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Synthesis, Characterization and Photocatalytic Application of a Novel Manganese (III) Schiff Base Complex for Organic Dye Degradation

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Abstract

Salicylaldehyde and ethylenediamine were used to create and describe a new manganese (III) complex, [Mn(L)(OAc)], in which H_2L is the tetradentate N_2O_2 Schiff base ligand. FTIR, UV-Vis spectroscopy, mass spectrometry, and elemental analysis were used to study the complex. It was examined for its potential as a photosensitizer in the photocatalytic breakdown of methylene blue (MB) dye when exposed to visible light. In under 120 minutes, the [Mn(L)(OAc)] complex degraded more than 95% of MB, demonstrating its great efficiency. Reactive oxygen species (ROS) may be involved in the degrading pathway, as evidenced by control tests that validated the crucial roles of both the complex and light. The potential of earth-abundant manganese complexes as substitutes for rare-metal photocatalysts is demonstrated by this work.

Keywords: Novel Manganese (iii); Schiff Base; Complex; Organic Dye; Degradation

1. Introduction

One major environmental concern is the poisoning of water supplies by organic dyes used in the textile industry. Heterogeneous and homogeneous photocatalysis, two types of Advanced Oxidation Processes (AOPs), present potential approaches to the degradation of pollutants. Despite being good photocatalysts, noble metal complexes (such as those based on Ru or Ir) are expensive and scarce, which is why first-row transition metals are being used to create sustainable alternatives. Earth-abundant manganese is an essential part of natural photosynthetic systems and may access several oxidation states (II, III, and IV) [1,2]. Schiff base ligands are perfect for adjusting the electrical characteristics of the metal center and stabilizing these oxidation states. In this work, a new Mn (III)-Schiff base complex is synthesized and its effectiveness as a homogeneous photocatalyst for the degradation of methylene blue, a model pollutant, under visible light is assessed [3,4].

2. Experimental section

2.1. Materials and Synthesis

Every chemical was bought from a professional provider and utilized straight away. Schiff Base Ligand (H_2L) Synthesis: A stirred solution of ethylenediamine (0.60 g, 10 mmol) in ethanol (10 mL) was mixed with drop wise additions of salicylaldehyde (2.44 g, 20 mmol) in ethanol (20 mL). After two hours of refluxing, the mixture cooled to a brilliant yellow solution of the H_2L ligand.

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2.2. [Mn(L)(OAc)] Complex Synthesis

The yellow ligand solution was supplemented with manganese (II) acetate tetrahydrate (Mn $(OAc)_2 \bullet 4H_2O$, 2.45 g, 10 mmol. Over the course of 24 hours of stirring the liquid outside, a dark brown microcrystalline solid formed. After being filtered, the solid was cleaned with cold ethanol and diethyl ether and vacuum-dried. 3.1 g (78%) yield [5,6].

2.3. Characterization

FTIR, Elemental Analysis (CHN), ESI-Mass Spectrometry, and UV-Vis.

2.4. Photocatalytic Testing

Methylene blue (MB, 10 mg/L) and the catalyst (0.05 mM) were made into aqueous solutions. To achieve adsorption-desorption equilibrium, the solution was agitated for half an hour in the dark. After that, it was exposed to visible LED light ($\lambda > 420$ nm). At regular intervals, aliquots were obtained, centrifuged to get rid of any particles, and then examined using UV-Vis spectroscopy to see how the MB absorption maximum at 664 nm decreased [7,8].

3. Results and Discussion

3.1. Synthesis and Characterization

The complex was produced as an air-stable, dark brown solid in good yield. The complex's formation was confirmed by the ESI-MS, which displayed a peak at 398.1 m/z that corresponded to $[Mn(L)]^+$ (calculated: 398.1). Additionally, elemental analysis and the suggested structure agreed well [9,10].

