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DETERMINATION OF OPTIMAL PARAMETERS AND MECHANISM OF REMOVAL OF REACTIVE DYE SUNZOL BLUE RS 150% (CI REACTIVE B19) BY MODIFIED FENTON PROCESS

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ABSTRACT

Kinetic models of the color degradation process based on the modified Fenton reaction (continuous H_2O_2 injection throughout the experiment), including first-order, second-order, and Behnajady–Modirshahla–Ghanbary (BMG) kinetic models, were used to evaluate the efficiency and mechanism of the removal of the reactive dye Sunzol blue RS 150% (BRS) for each parameter studied. The parameters studied included initial pH, Fe^{2+} concentration, stirring speed, H_2O_2 content, and BRS concentration. The results showed that the BRS removal process occurred in two stages: (i) fast in the first stage (20 min) and (ii) slowed down thereafter. The study determined the experimental optimal parameters, including C_0 100 mg/L, pH 3, Fe^{2+} 16.8 mg/L, stirring speed 300 rpm, H_2O_2 0.5%, and injection speed 1.5 mL/min. The BMG model was suitable for explaining the BRS removal mechanism. The research results confirm that the modified Fenton process is more effective, the BMG model is suitable to explain the color removal mechanism (2 stages), and is the basis for the design of color removal in water on a larger scale.

Keywords: Reactive Dye, Modified Fenton, BMG Model, HO• Radical, Fe^{2+} Catalyst

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INTRODUCTION

Sunzol blue RS 150% reactive dye from Korea (CI reactive 19) has the general formula $C_{22}H_{16}N_2Na_2O_{11}S_3$ and is a sulfonate salt with aromatic ring structures. Vietnam is one of the world's three largest importers of reactive dyes. Reactive dyes are persistent, bioaccumulative, and toxic to aquatic organisms, threatening drinking water supplies and human health.[1] The main treatment methods for dye removal are membrane, electrochemical, coagulation, biodegradation, ion exchange, and adsorption. These methods have many disadvantages, such as high removal cost, low efficiency (for color compounds with complex chemical composition), high dependence on operating conditions (long treatment time), and secondary pollution generation.[1-3] Several advanced oxidation processes (AOPs) have been investigated, including the Fenton process, photocatalysis, electrocatalysis, and ozonation for the treatment of reactive dye wastewater.[1] Among them, the homogeneous Fenton process with Fe^{2+} and hydrogen peroxide (H_2O_2) as an important precursor to produce HO•, HOO• has attracted the attention of many scientists, Table-1.[1, 4] In which, the hydroxyl radical •OH has the strongest reaction with a reduction potential of $E^0 = 2.80$ V.[5, 6] This radical has a strong tendency to accept electrons and will effectively decompose organic compounds. During the process, a series of single electron transfer reactions to create HO• and HOO• free radicals such as between Fe^{2+}, $Fe^{3+,}$ and H_2O_2 (reaction 1, reaction 2), causing loss of HO• free radicals (reaction 3, reaction 4, reaction 5 and reaction 6), creating weaker free radicals or losing H_2O_2 (reaction 7 and reaction 8).[7] The main advantages of the Fenton process are short reaction time, carried out at room temperature and atmospheric pressure, cheap and widely available chemicals used, and furthermore, it can be integrated into other wastewater treatment methods such as coagulation, filtration, and biological oxidation.[8]

Kinetic research is the basis for determining the reaction mechanism of a chemical process. Thereby, it is possible to control reaction conditions such as increasing the rate of the main reaction and reducing or slowing down the rate of side reactions, which is the basis for implementing industrial-scale design.[9, 10] Studies showed that the Fenton process kinetics include two stages. In the first stage, the oxidation of Fe^{2+} to form HO•, reaction (1) occurs quickly, and then in the second stage (occurs slowly), the regeneration of

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Fe^{2+} and the formation of HOO• radicals, reaction (2). There are many kinetic models used to explain dye removal mechanisms, including first-order kinetic models[5, 11, 12], second-order kinetic models,[13, 14] And the Behnajady–Modirshahla–Ghanbary model (BMG).[10]

Table-1: Reactions of the Fenton Process

$H_2O_2 + Fe^{2+} \rightarrow HO\bullet + Fe^{3+} + OH-$ $k=50\text{-}80\ M^{-1}\ s^{-1}$ [15]	(1)
$H_2O_2 + Fe^{3+} \rightarrow Fe^{2+} + HOO\bullet + H^+$ (Fenton-like reaction) $k=0.002\text{-}0.01\ M^{-1}\ s^{-1}$ [10]	(2)
$HO\bullet + Fe^{2+} \rightarrow Fe^{3+} + HO-$ $k=3.2\ 10^8\ M^{-1}\ s^{-1}$	(3)
$HOO\bullet + HO\bullet \rightarrow H2O + O2$ $k=1.2\ 107\ M^{-1}\ s^{-1}$ [16]	(4)
$2HO\bullet \rightarrow H_2O_2$ $k=5.3\ 10^9\ M^{-1}\ s{-}1$ [16] $HO\bullet + HOO\bullet \rightarrow 2H_2O + O_2$ $k=10^{10}\ M^{-1}\ s^{-1}$ [16] $2HOO\bullet \rightarrow H_2O_2 + O_2$ re-link [17].	(5)
$Fe^{3+} + HOO\bullet \rightarrow Fe^{2+} + O_2 + H+$ $k<2.10^3\ M^{-1}\ s^{-1}$ [18]	(6)
$HO\bullet + H_2O_2 \rightarrow H_2O + HOO\bullet$ [19]	(7)
$2H_2O_2 \rightarrow 2H_2O + O_2$	(8)

