

## **EFFECT OF ADDITIVES ON DECOMPOSITION OF METHYL ORANGE AND CONGO RED DYES FOUND IN INDUSTRIAL WASTEWATER**

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### **ABSTRACT**

A number of textile industries are using different types of dyes which can be a threat to the environment when they are directly exposed without any treatment. The advanced oxidation process (AOP) has become one of the popular methods in which the dye molecules are degraded using microwave irradiation. This method has become popular due to its eco-friendly and cost-efficient characteristics. In this article, the author has reported the degradation of two azo dyes, named methyl orange (MO) and congo red (CR), in the presence of inorganic salts, some additives like charcoal, H<sub>2</sub>O<sub>2</sub>, CCl<sub>4</sub>, *tert*-butyl alcohol (TBA), glucose and sucrose. A comparative study was conducted to find out the relative degradation rate enhanced by various additives. For both of the dyes, it was found that charcoal was the most effective additive and significantly enhanced the degradation rate mainly due to its high adsorption capability. TBA was found to be the least enhancer. The order of effectiveness based on the irradiation period for MO was 0.01g charcoal > 20 ml NaCl > 20 ml Na<sub>2</sub>SO<sub>4</sub> > 20ml glucose > 10 ml sucrose > 100 μL CCl<sub>4</sub> > 200 μL H<sub>2</sub>O<sub>2</sub> > pure MO > TBA. On the other hand, the order of effectiveness for CR for the same condition of irradiation was Combined additives > 0.01g charcoal > TBA > 100 μL CCl<sub>4</sub> > pure CR.

**Keywords:** Azo dyes, advanced oxidation process, microwave irradiation, inorganic salts, additives

### **1. INTRODUCTION**

Many industries like textile, tanning, food, and photoelectrochemical use various types of dyes very often, which regularly produce a huge number of colored wastewater. The use of organic dye is increasing day by day and as a result, it is obvious to find some solution to remove and dispose of these dye materials to reproduce clean and safe water [1,2]. There are different types of dyes, among them the majority production is of azo dyes (70%) all over the world [3]. These compounds are characterized by the chromophore group of azo bonds (–N=N–) linked to aromatic structures with functional groups such as –NH<sub>2</sub> and –SO<sub>3</sub>H [4,5]. As these dyes have high-cost effectiveness and better properties, they are being highly used in the textile, paper, leather, pharmaceutical and food industries [6]. These dye molecules have diverse effects on human health. The azo dyes and their intermediates, during the degradation, have toxic, carcinogenic and mutagenic effects which can cause severe health hazards to mankind [7]. So, it is necessary to remove these hazardous dye particles from wastewater. The diverse processes

used for the decolorization of dyes are based on physical or chemical methods including membrane filtration [8,9], electrochemical technology [10,11] and advance oxidation process (AOPs) such as photochemical oxidation [12], photocatalysis [13,14] and ultrasonic wave [15,16]. In order to remove the dye compounds from wastewater efficiently, the combination of two or more individual processes has been developed. Some of these include ultrasound/H<sub>2</sub>O<sub>2</sub> or ultrasound/ozone [17,18] UV-light/H<sub>2</sub>O<sub>2</sub> or UV/ozone [19,20] sono photocatalysis [21,22] Fenton, electro-Fenton, photo-Fenton processes [23,24,25] photocatalytic oxidation and electrochemical processes [26,27,28]. A different technique known as adsorption often makes use of activated carbon from high-carbon-content natural resources. Due to its convenience, simplicity, ease of use and ability to be reused, activated carbon is frequently used to remove colors from wastewater [29-34]. However, some commercial activated carbon continues to be an expensive material while being widely used in wastewater treatment [32–34]. As a result, there is growing interest in less expensive options to commercially available activated carbon that would nonetheless offer effective and affordable ways to remove colours from contaminated water. Due to this, inexpensive agricultural waste materials with the ability to act as adsorbents have been acknowledged as a sustainable wastewater treatment method that reduces waste, recovery, and reuse [35-42]. With the help of carbonization and activation by KOH, NaOH and H<sub>2</sub>SO<sub>4</sub>, powdered activated carbon (PAC) was able to remove colors from an aqueous solution. A study demonstrated that a pseudo-second order model suited the experimental data very well. According to the Langmuir isotherm, the maximal adsorption capacity for congo red (CR) and methylene blue (MB) removal was 458.43 mg g<sup>-1</sup> and 769.23 mg g<sup>-1</sup>, respectively. To remove colours from industrial effluent, the powdered activated carbon (PAC) can do it effectively and economically [43].

