Impact of Additives and ZnO Nanoparticles on the Sonodegradation of Methylene Blue

Abstract

Sonohemical method for the degradation of toxic dyestuffs is now considered as a promising technique. An important dye namely Methylene Blue (MB) was degraded using ultrasound irradiation at different pH. The impact of various additives like CCl_4, H_2O_2, NaCl and $Na_2SO_4$with different doses for MB degradation was observed. The degradation efficiency of ZnO nanoparticles was also measured.The sonolysis was conducted for 0, 5, 15, 25 minutes and the extent of degradation was measured by UV–vis spectrophotometer. Acidic media was more efficient for degradation compared to basic media.200 μM/L doses $H_2O_2$of 20 mM concentration showed highest efficiency (41.98%) to degrade the MB molecules. On the otherhand CCl_4, Na_2SO_4 and ZnO nanoparticles also showed maximum 35.63%, 34.88% and 41.72% degradation efficiency respectively. The efficiency increases in the presence of additives and ZnO nanoparticles in comparison to the efficiency (18.88%) obtained without these additives.

Keywords: Additives, Azo dye, Degradation, Methylene Blue, Nanoparticles

1. Introduction: Organic dyes are used in a wide range of sectors, including optical discs, food, cosmetics, medicines, solar cells, and traditional textile manufacturing. To ensure clean and safe water sources, it is critical to identify a method for extracting and discarding the organic dye chemicals used on a regular basis [1, 2]. Although there are numerous types of dyes, azo dyes which are organic compounds bearing the functional group R-N=N-R′, represent for 70% of total dye production globally [3]. These compounds are distinguished by the azo bond chromophore group (-N=N-), which is related with aromatic structures and functional groups such as $-NH_2$ and $-SO_3H$ [4, 5]. These dyes are widely used in the food, textile, paper, leather, and pharmaceutical industries because of their exceptional properties and low cost [6]. The effects of various dye compounds on human health vary. When azo dyes and their intermediates degrade, they can cause damage, mutagenesis, and carcinogenesis, posing a major risk to human health [7]. For example, MB is a major synthetic dye used in large quantity in water testing, sulfur analysis, medicine, and biology [8]. It also functions as a peroxide producer and redox indicator. However, they generally disturb ecosystems and are toxic, mutagenic, and carcinogenic to both aquatic and human life [9-11].It is important to develop a technology to remove this dye from

water. Numerous chemical, physical, and biological approaches have been developed to remove these compounds [11-13]. Advanced oxidation processes (AOPs) have received a lot of attention due to their ability to totally mineralize organic contaminants to CO_2 and H_2O. Ultrasonography has lately been used in alternative operating procedures for wastewater treatment [14-16]. The two main concepts of sonochemical reactions include pyrolysis reactions that occur at and inside hot regions during collapsing bubbles, as well as radical reactions by OH and H radicals produced by water pyrolysis [17].

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

Several research have looked at the sonochemical degradation of phenolic chemicals and dyes [11, 18-24]. The development of efficient additives to accelerate the decomposition of organic molecules has been the subject of extensive investigation. Recent research investigated how adding CCl_4 or C_6F_{14} impacts sonochemical degradation [24-15]. Although CCl_4 is a toxic chemical and hence heavily controlled in use, it is an effective addition for speeding up the sonochemical breakdown of the target organic molecules. Sponza et al. found that 19 mg $L^{-1}C_6F_{14}$ boosted the rate of phenol removal in olive mill wastewater [25].According to Zeng et al., the sonochemical degradation of phenol rose from 0.014 to 0.031 min^{-1} or from 0.014 to 0.032 min^{-1} in the presence of 150 M CCl_4 or 1.5 M C_6F_{14} [26]. According to research the action of CCl_4 or C_6F_{14} as a H atom scavenger is responsible for these positive benefits [25, 26],. Furthermore, it has been observed that the usage of specific chemicals can affect how quickly colors deteriorate [27–29]. Metal oxide semiconductors, on the other hand, have been extensively studied for their potential as photocatalysts for the removal of organic pollutants from water and air [30-33]. When the metal oxides are activated by the appropriate quantity of photon energy, active sites of electron-hole pairs form, increasing catalytic activity on the metal oxide surfaces [34]. TiO_2 is considered the oldest, most common, and best material among these metal oxides due to its outstanding stability and relatively simple fabrication technique [35-38]. 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 received substantial research as a photocatalyst for the breakdown of numerous organic contaminants. Furthermore, ZnO nanostructures are recommended over TiO_2 as photocatalyst alternatives for photodegradation due to their better solar spectrum absorption, lower cost, and nontoxicity [39]. Numerous studies have shown that ZnO nanostructures have significant photocatalytic activity for removing organic contaminants, such as organic dyes [40].

