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Modeling and Optimization of the Electrochemical Process for Cefixime Removal from Water

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Abstract- Pharmaceutical compounds such as Cefixime (CFX) as an antibiotic were detected in water resources. These compounds in water has caused many problems including bacterial resistance to antibiotics. Some processes have been used to remove these compounds from water sources. In this research the effect of electrocoagulation process was investigated on CFX removal from water. Experiments were carried out as a batch mode in electrochemical reactor that was made from Plexiglas with dimensions of 10×15×10 cm (1 L volume) and Aluminum/Aluminum electrodes (48 cm^2 effective area) that immersed vertically, monopole configuration and in parallel. Central Composite Design (CCD) utilizing a response surface methodology (RSM) was used with experimental points of pH, Initial CFX concentration, current intensity and Reaction time and employed by different model such as linear, interaction and quadratic models. The results of this study showed that the efficiency of four dependent parameters on CFX removal represent with four linear and six interactions effects that the reaction time and initial concentration of CFX have the most positive and negative effects on the removal of CFX (%), respectively. The optimum conditions for the electrocoagulation process was pH=7.5, a primary concentration of CFX=5.75 mg/L, a current intensity=6.0 amperes and a reaction time=72.5 minutes (CFX removal=85.6%).

Keywords- Antibiotic, Cefixime, Electrocoagulation, Aqueous Solutions

1. INTRODUCTION

In recent years, residues of pharmaceutical compounds were detected in aqueous solutions such as wastewater [1], ground water [2,3], surface water [4,5], streams [6] and drinking water [7]. according to research results in Europe, pharmaceutical compound completely isn't removed during the conventional wastewater treatment and 80% of them are discharged to the environment [8]. Today pharmaceutical residues in environmental is considered as a big problem [9,10]. Among the pharmaceutical compounds, antibiotics can increase the resistance in bacteria, genotoxicity and aquatic toxicity [10-15].

CFX is one of the most important of third generation cephalosporin antibiotic [16-18]. An orally CFX can be effectively used against various gram positive and gram negative bacteria, such as hemophilic influenza, Escherichia coli, staphylococcus, tonsillitis, throat infections, febrile streptococcus, etc. [19,20]. This drug is clinically used for the treatment of infection diseases including gonorrhea, otitis media, pharyngitis, pneumonia, bronchitis, syphilis and etc. [16,21,22].

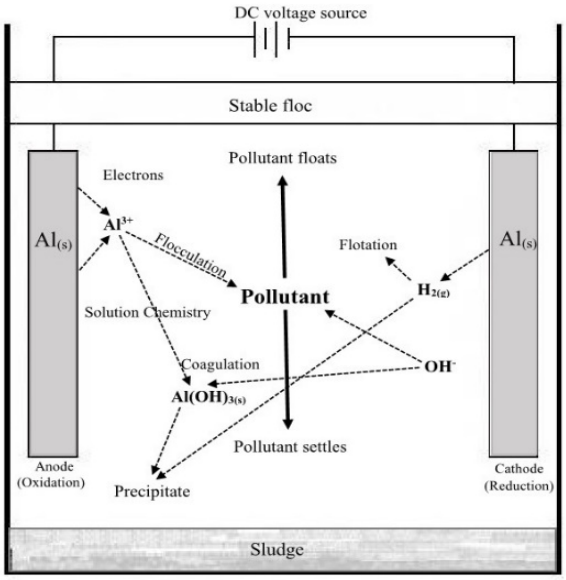


Fig. 1. Schematic of an electrocoagulation reactor

Several techniques such as ozonation [23], wetland [24], reverse osmosis [16], ion change [25], adsorption [26] and photocatalytic degradation [27] have been used for antibiotics removal from water and wastewater. However, among all of them, electrocoagulation (EC) process received providing satisfactory result for the removal of pollutants from wastewater [28,29]. EC reactor is an electrolytic cell with sacrificial electrodes. This process involves applying an electric current between two electrodes where the generates gas bubbles and coagulating agent [30,31]. Iron and aluminum are usually used as sacrificial electrode material in EC process [32]. The ions that are generated in during the process of electrolytic oxidation,

are similar to the addition of a chemical coagulant such as aluminum and ferric chloride in water. Interactions occurring within an EC reactor have been shown in Fig. 1 [33]. In interactions an EC, produces compounds of hydrolysis aluminum; such as $Al(OH)_2^+$, $Al_2(OH)_2^{4+}$, $Al(OH)_4^-$, $Al_6(OH)_{15}^{3+}$, $Al_7(OH)_{17}^{4+}$, $Al_8(OH)_{20}^{4+}$, $Al_{13}O_4(OH)_{24}^{7+}$, $Al_{13}(OH)_{34}^{5+}$ [29].

