Degradation of Dyestuffs from Industrial Effluents using an Ultrasonic Bath in the Presence and Absence of Various Inorganic Salts and H₂O₂

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Original Research Article

ABSTRACT

Textile industries consume various dyestuffs for dyeing of fabrics. These dyestuffs are often present in industrial wastewater, have a serious impact on the environment and personal health. This report describes a system developed to degrade the dyestuffs using sonochemical reactions. The degradation of dye molecules in aqueous solution was investigated using 50-60 Hz ultrasonic bath, and all solutions volumes were 50mL where the concentration of dye solution was 50mg/L. Experiments were performed at an initial concentration of two types of dye solutions, *i.e.* Methylene Blue (MB) and Methyl Orange (MO) in absence and presence of inorganic salts (NaCl, Na₂CO₃, Na₂SO₄, NaNO₃) and catalyst, *i.e.*H₂O₂ at room temperature. This research investigated the degradation of dye molecules with an aim of cost-effective method. In sonochemical degradation of dyestuffs in sonicator, high degradation rate for MB was obtained in the presence of additives H₂O₂ and followed the order: MB+H₂O₂> MB+ Na₂CO₃ > MB+Na₂SO₄> MB + NaCl> MB (0 mM salt & additive) > MB+NaNO₃. Whereas, in case of MO, high rate was obtained in the presence of Na₂SO₄ and then followed the order: MO+Na₂SO₄> MO + NaNO₃> MO+H₂O₂> MO + NaCl> MO (0 mM salt & additive) > MO+Na₂CO₃.

Keywords: Methyl orange; methylene blue; sonolysis; inorganic salts; H₂O₂.

1. INTRODUCTION

The organic dyes widely used in various industries like textile processing, paper making, printing and cosmetic industries etc. [1, 2]. Often

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produce significant amount of toxic coloured wastewater, which has led to severe pollution of surface water and groundwater, inducing severe threat to the natural ecosystem [3–6]. Several industries use the synthetic dyestuffs extensively, and it is estimated that 10–15% of the dyestuffs remain in the effluent during the dyeing processes. Some azo dyestuffs and their reaction products also have the carcinogenic effects. Thus, the treatment of these wastewaters containing dyestuffs is important for the protection of waters and the environment [7].The treatment of dye-containing wastewater by conventional methods such as biological treatment [8-9], coagulation [10-11], filtration [12] and adsorption [13–15], solvent extraction, incineration, chemical dehalogenation, biodegradation, flocculation, air sparging and are quite ineffective for the decolorization of wastewater since it is very difficult to remove the dyestuffs using these processes because of their low molecular weight and high-water solubility [16]. Other processes, such as coagulation and adsorption, hardly transfer the pollutant from the wastewater to other media that produce sludge and cause disposal problems [17]. In the past few years, advanced oxidation processes (AOPs), as the most promising and highly competitive innovative wastewater treatment methods for the removal of organic pollutants, have been widely developed [18–20]. AOPs are based on the physicochemical process that creates reactive oxygen species (ROS) at ambient temperature and pressure with or without catalyst and subsequently converts organic pollutants into CO_2 and H_2O without the production of any types of secondary wastes [21-22]. Among them, ultrasonic degradation of dyestuffs was found to be a very useful method for the removal of organic compounds [23-24]. The ultrasonic irradiation of liquids generates cavitation bubbles which grow during the compression cycles until they reach a maximum size, which is known as critical size and implodes generating heat and highly reactive radical species. Inside cavitation bubbles, the temperature and pressure rise to the order of 5000 K and 100 MPa, respectively. For such conditions, water molecules degrade releasing OH• and H• radicals [Equations. (1) – (8)].

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

$$H\bullet + OH\bullet \rightarrow H_2O \quad (2)$$

$$2H\bullet \rightarrow H_2 \quad (3)$$

$$2OH\bullet \rightarrow H_2O_2 \quad (4)$$

$$2OH\bullet \rightarrow O\bullet + H_2O \quad (5)$$

$$2O\bullet \rightarrow O_2 \quad (6)$$

$$O\bullet + 2H\bullet \rightarrow H_2O \quad (7)$$

$$O\bullet + H_2O \rightarrow H_2O_2 \quad (8)$$
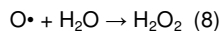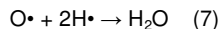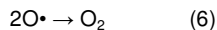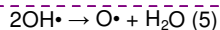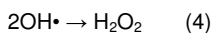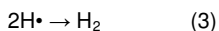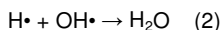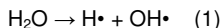
Inside the bubble or in the liquid shell surrounding the cavity, these radicals can combine in various ways or react with the gases and vapor present, leading to the detection of H_2O_2, HNO_2 and HNO_3 [25] in the medium as shown by Equations. (9) – (14).

