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Oxidative degradation of AOII dye by bicarbonate-activated hydrogen peroxide in the presence of $CaCu_{0.5}Fe_{0.5}O_3$ perovskite: Influence of initial pH

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ABSTRACT

The catalytic performance of Acid Orange II (AOII) degradation by bicarbonate-activated hydrogen peroxide in the presence of $CaCu_{0.5}Fe_{0.5}O_3$, and the influence of initial pH during the reaction, were investigated in this study. The introduction of bicarbonate ($NaHCO_3$) into the $CaCu_{0.5}Fe_{0.5}O_3/H_2O_2$ system significantly enhanced AOII degradation, achieving 94% removal within 5 minutes of reaction, which was 2.5 times higher than that observed in the absence of $NaHCO_3$ (38%). The BAP-$CaCu_{0.5}Fe_{0.5}O_3$ system effectively degraded AOII across a wide pH range, following a pseudo-second-order kinetic model. Notably, the rate constant at an unadjusted initial pH of 6.4 ($k = 0.0949$ $L \cdot mg^{-1} \cdot min^{-1}$) was approximately 1.7 times higher than that at pH 8 ($k = 0.0549$ $L \cdot mg^{-1} \cdot min^{-1}$), although both conditions achieved comparable AOII degradation efficiencies (>98%) after 30 minutes of reaction. The BAP–$CaCu_{0.5}Fe_{0.5}O_3$ system exhibited its most efficient catalytic performance and fastest AOII degradation rate at an unadjusted initial pH of 6.4, highlighting the critical influence of near-neutral pH on reactive species generation. These findings provide new insights into the role of bicarbonate and initial pH in enhancing the oxidative degradation of organic pollutants using $CaCu_{0.5}Fe_{0.5}O_3$ perovskite catalysts within the bicarbonate-activated hydrogen peroxide system.

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1. INTRODUCTION

Synthetic dyes are widely used across various industries, including textiles, cosmetics, food, and leather, owing to their vivid colouration, high stability, and strong colour intensity. Among these, azo dyes, characterised by the presence of azo linkages (−N=N−) and complex aromatic structures, account for approximately 60% of the total annual dye production [1]. These dyes are favoured for their excellent chemical stability and diverse industrial applications; however, they pose serious environmental and health concerns when discharged untreated into water bodies, owing to their resistance to biodegradation and inherent toxicity [2–4]. In addition, dye pollutants in aquatic environments can significantly reduce light penetration, thereby inhibiting photosynthesis and disrupting aquatic ecosystems [5].

Advanced oxidation processes (AOPs) including catalytic wet air oxidation, catalytic ozonation, catalytic wet peroxide oxidation, sonocatalysis and photocatalysis [6–10] have emerged as widely adopted technologies for wastewater treatment due to their high efficiency and low environmental toxicity. Among various AOPs, Fenton and Fenton-like oxidation processes utilising hydrogen peroxide (H_2O_2) to generate hydroxyl radicals (•OH) have been extensively investigated for the degradation of organic pollutants [11,12]. This activation can be facilitated by various types of catalysts, including soluble iron salts in homogeneous Fenton systems, insoluble iron-based catalysts in heterogeneous Fenton systems, and transition metal-based solid catalysts in heterogeneous Fenton-like processes [13,14]. However, homogeneous Fenton reactions are limited by a narrow optimal pH range (pH 2-3), and the generation of iron sludge during reactions results in secondary contamination, necessitating additional post-treatment separation steps [15]. To overcome these limitations, recent research has focused on heterogeneous Fenton and Fenton-like reactions.

Recently, various UV–visible-light-driven heterogeneous Fenton-like reactions have been developed. For instance, CNTs@Nb_2O_5 [16], MnCuFe-LDH [17], CFA-$CoFe_2O_4$ [18], and $FePO_4$ [19] have exhibited excellent performance in degrading synthetic azo dyes in aqueous systems under ultraviolet or visible light irradiation. In addition to their high degradation efficiency, these catalysts have also demonstrated considerable stability during the reaction process. These catalysts were effective over a relatively wide pH range (approximately 1.7–7), but they performed better under acidic conditions. Nevertheless, many of these catalysts require external energy input from ultraviolet or visible light to maintain activity, which restricts their practical applicability. Therefore, developing heterogeneous Fenton-like catalysts capable of maintaining high efficiency across a broad pH range without dependence on photoenergy input remains a key objective for real-world applications.

