

Effects of Additives on Sonolytic Degradation of Azo Dye Molecules Found in Industrial Wastewater

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Received 28 August 2020, Received in revised form 7 July 2021

Accepted 7 August 2021, Available online 30 January 2022

ABSTRACT

The growing number of industries is a threat to our environment, as they expose the polluted water directly into the natural water sources. Various processes have been expanded to evacuate these hazardous organic pollutants from water. Among them, advance oxidation process (AOP) is very popular, as the method is cost efficient and highly effective in the similar field. In this process ultrasound is exercised for the deterioration of these pollutants and occasionally some additives are also added to enhance the degradation. In this article, the author has reported the degradation of two azo dyes, named methyl orange (MO) and congo red (CR), in presence of some additives like, inorganic salts, charcoal, H₂O₂, CCl₄, tert-butyl alcohol (TBA), glucose and sucrose. The total experiment was conducted at room temperature and the power of the sonication machine was fixed to 40 kHz-120W. For both of the dyes, it was found that charcoal was most effective additive, which highly enhanced the degradation rate, which was mainly due to its high adsorption capability. TBA found to be the least enhancer for MO, on the contrary, pure CR showed the lowest degradation rate. The effectiveness of additives for MO can be shown as 0.01g charcoal > 20 ml glucose > 20 ml NaCl > 100 µL CCl₄ > 10 ml Na₂SO₄ > 400 µL H₂O₂ > 10 ml sucrose > pure MO > TBA, while for CR is 0.01g charcoal > combined additives > TBA > 100 µL CCl₄ > pure CR, after sonication for 15 minutes in a sonicator.

Keywords: Methyl orange; congo red; sonolytic degradation; additives; inorganic salts; charcoal; degradation efficiency

INTRODUCTION

As the dyestuffs are not promptly degradable and cannot be eliminated from these aqueous discharges by conventional sewage plant treatments forasmuch as of their high biochemical stability, relatively high molecular weight and the presence of aromatic rings, dye contaminated wastewater has recently become a major concern (Chen et al. 2004; Houas et al. 2001; Panizza et al. 2007). The two significant sources of dye compounds are the textile and dyestuff industries (Martínez et al. 2012). Effluents

discharged by textile dyeing, food additives, cosmetics and printing industries are rich in color and this create aesthetic problem to the public. Dyes can be classified according to their functional groups; such as, anthraquinone, azo, metal complex, azo metal complex, phthalocyanine and reflecting the chromosphere structures on their constituent molecules (Suslick et al. 1989). Biodegradation of azo dyes, an important class of dyes, under aerobic conditions are resisted due to taking on azo bonds (-N-N-) having complex structures (Vajnhandl et al. 2007). These azo dyes molecule contribute about 70% of the total dye used (García-Montaña et al. 2008) and are found to be genotoxic,

carcinogenic and mutagenic to human population and other aquatic living beings. These molecules are also responsible for some aesthetic serious environmental problems, as they form hazardous aromatic amines under anaerobic conditions or due to reduction in waters and empoison the soil and groundwater (de Lima et al. 2007; de Aragao Umbuzeiro et al. 2005; Gregory et al. 1981; McKay et al. 1985; Pinheiro et al. 2004; Tsuboy et al. 2007). A hypothesis of about 15% of the total world occurrence of dyes is lost in wastewater stream during the dyeing and ending operations (Zollinger et al. 2003). Turbidity problems and the structure of toxic deterioration products can be created due to the presence of organic dyes at very low concentrations (Maleki et al. 2005; Teo et al. 2001; Vajnhandl et al. 2007). So, it is very necessary to remove them from water.

