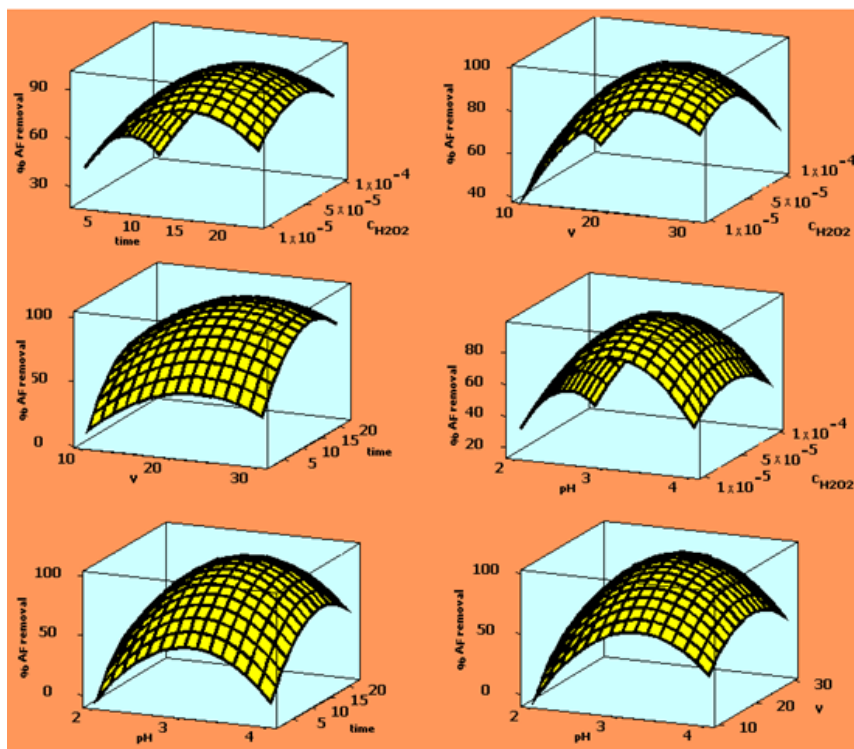


Figure 5. Response surface plots.

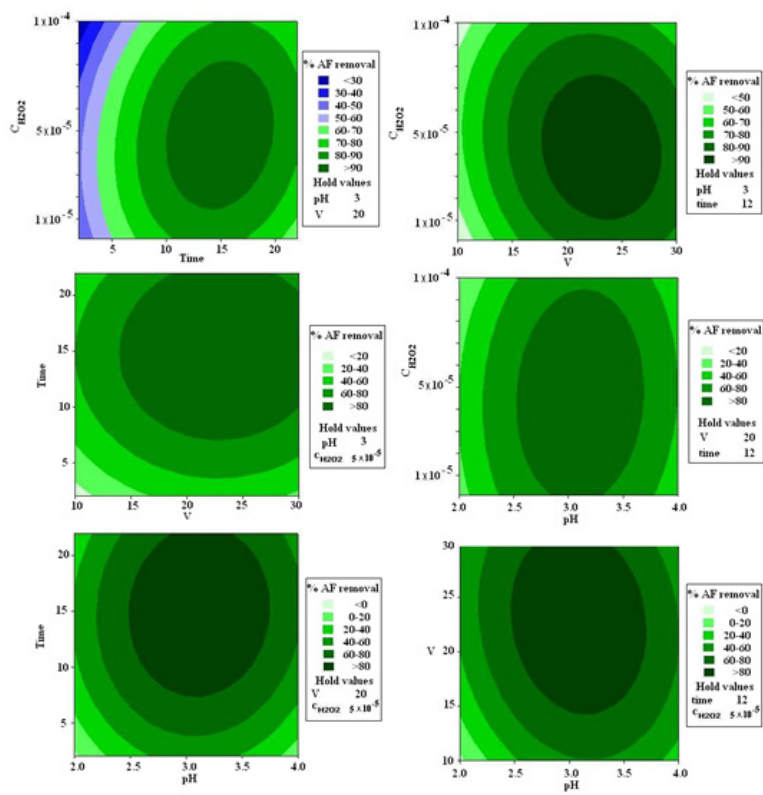


Figure 6. Contour plots. In each plot, two factors are varied but two factors are kept constant at center points. Different dyes indicate % AF removal vs. variation of two factors.