Recently another process has been developed named as Microwave (MW) radiation degradation process. MW radiation is a wavelength band of the electromagnetic spectrum and its frequency ranges from 300 MHz to 300 GHz [44]. MW irradiation exhibits higher heating rates, reduced activation energies [45,46] increased reaction rates [47,48], improved energy efficiency, and reduced equipment size [49]. In addition, MW irradiation exhibits selective and volumetric heating properties (which are not features of conventional heating), has different thermodynamic functions to those found for conventional heating, and has excellent controllability [49,50]. Due to the above advantages, MW irradiation has received considerable attention as an effective method for various analytical and chemical applications, such as organic and inorganic synthesis [51,52] remediation of contaminated soil [53,54] macromolecule degradation [55] etc. In particular, in the field of organic wastewater treatment, many kinds of organic pollutants in wastewater have been removed effectively, such as volatile organic compounds [56] dyes [57] ammonia nitrogen [58] phenolic compound [59] etc. MW equipment used for chemical reactions can be classified into two types, namely a multi-mode type and a mono-mode type, according to the number of working modes in the cavity [60,61]. In the organic wastewater treatment field, due to the large volumes of wastewater that are involved, multimode MW equipment is always used because of the larger capacity provided by the multimode cavity.

In this paper, degradation characteristics of two azo dyes Methyl Orange (MO) and Congo Red (CR) have been reported using microwave irradiation in the presence of inorganic salts: NaCl, Na<sub>2</sub>SO<sub>4</sub>, additives: charcoal, H<sub>2</sub>O<sub>2</sub>, CCl<sub>4</sub>, TBA.

## 2. MATERIAL AND METHODS

### 2.1 Chemicals

Methyl orange [IUPAC name: Sodium 4-[[4-(dimethyl amino) phenyl] diazanyl] benzene-1-sulfonate] (**Fig. 1a**), an azo dye, having a molecular weight of 327.34 g/mol and solubility in water 5 g/L at 20 °C [47] 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] diazanyl-naphthalene-q-sulfonate] (**Fig. 1b**) having a molecular weight of 696.68 g/mol and solubility in water 25 g/L at 20° [62] was purchased from Loba Chemie Pvt. Ltd., India. All other chemicals like NaCl, Na<sub>2</sub>SO<sub>4</sub>, H<sub>2</sub>O<sub>2</sub>, CCl<sub>4</sub>, glucose, sucrose and *tert*-butyl alcohol (TBA) were purchased from Merck chemical company, Germany, while charcoal from BDH chemical company, UAE.

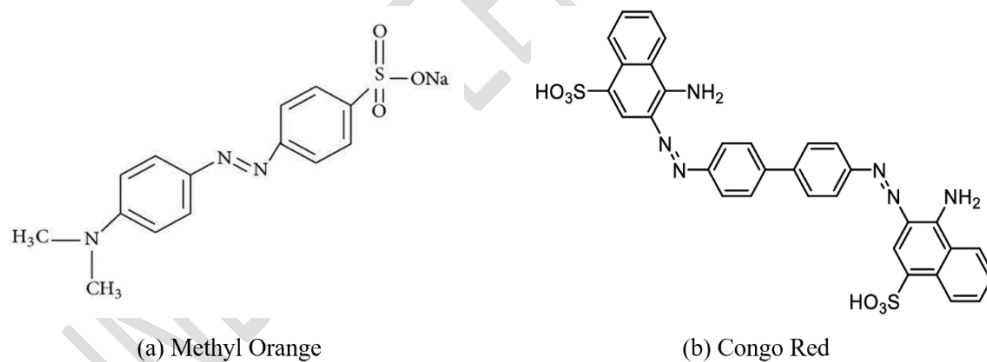


Figure 1: Structure of azo dyes

### 2.2 Preparation of Solutions

A 12.5 mg/L solution of MO and a 200 mg/L solution of CR were prepared. The solutions of the additives (NaCl, Na<sub>2</sub>SO<sub>4</sub>, glucose and sucrose) were prepared according to their respective highest saturation point in 100 ml distilled water (36g, 13.9g, 100g and 200g respectively). Dyestuff in water can adsorb and reflect light, thereby reducing the natural transparency of water, and can consume a lot of dissolved oxygen in water, causing water hypoxia and affecting aquatic life and microbial growth, and so, in turn, undermining the aquatic ecosystem [63]. Dye effluent normally contains about 10 to 50 mg/L, but

