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Contemporary application of microbubble technology in water treatment

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Abstract

Microbubble technology (MBT) constitutes a suite of promising low-cost technologies with potential applications in various sectors. This paper present contemporary assessments of microbubbles (MBs) in water treatment processes. A summary of the recent finding of MB in water treatment, discussion on the existing research gaps, challenges and limitations in upscaling of the technology, conclusion and future scope is detailed. An in-depth review of the cost and energy consumption is done to develop an insight into the steering transition from more expensive conventional technologies to eco-friendly MBT in water treatment.

Keywords: microbubble, cavitation, water treatment, aeration

Introduction

With the current surge in global environmental challenges, there is a shift towards more eco-friendly and sustainable technologies. Microbubble technology (MBT) has recently emerged as a viable option in water treatment. THIS IS NOT A PARAGRAPH AND NEEDAS MORE. WHY WATER TREATMENT NEES SOMEHTING NEW AS A SENTENCE OR TWO.

Air, oxygen and ozone microbubbles (MBs) have shown great potential for reducing the running cost and economising of the waste-water treatment plants (WWTP) [1]. Micro and nano bubbles have been reported to be effective in the processes of aeration, flotation and disinfection [2,3]. Microbubbles (MB) are tiny bubbles with diameter in the range of 10-100µm. The shared characteristics of these MBs, which make them unique is their longer residence time, slow buoyancy, self-pressuring effect, large gas-liquid interfacial area as compared to the conventional macrobubbles (REF).

MB consists of three segments: an inner gas phase, an outer liquid phase and a shell which separates these two different phases. Each bubble has a critical radius as defined by Young-Laplace equation (REF). Bubbles smaller than that radius have low buoyancy forces and tend to slowly diffuse the gas present within and shrink while ascending slowly (REF). Such bubbles finally collapse underneath the liquid surface, producing reactive free radicals. Conventional macro-bubbles on the other hand, during coalescence develops size larger than the critical radius, rise-up quickly and explode on the liquid surface [4]. Figure 1 shows the time-conditioned shrinkage mechanism of micro and macro-bubble. The presence of MB is known to influence the physico-chemical and mechanical property of the bulk liquid. This change in liquid property has shown to be beneficial for further chemical reaction like oxidation.

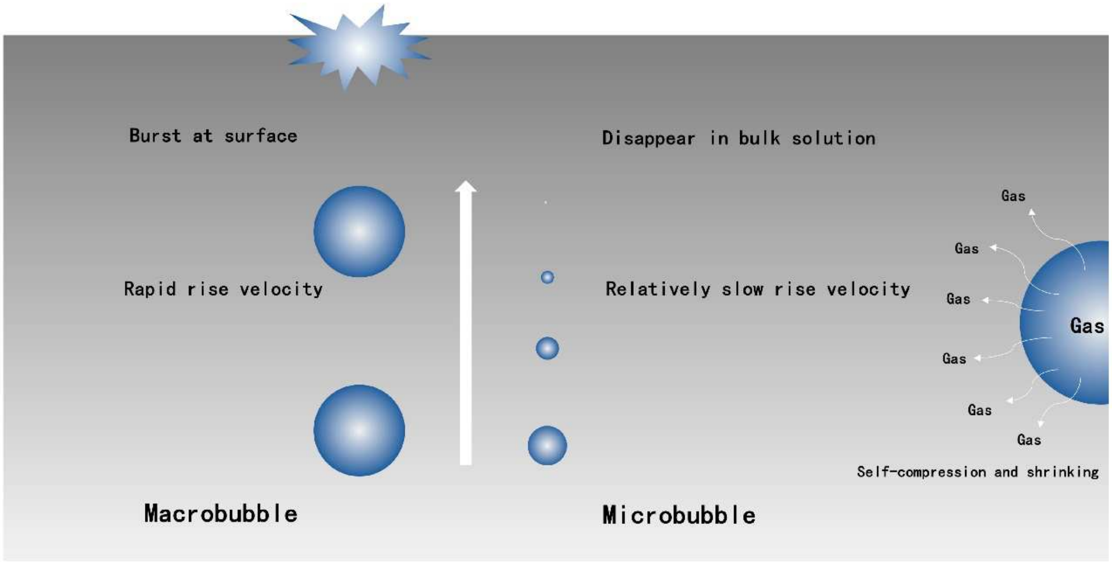

Figure 1: Schematic diagram of macrobubble and microbubble [5]

MB Generation

The formation of bubbles in the liquid is thought to be a static or quasi-static progression followed up by dynamic regime of coalescence and break up. This overall process of bubble inception, its growth and collapse is called the cavitation. The driving mechanism of cavitation is a reduction in static pressure of liquid below its vapour pressure, leading to vapor/gas filled cavities in the liquid. The cavitation is unlike the bubble formation in boiling where the process is primarily driven by temperature change.

Microbubble generator (MBG) has been chosen depending upon the application requirement. Whether it is in production on a laboratory scale, for research purpose, actual field trials or at an industrial scale. The capacity and concentration of microbubbles production required for the treatment and the cost of treatment also decides the MBG employed. All these factors decide the choice of MBG.

Based on source employed for cavitation, it is classified into the following types.

1. Hydrodynamic cavitation- This type of cavitation is induced as a result of local pressure and the velocity changes of the flowing liquid owing to the passage through restricted geometry of the system.

2. Acoustic cavitation- Cavitation is achieved through high intensity ultrasonic fields. The equipment and operational cost are higher for this type when scaled at the industry level.
3. Particle cavitation- Particle cavitation is caused by beam of elementary particle rupturing the liquid flow.
4. Electrolysis cavitation- The application of electric current to the fluid to produce cavitation
5. Optic cavitation-Such type of cavitation is produced as a result of interaction with high intensity light (laser), which breaks the continuum of liquid flow.

Out of these cavitation techniques hydrodynamic and acoustic cavitation has been most widely applied in waste-water treatment [6]. Optic, electrolytic and particle cavitation are not capable of stimulating any changes in the bulk liquid. However, hydrodynamic and acoustic cavitation has shown to bring about the desired physico-chemical changes in the bulk liquid [7]. Though the acoustic technique may seem to work for lab scale trials requiring high accuracy, but it is not suitable for large scale processes owing to the high associated costs with its operation (why and REF). When it comes to pilot and large-scale processing, hydrodynamic cavitation technology remains the first choice for MB formation in water treatment.

In water treatment processes, MB is generated by one of the below mentioned hydrodynamic process:

- Pressurised dissolution or decompression type generator- Gas is dissolved in the water stream by creating high pressure of about 304-405 KPa *i.e.*, super saturation of gas. Supersaturated gas being unstable escapes generating MBs.
- Cavitation by Venturi type generator- In this method, MB is generated by the passage of air and water simultaneously through the venturi tube. The system has three sections, namely a converging inlet, throat, and diverging outlet. Pressurised fluid enters through the inlet and velocity of the fluid increases at the cost of a decrease in static pressure in

narrow cross-section throat. This accelerated fluid generates cavitation and sucks in the gas. A multiphase flow of the gas and liquid is generated in the throat, creating microbubbles [8]. These MBs creates shock wave and highly localised temperature and pressure waves when they collapse and brings about splitting of water into reactive radicals $OH^{\cdot}$ and $H^{\cdot}$. This type offers the benefit of lesser pump power, and compacted size, apart from the power to pressurise the air-water mixture to the venturi additional power input is not required.

