The cooperative influence of Ag(I) and sodium lauryl sulfate micelles on the oxidation rate of D-mannitol by acidic $KMnO_4$

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ABSTRACT

Oxidation of organic substrate by $KMnO_4$ is fascinating because such process proceeds via different mechanistic pathway. No research has examined how metal catalysts affect carbohydrate oxidation by acidic $KMNO_4$ in micellar environments. The impact of sodium lauryl sulfate (SLS) micelles on the Ag(I) facilitated oxidation kinetics of D-mannitol by $KMnO_4$ has been investigated in the aqueous micellar media. The progression of the oxidation process is assessed by monitoring the decrease in absorbance, employing the pseudo-first-order condition as an indicator for [H^+], [Mn(VII)], [Na_2SO_4], [D-mannitol], [Ag(I)], and [SLS]. The examined reaction shows fractional-first-order kinematics for [D-mannitol], [H^+], and Ag(I), and first-order for [Mn(VII)] across the examined concentration range. The spectrophotometric kinetic investigation revealed that both Ag(I) and SLS catalyze the oxidation independently, yet their combined influence is significantly more pronounced. Ag(I) significantly increases the oxidation rate (5.5 times). Interestingly, the kinetic profile demonstrated an increase in the observed rate constant values (7.2 fold) corresponding to an increase in surfactant concentration. The combination of Ag(I) and SLS micelles produces an 11.5-fold enhancement in the oxidation rate of D-mannitol. The electrostatic attraction between the charged micelle and Mn(VII) promotes the proximity of the oxidizing species to the substrate (D-mannitol), which is typically solubilized in a micellar environment, thus enhancing the oxidation process. A credible mechanism that corresponds with the kinetic findings has been emphasized, alongside an analysis of the Piszkiewicz model, to elucidate the apparent catalytic influence of SLS micellar environments.

KEYWORDS

D-mannitol, Ag(I) catalyzed, Permanganate ion, Micellar medium, Rate enhancement

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INTRODUCTION

The exploration of carbohydrates represents a captivating domain within organic chemistry. Carbohydrates serve as the fundamental fuel for energy across humans, animals, and plants. Mannose is a significant sugar alcohol within the realm of carbohydrates, playing a crucial role in human metabolism, particularly in the glycosylation processes of specific proteins. A number of genetic problems of glycosylation are linked to enzyme mutations that play a role in mannose metabolism.[1] Mannitol, an epimer of glucose, can undergo oxidation to yield the analogous aldohexose, mannose, through the application of various oxidizing agents.[2-4] With the chemical formula $C_6H_8(OH)_6$, D-Mannitol is a significant organic molecule that is mostly utilized in chocolate-flavored topping ingredients for confections and ice cream. This polyol serves a primary function in the circuit prime of the heart-lung device during cardiac bypass surgery and is additionally utilized as a therapeutic agent for the management of acute glaucoma in veterinarians. It serves as a sweetening agent in candies designed to freshen breath and is frequently identified as a mild laxative suitable for infants.[5] The carbohydrate oxidation has been extensively examined within the realm of organic chemistry.[6-8] This research aims to investigate the oxidation of D-mannitol by permanganate in a sulfuric acid environment, facilitated by Ag(I) and SLS micelles.

Potassium permanganate is a highly adaptable oxidizing agent, frequently employed in acidic, neutral, and alkaline environments for the oxidation of organic molecules.[9] The permanganate oxidation process is environmentally benign when juxtaposed with other higher valent transition metals and has gained importance in the field of green chemistry.[10] This is attributed to the advancement of the technique for recycling the by-product manganese dioxide, which can be reconstituted into an oxidant or utilized in the production of catalysts, pigments, or batteries.[11] In an acidic environment, it manifests in various forms such as Mn_2O_7, $HMnO_3$, $H_2MnO_4^+$, and $HMnO_4$.[12] The specific nature of the reductant influences the assignment of the oxidant to either outer sphere or inner sphere pathways in their redox reactions.[13] The kinetics associated with the polyol oxidation by permanganate ions in both alkaline and acidic milieus have garnered significant interest over the past several years.[14-17] Transition metal ions like Ru(III), Cu(II), and Ag(I) are sought after as catalysts in the oxidation of organic moieties by permanganate ions. Researchers have proposed multiple mechanistic approaches for the oxidation of organic moieties by permanganate ions.[18,19] In light of this, it is considered important to examine the kinetic behavior of the specified reaction in the aqueous anionic micellar environment.

The influence of surfactants and micelles on chemical kinetics presents a captivating area of research for biologists and chemists.[20-24] Micelles represent dynamic assemblies of amphiphilic molecules that establish distinctly anisotropic interfacial regions at the interface of non-polar hydrocarbon and highly polar aqueous regions, thereby conferring novel physical and chemical characteristics to the system.[25] Micelles, along with other association colloids, function as microreactors that concentrate, separate, or dilute reactants, thus significantly affecting chemical reactivity.[23] Due to their unique properties, micelles have been frequently employed as models to explore the influence of heterogeneous environments and microenvironments on a diverse array of reactions.[26] Moreover, micellar catalysis has the capacity to create mild reaction conditions, effectively suppressing the occurrence of side reactions while simultaneously enhancing the efficacy of organic processes.[27] Micellar catalysis offers a compelling approach to addressing solubilization challenges; indeed, the aggregation of surfactants, propelled by the hydrophobic effect, facilitates the creation of polar nano-environments. In these environments, both substrates

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and catalysts can be effectively dissolved and interact with elevated local concentrations.[28,29] The availability of various types of surface-active agents is contingent upon the charge of the head group. The anionic surfactant SLS, characterized by its sulfate head group, unveils several advantages when it interacts with substances of opposite charge. The stabilization of these formulated anionic conjugated systems is achieved through electrostatic forces of attraction. Consequently, the chemical transformations mediated by aqueous SLS represent a compelling area of research that continues to be investigated, particularly with an emphasis on establishing their roles in environmentally friendly synthesis, green chemistry, and sustainable catalysis.