even at 1 mg/L, dyes are easily noticeable and thus may be perceived as being contaminants, and unacceptable [64]. As a result, the treatment of dye effluent has been investigated extensively in modern times [65].

### 2.3 Microwave Irradiation Process

The microwave irradiation process was conducted in batch method Microwave Oven ZR659YW 2.2 cu. ft. 1200W. The degradation of dye was carried out both in the absence and presence of additives (Fig. 2). 100 ml solution of dye was taken with different quantities of additives. MO was mixed with NaCl, Na<sub>2</sub>SO<sub>4</sub>, glucose and sucrose (10 ml and 20 ml each), H<sub>2</sub>O<sub>2</sub> and CCl<sub>4</sub> (100 μL, 200 μL and 400 μL each), tert-butyl alcohol (TBA) (1 ml) and charcoal (0.01g), while CR (200 mg/L) was mixed with 100 μL of CCl<sub>4</sub>, 1 ml TBA, 0.01g charcoal and combination mixture of the mentioned additives.

The irradiation process was carried out in a microwave-resistant beaker in a microwave oven for 30 seconds. The irradiated solution was taken (1ml) every 0, 10, 20 and 30 seconds by a glass syringe without exposing the sample to air. The extracted solutions were diluted with 3 times the volume of distilled water before the measurement of the absorbance.

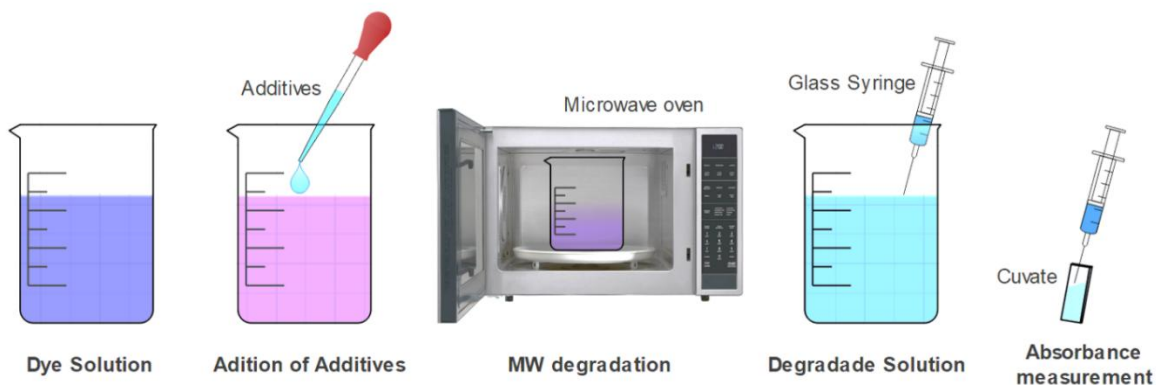


Figure 2: Microwave degradation process of dye solution with additives

### 3. RESULTS AND DISCUSSION

Figure 3 shows the dependence of the concentration of MO on the irradiation time at different periods. Each point shows the average data of at least two experimental runs. In the figure, the curve has shown the first-order kinetic model function of equation-1 [66].

$$X = ae^{-kt} \quad (1)$$

Here the constant  $a$  is an initial concentration of MO, 12.5 mg/L and  $k$  is the fitting parameter. From Figure 1 it is observed that the rate of degradation increased with increasing irradiation time. It also satisfies the first-order reaction.

