

# **Application of Ultrasonic Technology for Water and Wastewater Treatment**

*AH Mahvi*

*School of Public Health and Center for Environmental Research, Tehran University of Medical Sciences, Iran*

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## **Abstract**

Ultrasonic technology as an innovative technology may be used for water and wastewater treatment for pollution removal. This technology acts as an advanced oxidation process. Application of this technology leads to the decomposition of many complex organic compounds to much simpler compounds during physical and chemical compounds during cavitation process. In this article review, some applications of this valuable technology are presented.

**Keywords:** *Ultrasonic irradiation, Sonochemistry, Contaminant removal*

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## **Introduction**

Ultrasound irradiation is a novel advanced oxidation process that has emerged as an answer to the growing need for lower levels of contaminants in wastewater (1, 2). The basis for the present-day generation of ultrasound was established as far back as 1880 with the discovery of the piezoelectric effect by the Curies (3). Cavitation phenomenon was first identified and reported in 1895 (4). Destruction of microorganisms by ultrasonic has been of considerable interest since 1920's when studies of Harvey and Loomis were published. They showed that heating injure the bacteria, but ultrasonic appeared to have a greater effect (5). Since 1945, an increasing understanding of the phenomenon of cavitation has developed coupled with significant developments in electronic circuitry and transducers (i.e. devices which convert electrical to mechanical signals and vice versa). As a result of this there has been a rapid expansion in the application of power ultrasound to chemical processes, a subject that has become known as "Sonochemistry" (3, 6). In the 1960's, research concentrated on understanding the mechanisms of ultrasonic interaction with microbial cells. Cavitation phenomenon and associated shear disruption, localized heating and free radical forma-

tion were found to be contributory causes (7). By 1975 it was shown that brief exposure to ultrasonic lead to thinning of cell walls which was attributed to release cytoplasm membrane from the cell wall. Fecal coliforms inactivation most likely results from a combination of physical and chemical mechanisms which occur during acoustic cavitation, so it is expected that higher intensities will enhance inactivation rates. The correlation of chemical reaction rates and ultrasonic intensity has been reported previously. However, for most processes, increase in process rate not continues with higher sound intensities (8, 9). Since 1990, several studies have focused on the use of ultrasound to remove organic xenobiotics from water (10-13).

## **Sound theory**

Most modern ultrasonic devices rely on transducers which are composed of piezoelectric materials. Such materials respond to the application of an electrical potential across opposite faces with a small change in dimensions. This is the inverse of the piezoelectric effect. If the potential is alternated at high frequencies, the crystal converts electrical energy to mechanical vibration (sound) energy. At sufficiently high alternating potential, high frequency sound (ultrasound) will

be generated. When more powerful ultrasound at a lower frequency is applied to a system, it is possible to produce chemical changes as a result of acoustically generated cavitation (3, 6). Frequencies above 18 kHz are usually considered to be ultrasonic. The frequencies used for ultrasonic cleaning, range 20 kHz to over 100 kHz. The most commonly used frequencies for indus-

trial cleaning are those between 20 and 50 kHz (3, 14, 15). Ultrasound has wavelengths between successive compression waves measuring roughly  $10^{-3}$  cm. These are not comparable to molecular dimensions (Fig. 1). Because of this mismatch, the chemical effects of ultrasound cannot result from a direct interaction of sound with molecular species (6, 16).

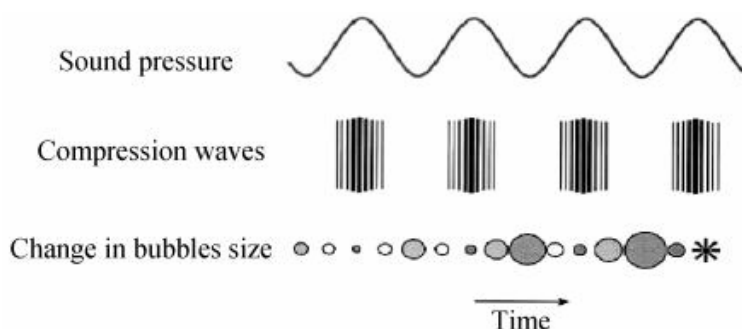
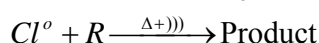
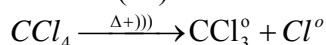


Fig. 1: Compression and expansion cycle of ultrasound (6, 16)

### Bubble cavitation

Ultrasound reactor technology (USRT) in a liquid leads to the acoustic cavitation phenomenon such as formation, growth, and collapse of bubbles (cavitation), accompanied by generation of local high temperature, pressure, and reactive radical species ( $^{\circ}\text{OH}$ ,  $^{\circ}\text{OOH}$ ) via thermal dissociation of water and oxygen. These radicals penetrate into water and oxidize dissolved organic compounds. Hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) is formed as a consequence of  $^{\circ}\text{OH}$  and  $^{\circ}\text{OOH}$  radical recombination in the outside of the cavitation bubble (17-19). Concentration of  $\text{HO}^{\circ}$  at a bubble interface can be as high as  $4 \times 10^{-3}$  M, which is  $10^8$ - $10^9$  times higher than that in the other advanced oxidation processes. Pyrolysis of pollutants could lead to radical formation and starting chain reactions, e.g. degradation of carbon tetrachloride (20):



The basis for ultrasound irradiation applications is that acoustic cavitation can create a number of mechanical, acoustical, chemical and biological changes in a liquid (21, 22).

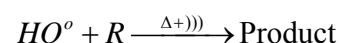
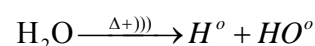
Bubbles form, grow and subsequently collapse through compression-rarefaction cycles. Temperature in collapsing bubbles can reach to 3000-5000°K and pressure to 500-10,000 atm. Under such extreme conditions, water molecules undergo homolysis to yield hydroxyl radicals and hydrogen atoms. Since oxidation by hydroxyl radical is an important degradation pathway, amount of the hydroxyl radicals present in the sonolysis system is directly related to the degradation efficiency (23).