Our research team has long been engaged in the exploration of diverse metal-catalyzed oxidative transformation processes, focusing on a range of organic substrates and various oxidizing agents within micellar environments.[30-34] While numerous studies have addressed the catalyzed and uncatalyzed oxidation of amino acids, carboxylic acids, polyols, and dyes by Mn(VII) in both aquatic and micellar milieus,[35-38] the metal-catalyzed oxidation of polyols by Mn(VII) in micellar medium has been relatively underexplored. This study presents the comprehensive kinetic analysis of the synergistic influence of Ag(I) and SLS on the rate enhancement of D-mannitol oxidation directed by permanganate ion, as this particular investigation has not been previously addressed within the micellar environment. In this scenario, Mn(VII) functions as a polar oxidant, while the catalyst carries a positive charge. Utilizing a micellar surface with a contrasting charge may significantly improve the oxidation rate. In addition, the other aims included identifying the primary kinetically reactive species, determining the principal oxidation product, deducing the rate law, and ascertaining the likely reaction site in the presence of a catalyst.

EXPERIMENTAL

Reagents and Instruments

D-mannitol (99 %, SRL, India), H_2SO_4 (98%, Merck, India), $AgNO_3$ (99.9 % HiMedia, India), SLS (99 %, Merck, India), and Na_2SO_4 (98%, Merck, India) were used without further purification. Preparation and standardization of potassium permanganate solution (99 %, Merck, India) was similar to that reported elsewhere.[39] All reaction mixtures were made using double-distilled water, which has a conductivity between 1 to 2 μS cm^{-1}. To eliminate the possibility of any photochemical degradation, reaction vessels were painted black.

An HTLP digital pH Meter (model HTLP-081) was employed to monitor the pH of the resultant mixture. The device's accuracy was confirmed using a recommended buffer solution. A Labtronics (LT-1900) UV-visible spectrophotometer was utilized for recording the spectral scans and measuring absorption values at a designated wavelength. An independently developed system of recirculating water was responsible for maintaining a constant temperature inside the cell compartment.

Kinetic Measurements

The oxidation of D-mannitol by Mn(VII), catalyzed by Ag(I), was performed at 303 K utilizing pseudo-first-order settings ([D-mannitol] >> [Mn(VII)]) throughout both aquatic and micellar conditions. The specified quantity of reactants (D-mannitol, surfactant, H_2SO_4, Na_2SO_4, and Ag(I)) was pre-immersed for 30 minutes in a thermostat to maintain temperature stability. The reaction commenced by mixing the requisite volume of thermally equilibrated oxidant solution with the reaction mixture. The reaction's progression was monitored spectrophotometrically by monitoring the decline in absorbance at 525 nm corresponding to [Mn(VII)]. The absorption peaks at 525 nm assigned to the orbitally allowed $^1A_1$ to $^1T_2$ transition.[40] The absorbance at zero time (A_0), at distinct points of time (A_t), and at infinite time (A_∞) was carefully computed. The non-linear fitting strategy, utilizing the exponential first-order rate equation (equation 1), was employed to ascertain the rate constant (k_{obs}) of the reaction.[41]

$$\left(A_\infty - A_t\right) = \left(A_\infty - A_0\right)e^{-kt} \tag{1}$$

RESULTS AND DISCUSSION

Reaction's stoichiometry, analysis of the product, and evaluation of free radical involvement

To determine the stoichiometry of the reaction, the measured amount of D-mannitol (1.0 × 10^{-2} M) was allowed to react with different mole ratios of Mn(VII) (1.0 × 10^{-2} M to 6.0 × 10^{-2} M) in 1.0 M H_2SO_4 within a tightly closed container at a temperature of 303 K, in both micellar and aquatic environments. The reaction was allowed to proceed to completion at that temperature. Determination of unreacted Mn(VII) suggested that two moles of Mn(VII) are required to oxidize five moles of D-mannitol, as demonstrated in equation 2.

$$5\,RCH_2OH + 2\,MnO_4^- + 6\,H^+ \xrightarrow{Ag(I)} 5\,RCHO + 2\,Mn^{2+} + 8\,H_2O$$

D-Mannitol D-Mannose (2)

Where R = $HOH_2C(CHOH)_4$

Following the conclusion of the reaction, the resulting mixture was meticulously shifted into a round-bottom flask, linked to fractional distillation equipment. The product of the distillation process was collected for subsequent analysis. The ultimate outcome of the reaction was identified as D-mannose; the detection and confirmation are achieved through spot tests utilizing a reagent solution composed of 0.25 g of stannous chloride and 5.0 g of urea dissolved in 12 mL of 45% sulfuric acid under heated conditions.[42] The resulting solutions exhibited a red hue. This hue signifies the emergence of aldohexose. The identical R_f value of the obtained product and pure D-mannose in TLC analysis, along with the melting point of the formed product, further corroborates the formation of D-mannose as the final product.

A research attempt was conducted to determine the presence of free radicals in the course of the reaction by employing a polymerization test. The reaction mixture was sustained in an inert environment for an extended period of 6 hours, employing a specific amount of acrylonitrile. The addition of methanol results in the formation of a white precipitate, signifying the emergence of a free radical intermediate within this reaction process.