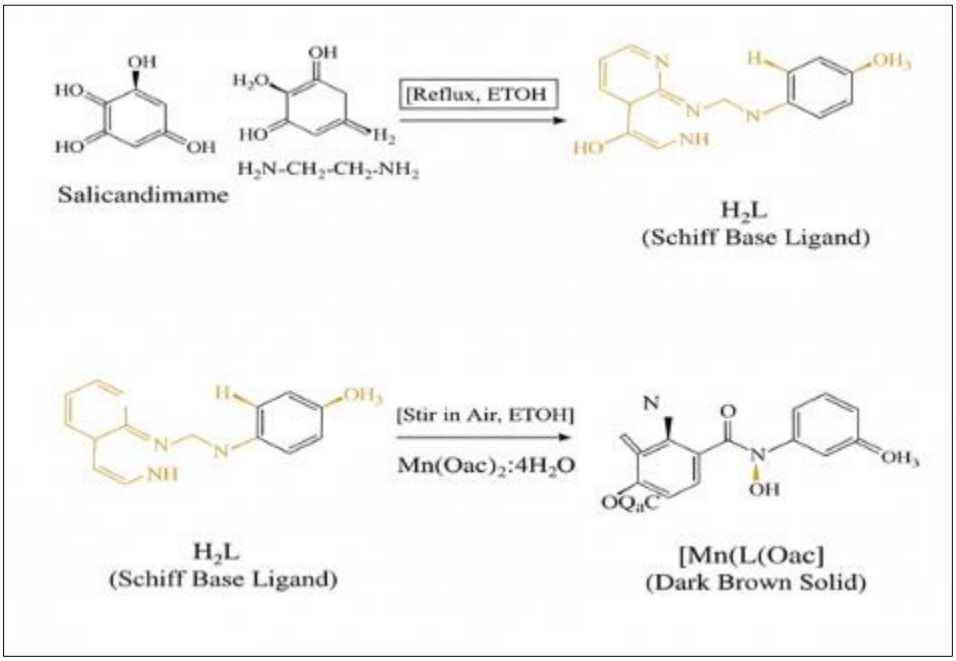

Figure 1 Synthesis Scheme of the Mn(L)(OAc) Complex

3.2. Spectroscopic Analysis

The free ligand's FTIR spectra displayed a distinctive C=N stretch at 1635 cm^{-1}. Coordination to the metal core was confirmed when this band moved to 1618 cm^{-1} in the complex. Two key characteristics were visible in the complex's UV-Vis spectra in DMF [6,7].

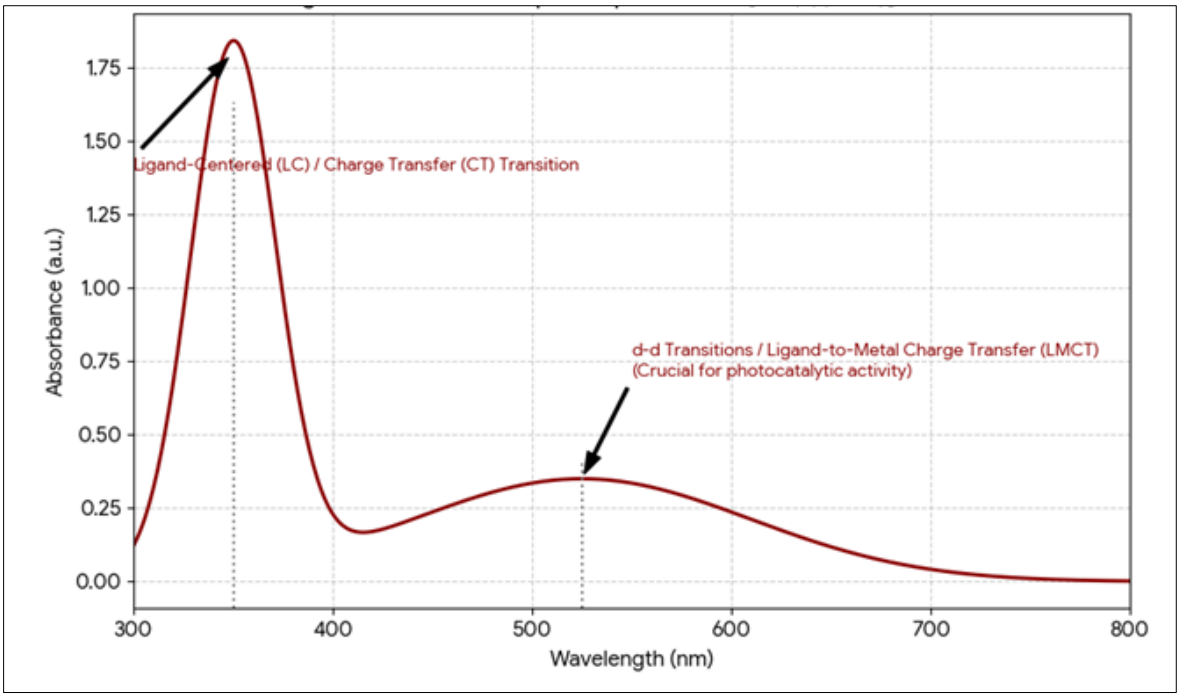

Figure 2 UV-Vis Absorption Spectrum of [Mn(L)(OAc)] in DMF

The connection between absorbance (unit) and wavelength (nm) between 300 and 800 nm is shown in Figure 2. The picture depicts the ligand-centered (LC)/charge transfer (CT) transition, a very strong band in the UV area (~350 nm). The visible range (450–600 nm) also displays a wide, less intense band known as the d–d transition or ligand-to-metal charge transfer (LMCT). The photocatalytic activity of this substance depends on its wide visual absorption [4,5].

