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WATER AND WASTEWATER DISINFECTION BY ULTRASOUND IRRADIATION-A CRITICAL REVIEW

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ABSTRACT

It is well known that chlorine and its compounds, traditionally utilized for water and wastewater disinfection, react with some organic matter to form undesirable by-products, hazardous to human health, known as Disinfection By-Products (DBPs). In many countries very stringent limits for chlorination by-products such as trihalomethanes were set for wastewater reuse. Accordingly, the use of different oxidation/disinfection systems should be evaluated as possible alternative to chlorine. Ultrasound (US) was recently found to be effective for this purpose.

Aim of this work is to review main US disinfection studies, pointing out ultrasound mechanisms as well as its effects in terms of different bacteria inactivation (*Total coliform, Escherichia coli, Pseudomonas aeruginosa, Bacillus subtilis, Saccharomyces cerevisiae, Klebsiella pneumonia*) at both laboratory scale and pilot-scale. To this end, several experimental results were discussed and both focal interest points and encountered problems were summarized.

Moreover the intensification of cavitation phenomena by combined oxidation processes was overviewed and main advantages and disadvantages were pointed out, in order to address future research and promote efficient large scale operations.

Keywords: cavitation, combined treatments, Escherichia Coli, frequency, microorganisms inactivation, pathogen, removal, sonication.

1. Introduction

Disinfection process is a conventional step used to remove pathogen microorganisms from both drinking water and wastewater, in order to eliminate waterborne microbiological contamination caused by pathogenic bacteria, viruses and protozoan parasites and protect public health as well as the environment.

Disinfection agents commonly used within both drinking water and wastewater treatment plants are chlorine and its related compounds, such as sodium and calcium hypochlorite and chlorine dioxide, with chlorine being by far the most widely used disinfectant (Winward *et al.*, 2008). The strong oxidizing potential of these reagents provides a minimum level of chlorine residual throughout the distribution system and adequate protection against microbial recontamination (Sadiq *et al.*, 2004).

However, in the early 1970s, it was found that chlorine reacts with the natural organic matter present in water and wastewater to produce various undesirable chlorinated disinfection by-products (DBPs). Among these, trihalomethanes (THMs) and haloacetic acids are the most dangerous by-products for public health since they are considered to be potentially carcinogenic and/or mutagenic (Rook, 1974). Therefore, the use of chlorine compounds as disinfectant in water and wastewater treatment requires the dechlorination of the effluent to minimize the potential toxic effects of low level chlorine residuals as well as to prevent the formation of DBPs in receiving water bodies.

This disadvantage has emphasized the need for exploring alternative disinfectants and new treatment technologies (Gopal *et al.*, 2007). In this respect, ozonation and UV-C irradiation have traditionally been tested and, to a certain extent, employed as disinfection methods due to the simplicity of operation and the relatively affordable operating and maintenance costs (Rice *et al.*, 1981; Symons *et al.*, 1989; Lin *et al.*, 1999; Bonacquisti, 2006; Wait *et al.*, 2007). In recent years research on disinfection has been directed to the use of (i) TiO_2 heterogeneous photocatalysis driven by artificial (UV or visible) or natural sunlight irradiation (Fernández *et al.*, 2005; Belgiorno *et al.*, 2007), and (ii) ultrasound (US) which represents a relatively innovative technique to conventional treatment technologies (Mason and Tiehm, 2001; Khan *et al.*, 2006; Naddeo *et al.*, 2007).

The science of ultrasound involves the study of the formation, impact and applications of sonorous waves occurring at frequencies higher than 20 kHz, which represents the upper audibility threshold of the human ear.

Ultrasound has been studied with reference to physical (Mason, 1976; Hoyler and Luke, 1984; Asher, 1987), chemical (Richards and Loomis, 1927; Flisak and Perna, 1977; Suslick and Doktycz, 1990; Thomas and De Vries, 1959; Srinivasan *et al.*, 1995), medical (Dunn, 1991; Byrne, 1984; Kulier and Kapp, 2011), industrial and environmental engineering applications (Brown and Goodman, 1965; Thakore, 1990; Hunicke, 1990; Floros and Liang, 1994; Ince *et al.*, 2001; Mason, 2007). The latter include (i) the degradation and removal of conventional and emerging contaminants in wastewaters (Naddeo *et al.*, 2007, Hoffman *et al.*, 1996; Lifka *et al.*, 2003; Gogate, 2008; Naddeo *et al.*, 2009a; Naddeo *et al.*, 2009b; Naddeo *et al.*, 2010; Mendez-Arriaga *et al.*, 2008; Secondes *et al.*, 2014), (ii) drinking water and wastewater disinfection (Harvey and Loomis, 1929; Mason *et al.*, 2003; Naddeo *et al.*, 2009c; Arrojo *et al.*, 2008), (iii) sludge (Tiehm *et al.*, 1997; Chu and Lee, 2004; Wang *et al.*, 2005; Bougrier *et al.*, 2005; Climent *et al.*, 2007; Naddeo *et al.*, 2009d) and solid waste (Chen *et al.*, 2008; Cesaro *et al.*, 2012; Cesaro and Belgiorno, 2013; Cesaro *et al.*, 2014) treatment prior to anaerobic digestion, (iv) landfill leachate pre-treatment (Lema *et al.*, 1998; Gonze *et al.*, 2003; Neczaj *et al.*, 2007), (v) extraction of substances from contaminated sediments (Mecozzi *et al.*, 2002; Collasiol *et al.*, 2004; Moreda *et al.*, 2004) and (vi) air purification (Grinthal and Ondrey, 1992; Serpone *et al.*, 1994).