Impact of varying [H$^+$] on k_{obs}

The study aimed to examine the oxidation rate constant across a range of [H$^+$] from 0.25 M to 2.5 M by computing the rate constant at various [H$^+$] values. The studied reaction reflects fractional-first-order kinematics in relation to [H$^+$], as demonstrated by the positive intercepted linear relationship between [H$^+$] and k_{obs} in Figure 1. At low levels of H$^+$, the reaction rate is moderate, but then it gradually increases in a precise manner as the concentration of H$^+$ increases within the assessed range. Mn(VII) predominantly manifests in its protonated state ($HMnO_4$) in an acidic medium.[18,19] The existence of a protonated form of Mn(VII) is thought to be accountable for the enhanced oxidation rate witnessed in an aquatic medium following an increase in [H$^+$]. Compared to an aqueous environment, the oxidation rate in SLS micellar media was significantly higher, as depicted in Figure 1.

Impact of varying [D-mannitol] on k_{obs}

The impact of variable [D-mannitol] upon the reaction rate was investigated at 303 K temperature within the spectrum of 3.0 × 10^{-3} M to 30.0 × 10^{-3} M, at the specified H$^+$ concentration. The graph illustrating the correlation between k_{obs} and [D-mannitol] (Figure 2) demonstrates a consistent increase in the k_{obs} value, characterized by positive intercepts. The results of the investigation demonstrate that in both the aquatic and SLS micellar milieus, the D-mannitol displays fractional-first-order kinetics. In contrast to the aquatic medium, the micellar environment of SLS displays a higher rate of D-mannitol oxidation by Mn(VII).

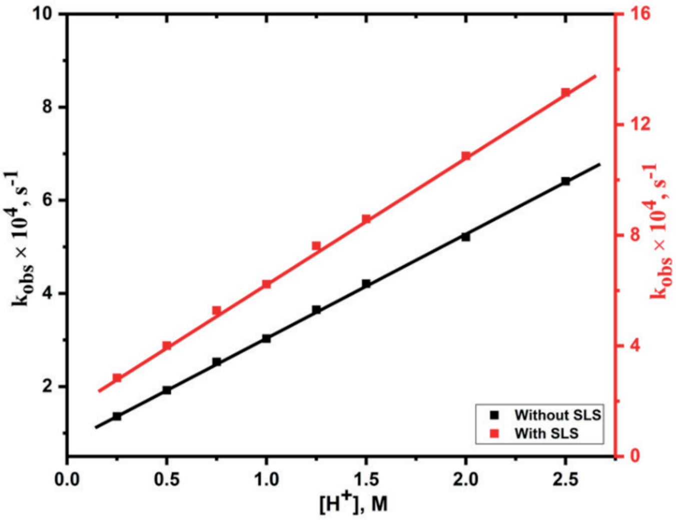

Figure 1: The correlation between k_{obs} and $[H^+]$ at $[KMnO_4]$ = 2.5 × 10^{-4} M, [D-mannitol] = 8.5 × 10^{-3} M, $[Na_2SO_4]$ = 0.25 M, [Ag(I)] = 7.5 × 10^{-5} M, Temperature = 303 K, and [SLS] = 7.75 × 10^{-3} M

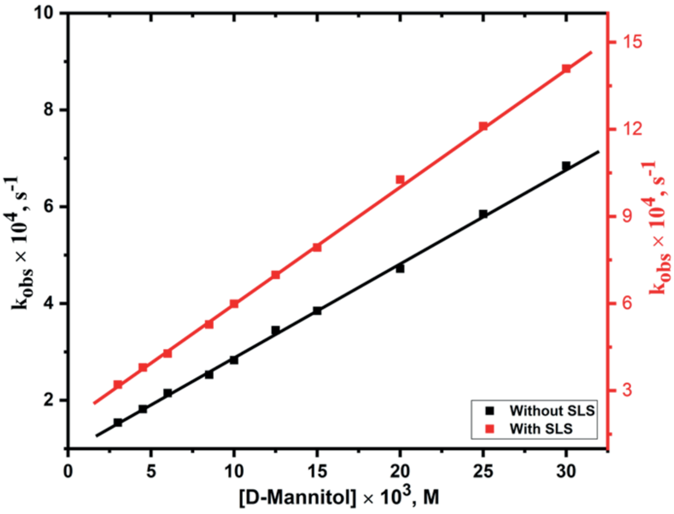

Figure 2: The correlation between k_{obs} and [D-mannitol] at $[KMnO_4]$ = 2.5 × 10^{-4} M, $[H_2SO_4]$ = 0.75 M, $[Na_2SO_4]$ = 0.25 M, [Ag(I)] = 7.5 × 10^{-5} M, Temperature = 303 K, and [SLS] = 7.75 × 10^{-3} M

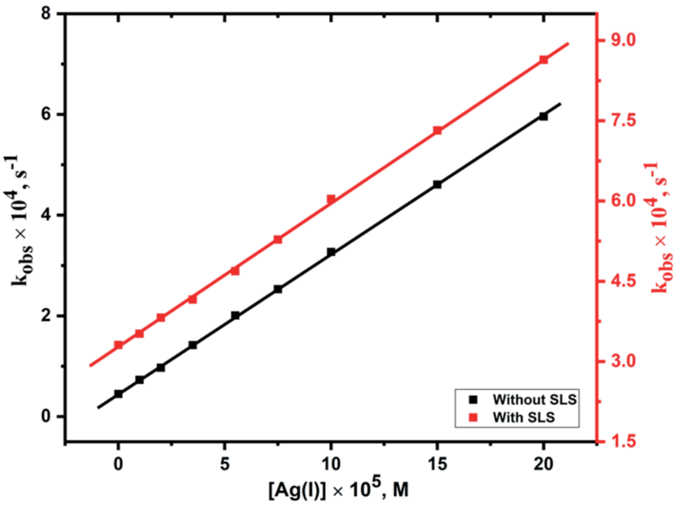

Figure 3: The correlation between k_{obs} and [Ag(I)] at $[KMnO_4]$ = 2.5 × 10^{-4} M, [D-mannitol] = 8.5 × 10^{-3} M, $[H_2SO_4]$ = 0.75 M, $[Na_2SO_4]$ = 0.25 M, Temperature = 303 K, and [SLS] = 7.75 × 10^{-3} M

