Degradation of Chrysoidine Y Dye with Sulphite Ion in Aqueous Acidic Phase using Kinetic Method of Analysis

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Kinetik Analiz Yöntemi Kullanılarak Sulu Asidik Fazda Sülfit İyonlu Krizoidin Y Boyasının Bozunması

ABSTRACT

Chrysoidine Y dye (CYD) is harmful to aquatic species and human beings, which has the tendency to induce cancer and mutation to living cells. Its degradation is key in creating a healthy environment and curbing pollution. Hence, a stoichiometric method is used to study its degradation with a pool of sulphite ions (SO_3^{2-}), under a constant ionic strength, [H^+], and 449 nm wavelength. The stoichiometry is observed to be 1:1 for CYD: SO_3^{2-}, which results in the formation of aniline and sulphonic acid as the main products of the degradation. The reaction is first-order with respect to CYD, first-order with respect to SO_3^{2-}, and a second-order-overall. Increase in the proton concentration impacts positively on the reaction rate of CYD degradation. Negative salt effect is observed as the dilapidation rate of the dye drops. Occurrence of counter ion catalysis is pronounced with large appreciable rate. The participation of a firm intermediate molecule is negative as revealed by the Spectroscopic Scanning Technique (SST) and Michaelis Menten's Type Plot (MMTP), which cancels the inner-sphere mechanism expectancy. The degradation of the dye was successfully carried out and the reaction points to the outer-sphere mechanism

Keywords: Degradation, Chrysoidine Y, Sulphite ion, Acidic phase, Kinetics

ÖZ

Krizoidin Y boyası (CYD), kansere ve canlı hücrelerde mutasyona neden olma eğiliminde olan su canlıları ve insanlar için zararlıdır. Bozunumu sağlıklı bir çevre yaratmada ve kirliliği azaltmada anahtar rol oynar. Bu nedenle, sabit iyonik güç [H^+] ve 449 nm dalga boyu altında bir sülfit iyonları havuzunda (SO_3^{2-}) bozunmasını incelemek için stokiyometrik bir yöntem kullanılır. Stokiyometrik oranın CYD: SO_3^{2-} için 1:1 olduğu gözlemlenir ve bu da bozunmanın ana ürünleri olarak anilin ve sülfonik asit oluşumuyla sonuçlanır. Tepkime CYD açısından birinci dereceden, SO_3^{2-} açısından birinci dereceden ve genel olarak ikinci derecedendir. Proton konsantrasyonundaki artış, CYD bozunmasının reaksiyon hızını olumlu yönde etkiler. Boyanın bozulma hızı düştükçe negatif tuz etkisi gözlemlenir. Karşı iyon katalizinin oluşumu büyük ve belirgin bir hızla belirgindir. Spektroskopik Tarama Tekniği (SST) ve Michaelis Menten Tip Grafiği (MMTP) tarafından ortaya konulduğu gibi, sağlam bir ara molekülün katılımı negatiftir ve bu da iç küre mekanizması beklentisini iptal eder. Boyanın bozunması başarıyla gerçekleştirildi ve reaksiyon dış küre mekanizmasına işaret etmektedir.

Anahtar Kelimeler: Bozunma, Krizoidin Y, Sülfit iyonu, Asidik faz, Kinetik

INTRODUCTION

Chrysoidine Y dye here after referred to as (CYD) is an azo-dye with nitrogen-nitrogen double bond functional group that is frequently used in the leather, polymer, food, and cosmetic industries.[1] The dye is well known to be resistant to biodegradation as well photo-stability. It is reported to be carcinogenic, mutagenic, and has toxicity which has traits to the environment and human system.[2] CYD removal has been studied with biosorbents produced from row-cork and cork stocked in Ca-alginate bead.[3] It has been demonstrated that oral administration of Chrysoidine Y results in liver-cell adenomas, carcinomas and leukemia in animals.[4] Modified Hummers and co-precipitation approach was employed by Hao et al. [5] to study the removal of CYD with nano-almagamated iron oxide-graphene oxide, as adsorbent. The process was pseudo-second-order compliance and was largely pH dependent, which is in collaboration with the report of Gote et al. [6] where the help of hydrogen peroxide and nickel-iron oxide catalyst, a sono-catalytic method was used to degrade chrysoidine R Also, the rate removal was chemi-sorption influenced, which is a pointer that chemical process can be a better means of degrading CYD dyes. Ensuring a clean environment Mittal and co-scientists[7] studied the removal and recovery of CYD dye with agricultural wastes (bottom ash and oil-free soya). The impact of the tested parameters (pH, contact time, particle size, and adsorbent concentration) had a significant effect on the removal process of the dye. The use of CYD for staining tissues and in textiles can course pollution in water bodies as well as humans and its environment.

