

Available Online

JOURNAL OF SCIENTIFIC RESEARCH

J. Sci. Res. 1 (1), 108-120 (2009) www.banglajol.info/index.php/JSR

Degradation of Commercial Textile Dye by Fenton's Reagent under Xenon **Beam Irradiation in Aqueous Medium**

M. M. Rahman^{†1}, M. A. Hasnat[#], and Kazuaki Sawada[†]

[†]Venture Business Laboratory, Department of Electrical and Electronic Engineering, Toyohashi University of Technology, Toyohashi, Aichi 441-8580, Japan

^{*}Department of Chemistry, Shah Jalal University of Science and Technology, Sylhet 3114, Bangladesh

Received 21 August 2008, accepted in final form 28 October 2008

Abstract

Degradation of commercial textile dye named Malachite green (MG) has been investigated by Fenton reagent under xenon beam irradiation (1501~1532 lux, $\lambda < 320$ nm) in an aqueous solution. The degradation process was initiated by the photolysis of Fe(III)-hydroxyl species, and accelerated by xenon beam irradiation, due to enhance photolysis of Fe(III) species, which enhances the regeneration of Fe(II) with concomitant production of hydroxyl free radicals. Influences of various experimental parameters, such as the concentration of H₂O₂, Fe (III), xenon irradiation source, and pH of the experimental solutions on the initial rate and photo-degradation extent of the MG dye degradation were assessed and optimized. Although the initial rate of degradation was not affected by the initial MG concentration, it was affected by the concentration of Fenton reagents [Fe(III) and H_2O_2 solution], pH of the experimental solutions, and the intensity of the xenon beam. A significant enhancement of the initial rate and extent of degradation of MG dve was observed at solution pH of \sim 3.1 under xenon beam radiation. Complete degradation of MG dye (>99.5 %) was achieved by xenon beam/Fenton's reagent process in aqueous solution (pH, 3.1).

Keywords: Malachite green; Organic dye, Dye mineralization, Fenton reagents, Xenon radiation.

©2009 JSR Publications. ISSN: 2070-0237 (Print); 2070-0245 (Online). All rights reserved. DOI: 10.3329/jsr.vlil.1059

1. Introduction

The untreated dyes in effluents from dyeing factories and leather industries are a group of hazardous chemicals as well as major sources of water pollution. The hazards are rendered more acute by their wide-spread presence [1, 2]. Dyes undergo chemical changes as well as biological changes in the aquatic system, consume dissolved O₂ and thus disturb the aquatic eco-system [3]. Consequently, survival of fishes and other lives becomes difficult

¹Corresponding author: mmrahmanh@gmail.com; mmrahman@vbl.tut.ac.jp

in wide spread, especially in environmental water. In presence of these organic compounds, water becomes unusable for practical uses. In many countries, most of the textile industries are using colour materials like dyes and pigments, and effluents are released as a textile effluent to canal, river, and sea without further treatments and purifications [4, 5]. It is well known that dyes and their degradation products are highly toxic, carcinogenic, and caused water pollution. It is therefore necessary to treat the water containing colour dyes and other organic compounds to discharge them [6].

Due to the complex aromatic structure and stability of these dyes, conventional biological pre-treatment methods are ineffective for degradation [7]. Also low efficiency and low reaction rate are associated with these methods caused the methods to be ineffective. Over the past few years, a number of physical and chemical techniques had been reported for the treatment of dye effluent including electrochemical oxidation, active sludge, flocculation, reverse osmosis and adsorption on activated carbon [8-13]. In most cases, the degradation is conducted for dissolved compounds in water with UV radiation. The Fenton process uses Fe²⁺ to react with H₂O₂ to generate radicals (OH) with powerful oxidizing ability. This process in the presence of visible light and UV-radiation in different temperatures has been used previously for the mineralization of organic pollutants [14-19]. Although hydroxyl radicals are powerful species, only Fenton reagent could not reach deep mineralization of organic dyes [1, 16]. Fenton reaction is powerful at initial stage and gradually loses its capability for pollutants degradation with reaction time [17]. Such deactivation is due to the interaction of Fe (III) with degradation intermediates, which is unfavourable to either Fe (III)/Fe(II) recycling [18] or to hydroxyl free radicals generation [19]. It is well known that Fenton reaction could be accelerated by UV light irradiation, due to photolysis of ferric species, which enhances the regeneration of Fe²⁺ with concomitant production of OH. Thus, it is worthwhile to see how these textile dyes could be degraded in aqueous medium under xenon beam irradiation in the presence of Fenton reagent. Accordingly, our attempt was to use the xenon beam (wavelength $\lambda \leq 320$ nm and intensity $1501 \sim 1532$ lux) with Fenton's reagent to mineralize textile dyes. Malachite Green was employed as model dye.