Bicarbonate ($NaHCO_3$)-activated hydrogen peroxide (H_2O_2) oxidation, commonly referred to as the BAP system, has attracted growing research interest due to its ability to generate a greater abundance of reactive oxygen species (HCO_4^-, CO_3^-) compared to conventional AOPs [20]. It is a relatively non-toxic anion and one of the most abundant species naturally occurring in aquatic environments, exhibiting excellent performance in water treatment under neutral to mildly alkaline pH conditions [21]. Besides, several studies have reported that the introduction of heterogeneous catalysts into the BAP system can improve the degradation efficiency of the organic pollutants. For instance, Marín-González et al. [22] reported that the degradation rate of Allura Red AC dye reached 99.43% within 1 hr when cobalt-impregnated pillared clay (Co/Al–PILC) catalyst was added to the BAP system. Meanwhile, Xia et al. [23] revealed that the $Ca_2Co_2O_5$-BAP exhibited excellent degradation efficiency of AO7 at 90.9% within 30 min of reaction, which was 5.4 times higher than achieved by the BAP system alone (16.8%).

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Furthermore, recent investigations [24] have highlighted the potential of copper (Cu)-substituted perovskite compounds for the degradation of organic pollutants. Specifically, Cu incorporation at the B-site of the $LaCu_xFe1\text{-}xO_3$ perovskite notably enhanced catalytic activity, achieving a bisphenol A degradation efficiency of 92% within 3 hours at a substitution level of x = 0.5, compared to only 20% removal by pristine $LaFeO_3$ under identical conditions. Despite these promising advancements, the catalytic behaviour of such Cu-substituted perovskites within BAP systems has yet to be explored. Therefore, this study aims to elucidate the role of bicarbonate as a co-oxidizing agent in H_2O_2 activation within the BAP–$CaCu_{0.5}Fe_{0.5}O_3$ perovskite system to achieve enhanced AOII degradation efficiency. Additionally, the influence of initial pH on AOII degradation in the BAP–$CaCu_{0.5}Fe_{0.5}O_3$ system will be systematically investigated.

2. MATERIALS AND METHODS

2.1 Materials

Calcium nitrate tetrahydrate ($Ca(NO_3)_2.4H_2O$; ≥99%), iron(III) nitrate nonahydrate ($Fe(NO_3)_3.9H_2O$; 99%), copper(II) nitrate trihydrate ($Cu(NO_3)_2.3H_2O$; ≥99%), ethylenediaminetetraacetic acid (EDTA), ammonium hydroxide (NH_4OH) solution, hydrogen peroxide (H_2O_2; 30% (w/w)), acid orange II (AOII), hydrochloric acid (HCl) and sodium hydroxide (NaOH) were supplied by Sigma Aldrich. Citric acid monohydrate ($C_6H_8O_7.H_2O$; ≥99%) and sodium bicarbonate ($NaHCO_3$) were purchased from QReC (Asia). All chemicals were of analytical grade and used as received without further purification.

2.2 Synthesis of $CaCu_{0.5}Fe_{0.5}O_3$ perovskite catalyst

A combined ethylenediaminetetraacetic acid (EDTA)-citric acid complexation method was used to synthesize $CaCu_{0.5}Fe_{0.5}O_3$ perovskite in this study. A set of molar ratios of metal ions, EDTA, citric acid, and ammonium hydroxide was kept constant at 1:1.1:2:10 at a fixed precursor molar concentration of 0.05 M. Ammonium hydroxide aqueous solution was added slowly to a beaker containing EDTA, under constant stirring until the EDTA was entirely dissolved and a clear solution formed. In another beaker, the metal ions and citric acid were dissolved in distilled water for 10 minutes. After that, the EDTA solution was added to the mixture under continuous stirring to get the required solution. The resulting solution was then heated to 100 ℃ with constant stirring again to evaporate most of the water until a viscous fluid formed. The viscous fluid was dried in an oven at 90℃ for 24 hours. The resultant sample was calcined in the muffle furnace at 450°C for 8 hours in air at a ramping rate of 5°C min^{-1}. The pre-calcinated sample was further calcinated at 800°C for 4 hours at a ramping rate of 5°C min^{-1}. The resultant sample was characterised using X-ray diffraction (XRD, D8 Advance, Bruker, USA) with Cu-Kα (X = 1.5406 Å) at 40 kV and 40 mA. The sample was analysed using Xpert High Score Software in the range of $10° \leq 2\theta \leq 60°$ to validate the formation of the perovskite phase in the resultant catalyst. Quantitative phase analysis was subsequently performed using the Rietveld refinement method implemented in the software.