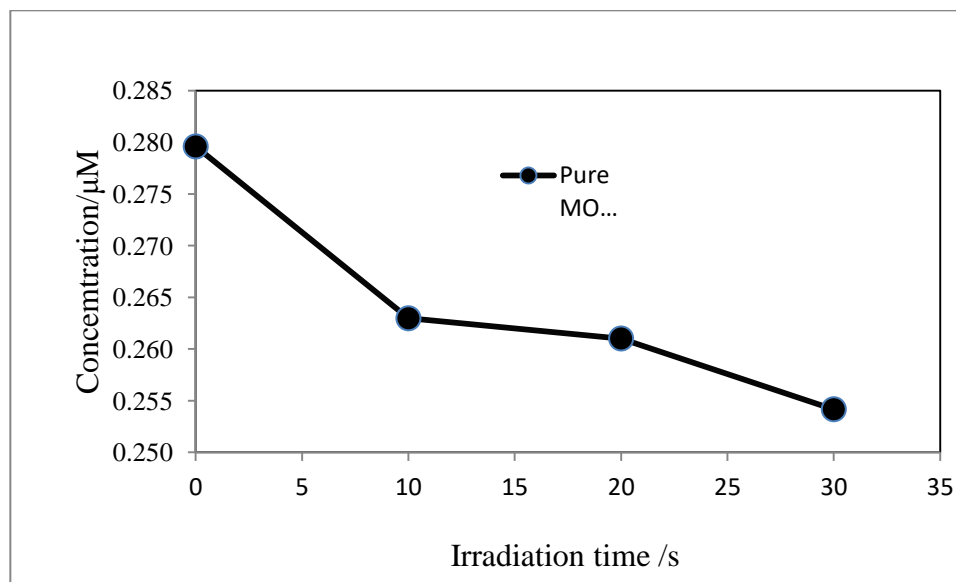


Figure 3: Time dependence of irradiation of MO in the open atmosphere

### 3.1 Microwave Degradation of MO in the Presence of Additives

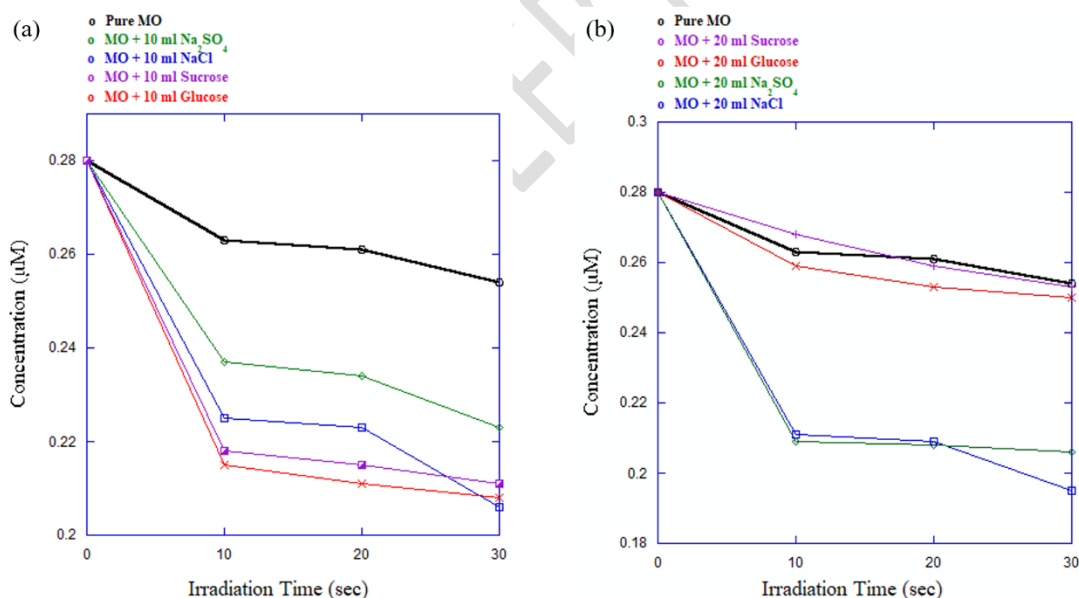
The degradation rates of MO with and without additives using the microwave irradiation process are shown and discussed below. It is clear from the data that, all additives drastically enhanced the degradation rate as compared to that of pure MO [67].