There are two main mechanisms in sonolysis system for pollutant decomposition:

Pyrolysis reactions in cavitation bubbles

Radical reactions by radical species ( $^{\circ}\text{H}$ ,  $^{\circ}\text{OH}$ ) from water sonolysis.

These two mechanisms are as below (20):



In elastic media such as air and most solids, there is a continuous transition as a sound wave is transmitted. In non-elastic media such as water and most liquids, there is continuous transi-

tion as long as the amplitude or loudness of the sound is relatively low. As amplitude is increased the magnitude of the negative pressure in the areas of rarefaction eventually becomes sufficient to cause the liquid to fracture because of the negative pressure, causing a phenomenon known as cavitation. Cavitation bubbles are created at sites of rarefaction as the liquid fractures or tears because of the negative pressure of sound waves in the liquid. As the wave fronts pass, the cavitation bubbles oscillate under influence of positive pressure, eventually growing to an unstable size. Finally the violent collapse of the cavitation bubbles results in implosions, which causes radiation of shock waves from the sites of the collapse. The collapse and implosion of myriad cavitation bubbles throughout an ultrasonically activated liquid result in the effect commonly associated with ultrasound (8, 24, 25). Thus, sonochemical destruction of pollutants in aqueous phase generally occurs as the results of imploding cavitation bubbles and involves several reaction pathways and zones such as pyrolysis inside the bubble and/or at the bubble-liquid interface and hydroxyl radical-mediated reactions at the bubble-liquid interface and/or in the liquid bulk (26).

### **Types of acoustic cavitation**

There are two types of acoustic cavitation: transient and stable (or controlled). Transient cavities exist for a few cycles, and are followed by a rapid and violent collapse, or implosion, that produces very high local temperatures. Ultrasonic cleaning frequencies transform low-energy/density sound waves into high-energy/density collapsing bubbles, producing transient acoustic cavitation. Transient acoustic cavitation can cause damaging surface erosion in more sensitive substrates (8, 21, 27, 28). Totally weaken or disrupt bacteria or biological cells by ultrasonic could be attributed to following processes:

Forces due to surface resonance of the bacterial cell are induced by cavitation. Pressures and pressure gradients resulting from the collapse of gas bubbles which enter the bacterial solution on or near the bacterial cell wall. Bacterial cell damage

results from mechanical fatigue, over a period of time, which depends on frequency (14).

Shear forces induced by micro streaming occurs within bacterial cells (27)

Chemical attack due to the formation of radicals during cavitation in aqueous media. These radicals attack the chemical structure of the bacterial cell wall and weaken the cell wall to the point of disintegration (27)

Amongst final products of this sonochemical degradation of water is hydrogen peroxide, which is a strong bactericide (29).

### **Advantages and disadvantages**

There are no additives introduced into the ultrasonic system and no by products generated by ultrasonic technology. Therefore, there are no anticipated environmental concerns associated with this technology (30).

In contrast to many other processes which are negatively affected when suspended solids of effluent increase, US efficiency may even improve by increase of turbidity or suspended solids (31).

Although the technology has been shown to be feasible on a small scale, the commercialization of sonolysis is still a challenge, due to the high energy requirement of the process (20).

### **Ultrasound applications**

In recent years, considerable interest has been shown in the application of ultrasound as an advanced oxidation process for the treatment of hazardous contaminants in water. Sonochemistry has been demonstrated as a promising method for the destruction of aqueous pollutants (32).

#### ***1- Applications of ultrasound in phenolic effluents treatment***

Phenol is one of the most abundant pollutants in industrial wastewater (33). Phenol is released to the environment from industries such as petroleum refining, coal tar, steel, tanning, pesticides, pharmaceuticals and etc (13, 34-36). Phenol has attracted public attention due to its presence in groundwater, rivers and drinking waters (13). Phenol even in small quantities causes toxicity and foul odor to the water. Most of the countries spe-

cify maximum allowable concentration of phenol in effluent to be less than 1 ppm (37). Several treatment methods such as chemical oxidation, biological treatment, wet oxidation, ozonolysis and activated carbon adsorption have been proposed for the removal of phenol from industrial effluents. In recent years advanced oxidation processes (AOPs) was developed (38, 39). One of these technologies is photolysis. This method is based on supplying energy to chemical compounds as radiation which is absorbed by reactant molecules that can pass to excited states and have sufficient time to promote reactions (40). Direct photoly-

sis has been always considered as one possible alternative because it is possible for molecules of most organic compounds to transform, to cleave bonds and even to undergo complete destruction in the presence of UV irradiation (41).

The photolysis degradation of the phenol at different initial concentration in the range 1-100 mg L<sup>-1</sup> was investigated by (42). Fig. 2 shows the degradation of phenol as function of time. Time required for complete degradation increased from 3 to 120 min when the initial concentration was increased from 1 to 100 mg/L.

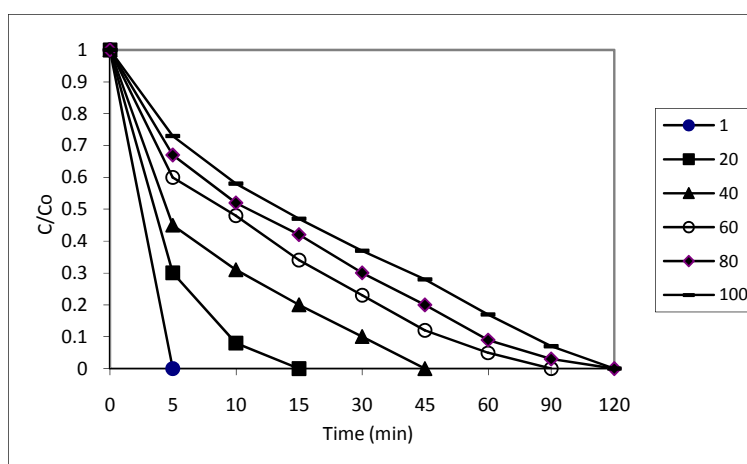
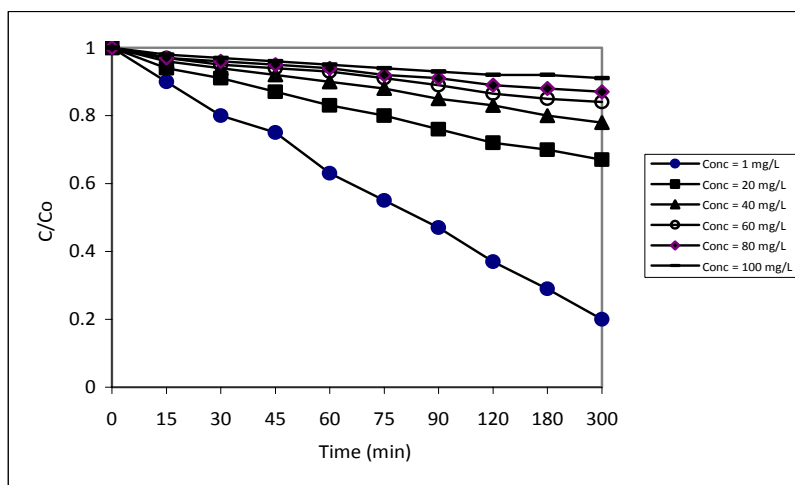


