A Review on SonochemicalTreatment of Dye Molecules

Abstract

The use of ultrasound is a very effective and suitable process for the degradation of harmful dyes. In order to elucidate the degradation mechanism, numerous studies have examined the breakdown of dye molecules in aqueous medium with different additives using ultrasonic irradiation. In this review, the effect of different additives like NaCl, Na_2SO_4, CCl_4, H_2O_2,ZnO nanoparticle etc. on sonolyticdegradation of the organic dyes is discussed. From the studies it is concluded that the acidic condition is favorable for most of the dye degradation. Assessing the results of earlier studies we have also found that the aforementioned additives aided the degradation of dye molecules depending on their amount used in sonication with relatively higher or lower extent. Therefore, dye molecule from environment especially from waste water can be effectively reduced by usingsonochemical irradiationprocess.

Keywords: Organic Dyes, SonochemicalTreatment, Additives, Degradation.

1. Introduction: Organic dyes finds their application in a wide range of industries, such as solar cells, food, cosmetics, medicines, optical data discs, and traditional textile manufacture, which frequently generates copious amounts of colored effluent. In order to maintain clean and safe water supplies, it is imperative to discover a way to extract and discard the organic dye ingredients used on a daily basis [1, 2]. Although there are many different kinds of dyes, azo dyes account for 70% of all dye manufacturing worldwide [3].The azo bond chromophore group (-N=N-) associated with aromatic structures including functional groups like $-NH_2$ and $-SO_3H$ characterizes these compounds [4,5]. These dyes are widely utilized in the food, textile, paper, leather, and pharmaceutical industries due to their superior characteristics and great cost effectiveness [6]. The impact of these dye compounds on human health are varied. When azo dyes and their intermediates break down, they can have harmful, mutagenic, and carcinogenic consequences that pose a serious risk to human health [7]. Therefore, it is imperative that wastewater be cleaned of these dangerous color particles. Many physical or chemical techniques, such as membrane filtering [8,9], electrochemical technology [10,11], and advanced oxidation processes (AOPs) like photochemical oxidation [12], photocatalysis [13,14], and ultrasonic wave [15,16], constitute the foundation for the various dye decolorization procedures. A method that effectively removes color chemicals from wastewater involves combining two or more separate procedures. Ultrasound/H_2O_2 and ultrasound/ozone are a couple of these [17, 18]. UV light/ozone or UV light/H_2O_2 [19,20], photocatalysis in sono Fenton, electro-Fenton[21, 22], and photo-Fenton processes [23, 24–25], as well as electrochemical and photocatalytic oxidation processes [26–27, 28].The possibility of fully mineralizing organic contaminants to CO_2 and H_2O has raised interest in advanced oxidation processes (AOPs). Ultrasonography has lately been used in AOPs for wastewater remediation [29-31]. The two main principles of sonochemical reactions are the pyrolysis events that take place at and inside hot regions during collapsing bubbles and the radical reactions by OH and H radicals [32] that are produced by the pyrolysis of water.

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

It has been discovered that one of the effective methods for degrading organic contaminants is sonochemical treatment. Sound waves with frequencies higher than those to which the human ear can respond are referred to as ultrasounds (N16 kHz). Ultrasound can be used in two general categories: low frequency ultrasound (20–100 kHz) and high frequency ultrasound (2–10 MHz) [33]. Although Alfred L. Loonis first identified the action of ultrasound in liquid media when 1927, significant advancements in sonochemistry occurred with the introduction of low-cost, high-intensity ultrasound generators in the 1980s [34]. Low frequency or power ultrasound is the primary ultrasound used in sonochemical studies [35]. According to Mason and Lorimer [33], the main uses of ultrasonography in recent years have been in organic, inorganic, and organometallic synthesis, polymer chemistry (including copolymerization, initiation, and degradation), and certain areas of catalysis. The use of ultrasonic technology to eliminate organic pollutants found in water and/or wastewater has gained popularity since 1990 [36]. Sonochemical degradation offers important benefits like safety, cleanliness, and energy conservation without causing secondary pollution when compared to other organic pollutant treatment technologies like UV light, high voltage corona, incineration, and solvent extraction [37, 36].Many research works have looked at the sonochemical breakdown of dyes and phenolic compounds [38 – 45]. This review focuses on the basic steps involved in sonochemical treatment and how sonochemistry is applied to the degradation of organic dyes in the presence of various additives.

