

Kinetic, Mechanistic & Computational Investigation of Oxidation of Sulfadiazine by Peroxymonosulphate in Acidic Medium

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ABSTRACT

Antibiotics are a novel class of chemical compounds exhibiting antibacterial and anti-inflammatory properties in clinical settings; still, their widespread use has serious adverse effects on the natural water environment. Sulfadiazine (SDZ) is a common antibiotic, and the treatment discusses the significance of studying sulfadiazine oxidation for environmental and health impacts and highlights the role of strong oxidizing agent peroxymonosulfate (PMS) in degrading pollutants.

The kinetics and mechanism of sulfadiazine oxidation by peroxymonosulfate have been examined, and the species of peroxymonosulfate are evaluated to clarify the role of activated species. A plausible reaction mechanism is presented, and the resulting rate law is consistent with all experimental observations. The activation parameters, specifically the activation energy and entropy, have been determined to be 12.912 kJ mol⁻¹ and -231.44 J K⁻¹ mol⁻¹, respectively, using the Eyring plot method. FTIR examination of the oxidation product demonstrated the formation of oxygenated sulfadiazine derivatives while keeping the aromatic sulfonamide framework. Density functional theory (DFT) simulations showed that sulfadiazine functions as an electron donor (HOMO = -6.72 eV), whereas PMS acts as an electron acceptor (LUMO = -2.45 eV), indicating a HOMO-LUMO mediated oxidation pathway.

Keywords: Sulfadiazine, kinetics, oxidation, mechanism, peroxymonosulfate, acidic medium, computational.

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1. INTRODUCTION

Chemical kinetics, often known as reaction kinetics, is a field of physical chemistry concerned with understanding the speeds of chemical reactions. It is different from chemical thermodynamics, which deals with the direction in which a reaction happens but says nothing about its rate. Chemical kinetics covers examinations of how experimental conditions affect the pace of a chemical reaction and offer information about the reaction mechanism and transition states.

The word “chemical kinetics” is derived from the Greek word “kinesis,” which means movement. Thermodynamics just talks about the feasibility of a reaction, but chemical kinetics talks about the rate of a reaction. It entails measuring reaction rates and analyzing experimental data to build a systematic collection of information that highlights all the quantitative kinetic details for every given reaction.

Kinetic research ascertains the shelf life of a drug under diverse environmental circumstances. It can facilitate the prediction of specific essential aspects that can alter the quality of medication during storage. Kinetic studies are important in stability analysis and guide the selection of a logical method for identifying drugs' kinetic behavior. Properly designed kinetic studies will help establish precise, robust conditions for pharmaceuticals.

Degradation kinetics is the study of the rate of drug degradation. Data gathered from degradation kinetics can be utilized to build a better understanding of the process behind drug degradation.

Sulfonamide antibiotics constitute an important class of antimicrobial agents that exert their therapeutic action by inhibiting bacterial folate biosynthesis¹. These compounds act as structural analogues of p-aminobenzoic acid and competitively inhibit the enzyme dihydropteroate synthase, thereby disrupting the formation of dihydropteroic acid, a crucial precursor in the synthesis of folic acid, purines, and nucleic acids. Owing to their broad-spectrum antibacterial activity, sulfonamides have been extensively used in both human medicine and veterinary practice for the treatment and prevention of bacterial infections².

Among the sulfonamide antibiotics, sulfadiazine is widely employed and is frequently detected in environmental compartments, particularly in surface water, groundwater, and wastewater systems³. The continuous discharge of sulfadiazine from pharmaceutical, agricultural, and livestock sources has raised significant environmental concerns due to its persistence, potential ecotoxicity, and contribution to the development of antibiotic-resistant microorganisms. Consequently, the occurrence of sulfadiazine in aquatic environments has become an

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emerging issue in water quality management ^{4,5}.

Advanced oxidation processes have attracted considerable attention as effective strategies for the degradation and removal of persistent pharmaceutical contaminants ⁶. In such systems, sulfadiazine functions as an oxidizable substrate rather than as an oxidizing agent. Investigating the kinetics and mechanisms of its oxidation by various oxidants, including ozone, permanganate, peroxymonosulfate, and persulfate, is essential for understanding its environmental fate and transformation behavior ^{7,8}. Kinetic studies provide valuable information regarding the influence of reaction parameters such as pH, temperature, oxidant concentration, and ionic strength on the degradation efficiency. Such information is critical to optimizing water treatment technologies aimed at the effective removal of sulfadiazine from contaminated water matrices. Furthermore, mechanistic insights into the oxidation pathways contribute to the prediction of transformation products and assessment of their potential environmental impacts, thereby supporting the

development of sustainable and efficient remediation strategies ^{9,10}.

The degradation of refractory organic compounds to harmless matter is one of the major concerns of environmentalists. Advanced oxidation processes are promising technologies producing the hydroxyl and sulfate radicals for pollutants degradation ^{11,12}. Recently, much attention has been paid to producing sulfate radicals ¹³. Now days, the use of PMS has been acquired popularity due to its high reactivity and high potential in generating sulfate radicals. It is becoming an alternative for hydrogen peroxide and persulfate. PMS is an unsymmetrical oxidant which can be activated to produce both hydroxyl and sulfate radicals. Sulfadiazine (SDZ), antibiotics, can be effectively degraded using peroxymonosulfate (PMS)-based advanced oxidation processes ¹⁴. This process is considered a promising alternative to conventional methods for removing recalcitrant pollutants like SDZ from environmental ^{15,16,17}.

