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Sonochemical Analysis of Methylene Blue with Additives and ZnO Nanoparticles in Aqueous Medium

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Authors' contributions

This work was carried out in collaboration between all authors. Author MSI designed the study, performed the statistical analysis, wrote the protocol and wrote the first draft of the manuscript. Authors MSA and MHU managed the analyses of the study and the literature searches. All authors read and approved the final manuscript.

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ABSTRACT

The use of ultrasound was found to be a very suitable and effective method for the degradation of hazardous dyestuffs. The degradation of dye molecules in aqueous media in the presence of various additives by ultrasonic irradiation was investigated in order to clarify the degradation

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mechanism. In this investigation, Methylene Blue (MB) has been used as a representative of azo dye and inorganic salts like NaCl and Na_2SO_4 were used as additives. Besides degradation in presence of CCl_4 and H_2O_2 solutions were observed. ZnO nanoparticle was also used for the investigation of degradation. The volume of all solutions was 100ml containing 25mg/L of MB. The sonication of dye molecules were conducted for 0, 5, 15, 25 minutes. The degradation mechanism was discussed in details adopting UV- visible spectra. From the experiments it is clear that the acidic condition is favorable for dye degradation. Evaluating the results we have also found that all additives aided the degradation comparatively higher or lower the extent depending on their used amount in solution. So, it is a clear indication that azo dyes from waste water can be efficiently removed by using sonochemical irradiation method under acidic condition.

Keywords: Azo dye; Methylene blue; Sonochemical analaysis; Additives; Decomposition.

1. INTRODUCTION

Organic dyes are utilized in many different industries, including optical data discs, food, cosmetics, pharmaceuticals, solar cells, and traditional textile manufacturing. For instance, Methylene blue (MB) is a significant synthetic dye that is utilized in water testing, sulfide analysis, medicine, and biology [1]. It also serves as a peroxide generator and a redox indicator. However, they generally disrupt ecosystems and are poisonous, mutagenic, and carcinogenic to aquatic and human life [2-4]. It is necessary to create the technology for eliminating organic dyes from water. To get rid of these chemicals, numerous techniques including chemical, physical, and biological ones have been devised [4-6]. Among these advanced oxidation processes (AOPs), electrochemical technologies and membrane filtration are mentionable. The possibility of fully mineralizing organic pollutants to CO_2 and H_2O has raised interest in advanced oxidation processes (AOPs). In AOPs for wastewater treatment, ultrasonography has recently been employed [7-9]. The pyrolysis reactions that occur at and inside hot regions during collapsing bubbles and the radical reactions by OH and H radicals [10] that are generated by the pyrolysis of water are the two fundamental principles of sonochemical reactions.