Therefore, in this work we have investigated the efficiency of additives like CCl_4, H_2O_2, NaCl, Na_2SO_4, and ZnO nanoparticles on the sonolytic degradation of MB at certain concentration and pH.

2. Experimental

2.1 **Materials and equiments:** Methylene Blue ($C_{16}H_{18}ClN_3S$), Merck Life Science Private Limited, Godraj, Vadhroli East, Mumbai-400079, Sodium Chloride (NaCl), Ranbaxy Fine chemicals Limited A-3, New Delhi-l10020 (ISO 9001: 2000 certified company), Sodium sulphate (Na_2SO_4), Merck Specialities Private Limited, Worli, Mumbai-400018, Hydrogen peroxide (H_2O_2), Carbon tetrachloride (CCI_4), Merck, D-6100 Darmstadt, FR. Germany. All the Chemicals were used as purchased. The sonicator machine used in this work is shown in figure 1.

Fig. 1: Sonicatorbath used in sonochemical degradation.

2. 2 Preparation of Solutions

MB solutions with a concentration of 25 mg/L were prepared. In 100 ml of distilled water, solutions of the inorganic salts NaCl and Na_2SO_4 were prepared using 36 and 13.9 grams of each salt, respectively, based on their maximum saturation points. One liter of distilled water was mixed with 1.02 mg and 2.04 ml of H_2O_2 to make 10 mM/L and 20 mM/L H_2O_2 solutions, respectively. A little syringe is used to prepare the 200, 400, and 600µM/L CCl_4.ZnO nanoparticle was prepared using sol gel technique using $ZnNO_3$ and KOH. Pure ZnO nanoparticle was used as catalyst with 12.5 mg dose.Water soluble dyes can absorb and reflect light, reducing the water's clarity naturally. Normally, dye effluent contains between 10 and 50 mg/L, although 1.0 mg/Ldye solutions are visible and may be considered as pollutants [41, 42].