The several processes used for decolorization of dyes are emerged on physical or chemical methods including membrane filtration [Lee et al. 2006; Satyawali et al. 2008; Tan et al. 2006.], electrochemical technology (Chatzisyameon et al. 2006; Flint et al. 1991; Wang et al. 2007) and advance oxidation process (AOP) such as photochemical oxidation (Sánchez-Polo et al. 2007), photocatalysis (Behnajady et al. 2007; Chin et al. 2007; Patsoura et al. 2007) and ultrasonic wave (Gültekin et al. 2006; He et al. 2007a; Shen et al. 2006; Wang et al. 2007). In order to remove the dye compounds from wastewater efficiently, the combination of two or more individual processes have been developed. Some of these include ultrasound/H₂O₂ or ultrasound/ozone (Gogate et al. 2004; Martins et al. 2006; Vajnhandl et al. 2005; Zhang et al. 2006), UV light/H₂O₂ or UV/ozone (Domínguez et al. 2005; Gogate et al. 2004a), sonophotocatalysis (Gogate et al. 2004b; González et al. 2008; Kaur et al. 2007), Fenton, electro-Fenton, photo-Fenton processes (Daneshvar et al. 2008; Guivarch et al. 2003; Huston et al. 1999; Li et al. 2007; Martínez et al. 2009; Pignatello et al. 2006), photocatalytic oxidation and electrochemical processes (Aarthi et al. 2007; Arellano et al. 2007; Domínguez et al. 2005; Houas et al. 2001; Panizza et al. 2007; Villanueva et al. 2007). Among them sonochemical treatment has been found to be one of the successful technologies for degradation of organic pollutants such as dye.

The sound waves having frequencies in the range of 18 kHz-10 MHz with amalgamated acoustic wavelength of 10-0.1 cm, is known as ultrasound waves. The outcome of ultrasound in aqueous medium was first discovered by Alfred L. Loomis in 1927 (Richards et al. 1927). As soon as the cheap high-intensity ultrasound generators arrived in 1980, an enormous development occurred in sonochemistry (Suslick et al. 1989). The use of ultrasound may be broadly divided into two areas: i) high frequency ultrasound (2-10 MHz) and ii) low frequency ultrasound

(20-100 kHz) (Mason et al. 1989). The low frequency ultrasound or power ultrasound is mostly used by the sonochemical researchers (Price et al. 1992). In this technique, temporary degeneration of cavitation bubbles, which is conducted by an ultrasound wave, creates the free radicals. Ultrasonic sound induces the generation of cavitation bubbles in the liquid through which it is transmitted. The microbubbles produce during the approximate compression-rarefaction cycles, until then they attain a critical size. Further compression conducts to the collapse of the bubbles (Figure 1), with the concomitant deliverance of heat energy and production of chemically active species during the last phase of the bubble collapse (Adewuyi et al. 2001; Colussi et al. 1998; Thompson, L.H., 1999). The chemical outcomes are a direct result of the elevated temperatures, on the order of 5000 K and pressure, in the range of hundreds of bars, which are related in the gaseous cavities when the cavities size is reduced in many orders of magnitude within a few microseconds (Flint et al. 1991). The particularly high temperature and pressure formed in collapsing cavitation bubbles in aqueous solutions lead to the thermal dissociation of water vapor into reactive hydroxyl radicals and hydrogen atoms (eq. 1-8) (Hart et al. 1987; Hart et al. 1985). The radical species generated can recombine, react with other gaseous species present in the cavity, or diffuse out of the bubble into the bulk liquid medium where they are capable to react with solute molecules. There are two major mechanisms for the sonolytic degradation of organic compounds in dilute aqueous solution, a) thermal decomposition of the volatile pollutant molecules entrapped inside the bubbles (Weavers et al. 1998), and b) the reaction of OH radicals with the solute adsorbed at the bubble interface, in the bulk, and to some extent within the bubbles (Hoffmann et al. 1996). The limitation of degradation in bulk liquid is limited by the quantity of hydroxyl radicals diffused into the water (Henglein et al. 1993).