#### 3.1.1 Effect of Inorganic Salts

In order to study the effects of the addition of the inorganic salt on the degradation of dye, 10 ml (**Fig. 4a**) and 20 ml (**Fig. 4b**) of NaCl and Na<sub>2</sub>SO<sub>4</sub> each was added in the 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. After comparison, it can be concluded that NaCl is more effective for the degradation than Na<sub>2</sub>SO<sub>4</sub> [68]. A research work published regarding the Sonochemical degradation of 4-chlorophenol, phenol, catechol and resorcinol in the absence and presence of Na<sub>2</sub>SO<sub>4</sub> or NaCl under Ar [68]. The rate of degradation of phenolic compounds obeyed a pseudo-first-order rate constant.

### 3.1.2 Effect of Glucose and Sucrose

The microwave degradation of MO solution was investigated in the presence of organic competitors such as glucose and sucrose. The effect of the addition of glucose and sucrose, 10 ml (**Fig. 4a**) and 20 ml (**Fig. 4b**) on the microwave degradation of MO has been shown in **below**. It was observed that the degradation of MO was enhanced due to the presence of glucose and sucrose. But in the case of sucrose, the rate of degradation was hindered, when it was exposed to microwave irradiation for a long time. Another graph (**Fig. 4b**) shows that the excess concentration of sucrose also creates obstacles to the degradation rate. Excessive amounts of sucrose may interfere with the introduction of the radiation in the liquid. So, it can be assumed that the use of glucose is more effective than sucrose **published** the decolorization of 200 **mg /L dye**, the increase of glucose concentration from 5 to 10 g/L obviously increased the color removal efficiency [69].



**Figure 4: Microwave degradation of MO in presence of inorganic salts, glucose and sucrose, (a) 10 ml solution of additives, (b) 20 ml solution of additives.**

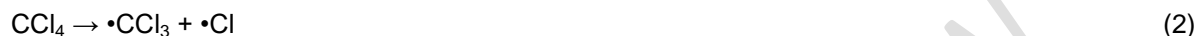
### 3.1.3 Effect of $\text{H}_2\text{O}_2$ and $\text{CCl}_4$

The microwave degradation of MO was also carried out in the presence of  $\text{H}_2\text{O}_2$  and  $\text{CCl}_4$ . The rate of dye degradation was increased by the addition of  $\text{H}_2\text{O}_2$  to the microwave oven (**Fig. 5a**). The

degradation rate increased with the increase of H<sub>2</sub>O<sub>2</sub> concentration from 100 μL to 400 μL. The enhanced degradation rate of MO is due to the attack of additional free radicals, generated from H<sub>2</sub>O<sub>2</sub> (eq. 1). Though, the degradation rate was not appreciably high, it shows far better degradation than pure MO.



On the other hand, CCl<sub>4</sub> appreciably enhanced the degradation rate (**Fig. 5b**), far better than H<sub>2</sub>O<sub>2</sub> (**Fig. 5a**). This improvement is due to the degradation of CCl<sub>4</sub> by pyrolytic cleavage, which conducts to the release of oxidizing agents that can react with dye molecules. The overall reaction mechanism can be written as (2–9).



The degradation of CCl<sub>4</sub> generates •Cl radicals that will lead to a series of recombination reaction conducting to the formation of additional active species, such as HClO, Cl<sub>2</sub> and chlorine-containing radicals (•Cl, •CCl<sub>3</sub> and •CCl<sub>2</sub>), having strong oxidizing properties, which will markedly accelerate the degradation of MO in aqueous solution.