2.Fundamentals of sonochemical treatment

The main principle of sonochemistry is acoustic cavitation, which is the emergence, expansion, and implosive collapse of bubbles in a liquid. By cavitation, the diffuse energy of sound is amplified [46]. According to Suslick [34], a liquid can experience both positive and negative pressure due to the compression and expansion cycles of ultrasonic waves. The average distance between molecules in a liquid would exceed the critical molecular distance required to keep the liquid intact when a large enough negative pressure is applied; as a result, the liquid would break down and voids or cavities would form, leading to the formation of cavitation bubbles [33]. Negative pressure depends on the kind and purity of the liquid since it acts against the liquid's tensile strength. While only a few atmospheres of negative pressure are needed to create bubbles in tap water, more than 1000 atm of negative pressure is needed for cavitation in pure water [34]. Once created, these gaps, cavities, or bubbles may continue to expand until the maximum negative pressure is achieved [33]. Fig. 1 depicts the cavitation bubble's creation and development. The cavitation bubble grows mostly in response to sound intensity. Occasionally, high-intensity ultrasound during the negative-pressure cycle causes the cavity to expand so quickly that the positive pressure in the following cycle is unable to stop the cavity's growth. In the case of low intensity ultrasound, the bubble surface area is relatively bigger during expansion cycles than during compression cycles because the cavity's size oscillates in phase with the expansion and compression cycles. The cavitation bubble has a critical size that is dependent on the ultrasonic frequency; at this size, the cavity can absorb ultrasonic energy more effectively and develop more quickly. However, a cavity that grows extremely quickly loses its ability to absorb sound waves' energy; as a result, liquid rushes in and the cavity implode [34]. Inside the hollow, vapors and gas are compressed, creating heat and, in the end, a transient localized hot spot (Fig. 2).In summary, huge local temperatures and pressures are produced by the cavitational collapse, which makes the chemical reaction environment peculiar [46]. The most advanced models forecast heating durations of less than a microsecond, pressures ranging from hundreds to thousands of atmospheres, and temperatures of thousands of degrees Celsius [34].

A sonochemical circumstance can be explained by one of three widely accepted theories: the hot-spot theory, the electrical theory, or the plasma discharge theory. The brief duration of the localized "hot spot" produced by the swift collapse of acoustic cavities (b10 μs) suggests the presence of exceptionally high rates of heating and cooling within the vicinity of 1010 K/s [47]. It is estimated that the bubble's interior temperature is close to 5000 K. It is possible to obtain light output of over 107 photons per flash because to the high temperature [48, 49]. Hot-spot theory states that blackbody radiation from gas inside the bubble during adiabatic collapse is essentially what causes sonoluminescence. Extensive experimental evidence was presented by Marguls [50] to support an electrical explanation of cavitation. He claims that during bubble creation and collapse, significant electrical field gradients are created, which trigger sonochemical processes and sonoluminescence. Lepoint and Mullie [51] proposed plasma theory to explain cavitation and noted similarities between coronachemistry and sonochemistry. Instead of thinking it was a real implosion, they thought it was a process of fragmentation brought on by a strong electrical field. In the field of environmental science, the hot-spot theory is the most frequently recognized theory to explain sonochemical processes [29].

