

Oxidative Degradation of Orange II Solution using CaMo_{0.67}Cu_{0.33}O₃ Perovskite: Parametric Optimization by Response Surface Methodology

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ARTICLE HISTORY

ABSTRACT

This study investigates the optimal operational parameters for oxidative Received degradation of orange II (OII) in heterogeneous chemical oxidation using 30 June 2022 CaMo_{0.67}Cu_{0.33}O₃ perovskite catalyst. Response surface methodologycentral composite design (RSM-CCD) experiments were used to optimize Accepted four critical operational parameters: catalyst dosage, initial concentration 18 August 2022 of OII dye, pH of solution and concentration of H_2O_2 on the percentage of OII removal. Based on the analysis of variance (ANOVA), the model Available online illustrated a high correlation coefficient (R^2) of 0.9941, which suggested that 1 September 2022 the empirical quadratic model fitted well to the experimental data. The optimal operational parameters were found to be at 0.99 g/L of catalyst dosage, 20.80 mg/L of initial concentration of OII dye, 5.42 pH solution and 0.87 g/L of H_2O_2 concentration. Interestingly, the experimental OII removal was found to be approximately 91.19% at the suggested optimal conditions. This experimental OII removal value aligns with the predicted value derived from the proposed quadratic model with a difference of 3.98%. Such findings have shown significant global interaction between four critical operational parameters towards OII removal during the heterogeneous chemical oxidation using CaMo_{0.67}Cu_{0.33}O₃ perovskite catalyst.

Keywords: response surface methodology; heterogeneous chemical oxidation; perovskite catalyst; CaMo_{0.67}Cu_{0.33}O₃; orange II

1. INTRODUCTION

Textile dyeing industries are considered significant contributors to environmental water pollution due to a considerable amount of dye-contaminated wastewater effluents that have been directly discharged to water bodies. The estimation of azo dyes usage in textile dyeing industries was represented by more than 70% compared to other types of dye [1-3]. Acid orange II (OII) is known as one of the typical types of azo dyes that are frequently used in the textile dyeing process. OII contains azo bond (-N=N-) that is toxic and carcinogenic to aquatic life, which led to harmful impacts to other living organisms as well as humans [2-5].

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Therefore, dye-contaminated wastewater must be treated with appropriate wastewater treatment technologies before being discharged into natural water bodies. Advanced oxidation processes (AOPs) have been identified as promising wastewater treatment technology in the oxidation of a wide range of aqueous organic contaminants pollution [6-7]. Among the many technologies employed in AOPs, researchers have come to the attention of heterogeneous chemical oxidation due to its high efficacy in decolorization and degradation of dye-contaminated wastewater [8], [9]. The oxidation involves highly oxidizing radicals, namely hydroxyl (OH⁻) and sulfate (SO4⁻⁻) radicals [8], [10-11] during heterogeneous catalysis. The generation of these radicals required activation of oxidants (e.g., hydrogen peroxide, (H_2O_2) , persulfate and peroxymonosulfate (POMS)) in presence of heterogeneous catalysts.

Recently, perovskites are known as one of potential heterogeneous catalysts in AOPs due to their decent physicochemical properties such as good thermal stability, electron mobility and redox behavior [6], [12]. In general, perovskite is a type of oxide with a chemical formula $A_xB_yO_3$. It also can be in the form of binary or ternary metal structures such as $A_xA'_{1-x}B_yO_3$, $A_xB_yB'_{1-y}O_3$ or $A_xA'_{1-x}B_yB'_{1-y}O_3$. The A-site cation refers to rare, earth or lanthanide metal (i.e. Na, Ca and K), whilst the B-site cation refers to a transition metal (i.e. Cu, Ni and Co). Almost 90 % of the A-site and B-site elements are from the periodic table, forming perovskite structures [8], [13]. Perovskites have shown good catalytic performances in degradation of dye-contaminated wastewater [10]. For instance, Lin *et. al* [8] used LaCoO₃ perovskite catalyst in activating POMS to form SO₄⁻⁻ radicals. During oxidation, such efficient generation of SO₄⁻⁻ radicals exhibited approximately 90 % rhodamine B (RhB) decolorization within 60 minutes. Moreover, Wu *et. al* [11] reported 70.8 % AO7's degradation from the activation of POMS by LaFeO₃ perovskite catalyst.