The EC process has various benefits including, the simple equipment required, easy handling, low operating cost, no addition of chemicals, high removal efficiency of toxic matters at short treatment times, and low sludge production [29,34].

RSM is a statistical technique for designing an experiment, has been employed to optimize, evaluate interactive effects of independent factors and reducing the number of experiments in chemical and biochemical processes [35]. In the present study, RSM was used to evaluate the main effects of various parameters, such as pH, initial CFX concentration, current intensity and reaction time, also their simultaneous interactions to achieve the optimum condition for EC process.

2. MATERIALS AND METHODS

2.1. Chemical and reagents

CFX ($C_{16}H_{15}N_5O_7S_2$, 453.45 mol.wt) were purchased from sigma Aldrich Co (Fig. 2). Other chemicals such as HCl, NaOH, NaCl and methanol, KCl, NaCl, were obtained from Merck Company and then used.

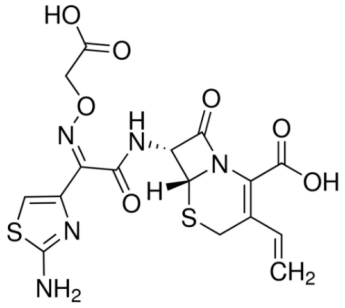


Fig. 2. Chemical structure of CFX

All the solutions were prepared from the stock by using de-ionized water. Experiments were carried out in a batch electrochemical reactor from Plexiglas with dimensions of 10×15×10 cm and 1L capacity (Fig. 3). The electrocoagulation was setup with Aluminum/Aluminum electrodes (48 cm^2 effective area) that immersed vertically, monopole configuration and in parallel. The distance between electrodes was 1.5 cm. A direct current was supplied by using a DC power source. The concentration of the electrolyte was maintained constant at a value of 1.2 ms/cm^2 by adding NaCl as the electrolyte. HCl or NaOH solution

were used for adjusting the pH. Operation of reactor was performed at room temperature (20±2 °C).

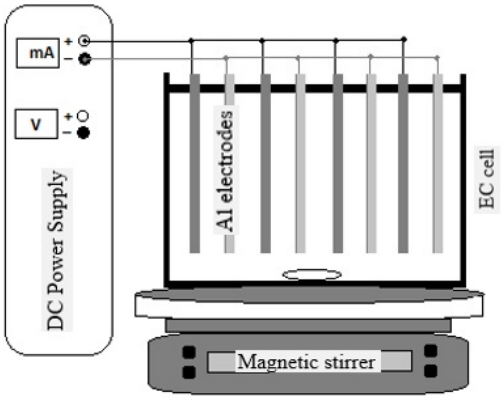


Fig. 3. Schematic experimental setup of the electrocoagulation cell

2.2. Cefixime measurement

CFX concentrations were determined by UV–VIS spectrophotometer (T80 UV/VIS Spectrophotometer, PG instruments Ltd) at the wavelength of 288.5 nm[16]. The standard calibration curve was generated using five solutions in the range of 0.1–20 mg/l with a regression coefficient greater than 0.99. Finally, the removal efficiency of CFX by the EC was calculated by following Eq. (1):

$$R = \frac{C_0 - C_t}{C_0} \times 100 \quad (1)$$

where R is the removal efficiency of CFX (%), C_0 and C_t imply on CFX initial and final concentration (mg/L), respectively.