The use of ultrasound for disinfection purposes has been extensively studied. This paper aims at reviewing main studies on the topic, thus pointing out action mechanisms of ultrasound and its efficiency as well as the variation in the process performance occurring after the combination of ultrasound with other oxidation processes.

Main advantages and drawbacks are also highlighted, in order to address future research and promote efficient large scale operations.

2. Disinfection mechanisms and reaction systems during ultrasound irradiation

The first report on the use of ultrasound as disinfectant was published by Harvey and Loomis (1929) in the late 1920s. In their pioneering work, authors reported the positive disinfectant action of ultrasound, pointing out that the process could not be considered of any practical or commercial importance due to its expense.

In early to mid 1970s, three other reports (Burgos *et al.*, 1972; Burleson *et al.*, 1975; Dahl, 1976) discussed the likely use of ultrasound as a disinfection agent due to its ability to decrease the heat resistance of bacterial spores (Burgos, 1972) or to act synergistically to ozone-induced disinfection of viruses and bacteria (Burleson *et al.*, 1975; Dahl, 1976).

Nowadays, it is well-documented that ultrasound disinfection power is related to the occurrence of cavitation phenomena (Mason and Peters, 2002). It consists of the production of micro-bubbles, which are generated when a great negative pressure is applied to a liquid (Mason and Peters, 2002). Compression and rarefaction waves rapidly move through the liquid media. If the waves are sufficiently intense they will break the attractive forces in the existing molecules and create gas bubbles. As ultrasound energy enters the liquid, the gas bubbles grow until they reach a critical size beyond which they either implode or collapse, thus releasing a great energy amount and promoting sonochemical reactions (Neppiras, 1980; Dehghani, 2005). In a cavitating liquid these reactions occur in three regions (Figure 1): the gas bubble; the interface between the gas phase and the liquid bulk; the liquid bulk itself (El' Piner, 1964).

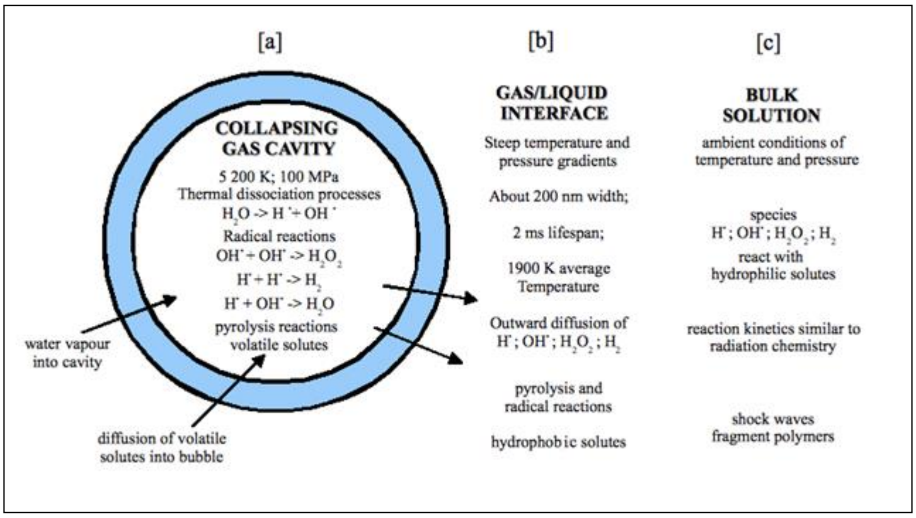

Figure 1. Schematic diagram of the regions in a cavitating liquid where chemical reactions take place

According to the "hot-spot theory", cavitational bubble collapse produces an intense increase in temperature, up to 5000 K, as well as in pressure, thus promoting the formation of free radicals (e.g., •OH, $HO_2^{\bullet}$ and O•) (Furuta *et al.*, 2004), with a strong oxidative power. Both physical and chemical effects are, therefore, promoted by cavitation: it has been extensively proved that physical phenomena are predominant at low frequency, while high frequency ultrasound mainly supports chemical effects (Carrére *et al.*, 2010).

Therefore, a number of physical, mechanical and chemical processes arise from acoustic cavitation, which can inactivate bacteria and de-agglomerate bacterial clusters (Furuta *et al.*, 2004; von Sonntag, 1986; Oyane *et al.*, 2009; Joyce *et al.*, 2003a). These effects include:

- 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;
- shear forces induced by micro-streaming occurring within bacterial cells;
- chemical oxidation by free radicals (•OH and •H) during cavitation in the aqueous medium. These radicals attack the chemical structure of the bacterial cell wall and weaken the cell wall to the point of disintegration.