The oxidative degradation of AOII in bicarbonate-activated hydrogen peroxide (BAP) in the presence of $CaCu_{0.5}Fe_{0.5}O_3$ was initiated by adding 22 mM of H_2O_2 and 440 mM of $NaHCO_3$ into 100 mL of AOII dye solution (35 mg L^{-1}) while dispersing 0.1 g of the catalyst into the reaction solution. The solution was stirred at 200 rpm for 60 min of reaction without pH adjustment. Similar procedures were repeated in the absence of H_2O_2 and $NaHCO_3$, respectively, while dispersing the catalyst in the solution mixture.

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Approximately 3 mL of the reaction suspension was withdrawn and filtered at regular intervals through a 0.2 µm syringe filter. The concentration of AOII was determined by measuring the absorbance of the sample solution at λmax of 486 nm using a UV-vis spectrophotometer (Perkin Elmer, Lambda 25).

The influence of the initial solution pH on the oxidative degradation of AOII in the bicarbonate-activated hydrogen peroxide (BAP)-$CaCu_{0.5}Fe_{0.5}O_3$ system was systematically investigated. The initial pH of the AOII solution was adjusted to 2.5, 3.0, 4.0, 6.4, 8.0, and 9.0 using 0.1 M HCl or 0.1 M NaOH. The unadjusted AOII solution exhibited a natural pH of 6.4 and was used without further adjustment. Following pH adjustment, the solutions were magnetically stirred for 5 min to ensure complete equilibration before use. The pH was verified using a calibrated pH meter immediately prior to the degradation experiments. Unless otherwise stated, the experimental conditions were maintained at an AOII concentration of 35 mg/L, H_2O_2 concentration of 22 mM, catalyst dosage of 1.0 g/L, and $NaHCO_3$ concentration of 440 mM. All experiments were conducted at room temperature. The percentage of AOII degradation was calculated using the following Eq. (1).

$$\text{Percentage of AOII degradation} = \frac{C_o - C_t}{C_o} \text{x } 100\% \quad (1)$$

where C_o is the initial concentration of AOII (mg L−1), and C_t is the final concentration at a specific reaction time (mg L^{-1}). The experiments were consistently reproducible with a standard deviation of less than 5%.

The kinetic evaluation of oxidative degradation for AOII dye solution at different initial pH values was investigated using pseudo-zero-order, first-order, and second-order kinetic models. Pseudo-zero-order, first-order and second-order kinetic models are defined according to Equations (2) - (4), respectively:

$$\frac{dC_t}{dt} = -k_0 \quad (2)$$

$$\frac{dC_t}{dt} = -k_1.C_t \quad (3)$$

$$\frac{dC_t}{dt} = -k_2.(C_t)^2 \quad (4)$$

The k_o (mg L^{-1} min^{-1}), k1 (min^{-1}), and k_2 (L mg^{-1} min^{-1}) are apparent kinetic rate constants of pseudo-zero-order, first-order, and second-order models, respectively, t is the reaction time, and C_t is the pollutant concentration at a given time t. The following Equations (5) - (7) are obtained when equations (2) - (4) are integrated and linearized, respectively:

$$C_t = C_o - k_t \quad (5)$$

$$\ln C_t = \ln C_o - k_1 t \quad (6)$$

$$\frac{1}{C_t} = \frac{1}{C_o} + k_2 t \quad (7)$$

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3. RESULTS AND DISCUSSION

3.1 Characterisation

The phase identification of the resultant catalyst was analysed by XRD analysis as shown in Fig. 1. The XRD pattern of the synthesized catalyst revealed the formation of the perovskite phase together with several metal oxide phases. The diffraction peaks at 2θ = 30.10° and 39.88° were assigned to the perovskite phases $CaCu_{2.79}Fe_{4.21}O_{12}$ (COD 96-434-4870) and $CaFeO_3$ (COD 96-152-6749), respectively. In addition, the diffraction peaks at 2θ = 24.04° and 33.40° corresponded to a perovskite-like structure (COD 96-152-0830) [25]. Metal oxide phases were identified as CuO (COD 96-721-2242), with characteristic reflections at 2θ = 35.52° and 38.70°, and Ca_2CuO_3 (COD 96-200-2258), with diffraction peaks at 2θ = 14.46°, 33.26°, and 44.36°. These results indicate that the synthesized $CaCu_{0.5}Fe_{0.5}O_3$ catalyst comprises a mixture of perovskite/perovskite-like and metal oxide phases.

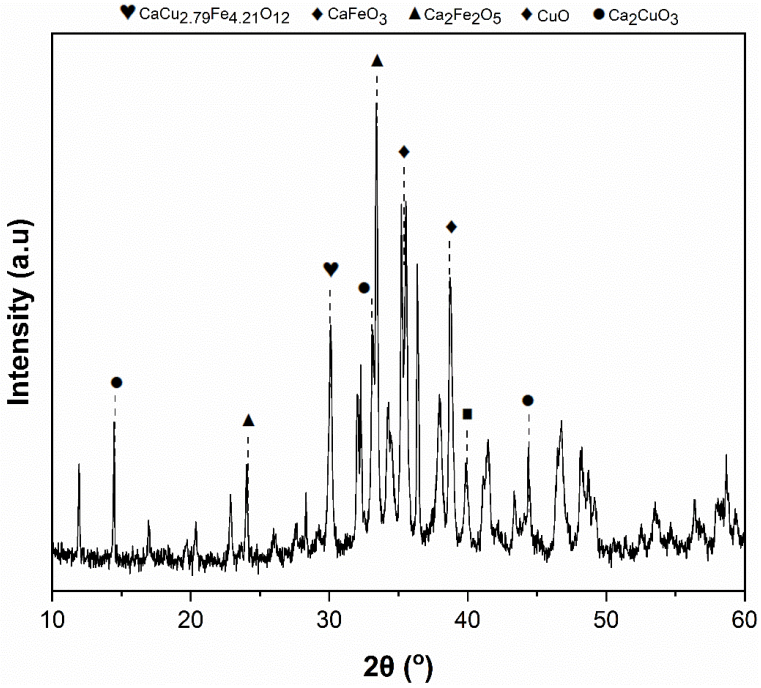

Fig. 1. XRD pattern of $CaCu_{0.5}Fe_{0.5}O_3$ perovskite catalyst.

3.2 Catalytic performance of $CaCu_{0.5}Fe_{0.5}O_3$ at different conditions

Fig. 2. illustrates the catalytic performance of $CaCu_{0.5}Fe_{0.5}O_3$ toward AOII oxidation under various conditions, including the presence and absence of H_2O_2 and bicarbonate ($NaHCO_3$). No noticeable removal of AOII was observed in the presence of H_2O_2 alone. It was found that in the absence of H_2O_2, the adsorption capacity of $CaCu_{0.5}Fe_{0.5}O_3$ toward AOII reached approximately 28% after 60 min. The relatively low adsorption efficiency can be attributed to the strong electrostatic repulsion between the negatively charged perovskite surface and the anionic AOII molecules [26]. Upon the addition of H_2O_2 and H_2O_2/$NaHCO_3$ to the $CaCu_{0.5}Fe_{0.5}O_3$-catalyzed system, AOII removal markedly increased to 97% and 99%, respectively. Furthermore, after 5 min of reaction, only 38% of AOII was degraded in the $CaCu_{0.5}Fe_{0.5}O_3$/H_2O_2 system in the absence of $NaHCO_3$. In contrast, the introduction of $NaHCO_3$ into the $CaCu_{0.5}Fe_{0.5}O_3$/H_2O_2 system