- Orifice type generator- In this method, air is sucked into the vacuum created by the movement of fluid through holes in the orifice plate. Unlike the venturi, orifice type generator generates intense cavitation conditions owing to immediate contraction and divergence sections. A higher cavitation yield can be achieved by optimizing the geometric parameters of the orifice plate like number of holes and hole sizes.
- Ejector type generator- In this type of reactor, shrinking or step by step enlargement of the fluid channels occurs producing complex pressure profiles as the liquid flows. At the lowest pressure point, gas will be sucked in generating MBs from the shear of the turbulent liquid flow [9].

Hydrodynamic cavitation appears to be the most practical and economical way to generate MB at pilot and industrial scale and at higher rates using a simple centrifugal pump along with a flow confiner like venturi tube, orifice plate, throttling valves (why?) . For more information on the micro-bubble generation methods and bubble characterisation techniques, one can refer to these recent articles [10, 11].

Some studies have reported generation of significant reactive oxygen species or free radicals by hydrodynamic cavitation as the MB collapse. For example, Khuntia et al. quantified the hydroxyl radicals produced from ozone MBs at different pH using p-chlorobenzoic acid radical probe. Their study revealed a higher generation of hydroxyl radical at acidic pH than that in the alkaline medium [12]. Zheng et al. ascribed the better degradation of biorefractory organic compounds by MB ozonation to the greater amount of unselective hydroxyl radicals produced using fluorescence detection [13]. However, some other studies highlighted that the treatment process with microbubbles is most likely to be thermal based rather than free radical based [14]. Though there is debate over the mechanism behind the organic water pollutant degradation. Still, all these studies identified feasibility of the hydrodynamic cavitation in water treatment. Bandala et al. claimed that hydrodynamic cavitation alone or coupled with other advanced oxidation technique is a promising practice to remove organic contaminant like azo dye and antibiotics entering the water cycle [14]. NEEDS A CONCLUSION SENTENCE HERE

The ever-increasing interest in MBT and its application to water treatment can be seen through a sheer increase in the number of publications related to this technology over the last few years (Figure 2). Though studies pertaining to microbubbles in bulk liquid have been followed from quite a long but still ambiguity exists regarding the mechanism of its operation and real-world application of this technology [15].

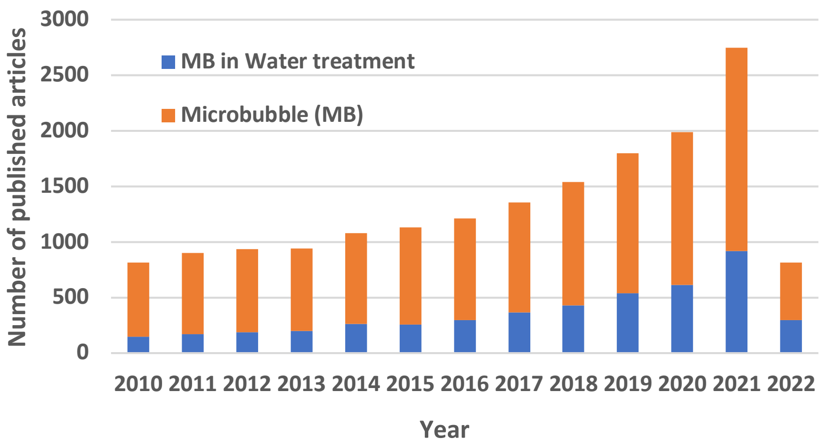

Figure 2: Publication related to microbubble and its application in water treatment over the years. Source: Science direct

Previous literature have focussed on generation techniques, operational conditions employed, bubble size and distribution measurements, and characterisation techniques (REFs). However, limited efforts are directed towards studying the application gaps of MB/NBs in water treatment technology. Though few reports accounted the possibility of scale up to industry level, but a comprehensive, up-to-date report on the existing knowledge gap in the application field is desired to explore the endless avenue of this technology. This article reviews and briefly discusses the various MB generation technologies employed and comprehends their applicability in the state-of-the-art water treatment processes.

Application of Microbubbles in water treatment

1. Aeration- Environment friendly biological treatment methods have been the preferred treatment of organic waste water. The metabolism of microorganisms is used in biological treatments like bio-activated sludge, biofilm, and membrane bioreactors to degrade the harmful chemicals. These processes are limited by the high cost incurred during aeration, sludge treatment and issues of membrane fouling. Aeration or oxygen supply in the conventional activated sludge system consumes 50-90% of total electricity of the WWTP and traditional aerators have extremely low oxygen transfer efficiency [16]. Microbubbles with long retention time and high gas mass transfer efficiency is conducive to energy conservation and cost reduction in WWTPs. Microbubbles makes it possible to diffuse oxygen more effectively in aerobic waste water treatment.

   A novel aeration system was proposed for waste-water treatment employing microbubble generator for faster oxygen supply to microorganisms. The oxygen absorption measurements and power requirements of various aerators were evaluated. Figure 3 shows the comparison of the specific power requirement of various microbubble generators and typical gas distributors. Though, the specific power consumption of microbubble generator was found to be higher than gas distributors, but they allowed faster dissolution of oxygen in water. So, overall reduction in cost of aerobic treatment is expected of microbubble generator as they aid in the downsizing of aeration tank and reduce the residence period of waste water [17].

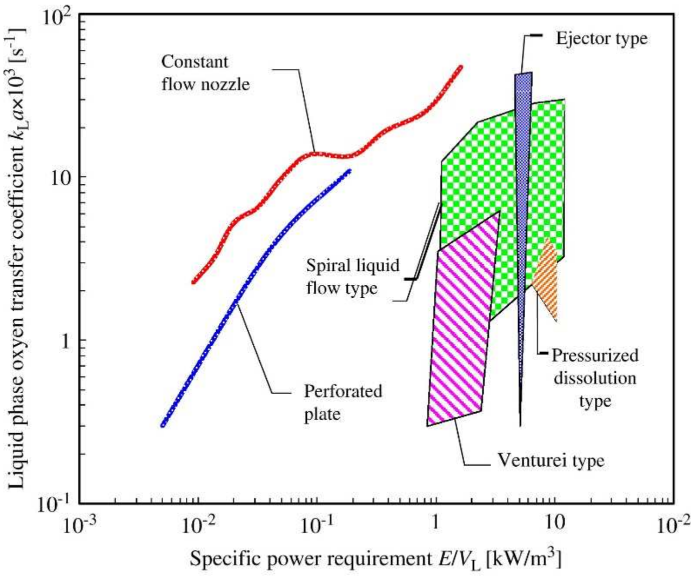

Figure 3: Specific power requirement of various aerators to dissolve oxygen into water [17].

Though MBT is believed to support biological aerobic waste-water treatment owing to its high oxygen mass transfer rates, but few studies have exposed the negative effect of MBT on the mixed liquor property of activated sludge during microbubble aeration. The high shear force generated during MB generation, was found to break sludge flocs, reducing microbe population available for oxidation of organic matter [18]. To tackle this problem Budhijanto et al. proposed to combine MBG with an attached growth aerobic system. The successful application of MBG as the aerator with higher soluble chemical oxygen demand (SCOD) removal efficiency in the low gas flow rates is seen. They emphasised on the careful design and selection of MBG configuration and its relative position (if more than one MBG used) in the reactor to avoid bubble coalescence [19]. Lei et al. have investigated the effect of microbubble aeration on the biofilm formation. The synthetic municipal waste-water was treated in fixed bed biofilm reactor using microbubbles generated from Shirasu porous glass (SPG) membrane system. Microbubble aeration led to 80% faster biofilm formation as compared to coarse bubble aeration. This enhancement was related to the improved attachment of the suspended microbes to the carrier surface by microbubbles. The SPG membrane area-based chemical oxygen demand (COD) removal capacity was found to be 6.88 kg COD/ (m^2 d) with a COD removal efficiency as high as 91.7% [20].