Chemical-coagulation approach was deployed by Mariyam and co-workers[8] to study the removal of chrysoidine R, an analogue of CYD Y dye, on an organo-metal framework. The impact of $[H_3O^+]$ and adsorption time were laudable with the implication of chemi-sorption as the rate controlling route. Ashraf et al., [9] contributed to the removal studies of CYD with sawdust and the striking observation was high rate of CYD adsorption in pseudo-second-order fashion. The submission buttressed the far fetching need for chemical degradation of CYD to ensure eco-friendly system.

Sulphite ion is used to reduce chromium (VI) to chromium (III), making it less toxic.[10] Sulphite ion acts as an antioxidant and preservative in foods and beverages to prevent browning and spoilage.[11] In photographic processing, sulphite reduce silver halides thereby helping to develop photographs. Furthermore, it is used in organic synthesis to reduce various functional groups, including ketones and aldehydes. Sulphite ion is equally employed in the pulp and paper industry to bleach wood pulp without the use of chlorine. In dyeing processes, sulphite can help reduce certain dyes and modify their properties. [12-15]

The aim of the study is to contribute to clean environment by exploiting the redox potential species, sulphite ion in the degradation of CYD and explaining its mechanistic pathway.

Much work has not been reported on the degradation of CYD which has prompted this research to see how CYD can be degraded to a less harmful compound.

This study is therefore carried out to see how CYD can be degraded using kinetic method of analysis due to the fact that chemical process is a better means of degrading CYD and this is not well documented in literature.

MATERIALS AND METHODS

Materials

Chrysoidine Y dye and sodium sulphite were obtained from Sigma-Aldrich, Germany. Sodium chloride (BDH, UK) was used to keep the reaction's salt effect constant. Magnesium chloride, potassium chloride, and sodium nitrate (May and Baker, Nigeria) were used to ascertain the effect of counter ions on the reaction rate. Hydrochloric acid (BDH, UK) was used to study the effect of $[H^+]$ on the rate of reaction. Acetone (May and Baker, Nigeria) was used to investigate the effect of solvent polarity (dielectric constant) on the reaction mixture. Bromine water, carbon disulphide, and sodium bicarbonate (BDH, UK) was used for product analysis. Acrylamide with methanol (May and Baker, Nigeria) was used to check the participation of unstable atoms in the reaction. Kinetic investigations were performed with a UV/Visible spectrophotometer (Spectrum Lab 752s) and characterisation of reaction products were carried out qualitatively.

Methods

Maximum Absorbance Determination: The maximum absorbance was determined by running a 0.1 mol dm^{-3} solution in a UV/Visible spectrophotometer in the range of 200 - 800 nm.[9]

Stoichiometry and product determination: The stoichiometry of the reaction was determined spectrophotometrically using the mole ratio method under the reaction conditions [CYD] = 1.0×10^{-4} mol dm^{-3}, $[SO_3^{2-}]$ = $(0.2 - 1.6) \times 10^{-4}$ mol dm^{-3}, $[H^+]$ = 1×10^{-2} mol dm^{-3}, I = 1.0 mol dm^{-3}, T = 30 ± 1 °C and λ_{max} = 449 nm. For a series of solutions containing constant [CYD], the $[SO_3^{2-}]$ (where [] symbolizes concentration) was varied and the absorbance of the reaction mixtures were measured for an interval of eight hours after the initial measurement before arriving at

constant absorbance after twenty hours.[16-20]