COD measurements with smartphone

Experiments were also carried out by using a smartphone instead of a spectrophotometer for COD measurement. The required analytical curves were attained for potassium hydrogen phthalate (KHP) and color processing involved the COLOR GRAF software.¹⁹ Therefore, the HSV (hue, saturation, value) parameter was used. The saturation of the images shows a linear behavior that can be used to plot the analytical curve and calculate the COD (Figure 7). Theoretically, 1 mg of KHP is equivalent to 1.171 mg L⁻¹ of COD.⁵ COD values of wastewaters before and after the treatment indicates the amount of organics degradation.²⁵ The COD of the colored solution before starting the electro-Fenton process was 56 mg L⁻¹. After completing the electro-Fenton process under optimal conditions, COD value was 19.6 mg L⁻¹ (65% decrease after 12 min).

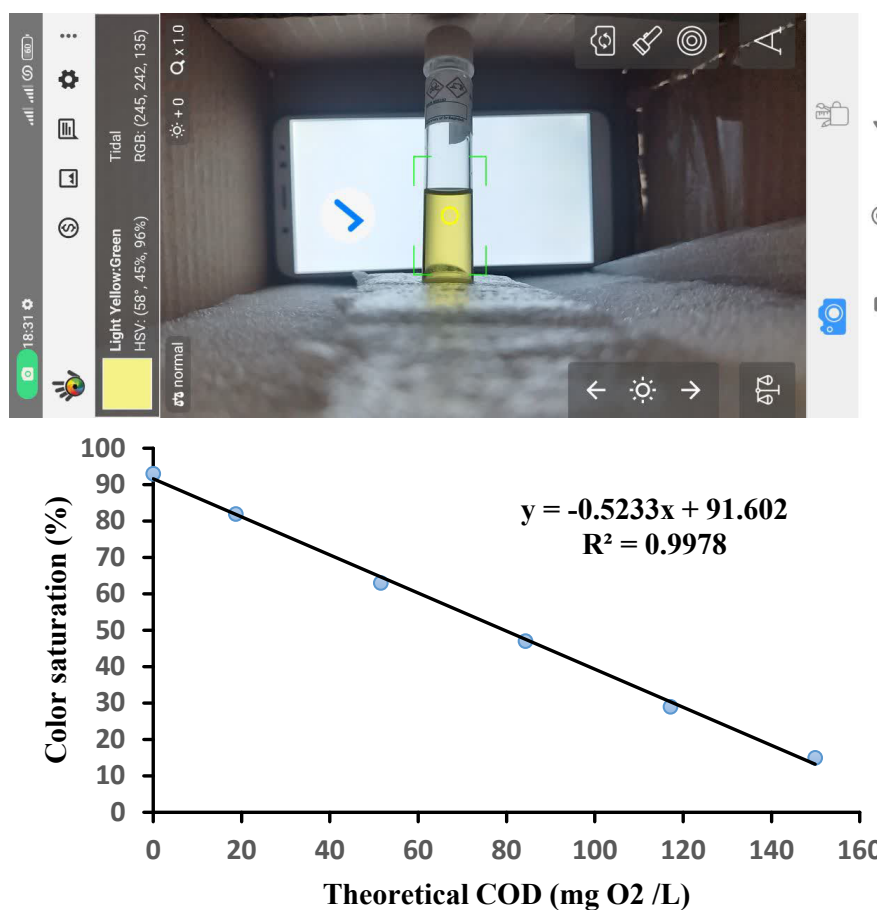


Figure 7. Graphical representation of the color saturation values of KHP standards against its theoretical COD.

