

Fe(III)-Aqua Complex Mediated Photodegradation of Methylene Blue Dye

Ashfeen Nawar, Md. Aatur Rahman, and Md. Mufazzal Hossain

Abstract—Industrialization in the current times has become extremely rapid. Some of these industries are responsible for discharging dye-containing wastewater into natural water bodies and hence causing environmental deterioration. The purpose of this research is to investigate an inexpensive and easy-to-set-up photodegradation process for the mineralization of methylene blue (MB) dye. The optimum conditions required for maximum degradation of the dye were explored by varying different experimental parameters such as the initial concentration of Fe(III) and dye, pH of the reaction mixture, nature of light sources, and intensity of ultraviolet (UV) light. Approximately 97% photodegradation of methylene blue was recorded at pH 2.30 for the optimum concentration of MB of 3.00×10^{-5} M and Fe(III) aqueous solution of 8.00×10^{-4} M when irradiated under UV light of intensity 3.31×10^{-9} Ein cm^{-3} s^{-1} . Under sunlight, with similar experimental conditions, 73% degradation of the dye was achieved. This is an environment-friendly, efficient, and low-cost degradation process of methylene blue.

Index Terms—Environmental chemistry, Fe(III)-aqua complex, methylene blue, photocatalysis, photodegradation.

I. INTRODUCTION

One of the main causes of environmental pollution worldwide is the discharge of dye-containing wastewater effluents into natural water bodies from textile, pharmaceutical, food processing, paper and pulp, and tannery industries [1]. There are a few reasons why their presence poses such a major threat to the surrounding ecosystem. Many of the dyes and their breakdown products are extremely poisonous and cancer-causing agents. If ingested for a longer period of time, they can be very harmful to living organisms. Moreover, a high concentration of textile dyes in water bodies inhibit the reoxygenation capacity of water as well as prevent sunlight from passing through and reaching the plants and organisms living underwater. As a result, the biological activity and photosynthetic process of aquatic life are greatly impaired [2].

Most of the dyes are not biodegradable because of their aromatic structure, and therefore, without proper treatment, they can persist in the environment for a long time [1], [3], [4]. Therefore, it is crucial to eliminate the organic dyes from wastewater so that their effect on the environment is minimized. There are numerous conventional methods such as biological, physical, and chemical processes or their combinations commonly employed for the removal of these

pollutants, but they all have several drawbacks, e.g., inefficient dye removal, the formation of toxic by-products, expensive, and time-consuming. Moreover, most of these processes only convert the pollutants from one phase to another, leading to secondary pollution [5], [6].

Advanced oxidation processes (AOPs) are alternative to traditional methods that involve the *in situ* generation of hydroxyl radical ($\cdot\text{OH}$). These reactive species are capable of mineralizing a broad range of organic pollutants rapidly and nonselectively [7], [8]. The AOPs have been broadly classified into homogenous and heterogeneous systems. Extensive research work has been done using TiO_2/ZnO as heterogeneous photocatalysts for the degradation of dye and pollutants under an artificial or natural light source. These materials are commonly used because they are photo stable, low priced, and have high redox activity and selectivity [9], [10]. However, one major disadvantage is that it is quite time-consuming and cost-inefficient to separate and recycle the extremely fine catalyst from the wastewater before discharge during large-scale operations.

Several studies have also been carried out using homogenous AOPs such as Fenton's reagent, light-assisted Fenton's oxidation, H_2O_2 /ultraviolet(UV) treatment, and ozonation at high pH [11], [12]. Despite the many advantages of using the Fenton reagent, the oxidation process might not be considered efficient for large-scale applications because of the toxicity and high cost of the oxidant H_2O_2 as well as modulation of pH during the process [13]. The drawback of the O_3 /UV process is that, in addition to it being cost- and energy-intensive, the ability of UV light to penetrate the solution is hindered by the turbidity of the system. Also, compounds such as nitrate are known to obstruct the absorbance of UV light [14].