The effectiveness of oxidative degradation for dye-contaminated wastewater was dependent on the operational reaction parameter involved during the reaction. The critical operational parameters are known to be catalyst dosage, initial concentration of OII dye, pH of solution, and concentration of oxidant (H_2O_2). However, the operational parameters' optimization using perovskite catalyst were performed using one-factor-at-time (OFAT) approach [1], [13-17]. This OFAT approach cannot directly correlate with the global interaction of other operational parameters due to time-consuming and requires too many experimental runs. Hence, response surface methodology (RSM) was introduced to overcome the drawbacks of OFAT.

Response surface methodology (RSM) is a collection of mathematical and statistical techniques used for process optimization whilst evaluating the significance of global interaction between parameters towards desired responses [18-20]. A standard RSM design called a central composite design (CCD) is suitable for fitting a quadratic surface, and it helps to optimize the effective parameters with a minimum number of experiments, as well as to analyze the interaction between the parameters [21-22]. Intriguingly, no work has been reported for detailed optimization of operational parameters in the oxidative degradation of dye-contaminated wastewater using perovskites with the aid of RSM. Such findings are vital as they influence the effective generation of radicals during heterogeneous catalysis leading to high oxidative

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degradation of organic pollutants. Hence, this work aims to determine the optimal operational parameters in the oxidative degradation of OII using CaMo_{0.67}Cu_{0.33}O₃ perovskite catalyst.

2. METHODOLOGY

2.1 Materials

All chemicals were used as received. Ethylenediamine tetra-acetic acid (EDTA, ≥ 99 %), ammonium hydroxide (NH₄OH, 25 %), ammonium molybdate ((NH₄)₂MoO₄, 99.98 %), and orange II sodium salt (C₁₆H₁₁N₂NaO₄S, ≥ 85 %) were purchased from Sigma-Aldrich. Calcium nitrate tetrahydrate (Ca(NO₃)₂.4H₂O, ≥ 99 %) and copper (II) nitrate trihydrate (Cu(NO₃)₂.2.5H₂O, ≥ 99 %) were purchased from Merck meanwhile citric acid monohydrate (C₆H₈O₇.H₂O, ≥ 99 %) from QRëc.

2.2 Synthesis of CaMo_{0.67}Cu_{0.33}O₃ Perovskite Catalyst

CaMo_{0.67}Cu_{0.33}O₃ perovskite catalysts were synthesized via a combined EDTA-citric acid complexation method [23-24]. A set of precursor molar ratios of total A metal ions: ethylenediamine tetra-acetic acid (EDTA): citric acid: ammonium hydroxide was kept constantly at 1:1.1:2:10. The precursor molar ratio of B:B' site metal ions was fixed at 1:0.5. Initially, 0.05 M of Ca(NO₃)₂.4H₂O, 0.05 M (NH₄)₂MoO₄, 0.25 M Cu(NO₃)₂.2.5H₂O and 0.1 M C₆H₈O₇.H₂O solutions were mixed and stirred for 15 min. After that, a mixture containing 0.05 M NH₄OH and 0.055 M EDTA solution was added into the former mixture and stirred for 15 minutes at ambient temperature until a homogeneous mixture was attained. The homogeneous mixture was then heated at 100 °C while magnetically stirred to evaporate most water until a viscous solution formed. The resultant gel sample was calcined at 2-stages calcination temperatures of (i) 450 °C (8 h) and (ii) 800 °C (4 h) in the air at heating and cooling rates of 5 °C min⁻¹. The resultant CaMo_{0.67}Cu_{0.33}O₃ perovskite samples were milled and stored prior to further use.