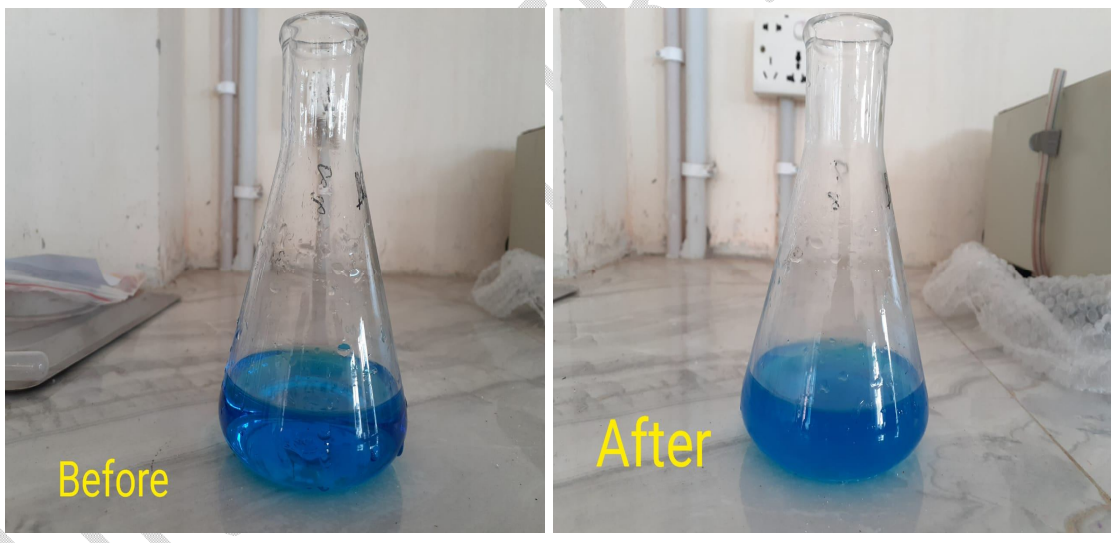

Fig. 2: Methylene blue solution

3. Results and Discussion

3.1Degradation percentage of MB at various pH

The degradation percentages of MB without additives used in sonochemical irradiation process under various situations are presented and discussed below. The degradation percentages were calculated using the following equation.

% of degradation = $(C_0\text{-}C)/C_0$ X 100 (2)

Where, C represents the final concentration obtained after sonication at different time and C_0 represents theinitial concentration of MB solution.Figure 3 depicts the degradation efficiency of MB at different pH. NaOH and H_2SO_4 solutions were used to change the solution's pH.

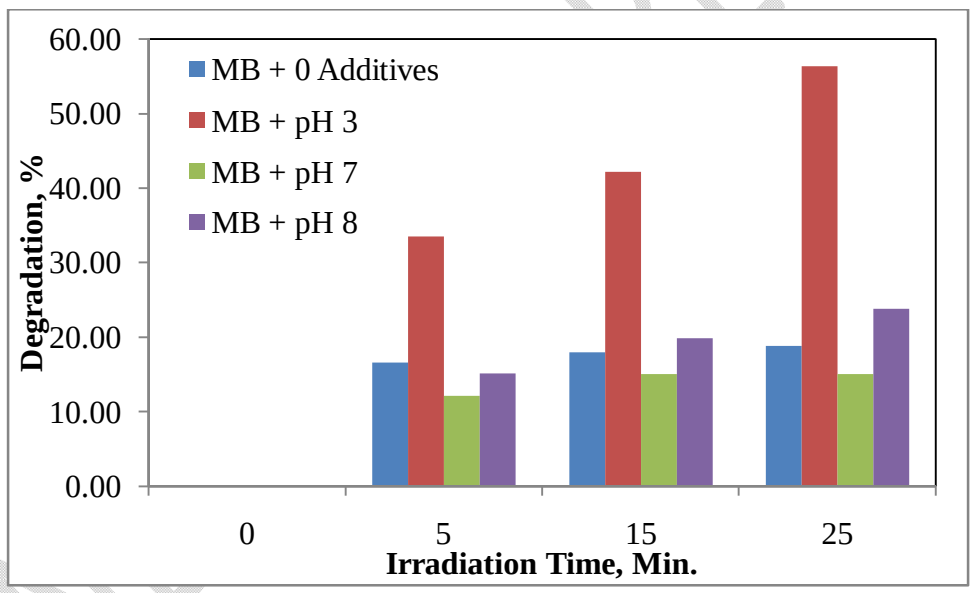

Fig. 3: Degradation efficiency forpure MB at various pH.

From the graph it is found that the efficiency at pH 3 is56.33% atpH 7 is 15.06% and atpH 8 is 23.82% where the efficiency for raw solution without changing the pH is18.88% i. e acidic media is more efficient for degradation than basic media.There is a considerable chance of enhancing degradation efficiency using additives that maintain an acidic environment during sonochemical breakdown. Previous research has shown that sonochemical degradation efficiency is higher in an acidic environment [27-29].

