

## Electrochemical Oxidation Approach towards the Treatment of Acetamiprid Pesticide from Polluted Water

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### ABSTRACT

**Introduction:** Acetamiprid (AP) is one of the most widely used pesticides in the neonicotinoid class. AP residues in the environment have received considerable due to their potential toxicity to humans. Therefore, it is important to remove AP from the aqueous solution.

**Materials and Methods:** In the current study, response surface methodology (RSM) was used as an efficient approach to optimize the removal of AP using the electro-Fenton (EF) process. The effects of the main variables, including reaction time, AP concentration, current density, and H<sub>2</sub>O<sub>2</sub> dosage were investigated and optimized. ANOVA technique was also used to identify the Fisher's value (F-value) and P-value of the model.

**Results:** The predicted AP removal efficiency by the model was in good agreement with the obtained experimental results with correlation regression of 0.98. The ANOVA test proved that the developed quadratic model was significant with very low P-values less than 0.05, the high F-value of 240.1, and regression coefficients close to 1 at a 95% confidence level. The optimum condition for AP removal efficiency of 99.02% was attained at the reaction time of 12 min, AP concentration of 3.5 mg L<sup>-1</sup>, the current density of 12 mA cm<sup>-2</sup>, and H<sub>2</sub>O<sub>2</sub> dosage of 86 μL.

**Conclusion:** RSM was employed as a suitable method to optimize the operating condition and maximize the AP removal. Herein, the EF process as an eco-friendly electrochemical advanced oxidation process (EAOP) successfully applied to remove AP from the water and wastewater.

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### Introduction

The lives of all biota, including humans, animals, and plants, are affected by the quantity and quality of water. Also, health, agricultural, industrial, and welfare activities are affected by water resources.

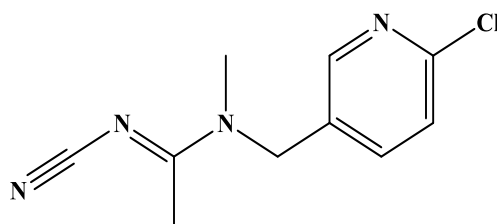
Due to the importance of water in the life of organisms, in recent decades, emerging pollutants

(Eps) (even in low concentrations) have been detected in water resources, which have caused a decrease in water quality and subsequent occurrence of diseases and environmental hazards<sup>1-3</sup>. Eps include surfactants, personal care products, pharmaceutical compounds, and pesticides<sup>4-6</sup>.

The presence of Eps in water resources has caused many concerns in human societies. Today, due to increasing population growth, the need for food has increased. The use of pesticides in agricultural activities to increase crop production and pest control has found special and inevitable applications. Researchers have frequently detected the residues of various pesticides in surface water, groundwater, soil, and sediments. Pesticide residues in the environment pose a

serious threat to the living biota, since they have the potential for bio-accumulation, bio-magnification, and remain in the environment for a long time without decomposition<sup>7-9</sup>.

Acetamiprid (AP) is one of the most widely used insecticides and belongs to neonicotinoid pesticides (Figure 1). In particular, AP has attracted the attention of many farmers due to its affordable price, availability, and ability to deal with a wide range of plant pests<sup>10-12</sup>.



**Figure 1:** Structure of AP as a neonicotinoid pesticide

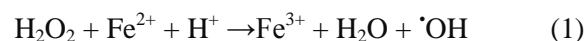
The presence of AP in the environment causes health problems, such as tremors, nausea, headaches, decreased or lost memory, weakened nervous system, and endocrine disruptors. In addition, studies have shown that AP presumably adversely affects the beneficial biota, including earthworms and bees<sup>10</sup>.

Therefore, its removal from water sources is of utmost importance. One of the most common environmental bottlenecks is focusing on designing efficient processes. Among the recent technologies, the electrochemical advanced oxidation processes (EAOPs) have proved to be a powerful oxidative approach for the degradation and removal of several organic pollutants. The electro-Fenton (EF) process is one of the most widely used EAOPs, which has shown good performance for treating resistant and refractory pollutants<sup>3</sup>. The basis of the EF process is the generation of hydroxyl radicals ( $\cdot\text{OH}$ ) through electrochemical reactions<sup>13-15</sup>.