In this study, we report on an easy and simply method based on xenon beam/Fenton reagent for the mineralization of Malachite green. The degradation process was initiated by the photolysis of Fe(III)-hydroxyl species, and accelerated by xenon beam irradiation, due to enhance photolysis of Fe(III) species, which enhances the regeneration of Fe(II) with concomitant production of hydroxyl free radicals. Influences of various experimental parameters, such as the concentration of H_2O_2 and Fe (III), xenon irradiation source, and pH of the experimental solutions on the initial rate and photo-degradation extent of the MG dye degradation were assessed and optimized.

2. Experimental

2.1. Materials

Malachite green oxalate dye (M.W. 927.03, 96%) (Fig. 1a) and hydrogen peroxide (32%)

were purchased from Sigma Aldrich, USA. Ferric sulphate n-hydrate (98%) was purchased from Showa Chemicals Inc., Japan. Other chemicals were in reagent grade and used as received without further purification. All solutions were prepared with de-ionized water (Resistivity, >18.2 M Ω -cm).

2.2. Apparatus and Method

The photochemical experiments were carried out at 25 ± 0.2 °C. The xenon light was used from the xenon radiation source from ICL technology, Model PS300-1, USA. Absorbance and pH of the solution were measured using Full-Wave Spectrophotometer (Ocean optics ICS, USA) and pH meter (PHS-25, China), respectively. MG dye degradation was carried out both in the dark and in the presence of xenon light. All experiments were conducted in a cylindrical Pyrex vial under magnetic stirring in an aerated solution at pH of ~ 3.1 (adjusted with HCl). All solutions were prepared fresh daily. The stock solution of Fe(III) sulphate (1.0 mM), MG (0.15 mM), and H₂O₂ (1.0 mM) was prepared using deionized water. The experimental concentration of H_2O_2 measured before performing experiments. The pH of the solutions was controlled by adding appropriate amount of acid solution. A reaction mixture of 100 mL, pH 3.1, was prepared by taking requisite amount of MG, Fe(III) sulphate, and H_2O_2 solutions of known concentration. From this solution, half of the solution (50.0 mL) mixture was transferred into a parallel beam photochemical reactor for photolysis under xenon beam and the rest of the solution was kept in the dark. The chemical changes (i.e., absorbance) were measured for both solutions under dark and xenon beam condition in every 10 minutes intervals with the spectrophotometer.

The extent of the dye degradation under xenon beam and dark condition by the proposed method depend on the different experimental parameters, such as the concentration of H_2O_2 , Fe(III), and dye solutions, and pH of the experimental media. Therefore, experimental parameters affecting the analytical performance of the MG degradation were assessed and optimized. The initial rate of MG degradation was calculated from the slope of the tangent drawn at the initial absorbance of dye versus time profile. Initial rate of degradation,

$$v = - d[MG]/dt = - (1/l.\varepsilon.) dA/dt$$
(1)

where, A = absorbance of MG, $\varepsilon =$ molar absorption coefficient of MG, and l = length of cell. The percent (%) of degradation (mineralization) was determined by following the Beer Lambert law,

$$A = \varepsilon. \text{ [MG]. } l, \text{ (for dilute solution)}$$
(2)

Since ε and l are constant, then

$$A \alpha [MG], A_o \alpha [MG]_o, and A_t \alpha [MG]$$
(3)

Therefore, percent (%) of degradation is calculated using the equation

% degradation=
$$(A_0 - A_t) 100/A_o$$
 (4)