Studies also showed that the suitability of the kinetic model depends on the type of dye used, the operating conditions (Fe^{2+}, H_2O_2 addition process).[12, 15, 20, 21] The previous Fenton process operations mostly involved a single H_2O_2 injection at the beginning of the experiment, [12, 15, 19-23] which would reduce the color removal efficiency due to the high amount of H_2O_2 at one time, causing free radical loss (Reactions 7 and 8).[8] Therefore, the modification of the H_2O_2 injection process to avoid the escape as presented has been raised. However, such research is currently lacking in information, specifically with the reactive dye Sunzol blue RS 150%. Furthermore, the studies have focused more on evaluating the color removal efficiency than on studying the color removal mechanism through kinetic models. The purpose of the study is to use kinetic models to determine the optimal conditions and parameters from the modified Fenton process: continuous H_2O_2 injection throughout the experiment.

EXPERIMENTAL

Experimental Design for BRS Removal

The experiment was carried out at room temperature (28 °C) in a 1000 mL beaker containing BRS solution at pre-prepared color concentrations, Fe^{2+} content was added next, and initial pH was adjusted with 0.1 N H_2SO_4 or 0.1 N NaOH using a BP3001 Hana pH meter (pH was not adjusted during the experiment), stirring speed was adjusted on a STUART UC152 heated magnetic stirrer (8x70 mm magnetic stirrer length), H_2O_2 feeding rate was fixed at 1.5 mL/min using a BQ50-1J-A peristaltic pump (0.0002 mL/min to 20 mL/min, made in China) with varying H_2O_2 contents. Samples (5 mL) were withdrawn at fixed times (5, 10, 20, 30, and 60 min) and centrifuged at 4000 rpm for 5 min on a Dlab M0630 centrifuge (USA). The concentration of BRS in the centrifuge solution was measured by the colorimetric method. All experiments were carried out three times. The first-order, second-order, and BMG kinetic models were used to select the optimal process parameters based on the correlation coefficient R2, the calculated results from the model, the experimental results, and the rate constants. The experimental treatments considered based on preliminary experiments included: (i) Fe^{2+} content (8.4; 16.8; 25.2 mg/L); (ii) Initial pH (1, 3, 5); (iii) Stirring speed (250, 300, 350 rpm); (iv) H_2O_2 content (1.5 mL/min with concentrations of 0.5; 1.0 and 2.0%); and initial dye content (50; 100 and 200 mg/L).

Chemicals and Analytical Methods

Chemicals, including Sulfuric acid 98%, NaOH 96%, H_2O_2 30%, and $FeSO_4$ $7H_2O$ of analytical grade from China, were used in the experiments. Active color Sunzol blue RS 150%, of Korean origin, was obtained from a trading company in Ho Chi Minh City, Vietnam. Color analysis of active dye Sunzol blue RS 150% by UV-Vis absorption spectrum (Model: GENESYS 10S UV-VIS, 6-/1-CELL) at 600 nm wavelength.

Experimental Data Processing

The concentration of BRS was calculated according to the Lambert-Beer law. The decomposition efficiency was calculated according to formula (9).

$$De_t = 100 * \frac{C_0 - C_t}{C_0} \quad (9)$$

Det: Color removal efficiency at time t (%); C_0 is the initial BRS concentration at time t = 0 (mg/L), and Ct is the BRS concentration obtained after centrifugation at time t (minutes).

First-order kinetic model equation, expression(10):[5, 11, 12]

$$Ln\left(\frac{C_t}{C_0}\right) = -k_1 t \quad (10)$$

Second-order kinetic model equation, expression (11):[14]

$$Open\left(\frac{1}{C_t}\right) = \frac{1}{C_0} + k_2 t \quad (11)$$

k_1 first order rate constant ; k_2 second-order rate constant.