In EF,  $\text{H}_2\text{O}_2$  (hydrogen peroxide) and  $\text{Fe}^{2+}$  ion promote a tremendous and fast generation of  $\cdot\text{OH}$  according to Fenton's reaction based on Eq. 1.

The  $\cdot\text{OH}$  species generated during the EF process attack the organic pollutants molecules,

transforming them into more safe products, such as water and carbon dioxide. In this process, the  $\text{Fe}^{2+}$  ions are regenerated from  $\text{Fe}^{3+}$  owing to their cathode reduction, according to Eq. 2. This confirms the generation of  $\cdot\text{OH}$  if the  $\text{H}_2\text{O}_2$  is available<sup>16-18</sup>.



In the present study, response surface methodology (RSM) was used as an efficient approach to optimize the removal of AP using the EF process. The important operational parameters, including reaction time, AP concentration, current density, and  $\text{H}_2\text{O}_2$  dosage, were considered. Finally, first-order and second-order kinetic models were investigated.

## Materials and Methods

### Chemicals

Analytical grade AP ( $\text{C}_{10}\text{H}_{11}\text{ClN}_4$ ) was obtained from Sigma-Aldrich Company. Acetonitrile ( $\text{CH}_3\text{CN}$ ) HPLC grade, sodium sulfate ( $\text{Na}_2\text{SO}_4$ ), sulfuric acid ( $\text{H}_2\text{SO}_4$ ), sodium hydroxide ( $\text{NaOH}$ ), and hydrogen peroxide ( $\text{H}_2\text{O}_2$ , 30% w/w) were purchased from Merck Company.

**EF reactor setup**

A cylindrical glass cell was used as the reactor. Two iron plates as anode and cathode electrodes with the same dimensions (4.0 × 0.8 × 0.1 cm) were placed parallel and at a constant distance (3.0 cm) from each other in the electrochemical reactor. The sample volume of 0.25 L with 0.05 M of Na<sub>2</sub>SO<sub>4</sub> as supporting electrolyte and pH solution of 5.0 was constant in all the experiments. The pH of samples was adjustment adding NaOH or H<sub>2</sub>SO<sub>4</sub> solutions. The current density was regulated using a DC power supply during the EF process. After

the electrochemical treatment process, the AP concentration was measured using the KNAUER Smartline HPLC system (C<sub>18</sub>-250 × 4.6, 0.5 mm). The wavelength was set at 242 nm. The mobile phases were acetonitrile (MeCN) and water (H<sub>2</sub>O) in the ratio (30:70 v/v).

**Experimental design**

Central composite design (CCD) was applied to optimize variables, such as AP concentration, current density, and H<sub>2</sub>O<sub>2</sub> dosage. The levels of independent variables for the AP removal using the EF process are summarized in Table 1.

**Table 1:** Level of independent variables for the AP removal using EF process

Variables (X <sub>i</sub> )	Level				
	-α	-1	0	+1	+α
X <sub>1</sub> = AP concentration (mg L <sup>-1</sup> )	1	2.5	5.5	8.5	10
X <sub>2</sub> = Current density (mA cm <sup>-2</sup> )	6	7.5	10	13.5	15
X <sub>3</sub> = H <sub>2</sub> O <sub>2</sub> dosage (μL)	20	34	60	87	100

The following equation was applied to evaluate and investigate the experimental results of CCD and to model the AP removal process using the EF process as follows<sup>19</sup>:

$$Y = \beta_0 + \sum_{i=1}^n \beta_i X_i + \sum_{i=1}^n \beta_{ii} X_i^2 + \sum_{i=1}^{n-1} \sum_{j=i+1}^n \beta_{ij} X_i X_j + \varepsilon \quad (3)$$

Where, Y (the removal percentage of AP using the EF process as a response), β<sub>0</sub> (intercept of the developed model), β<sub>i</sub> (linearity effects), β<sub>ij</sub> (interaction effects), β<sub>ii</sub> (quadratics effects), and ε denotes the error. The ANOVA was conducted at a (95% confidence level) for validating the model and significance of investigated variables using statistical parameters, such as the sum of squares, mean square, degree freedom (df), probability level (P-value), Fisher's test (F-value), determination coefficient (R<sup>2</sup>), and lack of fit<sup>20-22</sup>.