$$N_2 \rightarrow 2N\bullet \quad (9)$$

$$O_2 \rightarrow 2O\bullet \quad (10)$$

$$N\bullet + O\bullet \rightarrow NO \quad (11)$$

$$NO + O\bullet \rightarrow NO_2 \quad (12)$$

$$OH\bullet + NO \rightarrow HNO_2 \quad (13)$$

$$OH\bullet + NO_2 \rightarrow HNO_3 \quad (14)$$
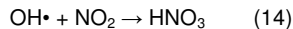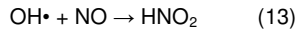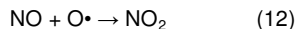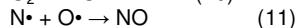
These radicals can oxidize the solute occasioning its degradation. A higher ultrasonic frequency would be more suitable for dyestuff degradation. The present study verifies the influence of the presence of salts (NaCl, $NaNO_3$, Na_2CO_3, Na_2SO_4) and additives (H_2O_2) on dyestuff degradation processes. Frequencies in the range of 50-60 Hz and power inputs of 320 W were tested in order to optimize the degradation conditions for Methylene Blue (basic dye) and Methyl Orange (azo dye) solutions. Ultrasonic power is one of the important factors for sonochemistry. Calorimetry is another one of the most frequently used methods for measuring the actual ultrasonic power input for a solution [26,27].Eq. (15) gives the calculated ultrasonic output power (P).

$$P = CpM(dT/dt) \quad (15)$$

Here,
C_p = heat capacity of water
M = mass of water
T = temperature of the sample solution
t = ultrasonic irradiation time.

The effects of sonic waves and wave properties on chemical systems are discussed under the study of sonochemistry. Ultrasound causing the chemical effects does not directly interact with molecular species. Studies have shown that sonochemistry or sonoluminescence cannot

consider any direct contact of the acoustic field with chemical species. Instead, sonochemistry is related to acoustic cavitation: the formation, growth, and implosive collapse of bubbles in a liquid. The growth of cavitation bubble mainly depends on the sound intensity. The formation and growth of the cavitation bubble are shown in (Fig.1). Sometimes the cavity expansion occurs very quickly by high-intensity ultrasound during the negative-pressure cycle so that the positive pressure in the next cycle fails to reduce the size of the cavity. For low intensity ultrasound, the size of the cavity oscillates in phase with the expansion and compression cycles and in this case the bubble surface area is comparatively larger during expansion cycles than during compression cycles. There is a critical size of cavitation bubble which depends on the ultrasound frequency and in this situation the cavity can absorb energy more efficiently from ultrasound and grow more rapidly. But when a cavity experiences a very rapid growth it can no longer absorb energy as efficiently from the sound waves and thus the liquid rushes in and the cavity implodes. Gas and vapors are compressed inside the cavity and this generates heat, which finally produces a short-lived localized hot spot (Fig. 2).

In brief, the cavitational collapse creates an unusual environment for a chemical reaction in terms of enormous local temperatures and pressures. Again, the most sophisticated models predict temperatures of thousands of degrees Celsius, pressures of from hundreds to thousands of atmospheres and heating times of less than a microsecond. Various experiments were done for sonochemical degradation of organic compounds in the presence of various chemical additives. The results are illustrated in Table 1:
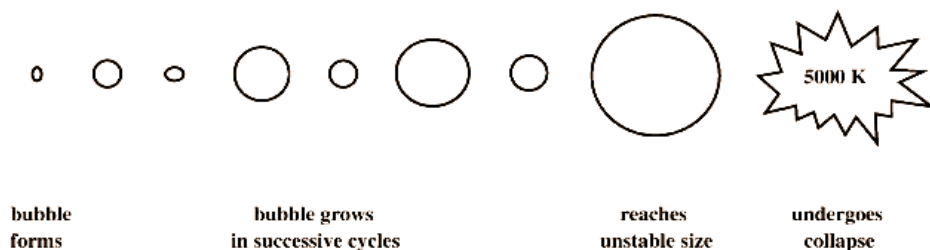
Fig. 1. Cavitation bubble formation and collapse
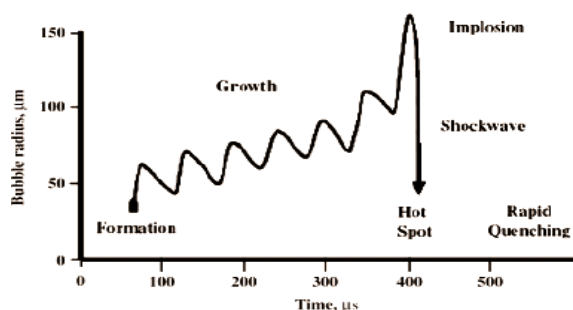
Fig. 2. Graphical representation of bubble growth and implosion (Suslick et al. 1989)