As a result, disinfection processes rely on two main phenomena occurring due to acoustic cavitation effects.

The first is bacterial splitting which breaks up bacterial agglomerates into a greater number of individual bacteria in a suspension. The second is bacterial killing (or inactivation) which results in less individual reproduction ability of bacteria being present in a suspension. The overall effect of applying ultrasound is thus the result of a competition between splitting and inactivation of bacteria in solution. For this reason disinfection efficiency is strongly influenced by both irradiation time and intensity as well as from the reactor configuration.

Several types of reaction systems (Figure 2) have been employed for ultrasound-induced disinfection studies, as reported in several papers (Reisse *et al.*, 1999; Gogate and Pandit, 2004; Gogate, 2007). In brief, the ultrasonic probe is suitable to treat relatively small volumes of liquid with the ultrasound irradiation being localized around the emitting horn and not distributed as in the case of ultrasonic bath, plug-flow reactor and the flow cell. The plug-flow reactor consists of immersed ultrasound transducers unlike the bath, where the transducers are not in direct contact with the liquid phase; moreover, it can treat larger volumes than the probe or the bath. On the other hand, the flow cell is a unique system that works under pressure and the water inside is sonicated all around.

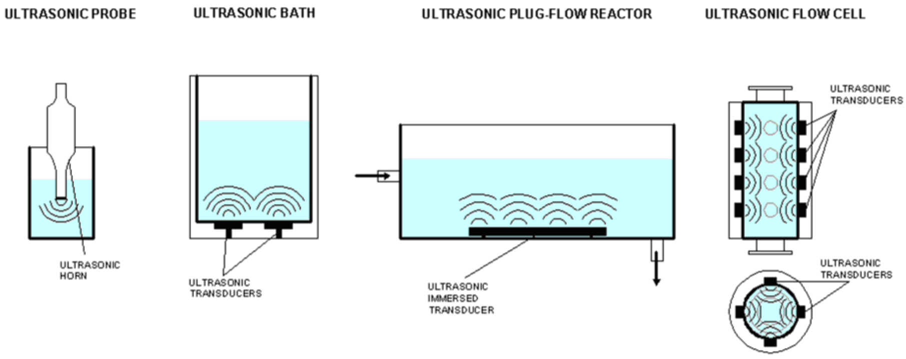

Figure 2. Ultrasonic systems typically used for sonochemical treatment

The following paragraphs overview several studies on the use of ultrasound, either alone or in combination with other advanced oxidation processes, to treat several types of microorganisms in different aqueous matrices.

3. Disinfection by ultrasound irradiation

Table 1 summarizes studies on the inactivation of various microorganisms induced by ultrasound irradiation as the sole disinfectant; relevant operating conditions and efficiencies are quoted.

Most studies focus on the inactivation of *Escherichia coli* in various aqueous matrices, mainly synthetic ones. The interest in this kind of microorganism is reasonably related to the evidence that it is one the most used faecal contamination indicator in Regulations and guidelines dealing with water and wastewater quality assessment.

E. coli inactivation exhibited pseudo-first order behaviour and its extent was found to enhance with increasing intensity: Hua and Thompson (2000) investigated the effect of ultrasound density in the range 270-460 W l^{-1} at a frequency of 20 kHz and they observed that the extent of *E. coli* inactivation became about 2.8 Log after 60 min at the highest density of 460 W l^{-1}.

Similarly, Antoniadis *et al.*, (2007), performed experiments at laboratory scale, using both synthetic municipal and septic tank wastewaters, in order to verify US effects on *E. coli* inactivation. In both cases, 100 ml of wastewater were subjected to ultrasonic irradiation by a device operating at a fixed frequency of 80 kHz and a variable electric power output up to 150 W, connected to a titanium-made horn with a 7 mm tip. For those experiments carried out at a frequency of 24 kHz, a horn-type sonicator operating at a variable electric power output up to 450 W was used instead.

It was observed that high power and low frequency ultrasound is capable of eliminating nearly completely the *E. coli* colonies in synthetic municipal wastewaters as well as the total microbiological load in actual municipal wastewaters at relatively short irradiation times.

Furuta *et al.*, (2004), investigated the inactivation of *E. coli* along with hydrogen peroxide formation due to water sonolysis, in order to assess the feasibility of hydrogen peroxide formation as an indicator of bacterial inactivation. A 27.5 kHz horn type sonicator was used; its operation was based on the "squeeze-film effect" (i.e. the film is defined as the space between the end of the probe of the sonicator and the bottom of the reactor) and the maximum power of this sonicator was 42 W ml^{-1}. When the amplitude on the vibration face was 3 mm, inactivation was 6 Log at room temperature. They observed that the ultrasonic shock wave was more important in killing microorganisms than the indirect effect of $^{\bullet}OH$ radicals formed by ultrasonic cavitation. Ultrasound waves at a frequency of 42 kHz were also used to treat aqueous suspensions of *E. coli* in the study by Dehghani, (2005); the author reported a 2.7 Log inactivation at a power density of 120 W l^{-1} and a sonication time of 90 min.