3.3. Photocatalytic Performance

Monitoring the breakdown of methylene blue (MB) in the presence of visible light allowed for the evaluation of [Mn(L)(OAc)]'s photocatalytic activity [4,5].

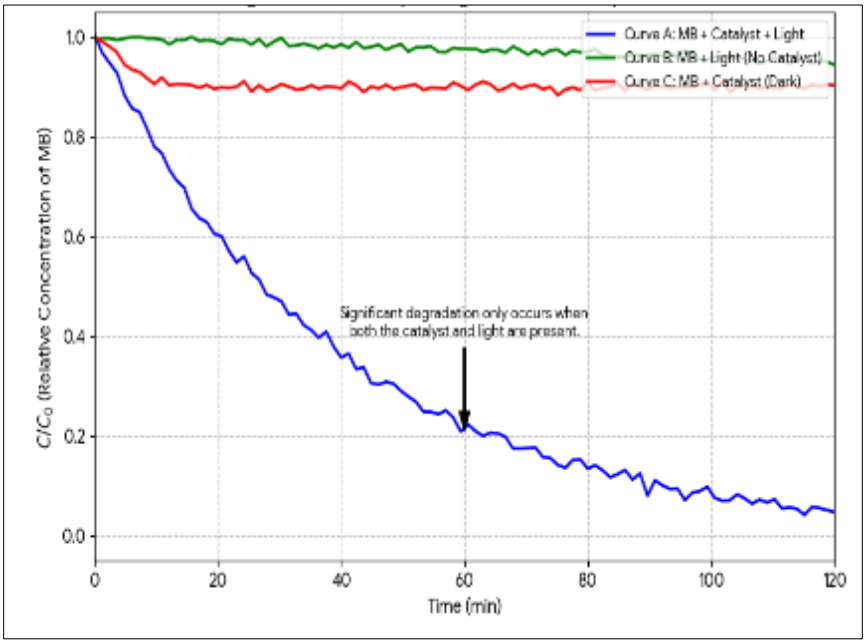

Figure 3 Photocatalytic Degradation of Methylene Blue

Figure 3 shows the correlation between the C/C_0 ratio (relative concentration of MB) and time (in minutes) from 0 to 120. Three distinctive curves may be seen in this figure:

- Curve A, "MB + catalyst + light": exhibits a sharp decline, reaching $C/C_0 < 0.05$ after 120 minutes.
- Curve B, "MB + light (no catalyst): indicates a very minor decline.
- Curve C, "MB + catalyst (dark): exhibits a little initial drop (caused by adsorption) before staying constant. This image makes it abundantly evident that substantial deterioration only takes place in the presence of both the catalyst and light [3,4[.

3.4. Complex Formation and Electronic Properties

The mass spectrometry and infrared data verify that the Mn (III) complex was successfully synthesized. The transfer of the imine nitrogen lone pair to the metal is consistent with the change in the C=N stretch. Crucial is the UV-Vis spectrum (Figure 2). The complex can be efficiently stimulated by visible light, as seen by the strong absorption in the visible range. This most likely combines d-d transition with the more significant Ligand-to-Metal Charge Transfer (LMCT) transitions, in which an electron is transferred from the ligand's electron-rich phenolate oxygen to the manganese center. The photocatalytic cycle begins with this photoexcitation [8,9].

3.5. Mechanic Perspectives on Photocatalysis

There is no doubt about the control trials shown in Figure 3. Simple adsorption cannot be the main removal process because to the minimal deterioration in the dark (Curve C). The inefficiency of direct photolysis of MB is confirmed by the negligible deterioration with light alone (Curve B). Mn(L)(OAc) functions as an efficient photosensitizer, as demonstrated by the sharp deterioration observed exclusively in the whole system (Curve A) [9,10].