Fig. 2: Effect of concentration on photodegradation of phenol (42)

It is clearly shown that lower pH values favored the phenol degradation. Degradation of phenol attained 94% at pH 3, 91.5% at pH 5, 71% at pH 9 and 62% at pH 11 (42). For photolysis of phenol other researcher have reported that the rate of degradation under acid condition were faster than in alkaline condition (1, 40, 43). It is found that *D. magna* is the most sensitive organism to phenol (44), so authors also studied phenol toxicity on *D. magna*. Results showed that phenol is toxic to *D. magna* and resulted in quite low LC<sub>50</sub> values (LC<sub>50</sub> 96 h of 15.7% v/v), 24 and 48 h LC<sub>50</sub> (% v/v) values ranged from 33.1 and 19.5 for phenol and 66.5 to 42.4 for effluent mixture, respectively. Comparison of Toxicity Unit (TU) between phenol and effluent toxicity showed that TU value for effluent was 2.18

times lower than that obtain to phenol (according to 48 h LC<sub>50</sub>). Thus, photolysis was able to eliminate the toxicity of by-products formed during the degradation of phenol (42). This reduction was achieved by phenol degradation and transformation of aromatics by-products to aliphatic products by ring opening reactions (45). Data of this study showed that bioassay can be used as a suitable method for evaluation of the efficiency of treatment procedures by ultraviolet waves (42).

In other study phenol degradation was carried out for 5 h irradiation time. Figure 3 shows the variations of phenol concentration with time. Only 13% degradation of phenol has been observed for 300 min sonication of 100 mg/L phenol solution (46).



**Fig. 3:** Effect of 35 kHz sonication on phenol decomposition at different initial phenol concentrations (46)

The experimental data from this study fitted well with first order reaction rate equation. Initial rate of ultrasonic degradation was high but later it reduced substantially. It demonstrated that lower pH values had favored the phenol degradation. The maximum and minimum efficiencies of phenol degradation were determined to be 37% and 19% at pH value of 3 and 11, respectively (46).

Photolysis degradation of phenol at different initial concentrations in the range 1-100 mg/L was investigated (47). Figure 3 shows degradation of phenol by the photolysis process at different pH. It was clearly showed that lower pH values favored the phenol degradation. The degradation of phenol attained 94% at pH 3, 91.5% at pH 5, 71% at pH 9 and 62% at pH 11 (47). For photolysis of phenol other researchers have reported that the rate of degradation under acid condition was faster than that in alkaline condition (1, 42, 39). In this study, ionic species of phenol is predominant when pH exceeds 10.0, but molecular species predominates when pH is less than the  $pK_a$ . Fraction in molecular state of phenol was larger when pH was smaller. Therefore, it has been concluded that photolysis of phenol is pH dependent and increases under more acidic conditions. This might be the reason why lower pH favored the ultrasonic degradation of phenol (47).

Francony and Petrier showed that the rates of reactions involving hydroxyl radicals ( $H_2O_2$  formation and phenol degradation) have a maximum value at 200 kHz compared with lower and higher frequencies (20, 500 and 800 kHz) (11). Goel and co-workers recognized that decomposition rates of non-volatiles were lower than volatiles (48).

Study on effect of temperature revealed that the destruction rate of 1, 2-DCA (dichloroethane) is almost independent of temperature (in the range of 15-30° C) (49).

Influences of various factors, such as initial pH, initial phenol concentrations and kinetic constant on the UV degradation of phenol have been studied (42). Also, they determined  $LC_{50}$  of the aqueous phenol solution before and after photolysis (reaction by-products) using *Daphnia magna* as the test organisms.

The degradation of phenol by ultrasonic equipment operating at 130 kHz has been studied (50). Also influences of various factors, such as initial pH and initial phenol concentrations on the ultrasonic degradation of phenol and  $LC_{50}$  of an aqueous phenol solution before and after sonication using *Daphnia magna* as the test organisms were studied. Phenol degradation was for 300 min irradiation time. Fig. 4 shows the change in concentration of phenol over time. They observed

that initially the rate of ultrasonic degradation of phenol is high but later it reduces substantially. This can be explained by the fact that whatever dissolved air is present in the solution, it is degassed after the initial period of sonication resulting in a decrease in the amount of hydroxyl radicals generated.

It has been reported that 96% removal for phenol ( $C_0 = 100 \text{ mg L}^{-1}$ ) by a bath UV equipment during 60 min irradiation (51). Also, 92% degradation for phenol has been reported (1) by means of UV at 254 nm (9 W) for initial phenol

concentration of about  $1.06 \times 10^{-4} \text{ mmol L}^{-1}$  during 60 min.

In other study, the effects of low frequency ultrasound (20 kHz) to remove organic contaminants containing aromatic compounds such as phenol (100 mg/L) in presence of catalysts and alone was evaluated. Results showed that phenol removal is about 10% after 180 min. Also the main mechanism of phenol removal is through reaction with  $^{\circ}\text{OH}$ . Phenol removal efficiency was increased using phenton process up to 85% in 120 min (52).