Table 1. Coded and actual values of numeric factors

Coded variables (X_i)	Factors (U_i)	Experimental field				
		-∝	-1 **level**	0	+1 **level**	+∝
X_1	A-Solution pH	3	4.5	6	7.5	9
X_2	B-Initial CFX concentration (mg/l)	1	5.75	10.5	15.25	20
X_3	C- current intensity (A)	0.3	0.4	0.5	0.6	0.7
X_4	D-Reaction time(min)	5	27.5	50	72.5	95

2.3. Design of the experiments

Experimental points for pH, CFX concentration, Reaction time, and current intensity were set using Central Composite Design (CCD) utilizing a response surface methodology (RSM) category of Design Expert 7 software. (Table 1) [36]. The number of points in CCD contains a center point runs (C_o), axial runs (2k) and factorial run (2^k), where k is the number of variables. Therefore, the total experimental runs (N) of CCD is given by: the sum of center points, axials and factorial ($N=2k+2^k+C_o$).[37] In this study, K was the equal to four. Therefore, all of the runs obtained thirty (16 factorials, 8 axials and 6 center points).

2.4. Data analysis

Experimental data from the CCD was analyzed by ANOVA test and employed by different model such as linear, interaction and quadratic models.(Eq. 2) [38] The statistical significance of model was checked using lack of fit, coefficient of determination (R^2), F-value and P-value at 95% confidence level [26,39].

$$Y = \underbrace{b_0}_{\text{constant}} + \underbrace{\sum_{i=1}^{k} b_i X_i}_{\text{single terms}} + \underbrace{\sum_{i=1}^{k} b_{ii} X_i^2}_{\text{square terms}} + \underbrace{\sum_{i=1}^{k-1} \sum_{j=i+1}^{k} \beta b_i X_i X_j}_{\text{interaction terms}} + \underbrace{\varepsilon}_{\text{residual}} \quad (2)$$

- Y is the predicted response of CFX removal
- X_{ij} represents the variable of the coded factor j at the ith run
- b_0 denotes the model intercept
- bi, bii, and bij refer to the coefficients for linear, quadratic and interaction terms, respectively
- ε is the error in experimental residue
- k offset term

3. RESULTS AND DISCUSSION

3.1. Model fitting and validation

Coefficient of variation (CV) was obtained 2.81% for the 30 experiments by ANOVA test, within the acceptable range of 10% (Table 2 and 3). The fitting model with the Design-Expert software suggested an interaction model based on Eq. 3.

$$Y \text{ (CFX reduction efficiency (\%))} = 64.19 - 0.3\, x_1 - 4.34\, x_2 + 0.92\, x_3 + 10.39\, x_4 - 0.67\, x_1x_2 + 0.69\, x_1x_3 + 1.47\, x_1x_4 - 0.44\, x_2x_3 - 1.88\, x_2x_4 + 0.87\, x_3x_4 \quad (3)$$

Eq. 3 represents the efficiency of four dependent parameters such as pH (x_1), initial concentration of CFX (x_2), current intensity (x_3) and reaction time (x_4) on CFX removal with four linear and six interactions effects. According to this equation, the reaction time and initial

concentration of CFX, with coefficients of 10.39 and 4.34, have the most positive and negative effects on the removal of CFX (%), respectively.

Table 2 shows the actual and predicted values of the model for CFX removal efficiency. Based on the results of the model, the correlation coefficient (R^2), adjusted correlation coefficient (R^2 adjusted), and predicted correlation coefficient (R^2 predicted) are 0.98, 0.97 and 0.95, respectively.