3. Results and Discussion

3.1. Characterization of MG dye

MG is a cationic dye and extensively used in dyeing and textile industries. The molecular structure and spectrum of MG in aqueous solution are shown in Fig.1. As shown in Fig 1b, MG shows two peaks at 426 and 617 nm in aqueous solution. At 617 nm, the molar absorption coefficient (ε) of MG at 25 °C is 1.55×10^5 L mol⁻¹ cm⁻¹. The MG dye was stable and no significant change in the spectrum was observed over the pH range of 3.0-4.5. At pH values of <3.0 and >4.5, the absorbance of MG was decreased. The points to be noted for dye mineralization by xenon beam/Fenton process are as follows: (i) lower concentration of MG should be used because of its high molar extinction coefficient, and (ii) the initial rate and extent of degradation depends on the concentration of H₂O₂, Fe (III), MG, temperature of the solution, and the xenon radiation source.

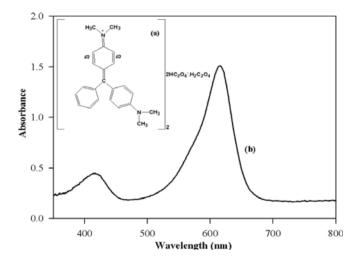


Fig. 1. (a) The molecular structure of MG dye. (b) A typical spectra of MG dye in aqueous solution (the concentration of dye solution, 0.01 mM (pH, 3.1).

3.2. Degradation of MG dye

The degradation of MG has been investigated spectrophotometrically to study whether mineralization occurs when the dye solution was treated with xenon beam/Fenton reagents in aqueous solution at 25 °C (pH, 3.1). Fig. 2 shows the degradation of MG dye under xenon beam and dark conditions with FR reagents treatment. Although the Fenton degradation of MG in the dark solution could be easily occur (Fig. 2a, solid line), the extent of such degradation was about 5-times less than that obtained under xenon beam/Fenton process. When the MG solution was irradiated by xenon beam, the MG degradation occurred continuously, and about 99% degradation occurred within 60 min

irradiation time (Fig. 2a. dash line). Fig. 2b (i) show a typical spectrum obtained for 0.1 mM MG dye solution in aqueous system at pH of 3.1 before irradiating by xenon beam. The adsorption peaks corresponding to the dye solution gradually diminished and finally disappeared under 60 min irradiation by xenon beam in the presence of Fenton reagents, indicating that 60 min irradiation time was sufficient to complete the mineralization of MG dye. The Fenton degradation of MG dye under the xenon beam irradiation was initiated by the photolysis of Fe(III)-hydroxyl species, other than through the electron transfer from the excited dye into ferric species.

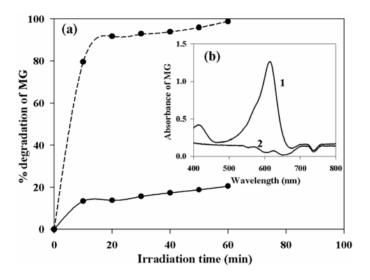


Fig. 2. (a) Degradation of MG dye in aqueous solution (pH 3.1) under dark (solid line) and xenon beam (dotted line) condition. Degradation conditions: pH of the solution 3.1, degradation time 60 min, and concentration of MG dye, Fe (III), and H_2O_2 were 0.1, 0.1, and 1.0 mM, respectively. (b) Typical spectrum obtained before (i) and after (ii) 60 min degradation under xenon beam/Fenton reagents process.

The mechanism of hydroxyl free radical generation is as follows: In presence of ferrous iron,

$$H_2O_2 + Fe(II) \rightarrow HO + OH + Fe(III)$$
(5)

In presence of ferric iron,

 $H_2O_2 + Fe(III) \rightarrow HOO^{-} + H^{+} + Fe(II)$ (6)

$$2HOO \rightarrow 2HO + O_2 \tag{7}$$

$$RH + HO \rightarrow R + H_2O \tag{8}$$

The free radical (R) scavenges oxygen molecules from environment and form highly reactive peroxy radical (ROO), and in presence of Fe(II), it is converted into stable products [20,21]. Recycling Fe (II) from Fe (III) or its complexes dominated the production of hydroxyl free radicals.