$$H_2O \rightarrow \bullet H + \bullet OH \quad (1)$$

Numerous studies have investigated the sonochemical degradation of phenolic compounds and dyes [4, 11–17]. The creation of efficient additives to speed up the decomposition of organic molecules has been the focus of intensive research. Recent studies [17–19] have looked into how adding CCl_4 or C_6F_{14} affects sonochemical degradation. Although CCl_4 is one of the hazardous chemicals and is therefore highly regulated in usage, it is a valuable additive to speed up the sonochemical breakdown of the target organic molecules. According to Sponza et al. [18], the presence of 19 $mgL^{-1}C_6F_{14}$ increased the elimination rates of phenol in the wastewater from the olive mill. Zeng et al. [19] reported that the sonochemical degradation of phenol increased from 0.014 to 0.031 min^{-1} or from 0.014 to 0.032 min^{-1}, respectively, in the presence of 150 M CCl_4 or 1.5 M C_6F_{14}. According to studies [18, 19], the action of CCl_4 or C_6F_{14} as a H atom scavenger is what causes these beneficial effects. Additionally, it has been noted that the use of certain chemicals can influence how quickly colors degrade [20-22]. On the other hand, metal oxide semiconductors have received much research into their potential as photocatalysts to purge water and air of organic contaminants [23-26]. The active sites of electron-hole pairs are produced when the metal oxides are activated by the right amount of photon energy, and this increases the catalytic activity on the metal oxide surfaces. [27]. Due to its excellent stability and relatively straightforward preparation process, TiO_2 is regarded as the oldest, most prevalent, and optimal material among these metal oxides [28-31]. Zinc oxide (ZnO), a metal oxide with an exciton binding energy of 60 m eV and a rather broad band gap of 3.37 eV, has been extensively studied as a photocatalyst for the degradation of various organic pollutants. Moreover, ZnO nanostructures are preferred over TiO_2 as photocatalyst alternatives for photodegradation due to their more favorable solar spectrum absorption, affordability, and nontoxicity [32]. In numerous studies, ZnO nanostructures demonstrated significant photocatalytic activity for the removal of organic pollutants, such as organic dyes [33]. Thus, in this study, we looked how several additives such as CCl_4, H_2O_2, NaCl, Na_2SO_4, and ZnO nanoparticles affected the sonochemical degradation of MB. Additionally, we have studied several parameters such as pH, additive concentrations (CCl_4, H_2O_2, NaCl, Na_2SO_4, and ZnO nanoparticle) in order to obtain the optimal towards the efficiency for the degradation rate of MB in water by sonochemistry.

2. METHODOLOGY

2.1 Materials and Equiments

Methylene Blue ($C_{16}H_{18}ClN_3S$), Merck Life Science Private Limited, Godraj One, 8th Floor, pirojshanagar, Eastern Express highway, Vadhroli East, Mumbai-400079, Sodium Chloride (NaCl), RANBAXY Fine chemicals Limited A-3, Okhla industrial area, phase-l, New Delhi-l10020 (ISO 9001: 2000 certified company), Sodium sulphate (Na_2SO_4), Merck Specialities Private Limited, Shiv Sagor Estate `A` DrAnnic Besant Road, Worli, Mumbai-400018, Hydrogen peroxide (H_2O_2), Merck KGaA, 64271 Darmstadt, Germany, Carbon tetrachloride (CCl_4), Merck, D-6100 Darmstadt, FR. Germany were used as purchased.

2.2 Preparation of Solutions

25.0 mg/l solutions of MB were prepared. In 100 ml of distilled water, solutions of the inorganic salts NaCl and Na_2SO_4 were produced using 36.0 and 13.9 grams of each salt, respectively, according to their greatest saturation points. One liter of distilled water has been mixed with 1.02 ml and 2.0 ml of H_2O_2, respectively, to create 10 mM/l and 20 mM/l H_2O_2 solutions. Using a tiny syringe, the 200, 400, and 600µM/L CCl_4 is adjusted. Dye stuffs in water can absorb and reflect light, decreasing the transparency of the water naturally. Normally, dye effluent includes between 10 and 50 mg/L, though 1.0 mg/L dye solutions are visible and might be considered as contaminants and objectionable [34, 35].

Fig. 1. Sonicator bath used in sonochemical degradation

Fig. 2. Prepared methylene blue solution

3. RESULTS AND DISCUSSION

3.1 Effect of Sonochemical Degradation of MB under Various pH

Based on the first-order kinetics model function, exponential curves are represented in the figure as solid curves.

$$X = ae^{-kt} \ldots\ldots\ldots\ldots\ldots \quad (2)$$

Where, the constant 'a' is primary concentration of MB, 25mg/l, k is a fitting parameter need to be optimized and t is the degradation time. The degradation rates of MB without additives using sonochemical irradiation processes in different conditions are shown and discussed below. NaOH and H_2SO_4 Solutions have been used to maintain the pH of the solution.

Fig. 3 shows the degradation rate of MB in acidic condition of pH: 3 is higher than that of the neutral, i.e. pH: 7 and basic condition, viz. pH: 8. With additives where the acidic condition is maintained in the sonochemical degradation, there is a high possibility of an increased of degradation rate of MB due to availability of OH radical.