resulted in a rapid AOII degradation of 94% within the same reaction time, which was 2.5 times higher than that observed without $NaHCO_3$. These findings demonstrate the superior catalytic performance of the BAP-$CaCu_{0.5}Fe_{0.5}O_3$ system in the oxidative degradation of AOII. The enhanced catalytic activity in the presence of bicarbonate can be attributed to its role in facilitating H_2O_2 activation and promoting the generation of reactive radical species (•OH and $•CO_3^-$) during the oxidation process. This observation is consistent with the findings of Du and Zhou [27], who reported that bicarbonate promotes the formation of oxidatively active surface complexes ($\equiv Co^{2+}/\equiv Co^{3+}$) at catalyst active sites, thereby enhancing radical generation. The increased formation of such active oxidant species within the catalytic system substantially improves the overall oxidation performance of the perovskite catalyst, leading to a higher degradation efficiency of AOII.

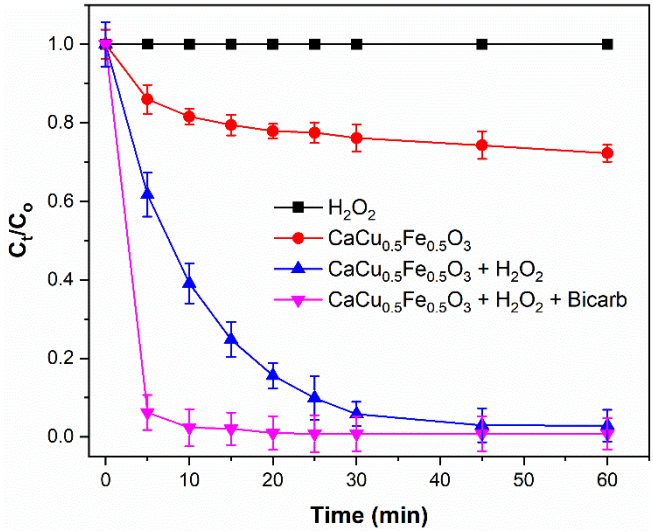

Fig. 2. Degradation profile of AOII solution in the presence of $CaCu_{0.5}Fe_{0.5}O_3$ perovskite catalysts at different conditions. Experimental conditions: AOII = 35 mg L^{-1}; H_2O_2 = 22 mM; $NaHCO_3$ = 440 mM; Catalyst = 1.0 g L^{-1}; room temperature and unadjusted pH (pH 6.4)

The influence of the initial pH on AOII degradation by bicarbonate-activated hydrogen peroxide in the presence of $CaCu_{0.5}Fe_{0.5}O_3$ (BAP-$CaCu_{0.5}Fe_{0.5}O_3$) was investigated over a pH range of 2.5–9, as shown in Fig. 3. At pH 2.5, AOII degradation reached 86% after 30 min of reaction. As the pH increased to 3, 4, 6.4, and 8, the degradation efficiency progressively improved to 88%, 95%, 99%, and 99%, respectively. However, a further increase in pH to 9 resulted in a decrease in degradation efficiency to 87%. This behaviour can be attributed to the partial decomposition of HCO_3^- into CO_2 or CO_3^{2-} under both highly acidic and highly alkaline conditions, thereby reducing the available concentration of HCO_3^- that serves as the H_2O_2 activator [28]. This finding contrasts with the conventional Fenton reaction, where a strongly acidic environment (pH ≈ 3) is typically required to achieve high degradation efficiency of azo dyes [29]. Gupta et al. [30] reported that deviations from the optimal pH ≈ 3 significantly diminish the efficiency of the Fenton process. Interestingly, the results of this study demonstrate that the BAP-$CaCu_{0.5}Fe_{0.5}O_3$ system can efficiently degrade AOII over a broad pH range. This observation is consistent with the findings of Pan et al. [31], who reported that the BAP system enables efficient activation of H_2O_2 across a wider pH window (6–9), corresponding to neutral and mildly alkaline conditions, thereby promoting •OH radical generation

and enhancing catalyst stability. Similarly, Guo et al. [32] and Xia et al. [23] observed comparable behaviour in the degradation of AOII using BAP-S-$CoFe_2O_4$ and BAP-Co_3O_4 systems, respectively, further confirming the beneficial effect of broad pH tolerance in bicarbonate-activated, perovskite-catalyzed oxidation processes.