Much research has also been carried out to study the effect of aeration by MBs in degradation of organic matter, seed germination and growth. A visible difference in aerial vegetative plant growth is seen with MB and macrobubble aeration [21]. MB aeration has also known to boost the growth of white shrimps *Litopenaeus vannamei* and biofloc [22]. Although small scale MB experiments are suggestive of aeration intensification and its energy saving capacity, however, it is intended to have further extended studies to replicate the results at larger scales and explore the efficiency of the process.

2. Physical Separation or Flotation- It is the most widely adopted means of removing contaminants of suspended oil, low density suspended solids and colloids in the waste-water treatment. The major steps leading to the separation are adsorption of gas bubbles on suspended particles forming bubble-particle aggregates. As formed aggregates being lighter, rises to water surface which can then be scraped off. Conventional air-flotation is limited by the bubble-particle interaction or collision and so, can separate only narrow particle size range. Several studies have shown that reducing the size of bubbles help in increasing the overall efficiency of flotation separation by promoting bubble-particle collision and attachment [23]. The MBT has been used to enhance the flotation

process [24]. Suwartha et al. have supported the application of smaller MBs to increase the mass transfer coefficient in flotation and aeration process.

A vortex type sparger producing a greater number of smaller bubbles, with slower rising velocity and longer residence time was found to benefit the floc capturing process and gas transfer [25]. Dissolved air flotation (DAF) with MBs and NBs have shown to remove emulsified crude oil in saline water. A flocculation polymer Dismulgan at an optimum concentration of 5 mg/L was used for destabilisation and flocculation of emulsion. The conditioning of oil flocs with NBs have promoted the oil removal efficiency of the process. NBs are believed to form aerated flocs by adhering to the inside of the flocculated oil droplet decreasing their density, which in a way helped the MBs in flotation [26]. The same authors have extended the DAF technology for the removal of Fe^{3+} precipitates and $Fe(OH)_3$ nanoparticles. Figure 4 shows the stages in the flotation with MBs and NBs. The removal efficiency reached as high as 99% with the initial iron feed of 30 mg/L [27]. A concluding sentence here

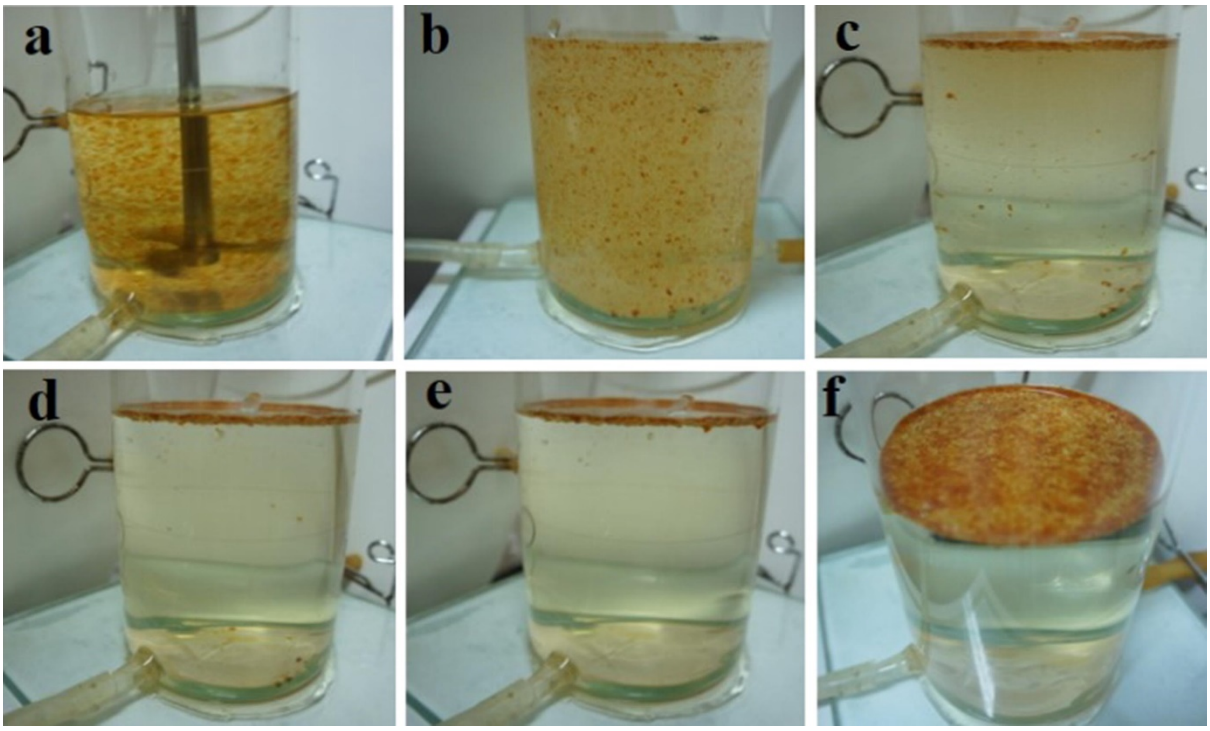

Figure 4: Photographs of different stages of Fe $(OH)_3$ precipitation, before and after flotation with MBs and NBs: (a) Precipitation; (b) Injection of MBs and NBs (c) Flotation at 30 s (d) Flotation at 1 min (e) Flotation at 5 min (f) Floated precipitates and treated water. Conditions $[Fe^{3+}]_{feed}$ = 30 mg /L; pH = 7; saturation time= 30 min [27].

The separation of contaminant from oil containing restaurant waste water has been undertaken by the novel microbubble air flotation. It was reported that highest oil removal efficiency was achieved when the microbubbles are of similar size to oil droplets. The maximum removal efficiency of oil, COD and turbidity achieved with microbubble air flotation is 97.6%, 83.6% and 97.5%, respectively [28]. Liu et al. reported a cost-effective and efficient ozone MB application in coagulation-MB flotation process for the treatment of coke waste-water containing refractory organic compounds. Enhanced flotation degradation by ozone MB as compared to oxygen and air MB was attributed to its highest zeta potential and greater production of hydroxyl radicals [29].

A superior microbial degradation and flotation separation has been suggested with microbubbles for waste-water treatment from the beverage industry. The study has supported the exploitation of bubble surface adsorption and flotation method for the removal of high nitrogen containing dissolved organic matter (DOM) from waste-water. As compared to macrobubbles, microbubbles have shown to support the growth of aerobic bacteria and accelerate the degradation of DOM [30].

Recently, an effective, and flotation simulation method has been proposed for the mixture optimisation between micro and nano bubble in flotation arrangement. The lab scale experimental study confirmed the deterioration effect of only nanobubbles (NB) presence to the water quality due to the long stagnation time of NB aggregate. However, same study argued the improvement in removal efficiency of fine particle as small as 25 µm of nylon, polyvinyl chloride (PVC) and kaolin by hybrid bubbles (MB and NB). NBs helps MBs to aggregate whereas, MBs increase the removal efficiency of NB aggregate in flotation process [31]. It is envisaged that with the use of competitive MBT in the flotation plants, otherwise fraught field of flotation or physical separation is making a strong comeback as compared to large settling tanks set-up. Microbubble flotation technique is flourishing with clarification of lightly laden waste-water on the grounds of energy saving, compactness and ease of operation.