In most of the reviewed studies, authors used low frequencies systems, which allow the achievement of the best performances in terms of microorganisms inactivation in comparison to high frequency ultrasonic technologies. Joyce *et al.* (2003a) studied the effects of sonication on *Bacillus subtilis* inactivation at different frequencies and found that both 20 and 38 kHz did not determine dramatic effect on the viability of the bacteria. Conversely, over the first 5 min, the higher frequencies produce an immediate rise in the concentration of microorganisms expressed as CFU (Colony-Forming Unit). Then a steady fall was observed but the level remains above the original concentration even after 15 min sonication.

These results suggest that the major effect of high frequency ultrasound is the declumping of bacterial agglomerates with little deactivation, while low frequency ultrasound, at higher powers, are characterized by a substantially higher kill rate.

Ultrasound has also been used in combination with other advanced oxidation processes. In the following paragraphs main combinations are pointed out, in order to highlight advantages and drawbacks.

4. Disinfection by ultrasound irradiation coupled to other processes

Although sonication can provide powerful disinfection, it is an energy-consuming system. In order to reduce US energy input, great interest has been directed towards the coupling of this technology with other processes, such as TiO_2 photocatalysis, UV irradiation, electrolysis and ozonation or other disinfectants such as sodium hypochlorite and hydrogen peroxide.

Table 2 summarizes studies on the inactivation of various microorganisms induced by ultrasound irradiation coupled to another process or disinfectant; relevant operating conditions and efficiencies are quoted.

4.1 Ultrasound and electrolysis disinfection efficiency

Among the studies dealing with ultrasound and electrolysis coupling, Joyce *et al.*, (2003b), investigated the efficiency of the combined process as a disinfection treatment to inactivate *Klebsiella pneumonia*. Experiments were conducted in a temperature-controlled ultrasonic bath (at a frequency of 40 kHz), with the electrodes immersed in the liquid phase. The electrode materials were carbon (felt and graphite), copper and stainless steel rods. It was found that the combined treatment was more efficient than ultrasound irradiation or electrolysis applied individually.

Disinfection happens as an effect of metal ions impacting microbial cells or through the generation of hydrogen peroxide, hydroxyl radicals and hypochlorous acid. At a frequency of 40 kHz and a density of 50 W l^{-1}, the inactivation of *Klebsiella pneumonia* was 50% after 2 min sonication and it proved to be complete after 5 min.

Table 1. Efficiency of various microorganisms inactivation by ultrasound irradiation only.

Microorganism	Aqueous mean	Ultrasonic system	Frequency [kHz]	Density [W l^{-1}]	Maximum sonication time [min]	Work highlights	Reference
Bacillus subtilis	Synthetic solution	Bath	38	180	15	Although US proved to be effective in bacteria declumping, the kill rate of low frequency high power systems is substantially higher	Joyce *et al.*, 2003a
		Probe	20	240			
			512 and 850	71 or 64			
Escherichia coli	Synthetic solution	Probe	20	270	60	E. Coli inactivation exhibits depends moderately on total power and power intensity at low frequency	Hua and Thompson, 2000
	Synthetic solution	Probe	27.5	42	3	Inactivation rate gradually increased with increasing amplitude of the vibration face	Furuta *et al.*, 2004
	Aqueous suspension	Bath	42	0.12	90	Low frequency US effective in E. Coli inactivation	Dehghani, 2005
	Synthetic municipal and septic tank wastewaters	Probe	24 and 80	1500-4500	120	Ultrasound provided a permanent elimination of cells	Antoniadis *et al.*, 2007
Saccharomyces cerevisiae	Synthetic solution	Probe	27.5	0.16	10	Yeast cells suffer bactericidal effects within early periods of sonication	Tsukamoto *et al.*, 2004
Total coliforms	Well water	Probe	22	2.4	15	Overall disinfection rate provided by the bath is higher than the one provided by the horn.	Jyoti and Pandit, 2004a
		Bath	20.5	0.06	15		
Gram-positive/gram-negative bacteria	Secondary treated municipal wastewaters	Probe	24	1.500	60	High removal efficiency for gram-negative bacteria than for gram-positive	Drakopoulou *et al.*, 2009

Table 2. Combination of ultrasound and other processes for microorganisms inactivation