Behnajady–Modirshahla–Ghanbary (BMG) kinetic model equation, expression (12):[10, 15]

$$\left(\frac{t}{1 - \frac{C_t}{C_0}}\right) = b.t + m \quad (12)$$

In which: b and m are two characteristic constants related to reaction kinetics and oxidation capacity. Determine the constants by plotting $\frac{t}{1-\frac{C_t}{C_{0_t}}}$ against t.[10, 20] When t is very short or approaches zero, the slope obtained is the inverse of the constant m, which represents the fast initial removal rate of the color, expression (13). Therefore, the higher 1/m, the faster the initial removal rate.

$$Open\left(\frac{d\frac{C_t}{C_0}}{dt}\right) = -\frac{1}{m} \quad (13)$$

As t is long and approaches infinity, the resulting intercept is the inverse of the constant b (1/b), which represents the maximum theoretical value of the color fraction removed, expression (14). It also represents the maximum oxidation capacity of the Fenton process.

$$\left(\frac{1}{b}\right) = 1 - \frac{C_t}{C_0} \quad (14)$$

SPSS 23.0 was used to determine the homogeneity of variance, then determine the difference in mean values between experiments with p-value < 0.05 by Tukey's post hoc test when Sig>0.05 or Tamhane when Sig<0.05.

RESULTS AND DISCUSSION

Evaluation of the Sunzol Blue RS 150 (BRS) reactive dye removal process was based on the color removal efficiency and kinetic parameters established experimentally.

Initial pH

The color removal ability of the Fenton process at initial pHs (1, 3, and 5) under laboratory conditions such as C_0 100 mg/L, Fe^{2+}: 16.8 mg/L, stirring speed 300 rpm, H_2O_2 1% loading rate 1.5 mL/min, temperature 28°C±1 has been presented in Fig.-1. The results showed that the color removal process at all pHs occurred in 2 stages: fast in the first 20 minutes, and gradually slowed down until 30 minutes, when it stopped. Specifically, at the initial 20 minutes, the color removal efficiency was 60.4, 92.6, and 28.8%, respectively, for initial pH of 1, 3, and 5. After the next 10 minutes, the efficiency only increased by 7.2, 1.5, and 6.8%, respectively. The ANOVA analysis results also showed that the color removal process took place in 2 stages: (i) a significant, rapid increase (20 minutes) and a slow, insignificant increase after about 20-30 minutes.

This can be explained that the initial stage of color content is large enough, and the amount of HO• formed from reaction (1) quickly removes the color. In stage 2, the amount of color in the solution decreased, and the amount of HO• needed more, or in other words, it needed to increase H_2O_2, but the H_2O_2 was continuously pumped in unchanged throughout the entire experiment, and the color removal efficiency

decreased. In addition, the amount of Fe^{3+} accumulated more, at that time, the reaction to form the HOO• radical, Reaction (2), was dominant. Because the reaction to form the HOO• radical took place much slower than the reaction to form the HO• radical, reaction (1), this also reduced the color removal efficiency.[10, 15]

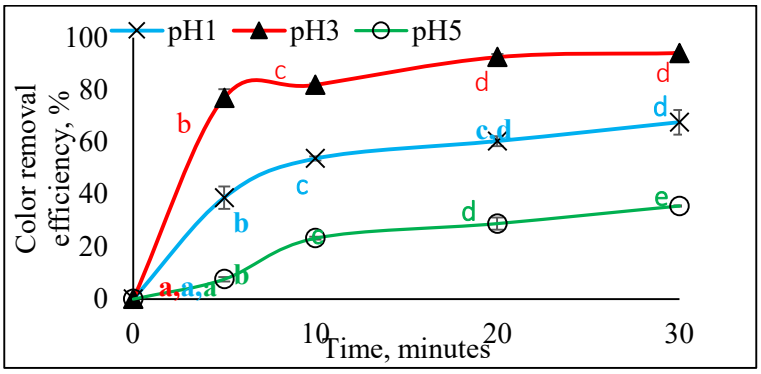

Fig.-1: Representation of Color Removal Efficiency Over Time at different Initial pHs. (The letters a,b,c,d,e in the Same Color Indicate Statistically Significant Differences.)

The results also showed that at each sampling time, the color removal efficiency under experimental conditions was lowest at pH 5 and highest at pH 3, Fig.-1. At the end of the 30-minute survey, the color removal efficiency was 35.6, 67.6, and 94.1%, respectively. Similar results were also found in the study of Ertugay & Acar, the Direct Blue 71 color removal experiment also showed high color removal efficiency at pH 3.[20] The results obtained can be explained by the fact that at pH 1, under the influence of H+, a part of H_2O_2 was converted to H3O2+, reducing the efficiency of H_2O_2.[18] Furthermore, due to the significantly high hydrolysis rate of Fe^{3+} (10^5–10^6 M^{-1} s^{-1}) and Fe^{2+} (10^4-10^5 M^{-1} s^{-1}) as pH increases [24] Iron sludge is easily formed, reducing the efficiency of the iron catalyst.