The products of the degradation were elucidated by the addition of six drops of bromine water to the product mixture and four drops of carbon disulphide solution.[21] Also, eight drops of sodium bicarbonate solution were added in a separate portion.[22]

Kinetic examination: The reaction order was determined by observing the bleaching of the reaction system as the reaction proceed at a maximum absorbance = 449 nm. The process was done using pseudo-first-order settings with the least concentration of SO_3^{2-} at 200-fold in excess over [CYD] and constant ionic strength of 1.0 mol dm^{-3}. A graph of absorbance (A) against time (s) was done and the observed rate constants, k_1 were obtained as the gradients of the graphs represented by equation 1: [21-24]

$$A_t/A_0 = exp(-k_1t) \qquad 1$$

where A_t and A_0 are the absorbance at the time t, and 0, respectively, k_1 is the observed rate constant and t is the time (seconds). The second-order rate constants, k_2 were determined from equation 2:

$$k_2 = k_1/[SO_3^{2-}] \qquad 2$$

The effect of hydrogen ion concentration was determined at the range of 0.5-3.0 mol dm^{-3}, the salt effect of the reaction medium within the range of 0.5 - 3.0 mol dm^{-3}, and the solvent polarity was varied by the addition of acetone (0.5 - 3.0) cm^3 at constant concentrations of the reactants and temperature 30 ± 1°C.[22-27]

The effect of added counter ions was investigated under a constant concentration of CYD, SO_3^{2-}, ionic strength, and at [ion] = $0.5 – 3.0 × 10^{-3}$ mol dm^{-3}. [28-30]

The investigation of the existence of unstable atoms in the reaction mixture was carried out by adding 0.2 cm^3 of acrylamide solution to an incompletely reacted CYD-SO_3^{2-} mixture and excess of methanol. [31-33]

The intermediate complex was examined spectroscopically by scanning the partially reacted mixture at wavelength 200–800 nm, and applying the Michaelis–Menten's plot of k_1^{-1} against $[SO_3^{2-}]^{-1}$. [34-36]

RESULTS

Maximum Absorbance of CYD: The maximum absorbance of CYD is observed at 449 nm.

Stoichiometry and Product Determination: A mole ratio of 1:2 (CYD: SO_3^{2-}) is obtained for the reaction between CYD: SO_3^{2-} (Figure 1). Indicative of 4e- contribution

from the sulphite ion for the breakdown of a mole of CYD and it is buttressed by equation 3.
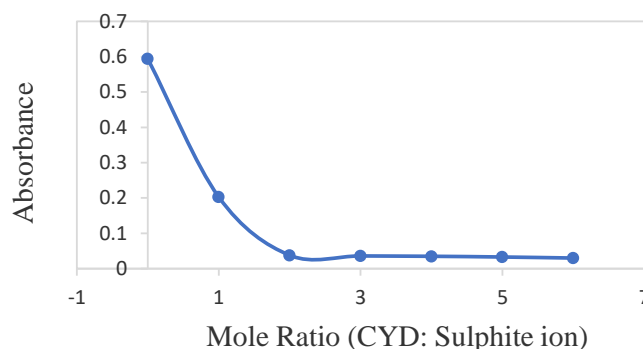

Figure 1: Plot of absorbance versus $[SO_3^{2-}]$ for the determination of the stoichiometry of CYD - SO_3^{2-} System.

On the addition of six drops of bromine water to the product mixture, the mixture turned to a white precipitate, indicating the deposit of aniline, which was confirmed by a mustard oil smell when four drops of carbon disulphide solution was added to a separate portion of the product mixture. Also, the sulphonic acid group was confirmed by the release of effervescence in the form of bubbles on the addition of eight drops of sodium bicarbonate solution to a portion of the product mixture.