3.2 Effect of CCl_4 on Sonolytic Degradation of MB

We examined the effect of CCl_4 at 200, 400, and 600 µM/L concentrations on the sonochemical degradation of MB. The result is represented in Fig. 4.

In presence of CCl_4, the rate of degradation increased compared to the pure MB and reaches maximum when 400 µM CCl_4 was present. In presence of 600µM/L CCl_4, the rate is also greatly increased. However, at 200µM/L, the rate of degradation first sharply declines before increasing with increasing sonication time, with the exception at 25 minutes. Our findings indicate that active Cl radicals or related species are produced during the sonolysis of CCl_4, and that these radicals or species would be useful for the breakdown of MB, despite some researchers' reports that CCl_4 could operate as a H atom scavenger [19]. According to earlier research [17, 36–39], the following responses ought to occur:

$$CCl_4 \ \bullet CCl_3 + \bullet Cl \quad (3)$$
$$\bullet CCl_3 : CCl_2 + \bullet Cl \quad (4)$$
$$2\bullet CCl_3 \ \ C_2Cl_6 \quad (5)$$
$$2{:}CCl_2 \ \ C_2Cl_4 \quad (6)$$
$$\bullet Cl + \bullet Cl \rightarrow Cl2 \quad (7)$$

The radicals that develop containing chlorine (Cl, CCl_3, CCl_2, Cl_2 etc.) may accelerate the degradation of MB in an aqueous solution. According to Merouani et al. [13], the presence of 200 mML^{-1} of CCl_4 enhanced the sonochemical degradation rate of rhodamine B at 300 kHz by 21 times. According to Okitsu et al. [31], the addition of 100 ppm CCl_4 enhanced the sonochemical breakdown rate of MO at 200 kHz by 41 times. The efficiency of CCl_4 for MB degradation we have found in this study agrees with these results. From the above viewpoints, though CCl_4 has high toxicity [40] it can be used as an additive.

3.3 Effect of H_2O_2 on the Rate of Degradation of MB

Sonolytic degradation of MO was also carried out in presence of H_2O_2. Fig. 5 shows that the rate of decomposition was increased by the addition of H_2O_2. It is clear from the data that the doses 20 and 200 µL of 10 mM/l H_2O_2 increased the rate of deterioration in comparison to pure MB but in presence of 100 µL the degradation rate decreased (Fig. 5a). The graphical representation of the degradation of MB in presence of H_2O_2 solution also noticed that lowest dose 20 µL and 200 µL of 20 mM/L H_2O_2 solution added to the MB solution shows the highest degradation rate as compared with the dose 100 µL of that H_2O_2 solution (Fig. 5b). H_2O_2 reaction mechanism for dye discoloration has been explained from the formation of free radicals as active species.

$$H_2O_2 \rightarrow \bullet OH + \bullet OH \quad (8)$$

Previous work has indicated that the US/UV/H_2O_2 approach was the most effective in degrading malachite green because it uses ultrasound to facilitate H_2O_2 scission in addition to photolysis [41]. Another recent study demonstrated that the impact of adding hydrogen peroxide was examined by varying its concentration to 0.05, 0.10, and 0.15 M [42]. The treatment with 0.10 M H_2O_2 and 45 W was successful and energetically feasible, resulting in a 62.9% absorbance removal for the water assessed [42]. The amount of dissolved gas dropped and the amount of water vapor in the bubbles increased as the temperature of the aqueous solution rise [43], which led to a lower rate of OH radical production. As a result, when solution temperatures raise the rates of MB degradation decreased. The sonolysis of 4-chlorophenol in aqueous solution at 20 and 500 kHz as a function of solution temperature was reported by Jiang et al. [43]. They found that, in the 10–40 ◦C range, the rates of H_2O_2 production and 4-chlorophenol degradation increased with the rising of solution temperatures at 500 kHz but reduced with rising solution temperatures at 20 kHz. The sonolysis of bisphenol A in aqueous solution as a function of solution temperatures at 489 kHz was reported by Uddin et al. [44]. The outcome shown that as the temperature of the aqueous solution increased, the rates of degradation increased within the range of 10–40 ^{0}C. When compared to earlier studies, it is evident that a variety of applications for hydrogen peroxide (H_2O_2) have been developed in the field of water treatment due to its relative safety and ease of use.