Microorganism	Aqueous mean	Process/reagent combined with US	Ultrasonic system	Frequency[kHz]	Density [W l^{-1}]	Maximum sonication time [min]	Reference
Escherichia coli	Synthetic solution	Sodium hypoclorite (1 mg l^{-1})	Bath	20 and 850	6	5	Duckhouse *et al.*, 2004
	Synthetic solution	TiO_2 photocatalyst	Bath	39	-	30	Dadjour *et al.*, 2005
	WWTP effluent	UV-C (200 W/lamp)	Reactor	39	5	30	Naddeo *et al.*, 2009c
Klebsiella pneumonia	Synthetic solution	Electrolysis at 100 mA	Bath	40	50	5	Joyce *et al.*, 2003b
Legionella pneumophila	Synthetic solution	TiO_2 photocatalyst (0-1g ml^{-1})	Bath	36	50	30	Dadjour *et al.*, 2006
Pseudomonas aeruginosa	Water samples	Cl_2 (1 mg l^{-1})	Bath	800	15000	20	Phull *et al.*, 1997
	Secondary treated municipal wastewaters	TiO_2 (5 mg l^{-1})	Probe	24	1500	30	Drakopoulou *et al.*, 2009
Total coliforms	Well water	H_2O_2	Bath	20.5	2400	15	Jyoti and Pandit, 2004a
	Well water	O_3	Bath	20.5	60	15	Jyoti and Pandit, 2004b
	Synthetic solution	TiO_2 (0.25-0.75 g l^{-1}) Cl_2 (1-5 mg l^{-1}) UV-A (9 W/lamp)	Probe	24-80	9000	30	Paleologou *et al.*, 2007
	WWTP effluent	UV-C (200 W/lamp)	Reactor	39	5	30	Naddeo *et al.*, 2009c
	Secondary treated municipal wastewaters	TiO_2 (5 mg l^{-1})	Probe	24	1500	30	Drakopoulou *et al.*, 2009

After 10 min electrolysis 100% kill was achieved using all electrodes, with copper being the most efficient. When lowering the current from 150 mA to 100 mA, the 40 kHz ultrasound produced faster removal rates than the ones achieved with electrolysis alone at the higher current of 150 mA.

This evidence suggests that the application of a combined US and electrolysis systems, under the optimal conditions, allows the reduction of energy inputs required to achieve comparable results by using the single technologies.

Schlager and Gorski, (2004), have recently patented a combined ultrasound and electrolytic disinfection apparatus that includes an electrolytic flow cell with electrodes forming a part of flow pipe or open channel through which water or wastewater passes. The electrodes are made of iron, stainless steel, carbon or copper. They are connected to a power supply voltage ranging between 20 and 100 V, establishing a power supply in the range 1 - 6 A. An ultrasonic transducer is connected to the electrodes and it enhances hydroxyl radical generation.

One of the most important aspects to be taken into account when electrolytic cells are employed is related to metal electrodes, which increase metal concentrations in solution, sometimes exceeding maximum contamination levels for silver, copper, lead or other metals. This represents one of the main disadvantages for the use of this technology in combination with US. Moreover, few papers on the topic are reported in literature, thus reducing the reliability of the combined process as well as the assessment of the economic feasibility.

4.2. Ultrasound and TiO_2 photocatalysist disinfection efficiency

The addition of a catalyst such as TiO_2 has been studied in order to reduce the ultrasonic energy input for disinfection purposes.

The combined process relies on the synergistic effect which occurs when ultrasonic energy is supplied to TiO_2 particles. In these conditions, excited electrons move from the valence band to the conduction band and positive holes are generated in the valence band. Near the surface of TiO_2 particles, holes react with water to generate more hydroxyl radicals (Cai *et al.*, 1992), which provide an additional disinfection effect. When an ultrasonic wave is propagated via the solvent, all TiO_2 particles are expected to contribute to the generation of radical species and the rate of disinfection would then be accelerated.

Several studies report the inactivation of *E. coli*, in the presence of TiO_2 photocatalyst. Dadjour *et al.*, (2005), used an ultrasonic bath at 39 kHz and found that 98% reduction in the concentration of viable cells was achieved in presence of TiO_2 during a 30 min period of irradiation. Only 13% reduction was observed when an ordinary ultrasonic irradiation system without TiO_2 was used. Thus, TiO_2 promotes the disinfection process by seven-fold under the investigated conditions. Authors stated that the effect of TiO_2 was related to the heterogeneous nucleation of bubbles, which enhances the cavitation power. This, in turn, may increase the pyrolysis of H_2O molecules and the formation of OH radicals, which are highly reactive and, therefore, short-lived. Moreover, TiO_2 pellets stabilize reactive species, resulting in a more intensive oxidation (Dadjour *et al.*, 2005).

Similar results were found on *Legionella pneumophila*. Dadjour *et al.*, (2006), found that only 18% of the initial viable cells were killed after 30 min of treatment but the concentration of viable cells was reduced to 3% of the initial concentration in the presence of 1.0 g ml^{-1} TiO_2 after a 30 min treatment period.

Other studies used the probe as US system in combination with TiO_2. Drakopoulou *et al.*, (2009), found that after 30 minutes sonication, 5.0 g l^{-1} TiO_2 results in disinfection performances ranging between 37 and 99%, according to the considered bacteria species. In particular, these authors stated that Gram-negative bacteria were more sensitive to sonication than Gram-positive ones.

Although the proposed method is capable of achieving disinfection standards for wastewater reuse, combined process optimization is still required to achieve the cost-effectiveness for large scale applications.

4.3 Ultrasound and ultraviolet disinfection efficiency

Short-wave ultraviolet light (UV-C) is a radiation in the range 200-280 nm in the UV spectrum, whose germicidal effect on bacteria, virus, protozoa, fungi and algae (Unluturk *et al.*, 2008) has been extensively investigated for disinfection purposes (Litved and Cripps, 1999; Sutton *et al.*, 2000).