Kinetic Examination

The reaction resulted in a first order with respect to the [CYD] due to consecutive linearity of the graphs of log $(A_t – A_∞)$ against time and a typical one is shown in Figure 2, and first - order with respect to the concentration of SO_3^{2-} as proven by the logarithmic plot of k_1 against $[SO_3^{2-}]$ that is one dimensional with a slope 0.9971 and a correlation coefficient R^2 = 0.9995 (Figure 3), which is reinforced by the consistency of the k_2 in Table 1.

The rate of the reaction is positively affected as the concentration of acid is increased for CYD – SO_3^{2-}. This result is contrary to the report of Edokpayi et al.[37]; Imam et al. [38] where increase in [H+] had no effect on rate of reaction between indigo carmine and sulphite ion; malachite green with sulphite ion systems, respectively.

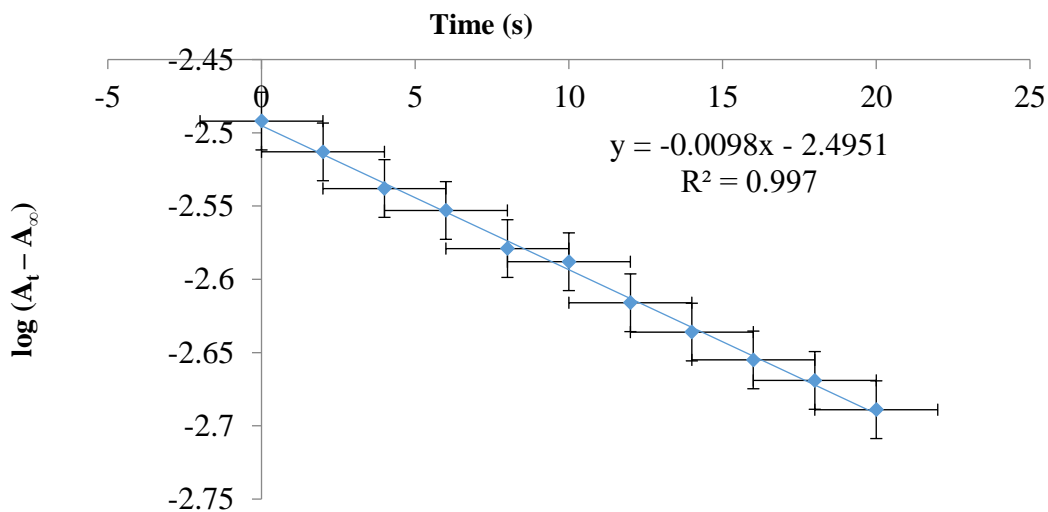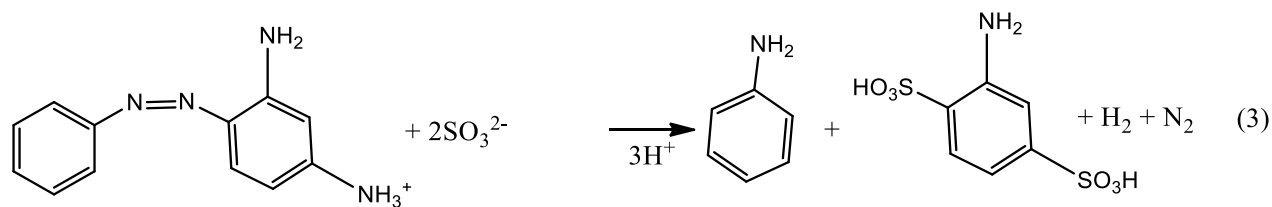$$+ 2SO_3^{2-} \xrightarrow[3H^+]{} \text{(aniline)} + \text{(sulfonated product)} + H_2 + N_2 \quad (3)$$