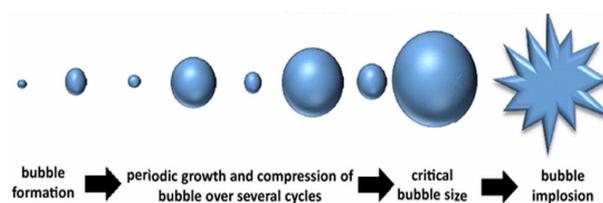
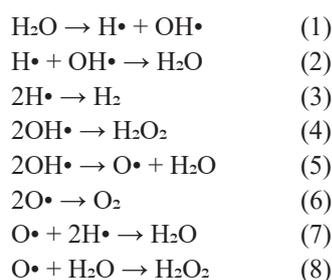


FIGURE 1. Bubble explosion and formation of hotspot



Compared with various technologies of organic pollutant dealing, sonochemical deterioration has significant dominances such as safety, cleanliness and energy conservation without causing secondary pollution (Hao et al. 2003; Teo et al. 2001). Successful sonochemical degradation has been carried out for different kind of dyes like Basic Blue 41 (Abbasi et al. 2008), C.I. Direct Red 23 (Song et al. 2007), Methyl Orange (Okitsu et al. 2008), C.I. Reactive Black 5 [He et al. 2007a], C.I. reactive Yellow 84 (He et al. 2007b) etc. have been reported. It has also been reported that, usage of various additives can affect the degradation of dyes (Islam et al. 2018).

In this report, degradation characteristics of two azo dyes [Methyl Orange (MO) and Congo Red (CR)], using ultrasonic wave in the presence of several additives, has been reported.

EXPERIMENT DESIGN AND PROCEDURE

2.1 Chemicals

Methyl orange [IUPAC name: Sodium 4- {[4-(dimethyl amino) phenyl] diaziny] benzene-1-sulfonate] (Figure 2a), an azo dye, having molecular weight of 327.34 g/mol and solubility in water 5 g/L at 20°C [Bejarano-Pérez et al. 2007] was purchased from Merck Life Science Private Limited. Another azo dye named congo red [IUPAC name: disodium 4-amino-3-[4-[4-(1-amino-4-sulfonatophthalen-2-yl) diazenylphenyl] phenyl] diaziny] naphthalene-q-sulfonate] (Figure 2b) having molecular weight of 696.68 g/mol and solubility in water 25 g/L at 20°C [Bejarano-Pérez et al. 2007] was purchased from Loba Chemie Pvt Ltd. All other chemicals like NaCl, Na₂SO₄, H₂O₂, CCl₄, glucose, sucrose and tert-butyl alcohol were purchased from Merck chemical company, while charcoal from BDH chemical company.

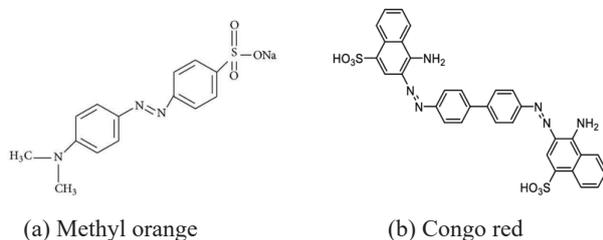


FIGURE 2: Structure of azo dyes

The ultrasonic waves were generated using an ultrasonic bath (Bandelin electronic RK 255H, Germany) at 40 kHz and 120 W. Absorbance measurements were performed using UV Mini Shimadzu 1240 spectrophotometer.

2.3 Experimental Procedure

2.3.1 Preparation of Solutions

12.5 mg/L solution of MO and 200 mg/L solution of CR were prepared. The solutions of the additives (NaCl, Na₂SO₄, glucose and sucrose) were prepared according to their respective highest saturation point in 100 ml distilled water (36g, 13.9g, 100g and 200g respectively).