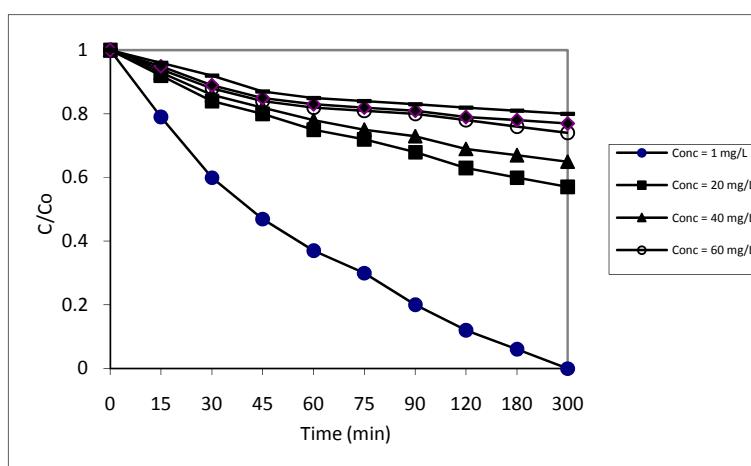


Fig. 4: Effect of the initial concentration of phenol on the sonodegradation at 130 kHz (50)

## 2- Algae removal

A novel method to inhibit growth of algal population is application of ultrasonic irradiation. Ultrasonic irradiation in a liquid medium has been used for many years to lyse biological cells. Ultrasonication may have the potential to reduce their capacity to float and control their buoyancy there by reducing their concentration near the surface of water bodies and reduction their growth and survival. Ultrasonication may also inhibit or reduce growth of algal population through its affect on metabolic processes (53). Application of ultrasonic irradiation to control algal population was evaluated in the laboratory conditions (54) and results showed that short exposure to ultrasonic irradiation collapsed algae gas vacuoles, which results in loss of buoyancy and regulating ability and thus localizing the cells. By 30, 60, 90,

120 and 150 seconds of sonication, respectively 8.55, 35.22, 67.22, 90.67 and 100% of the algal population were destroyed. Besides, results showed that increasing of sonication time has a considerable effect on algal removal. Results indicate that there is no significant reduction in algal population in less than 30 seconds contact time to 42 kHz but considerable reduction in control can be expected at higher periods. Experiments using Bransonic bath at 42 kHz for biological decontamination of water show that destruction of algal population occurs rapidly. It is concluded that using this frequency 100% of the algal population can be destructed in 150 sec (54).

## 3- Nematode Removal

There are more than 15,000 known species of roundworms and several thousands of individual nematodes. Conventional water treatment proc-

esses are not highly effective in nematodes removal. Nematodes are very resistant to inactivation by free chlorine and can pass through rapid sand filters (55). One approach nematode inactivation is ultrasonic (56).

In a research it has been shown that exposure to ultrasonic irradiation results in destruction of nematodes. 12 min sonication destroys 100% of the nematodes. Also results show that increasing of sonication time has a considerable effect on nematode removal. Results also indicate that there is no significant kill of nematodes in less than 8 min contact time to 42 kHz, but considerable levels in control can be expected at higher periods. By 2, 4, 6, 8, 10 and 12 minutes of sonication, respectively 23.75, 42.50, 53.5, 82.25, 89.25 and 100% of adults are destroyed, but by 2, 4, 6 and 8 min of sonication, respectively 38.0, 50.5, 58.75 and 100% of the larva are destroyed (57).

#### 4- Coliform Removal

Results of study showed that increasing of sonication time has a significant effect on bacterial kill. These results also indicate that there is no significant kill of fecal coliforms in less than 20 min contact time to 42 kHz but considerable levels all in activation can be expected at higher periods. When ultrasonic bath is used to sonicate smaller volumes of bacteria at low frequency, there is a resultant in the intensity of ultrasonic entering the system. Furthermore, this study showed removal efficiency in 90 min was highest. On the other hand, sonication of smaller volumes results in more rapid kill. Fig. 5 summarizes results of these experiments. As can be seen up to 99.95% reduction in bacteria concentrations were achieved with the majority of these reductions found to occur in the 90 min (58).

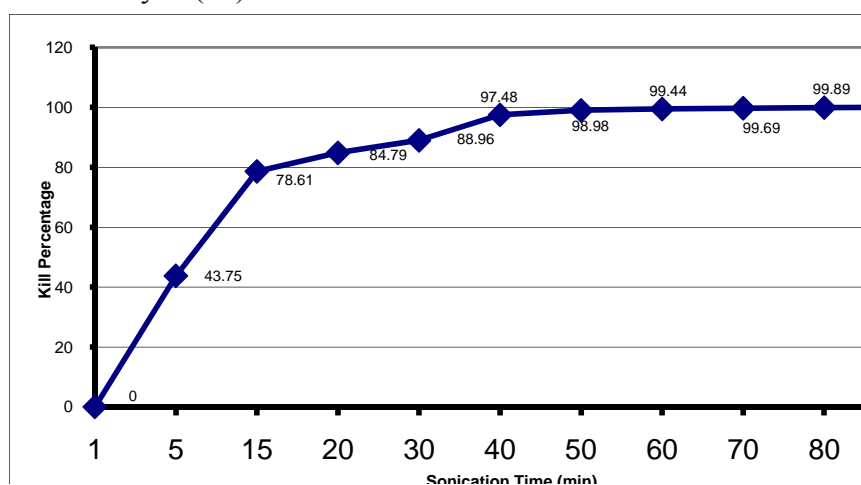


Fig. 5: Sonication time versus fecal coliform removal (58)

Experiments show that it is possible to decrease the number of organisms present in the water and that the process depends on exposure time, frequency and intensity of the ultrasound irradiation, as well as on the type of organisms (8).