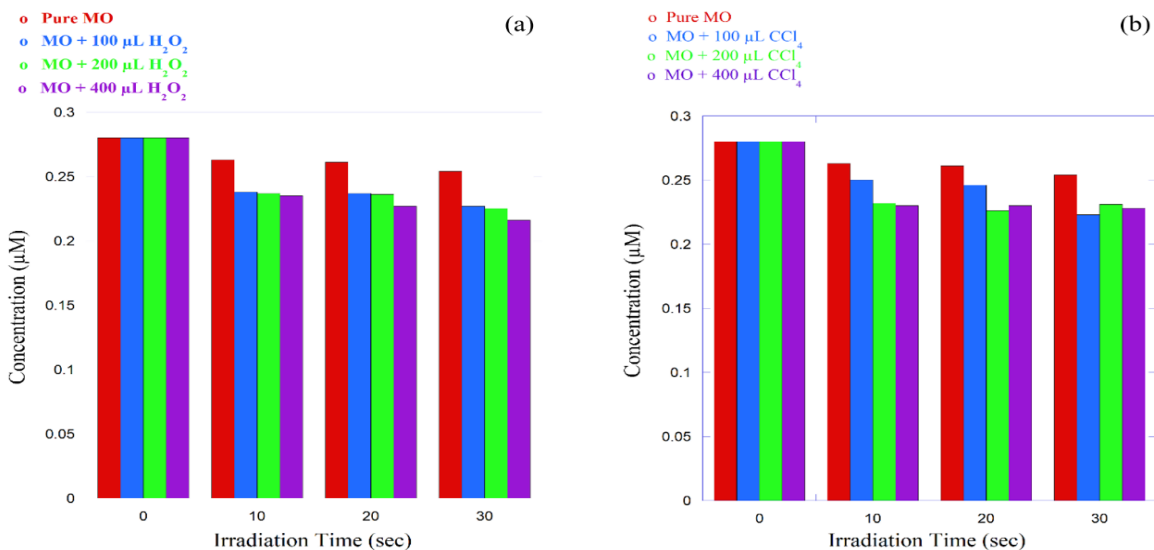


Figure 5: Microwave degradation of MO in presence of H<sub>2</sub>O<sub>2</sub> and CCl<sub>4</sub>, (a) H<sub>2</sub>O<sub>2</sub> as additive, (b) CCl<sub>4</sub> as additive

#### CCl<sub>4</sub> Concentration base degradation:

Figure 6 shows the time dependence of irradiation of MO in the presence of different concentrations of CCl<sub>4</sub>. Each point shows the average data of at least two experimental runs. The figure observed that the rate of degradation increased with the increasing irradiation time and doses of CCl<sub>4</sub>. The rate also significantly increased in the presence of 200 µL or 400 µL CCl<sub>4</sub>. During the experimental period, CCl<sub>4</sub> was directly added to the solution.

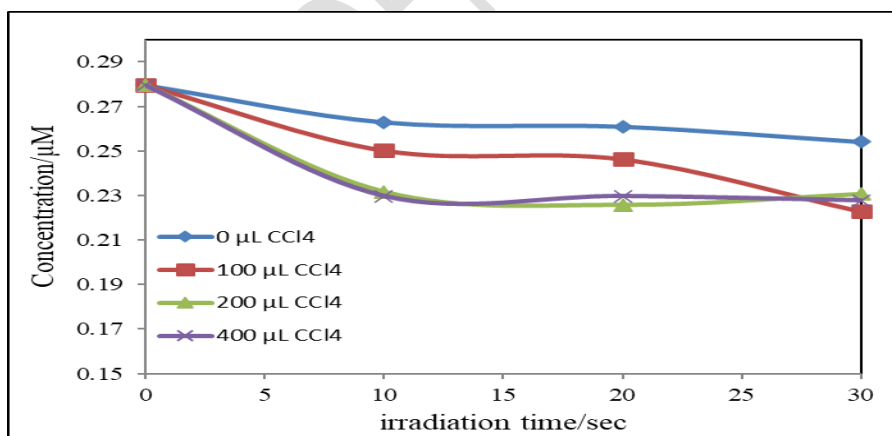


Figure 6: Time dependence of irradiation of MO in the presence of different concentrations of CCl<sub>4</sub>

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

Hydroxyl attack is the main reason for microwave degradation. TBA, an effective hydroxyl radical scavenger, has been taken under investigation to examine the fact. 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. Due to the scavenging property, TBA showed the lowest degradation rate than any other additives during microwave irradiation (Fig. 7a). Even it reduced the enhancing capability of  $\text{CCl}_4$  (Fig. 7b). On the contrary, charcoal (activated carbon) showed a higher degradation rate among all the additives used (Fig. 7b). 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  $\text{H}_2\text{O}_2$  in water, which then degrade to produce hydroxyl radicals to enhance the degradation. Further study is required in this field.