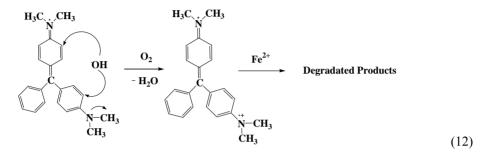
The mechanism of MG degradation is as follows:

$$HO' + ZH (MG dye) \rightarrow Z' + H_2O$$
(9)

$$Z' + O_2 \rightarrow ZOO' \tag{10}$$

 $ZOO + Fe(II) \rightarrow degraded product$ (11)

The mechanism of degradation large dye molecules is presented in Eq. (12).



There is also a possibility of decomposition of H_2O_2 .

$$2H_2O_2 \rightarrow 2H_2O + O_2 \tag{13}$$

Upon analysing the end products, pH was found to be decreased and the presence of the NH_4^+ , NO_3^- , and NO_2^- ions and evaluation of CO_2 were detected. Unidentified Fe(II) and Fe(III) complexes were also observed in the degraded products. These results indicate the complete mineralization of dye occurred via splitting into CO_2 , H_2O , NH_4^+ , HNO_2 , HNO_3 , unidentified Fe(II) and Fe(III) complex.

The rate of reaction (6) is dependent on the concentration of H_2O_2 and Fe(III) in Fenton reagent. Increasing any of these two not only increases the initial rate, but also increases the extent of dye degradation. Radiation and temperature are driving the reaction (6) to the forward direction and accelerate the formation of HO radical. Although dyes were degraded in the dark in presence of FR but the initial rate of degradation and the extent of degradation were enhanced by xenon light with FR. In this study, Fe(III) and H_2O_2 were used as the precursor of the radicals (OH), which ultimately degraded the MG dye in presence of xenon radiation.

3.3. Effect of Fenton reagents on the degradation of MG dye

Although there is no direct evidence for radical participation in the dye degradation, several indirect observations can lead to the conclusion that MG dye is oxidized by the hydroxyl radicals, which are generated in direct photolysis of ferric species [22]. It is well know that the ferric ion can catalyse H_2O_2 decomposition even in the dark and produce hydroxyl radical, and independent of organic molecules (Eq. 6). Accordingly, the concentration of hydroxyl radical (OH) is directly proportional to the concentration of

hydrogen peroxide and Fe (III) solution in Fenton reagents. Firstly, we tested the photomineralization of MG was under different concentration of H_2O_2 . The initial rate and extent of the degradation of MG at 617 nm increased with increasing the concentration of H_2O_2 (Fig. 3, Table 1). An improved behaviour in MG degradation observed under xenon beam/Fenton reagent process (Fig. 3a, dashed curves) when compared with MG degradation in the dark condition (Fig. 3a, solid line). Fig 3c shows typical spectra obtained at different concentration of H_2O_2 . It is noted that MG decomposition was terminated near 60 min of xenon beam irradiation and extent of degradation was calculated to be ~ 98.5 %.

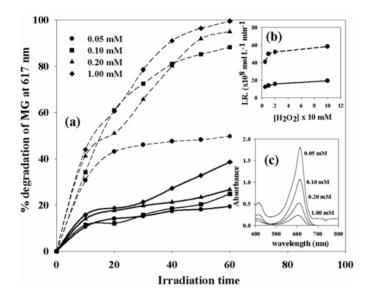


Fig. 3. Effect of H_2O_2 concentration on the (a) degradation and (b) initial rate of MG dye in aqueous solution under dark (solid line) and Xenon/Fenton reagent process (dotted line). (c) Typical spectrum obtained with 0.05, 0.1, 0.2, 1.0 mM H_2O_2 . Other experimental conditions: the pH of the solution, 3.1; degradation time, 60 min, and concentration of MG dye and Fe (III) solutions were 0.1 mM. Total volume of the solution is 50ml.