Several studies evaluated the effectiveness of ultrasound application as a pre-treatment step in combination with ultraviolet rays to optimize wastewater disinfection process (Blume and Neis, 2003).

Paleologou *et al.*, (2007), examined this combined system and compared it with various other combinations of both ultrasound and UV radiation with TiO_2 photocatalysis. Authors found that US allowed the halving of the reaction time necessary to obtain complete removal of *Total coliforms* by UV radiation alone.

Naddeo *et al.* (2009c) investigated the combined ultrasound and ultraviolet disinfection process in a pilot-scale configuration which is schematically shown in Figure 3.

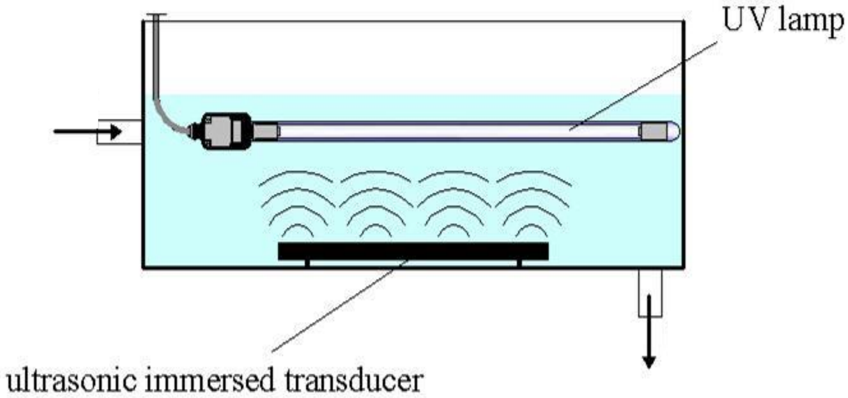

Figure 3. Experimental setup for combined ultrasound and ultraviolet disinfection (Adapted from Naddeo *et al.*, 2009c)

The reactor consists of a low frequency (39 kHz) ultrasonic transducer capable of operating at ultrasound power varying between 350 and 1400 W and two low pressure UV-C lamps of 150 W each. The treated volume was 80 L. Experimental tests showed that UV disinfection efficiency was enhanced in presence of US, especially when treating wastewater with low transmittance.

After approximately 55 h of continuous treatment, the disinfectant power was still up to 90% in the combined ultrasound/UV reactor, whereas in the UV reactor, the inactivation went down until 77%. This evidence was related to the influence of US on the formation of fouling on the lamps.

During the tests, lamps in UV reactor became dirtier and dirtier; in presence of US, UV lamps were perfectly clean, even after three days of treatment. The US cleaning effects was provided by the collapse of cavitation bubbles, which produced liquid jets on the surface of lamps. In this way, ultrasound broke the cake layer on the lamps making the UV beans emission achievable in wastewater.

The combined US/UV-C system has been recently studied for elimination of pathogens in recirculating aquaculture systems (Bazyar Lakeh *et al.*, 2013). To this end, dose-dependent inactivation rates were determined for the total viable counts and model organisms representing different taxa of common fish parasites. Authors found that a pre-treatment with low frequency ultrasound reduced the mean size of suspended solids in aquaculture water, thus increasing the germicidal effect of UV-C by up to 0.6 log units.

The innovative aspect of this recent investigation lays in the coupling of UV systems, which are commonly used for the prevention of bacterial, viral and fungal diseases in aquaculture facilities, with low frequency US that had not been studied yet in the aquaculture field.

The effectiveness of coupled UV-C and US process lays in the possibility of improving disinfection yields of the former by using the latter to reduce dissolved organic matter and suspended solid, which are recognized to provide significant interferences to UV irradiation.

In this way, UV reaction time can be conveniently shortened to ensure disinfection targets. The reduction of reaction time would result in the decrease of working volumes, with the consequent reduction in capital costs. Moreover, the positive influence of US on lamp fouling would reduce maintenance costs. Despite these advantages, US/UV process cost-effectiveness should take into account the energy requirements during the simultaneous operation of the combined system. Further studies are, therefore, necessary to assess energy balances as well as to evaluate the competitiveness of this system with other integrated processes.

4.4. Ultrasound and ozone disinfection efficiency

The use of ultrasound in combination with ozone, which is a powerful oxidant, enhances water and wastewater treatment by producing an emulsion from both suspended particles and dispersed microorganisms as well as by preventing the coalescence of ozone bubbles, ensuring that maximum bubble surface area is available for oxidation (Burleson *et al.*, 1975).

One of the first studies was performed by using a pilot plant processing 76 m^3 d^{-1} of sewage: it was found that 1 min of treatment with both ultrasound and ozone destroyed 100% of faecal bacteria and viruses (Chendke and Fogler, 1975).

Jyoti and Pandit, (2004a), also examined the application of acoustic cavitation and ozonation for the disinfection of water. Their lab-scale work was carried out with an ultrasonic horn operating at 22 kHz frequency and 240 W electrical power. They also used an ultrasonic bath (145×145×150 mm) characterized by a peak operative frequency of 20.5 kHz and an electrical power consumption of 120 W. After 15 min sonication, 100% efficiency in removing *Total coliforms* was achieved.