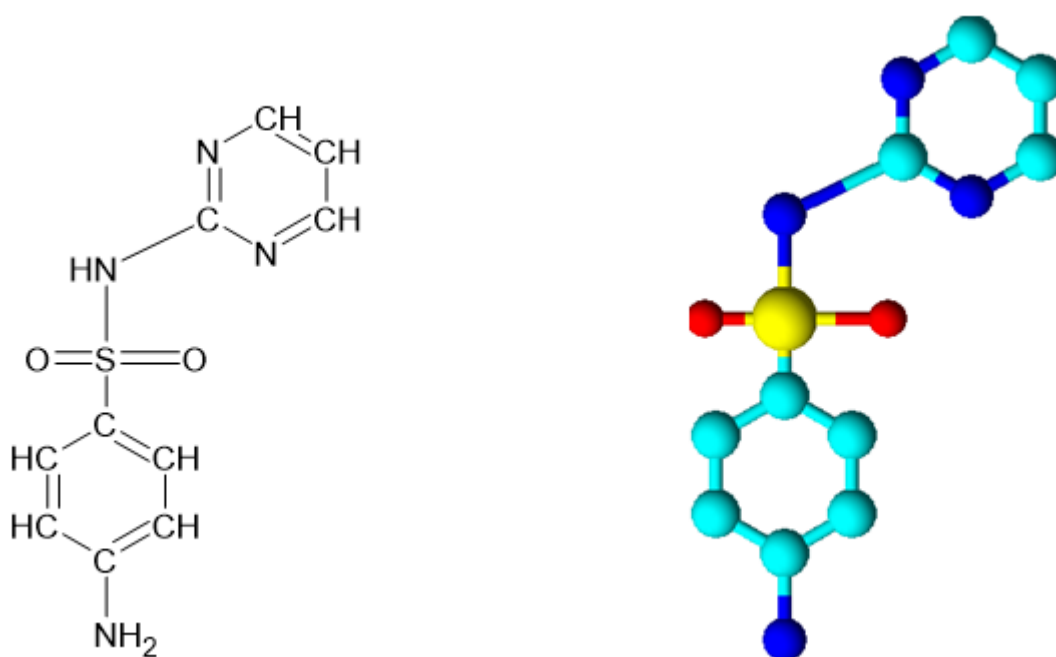


Fig. 1 Structure Diagram of Sulphadiazine.

As a typical sulfonamide antibiotic, sulfadiazine, with the structure diagram shown in Figure 1, has vigorous antibacterial activity and is widely used in animal

husbandry, so it is frequently detected in natural water ^{18,19}. The frontier molecular orbital values are shown in table 1.

Table-1 Frontier Molecular Orbitals for the Sulphadiazine.

Property	Value(eV)
HOMO	-6.72
LUMO	-2.09
HOMO-LUMO Gap	4.63

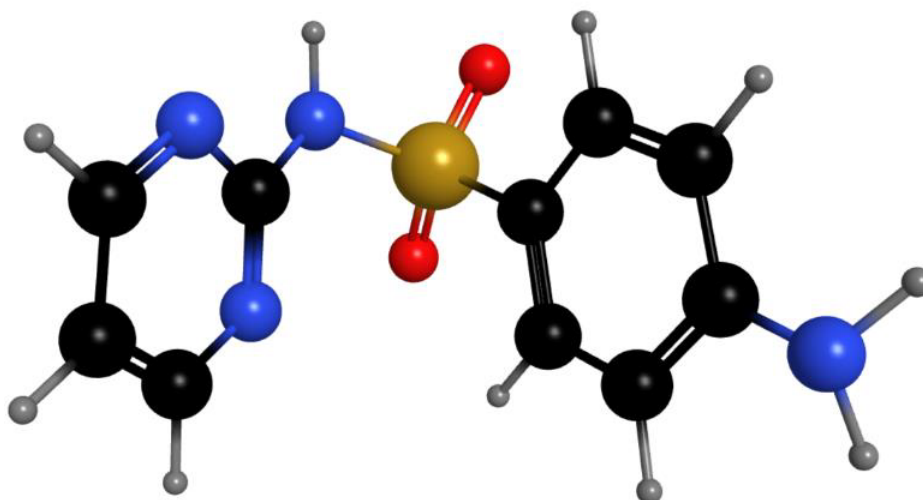


Figure 2. Sulfadiazine (SDZ) structure diagram (black represents C-atom, blue represents N-atom, red represents O-atom, gray represents H-atom, and yellow represents S-atom).