3. Degradation of organic and inorganic pollutants- MBT can enhance the oxidation and remediation to degrade the organic pollutants like organic nitrogen, organic halogens and hydrocarbons to the less toxic material. Ozone is the most powerful oxidising agent and has been practiced in different areas of water treatment process like breakdown of refractory organic matters and disinfection. Ozonation is the preferred choice as compared to chlorination because of no chemical residuals after the process completion, whereas chlorine leave behind carcinogenic by-products. The economics of treatment with ozone are a function of its size. A comparison of total organic carbon (TOC) and dimethyl sulphoxide (DMSO) removal with ozone microbubble (OMB) and millibubbles (OMLB) has been carried out to establish the effectiveness of OMB system. The TOC and DMSO removal profiles (Fig 5) at different pH depict the huge difference in the removal rates by OMB and OMLB system after 2.6 ks ozonation time. The better removal efficiency of DMSO as compared to TOC at all pH is related to the deficient $OH^{\cdot}$ to degrade the reaction intermediates produced during the ozonation of DMSO [32].

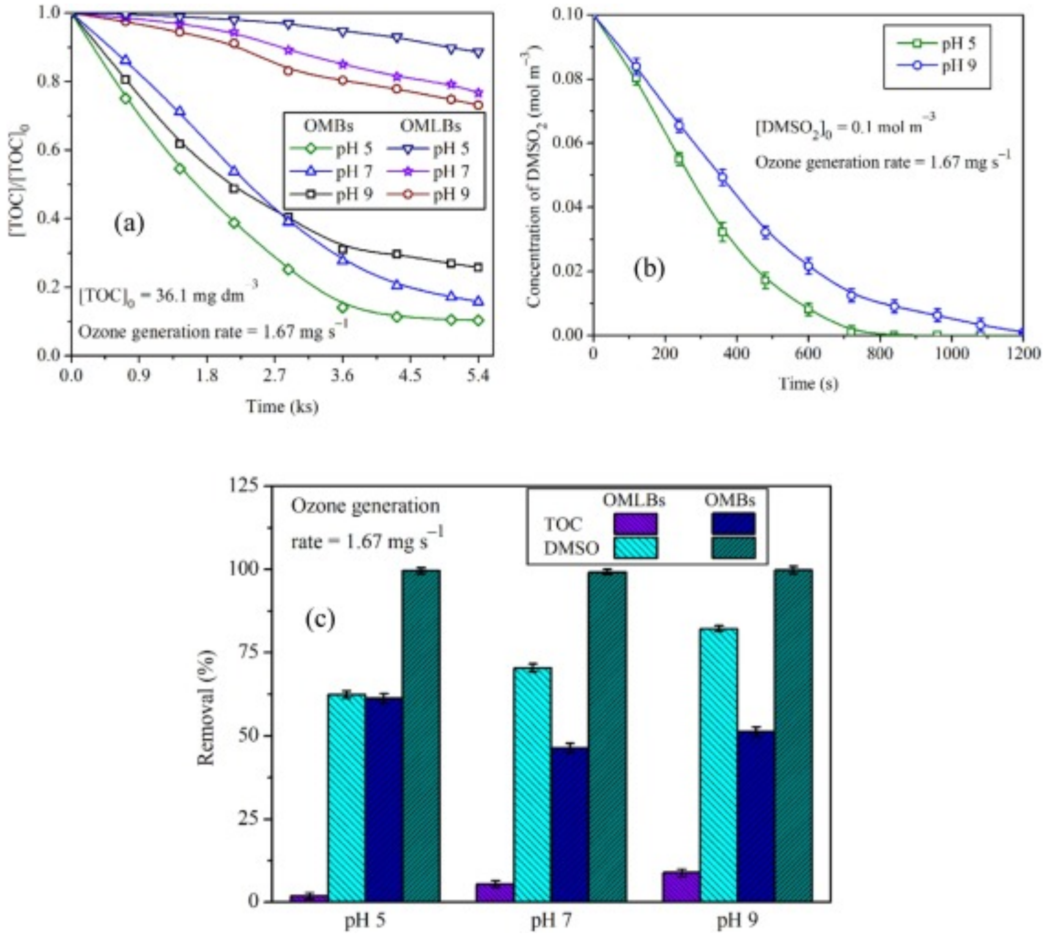

Figure 5: (a) TOC removal profiles at different pH, (b) oxidation of DMSO in water by the OMBs, and (c) comparison of the OMB and OMLB processes in terms of DMSO and TOC removal efficiencies at 2.16 ks of ozonation time [32].

A pilot scale ozone MB treatment with the possibility of scale up to large treatment plant is given by Ryskie et al. The continuous flow testing confirmed treatment efficiency as high as 99.1% at a flow rate of 1.1 L/min in removing ammonia-nitrogen from real mining effluents. However, the authors recommended testing of cyanide and thiocyanate presence in effluents prior to scale up as these contaminants decrease the removal efficiency with ozone MBs [33]. A successful demonstration of complete degradation and mineralisation of Butylated hydroxy toluene (BHT) pollutant in water by ozone MB is given by Achar et al. As compared to conventional ozonation, ozone MB produced 1.3-19 times enhancement in ozone gas mass transfer. The BHT removal rate was found to depend on the initial BHT concentration. A 60s treatment with ozone

MB resulted in complete removal of 0.34 and 0.45μM BHT however, 77% is removed when initial concentration is 0.90 μM (shown in figure 6). This was attributed to the production of additional metabolites produced during the treatment with higher initial BHT concentration which consumes $OH^{\cdot}$ radical [34].

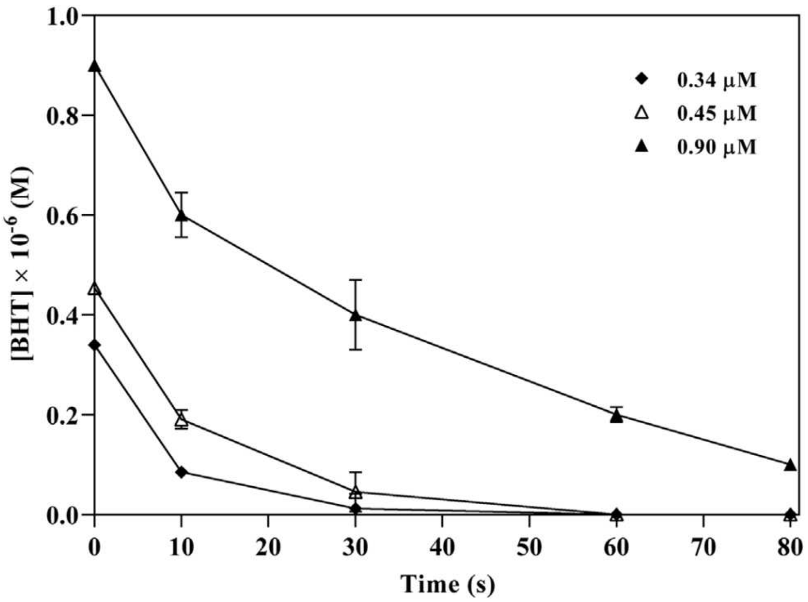

Figure 6: Effect of BHT concentrations on OMB treatment at pH 7 (mean ± SD, n = 3). Initial ozone concentration was 0.27 mM [34].