Table 1.Sonochemical degradation of various organic compounds in the presence of various chemical additives

No.	Organic pollutants	Additives	Reaction conditions	Important findings	References
1.	Acid Blue 25 (AB25)	$FeSO_4$, NaCl, $CaCl_2$ and$NaHCO_3$	100 mL of 50mg/L AB25; 1700kHz, 14W	The rate of degradation was increased with the addition of 10mg/L of Fe^{2+} and 386mg/L of H_2O_2.	[28]

| No.	Dye	Reagents	Conditions	Observations	Ref.
2.	Rhodamine B (RHB)	Iron (Fe^0 Fe^{2+}, Fe^{3+}), CCl$_4$, H$_2$O$_2$,t-butanol, Na$_2$SO$_4$, Sucrose and Glucose	300mL of 5mg/L RHB; 300kHz,60W	The rate of degadation was also increased in the presence of salts. The rate of degradation was increased with the addition of 5mg/L of Fe^{2+}, followed by Fe^{3+} and Fe^0. The rate of degradation was also increased by the addition of 50, 100 and 200mg/L CCl$_4$. The rate of degradation was also increased with the addition of 100mg/L of H$_2$O$_2$ and 2g/L of Na$_2$SO$_4$ and decreased with the addition of tert-butanol and sucrose and glucose.	[29]
3.	Malachite Green (MG)	Ethanol, 2-propanol and iso-butanol	1000mL of 5 mg/L MG; 35kHz and 49 W	The rate of degradation was decreased by using iso-butanol followed by 2-propanol and ethanol. It's a pseudo first-order reaction kinetics.	[30]
4.	C.I. Reactive Black 5 (RB5)	O$_3$	500mL of 500mg/L RB5; 20kHz, 88w/L	Decolorized by using the solution of US/O$_3$ within 10mins. It is a pseudo first-order reaction kinetics.	[31]
5.	Reactive red (RR)/Direct Yellow (DY)	CCl$_4$, tert-butanol, TiO$_2$, zero valent Cu (Cu^0)	100mL of 28.75μM RR/DY; 861kHz, 0.23 and 0.46 W/mL	The rate of degradation was increased in the presence of 2000 μM of CCl$_4$. The rate of degradation increases especially in the presence of 0.1-1.0g/L TiO$_2$ followed by CCl$_4$ and Cu^0	[32]

2. Experimental Section

The chemicals used for the experiment are:

i) Methylene Blue (Dye content ≥ 82%; Merck KGaA, Germany) IUPAC name 7-(dimethylamino) phenothiazin-3-ylidene]-dimethyl azanium chloride.

Methylene blue (or MB) is a basic aniline dye with the molecular formula $C_{16}H_{18}N_3SCl$. At room temperature, it appears as a solid, odorless, dark green powder that yields a blue solution when dissolved in water.
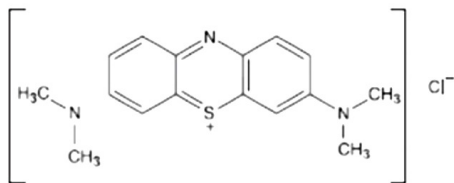
$C_{16}H_{18}C_lN_3S$ $MW = 319.86$

Fig. 3. Structure of methylene blue

ii) Methyl Orange (Merck KGaA, Germany) IUPAC name sodium;4- [[4- (dimethyl amino) phenyl] diazinyl] benzene sulfonate
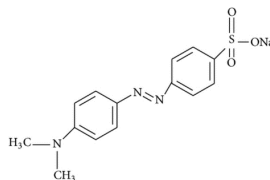
Fig. 4. Structure of methyl orange

Methyl orange is a pH indicator and due to its clear color change, it is very often used in titrations. Methyl orange changes colour at the pH of a mid-strength acid and is usually used in titrations for acids. Unlike a so called universal indicator, methyl orange does not have a full spectrum of colour change but has a sharper end point. Methyl orange is prepared from sulfanilic acid and N, N-dimethylaniline. The first product obtained from the coupling is the bright red acid form of methyl orange, called helianthin. In the base, helianthin is converted to the orange sodium salt, called methyl orange.