Table 1: Impact of varying $[KMnO_4]$ on k_{obs}
Experimental Condition: [D-mannitol] = 8.5 × 10^{-3} M, $[H_2SO_4]$ = 0.75 M, $[Na_2SO_4]$ = 0.25 M, [Ag(I)] = 7.5 × 10^{-5} M, Temperature = 303 K, and [SLS] = 7.75 × 10^{-3} M

$10^4 \times [KMnO_4]$, M	$10^4 \times k_{obs}$, s^{-1} (without SLS)	$10^4 \times k_{obs}$, s^{-1} (with SLS)
0.5	2.42 ± 0.10	5.33 ± 0.14
1.0	2.55 ± 0.09	5.17 ± 0.11
1.5	2.51 ± 0.11	5.20 ± 0.17
2.5	2.53 ± 0.08	5.28 ± 0.16
3.5	2.50 ± 0.10	5.31 ± 0.10
4.5	2.45 ± 0.09	5.29 ± 0.09
5.5	2.57 ± 0.07	5.22 ± 0.12
6.6	2.53 ± 0.12	5.35 ± 0.13
8.0	2.60 ± 0.14	5.25 ± 0.07

Table 2: Impact of varying $[Na_2SO_4]$ on k_{obs}
Experimental Condition: $[KMnO_4]$ = 2.5 × 10^{-4} M, [D-mannitol] = 8.5 × 10^{-3} M, $[H_2SO_4]$ = 0.75 M, [Ag(I)] = 7.5 × 10^{-5} M, Temperature = 303 K, and [SLS] = 7.75 × 10^{-3} M

$[Na_2SO_4]$, M	$10^4 \times k_{obs}$, s^{-1} (without SLS)	$10^4 \times k_{obs}$, s^{-1} (with SLS)
0.05	2.57 ± 0.12	5.18 ± 0.11
0.1	2.49 ± 0.11	5.29 ± 0.15
0.15	2.45 ± 0.10	5.25 ± 0.17
0.20	2.58 ± 0.13	5.30 ± 0.15
0.25	2.53 ± 0.08	5.28 ± 0.16
0.40	2.44 ± 0.09	5.33 ± 0.18
0.50	2.54 ± 0.10	5.20 ± 0.14

Impact of varying [Mn(VII)] on k_{obs}

By maintaining all other variables pertaining to the reaction constant and applying the optimum [D-mannitol] and H^+ conditions, the oxidation rate was calculated with respect to [Mn(VII)] within the 0.5 × 10^{-4} M to 8.0 × 10^{-4} M concentration range. Referring to the calculated k_{obs} value for each [Mn(VII)], as presented in Table 1, clearly indicates that the examined range of [Mn(VII)] exhibits first-order kinetics.

Impact of varying $[Na_2SO_4]$ on k_{obs}

By employing sodium sulfate to regulate the ionic strength (I) of the reacting mixture between 0.05-0.50 M, the impact of I on the oxidation rate was examined. The other variables were remained constant at $[KMnO_4]$ = 2.5 × 10^{-4} M, [D-mannitol] = 8.5 × 10^{-3} M, $[H_2SO_4]$ = 0.75 M, [Ag(I)] = 7.5 × 10^{-5} M, Temperature = 303 K, [SLS] = 7.75 × 10^{-3} M. A zero salt effect is suggested by the observed constancy in oxidation rate with increasing Na_2SO_4 concentration, as displayed in Table 2. The constancy of the oxidation rate regardless of the ionic strength indicates that the positively charged D-mannitol-Ag(I) complex and the neutral $HMnO_4$ are involved in the rate-determining step.

Impact of varying [Ag(I)] on k_{obs}

In light of the prospective utilization of Ag(I) promoted oxidative processes for the micro-level detection of Ag(I), it is vital to investigate the influence of [Ag(I)] on the k_{obs}. The research focused on analyzing the oxidation rate within the parameters of 1.0 × 10^{-5} to 20.0 × 10^{-5} M [Ag(I)] under optimal reaction conditions. This was accomplished through the calculation of the rate constant (k_{obs}) at different concentrations of Ag(I). Figure 3 displays a linear trend between k_{obs} and [Ag(I)], which suggests fractional-first-order kinematics in reliance on [Ag(I)] across the concentration range under investigation.

Impact of added Mn^{2+} and F^- ions on k_{obs}

The role of Mn(II) as a catalyst in the oxidation process facilitated by permanganate is well established. The anticipated oxidizing species in the oxidation process involving permanganate in acidic solutions are Mn(VII), Mn(IV), and Mn(III), as delineated in equation 3.[19,43]

$$\mathrm{MnO_4^- + 3\,Mn^{2+} + 8\,H^+ \longrightarrow 3\,Mn^{3+} + Mn^{4+} + 4\,H_2O} \tag{3}$$

If Mn(III) and Mn(IV) are the reactive species, the oxidation rate will increase with [Mn(II)], as documented by Verma et al.[44] In the current scenario, as the initial concentration of Mn(II) was increased, the rate exhibited a progressive decline (Table 3). This outcome suggests that neither Mn(III) nor Mn(IV) plays a role in the oxidation process. Moreover, if Mn(VI) or Mn(III) serve as the reactive species, the introduction of complexing agents such as fluoride ions would be expected to diminish the reaction rate.[19] The absence of any influence from fluoride ions on the rate indicates that Mn(VII) is the most likely reactive species.