3.2. Efficiency of CCl_4forsonolyticdegradation of MB

The efficiency of CCl_4 at 200, 400, and 600 µM/L concentrations on the sonochemical degradation of MB is represented in figure 4.

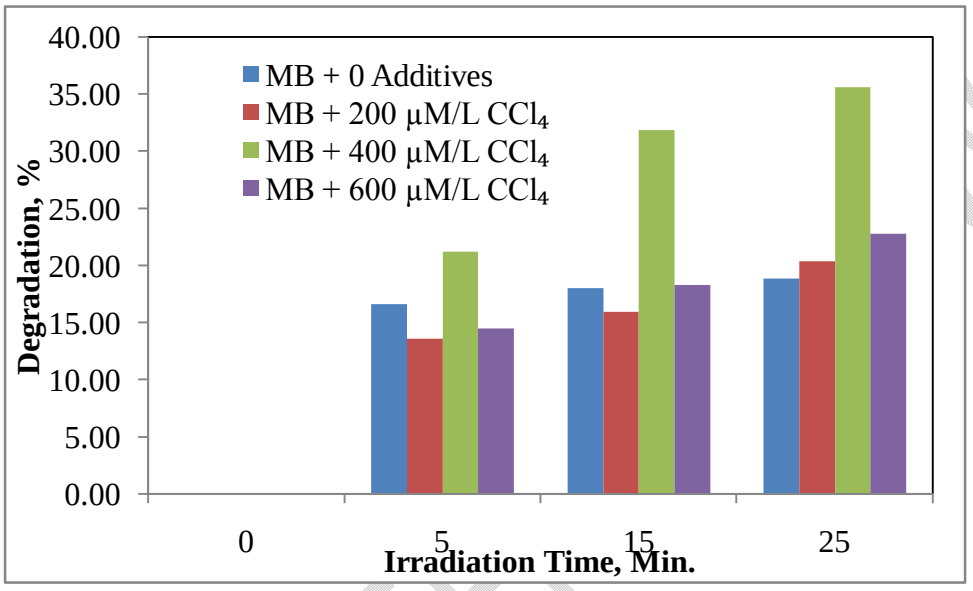

Fig. 4:Efficiencyof $CCl_4$for the degradation of MB

In presence of CCl_4, the highest efficiency of MB degradation is 35.63% when 400 µM/L CCl_4 was present. In the presence of 600µM/L CCl_4 the efficiency is 22.80% and at 200µM/L the efficiency of degradation is 20.39%. We suggest that active Cl radicals or related species are formed during CCl_4sonolysis, and that these radicals or species could be beneficial in the breakdown of MB, though some researchers' claims that CCl_4 could act as a H atom scavenger [26]. According to previous research [24, 43-46], the following responses might occur:

$CCl_4 \rightarrow \bullet CCl_3 + \bullet Cl$ (3)

$\bullet CCl_3 \rightarrow : CCl_2 + \bullet Cl$ (4)

$2\bullet CCl_3 \rightarrow C_2Cl_6$ (5)

$2 : CCl_2 \rightarrow C_2Cl_4$(6)

$\bullet Cl + \bullet Cl \rightarrow Cl_2$ (7)

The radicals that form, which contain chlorine (Cl, CCl_3, CCl_2, and Cl_2) and so on, may speed up

the breakdown of MB in an aqueous solution. Merouani et al. [20] reported that 200 mg L-1 of CCl_4 increased the sonochemical degradation rate of rhodamine B at 300 kHz by 21 times. According to Okitsu et al., adding 100 ppm CCl_4 increased the sonochemical breakdown rate of MO at 200 kHz by 41 times [38]. The effectiveness of CCl_4 for MB degradation discovered in this investigation is consistent with these findings. Another study found that the presence of CCl_4 improves the sonolytic breakdown. From the above discussion, though CCl_4has high toxicity [47]it can be used as an additive due its high degradation efficiency.