2.3 Operational Parameters' Optimization of Oxidative Degradation of OII Using Camo_{0.67}Cu_{0.33}O₃ Perovskite Catalyst

Response surface methodology-central composite design (RSM-CCD) (Design Expert software version 12, Stat-Ease Inc.) was chosen to initiate the optimization design for experimental runs. Four critical operational parameters were known as catalyst dosage (A), initial concentration of OII dye (B), pH of solution (C) and concentration of H_2O_2 (D). The response for this optimization process was the percentage of OII removal (Y). These four independent parameters were coded into five levels as tabulated in Table 1. Three types of experimental points are required in CCD, represented by 2^n factorial points, 2n axial points and 6 center points [25]. For four variables, 16 factorial points, 8 axial points and 6 replicates at the center points were employed, indicating a total of 30 experimental runs as shown in Equation 1 [26]:

$$N = 2^n + 2n + n_c = 2^4 + 2(4) + 6 = 30$$
⁽¹⁾

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where n is the number of operational parameter / variables, n_c is the center point's runs and N is the total number of experiments.

Variables (factors)	Unit	Code		Coded variables levels				
variables (lactors)	Unit		-α	-1	0	+1	-α	
Catalyst dosage	g/L	А	0.25	0.50	0.75	1.00	1.25	
Initial concentration OII dye	mg/L	В	10	20	30	40	50	
рН	-	С	3	4	5	6	7	
Concentration of H ₂ O ₂	g/L	D	0.40	0.60	0.80	1.00	1.20	

Table 1: Independent parameters and their coded levels for the central composite design (CCD)

The center points are used to determine the experimental error and the reproducibility of the data. The axial points are located at $(\pm \alpha, 0, 0)$, $(0, \pm \alpha, 0)$ and $(0, 0, \pm \alpha)$ where α is the distance of the axial point from center [26]. In this research work, the α -value was fixed at 2. The experimental sequence was randomized to minimize the effects of the uncontrolled factor. Each response was used to develop an empirical model which correlated with response to the four independent parameters using second-degree polynomial equation as shown in Equation 2 [22]:

$$Y = a_0 + \sum_{i=1}^k a_i x_i + \sum_{i=1}^k a_{ii} x_i^2 \sum_{i=1}^k \sum_{j=1}^k a_{ij} x_i x_j$$
(2)

where Y is the process response and a_0 is average of responses, k is number of patterns, i and j are the index numbers for the patterns. The x_i and x_j represent coded independent parameters, where i, ii and ij are linear, quadratic and interaction effects.

The oxidative degradation of OII was performed by dispersing CaMo_{0.67}Cu_{0.33}O₃ perovskite catalyst into 100 mL orange II (OII) solution at specific pH as presented in Table 2. The reaction was activated after adding a specific volume of hydrogen peroxide (H₂O₂) as an oxidant into the solution. The reaction mixture is stirred under a constant stirring rate of 200 rpm at room temperature. Then 5 mL aliquots were extracted and filtered using 0.2 µm syringe filters at 30 minutes of reaction. The concentration of filtrate OII samples was immediately analyzed using UV-Vis spectrophotometer (Perkin Elmer, Lambda 25) at $\lambda_{max} = 486$ nm. The percentage of OII removal (response) was calculated by following Equation 3:

OII removal percentage (%) =
$$[(C_0 - C_t)/C_0] \times 100$$
 (3)

where C_0 is the initial concentration of OII solution and C_t is the final concentration at 15 minutes reaction.

2.4 Model Fitting and Statistical Analysis

The experimental data were analyzed using a statistical software Design Expert software version 12 (Stat-Ease Inc., Minneapolis, USA) for regression analysis to fit the second-degree polynomial equation and evaluate the statistical significance of the suggested empirical model.