Impact of temperature on k_{obs}

The study looked at the relationship between temperature and oxidation rate between 298 and 318 K. The examination of the reaction under elevated temperatures was disregarded due to apprehensions regarding the possible deterioration of the finished product. The reaction proceeds precisely in accordance with the equation proposed by Arrhenius, which guarantees that the reaction rate rises with temperature. There is a noticeable pace of advancement in the reaction at 303 K. As a result, 303 K should be considered the optimal choice for further exploration of the reaction system. Activation energy (E_a) values were determined using the Arrhenius equation by plotting $\ln k_{obs}$ vs 1/T (Figure 4), and other activation parameters were calculated using the Eyring equation. The values of the evaluated activation parameters have been presented in Table 4. Moore and Hicks reported that the activation entropy, $\Delta S^{\#}$, for a series of MnO_4^- reactions is negative for those that occur through complex formation, specifically the inner-sphere mechanism, conversely, the $\Delta S^{\#}$ values for the outer-sphere reactions generally exhibit more positive values.[45] The heightened negative entropy of activation suggests a diminished degree of freedom, thereby facilitating the emergence of a complex between D-mannitol and Ag(I).

Impact of [SLS] on k_{obs}

Surfactants possess the ability to promote rate enhancement while simultaneously guaranteeing the consistent dispersion of organic reactants in an aqueous medium, dependent upon their charge attributes. The surfactants (SLS), spanning a broad spectrum of concentrations from pre- to post-micellar regions, have been utilized for an in-depth kinetic investigation. The examination of the kinetic outcome distinctly reveals that the presence of aqueous micellar media markedly accelerates the oxidation rate in comparison to the reaction taking place exclusively in aqueous media (Table S1). Figure 5 shows the effect of SLS and Ag(I) and combined impact of SLS + Ag(I) on reaction rate. Ag(I) acts as a catalyst and enhances the oxidation of d-mannitol by 5.5 times, whereas the negative charges SLS micelles attracts the positively charged Ag(I), polar D-mannitol, and neutral $HMNO_4$ in the stern layer and accelerates the rate of reaction by 7.2 times. The combined impact of SLS + Ag(I) is more pronounced, enhancing the reaction rate by 11.5 fold.

The graph depicting [SLS] versus k_{obs}, as presented in Figure 6, reveals a substantial rise in the reaction rate as [SLS] approaches 8.0×10^{-3} M, which is in close proximity to the CMC of SLS. Subsequent to this, the decrease in the oxidation rate was noticed throughout the ranges of the examined [SLS]. In the observed reaction condition, the computed CMC of SLS is 7.32×10^{-3} M, which is marginally less than the value recorded in the aquatic environment. The convergence of the two linear trajectories on the k_{obs} versus [SLS] graph elucidates this point. The apparent decline in CMC of SLS might be due to the electrostatic attraction between the positively charged Ag(I) and negatively charged head of SLS micelle, which mitigates the mutual electrostatic repulsion amongst the negative head groups of surfactant micelles.

Mechanism and rate law

In the oxidation of diverse organic moieties by acidic Mn(VII), it has been proposed that the accelerated oxidation rate observed at elevated hydrogen-ion concentrations is likely attributable to the protonation of the oxidant. The argument was corroborated by the documented spectral analyses that aligned with equation 4.[18,19]

$$\mathrm{MnO_4^- + H^+ \rightleftharpoons HMnO_4} \tag{4}$$

The effect of added ions (Mn^{2+} and F^-) on the reaction rate further suggests that the Mn(VII) will be the most reactive species in the current reaction conditions. The results of the kinetic study suggested

Table 3: Impact of added Mn^{2+} and F^- ions on k_{obs}
Experimental Condition: $[KMnO_4] = 2.5 \times 10^{-4}$ M, [D-mannitol] $= 8.5 \times 10^{-3}$ M, $[H_2SO_4] = 0.75$ M, $[Ag(I)] = 7.5 \times 10^{-5}$ M, Temperature = 303 K, and [SLS] = 7.75×10^{-3} M

$10^4 \times$ [Added Ion], M		$10^4 \times k_{obs}$, s^{-1} (without SLS)	$10^4 \times k_{obs}$, s^{-1} (with SLS)
Mn^{2+}	1.0	2.16 ± 0.07	4.31 ± 0.12
	2.0	1.63 ± 0.06	2.95 ± 0.09
	3.5	1.01 ± 0.04	1.57 ± 0.05
	5.0	0.43 ± 0.01	0.76 ± 0.05
F^-	2.0	2.51 ± 0.09	5.33 ± 0.15
	4.0	2.55 ± 0.10	5.20 ± 0.16
	6.0	2.48 ± 0.07	5.25 ± 0.11
	8.0	2.56 ± 0.08	5.36 ± 0.17
None		2.53 ± 0.08	5.28 ± 0.16

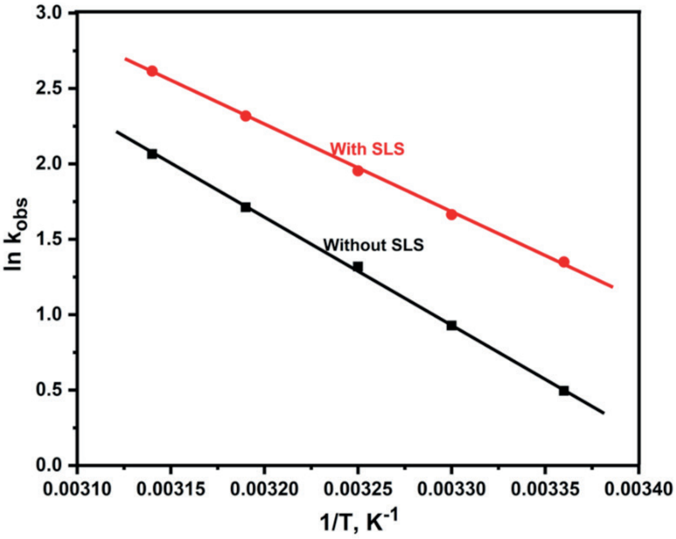

Figure 4: The correlation between $\ln k_{obs}$ and 1/T at $[KMnO_4] = 2.5 \times 10^{-4}$ M, [D-mannitol] $= 8.5 \times 10^{-3}$ M, $[H_2SO_4] = 0.75$ M, $[Na_2SO_4] = 0.25$ M, $[Ag(I)] = 7.5 \times 10^{-5}$ M, and [SLS] $= 7.75 \times 10^{-3}$ M