Figure 2: Graph of the logarithm plot of $(A_t - A_\infty)$ against time for the degradation of CYD by SO_3^{2-}. [CYD] = 1.0×10^{-4} mol dm^{-3}, [SO_3^{2-}] = $(2.0 - 10) \times 10^{-2}$ mol dm^{-3}, [H$^+$] = 1×10^{-2} mol dm^{-3}, I = 1.0 mol dm^{-3}, T = 30 ± 1 °C, and λ_{max} = 449 nm.
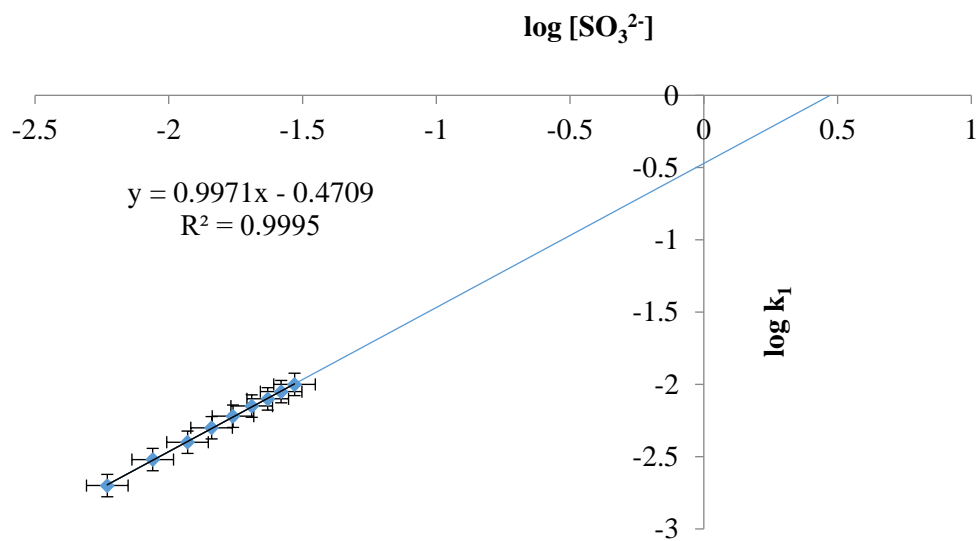
Figure 3: Logarithm plot of k_1 against [SO_3^{2-}] for the degradation of CYD by SO_3^{2-}. [CYD] = 1.0×10^{-4} mol dm^{-3}, [SO_3^{2-}] = $(2.0 - 10) \times 10^{-2}$ mol dm^{-3}, [H$^+$] = 1×10^{-2} mol dm^{-3}, I = 1.0 mol dm^{-3}, T = 30 ± 1 °C, and λ_{max} = 449 nm.

Table 1: Kinetics constant for the degradation of CYD by SO_3^{2-}. [CYD] = 1.0×10^{-4} mol dm^{-3}, [SO_3^{2-}] = $(2.0 - 10) \times 10^{-2}$ mol dm^{-3}, T = 30 ± 1 °C, and λ_{max} = 449 nm.