Table 2. Actual and predictive responses of CFX removal

Run number	Coded values				Actual values				Response CFX Removal (%)	
	X_1	X_2	X_3	X_4	A(pH)	B(mg.l^{-1})	C(A)	D(min)	Actual	Predicted
1	+1	+1	-1	-1	7.5	15.25	0.4	27.5	46.73	48.82
2	0	0	0	0	6	10.5	0.5	50	62.76	64.09
3	+1	+1	+1	-1	7.5	15.25	0.6	27.5	48.11	50.34
4	0	0	0	-2	6	10.5	0.5	5	47.53	43.47
5	-1	-1	+1	-1	4.5	5.75	0.6	27.5	57.67	58.73
6	0	0	0	0	6	10.5	0.5	50	62.02	64.09
7	-1	+1	-1	+1	4.5	15.25	0.4	72.5	68.03	67.25
8	-2	0	0	0	3	10.5	0.5	50	63.52	65.15
9	0	0	0	0	6	10.5	0.5	50	64.42	64.09
10	+1	-1	-1	+1	7.5	5.75	0.4	72.5	79.54	80.03
11	+1	+1	-1	+1	7.5	15.25	0.4	72.5	66.31	68.42
12	+1	-1	+1	-1	7.5	5.75	0.6	27.5	54.71	55.43
13	+2	0	0	0	9	10.5	0.5	50	65.72	63.02
14	-1	+1	-1	-1	4.5	15.25	0.4	27.5	54.12	52.12
15	+1	-1	-1	-1	7.5	5.75	0.4	27.5	53.24	53.92
16	0	-2	0	0	6	1	0.5	50	74.51	72.44
17	-1	+1	+1	-1	4.5	15.25	0.6	27.5	52.03	53.64
18	0	0	0	+1	6	10.5	0.5	72.5	63.13	64.09
19	0	0	+2	0	6	10.5	0.7	50	65.35	65.60
20	+1	+1	+1	+1	7.5	15.25	0.6	72.5	71.11	69.94
21	-1	-1	-1	-1	4.5	5.75	0.4	27.5	56.51	57.22
22	-1	-1	+1	+1	7.5	5.75	0.6	72.5	86.52	85.78
23	0	0	-2	0	6	10.5	0.3	50	63.13	62.57
24	+2	+2	0	0	6	20	0.5	50	58.57	55.74
25	-1	-1	-1	+1	4.5	5.75	0.4	72.5	78.52	78.86
26	0	0	0	0	6	10.5	0.5	50	64.42	64.09
27	0	0	0	0	6	10.5	0.5	50	63.31	64.09
28	0	0	0	+2	6	10.5	0.5	95	83.5	84.71
29	-1	-1	+1	+1	4.5	5.75	0.6	72.5	81.51	80.52
30	+1	+1	+1	+1	4.5	15.25	0.6	72.5	69.26	68.76

The closeness of the values of these coefficients reflects the high accordance of the predicted with real values which shows that the model has a high confidence level for

predicting values outside of the design space. In addition to, value of "adequate precision" (AP) that shows the signal-to-noise ratio, was equal to 38.6 that was greater than 4.

Other ANOVA results that can confirm the reliability and adequacy of the predicted model are values of F-value, P-value and lack of fit, which was equal to 98.02, 0.0001 and 0.0509 respectively. The F-value measures the data changes in the predicted model. Given that the value obtained is 0.92 in the model, it is only 0.01% that part of the data variables is not considered by the model. The value less than 0.05 and greater than 0.05 for P-value and lack of fit also indicates that the model is statistically significant (Table 3). According to Table 3, except for interactions of CD, BC, AC, AB, A, all the effects of factors and their interactions found to be significant in the CFX removal by EC process.

Table 3. ANOVA results for the response surface interaction model for CFX removal

Source	Analysis of variance				
	d.f.[a]	Sum of squares	Mean square	F-value	p-value
Model	10	3188.12	318.81	98.02	<0.0001 (S)
A-pH	1	2.16	2.16	0.66	<0.4252
B-Con	1	452.40	452.40	139.09	<0.0001
C-Cd	1	20.46	20.46	6.29	<0.0214
D-Time	1	2592	2592	796.93	<0.0001
AB	1	7.24	7.24	2.22	0.1522
AC	1	7.73	7.73	2.38	0.1397
AD	1	34.46	34.46	10.59	0.0042
BC	1	3.13	3.13	0.96	<0.3387
BD	1	56.40	56.40	17.34	0.0005
CD	1	12.04	12.04	3.70	<0.0695
Residual	19	61.81	3.25	-	-
Lack of fit	14	57.34	4.10	4.60	0.0509 (NS)
Pure error	4.46	5	0.89	-	-

R^2=0.981, R^2 adjusted = 0.971, R^2 predicted=0.953, AP=38.61, CV=2.81
[a]Degree of freedom.
NS=not significant p≤0.05
S=Significant at p≤0.05

The observation of the scanning electron microscopy (SEM) of the CFX surface and settled sludge after the electrocagulation process showed that the CFX particles (A) are needle-shaped crystals and rough surfaces. The sludge after the process (B) also showed the aluminum flocs with CFX particles (Fig. 4).