Table 4: Activation parameters for the oxidation reaction
Experimental Condition: $[KMnO_4] = 2.5 \times 10^{-4}$ M, [D-mannitol] $= 8.5 \times 10^{-3}$ M, $[H_2SO_4] = 0.75$ M, $[Ag(I)] = 7.5 \times 10^{-5}$ M, Temperature = 303 K, and [SLS] = 7.75×10^{-3} M

	E_a kJ mol^{-1}	$\Delta H^{\#}$ kJ mol^{-1}	$\Delta S^{\#}$ J $K^{-1}mol^{-1}$
Without SLS	59.33 ± 0.68	56.81 ± 0.53	-126.37 ± 0.78
With SLS	48.13 ± 0.55	45.61 ± 0.47	-157.22 ± 0.97

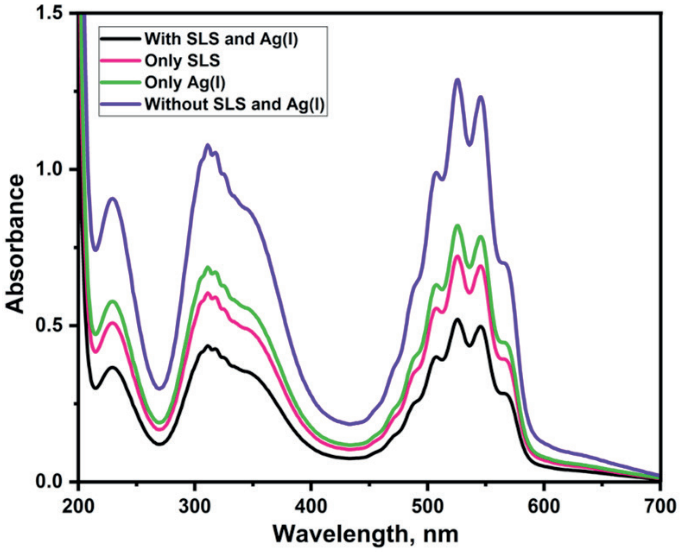

Figure 5: Independent and combined effect of Ag(I) and SLS on D-Mannitol oxidation by $KMnO_4$ at $[KMnO_4] = 2.5 \times 10^{-4}$ M, [D-mannitol] = 8.5×10^{-3} M, $[H_2SO_4] = 0.75$ M, $[Na_2SO_4] = 0.25$ M, $[Ag(I)] = 7.5 \times 10^{-5}$ M, Temperature = 303 K, and $[SLS] = 7.75 \times 10^{-3}$ M

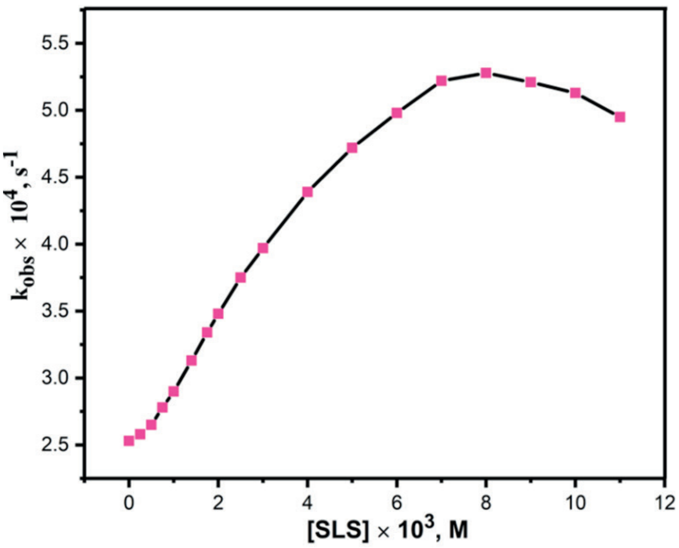

Figure 6: The correlation between k_{obs} and [SLS] at $[KMnO_4] = 2.5 \times 10^{-4}$ M, [D-mannitol] = 8.5×10^{-3} M, $[H_2SO_4] = 0.75$ M, $[Na_2SO_4] = 0.25$ M, $[Ag(I)] = 7.5 \times 10^{-5}$ M, and Temperature = 303 K

a first-order reliance on Mn(VII), a fractional-order relationship with respect to D-mannitol, H^+, and Ag(I). The fractional order dependence on both D-mannitol and Ag(I) indicates that the catalyst Ag(I) and the substrate D-mannitol engage in complex formation prior to the rate-determining step. The zero-salt effect reveals the participation of neutral $HMNO_4$ and positively charged D-mannitol-Ag complex in the critical rate-determining step. The research findings allow us to suggest the most plausible mechanistic framework for the Ag(I) facilitated oxidation of D-mannitol by Mn(VII) in an acidic environment through Eqs. 5-9, aligning with previous similar findings.[14-18,46]

$$MnO_4^- + H^+ \overset{K_1}{\rightleftharpoons} HMnO_4 \tag{5}$$

$$RCH_2OH + Ag(I) \overset{K_2}{\rightleftharpoons} RCH_2\overset{+}{O}H \rightarrow Ag(I) \tag{6}$$

$$RCH_2\overset{+}{O}H \rightarrow Ag(I) + HMnO_4 \xrightarrow[slow]{k} RCH_2O^{\bullet} + Ag(I) + MnO_3^- + H_2O \tag{7}$$

$$RCH_2O^{\bullet} + H_2O \xrightarrow{fast} RCHO + H_3O^+ \tag{8}$$

$$5\,MnO_3^- + 6\,H^+ \xrightarrow{fast} 3\,MnO_4^- + 2\,Mn^{2+} + 3\,H_2O \tag{9}$$