Peroxymonosulphate

Peroxymonosulfate (PMS) is a versatile and potent oxidant that has been extensively utilized in environmental remediation, particularly for water purification and disinfection. It is most typically utilized in the form of its potassium salt ($2\text{KHSO}_5 \cdot \text{KHSO}_4 \cdot \text{K}_2\text{SO}_4$, commercially known as Oxone). The efficiency of PMS in the removal of organic pollutants from wastewater stems from its capacity

to generate extremely reactive oxygen-centered radicals, such as sulfate radicals ($\text{SO}_4^{\bullet-}$) and hydroxyl radicals ($\bullet\text{OH}$), which possess exceptionally high oxidation potentials. These reactive species enable rapid and non-selective destruction of a wide spectrum of resistant organic contaminants, making PMS-based methods suitable for advanced oxidation applications.

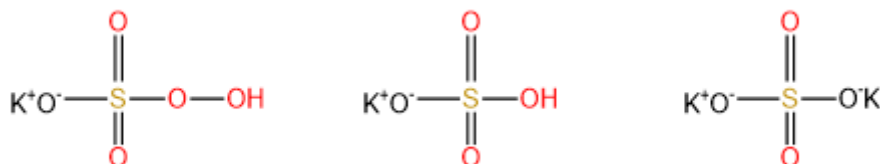


Fig. 3. Molecular structure of Oxone salt ($2\text{KHSO}_5 \cdot \text{KHSO}_4 \cdot \text{K}_2\text{SO}_4$)

Peroxomonosulfate, a hydrolytic derivative of peroxodisulfate, is regarded as a superior oxidant compared to its parent analogue, despite the significantly lower redox potential of the peroxomonosulfate–sulfate redox couple relative to that of the peroxodisulfate–sulfate redox couple.

Peroxomonosulfate serves as a possible oxidant, with several kinetic investigations documented in both acidic and alkaline environments [4–18], and its HOMO-LUMO is represented in fig. 4, and its gap is represented in fig. 5.



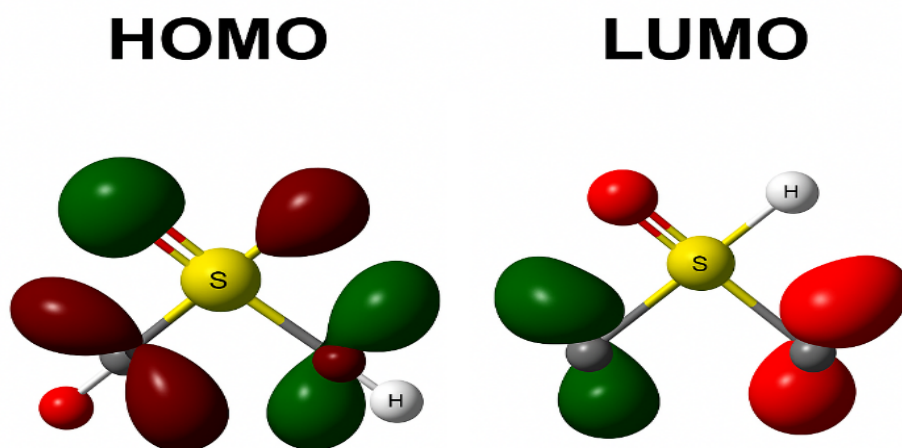


Fig. 4 HOMO-LUMO representation of peroxymonosulfate.

HOMO–LUMO Energy Diagram Peroxymonosulphate

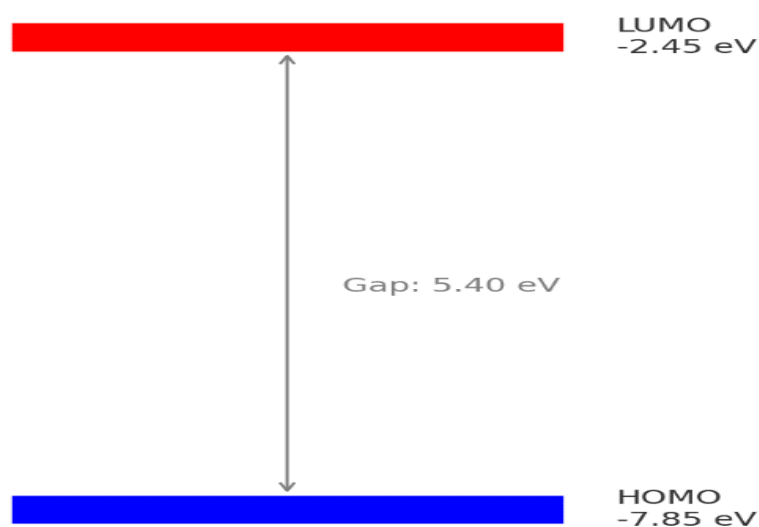
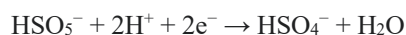


Fig. 5 HOMO-LUMO Gap.

2. EXPERIMENTAL

In the present investigation, peroxymonosulfate (PMS) acts as a powerful oxidizing agent. Although PMS acts as an oxidizing agent in an alkaline medium, for quantitative analysis, mostly acidic medium is used. The oxidizing action of (PMS) in the acidic medium can be represented by the following equation ²⁰.