Effectiveness of ultrasonic in treatment of total coliforms was studied (59). Results show that increasing in sonication time has considerable effect on bacterial kill. Also, there is no significant kill of total coliforms in less than 20 min contact time to 42 kHz but considerable levels

of inactivation can be expected at higher periods. When ultrasonic bath is used to sonicate smaller volumes of bacteria at low frequency, there is a resultant in the intensity of ultrasonic entering the system. The highest and lowest bacteria reduction after sonication for 300 mL and 600 mL volumes were 99.94% and zero. Also, for 800 mL volumes were 99.63% and zero, respectively. Furthermore, this study showed that removal efficiency in 90 min was highest. On the other hand, sonication of smaller volumes pro-

duced a more rapid kill. Also, up to 99.84% reduction in bacteria concentration was achieved with the majority of these reductions found to occur in the 90 min. they concluded that sonication leads to formation of dead bacterial cells or selectively destroying weak bacteria.

It was shown that by 5, 15, 20, 30, 40, 50, 60, 70, 80, 90 min of sonication, respectively 43.75, 78.61, 82.71, 85.62, 97.82, 98.99, 99.29, 99.50, 99.63 and 99.84% of the total coliforms are destroyed. Besides, the results show that increasing the sonication time has a significant effect on bacterial kill. Results also indicate that there is no significant kill of Total Coliforms in less than 20 min con-

tact time to 42 kHz but considerable levels of inactivation can be expected at higher periods. When ultrasonic bath is used to sonicate smaller volumes of bacteria at low frequency, there is a resultant in the intensity of ultrasonic entering the system. According results the highest and lowest bacteria reduction after sonication for 300 ml and 600 ml volumes were 99.94% and zero. Also, for 800 ml volumes were 99.63% and zero, respectively. Fig. 6 summarizes the results. As can be seen, up to 99.84% reduction in bacteria concentration was achieved with the majority of this reduction found to occur in the 90 min.

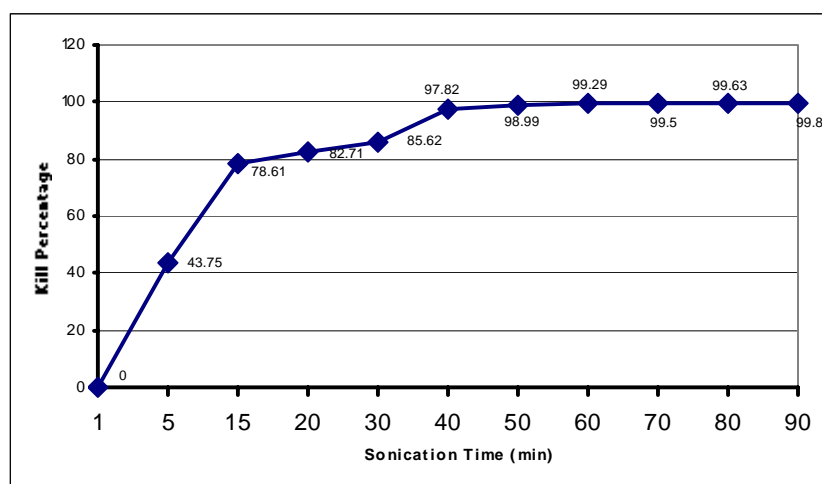


Fig. 6: Sonication time versus total coliform removal (59)

In another research the efficacy of various advanced oxidation processes based on ultraviolet and ultrasound irradiation to inactivate *Escherichia coli* in sterile water and total coliforms (TCs) in biologically treated municipal wastewater has been studied (60). They found that  $H_2O_2$ -assisted UV-A/ $TiO_2$  photocatalysis (9 W lamp) could generally lead to nearly complete *E. coli* destruction in 20 min with the extent of inactivation depending on the photocatalyst type and loading and oxidant concentration. Also Low frequency (24-80 kHz), high power (150-450 W) ultrasound irradiation was less effective than photocatalysis requiring longer contact times (i.e. 120 min) for *E. coli* inactivation (60).

### 5- Organic matters

Results of a study show that US reduces  $BOD_5$  of secondary effluent, but sanitation time had no considerable effect on the efficiency of this treatment. Suspended  $BOD_5$  was removed completely (approximately 100%), however soluble  $BOD_5$  was increased in some cases. Efficiency of total COD removal was determined to be 17-28%. Removal of suspended COD is better accomplished than SCOD. In this study most of COD removal was accomplished in initial sonication time and removal efficiency was not much increased by time. Better organics removal from secondary effluent is performed at 130 kHz compared with the lower frequency. Efficiency of treatment in 60 min



sonication at the frequency of 35 kHz was about 24%, but raised to about 28% at 130 kHz. H<sub>2</sub>O<sub>2</sub> formation at 130 kHz frequency was about 2.5 times higher than 35 kHz. In contrast to TCOD,

removal efficiency of suspended COD was better at 35 kHz. Figs. 7 and 8 shows summary of this study (61).

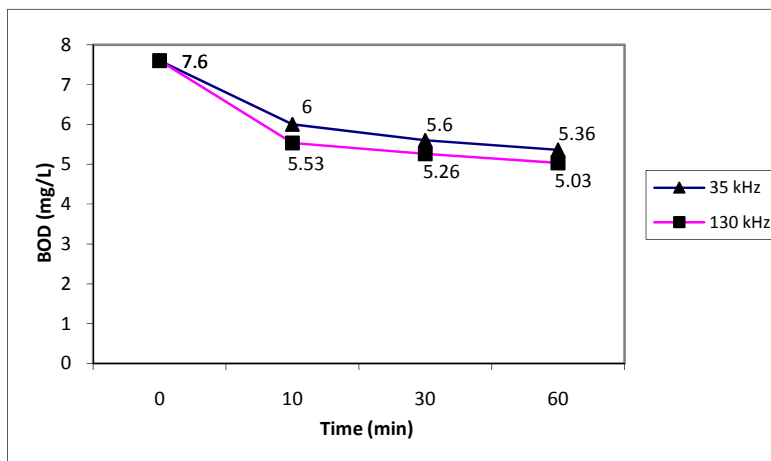


Fig. 7: BOD<sub>5</sub> variations of different effluent samples at two frequencies (61)