3.3 Efficiency of H_2O_2 for MB degradation

Sonolysis of MB with 20, 100 and 200 µL doses of H_2O_2 at two different concentration 10 mM and 20 mM were carried out and the degradation efficiency is demonstrated in figure 5.It is observed from the figure that highest degradation efficiency showed by 200 µL of 10 mM H_2O_2 is 25.97% (fig. 5 a) and200µLof 20 mM H_2O_2 is 41.98% (fig. 5 b) after 25 minutes irradiation i. e. higher doses and higher irradiation time increases the efficiency of degradation.The generation of free radicals as active species explains the H_2O_2 reaction process for dye discolouration.

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

According to research, the US/UV/H_2O_2 technique is the most successful in decomposing malachite green because it incorporates ultrasound to facilitate H_2O_2 scission in addition to photolysis [48]. Another recent study assessed the impact of introducing hydrogen peroxide at concentrations of 0.05, 0.10, and 0.15 M. The treatment with 0.10 M H_2O_2 and 45 W was effective and energy-efficient, removing 62.9% of the water's absorbance [49]. Sonolysis with 400 µL of H_2O_2 has been shown to improve methyl orange elimination efficiency [27]. When compared to previous research, it is clear that a variety of uses for hydrogen peroxide (H_2O_2) have emerged in the field of water treatment due to its relative safety and convenience of use.