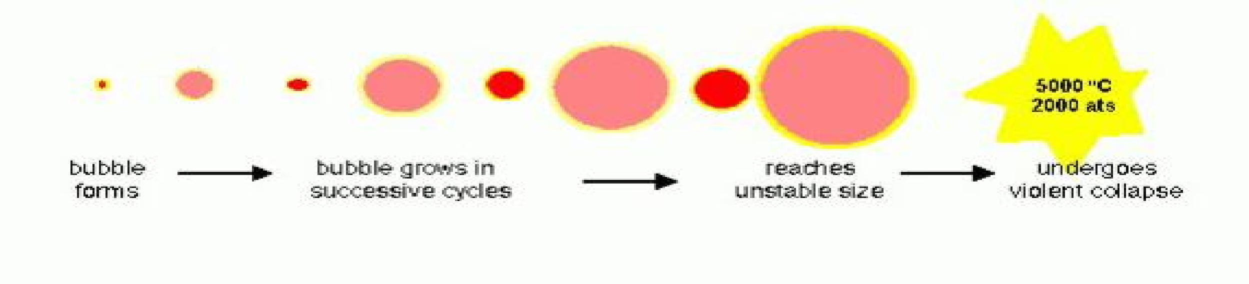

Fig. 1: Formation and collapse of cavitation bubble.

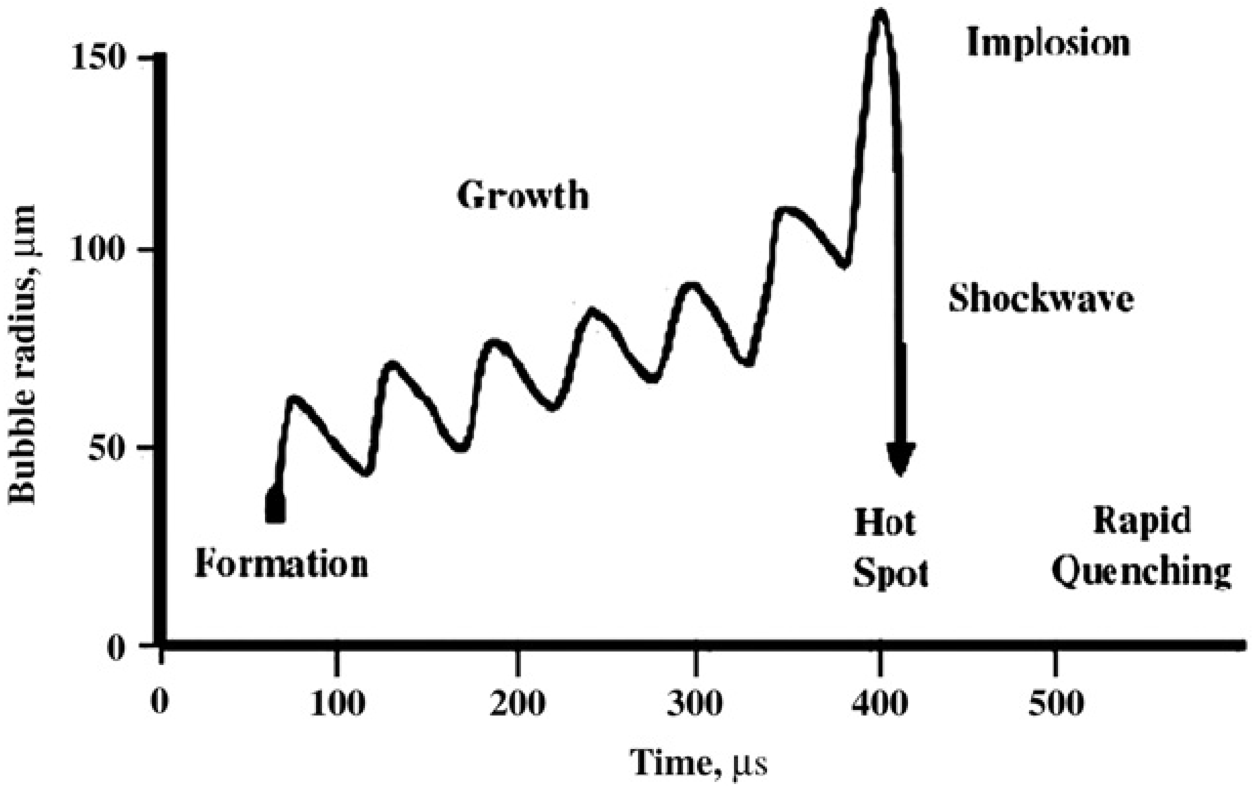

Fig. 2: Bubble growth andimplosion in sonicator [34,52].