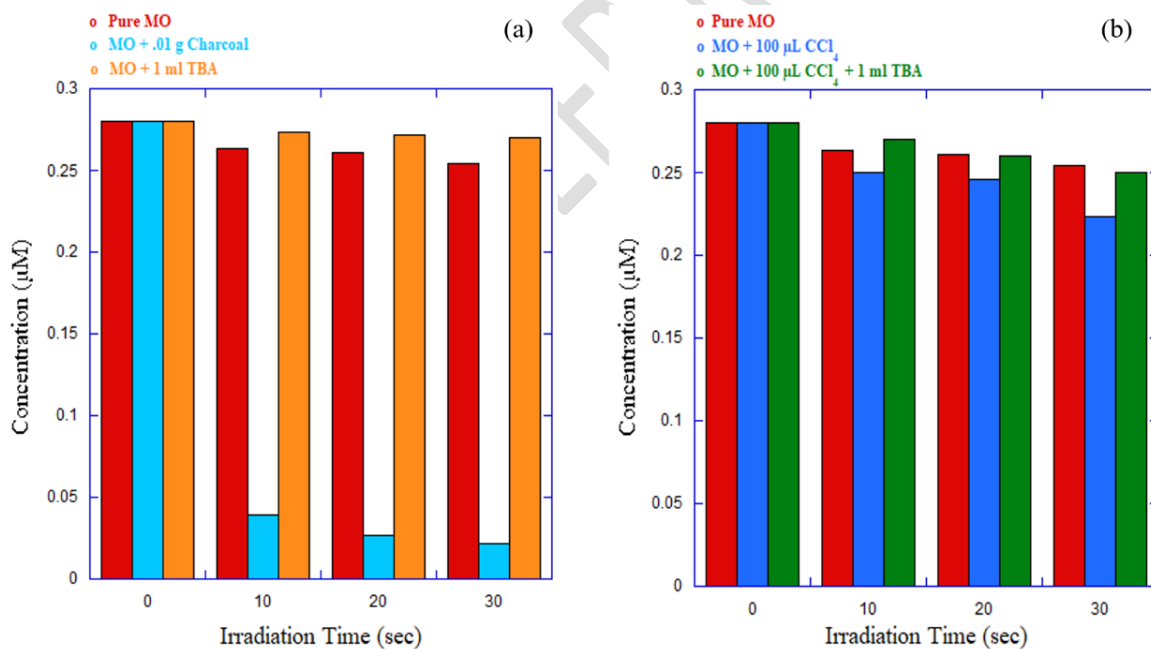


Figure 7: Microwave degradation of MO in the presence of TBA,  $\text{CCl}_4$ , and charcoal, (a) TBA and Charcoal as an additive, (b) TBA +  $\text{CCl}_4$  as an additive

## 3.2 Microwave Degradation of CR in the Presence of Additives

Another azo dye Congo red (CR) was also taken under investigation with a few numbers of additives like  $\text{H}_2\text{O}_2$ ,  $\text{CCl}_4$ , TBA and charcoal. The effects of different additives to degrade CR using the MW irradiation process are discussed below.

### 3.2.1 Effect of $\text{CCl}_4$

A 100  $\mu\text{L}$   $\text{CCl}_4$  was mixed with CR to determine the effect of  $\text{CCl}_4$ . From Fig. 6b, in the case of MO, we have seen that, 100  $\mu\text{L}$   $\text{CCl}_4$  enhanced the degradation. When it was mixed with CR, it enhanced the degradation largely as compared to pure CR (Fig. 8).

### 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 (**Fig. 8**). 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 ( $\bullet\text{CH}_3$ ), which are the main products of high-temperature pyrolysis of the additive [70]. 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 [71].

### 3.2.3 Effect of Charcoal

As mentioned before, charcoal is a very good adsorbent for dye. Fig. 8 shows that, in the presence of charcoal, the concentration of CR became almost zero. Adsorption capability along with microwave irradiation was used to remove the dye completely from the wastewater.