Where, RCH_2OH is D-mannitol

$$\text{Rate of Reaction} = -\frac{d\left[MnO_4^-\right]}{dt} = k\left[\text{Complex}\right]\left[HMnO_4)\right] \tag{10}$$

The ultimate rate law will be (derivation in supplementary information).

$$\text{Rate} = \frac{kK_1K_2\left[\left[\text{D-mannitol}\right]\left[Ag(I)\right]\left[MnO_4^-\right]\left[H^+\right]\right]}{\left(1+K_2\left[\text{D-mannitol}\right]\right)\left(1+K_1\left[H^+\right]\right)\left(1+K_2\left[Ag(I)\right]\right)} \tag{11}$$

The mentioned rate law delineates the entirety of the recorded kinetic order concerning various reaction parameters.

It can be inferred from our knowledge of chemistry that Ag(I) might act as a coordinator to facilitate the electron transport from D-mannitol to Mn(VII). Without Ag(I), the electron transfer process is a little sluggish. The enhancement of the rate by Ag(I) can be explained through the activation energy required for the progression of the rate-determining step in the oxidation process. It is plausible that Ag(I) lowers the activation energy linked to the electron transfer process, consequently decreasing the free energy of activation and thereby enhancing the reaction rate. The oxidation reaction advances rapidly as the concentration of Ag(I) within the reaction mixture increases.

Contingent upon their charge attributes (cationic, neutral, and anionic), surfactants can both increase the rate and ensure the uniform dispersion of reacting moieties in an aqueous media.[47,48] The catalytic activity observed below the CMC of SLS is likely attributed to the generation of catalytic micelles, which occurs through the aggregation of substrate moieties and monomeric SLS.[49] The processes facilitated by surfactants are profoundly influenced by premicellar conditions. Premicellar complexes provide a substrate that exhibits greater reactivity in comparison to micellar aggregates.[50] For the reaction in hand, the initial acceleration in oxidation rate with [SLS] might be ascribed to the establishment of a premicellar complex. Upon attaining the CMC, the premicellar structures undergo a collapse, instigating the emergence of micelles. Research indicates that an elevation in [SLS], particularly when [SLS] exceeds CMC, is unlikely to yield a significant concentration within the aqueous bulk phase. There is no notable alteration in the content of the non-micellar entity when exceeding the CMC of the surfactant. The reduction in nucleophile concentration due to the introduction of surfactant is responsible for the observed decline in reaction rate beyond the CMC of surfactant.[51-53]

In an aqueous surfactant environment, a comparable direct electron transfer pathway occurs, facilitating a rapid reaction progression. Surfactant exhibits a formidable ability to produce nanoscale micelles that facilitate the integration of all constituents in the SLS micellar solution.[50] Within the SLS micellar environment, the extended aliphatic chain of the SLS is located in the core of the micelle. On the other hand, the micelle's Stern layer keeps the polar components of the surfactant (SO_4^-), substrate (-OH), catalyst Ag^+, and reactive species Mn(VII) close together, thereby facilitating the oxidation of D-mannitol molecules (Figure 7). The enhanced accumulation of reactant molecules at the micellar interface enhances the frequency of effective collisions among the moieties involved in the reaction, thus significantly promoting the oxidation process. Micelle-facilitated reactions encompass three fundamental stages: the integration of substrates into micelles, the rearrangement of substrates at the micelle surface, and the ensuing formation of the product. Micelles exert an influence on the electron transfer process through the solubilization of reactants or by facilitating their distribution between the aqueous and micellar phases.

Piszkiewicz's Cooperativity Model

There are several suitable models for surfactants' catalytic action. Kinetic data in this study showed SLS's catalytic capacity at various

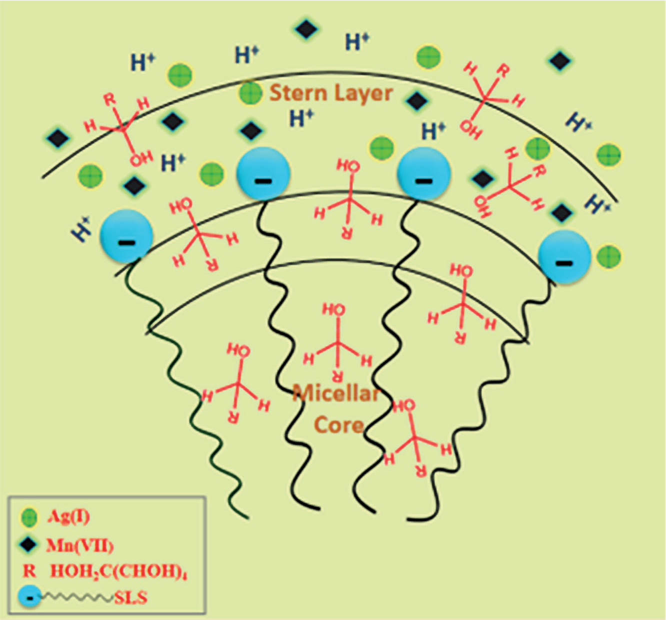

Figure 7: Conceptual illustration of Ag(I) catalyzed Oxidation of D-mannitol by $KMnO_4$ in an acidic SLS micellar medium