Table-2: Parameters of the Models at the Investigated pH Values

Kinetic model	1st order model			2nd order model			BMG model			
Parameters	R^2	k_1	C_0	R^2	k_2	C_0	R^2	1/b	1/m	E% TN
pH 1	0.94	0.02	64.0	0.97	0.0006	69.8	1.00	0.79a	0.17^a	67.6
pH 3	0.94	0.06	28.4	0.96	0.0056	71.5	1.00	0.99^a	0.59^b	94.1
pH 5	0.88	0.01	89.1	0.91	0.0002	90.3	0.49	0.96	0.02	35.6

E% TN: The color removal efficiency of the experiment was achieved in 30 minutes. Different letters a,b in the same column indicate that their values are significantly different. The results of calculating the parameters of the first-order and second-order kinetic models, Table-2, all show a high correlation with R2 >0.88 for all 3 investigated pHs (1, 3, and 5). However, when considering the calculated C_0 values and the experimental C_0 values (100 mg/L), they are very different. Specifically, the C_0 values calculated by the first-order model (64.0, 28.4, and 89.1 mg/L) and by the second-order model (69.8, 71.5, and 90.3 mg/L) corresponding to pHs 1, 3, and 5 are different from the experimental C_0 (100 mg/L). Therefore, these two models are not suitable to explain the BRS color removal mechanism under these experimental conditions. For the BMG model, in the case of pH 5, R2 <0.5 (0.49), so this model is not suitable. The cases of pH 1 and pH 3 have a very high correlation with R^2~1.0, and according to the calculation of 1/b value from the model, when the longest reaction time reaches 0.79 and 0.99, which is consistent with the experimental removal efficiency values of 67.6 and 94.1% when the reaction is conducted for 30 minutes. However, the calculated 1/m value representing the initial decomposition rate at pH 3 is significantly larger according to One-way ANOVA analysis (0.59 vs. 0.17). Therefore, the BMG model is suitable, and pH 3 is selected for the next experiments.

Fe^{2+} Content

The results of the study on the color removal ability of the Fenton process when changing the Fe^{2+} catalyst content under initial conditions such as pH 3, C_0 100 mg/L, stirring speed 300 rpm, H_2O_2 1% loading rate 1.5 mL/min, temperature 28°C±1 were presented in Fig.-2. The results showed that the color removal rate occurred in 2 stages: fast in the first 20 minutes and gradually slowed down afterward. After 30 minutes, it

was considered to stop. Specifically, in the first 20 minutes, the color removal efficiency was 65.0, 92.6, and 87.4%, respectively. Then, it increased slowly from the 20th to the 30th minute; the treatment efficiency only increased by 2.4%, 3.9%, and 1.5%, respectively. The results of the One-way ANOVA analysis also showed similar results. Increasing the Fe^{2+} content from 8.4 to 16.8 mg/L increased the color removal rate and the color removal efficiency. This can be explained by the fact that increasing the catalyst concentration increased HO•, reaction (1). However, when continuing to increase Fe^{2+} to 25.2 mg/L, the rate and efficiency increased insignificantly, Fig.-2. This can be explained by the fact that when Fe^{2+} increased too much, it would consume HO• to form OH^- and Fe^{3+}, reaction (3). A similar explanation was also found in the study of Ziembowicz & Kida.[8]

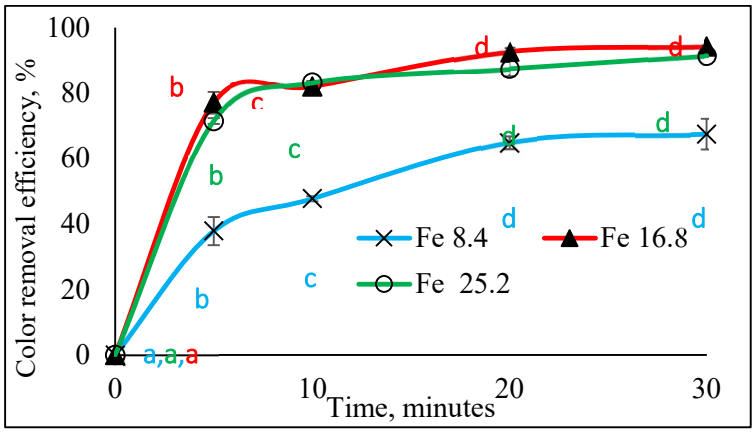

Fig.-1: Representation of Color Removal Efficiency Over Time at different Fe^{2+} Concentrations, (The letters ^{a,b,c,d} in the Same Color Indicate Statistically Significant Differences)

Furthermore, the results also showed that at each sampling time, the color removal efficiency at the Fe^{2+} content of 8.4 mg/L was lower than when using the Fe^{2+} content of 16.8 and 25.2 mg/L, Fig.-2. At the end of the 30-minute survey, the color decomposition efficiency was 67.4, 94.1, and 91.3%, respectively.