3.4 Effect of Inorganic Salt (NaCl and Na_2SO_4) on the Degradation of MB

Sonolytic degradation of MB was investigated in the presence of NaCl and Na_2SO_4. 10 ml and 20 ml of NaCl and Na_2SO_4 each were added into the MB solution in order to find the effects of the addition of inorganic salts on dye degradation. Fig. 6 and Fig. 7 shows the experimental result. It is seen in the figure, the addition of Na_2SO_4 improved the degradation of MB, but the addition of NaCl caused it to decrease. Previous studies have shown that various synthetic dye solutions with a number of mixtures of Na_2SO_4 and NaCl, Na_2SO_4 influenced the decolourization efficiency more modestly than NaCl [45]. Higher concentration of Na_2SO_4 did not hinder the decolorization process and even improved the effectiveness of reactive intense red K-2BP in dye solutions with the similar salt or Na^+ concentration [45]. Additionally, the deterioration rate accelerated due to the elevated salt concentration. From the above comparison, it can be said that Na_2SO_4 is superior to NaCl in terms of MB degradation. Sonochemical degradation in absence and presence of Na_2SO_4 or NaCl for 4-chlorophenol, phenol, catechol, and resorcinol under Ar was published by Uddin et al. in 2016 [46]. The rate of phenolic compound breakdown followed a pseudo-first order rate constant [47].

3.5 Effect of ZnO Nanoparticle on MB Degradation

Fig. 8 shows the degradation rate of Methylene blue with ZnO nanoparticle. The degradation of Methylene blue was significantly increased with the increasing of sonication time. Maedeh Asgharian et al. observed the maximum degradation of MB in presence of rGO/ZnO/Cu compound at 25 mg photocatalyst dosage [48]. Presence of ZnO nanoparticle is also showed second highest degradation of MB in this study. Crystalline structures of ZnO nanoparticle facilitate it to use as photo catalyst and porous nature increased the dye removal efficiency.