It has also been argued that the increase in the gas mass transfer was not the only reason for the speeding up of ozonation by microbubbles (REFs). One study emphasised on the increase in the concentration of ozone in the interfacial region for the enhancement of degradation of organic pollutant like phenol. Mathematical model-based simulation study further supported their claim by showing a steep ozone concentration gradient in the liquid film of microbubble as shown in figure 7 below [35].

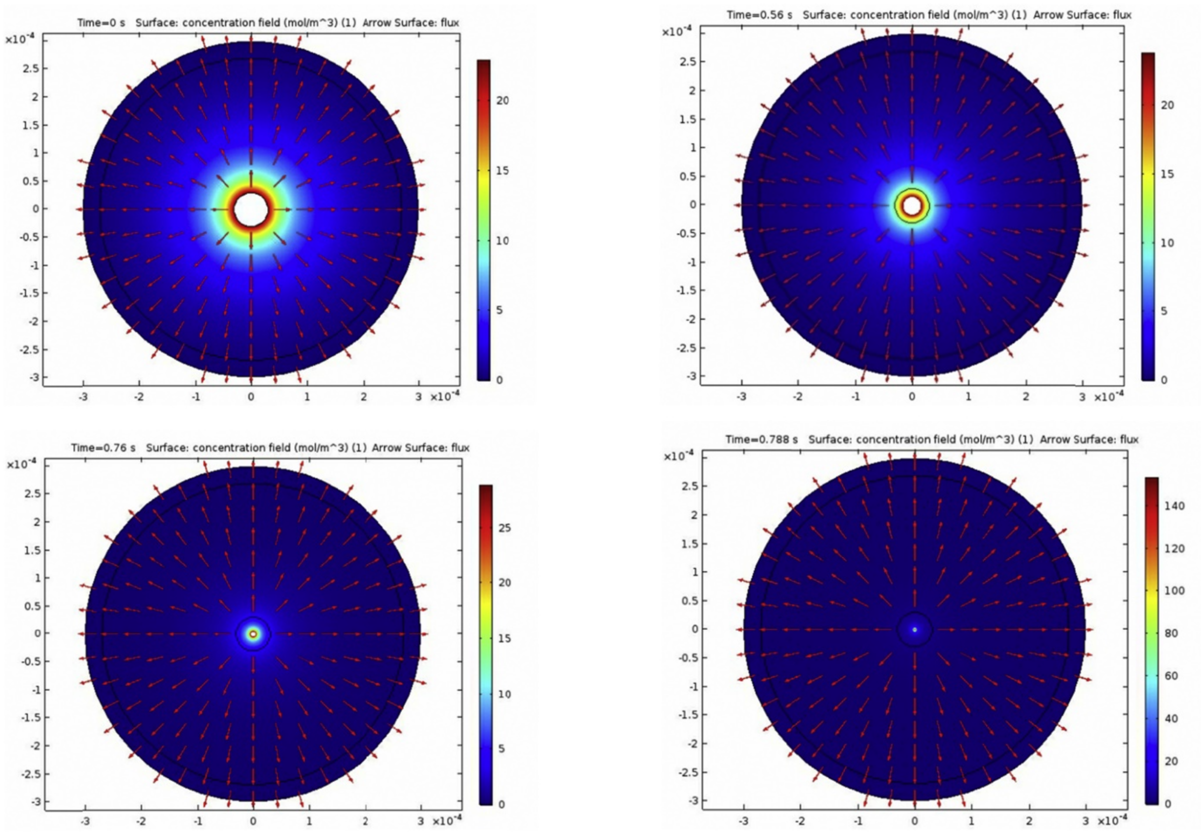

Figure 7: The change of bubble concentration in bubble surface during the bubble contraction process [35].

The hydroxyl ions accumulating on the microbubble gas-liquid interface is supposed to promote the self-decomposition of ozone and formation of ($OH^{\cdot}$) hydroxyl radicals. This increased generation of $OH^{\cdot}$ with microbubbles contribute to enhanced degradation of organic contaminant like Atrazine [36]. In a yet another study, 94% removal of diethyl phthalate (DEP) at pH of 7 and complete mineralisation was achieved at higher pH from ozone microbubbles. $OH^{\cdot}$ was believed to be the dominant reactive species responsible for oxidation of DEP micropollutant as compared to the direct oxidation with molecular ozone [37]. MBT can enhance the bioremediation in ground water. The applicability of hydrogen microbubbles to enhance the process of hydrogenotrophic denitrification (HD) and removal of nitrogen without leaving behind residual carbon is portrayed. Hydrogen microbubble reactor, performed better as compared to millibubble reactor in the HD system, achieving as high as 99% nitrogen removal efficiency as compared to less than 10% removal efficiency in the latter. This technology also afforded to reduce the energy consumption by increasing the hydrogen utilisation efficiency to 50% for biological consumption and hydrogen effectiveness to reach 1.21 g-N/g-H_2 [38].

In case of complex effluents, a combination or hybrid treatment approaches have been utilised for achieving enhanced removal efficacy. A case study of real industrial effluent treatment with a combination of HC and oxidants is presented. The extent of reduction in COD value by combination of HC and H_2O_2 (40%) is found to be significantly higher than the HC alone (7.9%) [39]. Microbubble ozonation have been effective as a pre-treatment in peat water treatment plant to remove carcinogenic disinfection by-products (DBPs) like trihalomethanes (THMs) and haloacetic acids (HAAs) formed during chlorination. The microbubble pre-ozonation treatment at pH 7 for 30 min decreased the concentration of THM4 to 33.73 ± 0.40 μg/L and that of HAA5 to 49.89 ± 0.09 μg/L acceptable as per USEPA standard [40].

The feasibility of microbubble ozonation and the presence of humic acids for abatement of pharmaceutical compound in feed water is systematically investigated. The degradation rate of pharmaceutical compound was enhanced with the higher solubilisation rate of O_3 and $OH^{\cdot}$ and enhanced gas mass transfer, which is related to the smaller size of microbubbles. The humic acid and temperature were found to have inhibitory and facilitatory effect, respectively, on the degradation efficiency [41].

A catalytic exhibition of microbubble ozonation for simulated printing and dyeing waste-water (methylene blue) is reported by Nkudede et al. A drastic increase in COD removal efficiency of up to 93.5% and fast decolourisation within 10 minutes time is recorded at high pH [42]. To summarise, the implementation of MBT is intended to be an efficient and eco-friendly approach to cut down on the chemical/oxidant dosage owing to excellent mass-transfer as compared to conventional ozonation.