Secondly, the concentration of Fe (III) in Fenton reagents also has influenced in MG degradation in aqueous media (pH 3.1). As shown in Table 1, the initial rate and the extent of degradation of MG were increased with increasing the concentration of Fe (III), suggesting almost complete decomposition (99.2 %) of dye achieved with 1.0 mM Fe(III) solution. These results indicated that the decomposition of MG dye initiated by the hydroxyl radicals (HO[•]) formed in the Fe(III) photo-redox reaction followed by the Eq. (6), other than via the reaction between the Fe(III) and excited dye or via the electron transfer from MG to the Fe(III) centre in their complex [23,24].

A particular amount of FR reagent produced a definite amount of hydroxyl radicals (OH). These free radicals are reacted only the proportional amount of MG molecules [25].

Accordingly, the initial rate increased and the extent of the degradation decreased as concentration of MG increased (Table 1). However, the spectrum of MG was unaffected during increasing the concentration of MG (not shown). It is noted that 99.2 % mineralization was achieved within 60 min when the concentration of Fe(III), H_2O_2 , and MG dye was 1.0, 0.1, and 0.1 mM (10 mol : 1 mol: 1mol), respectively, in an aqueous solution (pH 3.1). These results indicate 10 mol of H_2O_2 and 1 mol of Fe (III) molecule are needed to mineralize 1 mol of the MG dye molecule.

Table 1. Effects of concentrations of H_2O_2 , Fe(III), and MG on degradation of MG in aqueous medium at pH 3.1.^a

Conc. of Fe(III) (mM)	Conc. of H ₂ O ₂ (mM)	Conc. of MG (mM)	Extent of degradation (%) in 60 min		Initial rate (mol L ⁻¹ min ⁻¹)	
			Dark	Xenon beam	Dark	Xenon beam
0.10 0.15 0.50 1.0	0.10	0.10	26.7 38.1 53.7 66.4	72.0 91.4 96.6 99.2	14.3 34.0 34.3 36.2	43.3 48.1 53.1 60.0
0.10	0.05 0.10 0.20 1.0	0.10	19.4 24.8 26.7 38.6	49.7 75.2 95.9 98.5	12.2 13.8 15.7 19.4	40.7 45.8 52.1 58.2
0.10	0.10	0.01 0.05 0.10 0.15	64.6 26.7 21.9 16.4	96.6 79.3 71.9 68.9	8.15 10.6 14.2 17.6	16.9 33.1 48.1 53.0

^a Degradation time, 60 min.

3.4. Non-complexation between MG and Fe(III) ions

To test the possibility of Fe (III)-MG complex formation, we examined the adsorption spectra of MG dye in the presence of different concentration of FeCl₃ in an aqueous solution. No change in the absorption spectrum was observed upon analysing these mixture solutions. Furthermore, no decrease or increase in absorbance spectra at 617 nm and no shifting in the peak was observed even after keeping the MG solution with the Fe(III) solution for several days, indicating that Fe(III)-MG complexation is not significant under the experiment condition. This was possibly due to the inappropriate configuration of the lone pairs of N and O atoms in MG molecules [14]. Similar experiments were also carried in the presence of $Fe_2(SO_4)_4$, $Cu(NO_3)_2$, and $Fe(NO_3)_3$, but none of these cases changed the MG spectrum, indicating that Fe (III) or Cu(III) did not for any complex with MG molecule.

3.5. Effect of pH

The photo-degradation of MG was affected by pH of the experimental solution. The photo-degradation was performed at different pH values from 1.5 to 5.9 in the xenon beam/Fenton reagent process. It was found that λ_{max} and absorbance of MG spectrum was shifted with varying the pH of the solutions. Absorbance was increased rapidly with the increase in initial pH values (Fig. 4A). Further increase in the pH values resulted in a decreases in absorbance. It is noted that better λ_{max} of MG was found in the pH range 3.0 ~ 4.5. At <pH 3.0, H₂O₂ undergoes auto decomposition to form H₂O and O₂. On the other hand, ionic iron formed colloidal ferric species as Fe(OH)₃ at >pH 4.5. This tendency was checked to be consistence with the previously reported Fenton degradation of dyes [26]. Fig. 4B shows the effect of the pH on the degradation of the MG dye and initial rate. As shown in Fig 4B, the extent of degradation and initial rate was maximum at pH 3.1. These results clearly indicated that maximum hydroxyl free radical was generated at solution pH of 3.1. Thus, all subsequent experiments were carried out at the solution pH of 3.1.