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iii) Various Additives like:

a) Salts (NaCl, NaNO$_3$, Na$_2$SO$_4$, Na$_2$CO$_3$)
b) Additive (H$_2$O$_2$)

all were purchased from Merck KGaA 64271 Damstdt, Germany.
The Apparatus used are:

i. Beaker 500mL ii. 250 mL conical flask iii. Pipette iv. Micropipette
v. Digital Thermometer vi. Sample tube vii.Stand viii.Magnetic stirrer
ix. Measuring cylinder

The equipments used are:

i. Sonicator Machine (Soniclean 250HT; 50-60Hz; 320W)

ii. UV-Vis Spectrophotometer (UV Mini 1240; Shimadzu Company)
Procedure

The schematic diagram of the experimental setup to shown in the Fig. 5. First of all, the power of the sonicator is measured calorimetrically using equation-15. Then, a conical flask with a total volume of 100 ml of 50 mg/L dye solution was used for ultrasonic irradiation under air at room temperature around 24^{0}C. The sonicated solution was extracted every (0, 3, 7 and 10 min) by a glass syringe (1 ml) without exposing the sample to air. The glass vessel was flat bottomed and mounted at a constant position (4.0 mm from the oscillator). The sonicated solutions were analyzed by a UV-Vis spectrophotometer (Shimadzu UV-1650).

Fig. 5. Schematic diagram of the experimental set-up

3. RESULTS AND DISCUSSION

Fig. 6. The measurement of temperature change with increasing sonication time

3.1 Calorimetric Power Measurement

From the Table 2 and Fig. 6, Show that the temperature at the site of the reaction is increasing with sonication time. Again, the reaction rate increases with temperature. So, we can say that the degradation rate of the dye stuff will increase with the elongation of sonication time.

3.2 Degradation of Dyes

In case of Methylene Blue (MB) as it can be seen from Fig. 7, the concentration of MB decreased gradually as the sonication time increased. So, it can be said that MB degrades gradually with the increase in sonication time. When no sonication occurred the concentrations of MB were 0.1563 mM. After sonicating for 3mins, 7mins and 10mins, the concentration became 0.1524 mM, 0.1446mM and 0.1403mM respectively. With the addition of NaCl, degradation of MB was slightly enhanced up to 0.0014mM when compared with the absence of NaCl salt. The addition of NaNO$_3$ shows a higher degradation property at an initial state, but finally, it fails to keep the degradation rate higher than that of with the absence of NaNO$_3$ salt. The addition of Na$_2$CO$_3$ enhanced the degradation of MB up to 0.0053mM whereas

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Na$_2$SO$_4$ slightly enhanced the degradation of MB up to 0.0027mM. The positive effects of these inorganic salts on the destruction of MB were very little at an initial state. But finally, it has a great impact on the degradation except for NaNO$_3$.

Again, in the case of Methyl Orange (MO) from Fig. 8, the concentration of MO decreased gradually with the increase in sonication time. When no sonication occurred the concentrations of MO was 0.1528 mM. After sonicating for 3 mins, 7 mins and 10 mins, the concentration became 0.1518 mM, 0.1516 mM and 0.1496 mM respectively. The addition of NaCl, NaNO$_3$, Na$_2$CO$_3$, Na$_2$SO$_4$ enhanced the degradation of MB up to 0.0038 mM, 0.0175 mM, 0.1496 mM and 0.0209 mM respectively when compared with the absence of inorganic salts. The positive effects of these inorganic salts on the destruction of MO were very little at an initial state. But finally, it has a great impact on the degradation except Na$_2$CO$_3$. So, there may be an optimum amount of these inorganic salts to increase the degradation rate. An excessive amount of salts may interrupt the introduction of ultrasound into the liquid [33]. Addition of salts to aqueous solution pushes molecules from the bulk aqueous phase to the bulk–bubble interface. The presence of salt may increase the hydrophilicity, the surface tension and ionic strength of the aqueous phase and decrease the vapor pressure [34-35]. All these factors help in collapsing of the bubbles more violently, resulting in high degradation of dye.