concentrations. Piszkiewicz's model serves as an appropriate choice for explaining and clarifying the precise quantitative evaluation of current micelle-mediated kinetic information.[54] The Hill model is closely analogous, demonstrating the positive cooperative impact observed in the enzyme-catalyzed reactions.[55] This hypothesis posits that micellar catalysis results via the complementary interactions between substrate and surfactant molecules, propelled by hydrophobic and/or electrostatic forces, leading to the development of catalytically potent clusters. Afterward, the substrate facilitates further micellar aggregation, resulting in an increased concentration inside the micelles.[56] According to this concept, a substrate (S) and n surfactant molecules (D) interact to create catalytically responsive micellar aggregates (DnS), which subsequently undergo reactions to yield the final products, as illustrated in Eqs. 12-14.[54]

$$nD + S \xrightleftharpoons{K_D} D_nS \quad (12)$$

$$D_nS \xrightarrow{k_m} \text{Product} \quad (13)$$

$$S \xrightarrow{k_w} \text{Product} \quad (14)$$

In this instance, K_D stands for the micellar aggregate dissociation constant. The term k_w indicates the rate constants associated with the reaction occurring in the bulk (aqueous) phase, while k_m signifies the maximum k_{obs} value in the micellar medium. The rate constant (k_{obs}) for the micelle-catalyzed pseudo-first-order reaction can be described as a function of [surfactant] using the following equation.

$$\log\left[\frac{k_{obs} - k_w}{k_m - k_{obs}}\right] = \log P = n \log D - \log K_D \quad (15)$$

Where n denotes the cooperativity Index, while [D] represents the surfactant's overall concentration. Equation 15's advantage lies in its independence from the critical micelle concentration of the employed surfactant. While Eq. 15 was initially formulated for micelle-catalyzed reactions exhibiting an optimum rate accompanied by inhibition, the approach has been utilized in various studies to elucidate the micellar effect, wherein the reaction is either inhibited or catalyzed by the micelle throughout the entire spectrum. The slope and intercept of the linear graph of logP versus log[D] are utilized to determine the values of n and K_D and found to be 1.7104 and 4.43×10^{-5}, respectively (Figure 8). A value of n over one indicates positive cooperative binding, suggesting aggregation with an effective number of monomers. The presence of catalytically effective submicellar aggregates is indicated by values of n = 1-2, which are significantly lower than the aggregated number (20–100).[57] of surfactant molecules that form micelles. A higher submicellar aggregate number is indicated by a lower value of K_D and a higher value of n. This implies that the complex between the surfactant and the substrate is more stable.

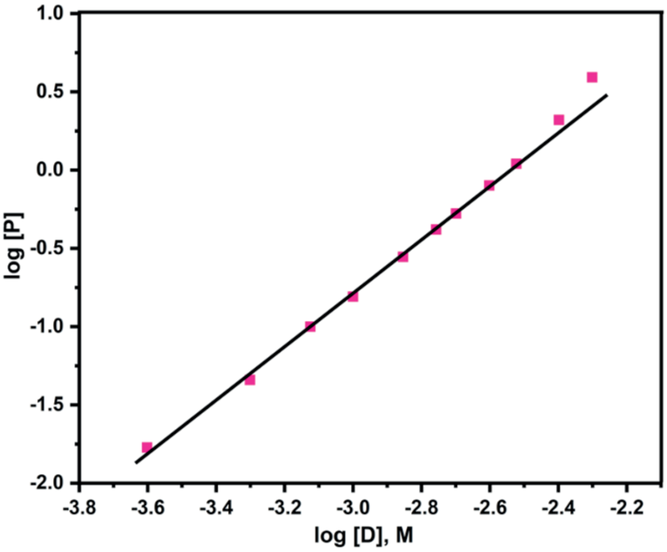

Figure 8: Examination of the experimental kinetic findings and their appropriate integration into Piszkiewicz's model

CONCLUSION

The electron transfer kinetics of D-mannitol are conducted both in the micellar phase and in an aqueous medium. In both mediums, the reaction proceeded along a comparable pathway, wherein an intermediate complex is established between the D-mannitol and Ag(I), preceding the rate-limiting step. Across the spectrum of concentrations explored, the reaction under consideration demonstrates fractional first-order kinetics with [Ag(I)], [H^+], and [D-mannitol], while displaying first-order reliance on [Mn(VII)]. The invariance of the oxidation rate with rising ionic strength demonstrates the presence of a zero salt effect. The oxidation of D-mannitol by Mn(VII) in an acidic environment has revealed that Ag(I) serves as a proficient metal catalyst. The combination of SLS and Ag(I) significantly enhances the reaction rate, demonstrating superior catalytic efficiency compared to Ag(I) alone. The Piszkiewicz model stipulates the catalytic activity of micellar-induced oxidative processes, highlighting positive cooperative binding ($n > 1$) and implying aggregation with an effective number of monomers.

SUPPLEMENTARY MATERIAL

Supplementary information for this article is provided in the online supplement.

AUTHOR CONTRIBUTION

A. Srivastava: Supervision, Conceptualization, Statistical analysis, Writing original draft. N. Srivastava: Investigation, Experimental, Statistical analysis, Formal Analysis, Writing original draft. V.K. Singh: Supervision, Conceptualization, Statistical analysis, Investigation, Formal Analysis, Writing original draft.

CONFLICTS OF INTEREST/COMPETING INTERESTS

None

GENERATIVE AI AND AI-ASSISTED TECHNOLOGIES

The authors declare that they have used generative AI and/or AI-assisted technologies in the writing process before submission, but only to improve the language and readability of their paper.

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