The same authors studied the coupling of ozone and US for the inactivation of *Total coliforms*, *Faecal coliforms* and *Faecal streptococci* in bore well water (2004b). In their investigation, ozone was supplied by a generator according to the corona discharge method. A dry air flow rate of 28 l s^{-1} was used as the feed gas. Sonication was applied through the same devices used in the previously mentioned investigation (Joyti and Pandit, 2004b). It was found that by using the combination of ultrasonic horn and ozone or hydrodynamic cavitation and ozone, the concentration of ozone required for disinfection was reduced to half or one-third depending upon the type of microorganism.

Although both ozone and ultrasound are effective technologies for water and wastewater disinfection, main disadvantages are related respectively to the high costs for ozone production and to the great energy consumption for ultrasound generation. However, their combination can allow the reduction of both ozone doses and ultrasound energy, with the consequent reduction of operating costs.

A further advantage from the coupling of ozone and US for disinfection purposes and, as a consequence, from the decrease of ozone doses is the elimination of toxic by-products associated to ozonation reactions and, in particular, to the oxidation of natural organic matter (NOM) or halogen compounds (von Gunten, 2003).

The assessment of the cost-effectiveness of the combined US/O_3 disinfection process requires further studies dealing with operating condition optimization, as already pointed out for the combined US/UV process. However, in this case, the high costs associated with ozone generation at full scale would represent a limit for the spread of the combined process, especially if compared with the coupling of US with other chemical processes.

4.5. Ultrasound and chemicals disinfection efficiency

A further option for water and wastewater disinfection is the application of ultrasound in combination with chemicals.

The effects of low (20 kHz) and high (850 kHz) frequency ultrasound on the biocidal efficiency of sodium hypochlorite against *E. coli* suspensions were studied by Duckhouse *et al.*, (2004). Operating at the lower frequency and a power intensity of 17 W cm^{-2}, maximum inactivation was achieved when ultrasound irradiation and hypochlorite were applied simultaneously to the suspension. Conversely, at the higher frequency of 850 kHz and a power intensity of 0.03 W cm^{-2}, efficiency was optimized when ultrasound was used as a pre-treatment immediately followed by hypochlorite addition under silent conditions. In particular, pre-treatment using 850 kHz proved to be very effective at 1 min exposure with an increase of kill by about 2-log reduction in comparison with the control experiment. On the other hand, pre-treatment at 20 kHz had a small adverse effect on the rate of kill of chlorine at all exposure times up to 5 min pre-treatment, while simultaneous application of hypochlorite and ultrasound resulted in almost a 2-log reduction in kill at either 1 or 5 min exposure.

Coupled ultrasound and chlorine treatment was applied for water disinfection in the study of Phull *et al.*, (1997). The experimental work was performed using both a probe and a bath on previously chlorinated samples. Authors observed that sonication amplified the effect of normal chlorination, thus promoting the reduction of the amount of chlorine required for disinfection.

Finally, Joyti and Pandit (2004a) investigated the effects of a combined US/H_2O_2 process on bacteria inactivation. To this end, for the experiments involving the HPC bacteria, 150 mg l^{-1} H_2O_2 was used and for the indicator microorganisms (*T. coliforms*, *F. coliforms* and *F. streptococci*) 5 mg l^{-1} H_2O_2 was used. This dosage was added to bore well water before subjecting it to ultrasonication. It was observed that the disinfection efficiency of acoustic cavitation was increased when hydrogen peroxide was added, with higher specific extent of disinfection for the combination of 5 mg l^{-1} of hydrogen peroxide with the ultrasonic bath rather than horn.

Similar results were obtained for wastewaters.

Ultrasound and chlorine dioxide were combined sequentially to improve *Escherichia coli* and *Total coliform* inactivation in raw wastewater (Ayyildiz *et al.*, 2011). A sequential application of ultrasonic density values of 150 or 300 W l^{-1} and a ClO_2 concentration of 2 mg l^{-1} provided about 3.2–3.5 log reduction in the number of microorganisms, while the sum of log reductions by the individual treatments were 1.4-1.9. This enhancement was attributed to the presence of high concentration of particles in raw wastewater and their influence in improving ultrasonic cavitation effects.

The comparison of those works highlighted the fundamental role played by the matrix under investigation and the need for studies on real wastewater. Although the use of synthetic solutions can promote the identification of disinfection mechanisms as well as the evaluation of specific aspects by eliminating interferences related to the presence of different substances, experimental results acquire a further reliability when validated under conditions as near as possible to the real ones.

Although treatment conditions are quite different in the reviewed studies, it can be concluded that short sonication times allow the decrease of chemical disinfectant amount. This item results in the reduction of operating costs associated with reagent supply and points out that US could be an interesting option for the upgrade of conventional chlorination systems.

Moreover, sonication times are even shorter than the ones applied in combination with TiO_2 addition or UV irradiation, suggesting that the combined US/chemical disinfectant process could be more competitive.

5. LIMITS AND POTENTIAL OF ULTRASOUND TECHNOLOGY

The use of ultrasound has been extensively studied for disinfection purposes, even in combination with other systems, so that main potentialities and limits can be highlighted.