Table 1: Summary of microbubble application

MBG/Bubble property	Water source	Target pollutant	Result	Reference
Pressurised dissolution/Decompression	Synthetic initial DEP concentration 0.18 mol m^{-3}	Diethyl phthalate (DEP)	• At pH 7, 97% of TOC removal efficiency achieved • Complete mineralisation at higher pH	Jabesa et al. [37]
200 L Pilot scale Ozone microbubble	Synthetic effluent and real mining effluent	Ammonia nitrogen	• Removal efficiency of NH_3-N with ozone MB was more at pH 9 than at pH 11 in batch and continuous flow testing	Ryskie et al. [33]
Ozone microbubble 14.64±2.08 µm diameter	Synthetic Initial BHT stock solution 0.90µM	Butylated hydroxy toluene (BHT)	• 1.3-19 fold improvement in ozone mass transfer • $OH^{\cdot}$ being dominant (82%) oxidation species	Achar et al. [34]
Hydrogen microbubbles (25 ±13 µm) from microbubble generator with oscillating mesh	Synthetic ground water	Nitrogen	• Microbubble enhanced biodegradation process achieving 99% nitrogen removal efficiency	Eamrat et al. [38]
Ozone microbubbles with average size of 30 µm in semi-batch mode	Synthetic water	Phenol Nitrobenzene	• Compared with 40 mg/L of ozone needed in conventional bubbling only 10 mg/L required for 80% removal of phenol with microbubble • Improved ozone utilisation efficiency	Wu et al. [39]
Ozone microbubble using rotating flow/vortex diffuser	Peat water from Riau Peatland, Indonesia	Haloacetic acids (HAAs) Trihalomethanes (THMs)	• Primary treatments reduce THM but not HAA • Microbubble pre-ozonation reduced HAA in all pH	Qadafi et al. [40]

			conditions except alkaline pH	
Ozone microbubbles by pressurised dissolution	Synthetic water	Atrazine	• Microbubble ozonation enhanced degradation at all pH • Self-decomposition of ozone supported by accumulation of OH^- on gas-liquid interface	Liu et al. [36]
Ozone microbubble	Simulated printing and dyeing waste water	Methylene Blue	• COD removal affected by pH and ozone dosage • Ozone dosage of 0.4 L/min and 0.5 L/min recorded more than 94% COD degradation	Nkudede et al. [42]

4. Disinfection

Chlorine is one of the most used chemical disinfectant for treating drinking water. However, carcinogenic by-products of chlorine disinfection and its ineffectiveness in destroying hidden microorganism in bio-films are the major cause of concern. Apart from this, ultrasonication is known to be active in decomposing microorganisms by acoustic cavitation effect. The high energy shock waves produced by the gas bubble collapse forming reactive oxygen species that can help in disintegrating the bacteria. But this technique is not of practical importance because of the allied cost factor. However, hydrodynamic cavitation producing similar effects as acoustic cavitation can be a low-cost viable water treatment technology to be scaled up to an industrial level. Reflection from disinfection experimentation by air or ozone MB suggests faster *E. coli* inactivation kinetics, reduced reactor size and lesser ozone dose as compared to conventional ozonation system [43]. In a yet another similar study, a novel ozonation system based on microbubble technology is suggested to overcome the lower utilisation efficiency associated with conventional ozonation disinfection. A reduction in operating cost was confirmed with the enhanced log reduction of *Bacillus subtilis* spores for the same ozone dosage by MBT with high inlet ozone concentration [44].

A meaningful insight into the disinfection mechanism of both gram-positive and gram-negative bacteria are provided by Jain et al. based on rotating flow cavitation device. The study demonstrated practically complete removal (99%) of *E. coli* with 1 h of cavitation treatment at 0.5 bar pressure drop. In comparison, a lower disinfection rate of 60% (seen in figure 8) was achieved for gram-positive bacteria *S. aureus* under similar conditions. This discrepancy in deactivation is eliminated at higher pressures (pressure drop of 1 and 2 bar) where elimination rate of *S. aureus* also reaches to a value close to 98% [45].

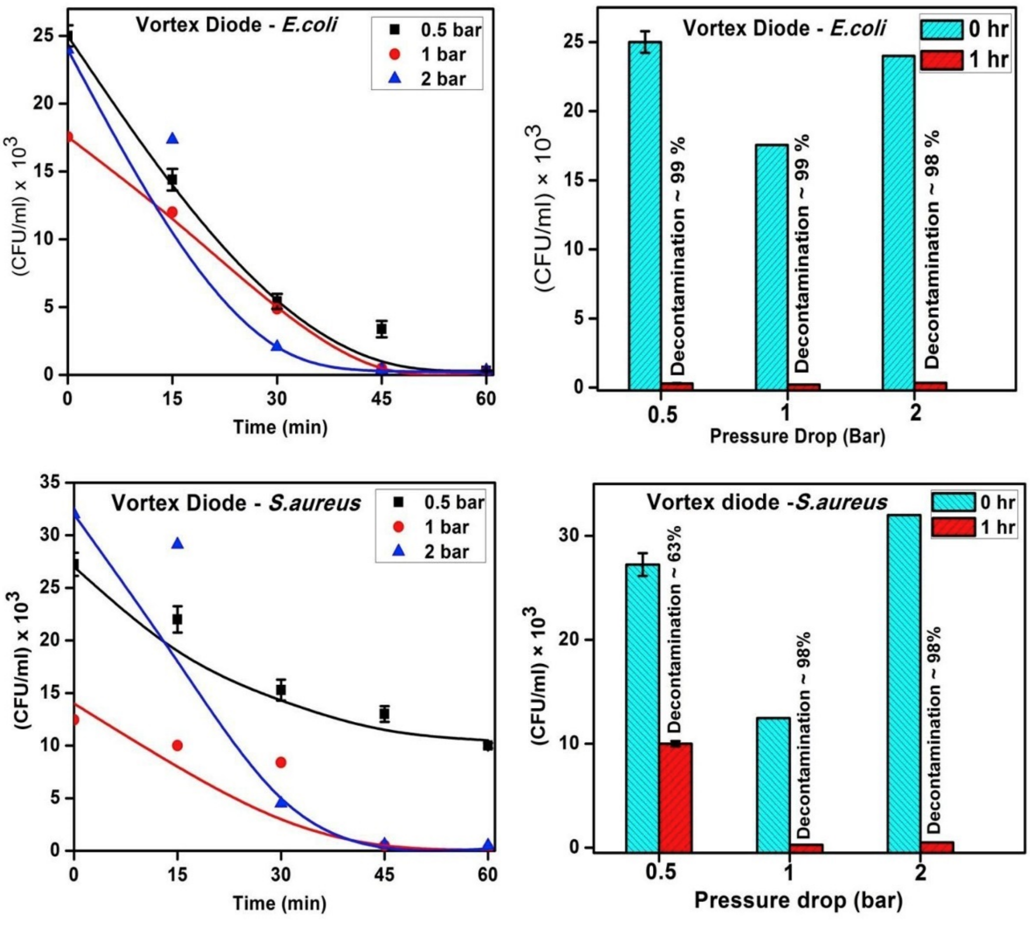

Figure 8: Effect of pressure: Disinfection of *E. coli* and *S. aureus* by vortex diode [45]

A faster and greater disinfection was suggested on *Fusarium oxysporum* f. sp. *melonis* Spores by ozone microbubbles (OMB) compared to their larger counterpart. Transmission electron microscopy (TEM) images (Figure 9) depicted the appearance of wave-like deformation of cell membrane and a gap between the cell wall and cytoplasm of f. sp. *melonis*. Spores treated for 180 s with OMB. It was considered that hydroxyl radicals produced of OMB induced leakage and coagulation of intracellular component following on to lysis of spore and final cell death [47]. Another case study of domestic waste-water treatment in Carhuaz, Peru highlighted the role of air-ozone micro-nano bubble in reducing faecal and total coliform counts. The application of micro-nano bubble treatment on the waste water with initial faecal coliform count of 130,000 CFU/100 mL achieved a reduction of up to 100 CFU/100 mL (99.92%) [48].