**Ethical issue**

The current study was conducted in the spring and summer of 2020, after receiving approval from

the ethics committee of Kerman University of Medical Sciences [IR.KMU.REC.1398.674].

**Results****Effect of reaction time on the AP removal efficiency**

The effect of reaction time was investigated from 2 to 15 min on AP removal. The AP concentration, pH solution, current density, and H<sub>2</sub>O<sub>2</sub> dosage were kept constant at 4.0 mg L<sup>-1</sup>, 5.0, 9 mA cm<sup>-2</sup>, and 50 μL, respectively, the results of which are shown in Figure 2. After 10 min of EF process, the AP removal efficiency significantly increased, but after 12 min, the AP removal efficiency reached the constant value of about 83%. Thus, the optimum reaction time was determined to be 12 min.

**RSM model fitting**

The designed matrix and experimental result of AP removal using the EF process are shown in Table 2.

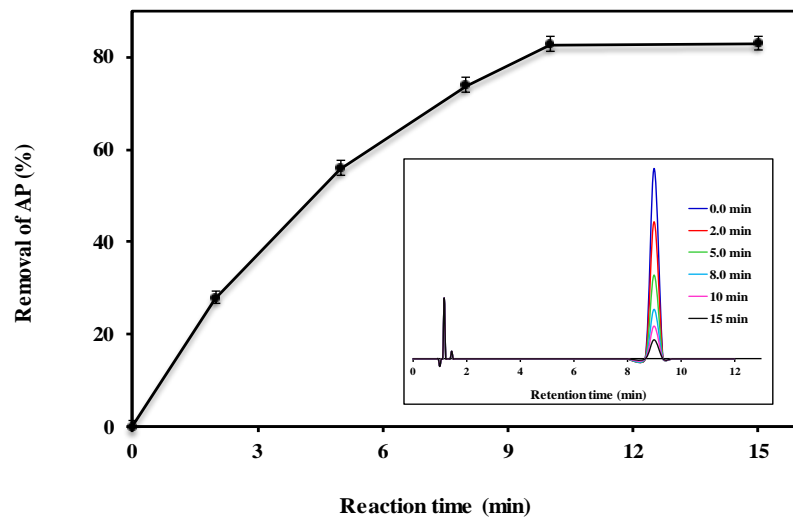


Figure 2: The effect of reaction time on the AP removal efficiency

Table 2: Designed matrix and experimental result of AP removal using EF process

Run	Actual value			Coded value			AP removal (%)
	X <sub>1</sub>	X <sub>2</sub>	X <sub>3</sub>	X <sub>1</sub>	X <sub>2</sub>	X <sub>3</sub>	
1	8.50	7.50	86	1	-1	1	75.9
2	2.50	13.50	86	-1	1	1	98.1
3	5.50	6.00	60	0	-1.5	0	70.5
4	1.00	10.50	60	-1.5	0	0	94.4
5	8.50	13.50	86	1	1	1	91.2
6	10.0	10.50	60	1.5	0	0	78.3
7	5.50	15.00	60	0	1.5	0	85.6
8	5.50	10.50	60	0	0	0	86.8
9	2.50	7.50	86	-1	-1	1	84.8
10	5.50	10.50	60	0	0	0	86.1
11	8.50	13.50	33	1	1	-1	76.7
12	5.50	10.50	100	0	0	1.5	93.1
13	5.50	10.50	60	0	0	0	85.8
14	5.50	10.50	60	0	0	0	85.6
15	5.50	10.50	20	0	0	-1.5	80.1
16	8.50	7.50	33	1	-1	-1	72.6
17	5.50	10.50	60	0	0	0	85.3
18	5.50	10.50	60	0	0	0	85.4
19	2.50	13.50	33	-1	1	-1	84.9
20	2.50	7.50	33	-1	-1	-1	81.3

The relevance between the AP removal efficiency as the response and the significant variables demonstrated by a quadratic model is as follows:

$$\text{Removal AP}(\%) = 86.21 - 4.55X_1 + 4.72X_2 + 4.32X_3 + 2.61X_2X_3 - 3.38X_2^2 \quad (4)$$

In this equation, Y is the AP removal (%), and X<sub>1</sub> to X<sub>3</sub> denote the coded independent factors of AP concentration, the current density, and H<sub>2</sub>O<sub>2</sub> dosage, respectively. The adequacy of the

proposed model was investigated using the ANOVA test and summarized in Table 3. The correlation coefficient between the predicted and actual values of AP removal efficiency was computed to be 0.98, indicating that the developed model could not describe only 1.15% of the total variance in the response. Moreover, the observed variation of less than 0.20 between the adjusted R<sup>2</sup> (Adj. R<sup>2</sup> = 0.98) and the predicted R<sup>2</sup> (Pred. R<sup>2</sup> = 0.97) confirmed the significance of the model.