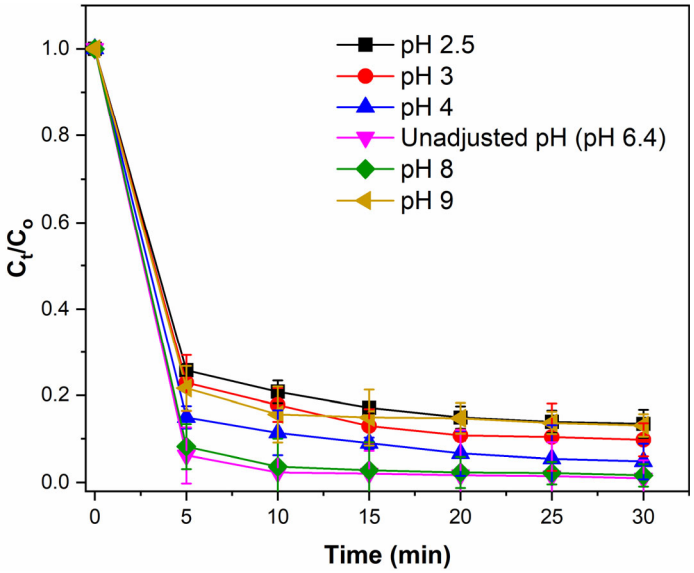

Fig. 3. AOII degradation profile by bicarbonate-activated hydrogen peroxide in the presence of $CaCu_{0.5}Fe_{0.5}O_3$ at different pH values. Experimental conditions: AOII = 35 mg L^{-1}; H_2O_2 = 22 mM; $NaHCO_3$ = 440 mM; Catalyst = 1.0 g L^{-1}; and room temperature

The kinetic behaviour of AOII degradation by BAP-$CaCu_{0.5}Fe_{0.5}O_3$ at different initial pH (Fig. 4.) was further analysed using three kinetic models: pseudo-zero-order, pseudo-first-order, and pseudo-second-order. The correlation coefficients (R^2) and apparent kinetic constants (k_0, k_1, and k_2) of AOII degradation at different initial pH values are summarised in Table 1. As shown in Table 1, the pseudo-second-order kinetic model provided the best fit for AOII oxidation by BAP-$CaCu_{0.5}Fe_{0.5}O_3$ over the pH range of 2.5–9, exhibiting higher R^2 values ($R^2 > 0.9$) than the zero-order and first-order models. This result indicates that the catalytic reaction of BAP-$CaCu_{0.5}Fe_{0.5}O_3$ depends on both the AOII concentration and the concentrations of the oxidants ($NaHCO_3$ and H_2O_2). Moreover, the pseudo-second-order model is often associated with Fe^{3+}-containing catalytic processes, suggesting that the AOII degradation proceeds primarily through a single degradation stage [33].

The reaction rate constant (k_2) for AOII degradation by BAP-$CaCu_{0.5}Fe_{0.5}O_3$ at different initial pH values is presented in Fig. 5. As shown, the rate constant at an initial pH of 6.4 (k = 0.0949 $L \cdot mg^{-1} \cdot min^{-1}$) was approximately 1.7 times higher than that at pH 8 (k = 0.0549 $L \cdot mg^{-1} \cdot min^{-1}$), despite both conditions achieving similar AOII degradation efficiencies (99%) within 30 min of reaction. This finding suggests that near-neutral conditions are particularly favourable for reactive species generation in the BAP-$CaCu_{0.5}Fe_{0.5}O_3$ system. Therefore, the unadjusted pH (pH 6.4) represents the optimal condition for AOII degradation by BAP-$CaCu_{0.5}Fe_{0.5}O_3$, owing to its higher reaction rate constant and superior overall

degradation performance. Similar trends have been reported for dye degradation in bicarbonate-activated hydrogen peroxide systems in the presence of heterogeneous catalysts [27,31,34].