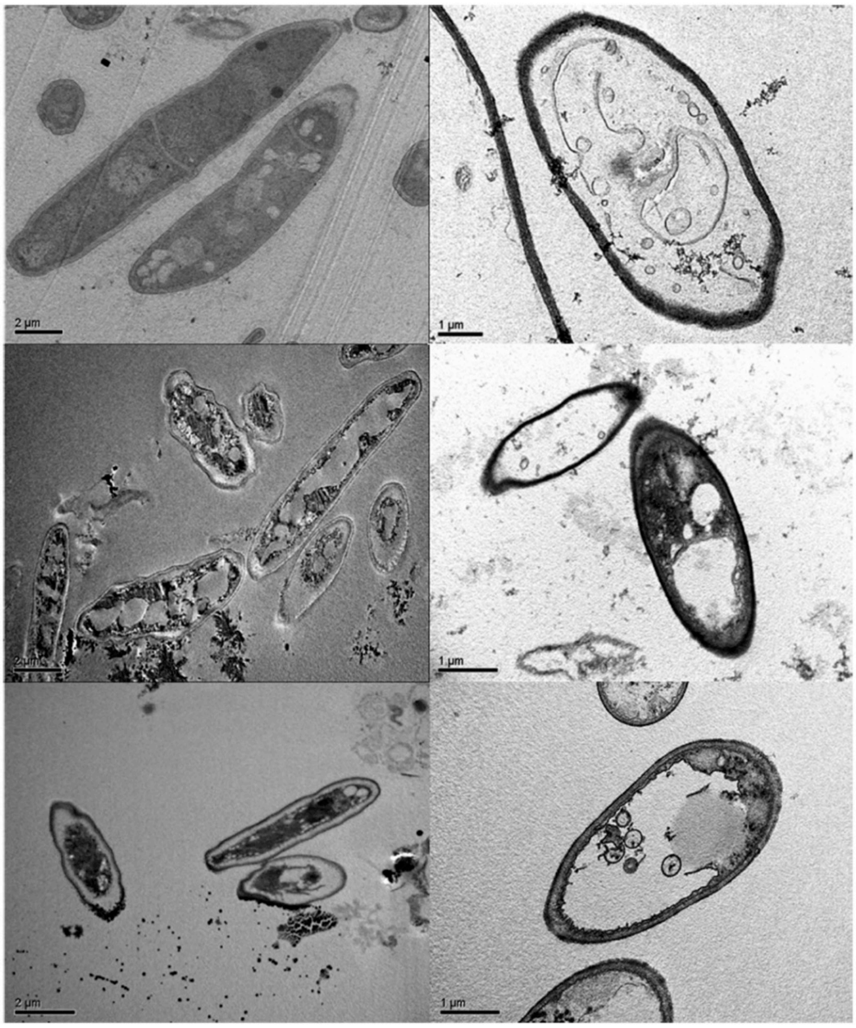

Figure 9: TEM images of *F. oxysporum* f. sp. *Melonis* spores. Top: Non-treated, Middle: After treatment with O3MMB for 180 s, Bottom: After treatment with O3MB for 180 s [47].

Harmful algal bloom (HAB) consisting of algae and cyanobacteria because of their more frequent occurrence in water bodies like lakes and rivers is becoming an epidemic. The presence of these organisms in water results in decrease of dissolved oxygen content, penetration of light and the exchange of other gases which is a doom for the aquatic life. Though certain chemical treatments are prevalent for remediation from HABs. However, these treatments disrupt the ecological balance of the environment. Again, some physical methods have also been employed in the past. But physical methods are ineffective against the toxins released as a result of algal cell damage. This sizeable challenge of removing algae as well as the generated toxins is very well taken up by the working miniatures called MBs [49]. Hydrodynamic cavitation producing micro and nano bubbles is believed to be a feasible approach suggested for eliminating surface blooms and associated toxins. A pilot study on Lake Neatahwanta, New York, United States with field blooms suggested a 50% reduction in cyanobacteria chlorophyll after 72 hr of microbubble treatment by hydrodynamic cavitation. The higher decline percentage of 80% was reported with additional treatment with peroxides [50]. A study by Thomas et al. have shown the potential of MBs induced by small, inexpensive nozzle and water circulation system to selectively destroy the harmful cyanobacterial bloom while leaving behind the beneficial algae that lacks gas vacuoles. This study demonstrated a dual phase treatment in which the strong turbulence/shear produced by hydrodynamic cavitation damages the cyanobacterial membrane or cell wall of algae while the free radicals generated in the MB process oxidised the cyanotoxins released from the disrupted cell wall. The efficacy of the treatment of algae was found to be dependent on inlet pressure of nozzles, treatment time and type and concentration of algae. MB treatment was found to be more effective for removing vacuolated algae than vacuole-negative algae. This variability in treatment was attributed to the different cell structure and the presence of cellulose in cell wall and the absences of gas vacuole which is known to initiate the secondary production of free radicals [51].

Cost and Energy Consumption Analysis

There is an interesting back-and-forth relationship between treatment efficiency, cost and energy consumption in the selection of technology for large-scale application of microbubble technology. Each of these parameters is to be discussed in tandem in deciding the feasibility of the process. Though, laboratory scale set up does not give an accurate idea of energetics of upscaling, but it can provide a suggestion on the probable trend. An economic efficiency study was carried out by Andinet et al. to assess the running cost benefit of supplying air or oxygen microbubble for aeration of 20 minutes. The air was found to be a more economical option at lower pressure conditions, whereas the cost of oxygen gas per unit dissolution decreased at higher pressure conditions. So, a more economical means of pressurised dissolution with a reduced gas flow rate is suggested to improve the natural water bottom area by oxygen MB [52]. An alleviation in membrane fouling is seen in the vacuum membrane distillation desalination process by MB aeration. Inclusion of MB aeration is merely contributing 2.8 ±0.3% of the total energy consumption of the process at different pump pressures. MB aeration is shown to be evidently effective in improving the specific energy consumption, specifically while operating at increased pump pressures. Further investigation is recommended to yield a cleaner method of treatment without chemicals and to improve the specific energy consumption in all [53].

A low-cost, efficient mineralisation of total petroleum hydrocarbon (TPH) is achieved with microbubble ozonation by synergetic utilising of oxidation and flotation phenomena. A 70.9%

reduction in TPH was observed in 120 min of microbubble treatment with 0.27 gO_3/g total solid (TS). The total operation fee for this innovative process, including labour fee, power and additive consumption at the stated ozone dosage was calculated to be 28.7 CNY/gTPH. This is very less when compared to other conventional oxidation processes [54]. Again, a positive effect of HC in hybrid treatment is evident in a dye degradation experiment. The degradation of methyl orange dye reached as high as 90% by integrating HC with H_2O_2 and metal. The author reported that the energy required and hence the operating cost was 21 times smaller with this favourable integration of HC. The total operation cost to treat 1 m^3 of waste-water containing 5 ppm of methyl orange was estimated to be 1985, 728 and 93 euro/m^3 for HC only, HC + H_2O_2, HC+H_2O_2+ metal system, respectively [55].