Table 3: ANOVA results of the developed model

Source	Sum of squares	df	Mean square	F-value	p-value
Model	942.13	5	188.43	240.10	< 0.0001
X <sub>1</sub>	258.55	1	258.55	329.46	< 0.0001
X <sub>2</sub>	278.01	1	278.01	354.25	< 0.0001
X <sub>3</sub>	233.28	1	233.28	297.26	< 0.0001
X <sub>2</sub> X <sub>3</sub>	54.60	1	54.60	69.58	< 0.0001
X <sub>2</sub> <sup>2</sup>	117.69	1	117.69	149.96	< 0.0001
Residual	10.99	14	0.78	-	-
Lack of Fit	9.45	9	1.05	3.43	0.0943
Pure Error	1.53	5	0.31	-	-
Cor Total	953.12	19	-	-	-
R <sup>2</sup> = 0.9885		Adjusted R <sup>2</sup> (Adj. R <sup>2</sup> ) = 0.98		Predicted R <sup>2</sup> (Pred. R <sup>2</sup> ) = 0.97	

In Figure 3-a, the obtained experimental values for the removal efficiency of AP were compared to the predicted values by the model. Also, the data were investigated to assess the normality of the residuals. The normal probability

plots of the internally studentized residuals are presented in Figure 3-b. The adequate agreement between the actual (experimental values) and the predicted values confirmed the normality of the results.

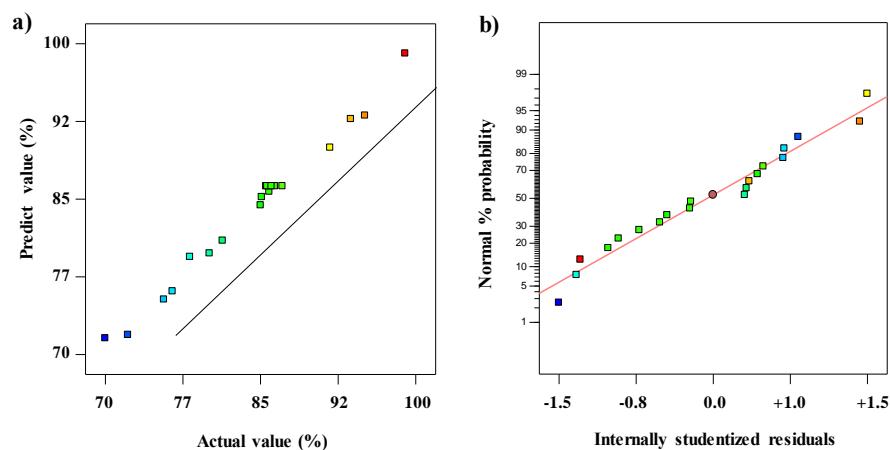
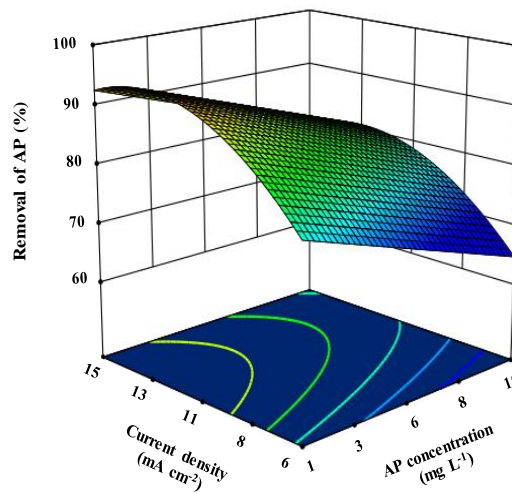