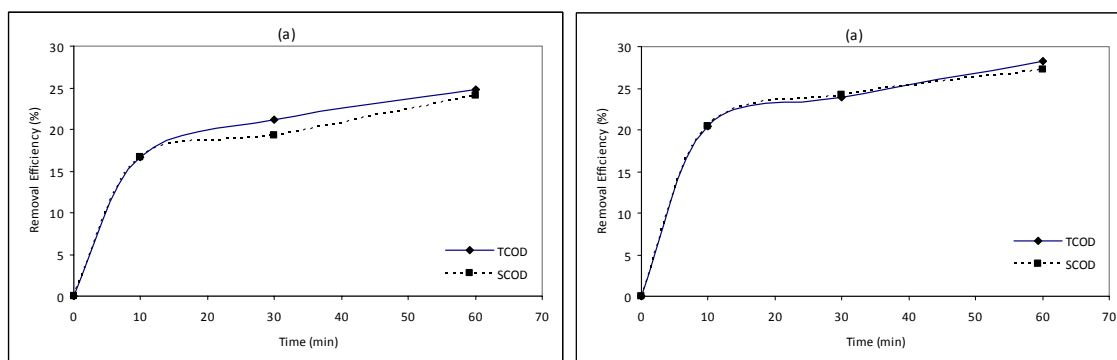


Fig. 8: TCOD and SCOD removal efficiency by ultrasound at two frequencies: (a: 35 kHz and b:130 kHz) (61)

Treatment of raw sewage by sonuv (combined sonication and UV irradiation) in 90 min was not effective to mineralize the organic matter. A significant reduction of COD was observed after 4 h of sonuv treatment (62).

Ultrasonic can decompose other organic substrates such as chlorinated hydrocarbons, pesticides, phenol, explosives such as TNT, and esters, and transform them into short-chain organic acids, CO<sub>2</sub> and inorganic ions as the final products. The time for complete degradation ranges from minutes to hours (63).

The application of ultrasound to remove low-concentration bisphenol A (BPA) in aqueous solution

at the frequency of 20 kHz, and evaluation of ultrasonic intensity and ozone on BPA removal was studied (64). BPA was degraded under US in the presence of CCl<sub>4</sub>. Also they identified the main intermediates resulting from BPA ultrasonic degradation by GC-MS. They found that OH radical induced oxidation is the major destruction pathway during BPA sonolysis (64).

The degradation of bisphenol A (BPA) upon ultrasonic action under different experimental conditions and evaluation of saturating gas, BPA concentration, ultrasonic frequency and power has been studied (65). They found that for 118 μmol/L BPA solution, the best performance obtained at

300 kHz, 80 W, and oxygen as saturating gas. In these conditions, BPA readily eliminated by ultrasound process (90 min). Identified intermediates were: monohydroxylated bisphenol A, 4-isopropenylphenol, quinone of monohydroxylated bisphenol A, dihydroxylated bisphenol A, quinone of dihydroxylated bisphenol A, monohydroxylated-4-isopropenylphenol and 4-hydroxyacetophenone (65).

A novel hybrid advanced oxidation technique (sonoelectro-Fenton process) was applied for the degradation of organic pollutants in aqueous medium (66). They coupled ultrasound irradiation and the in-situ electrogeneration of Fenton's reagent. They studied synergistic action of sonication in the sono-EF process at low and high frequency. It was demonstrated that destruction of herbicides 4, 6-dinitro-o-cresol (DNOC) and 2, 4-dichlorophenoxyacetic acid (2, 4-D) is significantly accelerated. They concluded that improvement yielded by sonoelectro-Fenton process is due to various contributions: (i) enhanced mass transfer rate of reactants towards cathode, (ii) additional generation of OH by sonolysis, and (iii) pyrolysis of organics due to cavitation generated by ultrasound irradiation (66).

The potential of using ultrasonic irradiation for the removal of sodium dodecylbenzene sulfonate (SDBS) at concentrations of 15, 30 and 100 mg/L from aqueous solutions with power values of 45, 75 and 150 W was studied (67). Results showed that SDBS conversion decrease with increasing temperature and initial solute concentration and decreasing power and frequency. Investigations using the radical scavengers 1-butanol and KBr revealed that SDBS degradation proceeds through radical reactions occurring predominately at the bubble-liquid interface and, to a lesser extent, in the liquid bulk. In this research addition of NaCl or H<sub>2</sub>O<sub>2</sub> had little or even an adverse effect on SDBS conversion (67).

In another research the effect of various operating conditions and the presence of matrix components on the sonochemical degradation of naphthalene, acenaphthylene and phenanthrene in water

was studied (68). At the operating conditions in question (initial concentrations of 150, 300 and 450 µg/l, temperatures of 20 and 40° C, applied power of 45, 75 and 150 W and ultrasound frequencies of 24 and 80 kHz), all PAHs were susceptible to sonochemical treatment and, in most cases, complete degradation could be achieved in up to 120 min of treatment. Conversion was found to decrease with increasing initial concentration and temperature and decreasing power and frequency as well as in the presence of an excess of dissolved salts (68).

### **5- Fungi removal**

The results of disinfection during sonicating 500 ml fungi suspension at eight different samples (200, 1000, 2000, 3500, 5500, 6500, 10000 and 17000 CFU/ml) are shown in Fig. 9. Number of fungi decreases with increasing in disinfection period. Results showed that increasing in disinfection time has considerable effect on fungi reduction. Also, there is no significant reduction of fungi in less than 15 min exposure time to 42 kHz but considerable levels of reduction can be expected after longer periods (99.92%) (69).

It is suggested that USRT at a frequency of 26 kHz is capable to some degree of inactivating fungi cells (9). Experiments at 42 kHz can be seen to be more effective than operation at less than this frequency.