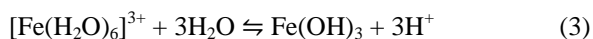
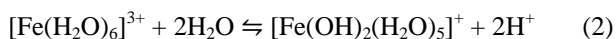
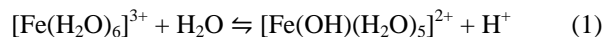
Photodegradation of organic pollutants and dyes under artificial or solar light has been studied using Fe(III) complexes as homogenous photocatalyst. Unlike the Fenton process, it does not involve the use of H_2O_2 , and hence, this AOP is more environment friendly. Among the different Fe(III) complexes investigated as photocatalysts, Fe(III) in aqueous solution (Fe(III)-aqua complex) have several advantages and yielded promising photodegradation results [15], [16]. This process is more economical because Fe(III) ions are easily found and available in the natural environment. Fe(III)-aqua complex is easier to handle and use because there is no necessity to conduct any intensive or complicated pre-/post-treatment of the dye/catalyst. Therefore, the degradation process photoinduced by Fe(III)-aqua complex can be expected to be efficient for the removal of pollutants from aqueous solution. Other Fe(III) complexes have also been found to act as efficient photocatalyst in the degradation

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of pollutants and dyes. The Fe(III)–citrate–oxalate binary system has been used for the successful photodegradation of amitriptyline [17]. Fe(III)–oxalate-mediated photodegradation of organophosphorus pesticide diazinon and para-arsanilic acid have also been studied [18], [19].

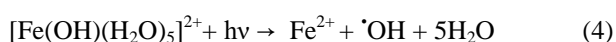
The hydrolysis of Fe(III) in dilute aqueous solution is considered to occur through the following scheme [16], [20]. Coordinated water molecules have been included here.



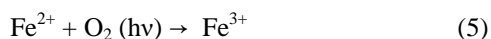
According to a study by Faust and Hoigne [21], $[\text{Fe}(\text{OH})(\text{H}_2\text{O})_5]^{2+}$ is the predominant monomeric Fe(III) hydroxyl complex formed in the pH range of 2.5 to 5.0.

Although other Fe(III) hydroxyl species such as the hexaquo Fe(III) complex are photoreactive, their quantity is insignificant at $\text{pH} \geq 2.5$. Hence, their contribution to the production of $\cdot\text{OH}$ radicals can be ignored. Therefore, by maintaining the pH, $\cdot\text{OH}$ radicals are being generated by irradiation of the major Fe(III)–aqua complex $[\text{Fe}(\text{OH})(\text{H}_2\text{O})_5]^{2+}$.

Upon irradiation, Fe(III)–aqua complex goes through a photochemical process and consequently generates $\cdot\text{OH}$ radicals and Fe(II) through an internal electron transfer.



Fe(II) is capable of being reoxidized by oxidants (e.g., dissolved oxygen) to Fe(III) again [22].



A synthetic organic dye found in wastewater is methylene blue (MB) also known as methylthioninium chloride, $\text{C}_{16}\text{H}_{18}\text{ClN}_3\text{S}$. It is a thiazine dye and is a formal derivative of phenothiazine.

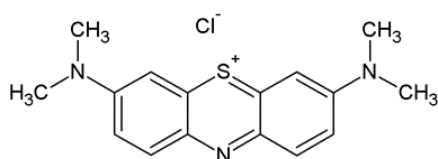


Fig. 1. Methylene blue dye.

In a recent study, a series of CuO catalysts were used for the discoloration of MB in the presence of H_2O_2 [23]. Suhaimy and Abdullah [24] conducted a research on the removal of MB dye from aqueous solution through adsorption using slag from an electric arc furnace. Although the process is fairly efficient in removing MB, the dye still remains in the environment in adsorbed form and can give rise to polluting effects. The slag containing the adsorbed dye would have to undergo further treatment for MB to be eradicated completely. The photodegradation mechanism of MB in water under UV light was investigated by using TiO_2 as photocatalyst. The study concluded that complete mineralization of the dye is possible and the final products of the photodegradation process were identified to be CO_2 , SO_4^{2-} , NH_4^+ , and NO_3^- [25].

To the best of our knowledge, no literature has yet been

found that investigated Fe(III)-mediated photodegradation of MB dye. Herein, we present the study of the catalytic efficiency of Fe(III)–aqua complex as photocatalyst for the mineralization of the MB dye and investigate the optimum conditions required for maximum photodegradation.

II. EXPERIMENTAL

A. Chemicals and Reagents

The analytical grade MB dye and $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ obtained from MERCK, Germany, were used without any further treatments. MB stock solution of 4.0×10^{-4} M was initially prepared, and further dilutions were made whenever necessary. Solutions of $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ were freshly prepared before every experiment. NaOH and HNO_3 were also obtained from MERCK, Germany.