Chemicals and materials: Peroxymonosulfate is a triple salt composed of (2KHSO₅.KHSO₄. K₂SO₄) and is commercially referred to as “oxone.” It is a monosubstituted derivative of hydrogen peroxide. Peroxymonosulfate (Aldrich) was utilized as provided, and its concentration was verified iodometrically with 96% peracid content. Sulfadiazine of 99% purity was purchased

from Sigma-Aldrich Co. LLC (St. Louis, MO, USA). All additional reagents utilized in this investigation were of either AR or GR grade and were used as received. Doubly distilled water was utilized throughout the investigation; the second distillation was performed from an alkaline permanganate solution in a fully glass commercial apparatus ^{21, 22, 23}.

Kinetic procedure

The reaction kinetics were assessed by iodometric assay of peroxymonosulfate. The reactions were conducted in glass-stoppered corning containers, maintained at a temperature of $\pm 0.1^\circ\text{C}$ in a water bath, unless stated otherwise. The reaction commenced with the addition of a temperature-equilibrated solution of peroxymonosulfate (PMS), with the initiation time documented when half of the pipette's contents were dispensed into the reaction

mixture. A 5 cm³ aliquot of the reaction mixture was periodically extracted and subsequently introduced into a freshly produced 10% potassium iodide solution. The released iodine was titrated with thiosulfate solution, using starch as an indicator^{24, 25}.

3. RESULTS AND DISCUSSION

3.1 Effect of Temperature: - In this reaction, the effect of temperature variation has been investigated on the rate of reaction as shown in Figure 6. The rate constant increases with increasing temperature. The relationship between rate constant (k) and temperature (T) has been described by the Eyring plot.

The activation parameters, specifically the activation enthalpy and entropy, have been determined to be 12.912 kJ mol⁻¹ and -231.44 J K⁻¹ mol⁻¹, respectively, using the

Eyring plot method. At 40 °C, the Gibbs free energy of activation was calculated to be 85.36 kJ mol⁻¹.

The low activation energy (12.912 kJ mol⁻¹) suggests that the oxidation of sulfadiazine by peroxymonosulphate occurs via a kinetically favourable pathway, aligning with an outer-sphere electron transfer mechanism rather than oxidation governed by bond cleavage. The substantial negative entropy of activation (-231.44 J K⁻¹ mol⁻¹) indicates the development of a highly ordered transition state, characterized by robust electrostatic contacts and solvent structuring between the protonated sulphadiazine and HSO₅⁻. These conditions suggests that the reaction is mostly entropy-controlled, with the rate determined by the establishment of a tightly bound encounter complex prior to electron transfer.

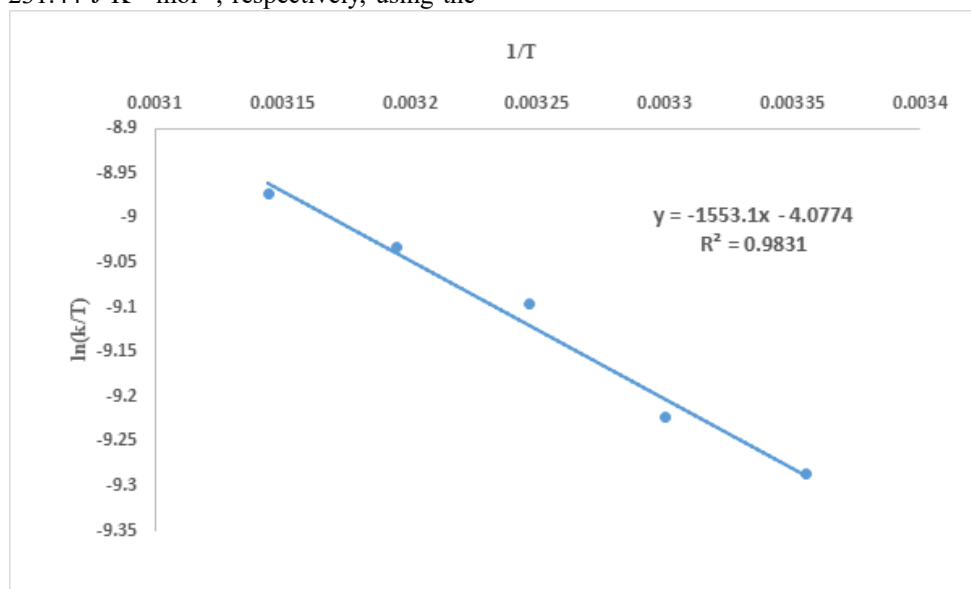


Figure 6. Effect of Temperature on degradation of SDZ by oxidant PMS.

Temperature= 298K, 303K, 308K, 313K, 318K,
[SDZ] = 0.02M [PMS]= 0.002M, HClO₄=0.02M.

3.2. Effects of Sulfadiazine (SDZ) Concentration: In this investigation the effect of sulfadiazine variation,

respectively, on the rate of reaction is shown in Figure 7. An increasing effect has been indicated with increasing concentrations of sulfadiazine.