In another study it was suggested that in a squeeze-film-type sonicator, more than 90% inactivation of fungi was achieved for 60 min (70). In this experiment, sonolytic inactivation of fungi cells was investigated using a horn-type sonicator at 27.5 kHz frequency. Results of the fundamental investigation included effect of USRT power, cell numbers, and flow rate on the inactivation of the fungi cells using a horn-type sonicator and a squeeze-film-type sonicator. Inactivation by USRT was fastest at the lowest initial cell numbers (70).

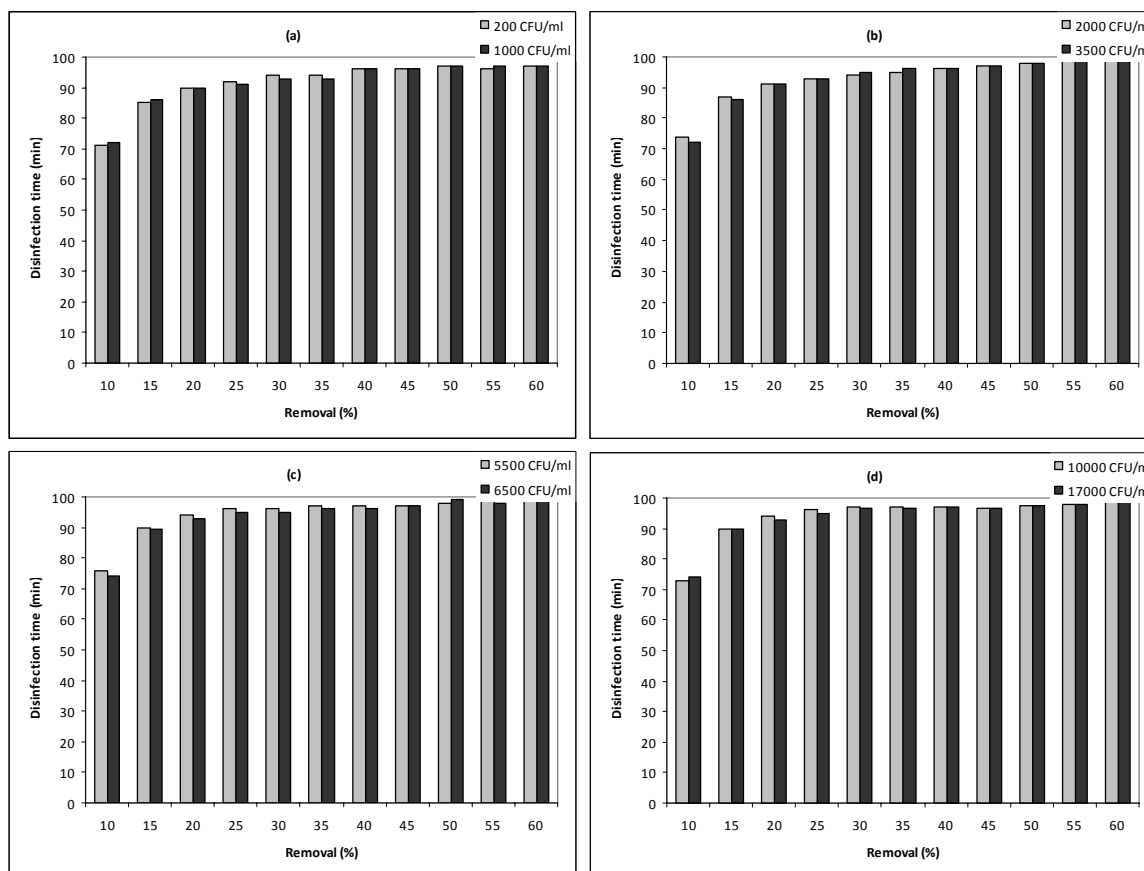


Fig. 9: Comparison of plate count at different samples (69)

### 6- Others Pollutants

LASs are anionic surfactants, found in relatively high amounts in domestic and industrial wastewaters. In a study, effectiveness of acoustical processor for LAS degradation was evaluated with emphasis on effect of treatment time and initial LAS concentration (71). Initial LAS concentrations were 0.2, 0.5, 0.8 and 1 mg/L, acoustic frequency was 130 kHz, applied power was 500 W and temperature was 18-20° C. Results showed that LAS degradation increases with increasing of sonochemical time. In addition as concentration increased, LAS degradation rate decreased in acoustical processor reactor (71). The effect of 1 MHz ultrasound on inactivation of *Cryptosporidium parvum* was studied (72). They found that continuous irradiation of ultrasound (20 min) increases temperature due to cavitation phe-

nomena. Ultrasound irradiation of liquid containing *C. parvum* showed significant quantitative changes in pH, temperature and inactivation of *C. parvum* (102.7 oocysts killed/s) with a minimum energy consumption (0.05 oocysts/s) (72). In another work, the influence of ultrasounds of diversified intensity (22 and 24 kHz) on iron in water was studied (73). Variable operational parameters were vibration amplitude and the exposure time (1–5 min). Effect of ultrasounds was studied as a result of influence of sonochemical oxidation processes and ultrasound coagulation on iron in the ionized form and on iron-organic complexes in the low ionized or colloidal form. The effectiveness of the researched processes was analyzed from the point of view of the possibility of determinate intensity ultrasound usage as an unconventional method of the removal of iron from water.

The degradation of Acid Orange 52 in aqueous solutions using three processes (photocatalysis, sonolysis, and photocatalysis with sonication) was investigated (74). In the case of photocatalysis, concentration of Acid Orange 52 decreased to 35% in 480 min, but it was decomposed completely in 300 min using sonolysis. Also concentration of Acid Orange 52 using photocatalysis with sonication reached to 0 in 240 min. They showed that ultrasonic irradiation enhanced the photocatalytic degradation (74).

In another study the feasibility of sonochemical reaction technology was studied for degradation of reactive yellow dye from aqueous solution. In this study it was shown that the process had very good results in detention time of 120 min at 130 kHz and 500 W (75).