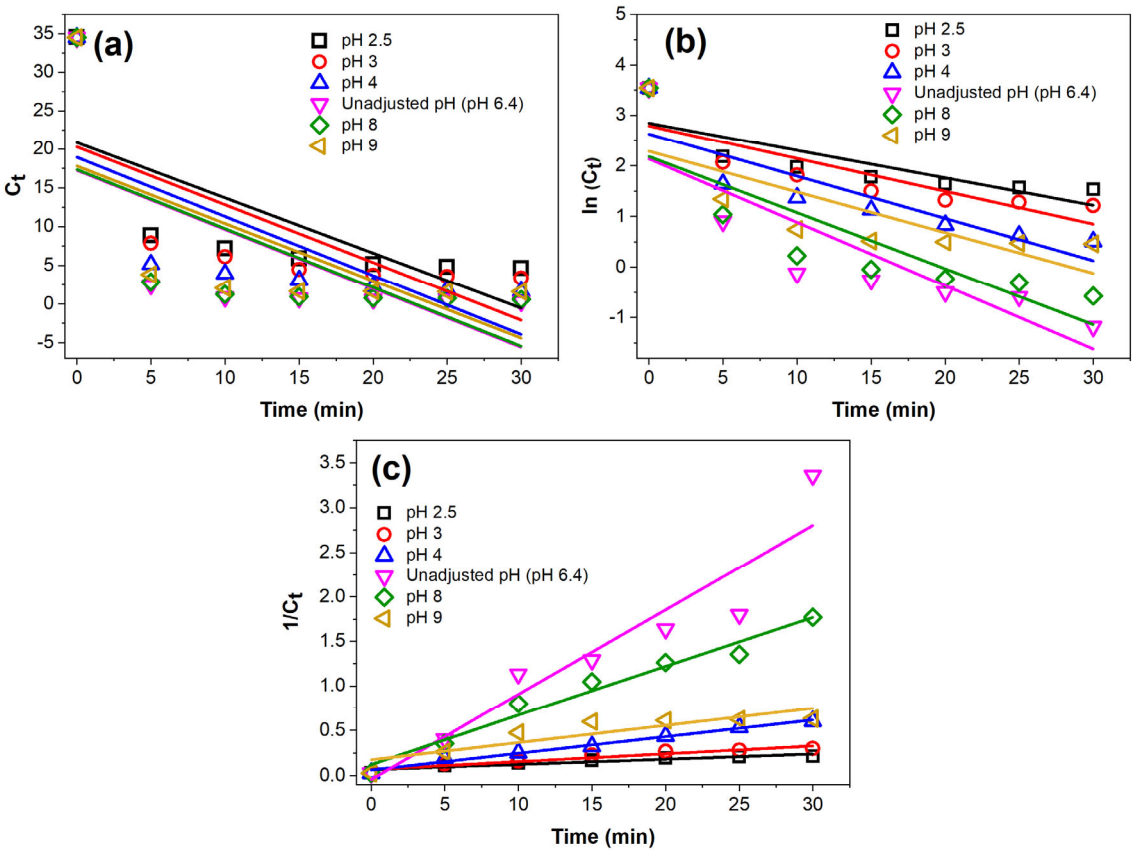

Fig. 4. AOII degradation profile by bicarbonate-activated hydrogen peroxide in the presence of $CaCu_{0.5}Fe_{0.5}O_3$ at different pH values using different kinetic models. Experimental conditions: AOII = 35 mg L^{-1}; H_2O_2 = 22 mM; $NaHCO_3$ = 440 mM; Catalyst = 1.0 g L^{-1}; and room temperature

Table 1. Apparent kinetic rate constants of the zero-order (k_0), first-order (k_1), and second-order (k_2) and correlation coefficients (R^2) obtained after data fits for degradation of AOII (30 min) by using bicarbonate-activated hydrogen peroxide in the presence of $CaCu_{0.5}Fe_{0.5}O_3$ at different initial pH values.

pH	Zero Order k_o (mg L^{-1} min^{-1})	R^2	First Order k_1 (min^{-1})	R^2	Second Order k_2 (L mg^{-1} min^{-1})	R^2
pH 2.5	0.6438	0.4874	0.0531	0.7187	0.0058	0.9074
pH 3	0.6540	0.4669	0.0623	0.7430	0.0087	0.9382
pH 4	0.6394	0.3873	0.0860	0.7447	0.0240	0.9866
pH 6.4	0.6251	0.3401	0.1232	0.7445	0.0949	0.9504
pH 8	0.6450	0.3189	0.1097	0.7606	0.0549	0.9468
pH 9	0.6165	0.3718	0.0780	0.7244	0.0190	0.9003