2.3.2 Degradation Process

Sonolytic deterioration process was conducted in batch method. The deterioration of dyes were carried out both in absence and presence of additives. Each time 100 ml solution of dye was taken with different quantities of additives. MO was mixed with NaCl, Na₂SO₄, glucose and sucrose (10 ml and 20 ml each), H₂O₂ and CCl₄ (100 μL, 200 μL and 400 μL each), tert-butyl alcohol (TBA) (1 ml) and charcoal (0.01g), while CR was mixed with 100 μL of CCl₄, 1 ml TBA, 0.01g charcoal and combination mixture of the mentioned additives.

The sonication process was carried out in open air at room temperature for 15 minutes. The commercial glass vessel was flat bottomed and mounted at a constant position (4.0 mm from the oscillator). The sonicated solution was extracted (1ml) after every 0, 5, 10 and 15 minutes by a glass syringe without exposing the sample to air. The extracted solutions were diluted with 3 times volume of distilled water before the measurement of the absorbance.

3. Results and Discussion

3.1 Ultrasonic Degradation of MO in Present of Additives

The degradation rates of MO with and without additives are shown in Figure 3-5. It is clear from the data that, all additives drastically enhanced the degradation rate as compared to that of pure MO.

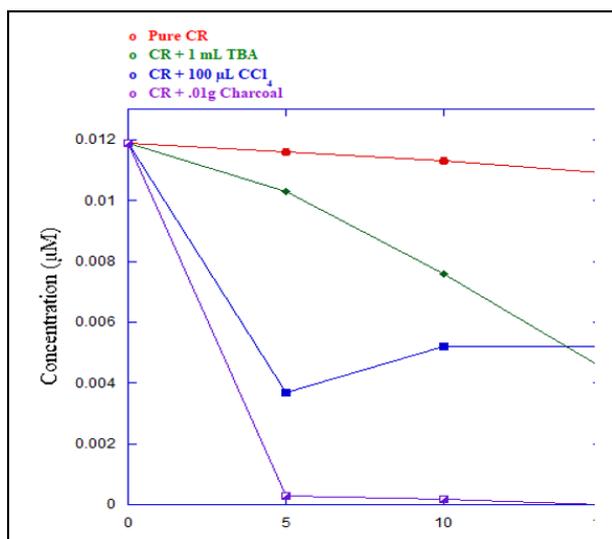
3.1.1 Effects of Inorganic Salts

In order to study the effect of inorganic salt addition on the degradation of dye, 10 ml [Figure 3a] and 20 ml [Figure 3b] of NaCl and Na₂SO₄ each were added in solution of MO. It can be seen from the figure, the addition of both salts enhanced the degradation of MO. Moreover, the increased concentration of the salts increased the degradation rate. This phenomenon can be explained as; addition of salt to dye solution collisions the dye molecules from the bulk liquid phase to the bulk-bubble interface [Findik et al. 2007; Seymour et al. 1997]. The presence of salts may increase the hydrophilicity, the surface tension and ionic strength of the aqueous phase and decrease the

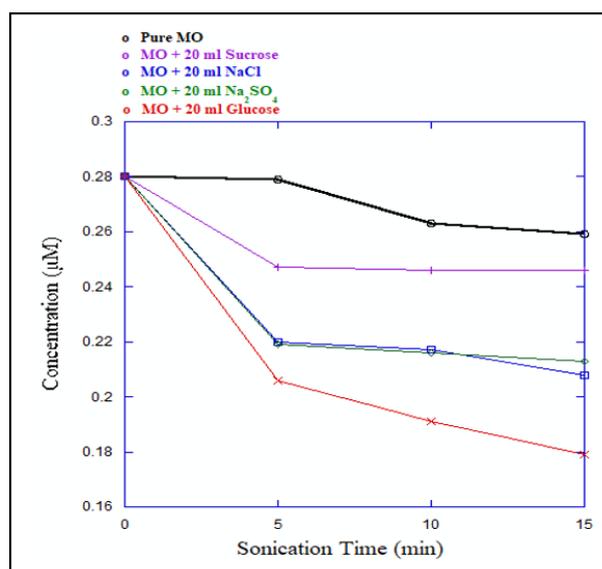
vapor pressure [Findik et al. 2007; Seymour et al. 1997]. All these factors help in collapsing of the bubbles more violently, resulting in high degradation of MO. From the comparison, it can be accomplished that NaCl is more effective for the degradation than Na₂SO₄.