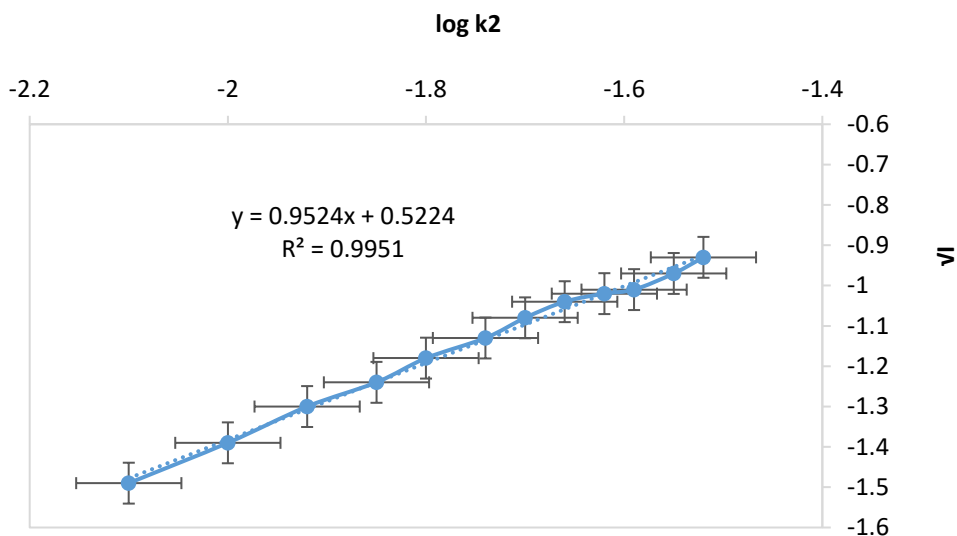| 10^2 [SO_3^{2-}], mol dm^{-3}	I, mol dm^{-3}	10^2[H$^+$], mol dm^{-3}	$10^2 k_1$, s^{-1}	k_2, dm^3 mol^{-1} s^{-1}
2.0	1.0	1.0	2.26	1.13
3.0	1.0	1.0	3.40	1.13
4.0	1.0	1.0	4.60	1.15
5.0	1.0	1.0	5.70	1.14
6.0	1.0	1.0	6.90	1.15
7.0	1.0	1.0	8.00	1.14
8.0	1.0	1.0	8.90	1.11
9.0	1.0	1.0	10.00	1.11
10.0	1.0	1.0	11.00	1.10
3.0	1.0	0.5	3.40	1.13
3.0	1.0	1.0	3.42	1.14
3.0	1.0	1.5	3.64	1.21
3.0	1.0	2.0	4.10	1.37
3.0	1.0	2.5	4.30	1.43
3.0	1.0	3.0	5.30	1.77
3.0	0.5	1.0	3.67	1.22
3.0	1.0	1.0	3.42	1.14
3.0	1.5	1.0	3.22	1.07
3.0	2.0	1.0	3.05	1.02
3.0	2.5	1.0	2.89	0.96
3.0	3.0	1.0	2.71	0.90

log k2

y = 0.9524x + 0.5224
R² = 0.9951

Figure 4: Graph of salt effect of k_2 versus $\sqrt{I}$ for the degradation of CYD by SO_3^{2-}. [CYD] = 1.0×10^{-4} mol dm^{-3}, [SO_3^{2-}] = 3.0×10^{-2} mol dm^{-3}, [H$^+$] = 1.0×10^{-2} mol dm^{-3}, T = 30 ± 1 °C, and λ_{max} = 449 nm.

The overall rate equation can be represented by equations 4 as follows:

$$-\frac{d[CYD]}{dt} = [CYD][SO_3^{2-}] \qquad 4$$

A negative salt effect is observed in the redox reaction as the rate of reaction is found to decrease with increase in the ionic strength of the reaction medium (Figure 4). The observed slope of less than one could be due to formation of ion – pair during the reaction. The negative Brønsted – Debye salt effect observed indicates that the species at the activated complex are of unlike charges.

The effect of change in dielectric constant is studied by using a binary solvent mixture of water and acetone (0.5 - 3.0) cm^3. It is observed that as the concentration of acetone increased, the rate of reaction increased for CYD - SO_3^2 (Table 2).

Added counter ions (Mg^{2+}, K$^+$, and NO$_3^-$) increased rate of reaction as the concentration increased (Table 3). Ibrahim et al. [39], Abdulsalam et al. [40], Oladunni et al. [41] reported that added ions are expected to catalyse reactions occurring by the outer-sphere mechanism. As such, this reaction is most probably in the favour of an outer – sphere mechanism.

The addition of 2 cm^3 acrylamide solution to the partially reduced CYD mixture shows no gel formation even in large excess of methanol, suggesting the probable absence of free radicals in the reaction systems.

The plot of k_1^{-1} versus $[SO_3^{2-}]^{-1}$ gives a straight line, which passes through the origin (Figure 5). This suggests absence of intermediate complex formation prior to redox reaction. In addition, the result of the spectroscopic study indicates no significant shift from the absorption maxima of λ_{max} = 449 nm. This suggests the absence of the formation of an intermediate complex in the reaction.

Table 2: Effect of system permittivity on the reaction rate for the degradation of CYD by SO_3^{2-}. [CYD] = 1.0×10^{-4} mol dm^{-3}, $[SO_3^{2-}]$ = 3.0×10^{-2} mol dm^{-3}, [H$^+$] = 1.0×10^{-2} mol dm^{-3}, I = 1.0 mol dm^{-3}, T = 30 ± 1 ^{o}C, and λ_{max} = 449 nm.