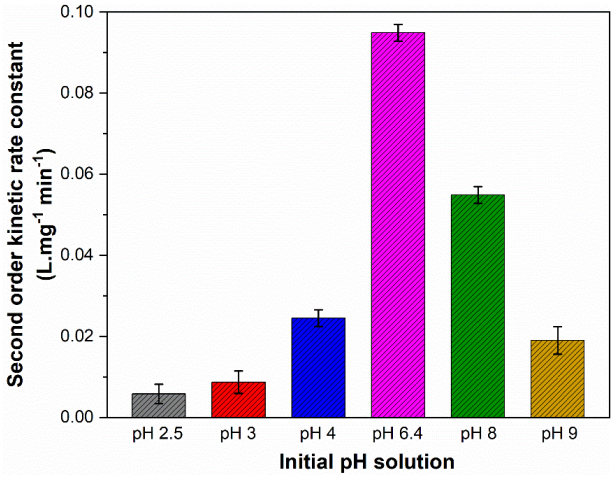

Fig. 5. Reaction Kinetic Rate Constant (k_2) of AOII degradation by bicarbonate-activated hydrogen peroxide in the presence of $CaCu_{0.5}Fe_{0.5}O_3$ at different pH values. Experimental conditions: AOII = 35 mg L^{-1}; H_2O_2 = 22 mM; $NaHCO_3$ = 440 mM; Catalyst = 1.0 g L^{-1}; t = 30 min; and room temperature

4. CONCLUSION

The catalytic performance of $CaCu_{0.5}Fe_{0.5}O_3$ toward AOII oxidation was evaluated under various conditions, including the presence and absence of H_2O_2 and bicarbonate ($NaHCO_3$). In the BAP-$CaCu_{0.5}Fe_{0.5}O_3$ system, 94% AOII degradation was achieved within 5 minutes, representing a significant improvement compared to $CaCu_{0.5}Fe_{0.5}O_3/H_2O_2$, $CaCu_{0.5}Fe_{0.5}O_3$ or H_2O_2 alone (0.5–38%) under similar conditions. The enhanced catalytic performance was attributed to the generation of highly reactive radicals (•OH and $•CO_3^-$) which facilitated rapid AOII oxidation. The BAP-$CaCu_{0.5}Fe_{0.5}O_3$ system exhibited excellent degradation efficiency (99%) across a broad pH range (6.4–8) within 30 minutes, following a pseudo-second-order kinetic model. The rate constant at an initial pH of 6.4 ($k = 0.0949\ L \cdot mg^{-1} \cdot min^{-1}$) was approximately 1.7 times greater than that at pH 8 ($k = 0.0549\ L \cdot mg^{-1} \cdot min^{-1}$), despite both achieving comparable degradation efficiencies (99%). The unadjusted initial pH of 6.4, which is a near-neutral condition was determined to be optimal for AOII degradation in the BAP-$CaCu_{0.5}Fe_{0.5}O_3$system due to its superior reaction rate and overall catalytic performance.

5. ACKNOWLEDGEMENTS

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6. CONFLICT OF INTEREST STATEMENT

The authors affirm that there is no conflict of interest pertaining to the publication of this paper.

7. AUTHORS' CONTRIBUTION

Ahmad Azizi Abdul Razak: Material preparation, data collection and analysis; **Rasyidah Alrozi**: Conceptualisation, methodology, formal analysis, investigation and writing the original draft; **Riza Lydia Liyana Rizalme**n: Methodology and analysis**; Nor Aida Zubir**: Conceptualisation, methodology, resource, analysis, validation, writing-review and editing; **Mohamad Anuar Kamaruddin**: Validation, writing-review and editing.

8. DECLARATION OF GENERATIVE AI IN THE WRITING PROCESS

During the preparation of this work, the authors used ChatGPT to improve the readability and language quality of the manuscript. Following the use of this AI tool, the authors reviewed and edited the content as necessary and took full responsibility for the final content of the publication.

9. DATA AVAILABILITY

Data will be made available on request.

10. ETHICS STATEMENT

The authors declare that this research did not involve human or animal subjects. All experimental procedures were performed following the institutional Safety, Health, and Environmental (HSE) protocols of Universiti Teknologi MARA (UiTM), Cawangan Pulau Pinang, Malaysia.

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