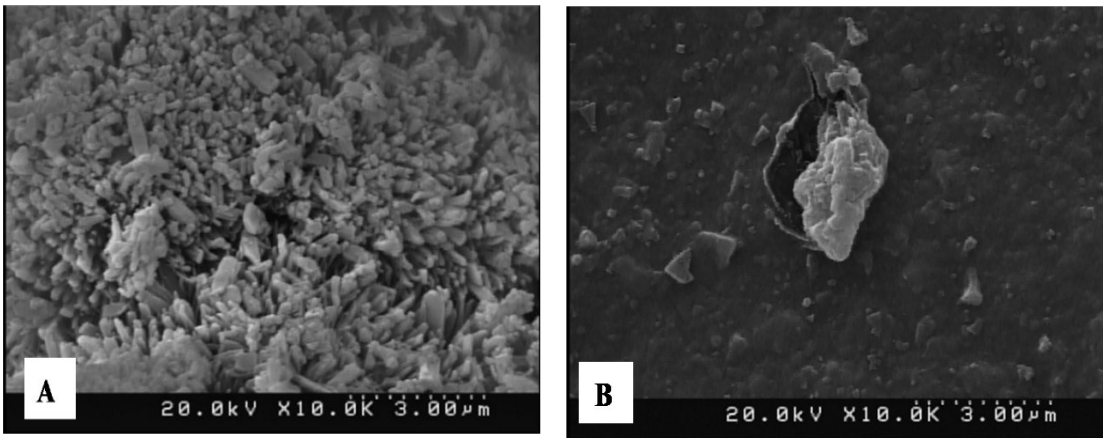


Fig. 4. SEM images, A: CFX powder; B: sludge of the process in an optimal condition

3.2. Effect of dependent parameters on CFX removal efficiency (%)

In order to compare the effect of dependent parameters on a specific point of the design space, a perturbation plot was used (Fig. 3). In this chart, the range of four parameters is proportional to the values listed in Table 1, coded by codes 1, 0.5, 0, -0.5 and -1. Code 0 is the midpoint in which the values of the variables are kept constant for comparison. The slope of each line in the perturbation plot indicates the sensitivity of the response to that parameter.

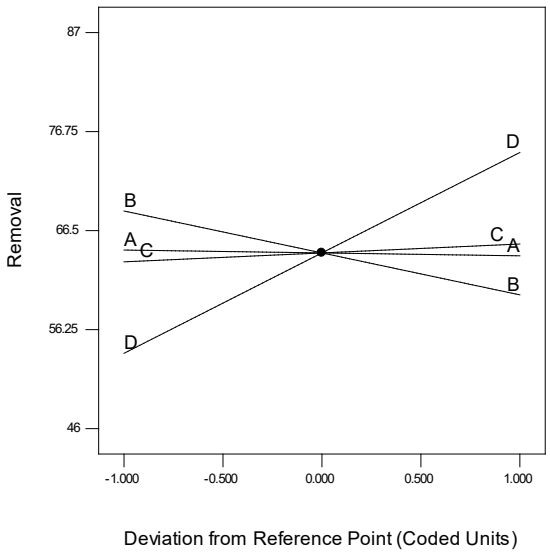
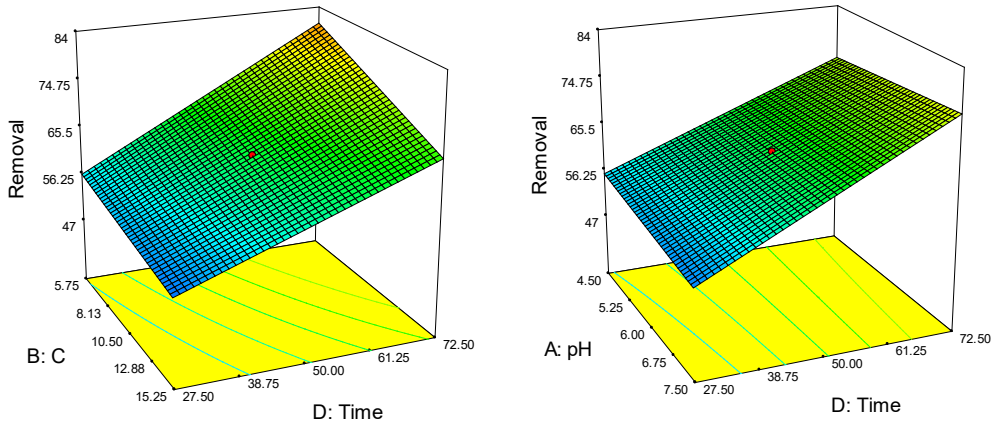