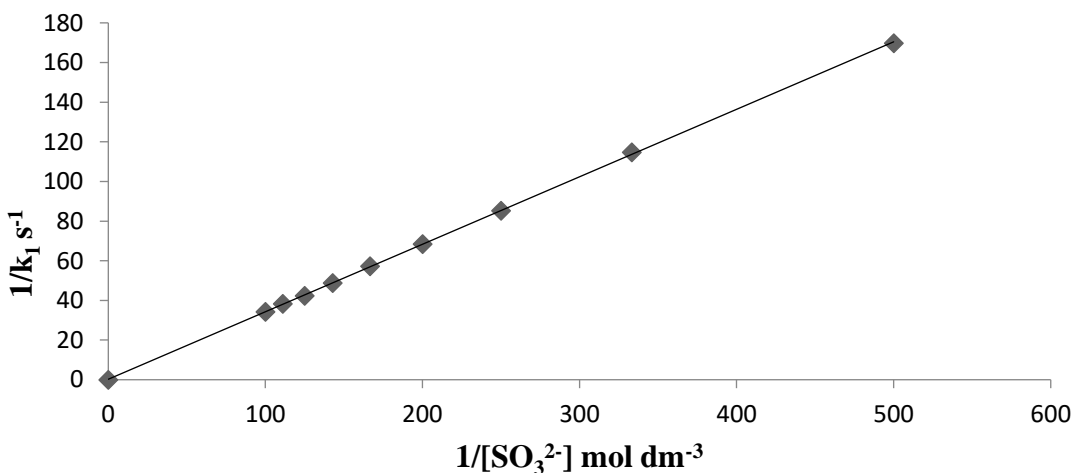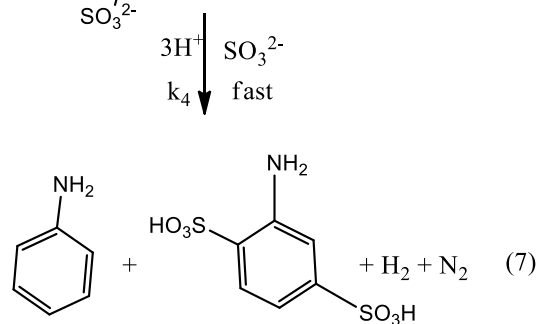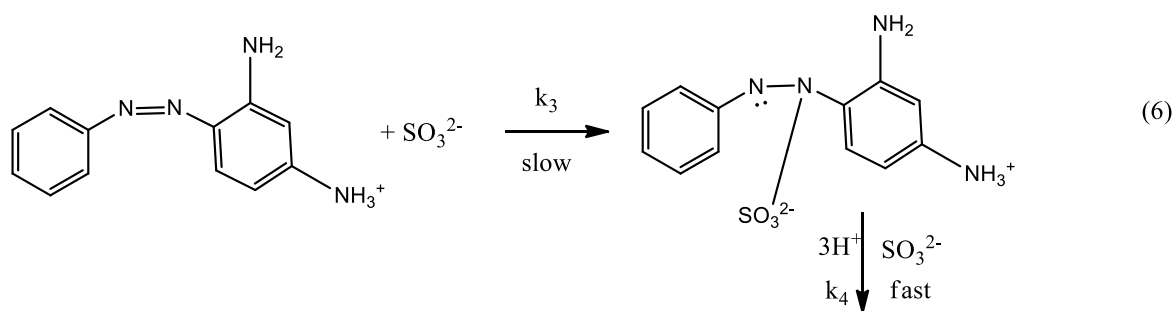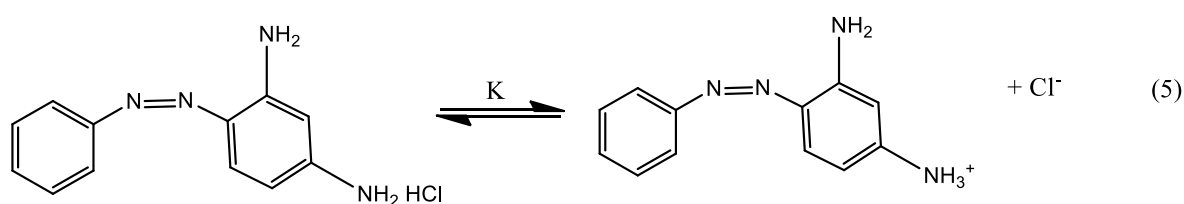| Acetone-H$_2$O (cm^3)	$10^1 k_1$, s^{-1}	k_2, dm^3 mol^{-1} s^{-1}
0.5	7.83	2.61
1.0	8.73	2.91
1.5	9.65	3.22
2.0	10.57	3.52
2.5	11.56	3.85
3.0	12.43	4.14