B. Preparation of the Sample

A certain volume of MB and $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ stock solutions were taken in a 100-mL beaker to prepare a reaction mixture of the desired concentration. Because Fe(III) ions form precipitate at a pH higher than 3.50, the pH of the reaction mixture was adjusted and maintained below 3.50 in every experiment. This was done using 0.01 M NaOH and 0.01 M HNO_3 .

C. Photodegradation Procedure



Fig. 2a. UV light source.



Fig. 2b. Visible light source



Fig. 2c. Instrumental setup for irradiation in sunlight.

A typical experiment was done in the following way. The reaction mixture along with a magnetic bar was placed on a magnetic stirrer plate, and the distance of the beaker from the light source was kept constant in every experiment. After every 10 min for 60 min, a certain portion of the irradiated solution was taken out, and then, the absorbance of the taken solution was measured using a UV spectrophotometer. The effect of three different light sources was investigated. The UV radiation source (Helios quartz group) was composed of 10 lamps (Fig. 2a) emitting monochromatic radiation of 320.0 nm. The light intensity of the UV light source was varied using 2, 4, 6, 8, and 10 lights corresponding to intensities 1.03×10^{-9} , 1.21×10^{-9} , 1.36×10^{-9} , 2.27×10^{-9} , and 3.31×10^{-9} Ein $\text{cm}^{-3} \text{s}^{-1}$, respectively [26]. The visible light source was enclosed with a wooden box, inner surfaces of which were covered with aluminum foil paper (Fig. 2b). For solar light, the reactor containing reaction mixture was placed on a magnetic stirrer under sunlight between 12.00 pm and 1.00 pm on sunny days (Fig. 2c) in April 2019 when the temperature was found to be 35 °C–38 °C. The percent degradation was calculated using the equation, %degradation = $\{(A_0 - A)/A_0\} \times 100$ where, A_0 and A are the initial absorbance of dye and absorbance of dye after a certain period of irradiation, respectively [27].

III. RESULTS AND DISCUSSION

A. Absorption Spectra

The absorption spectrum of pure MB solution is shown in Fig. 3. MB absorbed strongly in the visible and UV region with λ_{max} at 664.5 nm. All investigations were carried out at this wavelength to monitor the change of concentration of MB in the subsequent experiments. The molar extinction coefficient of the MB dye solution was found to be 55,470 $\text{L mol}^{-1} \text{cm}^{-1}$ using a calibration curve.

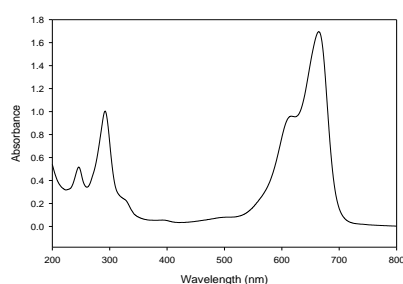


Fig. 3. Spectrum of methylene blue solution (concentration = 3.00×10^{-5} M).

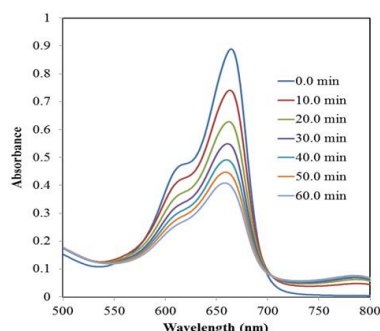


Fig. 4. Spectrum for photodegradation of methylene blue. $[\text{MB}]_0 = 1.50 \times 10^{-5}$ M, $[\text{Fe(III)}]_0 = 8.00 \times 10^{-4}$ M, light source = UV light, light intensity = 3.31×10^{-9} Ein $\text{cm}^{-3} \text{s}^{-1}$, pH = 2.30.

The absorption spectrum of MB during photodegradation is shown in Fig. 4.

B. Effect of the Initial Concentration of Fe(III)–Aqua Complex

Fig. 5 shows the effect of initial concentrations of Fe(III) ions on the photodegradation of MB under UV light. The concentration of Fe(III) solution was varied from 5.00×10^{-4} M to 1.00×10^{-3} M at a regular interval, whereas the concentration of MB and light intensity were kept fixed at 3.00×10^{-5} M and 3.31×10^{-9} Ein $\text{cm}^{-3} \text{s}^{-1}$, respectively.