In order to evaluate the effect of H$_2$O$_2$ alone on the degradation of dyes, control experiments were carried out by adding H$_2$O$_2$. The dosage of added hydrogen peroxide was the same as the amounts used in the presence of ultrasound. The results indicate that a similar dosage of H$_2$O$_2$ in absence of ultrasound produce low degradation of dye molecules, even after long times (240 min). Thus, hydrogen peroxide alone has a very limited effect on the degradation rate of dye molecules. Throughout the addition of hydrogen peroxide in the presence of ultrasound enhances the degradation rate of both MB and MO due to additional free radicals generation. It can be concluded that the controlling mechanism of dye molecule degradation is the free radical attack. The degradation rate was appreciably increased by the addition of H$_2$O$_2$. The H$_2$O$_2$ may be mainly present not inside the cavitation bubbles but in the bulk solution due to the high-water solubility and the low volatility. The lower intensification of degradation rate in the presence of high concentrations of H$_2$O$_2$ may be attributed to the increased level of OH• radical scavenging by H$_2$O$_2$ itself (reaction (16)).

$$H_2O_2 + OH• \rightarrow H_2O + HOO• \qquad (16)$$

Similar results for the enhancement of sonolytic degradation of dyes in the presence of hydrogen peroxide were reported in literature [36-37].

Table 2. Measurement of temperature changes

Observation	Sonication Time(sec)	Initial Temperature(^{0}C)	Final Temperature(^{0}C)	Temperature Change(^{0}C)	Actual power(Watt)
1	30	24.7	25.0	0.3	4.180
2	60	25.2	25.6	0.4	2.787
3	90	25.6	26.1	0.5	2.322
4	120	26.0	26.6	0.6	2.090
5	150	26.4	27.1	0.7	1.951
6	180	26.7	27.6	0.8	1.858

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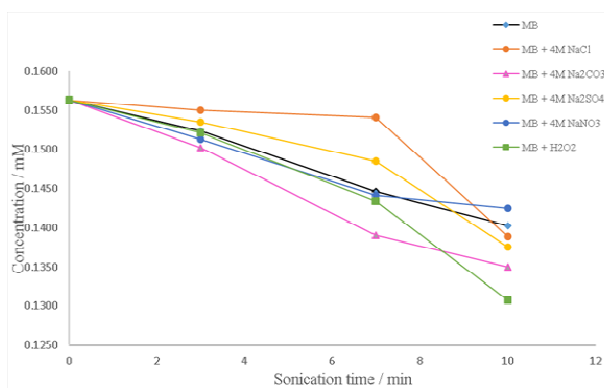
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Fig. 7. Comparison of degradation of MB with and without salt and additive
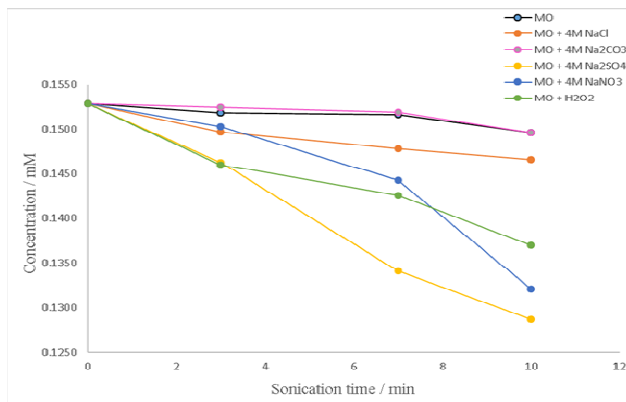

Fig. 8.Comparison of degradation of MO with and without salt and additive

Finally, from the Figs. 7 and 8 it can be stated that, in sonochemical degradation of dyestuffs in sonicator, high degradation rate for MB was obtained in the presence of additives H_2O_2 and followed the order: $MB+H_2O_2> MB+ Na_2CO_3 > MB+Na_2SO_4> MB + NaCl> MB$ (0 mM salt & additive) > $MB+NaNO_3$. Whereas, in case of MO, high rate was obtained in the presence of Na_2SO_4 and then followed the order: $MO+Na_2SO_4> MO + NaNO_3> MO+H_2O_2> MO + NaCl> MO$ (0 mM salt & additive) > $MO+Na_2CO_3$. But in both cases, it can be seen that H_2O_2 has a great impact on degradation of dye molecules, due to formation OH• radicals. So, H_2O_2 can be used as an additive effectively. He et al. 2016 [38] investigated the sonochemical degradation of MO in the presence of Bi_2WO_6 under ultrasonic irradiation is related to the generation of reactive oxygen species (ROS), in which both singlet molecular oxygen ($^1O_2$) and hydroxyl radical (• OH) play important roles in the sonochemical degradation of MO. Our research has also supported that results.

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4. CONCLUSION

The present work has shown that Dye can be effectively removed from water by ultrasound irradiation. This study demonstrates that sonochemical degradation occurs mainly through reactions with hydroxyl radicals. It was found that the initial degradation of dye molecule increased. The addition of salt and additives enhanced the rate of degradation. So, it can be concluded after this research work that, the dye stuffs can be removed from water by using the ultrasound system in presence of salt and additives effectively.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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