Table 2: Cavitational yield and operational cost based on energy for different treatment approaches

Contaminant	Treatment	Extent of degradation (%) COD/TOC	Cavitational yield (mg/J)	Energy required (KWh)	Total operational cost based on energy	Reference
Total petroleum hydrocarbon (TPH)	O_3 Microbubble (0.27 g O_3/gTS)	70.9			28.7(CNY/g TPH)	Sun et al. [54]
Dimethoate	HC(slit venturi) HC+UV HC+H_2O_2 HC+Fenton	14.63 30.8 72.5 100	2.95×10^{-5} 4.24×10^{-5} 14.6×10^{-5} 20.1×10^{-5}	0.094 0.065 0.019 0.013	0.85(Rs/L) 0.59 0.17 0.12	Thanekar et al. [56]
Methyl Orange	HC (venturi tube) HC + H_2O_2 HC+H_2O_2+metal	53.69 53.11 90	4.90×10^{-7} 5.47×10^{-7} 1.2×10^{-6}	1.44 1.56 0.20	1985 euro/m^3 728 93	Innocenzi et al. [55]
2,4,6-Triamino-1,3,5-trinitrobenzene (TATB)	HC (orifice) ClO_2 HC+ClO_2	13.59 20.38 65.9	5.52×10^{-3} 0 26.78×10^{-3}	0.2721 0 0.0561	0.1361(CNY/L) 0.045 0.0731	Wang et al. [58]
Complex industrial waste-water	HC+O_2+Fenton(orifice)	63	47×10^{-3}	2084.2 kWh/m^3	398 US$/$m^3$	Joshi et al. [57]

Thanekar et al. have compared the cavitation yield and operation cost of different type of treatments for degradation of dimethoate pollutant based on power consumption. The HC treatment alone was found to cost 0.85 Rs/L with a cavitation yield of 2.95×10^{-5} mg/J whereas the combined treatment approach of HC+Fenton resulted in total operational cost of 0.12 Rs/L with a cavitation yield of 20.1×10^{-5} mg/J [56]. In a similar study, synergistic combination of HC and AOP (advanced oxidation process) was applied for the treatment of industrial waste water at a pilot scale (70 L capacity). Energy efficacy and cost analysis revealed a maximum COD removal efficiency of 63% in 180 min of tandem treatment estimating a total combined cost of 398 US$/m^3 for electricity and additive [57].

An economical and suitable large-scale commercial operation of HC/chlorine dioxide (ClO_2) composite process is proposed for treatment of explosive containing waste-water. The cost calculations and cavitation rate comparison suggest the reduced treatment cost of 0.0731 CNY/L for HC/ClO_2 treatment as compared 0.1361 CNY/L for single cavitation along with an enhanced cavitation rate of 26.78×10^{-3} mg/J for the former [58]. Table 2 shows the cavitation yield and operational costs of different treatment approaches based on energy requirement. So, it could be concluded that combination treatment approaches help in cutting down of the cost of the treatment.

Mukherjee et al. have reported an economically feasible process to recycle the grey water from a real-life kitchen stream. The HC treatment for 120 min resulted in a 25% and 15% reduction in COD and TOC value, respectively, which is thought to be higher than that produced using H_2O_2 and ozonation treatment. However, HC treatment efficiency was not enough to be adopted for real field study. A synergistic combination of HC with other AOPs like H_2O_2 and ozone increased % COD and TOC reduction to 98.25 and 76.26%, respectively which was acceptable for large scale applications. Energy consumption and time needed for treatment is considered in tandem for technological integration and total cost in terms of operation cost and additive cost is summarised in Table 3. Energetic and cost calculations support HC $+H_2O_2$ (5 g/L) as the most feasible treatment since the cost of this process is the least with comparable cavitation yield as HC $+H_2O_2$ $+O_3$ [59].

Table 3: Comparison of cavitational yield and cost effectiveness of various processes.

Process	**Time required for 75% COD reduction (min)**	**Cavitational yield (mg/J)**	**Energy consumption for 75% COD reduction (kwh/m^3)**	**Cost (\$/m^3)**	**Additive cost (\$/m^3)**	**Total cost (\$/m^3)**
HC	360	0.005	32.05	2.24	0	2.24
HC+H_2O_2(2 g/L)	180	0.013	16.02	1.12	0.23	1.35
HC+H_2O_2(5 g/L)	50]	0.04	4.27	0.29	0.58	0.88
HC+H_2O_2(7 g/L)	55	0.03	4.80	0.33	0.82	1.16
HC+O_3(3 g/h)	75	0.19	29.17	2.04	0	2.04
HC+H_2O_2(5 g/L)+O_3(3 g/h)	30	0.34	11.67	0.81	0.58	1.40

A novel effort has been put forward by Ranade et al. to present a multi-scale modelling of HC devices for degradation of pollutants at four different scales (scaling of approx. 200 times of original capacity). The extended per-pass modelling described a decrease in degradation rate with increase in scale up until a finite limiting value is reached [60].

Any specific conclusions cannot be drawn as to which hybrid technology is finest as such a judgement would be vague. A hybrid technology which is best suited for a particular type or nature of pollutant may not be as effective for other type of pollutants. So, more efforts are to be directed to this end to further explore the sensible decision-making factors from the aspect of degradation performance, operating costs and energy savings in choosing the best hybrid technology to expand to industrial scale.

Challenges in implementation at Industrial scale

- Although much research and laboratory studies have shown immense potential of MBT in water treatment, the knowledge regarding the upscaling of methods and application of MB at the industrial level has been limited. More field size studies for extended periods is the need of the hour to evaluate the efficiency of the process and to get the working knowledge and experience.
- Simulation studies on MBs generation, and reaction with contaminants are limited. Simulation aids in understanding about the microbubble assisted processes without carrying out the experiments. Simulations can be done before upscaling of the process and can significantly cut the design and operation cost if used for optimisation of the process.
- Though theoretical models have been developed to study the mass transfer and stability of MBs, but these models should be extended to comprehend the interaction of MBs with microorganisms and organic contaminants taking into account the possible by-products of their reaction. Such studies would be beneficial for the optimal designing of novel systems retrofitted with MBT.
- Majority of published literature has concentrated on mass-transfer efficiency of MBT but very less importance is being given to the heat transfer. MBG during their course of operation are certainly generating heat (rise of the temperature of the medium) which could be prominent to several reaction mechanisms and can be further explored.

Conclusions

We identified the significant relevance of microbubbles in water treatment technology. Broad application and the prospect of MBT in water treatment is with reference to its enhanced gas solubility leading to efficient gas-liquid mass transfer and generation of oxidative free radicals. The application of microbubbles in disinfection entails from its dual phase treatment with mechanical shear produced during bubble collapse and oxidisation from free radical production. The results of implementation of MBT in aeration and ozonation overcoming the limitation of low gas utilisation efficiency and higher oxidant dosages of conventional methods have been predominantly encouraging.

Many studies have stressed on the role of MBT in downsizing of the treatment plant and reduced operation cost of WWTP along with increased contaminant removal efficacy. The cost and energy consumption analysis are indicative of the potency of MBT to replace the existing expensive processes in water treatment. However, more industrial scale studies are recommended for bridging the gaps between laboratory scale research and implementation at larger scales. In order to achieve the vision of the real-world application of MBT in waste-water treatment, a comprehensive assessment of degradation performance, energy efficiency and economic evaluation is to be carried out on model waste-water with different type and concentration of pollutants and their treatment time. The economic evaluation should reflect a synergistic combination of MBG construction and maintenance cost, chemical dosage expenditure, and power consumption through the life-time of treatment system.

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