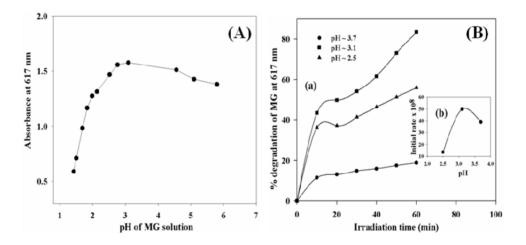


Fig. 4. (A) Effect of pH on the absorbance of MG dye in aqueous solutions. Typical spectrum obtained at the solution pH of 1.5, 1.7, 1.9, 2.0, 2.2, 2.3, 2.6, 2.8, 3.1, 4.5, 5.1, and 5.9. (B) Effect of pH on the degradation (a) and initial rate (b) of MG in aqueous solution under xenon/Fenton reagent process. Dye degradation conditions: degradation time 60 min, and concentration of MG dye, Fe(III), and H_2O_2 were 0.1, 0.1, and 0.15 mM, respectively. Total volume of the solution is 50ml.

3.6. Effect of light sources

MG does not undergo any degradation by photolysis in the xenon beam (wavelength, $\lambda \le$ 320 nm and intensity, 1501 ~ 1532 lux) in absence of FR reagent. In a control experiment, photo-degradation of MG dye was tested for an hr without Fenton reagents in visible and xenon light irradiation, but showed no observable or significant changed in adsorption spectra. This indicated that no mineralization of MG dye was observed upon visible or

xenon light irradiation in the absence of Fenton reagents. However, the photo-Fenton degradation of MG could be occurred under visible light irradiation. This degradation was possibly initiated from the hydroxylation of the dye molecules by 'OH radical generated from the Fe³⁺ assisted decomposition of H₂O₂. The degradation of dye caused due to opening of conjugated rings of hydroxylated dye molecules [27].

In the presence of the Fenton reagents, the initial rate and extent of the MG dye degradation accelerated with increasing the irradiation time, whereas in the visible light or in the dark condition much lower degradation was observed (Fig. 5).

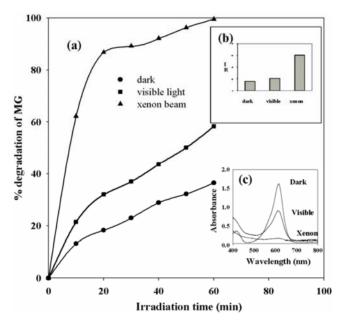


Fig. 5. Effect of light sources on the degradation (a), initial rate (b) and (c) intensity of the absorbance of MG dye in aqueous solution. The concentration of MG dye, Fe (III), H_2O_2 solutions were 0.01, 0.1, and 0.1 mM, respectively. Other experimental conditions were same as Fig. 4.

The extent and the initial rate of degradation within 60 min under the xenon beam irradiation were 99.5% and 60.6×10^{-8} mol L⁻¹ min⁻¹, respectively. In contrast, these values were 36.5% and 15.5×10^{-8} mol L⁻¹ min⁻¹, respectively, in dark condition. We anticipated that xenon beam enhanced the rate of degradation by increasing the production of hydroxyl radicals (OH) in the reaction media. Moreover, it enhanced the temperature of the reaction solution which accelerated the reaction between the exited MG molecules and OH radicals. In summary, MG could be photo-degraded completely within 60 min by the xenon beam/Fenton process, whereas it took longer time upon visible light irradiation or in dark conditions. These suggest that the faster degradation of MG dye by the present method, which could be tentatively explained taking into account the effect of xenon beam upon the 'OH radical's production and temperature of the solution.