There are numerous ways in which sonochemical reactions are different from chemical reactions. According to Suslick [34], ultrasonic energy affects chemical reactions by generating reactive free radicals or generating a great deal of heat (pyrolysis). Once more, turbulence caused by ultrasonic waves accelerates the mass transfer rate in an aqueous solution [53, 54]. According to Figure 3, there are primarily three reaction sites: bulk liquid, gas–liquid interface, and hollow interior. Water molecules are pyrolyzed inside the cavitation bubble, producing ·OH and ·H radicals in the gas phase. The substrate either experiences pyrolysis or a reaction with the hydroxyl radical. An aqueous phase of a comparable reaction takes place at the interfacial area. The recombination of ·OH radicals to generate H_2O_2 is the extra reaction. The bulk phase reactions primarily involve the substrate and the H_2O_2 or ·OH radical. According to Adewuyi, homogeneous sonochemistry takes into account all of these processes. While hydrophilic substances react at the bulk phase, the majority of hydrophobic compounds react inside the cavitation bubble [29, 55]. While the heterogeneous systems adhere to the same physical mechanism, they differ in terms of high-speed liquid jet, cavitational threshold, and other factors [55].

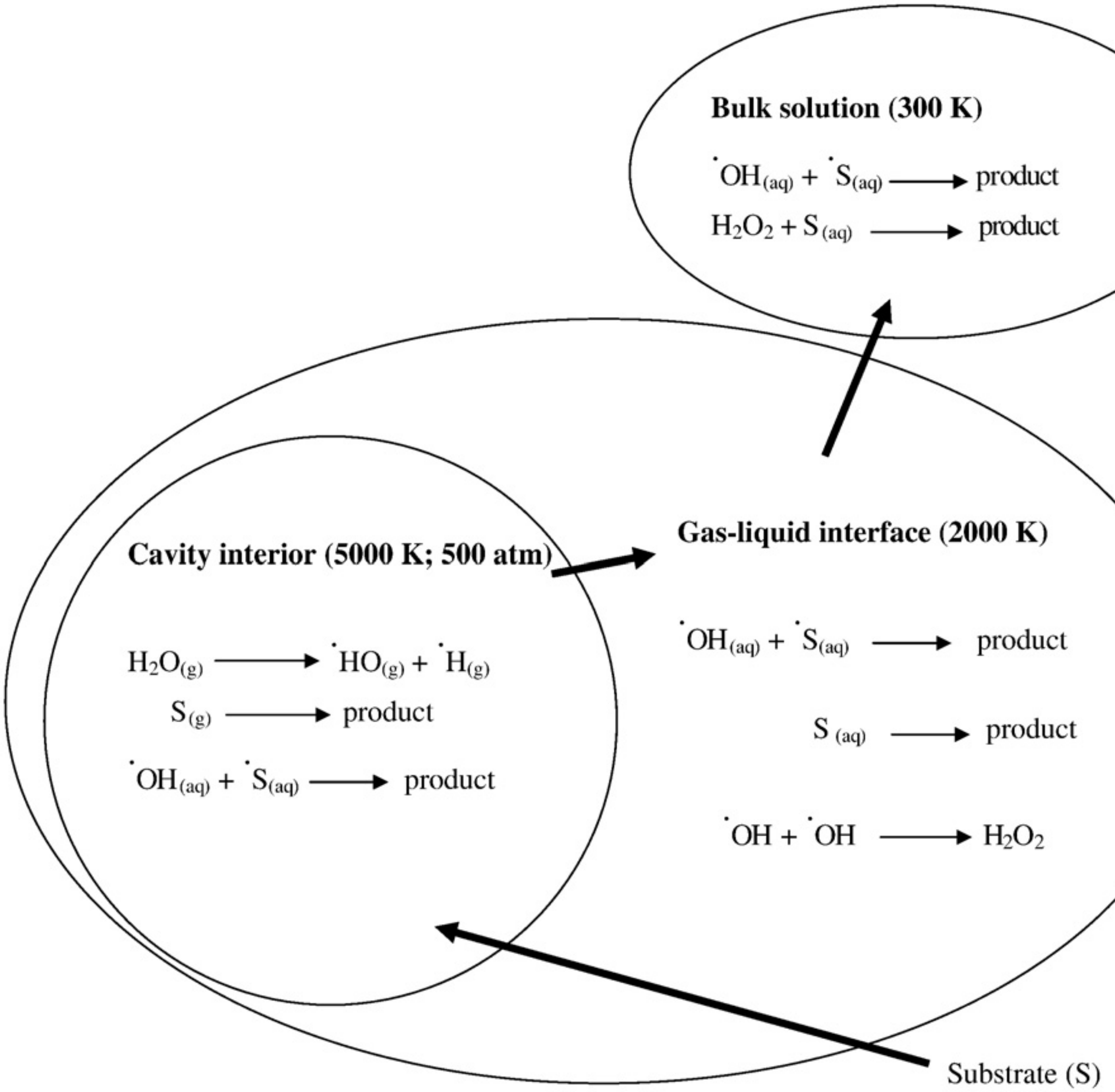

Fig. 3: Reaction zones in sonolysis[29,52]

In order to track various oxidative species (H_2O_2 and ·OH radicals) during the ultrasonic degradation of reactive dye, RB 5, Vajnhandl and Marechal [56] employed an iodine dosimeter and a Fricke. When comparing the plate type system to the probe type system at the same acoustic power, the radical production rate was 20–25 times greater. A kinetic model based on a heterogeneous reaction system was presented by Okitsu et al. [57] for the ultrasonic degradation of azo dye. Their concept for heterogeneous gas–solid catalytic reactions was comparable to the Eley–Rideal mechanism or the Langmuir–Hinshelwood mechanism. They guessed the existence of local reaction sites at the cavitation bubble interface region, where the extraordinarily high concentration of ·OH radicals