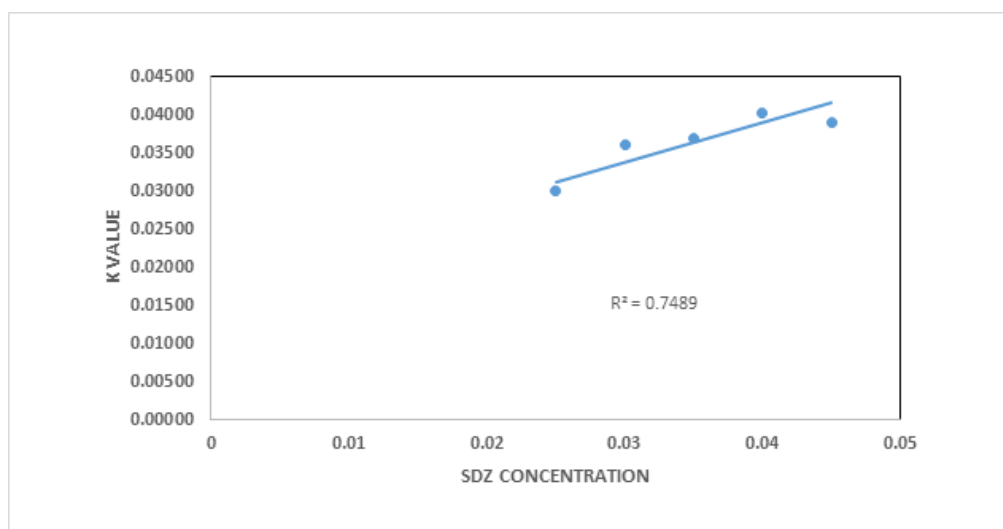


Figure 7. The effects of different [SDZ] processes.

SDZ = 0.025, 0.03, 0.035, 0.04, 0.045M

[HClO₄] = 0.02, [PMS] = 0.002M, Temp = 313 K,

the rate of reaction using sodium thiosulphate as shown in Figure 8. It has increasing rate of reaction with concentration of HClO₄.

3.3. Effects of HClO₄ Variation: - In this reaction we studied the effect of HClO₄ variation respectively on

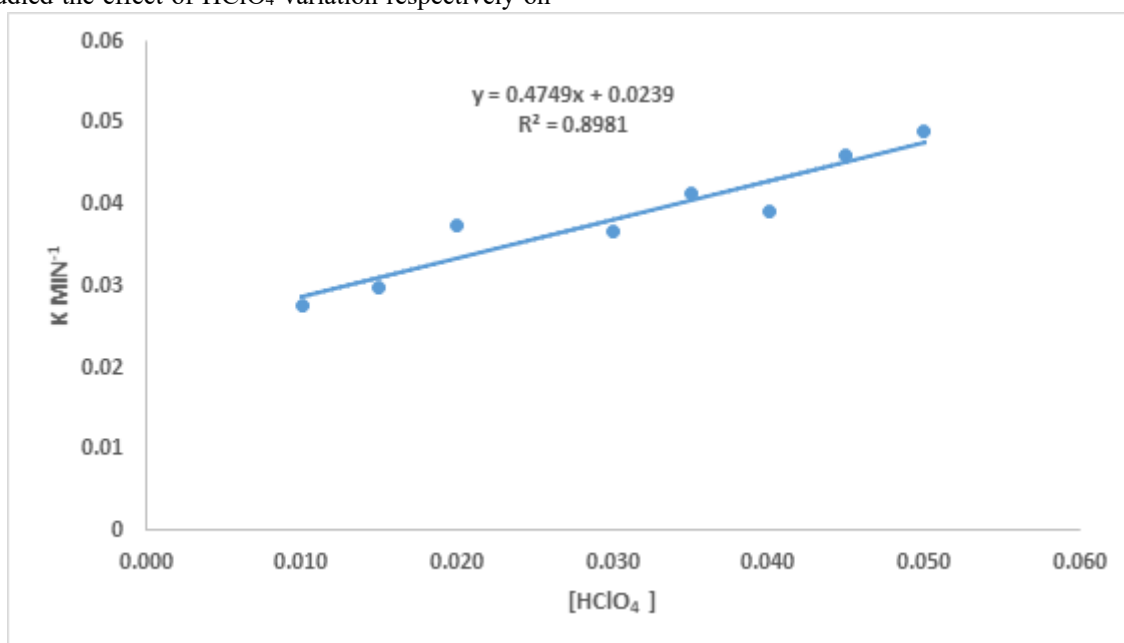


Figure 8. The effects of different HClO₄ processes

[SDZ] = 0.02M, Temperature = 313 K, HClO₄ = 0.01M, 0.015M, 0.02M, 0.03M, 0.035M, 0.04M, 0.045M, 0.05M, [PMS] = 0.002M

thiosulfate is shown in Figure 9. The degradation rate of SDZ increased with increasing PMS concentrations under investigation.