Advantages and limitations

Smartphone colorimetry is fast, portable, low-cost, effective, and promptly available. On the other hand, light conditions of the environment, the distance of the camera from the sample, camera parameters (sensitivity, white balance, aperture, etc.) may affect the output image. Colorimetric tests based on smartphones are usually performed under LED light, fluorescence, and sunlight, which yield different results.²⁶ Generally, the sample is placed inside a box with constant lighting. The distance from the samples is maintained consistently for all samples. Additionally, a smartphone is used throughout all stages of photography to minimize the effects of camera parameters on the output images. However, this procedure still requires improvement and development.

CONCLUSIONS

Compared to Fenton, coagulation, and electrocoagulation methods, the electro-Fenton process is a speedy, low-cost, effective, and less sludge-producing method. It becomes more environmental friendly if solar power source provides the required voltage. Additionally, coupling it with a smartphone-based colorimetry provides a portable, fast and cost-effective method for concentration determination. Results obtained with smartphone-based colorimetry were in agreement with those obtained by spectrophotometry. Green analytical chemistry (GAC) aims to reduce the negative effects on the environment by addressing issues such as use of solvents and reagents dangerous for the environment and human health, toxic waste, energy consumption, etc.²⁷ Evaluating the greenness of a method requires specialized tools such as Greenness (A Gree). The criterion for calculating the greenness of a technique is based on the 12 principles (P) of green analytical chemistry (SIGNIFICANCE),²⁸ which is scored from 0 to 1. The method of removing AF color using the EF process coupled with a smartphone was investigated in 12 steps with the help of A Gree software. The resulting pictogram in the Figure 8 indicates relatively suitable compatibility of the method with the environment and human health. In the first stage, the sampling procedure was chosen as at line (P1). A sample of 42 mL is required (P2). The position of the analytical tool is at line (P3), and the mentioned method includes 5 different steps (P4). The process is manual and non-miniature (P5), and no derivative factor is used (P6). The amount of decomposition waste is less than 0.1 g (P7). One sample was measured at each start time, and according to the 12-minute duration of the process, 5 samples can be measured in one hour (P8). Solar light was used to provide the necessary energy for the process (P9), and none of the reagents were from bio-based sources (P10). The solvent used in the whole process was water (P11). Due to the use of small amounts of hydrogen peroxide, there is a small possibility of oxidation and corrosion risks (P12).

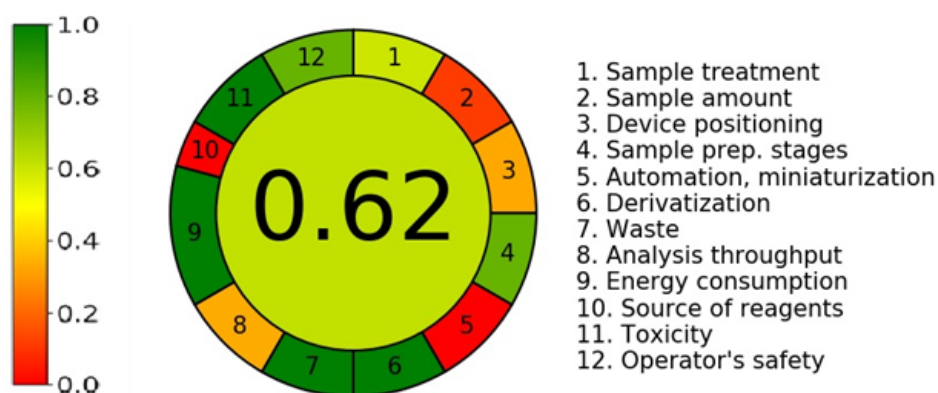


Figure 8. The results of the AGREEprep assessment of the electro-Fenton coupled with smartphone-based colorimetry.

Conflicts of interest

The authors declare no conflicts of interest.

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