caused the oxidative degradation of azo dyes to occur very quickly. They also suggested that their model may be used to different sonochemical reactions that take place in an aqueous media.According to Inoue et al. [58], Rhodamine B degrades more quickly than Orange II. As input power grew, so did the rate constant. In essence, .OH radical reactions were used to describe the degradation mechanism. While degradation was successful at high frequencies, the rate of ·OH radical generation and degradation was slower at lower frequencies.

3Effect of pH on the degradation

Several researchers have demonstrated that the sonochemical degradation rate falls with increasing solution pH [59–61]. If the pH is lower and there is a finite vapor pressure, then more of the neutral form can enter the cavitation bubble. It may at least stay at the cavitation-water interface because it is basically hydrophobic. However, the ionic form prefers to remain in water because the electric charge polarizes solvent molecules and lowers their energy. Ionic forms that stay away from the cavitation are more resilient to hydroxyl radical attack [62]. The multi-bubble sonoluminescence of organic acids also demonstrates the interplay between the neutral and ionic forms [63]. The neutral form has the ability to penetrate the cavitation bubble at very low pH values, hence inhibiting the sonoluminescence. On the other hand, Kidak et al. [64] discovered that at 300 kHz, pH 2 produced higher rates of phenol degradation than pH 10.Alkaline pH decreased the availability of ·OH radicals in the system, which in turn decreased the breakdown of the dye by creating ·H. [56].

$-SO_3H \rightarrow -SO_3^- + {}^{\cdot}H$ (2)

$-NH_3^+ \rightarrow -NH_2 + {}^{\cdot}H$ (3)

$Ar\text{-}OH \rightarrow Ar\text{-}O^- + {}^{\cdot}H$ (4)

${}^{\cdot}OH + {}^{\cdot}H \rightarrow H_2O$ (5)

Shaharul et al showedthat degradation rate of methylene blue at pH 3 is higher than that of the pH 7 and pH 8 [65]. There is a high chance that the sonochemical degradation process will degrade methylene blue, methyl orange and congo red more quickly when additives are used in an acidic environment.[65 - 68]