3.4. Effects of Peroxymonosulfate: The effect of PMS variation studied on the rate of reaction using sodium

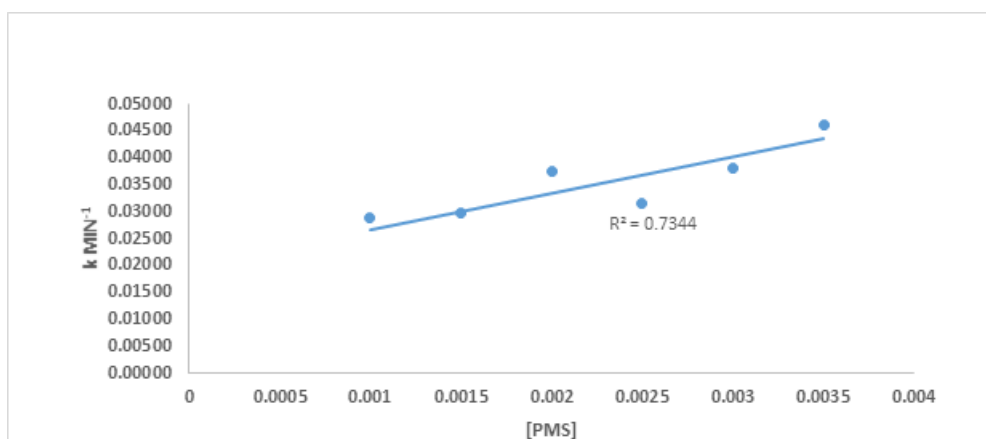


Figure 9. The effects of different oxidant dosages

[PMS] = 0.001, 0.0015, 0.002, 0.0025, 0.003, 0.0035M at
[SDZ] = 0.02M, [HClO₄] = 0.02M, Temp. = 313K

(SDZ) with peroxymonosulfate (PMS) is primarily related to its influence on ionic strength as represented in fig. 10.

3.5. Variation of NaClO₄ :-

The ionic effect of NaClO₄ (sodium perchlorate) on the reaction of sulfadiazine

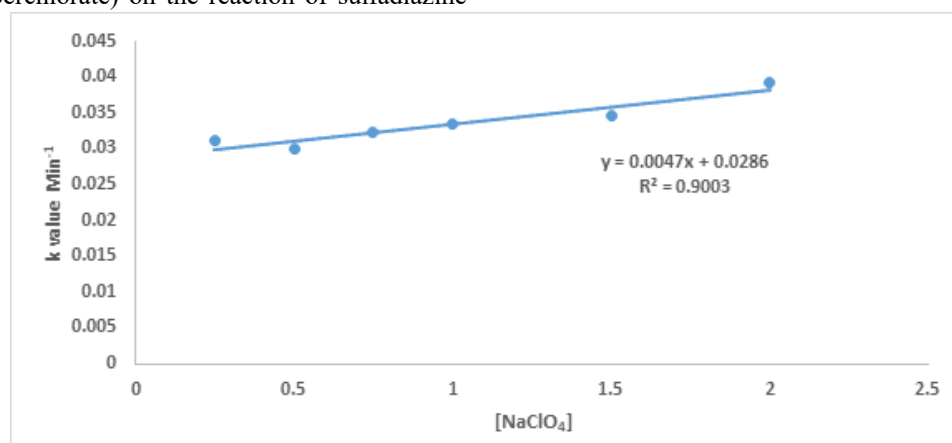
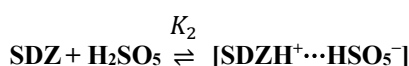
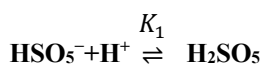


Figure 10. The effects of different concentrations of NaClO₄

[PMS] = 0.002, SDZ = 0.02M, HClO₄ = 0.02M, Temp = 313 K.

4. DISCUSSION

The oxidation of sulfadiazine by peroxymonosulfate in acidic medium shows an increasing rate with [SDZ], [PMS], [HClO₄], and [NaClO₄]. This indicates that the reaction proceeds through an acid-assisted ionic pathway involving a more reactive protonated oxidant species and an ordered activated complex.



Activated Complex



$$\text{Rate} = k_2 K_1 K_2 [\text{SDZ}] [\text{HSO}_5^-] [\text{H}^+]$$

In an acidic medium, peroxymonosulfate exists mainly as HSO_5^- . On increasing perchloric acid concentration, HSO_5^- becomes protonated to form the more reactive peroxomonosulfuric acid species:

The increase in rate with $[\text{HClO}_4]$ suggests that H_2SO_5 is the active oxidizing species. This protonated PMS species is more electrophilic and can oxidize sulfadiazine more easily, and Sulphadiazine forms an intermediate complex with the activated oxidant:

This complex formation is supported by the positive dependence on sulfadiazine concentration. The increasing rate with PMS concentration also shows that the oxidant participates directly in the rate-determining step.

The positive effect of sodium perchlorate is also significant. Since NaClO₄ does not chemically oxidize the substrate, its role is mainly to increase the ionic strength of the medium. The rate enhancement with NaClO₄ indicates

that the transition state is more polar or more charge-separated than the reactants. Therefore, higher ionic strength stabilizes the activated complex and increases the rate.