Figure 3: Statistical analysis of the AP removal efficiency model (a) the actual vs. predicted value, (b) normal% probability vs. internally studentized residual

### Effect of significant parameters on the removal efficiency of AP

Figure 4 shows a 3D response surface plot of removal as a function of AP concentration and the current density at the constant values of H<sub>2</sub>O<sub>2</sub>

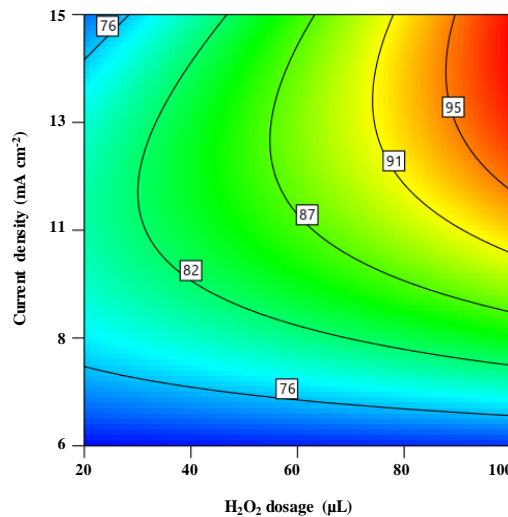
dosage of 60 μL, pH solution of 5.0, and reaction time of 12 min. The result shows that the AP removal decreased from 92.7% to 79.4% by increasing the AP concentration from 1 mg L<sup>-1</sup> to 10 mg L<sup>-1</sup>.



**Figure 4:** 3D plot of the current density vs. AP concentration ( $\text{H}_2\text{O}_2$  dosage of  $60 \mu\text{L}$ , pH solution of 5.0, and reaction time of 12 min)

The removal efficiency of AP increased by increasing the current density. In contrast, the current density was raised from  $6$  to  $12 \text{ mA cm}^{-2}$ , the removal efficiency rose from  $71.2\%$  to  $88.6\%$ . However, by increasing the current density from  $12$  to  $16$ , the removal efficiency decreased from  $88.6\%$  to  $85.4\%$ .

$\text{H}_2\text{O}_2$  dosage played a critical role in the AP removal efficiency throughout the EF process. Usually, the removal efficiency of pollutants increases by increasing  $\text{H}_2\text{O}_2$  concentration. Herein, the removal efficiency increased from  $79.2$  to  $92.8\%$  by increasing the  $\text{H}_2\text{O}_2$  dosage from  $20$  to  $100 \mu\text{L}$  (Figure 5).



**Figure 5:** Contour diagram of the current density vs.  $\text{H}_2\text{O}_2$  dosage (AP concentration of  $5.5 \text{ mg L}^{-1}$ , pH solution of 5.0, and reaction time of 12 min)

### Kinetics study

To find out the exact mechanism of the AP removal, kinetics investigations were carried out using first and second-order models. The obtained

experimental results are displayed in Table 4, which indicates that the first-order model with the satisfactory correlation coefficient of  $0.99$  best fitted to the achieved results<sup>23-26</sup>.

Table 4: Parameters of kinetic models for the AP removal

Kinetics model	Equation	$k_{app}$	$R^2$
First-order	$\ln\left(\frac{C_0}{C_t}\right) = +kt$	$0.174 \text{ min}^{-1}$	0.99
Second-order	$\frac{1}{C_t} = kt + \left(\frac{1}{C_0}\right)$	$0.0925 \text{ L.mg}^{-1}\text{min}^{-1}$	0.91

## Discussion

### Statistical analysis

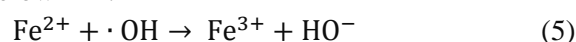
The results of ANOVA are presented in Table 3. The model was significant and can be applied successfully for estimating the AP removal efficiency using the EF process. The regression coefficient ( $R^2 = 0.98$ ) agrees well with the adjusted regression coefficient ( $\text{Adj.}R^2 = 0.98$ ). The Adeq precision describes the signal-to-level noise ratio, where the ratio of 56.66 is greater than 4, indicating the adequacy of the developed model<sup>27, 28</sup>. In Figure 3, the appropriate and close distribution of data points around a straight line proved the potential and ability of the model to suitably predict values and normally distribute experimental data.