4.Effect CCl_4on sonolyticdegradation of dyes

A great deal of effort has gone into developing effective additives that hasten the breakdown of organic compounds. The effects of adding CCl_4 or C_6F_{14} on sonochemical degradation have been examined in recent investigations [45, 69–70]. Despite being one of the more hazardous chemicals, CCl_4 is used with extreme caution and is a useful addition for accelerating the sonochemical degradation of organic molecules. Sponza et al. [69] reported that the presence of 19 $mgL^{-1}C_6F_{14}$ enhanced the rates at which phenol was eliminated from the olive mill's wastewater. In the presence of 150 M CCl_4 and 1.5 M C_6F_{14}, respectively, Zeng et al. [70] observed that the sonochemical degradation of phenol rose from 0.014 to 0.031 min^{-1} and from 0.014 to 0.032 min^{-1}. Based on research [69, 70], these advantageous effects are attributed to the H atom scavenger properties of CCl_4 or C_6F_{14}.

The rate of methylene blue degradation is36% (table 1) in presence of 400 μL CCl_4at 25 minutes[65]. According to Monira et al. Congo red and methyl orange can beremoved with 100 μL $CCl_4$by sonolysis[66]. Despite suggestions from some researchers that CCl_4 could function as an H atom scavenger [70], shaharul et al. Suggested that active Cl radicals or similar species are formed during the sonolysis of CCl_4, and that these radicals or species would be advantageous for the breakdown of metheleneblue[65]. Previous studies [45, 71–74] indicate that the following reactions should take place:

$CCl_4 \rightarrow \bullet CCl_3 + \bullet Cl$ (6)
$\bullet CCl_3 \rightarrow : CCl_2 + \bullet Cl$ (7)
$2 \bullet CCl_3 \rightarrow C_2Cl_6$ (8)
$2 : CCl_2 \rightarrow C_2Cl_4$ (9)
$\bullet Cl + \bullet Cl \rightarrow Cl_2$(10)

In an aqueous solution, the radicals that form that contain chlorine (Cl, CCl_3, CCl_2, Cl_2, etc.), may hasten the breakdown of methylene blue. Merouani et al. [41] reported that the sonochemical breakdown rate of rhodamine B at 300 kHz was 21 times higher in the presence of 200 mg L^{-1} of CCl_4. The addition of 100 ppm CCl_4 increased the sonochemical breakdown rate of methyl orange at 200 kHz by 41 times, according to Okitsu et al. [39]. Despite its high toxicity CCl_4 can be employed as an additive, according to the aforementioned perspectives[75].

5. Effect of H_2O_2on the rate of degradation of dyes

The rate of methylene blue degradation is 26% (table 1) in presence of 200 µL H_2O_2 at 25 minutes [65]. Monira et al. reported that concentration of methyl orange reduced with 400 µL H_2O_2 by sonolysis [66]. The production of free radicals as active species has been used to explain the H_2O_2 reaction process for dye discolouration.

$$H_2O_2 \rightarrow \bullet OH + \bullet OH \qquad (11)$$

The US/UV/ H_2O_2 method, which combines ultrasonic to promote H_2O_2 scission in addition to photolysis, has been shown to be the most successful in breaking down malachite green [76]. In a different recent investigation, the effects of introducing different concentrations of hydrogen peroxide—0.05, 0.10, and 0.15 M—were investigated. The water evaluated showed a 62.9% absorbance removal after the treatment with 0.10 M H_2O_2 and 45 W, which was effective and energetically feasible [77]. As the temperature of the aqueous solution rose, the amount of dissolved gas decreased and the amount of water vapor in the bubbles increased [78], which resulted in a lower rate of OH radical generation. Consequently, the rates of methylene blue degradation decreased as solution temperatures increased. Jiang et al. [78] reported on the sonolysis of 4-chlorophenol in aqueous solution at 20 and 500 kHz as a function of solution temperature. They discovered that the rates of 4-chlorophenol degradation and H_2O_2 production rose with growing solution temperatures at 500 kHz but decreased with rising solution temperatures at 20 kHz in the 10–40 ◦C range. Uddin et al. [79] reported on the sonolysis of bisphenol A in aqueous solution as a function of solution temperatures at 489 kHz. The results demonstrated that rates of degradation within the range of 10–40 ◦C increased as the temperature of the aqueous solution increased. Given its relative safety and convenience of use, hydrogen peroxide (H_2O_2) has been used in the field of water treatment for a wide range of purposes, as evidenced by comparisons with previous studies.