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	Coded variables level					Independer	OII removal, Y (%)			
Run	A	В	С	D	Catalyst dosage, A (g/L)	Initial concentration of OII dye, B (mg/L)	pH of solution, C	Concentration of H ₂ O ₂ , D (g/L)	Experimental	Predicted
1	-1	-1	-1	-1	0.5	20	4	0.6	40.89	40.83
2	+1	-1	-1	-1	1.0	20	4	0.6	71.58	73.90
3	-1	+1	-1	-1	0.5	40	4	0.6	37.76	34.54
4	+1	+1	-1	-1	1.0	40	4	0.6	62.51	60.97
5	-1	-1	+1	-1	0.5	20	6	0.6	54.45	55.23
6	+1	-1	+1	-1	1.0	20	6	0.6	89.37	86.21
7	-1	+1	+1	-1	0.5	40	6	0.6	38.05	37.89
8	+1	+1	+1	-1	1.0	40	6	0.6	59.10	62.18
9	-1	-1	-1	+1	0.5	20	4	1.0	47.63	46.35
10	+1	-1	-1	+1	1.0	20	4	1.0	81.70	80.10
11	-1	+1	-1	+1	0.5	40	4	1.0	44.04	45.45
12	+1	+1	-1	+1	1.0	40	4	1.0	71.58	72.56
13	-1	-1	+1	+1	0.5	20	6	1.0	56.19	55.98
14	+1	-1	+1	+1	1.0	20	6	1.0	91.70	87.59
15	-1	+1	+1	+1	0.5	40	6	1.0	44.50	43.98
16	+1	+1	+1	+1	1.0	40	6	1.0	70.64	68.95
17	-2	0	0	0	0.25	30	5	0.8	31.98	33.61
18	+2	0	0	0	1.25	30	5	0.8	93.33	91.65
19	0	-2	0	0	0.75	10	5	0.8	85.12	84.22
20	0	+2	0	0	0.75	50	5	0.8	58.43	59.29
21	0	0	-2	0	0.75	30	3	0.8	46.04	47.56
22	0	0	+2	0	0.75	30	7	0.8	59.97	58.40
23	0	0	0	-2	0.75	30	5	0.4	43.69	44.67
24	0	0	0	+2	0.75	30	5	1.2	57.99	56.96
25	0	0	0	0	0.75	30	5	0.8	75.73	73.00
26	0	0	0	0	0.75	30	5	0.8	71.26	73.00
27	0	0	0	0	0.75	30	5	0.8	73.84	73.00
28	0	0	0	0	0.75	30	5	0.8	70.58	73.00
29	0	0	0	0	0.75	30	5	0.8	74.46	73.00
30	0	0	0	0	0.75	30	5	0.8	72.11	73.00

Table 2: Experimental design matrix for independent parameters of OII removal

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3. RESULTS AND DISCUSSION

3.1 Development of regression model equation

In this research work, the global interactions between four independent parameters and the corresponding response of OII removal were well fitted with the quadratic model. The empirical quadratic model for OII removal obtained from RSM-CCD was presented in Equation 4:

Y = 73.0 + 14.89A - 6.61B + 3.09C + 3.45D - 2.23AB + 0.0356AC + 0.7406AD - 3.34BC + 0.7756BD - 0.6344CD - 2.50A² - 0.2159B² - 4.91C² - 5.45D²(4)

where A, B, C, D and Y is the coded factor for catalyst dosage, dye concentration, pH of solution, H_2O_2 concentration and percentage of OII removal, respectively.