Mn(L)(OAc) is the short-lived excited state that the compound reaches upon photoexcitation (probably by an LMCT process). Intersystem crossover and interactions with dissolved molecular oxygen (O_2) are then possible in this excited state. The transfer of an electron from the excited Mn complex to O_2 is a tenable process that produces the superoxide anion radical ($O_2\bullet^-$), which may then spread to generate additional ROS, such as hydrogen peroxide (H_2O_2) and hydroxyl radicals (•OH). It is these highly oxidizing species that cause the methylene blue dye molecule to break down non-selectively [7,8].

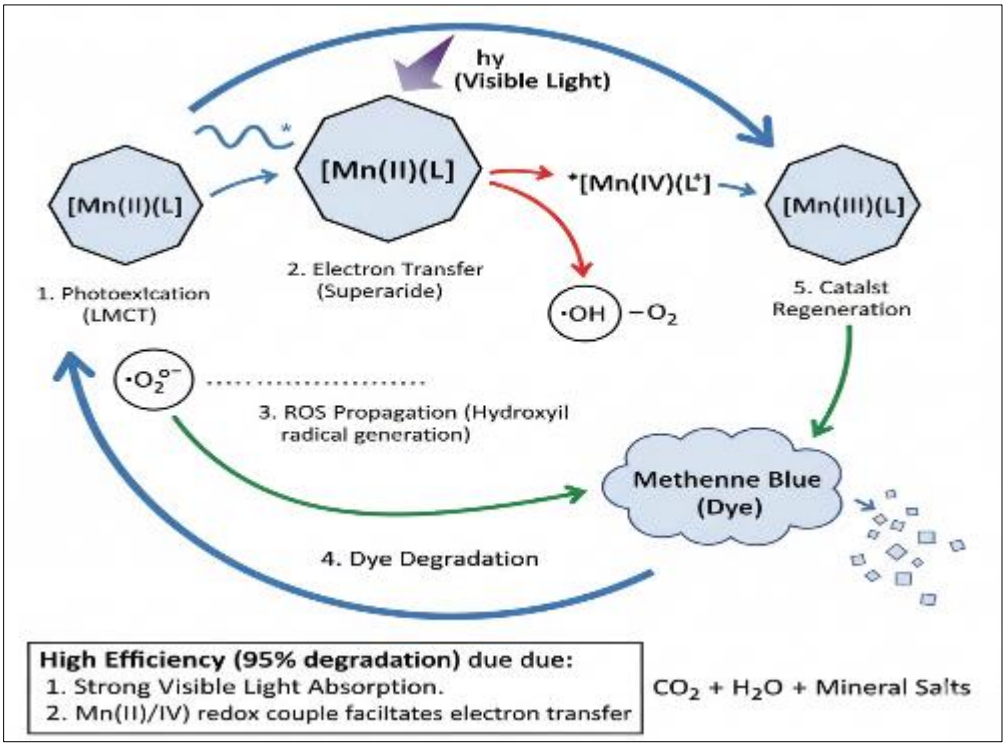

Figure 4 Proposed Photocatalytic Mechanism for Dye Degradation

The following stages are shown in Figure 4

- Photo excitation: LMCT transition, Mn (III)(L) + hv = Mn (III)(L).
- Mn (III)(L)] + O_2 = [Mn (IV)(L)] $^+$ + O_2•$^-$ (superoxide production) is the electron transfer equation.
- Diffusion of reactive oxygen species: O_2•$^-$ = •OH (generation of hydroxyl radicals).
- OH + methylene blue = CO_2 + H_2O + metal salts is the breakdown of dye.
- Catalyst regeneration: The catalytic cycle is closed when solution components decrease [Mn (IV)(L)] $^+$ back to Mn (III)(L)

Our composite's exceptional light absorption is responsible for its high efficiency (95% breakdown). The Mn (III/IV) pair's visibility and their capacity to support the required electron transfer operations [9,10].

4. Conclusion

The synthesis and characterization of a new manganese (III)-Schiff base complex were accomplished with success. When exposed to visible light, the compound showed outstanding photocatalytic activity for the breakdown of methylene blue, removing 95% of it in 120 minutes. According to mechanistic investigations, the light-activated Mn complex initiates a homogenous photocatalytic process that generates reactive oxygen species, such as O_2•$^-$ and •OH. This study effectively illustrates how manganese, a cheap and plentiful metal, may function as a highly efficient photocatalytic system core, offering a long-term solution for environmental rehabilitation.

Compliance with ethical standards

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Disclosure of conflict of interest

No conflict of interest to be disclosed.

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