3.1.2 Effect of Glucose and Sucrose

The sonolytic degradation of MO solution was investigated in the presence of organic competitors such as glucose and sucrose. The effect of glucose and sucrose addition, 10 ml [Figure 3a] and 20 ml [Figure 3b] each, on the sonochemical degradation of MO has been shown in Figure 3. It was observed that the sonolytic degradation of MO was enhanced due to the presence of glucose and sucrose. But in case of sucrose, the rate of degradation was hindered, when it was exposed to the ultrasonic wave for a long time. Another graph [Figure 3b] shows that, the excess concentration of sucrose also creates obstacles for the degradation rate. Excessive amount of sucrose may interfere with the introduction of ultrasound in the liquid. So, it can be assumed that, glucose is best option here.



(a) 10 ml solution of additives



(b) 20 ml solution of additives

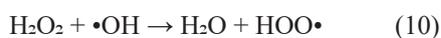
FIGURE 3. Sonolytic degradation of MO in presence of inorganic salts, glucose and sucrose

3.1.3 Effect of H₂O₂ and CCl₄

Sonolytic degradation of MO was also carried out in presence of H₂O₂ and CCl₄. The rate of dye degradation was increased by the addition of H₂O₂ to the ultrasound system [Figure 4a]. This is due to the decomposition of hydrogen peroxide into hydroxyl radicals causing high degradation rate [Behnajady et al. 2008; Teo et al. 2001]. The degradation rate increased with the increase of H₂O₂ concentration from 100 μL to 400 μL. The enhanced degradation rate of MO is due to the attack of additional free radicals, generated from H₂O₂ (eq. 9)

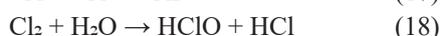
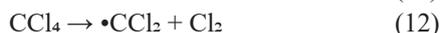


Though, the degradation rate was not appreciably high, it shows far better degradation than pure MO. Due to the high-water solubility and the low volatility, H₂O₂ may be mainly present not inside the cavitation bubbles but in the bulk solution. From previous research work it is clear that, H₂O₂ shows lower intensification of degradation rate in the presence of high concentrations due to the increased level of OH radical scavenging by H₂O₂ itself (eq. 10) [Merouani et al. 2010].



On the other hand, CCl₄ appreciably enhanced the degradation rate [Figure 4b], far better than H₂O₂ [Figure 4a]. This improvement is due to the degradation of CCl₄

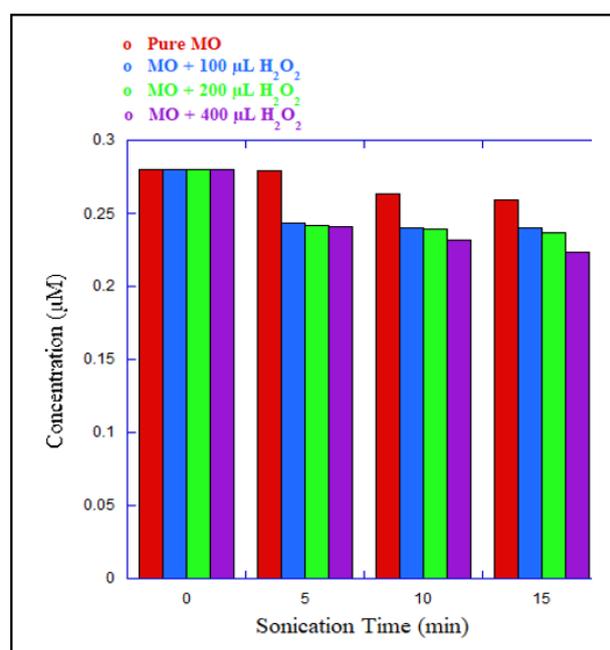
by pyrolytic cleavage in cavitation bubbles, which conducts to the release of oxidizing agents that can react with dye molecules. The sonolytic degradation of CCl_4 has been studied by several research groups [Bhatnagar et al. 1994; Francony et al. 1996; Hua et al. 1996; Pétrier et al. 1997]. The overall reaction mechanism can be written as [11-18].