Table-3: Calculation Results of the Parameters of the Models at Fe^{2+} Concentrations

Kinetic model	1st order model			2nd order model			BMG model			
Parameters	R^2	k_1	C_0	R^2	k_2	C_0	R^2	1/b	1/m	E% TN
Fe 8.4	0.92	0.03	66.0	0.94	0.0006	71.7	0.99	0.82^a	0.131^a	67.4
Fe 16.8	0.94	0.06	28.4	0.96	0.0056	71.5	1.00	0.99^b	0.591^b	94.1
Fe 25.2	0.90	0.04	30.1	0.96	0.0027	38.9	1.00	0.95^b	0.597^b	90.6

Different letters in the same column indicate that their values are significantly different. The results of calculating the parameters of the kinetic models all showed a high correlation with R2>0.90 for all 3 investigated cases, Table-3. However, when considering the C_0 values calculated from the first-order and second-order kinetic models, they were very different from the experimental C_0. Specifically, the C_0 calculated from the first-order model was 66.0, 28.4, 30.1, respectively, and that of the second-order model was 71.7, 71.5, 38.9, corresponding to Fe 8.4, Fe 16.8, and Fe 25.2 mg/L, which was very different from the experimental C_0 of 100 mg/L. Meanwhile, the BMG model, in addition to the very high R_2 value >0.99, also has a suitable 1/b value, specifically, 1/b is 0.82; 0.99, and 0.95 respectively, corresponding to the experimental efficiency after 30 minutes of 67.4; 94.1 and 90.6%, corresponding to the Fe^{2+} content values (8.4, 16.8 and 25.2 mg/L). In which 1/b in the Fe^{2+} condition of 8.4 mg/L is significantly lower than the other two experiments, and there is no significant change between the other two experiments, Table-3 (according to One-way ANOVA analysis). When considering the 1/m value, which represents the reaction rate, the 1/m of Fe^{2+} concentrations of 16.8 and 25.2 mg/L are considered equivalent, not significantly different (0.591 and 0.597), and significantly higher than that of Fe^{2+} 8.4 mg/L (0.131). When the Fe^{2+} concentration is low, the catalytic efficiency of Fe ions is limited and leads to less decomposition of H_2O_2 to form HO• radicals, Reaction (1). The use of large amounts of Fe^{2+} (either in dissolved or precipitated form) in the reaction water requires additional treatment, which incurs costs.[8] The Fe^{2+} content used in the study was much lower than the previous study, when removing Congo Red and Rhodamine B by Fenton

reaction with Fe^{2+} was 57.68 to 58.13 mg/L (Ghribi & Bagane, 2023). The study results showed that the BMG model was suitable under experimental conditions and parameters such as pH 3 and Fe^{2+} 16.8 mg/L were selected for further studies.

Stirring Speed

The effect of stirring speed on the color removal ability of the Fenton process under laboratory conditions, such as pH 3, C_0 100 mg/L, Fe^{2+} 16 mg/L, H_2O_2 loading rate 1% 1.5 mL/min, temperature 28°C±1, has been presented in Fig.-3. The results show that the color removal rate at all stirring speeds occurs in 2 fast stages in the first 20 minutes and gradually slows down until 30 minutes, considered to stop at all 3 stirring levels of 25, 300, and 350 rpm. Specifically, in the first 20 minutes, the color decomposition efficiency is 88.1%, 92.6%, and 90.5%, respectively. The color removal efficiency only increases slightly after 30 minutes of the experiment, respectively 1.6, 1.5, and 1.9%. The results of the One-way ANOVA analysis also gave similar results, Fig.-3. The results showed that the color removal efficiency when changing the stirring speed did not fluctuate much for all 3 cases, specifically the treatment efficiency was 89.7, 94.1, and 92.4%. This could be due to the rapid mass transfer of the homogeneous Fenton reactions.[24]

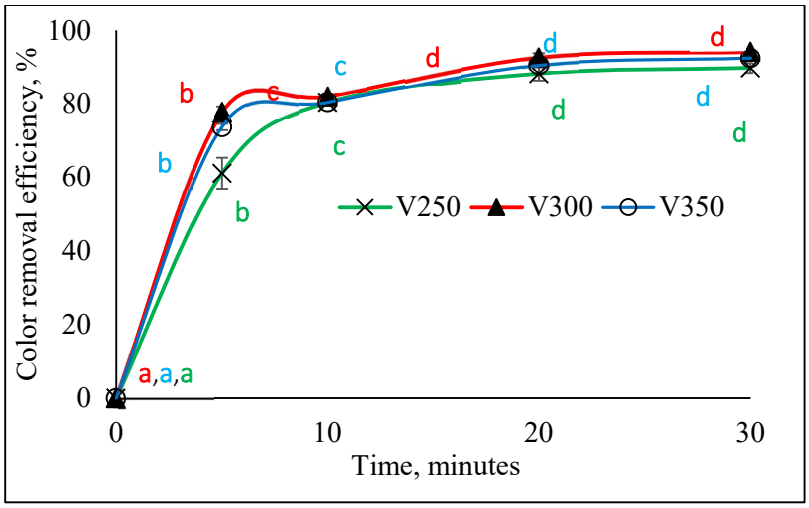

Fig.-2: Performance of Color Removal Efficiency Over Time at Different Stirring Speeds. The Letters [a,b,c, and d] in the Same Color Indicate Statistically Significant Differences.