6.Effect of NaCl and Na_2SO_4: It has beenobserved that the addition of 20 ml Na_2SO_4degrade methylene blue by 35% at 25 minutes (table 1)[65]. Another research done by Monira et al. showed that methyl orange can be removed by sonochemical treatment in presence of 20 ml NaCl solution and Na_2SO_4solution(table 1)[66].The decolorization efficiency was shown to be more moderately influenced by Na_2SO_4 than by NaCl in a number of synthetic dye solutions containing mixes of the two, according to earlier investigations. Increased Na_2SO_4 concentrations did not impede the decolorization process and, in fact, enhanced the efficacy of reactive bright red K-2BP in dye solutions containing comparable concentrations of salt or Na^+ [80]. Furthermore, it has been observed that the rate at which colors reduce can be affected by the application of specific chemicals such as NaCl and Na_2SO_4 [65–68]. Furthermore, the increased concentration of salt quickened the rate of deterioration. Uddin et a[81] presented their sonochemical degradation results for 4-chlorophenol, phenol, catechol, and resorcinol under Ar in the absence and presence of Na_2SO_4 or NaCl. A pseudo-first order rate constant was observed by another research in the breakdown rate of phenolic compounds [82].

7. Effect of ZnO nanoparticle: There has been a lot of study done on metal oxide semiconductors' potential as photocatalysts to remove organic pollutants from air and water [83–86]. When metal oxides are exposed to the appropriate photon energy, they become active sites of electron-hole pairs, which raises the catalytic activity on their surfaces [87]. Of all these metal oxides, TiO_2 is thought to be the most optimum material, the oldest and most common because of its outstanding stability and relatively simple fabrication method [88–91].With a rather wide band gap of 3.37 eV and an exciton binding energy of 60 m eV, zinc oxide (ZnO), a metal oxide, has been thoroughly investigated as a photocatalyst for the destruction of several organic contaminants. Furthermore, because of their nontoxicity, affordability, and superior absorption of the solar spectrum, ZnO nanostructures are favored over TiO_2 as photocatalyst substitutes for photodegradation [92]. ZnO nanostructures have shown strong photocatalytic activity for the elimination of organic contaminants, including organic dyes, in a number of investigations [93].Earlier studiesshowed that the concentration of Methylene blue decreased by 42% at 25 minutes (table 1) in presence of ZnO nanoparticle[65]. The highest degradation of methylene blue was found by Maedeh Asgharian et al. [94] when rGO/ZnO/Cu combination was present at a dosage of 25 mg photocatalyst.

Table 1: Degradation efficiency of sonochemical treatment for some dyes

Sonochemical system parameter (Frequency = F, Power = P, Volume = V)	Name of Dyes, Initial Concentration	Medium	Result (Irradiation time)	Reference
F: 489 kHz P: 30 W V: 100ml	Methylene blue, 25 mgL^{-1}	Deionized water with 200 µL H_2O_2	26% degraded(25 min)	[65]
F: 489 kHz P: 30 W V: 100ml	Methylene blue, 25 mgL^{-1}	Deionized water	19% degraded(25 min)	[65]
F: 489 kHz P: 30 W V: 100ml	Methylene blue, 25 mgL^{-1}	Deionized water with 400 µL CCl_4	36% degraded(25 min)	[65]
F: 489 kHz P: 30 W V: 100ml	Methylene blue, 25 mgL^{-1}	Deionized water with 20 ml Na_2SO_4	35% degraded(25 min)	[65]
F: 489 kHz P: 30 W V: 100ml	Methylene blue, 25 mgL^{-1}	Deionized water with ZnO nanoparticle	42% degraded(25 min)	[65]
F: 40 kHz P: 120 w V: 100 ml	Congo red, 200 mgL^{-1}	Deionized water	degraded(15 min)	[66]
F: 40 kHz P: 120 w V: 100 ml	Congo red, 200 mgL^{-1}	Deionized water with tert-butyl alcohol (TBA)	degraded(15 min)	[66]
F: 40 kHz P: 120 w V: 100 ml	Congo red, 200 mgL^{-1}	Deionized water 100 µL CCl_4	degraded(15 min)	[66]
F: 40 kHz P: 120 w V: 100 ml	Congo red, 200 mgL^{-1}	Deionized water with .01 g Charcoal	degraded(15 min)	[66]
F: 40 kHz P: 120 w V: 100 ml	Methyl Orange, 12.5 mgL^{-1}	Deionized water	degraded(15 min)	[66]
F: 40 kHz P: 120 w V: 100 ml	Methyl Orange, 12.5 mgL^{-1}	Deionized water 20 ml Sucrose	degraded(15 min)	[66]