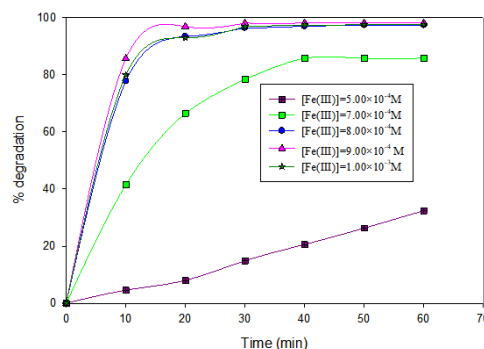


Fig. 5. Photodegradation of MB for various initial concentrations of Fe(III) aqueous solution. $[\text{MB}]_0 = 3.00 \times 10^{-5}$ M, light source = UV light, Light intensity = 3.31×10^{-9} Ein $\text{cm}^{-3} \text{s}^{-1}$.

Fig. 5 reveals that 97.3% (± 0.8) degradation of MB was achieved within 30 min with an initial concentration of Fe(III) $\geq 8.00 \times 10^{-4}$ M, although the same level of degradation was found to take place for $[\text{Fe(III)}]_0 = 9.00 \times 10^{-4}$ M within 15 min time. It clearly indicates that the higher the concentration of Fe(III) ions, the faster the degradation kinetics. Higher concentration of Fe(III) ions facilitate more production of $\cdot\text{OH}$ radicals that initiate the degradation process. This result agrees well with the findings of Li et al [8]. At the lowest initial concentration of Fe(III), only 30% of degradation occurred in 60 min time. Because for this particular source, light intensity reaching the reaction mixture in the reactor is constant, the excess of Fe(III) ions remain ineffective at concentrations higher than 8.00×10^{-4} M, and hence, a limiting value of degradation is reached at approximately $[\text{Fe(III)}]_0 = 8.00 \times 10^{-4}$ M.

C. Effect of the Initial Concentration of MB

In order to determine the effect of initial dye concentration on the percent degradation of dye, experiments were carried out varying the initial concentration of MB from 1.50×10^{-5} M to 3.50×10^{-5} M, whereas the concentration of Fe(III) ions was kept fixed at 8.00×10^{-4} M, and the pH of the solution was adjusted to 2.30. In each experiment, the reaction mixture was irradiated with UV light, keeping the intensity fixed at 3.31×10^{-9} Ein $\text{cm}^{-3} \text{s}^{-1}$. The results are shown in Fig. 6.

As seen, the optimum condition of the initial concentration of MB is in the range of 2.50×10^{-5} M to 3.00×10^{-5} M for the nearly complete degradation ($\approx 97\%$) of the dye. As the initial concentration of MB is increased from 1.50×10^{-5} M to 3.00×10^{-5} M, the percent degradation of the dye increased from $\approx 55\%$ to $\approx 97\%$. At the highest initial concentration of 3.50×10^{-5} M, only 20% degradation took place in 60 min time, whereas $\approx 97\%$ degradation occurred at the optimum

condition of the initial concentration in 15 min. The initial increase in degradation with increasing concentration of MB is due to the greater number of dye molecules available for $\cdot\text{OH}$. The decrease in degradation with an increase of dye concentration after the optimum value might be because a fixed amount of Fe(III) catalyst produces a fixed amount of $\cdot\text{OH}$ radicals, and these radicals can only attack a proportional amount of MB. This is consistent with the findings of Banat et al. [28] in their investigation of the photodegradation of MB dye in aqueous solution using a laboratory-scale UV lamp in the presence of H_2O_2 . They suggested that when MB is present at a higher level in the reaction mixture, the available free radicals generated from a particular amount of catalyst are not adequate for the photodegradation process.