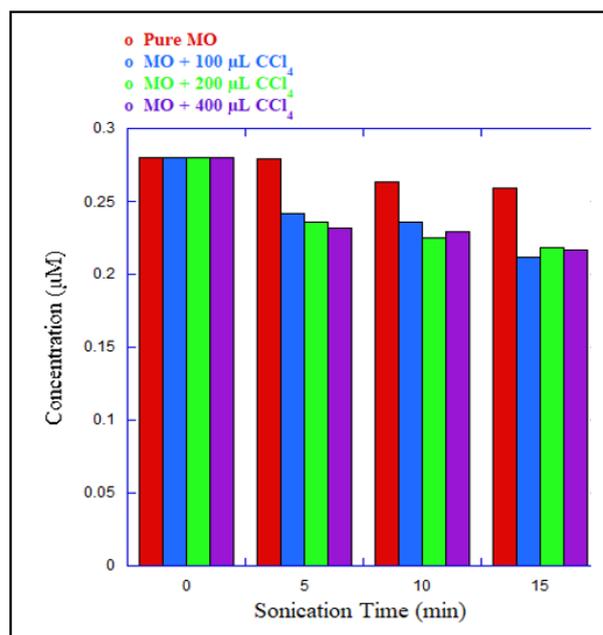
The sonolytic degradation of CCl_4 generates $\bullet\text{Cl}$ radicals that will lead to a series of recombination reaction conducting to the formation of additional active species, such as HClO , Cl_2 and chlorine-containing radicals ($\bullet\text{Cl}$, $\bullet\text{CCl}_3$ and $\bullet\text{CCl}_2$), having strong oxidizing property, which will markedly accelerate the degradation of MO in aqueous solution.

3.1.4 Effect of tert-Butyl Alcohol (TBA) and Charcoal

Hydroxyl attack is the main reason for the sonolytic degradation. TBA, an effective hydroxyl radical scavenger, has been taken under investigation to examine the fact.