118 Degradation of Commercial

3.7. Degradation of the MG dye under different reaction conditions

A series of experiments was performed at seven different experimental conditions to check the analytical advantages of the xenon beam/Fenton reagent process in terms of degradation extent and time. The results of these experiments were shown in Fig 6. In the absence of Fenton reagent/xenon beam irradiation process, 0.01 mM MG dye solution did not show any degradation. Similarly, solution temperature alone does not have any significant effect on the MG dye degradation.

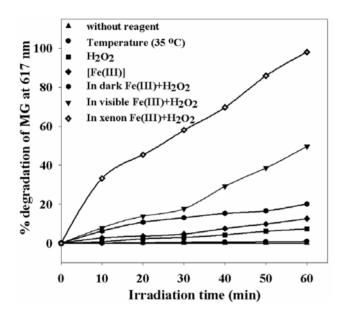


Fig. 6. The degradation of MG dye under different experimental conditions: \blacktriangle , blank analysis (0 mM Mg dye + 0 mM H₂O₂ + 0 mM Fe(III)); •, at 35 ^oC solution temperature (0.01 mM MG); •, with 1.0 mM H₂O₂ (0.01 mM MG); □, with 0.1 mM Fe (III) solution (0.01 mM MG dye); , \triangle , and \diamond are the Fenton reagent processes in dark, visible light, and xenon beam, respectively. The concentration of MG dye, H₂O₂, and Fe (III) in Fenton reagents processes is 0.01, 1.0, 0.1 mM, respectively.

No substantial degradation of 0.01 mM MG dye was observed upon increasing the solution temperature at 35 °C without using Fenton reagent/xenon beam irradiation. In the presence of H_2O_2 or Fe (III) alone (without xenon beam irradiation), about 10% degradation of 0.01 mM MG dye was observed within 1 h. It is well known that the Fenton reagents can degraded even in dark condition. Thus, we have studied the effect of the Fenton reagent on MG dye degradation in dark condition. About 20% degradation of MG dye occurred within 1 hr, whereas about 50% degradation occurred upon visible light irradiation. About 98.5% degradation of MG dye was obtained when 0.01 mM dye was treated under xenon beam/Fenton reagent process.

4. Conclusions

We have developed a simple and fast method for the mineralization of Malachite green (MG) in aqueous solution based on xenon beam irradiation ($\lambda \leq 320$ nm) and Fenton reagent. The extent and initial rate of degradation were increased by increasing the concentration of Fenton reagents (i.e. concentration of Fe(III) and H₂O₂) at pH 3.1. Hydroxyl radicals were the most important reactive species that played the dominating role in the degradation of dye molecules by the present method. Xenon beam/Fenton process enhanced the rate of degradation by i) increasing the production of hydroxyl radicals (OH) in the reaction media, and ii) increasing the temperature of solution which accelerated the reaction between the exited MG molecules and OH radicals. The method provided an economical and fast approach in the treatment of textile pollutants and could be useful for the large scale mineralization of the industrial effluents.

Acknowledgement

This work was supported by "The Toyohashi University of Technology Global COE" Program. "Frontiers of Intelligent Sensing" supported by Japan's Ministry of Education, Culture, Sports, Science and Technology (MEXT).