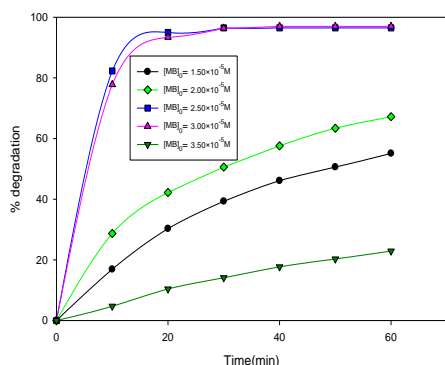


Fig. 6. Photodegradation of MB for various initial concentrations of MB. $[\text{Fe(III)}]_0 = 8.00 \times 10^{-4} \text{ M}$, light source = UV light, light intensity = $3.31 \times 10^{-9} \text{ Ein cm}^{-3} \text{ s}^{-1}$, pH = 2.30.

D. Effect of pH

The effect of pH on the photodegradation of MB in the presence of Fe(III)–aqua complex as a photocatalyst under UV irradiation was investigated, and the results are shown in Fig. 7.

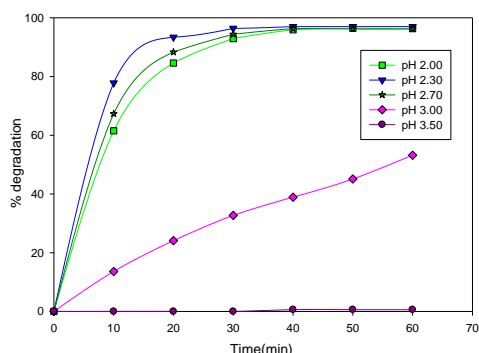


Fig. 7. Photodegradation of MB at various pH. $[\text{MB}]_0 = 3.00 \times 10^{-5} \text{ M}$, $[\text{Fe(III)}]_0 = 8.00 \times 10^{-4} \text{ M}$, light source = UV light, light intensity = $3.31 \times 10^{-9} \text{ Ein cm}^{-3} \text{ s}^{-1}$.

Fe(III) ions are well known to form precipitate at a pH higher than 3.50. So the photodegradation of MB dye was investigated over the pH range of 2.00–3.50 while keeping all other variables – initial concentration of MB and Fe(III) solution, light source, and light intensity – constant. Fig. 7 reveals that 97% of degradation occurred between pH 2.00 and 2.70 within 40 min time. However, at pH 2.00,

photodegradation is slightly slower than that at pH 2.30. The most rapid and maximum degradation was observed at pH 2.30, which might be due to the production of the highest concentration of $[\text{Fe(OH)(H}_2\text{O)}_5]^{2+}$ species that eventually makes $\cdot\text{OH}$ radical more available. No photodegradation was observed at pH 3.50, which is likely due to precipitation of Fe(OH)_3 . Our result agrees well with the findings of Hossain et al. [29] who investigated the photocatalytic effect of Fe(III)–aqua complex on photodegradation of orange green dye. They reported that maximum degradation occurred at pH 2.5.

E. Effect of Light Sources on Photodegradation

A mixture of an aqueous solution of MB ($3.00 \times 10^{-5} \text{ M}$) and Fe(III) ($8.00 \times 10^{-4} \text{ M}$) was exposed to natural sunlight. The reaction mixture was also irradiated with UV light ($3.31 \times 10^{-9} \text{ Ein cm}^{-3} \text{ s}^{-1}$) and visible light. The pH of the reaction mixture was kept fixed at 2.30 in all three experiments. Fig. 8 represents the comparison of the percent degradation of MB that occurred in three different light sources.

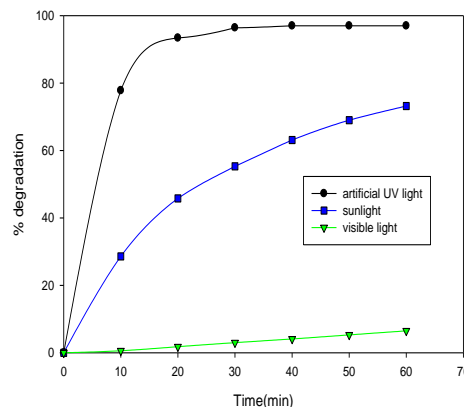


Fig. 8. Photodegradation of MB for various light sources. $[\text{MB}]_0 = 3.00 \times 10^{-5} \text{ M}$, $[\text{Fe(III)}]_0 = 8.00 \times 10^{-4} \text{ M}$ and pH = 2.3.