(a) H_2O_2 as additive

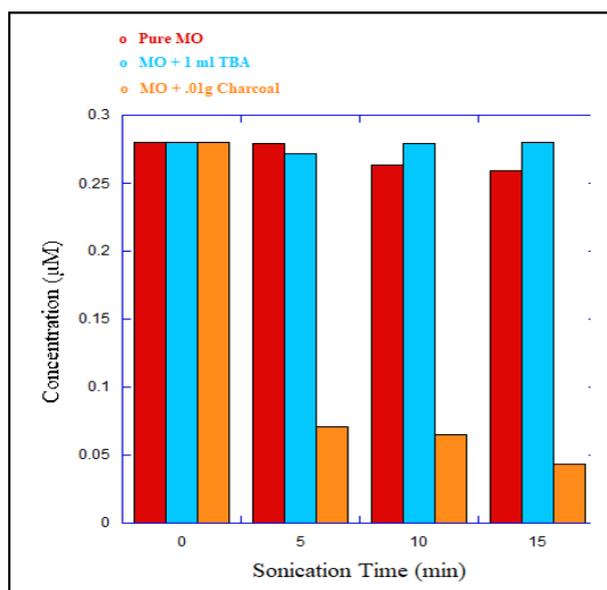


(b) CCl_4 as additive

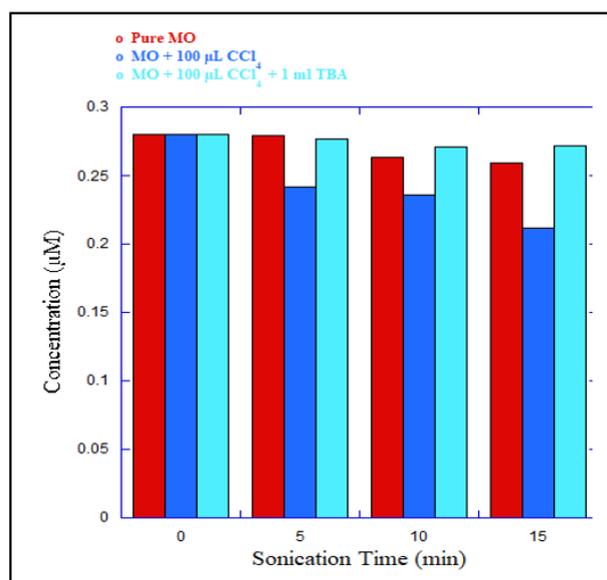
FIGURE 4. Sonolytic degradation of MO in presence of H_2O_2 and CCl_4

The tert-butyl alcohol is able to scavenge OH radicals in the bubble and prevent the accumulation of OH radicals at the interface of the bubble. Extensive work on the sonochemistry of tert-butyl alcohol has been reported earlier [Tauber et al. 1999]. Due to the scavenge, TBA showed the lowest degradation rate than any other additives during ultrasonic degradation [Figure 5a]. Even it reduced the enhancing capability of CCl_4 [Figure 5b].

On the contrary, charcoal (activated carbon) showed the higher degradation rate among all the additives used [Figure 5a]. A little amount of activated carbon brought a huge change in the degradation. There can be two reasons: (i) activated carbon can readily adsorb the dye materials and (ii) it helps in producing more H_2O_2 in water, which then degrade to produce hydroxyl radicals to enhance the degradation. Further study is required in this field.



(a) TBA and Charcoal as additive

(b) TBA + CCl₄ as additiveFIGURE 5. Sonolytic degradation of MO in presence of TBA, CCl₄ and charcoal

3.2 Ultrasonic Degradation of CR in Presence of Additives

Another azo dye congo red (CR) was also taken under investigation with a few numbers of additives like H₂O₂, CCl₄, TBA and charcoal.

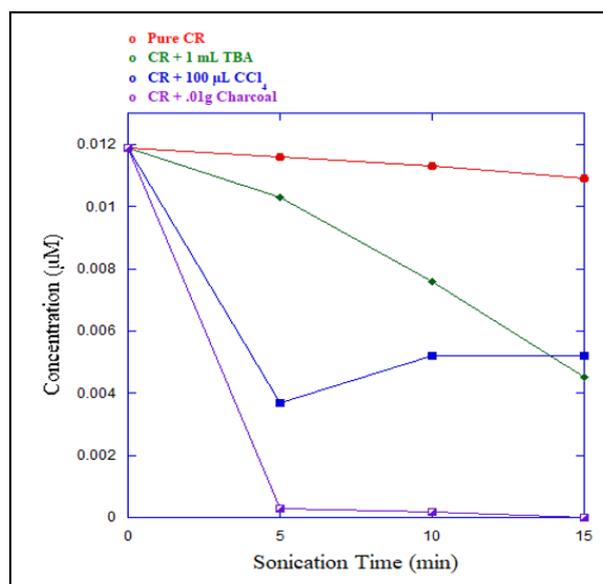
3.2.1 Effect of CCl₄

100 µL CCl₄ was mixed with CR to determine the effect of CCl₄. From Figure 4b, in case of MO, we have seen that,

100 µL CCl₄ enhanced the sonolytic degradation. But when it was mixed with CR, it showed some deviations from its character [Figure 6]. Though it enhanced the degradation largely as compared to pure CR, degradation rate was not the same all the time. In the beginning period (after 5 mins) it highly helped in the degradation of the dye. But, afterwards, the degradation rate was lower than the previous ones (15mins<10mins<5mins)