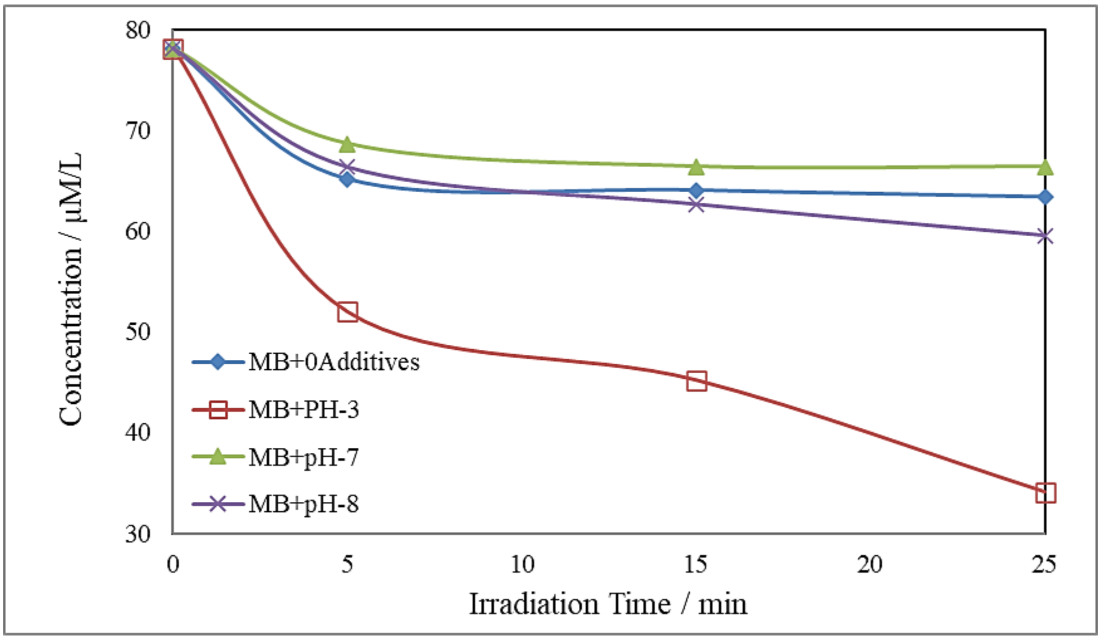

Fig. 3. Effect of Sonochemical degradation of pure MB at different time for various pH