Coefficient with one factor represents the effect of an individual operational parameter/factor. Meanwhile, the coefficients with two factors and those with second-order terms represent the global interaction between factors and their quadratic effect, correspondingly. The positive sign for each coded factor represents a synergistic effect, while the negative sign represents an adverse effect [21], [27] towards the response (percentage of OII removal). Therefore, from equation 4, it was found that the catalyst dosage (A), pH of the solution (C) and concentration of H₂O₂ (D) possessed a synergistic effect interaction in enhancing the OII removal. Interestingly, it was observed that OII removal increased with the increase of catalyst dosage in the presence of H_2O_2 over a wide pH range of the solution. This synergistic interaction was attributed to the increased number of available active sites responsible for the activation of H₂O₂ to hydroxyl radicals (•OH). Therefore, effective generation of •OH radicals resulted in rapid degradation of OII during catalysis. Similar findings on the effect of catalyst dosage, solution pH and concentration of H₂O₂ towards enhancement of dye-contaminated wastewater degradation were also reported elsewhere [1], [13]. On the contrary, the initial concentration of OII (B) showed an adverse effect on the response. The percentage of OII removal decreased when the initial concentration increased from 10 mg/L to 50 mg/L. The higher the initial concentration of AOII was, the more pronounced the inductive effect became. This was due to the fact that the available active sites on the surface of the catalyst were predominantly covered by a high concentration of AOII, which could hinder the free access of H_2O_2 towards the active sites. Consequently, the limitation of free accessibility of H_2O_2 reduced the effective generation of •OH radicals, which led to further decrease in AOII removal during catalysis. This finding is in concordance with Bresolin et. al [15] that observed the degradation of methylene blue decreasing as the initial concentration of methylene blue increases.

3.2 Analysis of Variance (ANOVA)

The statistical analysis of variance (ANOVA) for the empirical model of OII removal demonstrated that the model is statistically significant owing to the low probability value (p-value < 0.05) as well as the calculated Fisher variation reaction (F-value) as summarized in Table 3. The coded factors are considered significant when the p-value is less than 0.05 [21], [27]. The model F-value for OII removal in heterogeneous chemical oxidation corresponded to

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180.63 with a p-value <0.0001 that implied a significant empirical model with 95 % confidence level.

From ANOVA analysis, it was found that all the operational parameters (catalyst dosage (p-value < 0.0001), initial concentration OII dye, pH of solution and concentration of H_2O_2) played significant role during oxidation reaction towards the response of OII removal due to low p-value <0.0001. An insignificant lack of fit is highly anticipated during the model fitting. This is because a significant lack of fit (p-value > 0.05) reflects the regressor-response relationship's possibilities that suggested the empirical model's inadequacy to explain the experimental data variations [25]. The regressed model exhibited an insignificant lack of fit with a p-value of 0.6623, implying model adequacy and fitting well with experimental data. The coefficient of variance (CV) value of less than 15 % indicated a high degree of precision and acceptable variability in data analysis [25]. In this work, the model's CV value was 2.94 % which implied good reliability of experimental data.

Source	Sum of squares	dF	Mean square	F value	Prob > F	
Model	8545.34	14	610.38	180.63	< 0.0001	Significant
А	5321.39	1	5321.29	1574.75	< 0.0001	U
В	1049.54	1	1049.54	310.59	< 0.0001	
С	229.22	1	229.22	67.83	< 0.0001	
D	286.14	1	286.14	84.68	< 0.0001	
AB	79.70	1	79.70	23.59	0.0002	
AD	8.78	1	8.78	2.60	0.1279	
BC	178.96	1	178.96	52.96	< 0.0001	
BD	9.63	1	9.63	2.85	0.1121	
CD	6.44	1	6.44	1.91	0.1877	
A^2	170.87	1	170.87	50.57	< 0.0001	
\mathbf{B}^2	1.28	1	1.28	0.3785	0.5476	
C^2	660.83	1	660.83	195.56	< 0.0001	
D^2	814.60	1	814.60	241.06	< 0.0001	
Residual	50.69	15	3.38			
Lack of fit	30.72	10	3.07	0.7693	0.6623	Not significant
Pure error	19.97	5	3.99			-
Cor Total	8596.03	29				
\mathbb{R}^2	0.9941		Std. Dev.	1.84		
Adjusted R ²	0.9886		Mean	62.54		
Predicted R ²	0.9761		C.V. %	2.94		
Adeq Precision	45.9161					

Table 3: Analysis of variance (ANOVA) for the regression model of OII removal

Figure 1 shows the model response on the percentage of OII removal for predicted versus experimental values in each experimental run. A high data correlation was observed between the predicted and experimental values. Validation of empirical model adequacy can be well correlated with the acquired model's regression coefficient (R^2), adjusted R^2 and predicted R^2

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values, respectively. The empirical model exhibited a high R^2 of 0.9941, which indicated good estimation of predicted response.