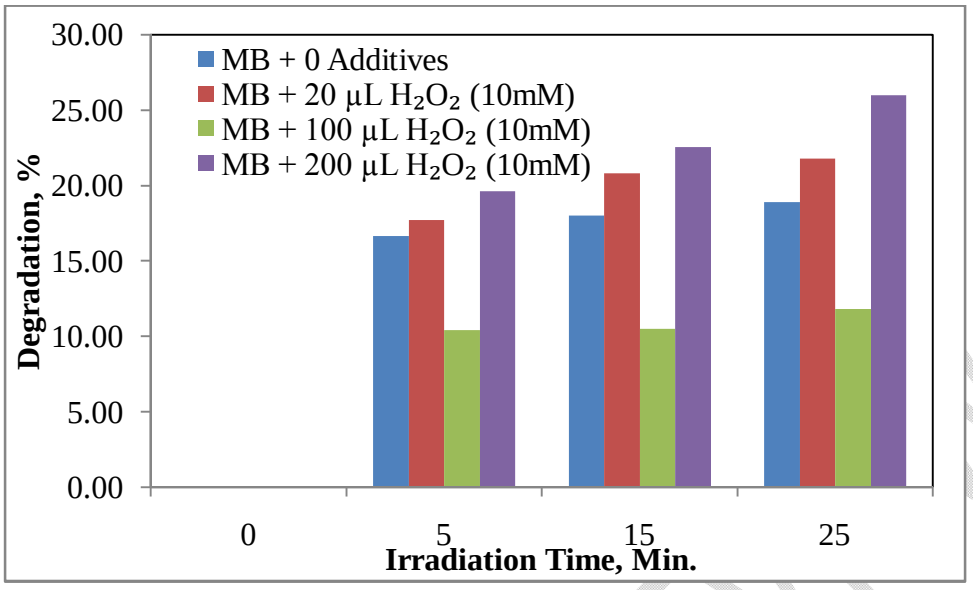

Fig. 5 a:Efficiency of H_2O_2(10mM) for the degradation of MB

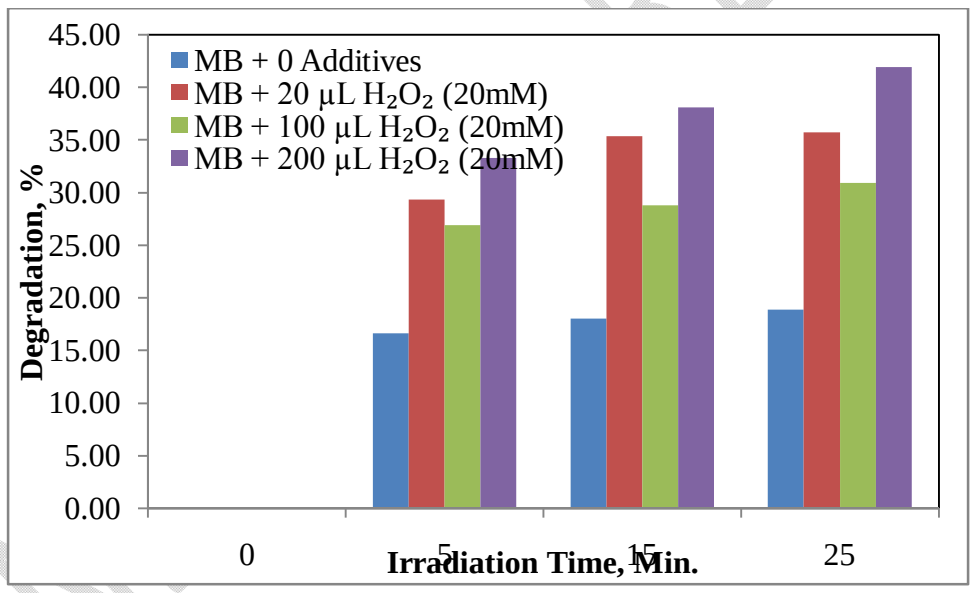

Fig. 5 b: Efficiency of H_2O_2(20mM) for the degradation of MB

3. 4Efficiency of NaCl and Na_2SO_4 for the degradation of MB:10 ml and 20 ml NaCl and Na_2SO_4 were added in to the MB solution in order to find the efficiency of the inorganic salts on dye degradation.Figure 6 and 7 represents the observed result. It is seen in the figure that highest degradation (34.88%) obtained by 20 ml Na_2SO_4 indicates the addition of Na_2SO_4 improved the degradation efficiency for MB. On the other hand the addition of NaCl caused it to decrease as compared to the degradation of pure MB due to the Cl^- ion produced.Previous research has

demonstrated that in synthetic dye solutions containing a variety of Na_2SO_4 and NaCl mixer, Na_2SO_4 had a more modest effect on decolourization efficiency than NaCl. A higher concentration of Na_2SO_4 did not impede the decolorization process and even boosted the effectiveness of reactive bright red K-2BP in dye solutions with comparable salt or Na^+ concentrations [50]. Furthermore, the degradation rate has accelerated due to the increased salt concentration. Following comparison, it can be concluded that Na_2SO_4 outperforms NaCl in terms of MB degradation. Uddin et al. published a study in 2016 on the sonochemical degradation of 4-chlorophenol, phenol, catechol, and resorcinol in the absence and presence of Na_2SO_4 or NaCl [51]. The rate of phenolic compound decomposition adhered to a pseudo-first order rate constant [52].Another study conducted by Monira et al. found that the effectiveness of methyl orange degradation increased in the presence of a 20 ml Na_2SO_4 solution.