When changing the stirring speed, the results of calculating the parameters of the first-order, second-order, and BMG kinetic models all showed a high correlation R2 (>0.85), Table-4. However, the C_0 values calculated from the first-order and second-order kinetic models were very different from the experimental Co. Specifically, the C_0 calculated from the first-order model were 39.5; 28.4; 30.6 mg/L, respectively, and those of the second-order model were 56.6; 71.5; 55.5 mg/L, respectively, for the conditions with stirring speeds of 250, 300 and 350 rpm, which were very different from the experimental C_0 (100 mg/L). Meanwhile, the BMG model, in addition to the very high R^2 value (>0.99), also has a suitable 1/b value, specifically 1/b is 0.98, 0.99, and 0.98, corresponding to the experimental efficiency after 30 minutes of 89.7, 94.1, and 92.4%, respectively. The increase in 1/b at 300 rpm is significantly higher than the other two stirring speeds (according to Oway-ANOVA analysis), Table-4. For the 1/m value, representing the reaction rate, the stirring speeds of 300 and 350 rpm have significantly higher values than the stirring speed of 250 rpm. However, the 1/m at the two stirring speeds of 300 and 350 rpm are not significantly different. Therefore, based on energy saving, the stirring speed of 300 rpm was selected. From the research results of pH 3, Fe^{2+} 16.8 mg/L, and stirring speed 300 rpm were selected for the following experiments.

Table-4 Calculation Results of the Parameters of the Models at Different Stirring Speeds

Kinetic model	1st order model			2nd order model			BMG model			
Parameters	R^2	k_1	C_0	R^2	k_2	C_0	R^2	1/b	1/m	E% TN
250 rpm	0.85	0.050	39.5	0.93	0.0028	53.6	1.00	0.98^a	0.38^a	89.7
300 rpm	0.94	0.057	28.4	0.96	0.0056	71.4	1.00	0.99^b	0.59^b	94.1
350 rpm	0.95	0.052	30.6	0.98	0.0042	55.4	1.00	0.98^a	0.52^b	92.4

Different letters[a,b] in the same column indicate that their values are significantly different.

H_2O_2 Content

Effects of changing the H_2O_2 content on the color removal efficiency under laboratory conditions, such as pH 3, C_0 100 mg/L, Fe^{2+} 16 mg/L, stirring speed 300 rpm, temperature 28°C±1, were presented in Fig.-4. The results showed that the color removal efficiency at all speeds occurred in 2 fast stages in the first 20 minutes, and gradually slowed down after that, until 30 minutes, it was considered to stop. Specifically, in the first 20 minutes, the color removal efficiency was 81.3, 91.9, and 82.8%, respectively. After 30 minutes, the removal efficiency only increased by 1.6, 2.0, and 1.5%, respectively. The results of the One-way ANOVA analysis also showed that the color removal efficiency occurred in 2 stages: a significant, rapid increase (20 minutes) and an insignificant increase thereafter.

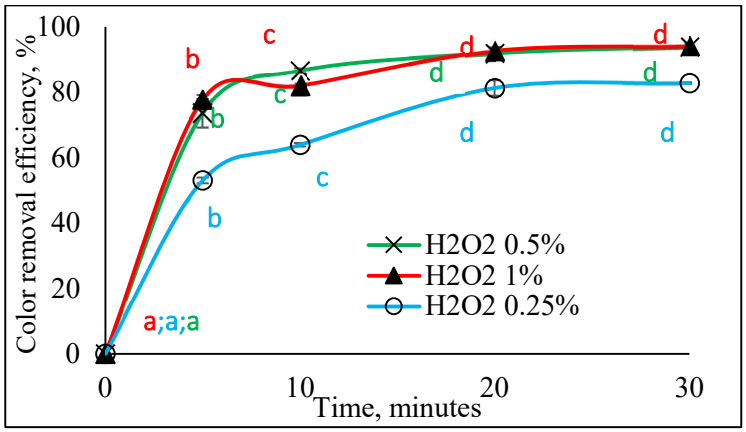

Fig.-3: Representation of Color Removal Efficiency Over Time at H_2O_2 Contents, The Letters a,b,c,d in the Same Color Indicate Statistically Significant Differences