The sonochemical decolorization and decomposition of azo dyes, such as C.I. Reactive Red 22 and methyl orange was investigated (76). They found that azo dye solutions were readily decolorized by the irradiation. The sonochemical decolorization was also depressed by the addition of the t-butyl alcohol radical scavenger. These results indicated that azo dye molecules were mainly decomposed by OH radicals formed from the water sonolysis. They also proposed a new kinetics model taking into account the heterogeneous reaction kinetics similar to a Langmuir-Hinshelwood mechanism or an Eley-Rideal mechanism (76).

Application of ultrasound to remove and recover ammonia from industrial wastewater was studied (77). They used three different concentrations of ammonia [5, 10, 15 Vol%] to study the efficiency of removing ammonia from water. These concentrations are exactly similar to what may be found in wastewater resulting from strippers at petroleum refinery. They found that the ultrasound has the ability to remove ammonia with 5% concentration to meet the local standard of treated wastewater within less than 2 h for 0.080 L solution. They also found that as the concentration of the ammonia increases the removing of ammonia within 2 h decreases, still the concentration of the ammonia meets the standard of

the treated wastewater. The ability of the ultrasound to remove the ammonia failed to produce any mist when the height of the liquid solution increased, namely when the height reached (0.0337 m). It means that the device capacity to remove ammonia has certain limitations based on liquid heights. The best condition for ammonia removal was obtained at 5% concentration and 0.080 L liquid volume (equivalent to 0.0165 m) (77).

## **Conclusion**

Cavitation is a nonthermal mechanism of ultrasonic irradiation that occurs when the gas vesicles are acted upon by a sufficiently intense ultrasonic irradiation of 42 kHz. Observation of differential interference microscopy showed the collapse of the gas vesicles after irradiation, for the collapse caused parts of the cell wall to cave in and consequently the cell surface became uneven. Furthermore, free radical and sonochemical effects can arise when inertial cavitation occurs, which greatly affects passive membrane permeability's, active transport processes and metabolic rates (54).

Experiments suggest that ultrasonic in low-kilohertz frequency range has some efficacy in inactivating some disease agents in water. This would suggest that transient cavitation is the physical mechanism responsible for affecting the microorganisms. The stable cavitation mechanism would appear to require much higher intensity levels for such effects (24).

Studies indicate that some degree of an ultrasonic-induced germicidal effect can be obtained against fecal coliforms in water. However, absolute definitive answers have not been achieved in these experiments. Therefore, additional quantitative studies will be requiring defining more fully the exact exposure condition which might ensure complete germicidal efficacy. Also, results show that increasing of sonication time proves fecal coliforms kill as expected. After 90 min of sonication, 99.95% of bacteria are inactivated and sonication of smaller volumes produced a more rapid kill. But in large-scale water treatment plants

90 min sonication times would prove to be uneconomical at the power used in this work. Therefore, using higher ultrasonic power is more beneficial in above process than using low power and leads to greater efficiency in destruction of bacterial cells (58).

Treatment of secondary effluent by ultrasonic can reduce about 30% of the remained organics in these effluents. This treatment efficiency is probably the result of organics characteristics. Most of the organics in secondary effluent are low-volatile. Besides, it is predictable that most of the remained matter in effluent have hydrophilic characteristics. Therefore, it is probable that the main mechanism of organics removal is treatment by °OH radicals in bulk solution. Pollutants which decompose in this region are less degradable by ultrasound than pollutants which decompose in gas phase. Besides, secondary effluent contains different organic compounds with specific characteristics. Thus, each have different behavior in treatment by ultrasonic. Moreover, these different compounds may interfere with the decomposition process of each other and deteriorate or enhance the ultrasonic treatment. Inorganic matter can affect the decomposition of organics too. Sometimes, treatment by US converts complex organics to much smaller compounds and it is obvious that much sonication times are needed for complete demineralization. Often, relative conversion of organics suffices for meeting much of the requirements. As these simple compounds have organic nature, the effect of treatment can not be detected by routine tests of COD and BOD<sub>5</sub> and in other words, by these tests it is difficult to show the effect of ultrasound on organics decomposition. For example, in Sono-oxidation of humic acids (78), complete degradation of these compounds occurred in 60 min whereas, reduction of TOC was only 40%. Suspended COD has converted to SCOD during sonication. Previous works on SCOD of wastewater sludge confirm our result about conversion of suspended COD to SCOD. For example, one of the previous studies showed considerable increase of SCOD of sludge after sonication

such that the SCOD was reported to increase from 620 mg/L to 2100 mg/L after 2.5 min and to 4200 mg/L after 10 min (79). The mechanical shear forces caused by ultrasonic may be the dominant factor for the disintegration enhancement (80).

USRT substantially improves the effectiveness of removing sewage fungi through the effects of acoustic cavitation in water. Transient cavitation and stable cavitation need to be considered in order to gain an understanding of what cavitation like activity might be responsible for the reduction of sewage fungi. In propagated ultrasound reactor, transient cavitation process occurs more easily at lower ultrasound frequency. As a result, USRT is suitable for disinfection of sewage fungi. For effective reduction of fungi using USRT alone it is almost certain that USRT would need to be applied in combination with another common disinfection technologies used in water treatment including ultraviolet irradiation, ozone or chlorination. USRT is a very small unit that easily can be installed at any place in a treatment plant. Quality USRT can replace sand filters that usually serve as a step to remove suspended solids prior to disinfection. There is scientific and economic potential in the development of combined disinfection processes. In order to definitely damage sewage fungi walls higher USRT energy input is necessary. Also, combination with other disinfectants applications is useful.

Experiments on LAS degradation showed that treatment time is the most important parameter for LAS degradation. Acoustical reactors alone may not be useful for reducing completely complex wastewaters of high surfactant load and could be improved by coupling with other treatment processes including ozone, UV, chlorination and H<sub>2</sub>O<sub>2</sub>. (71)

From these studies of the effects of ultrasonic upon the destruction of microorganisms, it can be seen that ultrasonic is suitable for water disinfection and can achieve the following:

Remove chlorine from water efficiently (81).

Ultrasonic reduces the amount of chlorine required for disinfection (81).

Sonication leads to the formation of dead bacterial cells or selectively destroying weak bacteria (29). Sonication of smaller volumes produced a more rapid kill (8).

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