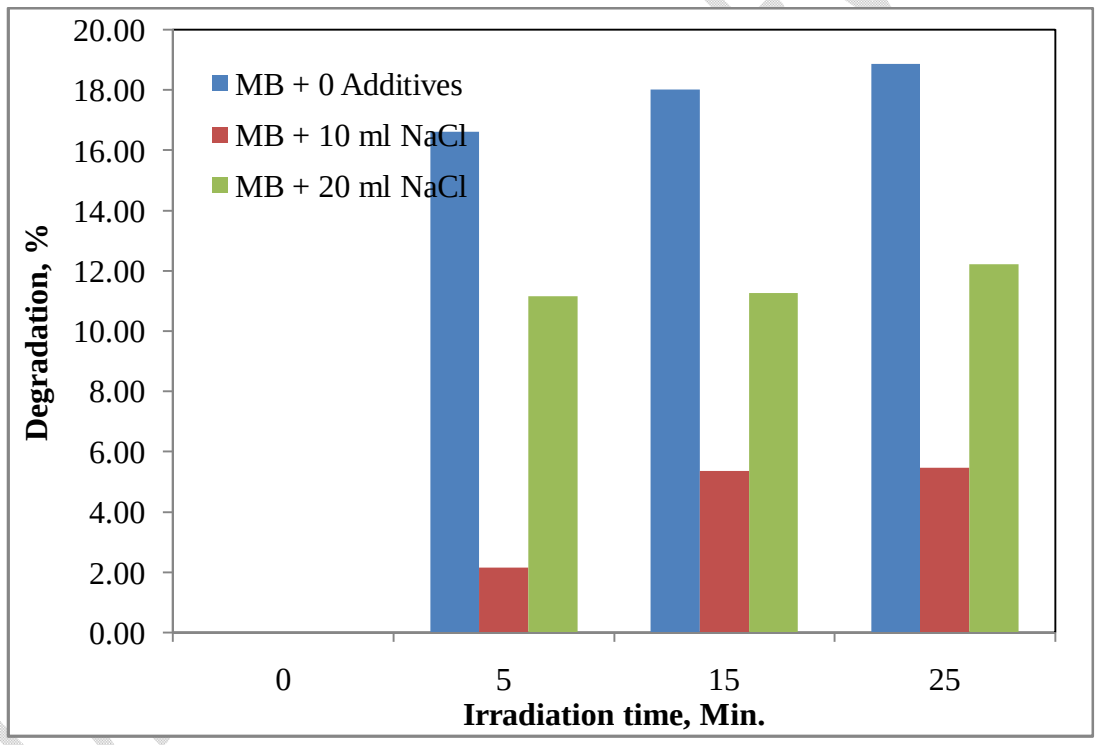

Fig. 6: Effect of NaCl on the degradation of MB

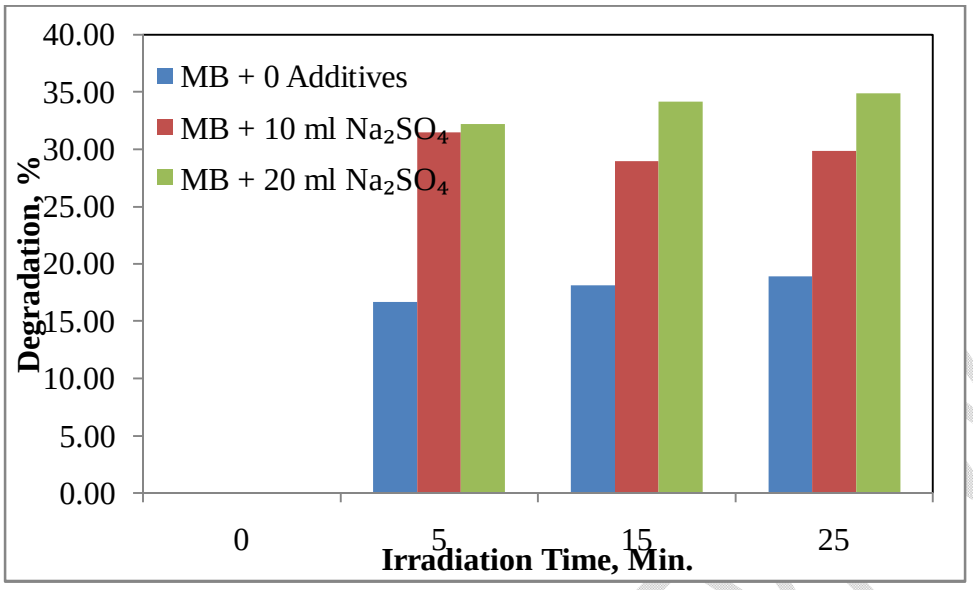

Fig. 7: Effect of Na_2SO_4 on the degradation of MB

3.5 Effect of ZnO nanoparticle on MB degradation: Figure 8 displays the ultrasonicdegradation efficiency of ZnO nanoparticle for MB. The efficiency of degradation isupto 41.73% after 25 minutes sonication. Due to advantageous spectrum absorption, porus nature and crystalline structure ZnO nanoparticle increases the degradation efficiency by producing OH radical. Maedeh Asgharian et al. observedthe maximum efficiency to degrade MB in presence of rGO/ZnO/Cu compound in presence of 25 mg photocatalyst doses[53].Kamaraj et al. showed that 99% BPA degradtion caused by ZnO nanoparticles when exposed to sunshine [54]. Barik et al. was also showed the increased degradation efficiency for 2, 4-DCP by ZnO nanoparticle [55]. ZnO nanoparticle is also showed one of the highest degradation efficiency of MB in this work.

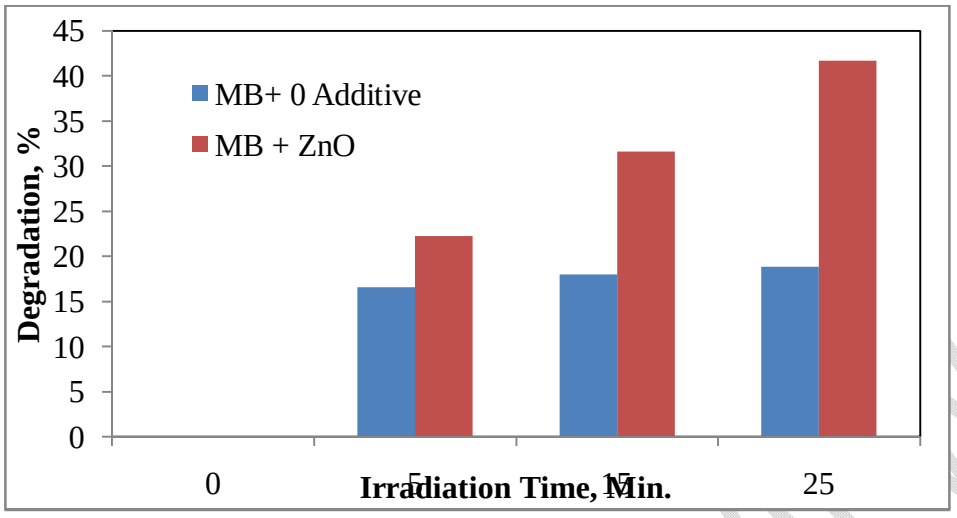

Fig. 8: Efficiency of ZnO on the degradation of MB

4. Conclusions

Theultrasonic degradationefficiency of CCl_4, H_2O_2, NaCl, Na_2SO_4 and ZnO nanoparticle for MB degradation was studied at different pH, doses and concentration.The efficiency of degradationwas higher atlow pH compared to higher pH. 400 µM/L dose of CCl_4 is more efficient to degrade the MB compared to other doses. On the other hand highest degradation efficiency obtained by H_2O_2 at 200 µL doseswith 20 mM concentration. H_2O_2also showed the highest degradation efficiency than the other additives used in this study. Furthermore, Na_2SO_4 increases the degradation percentages of MB But NaCl decreases the degradation percentages in compared to the degradation occurred for pure MB.ZnO nanoparticle enhanced the degradation efficiency considerably.Therefore, all these additives and nanoparticlesexcept NaCl are appropriate for sonolytic degradation of methylene bluein water.

Disclaimer (Artificial intelligence)

Option 1:

Author(s) hereby declare that NO generative AI technologies such as Large Language Models (ChatGPT, COPILOT, etc.) and text-to-image generators have been used during the writing or editing of this manuscript.

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