F: 40 kHz P: 120 w V: 100 ml	Methyl Orange, 12.5 mgL^{-1}	Deionized water with 20 ml NaCl	degraded(15 min)	[66]
F: 40 kHz P: 120 w V: 100 ml	Methyl Orange, 12.5 mgL^{-1}	Deionized water with 20 ml Na_2SO_4	degraded(15 min)	[66]
F: 40 kHz P: 120 w V: 100 ml	Methyl Orange, 12.5 mgL^{-1}	Deionized water 100 µL CCl_4	degraded(15 min)	[66]
F: 40 kHz P: 120 w V: 100 ml	Methyl Orange, 12.5 mgL^{-1}	Deionized water 400 µL H_2O_2	degraded(15 min)	[66]
F: 300 kHz P: 30 W V: 300 mL	Rhodamine B, 5 mgL^{-1}	Seawater	100% degraded (120 min)	[95]
F: 600 kHz P: 30 W V: 300 mL	Basic fuchsin, 10 mgL^{-1}	Seawater	100% degraded (120 min)	[95]
F: 600 kHz P: 30 W V: 300 mL	Acid orange 7, 20 mgL^{-1}	Seawater	100% degraded (120 min)	[95]
F: 585 kHz P. 80 W V: 300 mL	Naphthol blue black, 10 mgL^{-1}	Seawater	100% degraded (40 min)	[95]
P : 80 W V: 300 ml	Rhodamine B, 0.5 mgL^{-1}	Bicarbonate-rich water	99.9% degraded (42 Min)	[96]
P: 100 W V: 400ml	Naphthol blue black, 8.11 µmolL^{-1}	Bicarbonate-rich water	80% degraded (60 min)	[97]
P: 60W V: 400 ml	Ethyl violet, 10 mgL^{-1}	Sea water	80% degraded (30 min)	[98]

Despite the effect of above additives Table 1 shows the sonochemical degradation efficiency for some dyes molecules at different condition without additives obtained from the previous studies.Sonochemical treatment reported by Monira et al. in presence of Tert-butyl alcohol and charcoal also showed highly degradation of Congo red [66]. Similar result was also obtained for methyl orange in presence of sucrose [66].Hamdaoui and Merouani carried out the sonochemical treatment of different dyes like Malachite green, Naphthol blue black, Basic red 29, Rhodamine B, Basic fuchsin, Acid orange 7 in sea water and showed that almost 100% dyes are degraded after certain time(table 1)[95]. According to Merouaniet al. and Dalhatou in bicarbonate rich water degradation of Rhodamine B and Naphthol blue blackare possible by sonochemicaltreatment[96, 97]. Research conducted by Rayaroth et al. reported that 80% Ethyl violet in sea water medium can be removed[98] by sonolysis.

8. Conclusions

Removal of organic pollutants from environment as well as from water is very important for the aquatic life and human being. In this sense use of ultrasound i.e. the sonochemical treatment is now considered as a promising technique. The study of this review revealed that the Organic dyes which are very dangerous pollutants found in water mixed from different industrial and household sources can be efficiently removed by sonochemical treatment. Additives like NaCl, CCl_4, H_2O_2, NaCl, Na_2SO_4, Charcoal, Sucrose, Tert-butyl alcohol and ZnO nano-particle increases the rate of sonochemical degradation. To get the benefit from the sonochemical process it is required to apply this process on large scale.

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