Fig. 5. Perturbation plots for CFX reduction efficiency (A) pH, (B) Initial concentration and (C) current intensity, (D) Reaction time

According to equation 3, the variables D (reaction time) and B (initial concentration CFX) have the greatest effect on the removal of the CFX. However, the pH and current intensity have

a negative and positive slope, respectively, which means that they have less effect on CFX removal efficiency than the other two parameters. Contrary to the fact that the perturbation plot compares the effect of parameters with each other, it cannot show the interaction of several parameters together.

Fig. 6 shows the interaction between parameters on CFX removal efficiency as 3D contours. As shown in Fig. 6a, reducing the initial concentration of CFX from 15.25 to 5.75 mg/L, the CFX removal efficiency increased from 51.33 to 56.25%. By increasing the reaction time from 27.57 to 72.5, the removal efficiency increase from 51.33 up to 65.5%. The effect of decreasing the initial concentration and increasing the reaction time alone increased the efficiency of CFX removal by less than 15%, while, in accordance with Fig 5a, the interaction of two parameters can increase the removal efficiency from 51.33% to 84%.



(a) **(b)**

Fig. 6. Response surfaces plots for CFX removal as a function of (a) initial concentration of CFX and time; (b) pH and time

Fig. 6(b) also shows the interaction between pH and reaction time. In this chart, increasing the reaction time from 27.5 to 72.5 min, and pH from 4.5 to 7.5, the CFX removal efficiency increased from 47 to 74.75 percent.

PH has always been the most effective parameters in the electrocoagulation process[29]. The pH between 6 and 8 is considered as the application range[34]. In the present study, the pH range was studied between 3-9, with the highest removal efficiency at pH 7.7. At pH 7.5, regarding the surface charge of CFX ions depends on the pH value and the amount of CFX (pK_a is 3.73 and 2.1 because of two carboxyl groups), the mainly CFX is in the form of negative ions [40]. On the other hand, at pH between 4-10, the Al^{3+} and OH^- ions produced by the electrodes are in the form of a variety of different monomeric aluminum with a wide positive charge. Additionally, to pH, increasing the current intensity also produces more Al^{3+} and OH^- ions. Thus, the CFX anions are absorbed by aluminum monomers and eventually removed from the solution as settling forms[41]. By increasing the initial concentration of CFX, the active

positions of the saturated aluminum and then the removal efficiency are reduced. The results of our study is similar to Yoosefian's et al study[26].

The reaction time is another important parameter of this process. The effect of this parameter on interaction with pH and initial concentration of CFX can provide enough time to produce more Al^{3+} and OH^- ions, and thus aluminum monomers. It is worth noting that a significant increase in reaction time can increase the amount of aluminum cationic ions that results in the repulsion process and then the reduction of efficiency. In the present study, with an increase in reaction time from 2 to 75 min, the removal efficiency increased by more than 86%. After that, increasing the reaction time to 95 min reduced the CFX removal efficiency.

4. CONCLUSION

The results of this study showed that the optimum conditions for the electrocoagulation process with the design of the experiment is pH=7.5, a primary concentration of CFX=5.75 mg/L, a current intensity=6.0 amperes and a reaction time=72.5 min (CFX removal=85.6%) from the aquatic solutions. The CFX removal experiments were performed by the same authors using the electrocoagulation process without the design of the experiment. The results of the two studies are in agreement with each other[42]. Among the advantages of the design of experiment, it is possible to optimize and investigate the interactive effects of independent factors in the process and reduce the number of experiments in chemical and physical processes. Due to the different physico-chemical characteristics of different wastewaters, the design of the experiment provides a model with the possibility of predicting the efficiency of the removal and improvement of the conditions of operation. So far, electrocoagulation has a High performance on the removal of antibiotics, however, in the physico-chemical processes, several factors can affect the process. In the present study, the effect of four main variables was investigated. Therefore, it is suggested that the efficiency of this process be investigated on different wastewaters with different interventions. Although EC process was successfully applied to remove CFX from water, the concentration range employed in this study was high and its degradation should be studied in much lower concentrations.

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