References

- J. Ma, W. Ma, W. Song, C. Chen, Y. Tang, J. Zhao, Y. Huang, Y. Xu, and L. Xang, Environ. Sci. Technol. 40, 618 (2006). <u>doi:10.1021/es051657t</u>
- 2. Y. M. Slokar and A. M. L. Marechal, Dye Pigmen. **37**, 335 (1998). <u>doi:10.1016/S0143-7208(97)00075-2</u>
- 3. Li. Huang, D. Gu, L. Yang, L. Xia, R. Zhang, and H. Hou, J. Environ. Sci. 20, 183 (2008).
- K. Swaminathan, S. Sandhya, A. C. Sophia, K. Pachhade, and Y. Subrahmanyam, Chemosphere 50, 619 (2003). <u>doi:10.1016/S0045-6535(02)00615-X</u>
- H. Kusic, N. Koprivanac, and L. Srsan, J. Photochem. Photobio. A: Chem. 181, 195 (2006). doi:10.1016/j.jphotochem.2005.11.024
- E. Santos and W. Schmickler, Chem. Phys. 332, 39 (2006). doi:10.1016/j.chemphys.2006.11.033
- B. Yang, M. Zhou, and L. Lei, Chemosphere 60, 405 (2005). doi:10.1016/j.chemosphere.2004.11.091
- M. M. Uddin, M. A. Hasnat, A. J. F. Samed, and R. K. Majumdar, Dye. Pigmen. 75, 207 (2007). doi:10.1016/j.dyepig.2006.04.023
- F. Chen, W. Ma, J. He, and J. Zhao, J. Phys. Chem. A 106, 9485 (2002). doi:10.1021/jp0144350
- A. Umar, M. M. Rahman, S. H. Kim, and Y. B. Hahn, Chem. Commun. 166-168 (2008). <u>doi:</u> <u>10.1039/b711215g</u>
- 11. Q. Wang and A. T. Lemle, Environ. Sci. Technol. 35, 4509 (2001). doi:10.1021/es0109693
- F. J. Rivas, J. Frades, M. A. Alonso, C. Montoya, and J. M. Monteagudo, J. Agric. Food Chem. 53, 10097 (2005). <u>doi:10.1021/jf0512712</u>
- M. A. Hasnat, I. A. Siddiquey, and A. Nuruddin, Dye. Pigmen. 66, 185 (2005). <u>doi:10.1016/j.dyepig.2004.09.020</u>
- K. Dutta, S. Bhattacharjee, B. Chaudhuri, and S. J. Mukhopadhyay, Environ. Sci. Health 38, 1311 (2003). doi:10.1081/ESE-120021128

- 120 Degradation of Commercial
- 15. A. Umar, M. M. Rahman, and Y. B. Hahn, Talanta **77**, 1376 (2009). doi:10.1016/j.talanta.2008.09.020
- M. M. Rahman, M. J. A. Shiddiky, M. A. Rahman, and Y. B. Shim, Anal. Biochem. 384, 159 (2009). <u>doi:10.1016/j.ab.2008.09.030</u>
- 17. F. J. Benitez, and B. J. Heredia, L. J. Acero, F. Rubio, J. Ind. Eng. Chem. Res. 38, 1341 (1999). <u>doi:10.1021/ie980441f</u>
- J. Fernandez, M. R. Dhananjeyan, J. Kiwi, Y. Senuma, and J. Hilborn, J. Phys. Chem. B 104, 5298 (2000). <u>doi:10.1021/jp9943777</u>
- 19. Y. F. Sun and J. J. Pignatello, J. Agric. Food Chem. 41, 1139 (1993).
- 20. A. L. Teel and R. J. Watts, J. Hazard. Mater. B 94, 179 (2002). <u>doi:10.1016/S0304-3894(02)00068-7</u>
- 21. J. Zhao, K. Wu, H. Wu, H. Hidaka, and N. Serpone, Environ. Sci. Tech. 32, 2394 (1998).
- 22. G. Puppert, R. Bauer, and G. Heisler, Chemosphere 28, 1447 (1994). <u>doi:10.1016/0045-6535(94)90239-9</u>
- 23. C. Galindo, P. Jacques, and A. Kalt, Chemosphere **45**, 997 (2001). <u>doi:10.1016/S0045-6535(01)00118-7</u>
- 24. M. V. B. Zanoni, J. J. Sene, and M. J. Anderson, Photochem. Photobiol. A. Chem. **157**, 55 (2003). <u>doi:10.1016/S1010-6030(02)00320-9</u>
- 25. M. Saquib and M. Muneer, Dye. Pigmen. **53**, 237 (2002). <u>doi:10.1016/S0143-7208(02)00024-4</u>
- 26. M. Perez, F. Torrades, X. Domenech, and J. Peral, Wat. Res. **36**, 2703 (2002). <u>doi:10.1016/S0043-1354(01)00506-1</u>
- 27. G. Ruppert and R. Bauer, J. Photochem. Photobiol. A Chem. **73**, 75 (1993). <u>doi:10.1016/1010-6030(93)80035-8</u>