The model also demonstrated approximately 99% of targeted function variation for the global parametric interactions. The adjusted R^2 and predicted R^2 are 0.9886 and 0.9761, respectively suggesting excellent correlations between the independent variables. Such findings concluded that the data were well-fitted to the empirical model, providing a good estimation of response convincingly.



Figure 1: Predicted versus actual percentage of OII removal

3.3 Global Operational Parameters Interactions

Among all operational parameters, catalyst dosage (A) shows the most significant variable due to the highest F-value (1574.75), followed by initial concentration OII dye (B), concentration of H_2O_2 (D) and pH of solution (C), as summarized in Table 3. The plot displayed in Figure 2 shows the relationship of all operational parameters towards OII removal. The OII removal increased up to 80% after 15 mins at a catalyst dosage range of 0.9 - 1.0 g/L, lower range of initial concentration OII dye (20 - 25 mg/L), concentration of H_2O_2 of 0.8 - 1.0 g/L and pH of solution in a range of 5.5 - 6.0, respectively. At high catalyst dosage, the number of active catalytic sites also increases, which are responsible for the activation of H_2O_2 into •OH radicals. The effective generation of •OH radicals during oxidative reaction leads to an increase in OII removal.

3.4 Experimental Validation of Proposed Optimized Conditions

The optimum operational parameters were found to be at 0.99 g/L catalyst dosage, 20.80 mg/L initial dye concentration, 5.42 pH of solution and 0.88 g/L concentration of H_2O_2 , resulting in

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94.98 % of OII removal with a desirability value of 1. Further validation of the proposed optimized condition was performed experimentally. Table 4 summarizes the experimental data validation of the proposed optimized conditions. Interestingly, it was found that the experimental values obtained were in good agreement with the values predicted from the proposed empirical quadratic model, with a relatively small deviation of 3.98%. Detailed predicted and experimental results of OII removal obtained at optimum conditions are tabulated in Table 4. Such findings present new insight in obtaining a proper model for predicting oxidative dye degradation's performance based on the direct global interactions of critical operational reaction conditions.



Figure 2: Multiple global interaction of independent parameters (catalyst dosage (A), initial concentration of OII dye (B), pH of solution (C) and concentration of H₂O₂ (D)) towards response (OII removal)

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Model desirability	Catalyst dosage (g/L)	Initial			OII removal (%)		
		concentration of OII dye (mg/L)	pH of solution	Concentration of H ₂ O ₂ (g/L)	Predicted	Experimental	Error (%)
1.0	0.99	20.80	5.42	0.87	94.98	91.19	3.98

4. CONCLUSION

The work successfully investigated the effects of catalyst dosage, initial concentration of OII dye, pH of solution and concentration of H₂O₂ on OII removal using RSM-CCD. All operational parameters have significant impacts on OII removal that can be represented in the sequence of catalyst dosage (A) > initial concentration of OII dye (B) > concentration of H₂O₂ (D) > pH of solution (C). The optimum OII removal was obtained using catalyst dosage of 0.99 g/L, OII concentration of 20.80 mg/L, 5.42 pH of solution and H₂O₂ concentration of 0.88 g/L, resulting in 91.19% of OII removal experimentally. The experimental values were in good agreement with the predicted values with negligible differences of a high regression coefficient value of 0.9941. Such findings present an in-depth understanding of detailed global parametric interactions among the operational parameters during catalysis towards OII oxidative degradation using CaMo_{0.67}Cu_{0.33}O₃ perovskite catalyst.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interest regarding the publication of this paper.

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