The HOMO–LUMO values of sulfadiazine confirm its role as an electron donor in the oxidation reaction. The HOMO–LUMO gap of sulfadiazine, 4.63 eV, also implies moderate molecular reactivity. The HOMO–LUMO energy values of peroxymonosulfate provide additional evidence for the suggested oxidation mechanism. The comparatively wide HOMO–LUMO gap of PMS (5.40 eV) shows that PMS is stable in the absence of activation;

consequently, protonation in acidic media is required to boost its electrophilic nature. The low-lying LUMO of PMS (-2.45 eV) suggests its ability to accept electron density from the electron-rich centers of sulfadiazine. The HOMO of sulfadiazine (-6.72 eV) can interact with the low-lying LUMO of PMS (-2.45 eV), yielding a donor–acceptor energy differential of roughly 4.27 eV. Thus, the reaction may proceed through a {HOMO-(SDZ) → (LUMO-PMS)} interaction in the encounter complex. This facilitates electron transport from sulfadiazine to PMS during the rate-determining step. These data also indicate an acid-assisted electron/oxygen-transfer process involving a highly organized transition state.



Agilent Technologies

Sample ID: pms+sdz
Sample Scans: 32
Background Scans: 32
Resolution: 8
System Status: Good
File Location: C:\Users\Public\Documents\Agilent\MicroLab\Results\pms+sdz_2025-11-19T14-16-16.a2r

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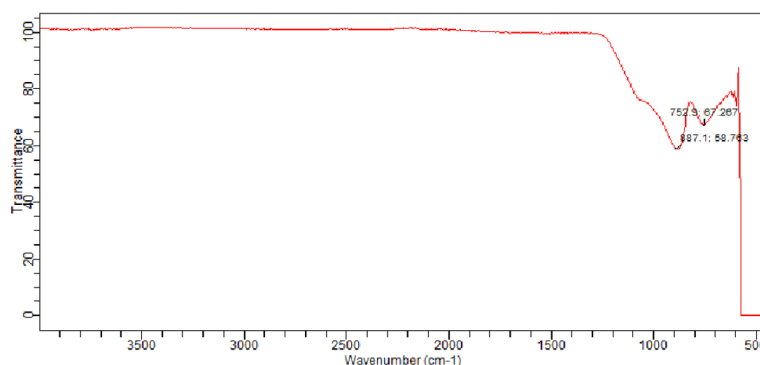
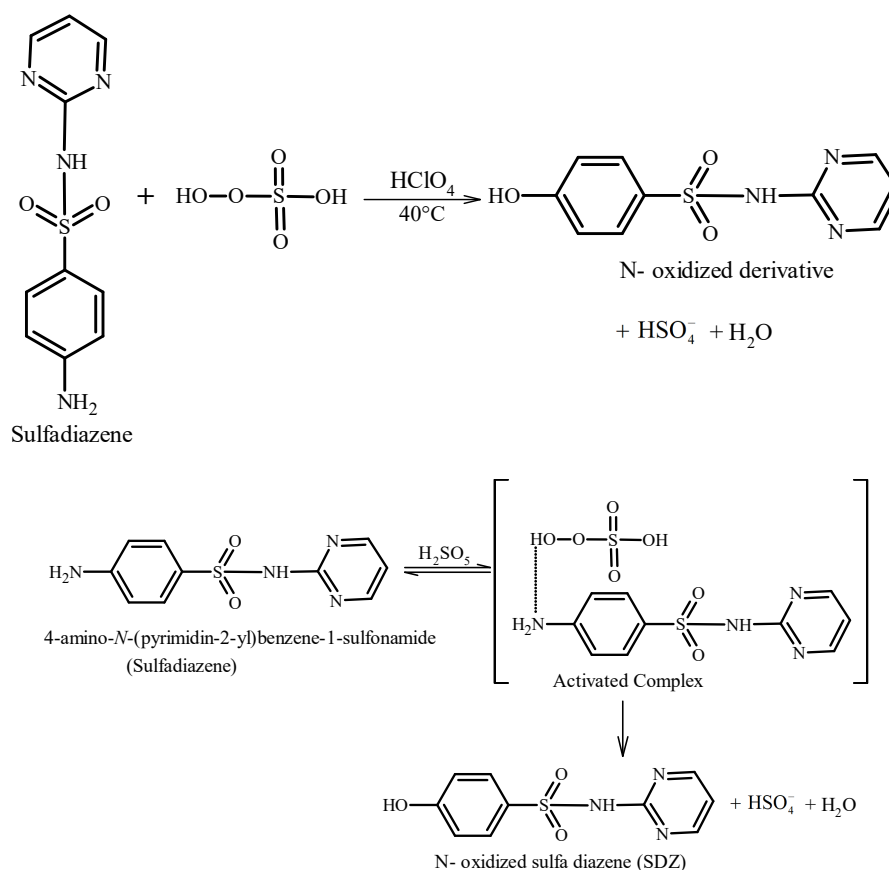


Fig. 10 FTIR analysis of the product for the SDZ-PMS reaction.