Table 3 summarises main advantages and disadvantages of ultrasound induced disinfection systems.

As pointed out in previous paragraphs, ultrasound process can determine the removal of different kind of pathogens from both water and wastewater and inactivation yields are usually high and quite close to the complete removal, notwithstanding the kind of microorganism under investigation. This evidence can be related to the action mechanism of ultrasound, whose effects are mainly produced by the mechanical disintegration of bacteria cell, especially when operating in the field of low frequencies. Moreover, it has been recognized that US is also effective in the degradation of chemical pollutants, so that not only disinfection but also an improvement of the process effluent can be achieved. This item is even more interesting if the generation of disinfection by-products is considered: differently from other conventional systems, such as chlorination or ozonation, ultrasound does not provide the formation of toxic compounds, potentially harmful for human health.

Table 3. Advantages and disadvantages of ultrasound-induced disinfection systems

Advantages	**Disadvantages**
Simple, flexible design with low capital costs	Design criteria still developing
Easy upgrading of conventional treatment unit	Increase of water turbidity
High efficiency of several bacteria inactivation	Energy consumption
Oxidation of natural organic matter and degradation of chemicals pollutants	Maintenance/replacement of ultrasound probe
No production of conventional disinfection by-products (THMs, etc.)	Lack of remaining disinfection capacity
High synergy/improved efficiency in combination with conventional disinfection treatments (O_3; Cl_2; UV).	

On the other hand, the turbidity increase should be highlighted as one of the main disadvantages of US application, along with the lack of a remaining disinfection capacity.

Under an operational point of view, ultrasonic reactors are compact and flexible, often commercialized as modular units, so that their implementation for the upgrade of existing disinfection units can be considered. In this case, the high synergy with conventional disinfection treatments ensures the achievement of high pathogen removal yields.

It should be pointed out that, up to now, the design features of most devices have been developed following empirical data: theoretical criteria are still being developed in order to rationalise the design of these systems as well as their application to larger volumes.

From an economic point of view, despite a relatively low capital cost, ultrasound allows the economic treatment only for small volumes, mainly due to the high energy consumption associated to ultrasound generation. Further operating costs are related to the maintenance and/or replacement of the devices, which tend to be eroded by the ultrasonic action itself.

6. CONCLUSIONS

In recent years the increasingly restrictive limits for water and wastewater disinfection has addressed the research towards the study of innovative methods to ensure high yields in pathogenic bacteria removal. The ultrasound process, even in combination with traditional disinfection methods, showed interesting results that were summarised in this work.

This review showed that for both *Escherichia Coli* and *Total Coliform* the best results in water disinfection were obtained through the use of ultrasound treatments with low frequencies (20 - 40 kHz), medium-low power (< 120 W), high density and sonication times varying between 3 to 15 min.

Similar treatment conditions can determine bacteria inactivation usually greater than 90%, notwithstanding the kind of microorganism under investigation. However, some species of bacteria, which can agglomerate into clusters thus proving to be resistant to conventional disinfection methods, were removed more easily by means of ultrasonic processes.

Literature data also pointed out that sonolysis efficiency in bacteria inactivation can be further increased by coupling this technology with other oxidative systems. As an additional advantage, this condition is often obtained with a significant reduction in reaction time. Pilot scale experiments proved that only 1 minute of treatment by means of ozonation and sonolysis allowed 100% microorganism inactivation.

On the basis of literature review, further studies should be addressed towards the definition of relations between disinfection yields and main parameters affecting process efficiency, such as contact time, radiated power and reactor features. The in-depth analysis of these aspects could also support the study of theoretical models able to describe the acoustic cavitation field provided by US, even in combination with other processes, as well as to predict the effects of specific treatment conditions on different kinds of matrix to be disinfected.

The analysis of scientific literature shows that most studies were performed using synthetic wastewater. The civil wastewater use is an essential factor for the reliable estimation of disinfection process efficiency, as it allows the investigation of conditions close to the real ones, thus paving the way for process scale-up considerations.

Experimental results are certainly encouraging, even with reference to possible reuse of investigated aqueous matrixes, ensuring high pathogenic removal yields without the risk of formation of toxic by-products, such as trihalomethanes (THMs). Further benefits from US disinfection treatment are related to: the guarantee of high efficiency bactericide for various viral and bacterial species, even with reference to those chlorine-resistant; the high oxidant power which can reduce concentrations of organic matter and many toxic compounds, such as pharmaceuticals, usually found in wastewater; the possibility of applying this technology for the upgrade of existing plants. Conversely, the disadvantages are primarily associated with increasing turbidity, which is generated by long delays contact times, high density and transducers wear.

In the perspective of real scale applications of ultrasound disinfection treatments, a fundamental aspect to be better clarified is the possible formation of harmful by-products: to this end, toxicological analyses are required.

Finally, the feasibility of this technology full-scale needs to be assessed. More studies should, therefore, be carried out in order to establish energy consumption levels, in order to verify the technical and economic competitiveness of ultrasound towards conventional technologies.

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