The effect of changing the H_2O_2 content loaded into the reaction system on the color removal efficiency under the experimental conditions is shown in Fig.-4. When the H_2O_2 content was 0.25%, the color removal efficiency was lower at all sampling times. Specifically, at the times of 5, 10, 20, and 30 minutes, the color decomposition efficiency was 53.0, 63.9, 81.3, and 82.8%, respectively, which were lower than the other two experiments, Fig.-4. The results can be explained by the fact that when the H_2O_2 content is too low, the reaction efficiency is decreased due to the lack of HO• radicals produced. On the contrary, increasing the dosage of H_2O_2 increased the generation of HO• radicals, reactions (1) and (2), but when the H_2O_2 content is too high, it can cause the binding of HO• radicals, causing free radical loss (reactions (3), (4), (5), and (8))[8], which can lead to a decrease in color removal efficiency. The results of calculating the parameters of the first-order, second-order, and BMG kinetic models all showed a high correlation R2 (>0.89), Table-5. However, when considering the C_0 values calculated from the first-order and second-order kinetic models, they were very different from the experimental C_0. Specifically, the C_0 calculated from the first-order kinetic model was 50.8; 27.4; and 28.4 mg/L, respectively, and that of the second-order kinetic model was 67.9; 48.2; 71.5 mg/L, respectively, under conditions with H_2O_2 content of 0.25; 0.50 and 1.00%, which was very different from the experimental C_0 (100 mg/L). Meanwhile, the BMG model has a very high R^2 correlation value (~1.0), the 1/b value is also suitable, specifically, 1/b is 0.95, 0.99, and 0.99, respectively corresponding to the experimental efficiency after 30 minutes of 82.8, 93.9, and 94.1%. In which, the 1/b value at H_2O_2 content of 0.5 and 1.0% is not significantly different, Table 5. When considering the 1/m value, representing the reaction rate, it shows that 1/m increases when increasing the H_2O_2 content. However, when increasing the H_2O_2 content from 0.5 to 1.0%, the 1/m value changes insignificantly (0.62 compared to 0.59) according to One-way ANOVA analysis. Therefore, the experimental condition with H_2O_2 content of 0.5% was selected for the following experiments.

Initial Dye Content

The effect of BRS dye content on color removal efficiency under experimental conditions: pH 3, Fe^{2+} 16.8 mg/L, stirring speed 300 rpm, H_2O_2 0.5%, loading rate 1.5 mL/min, and temperature 28°C±1 was presented

in Fig.-5. The results showed that the color removal efficiency in the experiments all occurred in 2 stages: fast in the first 20 minutes and gradually slowed down after that, stopping after 30 minutes.

Table-5: Calculation Results of Parameters of Kinetic Models at H_2O_2 Concentrations

Kinetic model	1st order model			2nd order model			BMG model			
Parameters	R^2	k_1	C_0	R^2	k_2	C_0	R^2	1/b	1/m	E% TN
H_2O_2 0.25%	0.91	0.043	50.8	0.92	0.002	67.9	1.00	0.95^a	0.22^a	82.8
H_2O_2 0.5%	0.89	0.055	27.4	0.96	0.005	48.2	1.00	0.99^b	0.62^b	93.9
H_2O_2 1%	0.94	0.057	28.4	0.96	0.006	71.5	1.00	0.99^b	0.59^b	94.1

Different letters[a,b] in the same column indicate that their values are significantly different.

Specifically, at the first 20 minutes, the color removal efficiency was 79.8, 91.9, and 83.3%, respectively, corresponding to BRS content of 50, 100, and 200 mg/L. After 30 minutes, the color removal efficiency only increased slightly, respectively, to 3.9, 2.0, and 0.5%. One-way ANOVA analysis results also showed that the color removal efficiency occurred in two statistically significant rapid stages (20 min) and increased insignificantly thereafter. The results showed that under the same experimental conditions, the initial C_0 content of 100 mg/L had a higher color removal efficiency than the case of C_0 being 50 and 200 mg/L at all sampling times, Fig.-5. This can be explained by the fact that at low BRS content, the number of color elements participating in the reaction is low, and even less after a period of reaction time. Therefore, to increase the color removal efficiency, more HO• groups are needed to shift the equilibrium. However, because H_2O_2 and the catalyst remained unchanged, the color removal efficiency did not increase further. In the case of continuing to increase the BRS content from 100 to 200 mg/L, the color removal efficiency decreased sharply from 93.9 to 83.3%. This may be due to the excessive dye content, at the same time requiring more HO• radicals. However, under the experimental conditions, the amount of H_2O_2 and Fe^{2+} catalyst remained unchanged, so the color removal efficiency also decreased. A similar explanation was found in the study of Gao *et al.*[25] Similar conclusion was also found in the study of Giwa et al, which stated that color degradation decreased as the dye concentration increased, specifically when the initial concentration of methylene blue (MB) increased from 10 to 50 ppm, the color removal rate decreased from 95.6% to 84.2% after 60 min of reaction.[26]