The FTIR spectrum of the oxidation product indicates that the sulfonamide aromatic framework of sulfadiazine remains largely intact after oxidation by peroxymonosulfate in acidic medium (fig. 10). The observed absorptions in the 1000–700 cm^{-1} region, particularly near 987, 887 and 752 cm^{-1} , suggest the formation of oxygenated derivatives while retaining para-substituted aromatic character. The absence of a strong

carbonyl absorption near 1700 cm^{-1} indicates that extensive ring cleavage or aldehyde/ketone formation does not occur under the employed conditions. The spectral features are therefore consistent with hydroxylated or N-oxidized sulfadiazine derivatives formed through selective oxygen-transfer oxidation by protonated peroxymonosulfate species.



Proposed Scheme for the Oxidation of Sulfadiazine with Peroxymonosulfate.

5. CONCLUSION

The oxidation of sulfadiazine by peroxymonosulphate in acidic medium has been effectively explored by kinetic, thermodynamic, spectroscopic, and mechanistic techniques. The reaction follows an increasing reliance on sulfadiazine, PMS, perchloric acid, and sodium perchlorate concentrations, showing the inclusion of protonated oxidant species and a polar activated complex in the rate-determining step. The observed positive ionic strength effect supports an ionic mechanism involving an ordered encounter complex between sulfadiazine and protonated peroxymonosulphate.

The low activation energy (12.912 kJ mol⁻¹) suggests that the oxidation occurs by an energetically simple pathway, while the highly negative entropy of activation (-231.44 J K⁻¹ mol⁻¹) implies the creation of a compact and highly organized associative transition state. The positive Gibbs free energy of activation at 40 °C (85.36 kJ mol⁻¹) reveals that the reaction is largely entropy regulated. HOMO–LUMO calculations further support the proposed mechanism, where sulfadiazine behaves as an electron donor and PMS acts as an electron acceptor through a $HOMO_{SDZ} \rightarrow LUMO_{PMS}$ interaction.

FTIR examination of the oxidation products reveals the development of hydroxylated and N-oxidized sulfadiazine derivatives while essentially keeping the aromatic sulfonamide framework. These data support a selective oxygen-transfer or outer-sphere electron-transfer mechanism rather than extensive mineralization or radical chain destruction.

The present work is environmentally noteworthy since sulfadiazine is a persistent sulfonamide antibiotic frequently discovered in wastewater, hospital discharge, surface water, and agricultural runoff. Its constant release into aquatic systems contributes to antibiotic resistance, toxicity for aquatic creatures, and ecological imbalance. The effective oxidation of sulfadiazine by PMS at mild acidic conditions demonstrates the potential applicability of this reaction for the removal of pharmaceutical pollutants from water matrices. The molecular insight provided from this study may potentially contribute to the development of selected enhanced oxidation methods for wastewater remediation and pharmaceutical pollution control.

Future investigations may include LC–MS and NMR identification of oxidation products, toxicity assessment of intermediates, catalyst-assisted PMS activation, and experiments in real wastewater systems involving competing ions and natural organic matter.

Table 2: Reaction of sulphadiazine with peroxymonosulphate in acidic medium.

S.NO	[SDZ]	[PMS]	[HClO ₄]	TEMP °C	[NaClO ₄]	K Min ⁻¹
1	0.02	0.002	0.02	25	0	0.02763

2	0.02	0.002	0.02	30	0	0.02990
3	0.02	0.002	0.02	35	0	0.03450
4	0.02	0.002	0.02	40	0	0.03742
5	0.02	0.002	0.02	45	0	0.04030
6.	0.02	0.002	0.02	40	0	0.03742
7.	0.025	0.002	0.02	40	0	0.02993
8.	0.03	0.002	0.02	40	0	0.03598
9.	0.035	0.002	0.02	40	0	0.03689
10	0.04	0.002	0.02	40	0	0.04030
11	0.045	0.002	0.02	40	0	0.03886
12	0.02	0.002	0.01	40	0	0.02763
13	0.02	0.002	0.015	40	0	0.02993
14	0.02	0.002	0.02	40	0	0.03742
15	0.02	0.002	0.03	40	0	0.03680
16	0.02	0.002	0.035	40	0	0.04145
17	0.02	0.002	0.04	40	0	0.03910
18	0.02	0.002	0.045	40	0	0.04600
19	0.02	0.002	0.05	40	0	0.04893
20	0.02	0.001	0.02	40	0	0.02878
21	0.02	0.0015	0.02	40	0	0.02990
22	0.02	0.002	0.02	40	0	0.03742
23	0.02	0.0025	0.02	40	0	0.03166
24	0.02	0.003	0.02	40	0	0.03822
25	0.02	0.0035	0.02	40	0	0.04606
26	0.02	0.002	0.02	40	0.25	0.03109
27	0.02	0.002	0.02	40	0.5	0.02993
28	0.02	0.002	0.02	40	0.75	0.03224
29	0.02	0.002	0.02	40	1	0.03339
30	0.02	0.002	0.02	40	1.5	0.03454
31	0.02	0.002	0.02	40	2	0.03915

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Conflict of Interest

The authors have no competing interests to declare that are relevant to the content of this article.

Credit Taxonomy

Anita Meena: Conceptualization, Methodology, Supervision, Editing, and Drafting.

Suraj Prasad: Investigation, Writing-original draft.

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