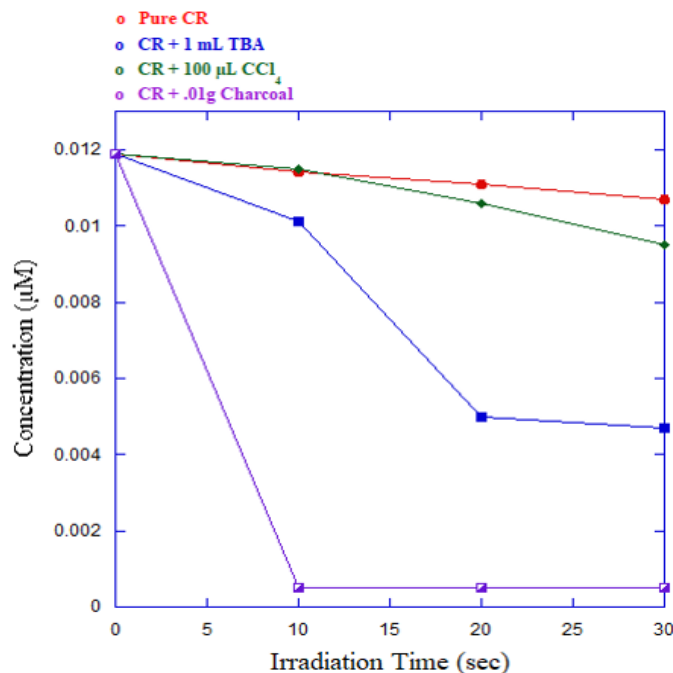


Figure 8: Microwave degradation of CR in the presence of TBA, CCl<sub>4</sub> and charcoal.

### 3.2.4 Effect of Combined Additives (CA)

A 100 μL CCl<sub>4</sub>, 1 ml TBA and 0.01g charcoal were combined together with CR during the microwave irradiation process to see the effect of additives when combined. From Fig. 9, 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 occurred. This may be due to the combined scavenge effect of TBA and CCl<sub>4</sub>.

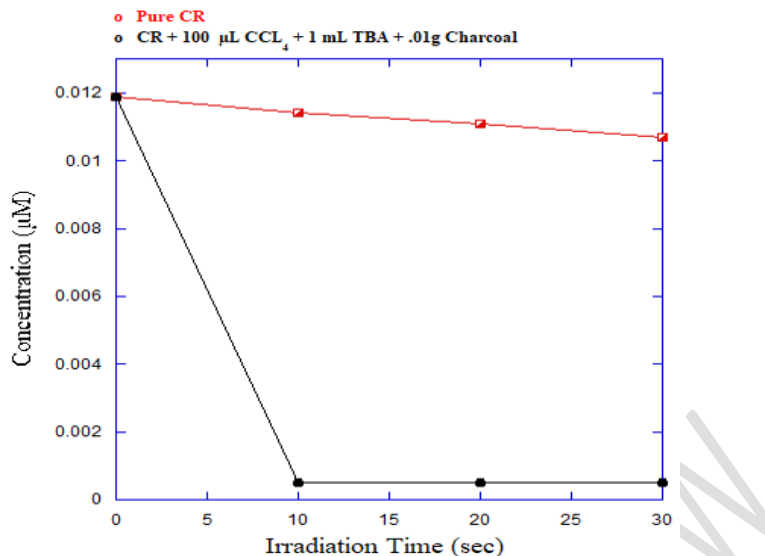


Figure 9: Microwave degradation of CR in the presence of TBA, CCl<sub>4</sub> and charcoal

#### 4. CONCLUSION

From the above discussion, it can be observed that all the additives highly increased the degradation rate of the mentioned azo dyes MO and CR as compared to their raw solution. Some of the additives have been showing an excellent degradation rate at the beginning with low concentration and after that, the degradation rate started to decrease with the increased concentration and irradiation time. For MO, it can be concluded that the consumption of charcoal was the most beneficial though TBA failed to show any satisfactory progress also TBA is a radical scavenger. The order of effectiveness for MO of irradiation was 0.01g charcoal > 20 ml NaCl > 20 ml Na<sub>2</sub>SO<sub>4</sub> > 20 ml glucose > 10 ml sucrose > 100 µL CCl<sub>4</sub> > 200 µL H<sub>2</sub>O<sub>2</sub> > pure MO > TBA. On the other hand, the order of effectiveness for CR for the same condition of irradiation was combined additive > 0.01g charcoal > TBA > 100 µL CCl<sub>4</sub> > pure CR. In both cases, charcoal has shown the greatest enhancement in the degradation rate. Also our experimental observation combined additives such as charcoal with CCl<sub>4</sub> and TBA may be considered for degradation.

**Ethical Approval** This article does not contain any studies with human participants or animals performed by any of the authors.

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