3.2.2 EFFECT OF TBA

TBA is an effective hydroxyl radical scavenger. But unexpectedly, the rate of CR degradation was accelerated when TBA was added [Figure 6]. This enhancement may be attributed to the relatively low dose of TBA, at which competition for OH radical is unlikely and the rapid decomposition of the additive to produce reactive methyl radicals (\bullet CH₃), which are the main products of high temperature pyrolysis of the additive [Ince et al. 2009]. Additionally, TBA, a surfactant substance, can reduce the surface tension and thus reduce the cavitation threshold and facilitate the generation of bubbles and, therefore, the production of hydroxyl radicals [Merouani et al. 2010].

FIGURE 6. Sonolytic degradation of CR in presence of TBA, CCl₄ and charcoal

3.2.3 Effect of Charcoal

As mentioned before, charcoal is a very good adsorbent for dye. Figure 6 shows that, in presence of charcoal, the concentration of CR become zero. Adsorption capability along with the sonolytic degradation was used to remove the dye completely from the wastewater.

3.2.4 Effect of Combined Additives (CA)

100 μL CCl_4 , 1 ml TBA and 0.01g charcoal was combined together with CR during the sonication process to see the effect of additives when combined. From Figure 7, it is clear that, the combination of additives enhanced the degradation process almost like only charcoal is present for the first 5 minutes. Then no more degradation occurs. This may be due to the combined scavenge effect of TBA and CCl_4 .

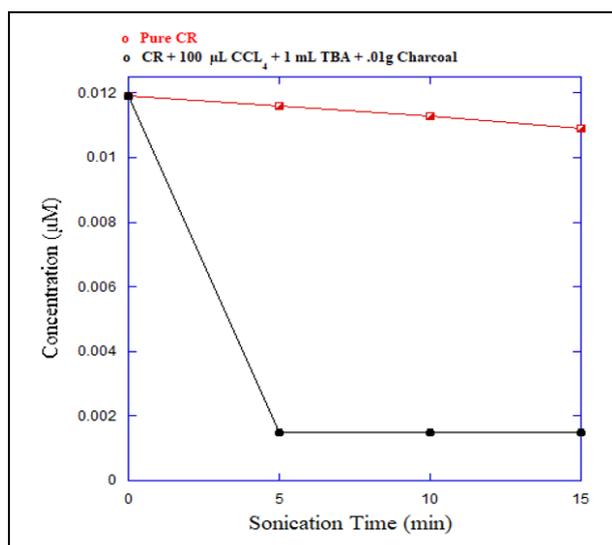


FIGURE 7. Sonolytic degradation of CR in presence of TBA, CCl_4 and charcoal

CONCLUSION

The present work has shown the effect of various additives on the sonolytic degradation of two types of azo dyes, MO and CR, at room temperature. From the research work, it is apparent that, all the additives highly enhanced the degradation rate as compared with pure dye solution. But the fact is, some of the additives showed a good degradation rate with low concentration at the beginning, but then the activity started to fall with the increased concentration and sonication duration. From the results of MO, it can be concluded that, the consumption of charcoal was the most beneficial, while TBA failed to show any satisfactory progress. After 15 minutes of sonication, the order of effectiveness is 0.01g charcoal > 20ml glucose > 20 ml NaCl > 100 μL CCl_4 > 10 ml Na_2SO_4 > 400 μL H_2O_2 > 10 ml sucrose > pure MO > TBA. On the other hand, the order of effectiveness for CR after 15 minutes of sonication is 0.01g charcoal > CA > TBA > 100 μL CCl_4 > pure CR, where charcoal showed the greatest enhancement in the degradation rate.

ACKNOWLEDGEMENT

The authors are highly thankful to Islamic University, National University, Jessore University of Science and Technology, Osaka Prefecture University and Bangladesh Army University of Engineering & Technology (BAUET) on its excellent help and support to conduct this research.

DECLARATION OF COMPETING INTEREST

None

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