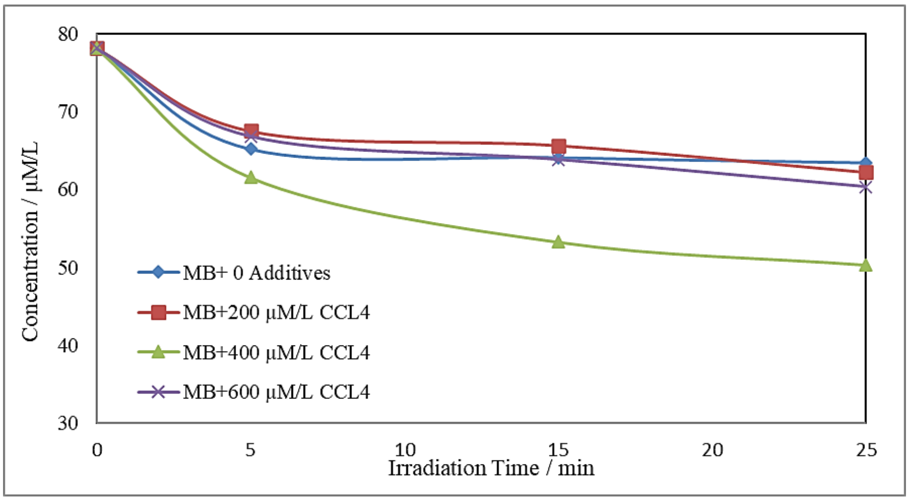

Fig. 4. Effect of CCl_4 on the degradation of MB

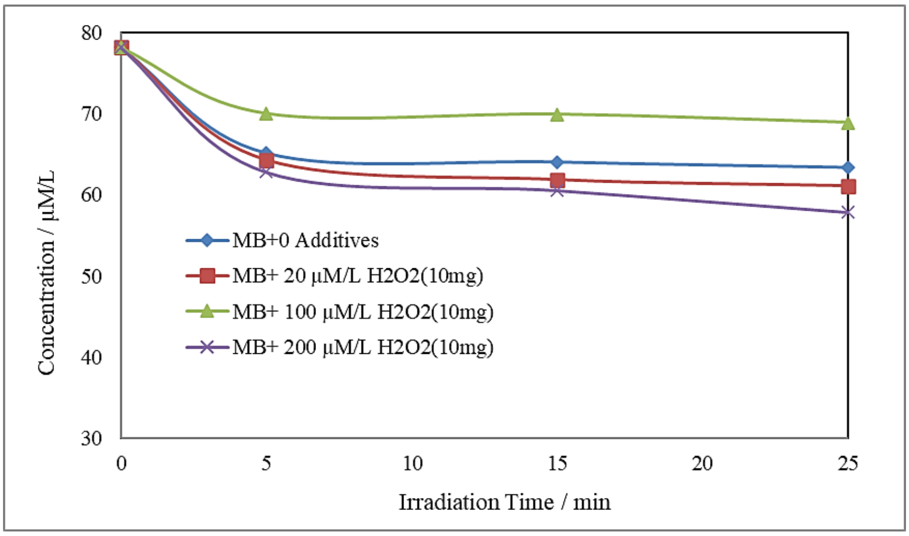

Fig. 5a. Effect of 10mM H_2O_2

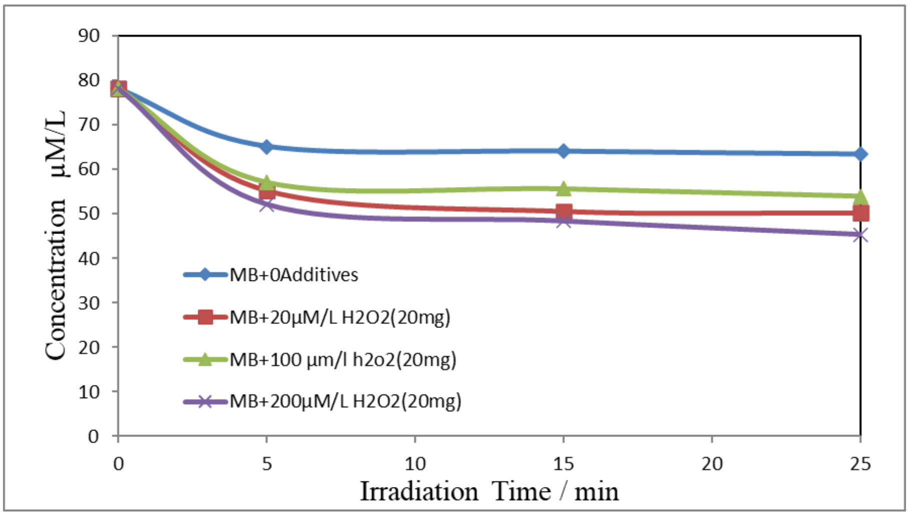

Fig. 5b. Effect of 20mM H_2O_2

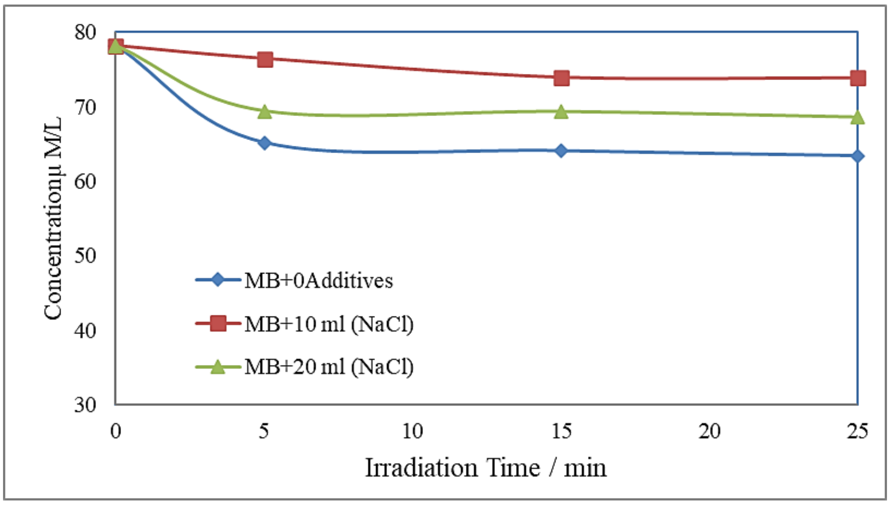

Fig. 6. Effect of NaCl on the degradation of MB

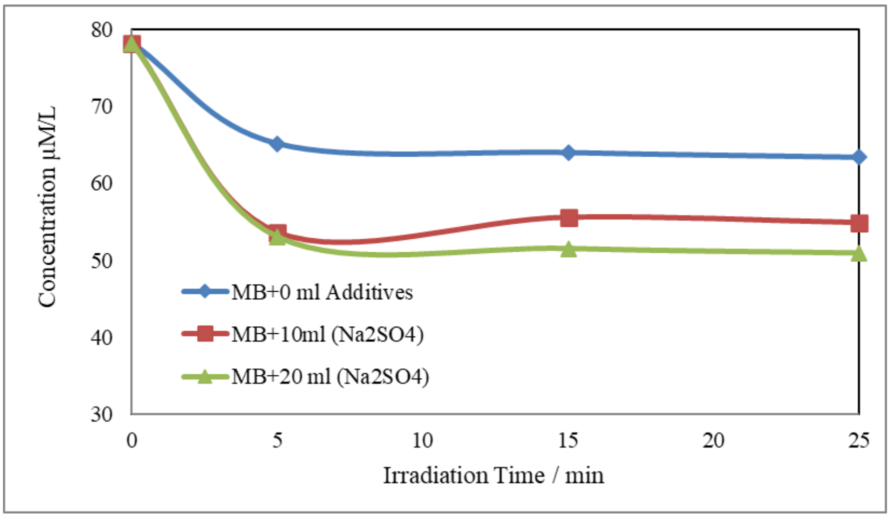

Fig. 7. Effect of Na_2SO_4 on the degradation of MB

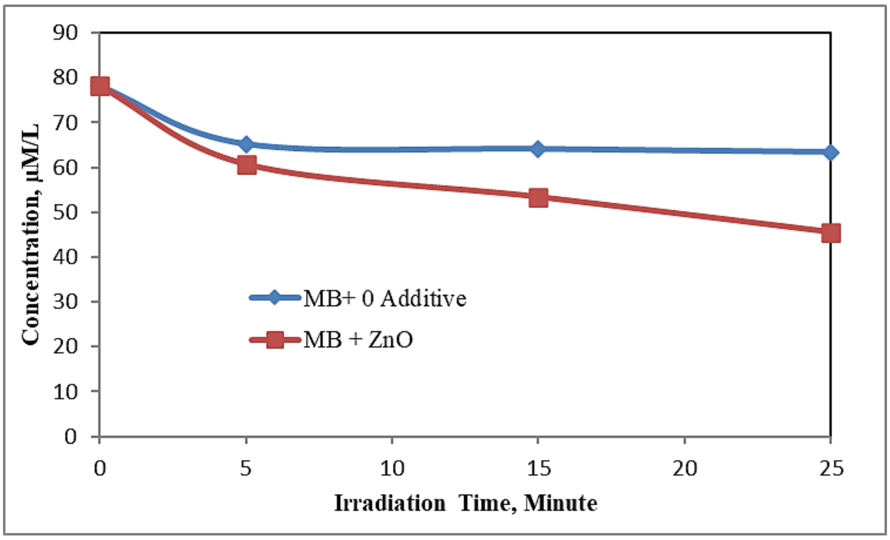

Fig. 8. Effect of ZnO on the degradation of MB

4. CONCLUSIONS

The sonochemical degradation rate of MB was studied with several parameters, such as pH, addition of several additives (CCl_4, H_2O_2, NaCl, Na_2SO_4 and ZnO nanoparticle). From the research work we found that the degradation of MB increased under acidic pH condition. 400 µM/L doses of CCl_4 is more efficient to degrade the MB compared to 200 and 600 µM/L doses. It is thought that the degradation of CCl_4 created chlorine species and radicals, which accelerated the destruction of MB. The decomposition rate is also higher in presence of H_2O_2 compared to the pure MB and 200 µM/L doses of 20mM/L H_2O_2 shows the highest degradation rate than the other used doses of H_2O_2 in this study. On the other hand the degradation power of Na_2SO_4 is higher than the NaCl. Use of ZnO nanoparticle improved the degradation rate significantly. The results showed that the MB degradation rate increases as the order of H_2O_2 > ZnO > CCl_4 > Na_2SO_4. Therefore, all these additives and catalysts are suitable for ultrasonic degradation of methylene blue in aqueous medium which can be effectively used for the removal of dye from water.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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