It is seen that the degradation of MB dye in visible light is negligible after irradiating the reaction mixture for 60 min. But when the reaction mixture was irradiated with UV light, approximately 97% of the dye was degraded after 30 min. This is because the absorption of UV photons induced the photocatalytic reaction of MB [30]. It has been reported [24] that upon irradiation at 365 nm, the light is absorbed by Fe(III) species and thus facilitates the formation of $\cdot\text{OH}$ radicals. Under sunlight, approximately 73% degradation of the MB dye occurred within 60 min. In comparison, approximately 50% degradation of tributyltin chloride (TBT) in a mixture of TBT and Fe(III) solution was found to occur in solar light after 2 h [20].

F. Effect of Intensity of UV Light

Because maximum degradation was observed under the UV light source as shown in Fig. 8, the effect of UV light intensity on photodegradation of MB was investigated. The UV light intensity was varied from $1.03 \times 10^{-9} \text{ Ein cm}^{-3} \text{ s}^{-1}$ to $3.31 \times 10^{-9} \text{ Ein cm}^{-3} \text{ s}^{-1}$ at a regular interval. Concentrations of MB and Fe(III) aqueous solution were fixed at $3.00 \times 10^{-5} \text{ M}$ and $8.00 \times 10^{-4} \text{ M}$, respectively, and the pH of the solution was maintained at 2.30. The results are shown in Fig. 9.

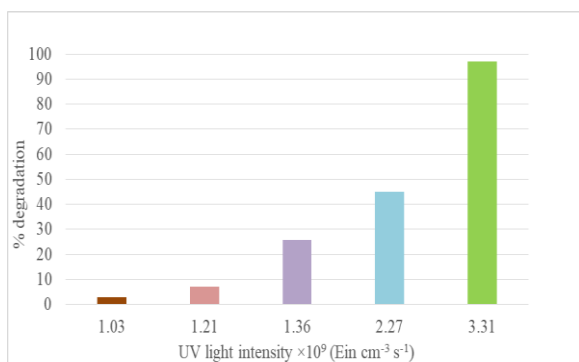


Fig. 9. Photodegradation of MB for various intensities of UV light. $[\text{MB}]_0 = 3.00 \times 10^{-5} \text{ M}$, $[\text{Fe(III)}]_0 = 8.00 \times 10^{-4} \text{ M}$ and $\text{pH} = 2.30$.

With the increase of light intensity, the number of photons also increases, which enhances the production of $\cdot\text{OH}$ radicals. A higher amount of $\cdot\text{OH}$ radicals causes more efficient degradation. Hence, Fe(III) -aqua complex-mediated photodegradation increases with the increase in the intensity of UV light.

IV. CONCLUSIONS

Fe(III) photoinduced degradation was found to be affected by various parameters such as concentration of Fe(III) ions and MB dye, pH , the intensity of UV light, and the type of light source.

In the Fe(III) aqueous system, with increasing initial concentration of Fe(III) solution, the percent degradation of MB increased and reached a limiting value at $8.00 \times 10^{-4} \text{ M}$. With increasing initial concentration of dye, degradation was found to increase, reach an optimum value of $3.00 \times 10^{-5} \text{ M}$, and then decrease. The degradation process was found to be efficient at lower pH , and maximum degradation occurred at $\text{pH} 2.30$.

Percentage degradation of 97% of MB was found by irradiation in UV light for 60 min. In comparison, 73% degradation of the dye was observed under sunlight with the same reaction conditions. Hence, it can be concluded that the MB dye can be readily degraded using Fe(III) -aqua complex as photocatalyst if kept under sunlight for a longer period of time without the use of any expensive UV light source. Therefore, this process is much more environment friendly and cost-effective. Negligible percentage degradation of the dye was observed under visible light.

The concentration of Fe(III) used in the process to achieve maximum degradation is quite low, so the treated solution can be expected to be compatible with the aquatic environment. The acidity of the treated effluent can easily be neutralized using sodium bicarbonate solution. Thus, in recent times when environmental deterioration is rapidly becoming the cause of utmost concern to humankind, this photocatalytic system could be a useful technology for the mineralization of MB and similar dyes and limit water pollution to some extent.

CONFLICT OF INTEREST

The authors declare no conflict of interest

AUTHOR CONTRIBUTIONS

Ashfeen Nawar conducted the laboratory work, analyzed the data, and wrote the paper. Md. Ataur Rahman cosupervised the research work. Md. Mufazzal Hossain supervised the research and reviewed the paper. All authors approved the final version.

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