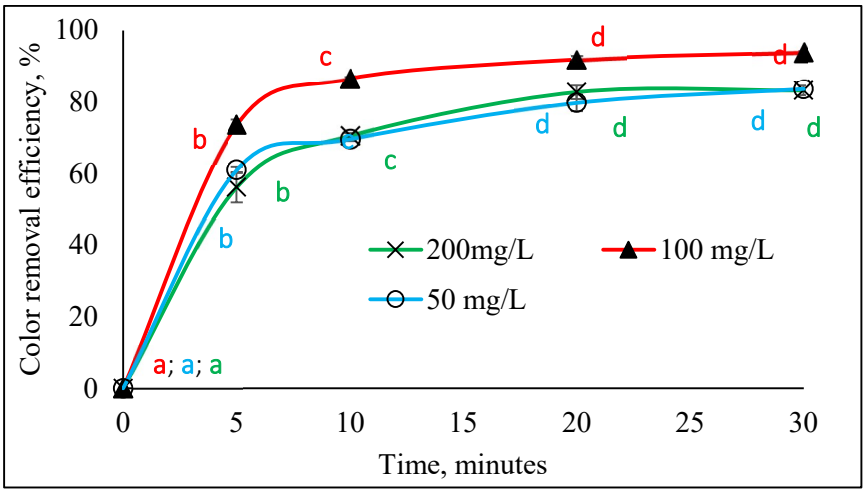

Fig.-4: Performance of Color Removal Efficiency Over Time at BRS Concentrations, The Letters [a,b,c,d] in the Same Color Indicate Statistically Significant Differences)

Table-6: Calculation Results of Model Parameters at BRS Contents

Kinetic model	1st order model			2nd order model			BMG model			
Parameters	R^2	k_1	C_0	R^2	k_2	C_0	R^2	1/b	1/m	E% TN
50 mg/L	0.97	0.035	21.9	0.99	0.0030	26.6	1.00	0.91^a	0.33^a	83.7
100 mg/L	0.94	0.057	28.4	0.96	0.0056	71.5	1.00	0.99^b	0.59^b	94.1
200 mg/L	0.85	0.039	91.4	0.88	0.0008	106.0	1.00	0.93^a	0.30^a	83.3

Different letters[a,b] in the same column indicate that their values are significantly different.

The results of calculating the parameters of the first-order, second-order, and BMG kinetic models all showed a high correlation R2 (>0.85) for all three cases, Table-6. However, when considering the C_0 values calculated from the first-order and second-order kinetic models, they were very different from the experimental C_0. Specifically, the C_0 calculated from the first-order model was 21.9, 28.4, and 91.4 mg/L, respectively, and that of the second-order model was 26.6, 71.5, and 106.0 mg/L, respectively, compared to the experimental C_0 content of 50, 100, and 200 mg/L.

Meanwhile, the BMG model has a very high R2 value (~1.0), and the 1/b value is also suitable, specifically, 1/b is 0.91, 0.99, and 0.93, respectively, corresponding to the experimental removal efficiency after 30 minutes of 83.7, 93.9, and 83.3%. The increase in 1/b at BRS content of 100 mg/L is significantly higher than that of BRS at 50 or 200 mg/L, Table-6 (according to One-way ANOVA analysis).

When considering the 1/m value, representing the reaction rate, it shows that the decolorization rate also gives similar results. The results of the study with the modified Fenton process (continuous H_2O_2 injection) showed that with a BRS content of 100 mg/L, the amount of Fe^{2+} (16.8 mg/L) and H_2O_2 (225 mg/L) consumed in the study was much lower than other studies such as the study of removing Congo Red (30 mg/L CR) and Rhodamine B (50 mg/L RB) which consumed Fe^{2+} (58.1 mg/L) and H_2O_2 (420 mg/L) [21], Anionic Reactive Black 5 dye (120 mg/L) was (25 mg/L) and (240 mg/L) [22], methylene blue (20 mg/L) was (224 mg/L) and (714 mg/L).[26]

CONCLUSION

The study used the modified Fenton process (continuous H_2O_2 injection throughout the experiment) to evaluate the color removal efficiency and determine the color removal mechanism (based on kinetic studies) of the reactive dye Sunzol blue RS 150% (CI reactive B19). The results showed that the BRS color removal process took place in 2 stages: (i) fast in the first stage (20 minutes) and (ii) slowed down thereafter. Thereby, the optimal experimental parameters were determined, including C_0 100 mg/L, pH 3, Fe^{2+} 16.8 mg/L, stirring speed 300 rpm, H_2O_2 0.5%, and injection speed 1.5 mL/min. The BMG model is suitable to explain the BRS color removal mechanism by the modified Fenton process, with R2~1, color removal efficiency 1/b is 99%, and rate coefficient 1/m is 0.59. The research results confirm that the modified Fenton process is more efficient (saving Fe^{2+} and H_2O_2) and the BMG model is suitable to explain the color removal mechanism (2 stages) of the modified Fenton reaction.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interest.

AUTHOR CONTRIBUTIONS

The present work is related to *SDG-6: Clean water and Sanitation*. The author contributed significantly to this manuscript, participated in reviewing/editing and approved the final draft for publication. The research profile of the author can be verified from their ORCID ids, given below:

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