Figure 5: Plot of k_1^{-1} versus $[SO_3^{2-}]^{-1}$ for the degradation of CYD by SO_3^{2-}.

Table 3: Effect of counter ions on the reaction rate for the degradation of CYD by SO_3^{2-} [CYD] = 1.0×10^{-4} mol dm^{-3}, [SO_3^{2-}] = 3.0×10^{-2} mol dm^{-3}, [H^+] = 1.0×10^{-2} mol dm^{-3}, I = 1.0 mol dm^{-3}, T = 30 ± 1 $^{\circ}$C and λ_{max} = 449 nm.

Ion	10^3 [Ion], mol dm^{-3}	$10^3 k_1$, s^{-1}	k_2, dm^3 mol^{-1} s^{-1}
Mg^{2+}	0.5	7.59	2.53
	1.0	8.74	2.91
	1.5	9.89	3.30
	2.0	11.04	3.68
	2.5	12.19	4.06
	3.0	13.34	4.45
K$^+$	0.5	7.74	2.58
	1.0	8.73	2.91
	1.5	9.72	3.24
	2.0	10.70	3.57
	2.5	11.67	3.89
	3.0	12.65	4.22
NO$_3^-$	0.5	7.29	2.43
	1.0	8.73	2.91
	1.5	10.18	3.39
	2.0	11.63	3.88
	2.5	13.05	4.35
	3.0	14.49	4.83

Reaction Mechanism:

Following the outcome of the kinetic evidence from the effect of ionic strength, acid, added ions, MMTP, and absence of stable and detectable free-radicals, an outer-sphere mechanism (equation 5 - 7) is considered.

DISCUSSION

The study of degradation of CYD with SO_3^{2-} ion using a kinetic method of analysis portrays a sustainable and efficient approach in determining the pathway of the degradation with rate input.

A maximum absorbance of 449 nm was obtained for this research, Ashaf et al.[9] reported the same wavelength and 452 nm was reported by Gote et al.[6] for sonocatalytic degradation of chrysoidine R dye, an analogue of CYD, using ultrasonically synthesized $NiFe_2O_4$ catalyst.

A contrary stoichiometry of 2: 1 was observed for the reaction between [Mn(cydta)(OH,]$^-$ and SO_3^{2-} as reported by Chandrawat et al.[42] for the Kinetics and mechanism of the oxidation of the sulphite ion by Mn(lll)-cydta complex ion .

The order of the degradation is first order with respect to the concentrations of the two reacting species (CYD and SO_3^{2-}) indicating second- order kinetics. A similar order was observed for the degradation of chrysoidin R dye[6].

Acid medium increased the rate of reaction positively which is contrary with the result of [37,38] but is however in agreement with the report of.[42]

The ionic strength accounts opposite charged species interaction which is also contrary to the report of [42] where similar charges are in operation during the redox reaction.

Free radicals are absent in the process added added ions had effect on the rate of reactions. Thus, the result is suggestive of an outer-sphere mechanism degradation process. These results are contrary to the results reported by Umoru, Babatunde 2019[43] for the Kinetics and Mechanism of the Oxidation of Sulphite Ion by Di-μ-oxo-tetrakis (2, 2' bipyridine)-Dimanganese (III, IV) Perchloratein Aqueous Acidic Medium.

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