### Effect of significant parameters

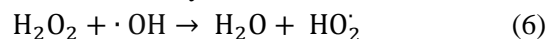
The influence of AP concentration in the range of 2 to 10 mg L<sup>-1</sup> was investigated. The obtained results showed that the AP removal efficiency decreased by increasing the AP concentration, which can be ascribed to the limited number of  $\cdot\text{OH}$  radicals compared to the increasing number of AP molecules<sup>29-31</sup>.

The results of the investigation of current density showed that increasing the current density causes an improvement in the AP removal efficiency. By increasing the current density, the amount of anodic dissolution of iron increased, resulting in a much great generation of  $\text{Fe}^{2+}$ . The increasing concentration of  $\text{Fe}^{2+}$  iron could enhance the Fenton reaction, which produced the  $\cdot\text{OH}$  radical as a favorable driving force of the treatment process. Moreover, by increasing the current density, the level of  $\text{H}_2$  bubble generation increased, and the bubble size decreased, which both benefited the treatment process through the flotation mechanism. However, beyond the

optimal concentration of  $\text{Fe}^{2+}$ , the slight reduction in the removal efficiency was observed, which is probably due to the following reaction. In the high concentration of iron cations, the  $\cdot\text{OH}$  radicals are trapped by  $\text{Fe}^{2+}$  as described below<sup>32-34</sup>.



To obtain the maximum AP removal efficiency, the optimal dosage of  $\text{H}_2\text{O}_2$  should be employed. Although increasing the dosage of  $\text{H}_2\text{O}_2$  enhanced the AP removal efficiency due to accelerating the generation of  $\cdot\text{OH}$  (see reaction 1), it cannot be added without any limitation. The excessive  $\text{H}_2\text{O}_2$  not only increases the costs of operation, but also increases the scavenging effect of  $\cdot\text{OH}$  by  $\text{H}_2\text{O}_2$  (reaction 4), which has a negative effect on the removal efficiency of AP<sup>35-37</sup>.



Racar et al.,<sup>38</sup> studied the removal of various CECs, including AP, from municipal wastewater (MWW) using membrane bioreactor (MBR). In a mixture of CECs and at a concentration of  $2.32 \pm 3.22 \mu\text{g L}^{-1}$  of AP in the influent, a removal percentage of 39.36% was obtained for MBR<sup>38</sup>. Many CECs are effectively degraded using UV radiation and ozonation techniques. However, the irradiation experiments using marketed mixtures containing AP resulted in a variety of photoproducts, which were also toxic for vertebrates and non-target species<sup>9</sup>.

Cruz-Alcalde et al.,<sup>10</sup> reported that the toxicity of transformation products (TPs) in the solution increased with increasing the ozone dose and then decreased. Similar toxic TPs were observed during photo-Fenton treatment of AP in race-way ponds<sup>12</sup>. Yao et al.,<sup>39</sup> fabricated ytterbium (Yb) doped- $\text{PbO}_2$  electrodes for electrochemical oxidation of AP. All the degradation products were decontaminated into  $\text{CO}_2$  and  $\text{H}_2\text{O}$ .

### Process optimization

In optimizing the AP removal process by design expert software, the desired ranges for each factor and response were selected as the final goal. The numerical optimization provided an opportunity to find a treatment condition with the maximum desirability function. Herein, the main goal was to maximize the removal of AP by the EF process. The optimum AP removal efficiency of 99.02% was attained at the 3.5 mg L<sup>-1</sup> AP concentration, the 12 mA cm<sup>-2</sup> current density, and 86 µL H<sub>2</sub>O<sub>2</sub> dosage.

### Conclusion

In the current study, the RSM was used to assay the effects of important parameters of the AP removal using the EF process using iron electrodes. The maximum removal efficiency of AP (99.02%) was achieved in the condition of 3.5 mg L<sup>-1</sup> AP concentration, 12 min reaction time, 86 µL H<sub>2</sub>O<sub>2</sub> dosage, and 12 mA cm<sup>-2</sup> current density. According to the developed model, the current density was identified as the most effective parameter in the removal AP using the EF process.

The first-order model well fitted the kinetics data obtained from the batch experiments.

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### Conflict of interest

The authors declare that they have no conflict of interest regarding the publication of the current paper.

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