


Contemporary application of microbubble technology in water treatment

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ABSTRACT

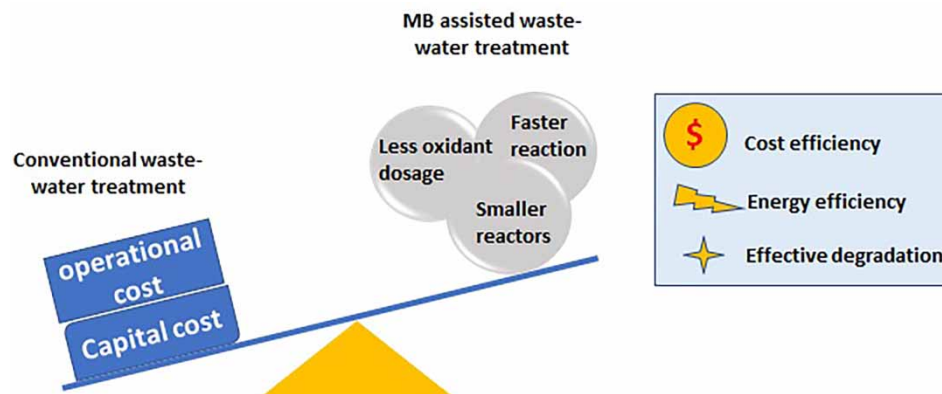
Microbubble (MB) technology constitutes a suite of promising low-cost technologies with potential applications in various sectors. Microbubbles (MBs) are tiny gas bubbles with diameters in the micrometre range of 10–100 μm . Along with their small size, they share special characteristics like slow buoyancy, large gas–liquid interfacial area and high mass-transfer efficiency. Initially, the review examines the key dissimilarities among the different types of microbubble generators (MBG) towards economic large-scale production of MBs. The applications of MBs to explore their effectiveness at different stages of wastewater treatment extending from aeration, separation/ flotation, ozonation, disinfection and other processes are investigated. A summary of the recent advances of MBs in real and synthetic wastewater treatment, existing research gaps, and limitations in upscaling of the technology, conclusion and future recommendations is detailed. A critical analysis of the energetics and treatment cost of combined approaches of MB technology with other advanced oxidation processes (AOPs) is carried out highlighting the potential applicability of hybrid technology in large-scale wastewater treatment.

Key words: aeration, cavitation, microbubble, water treatment

HIGHLIGHTS

- Extensive comparison of microbubble technology with existing water treatment processes (WTPs).
- Dual phase treatment with microbubbles in water disinfection.
- Microbubble technology overcomes the limitation of low gas utilisation efficiency of conventional methods.
- Cost and energy consumption analysis indicates potency of microbubble technology to replace expensive WTPs.
- Opportunities for downsizing of water treatment plant.

GRAPHICAL ABSTRACT



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1. INTRODUCTION

With the current surge in global environmental challenges, there is a shift towards more eco-friendly and sustainable technologies. Microbubble (MB) technology has recently emerged as a viable option in water treatment. The increasing demand for water following the dramatic population and economic growth has led to more efforts directed towards wastewater treatment. Conventional water treatment technologies like flocculation, sedimentation, filtration, UV degradation have been shown to reduce water pollutants, but these technologies generally require a series of steps which increases the overall cost of the process (Rajasulochana & Preethy 2016). So, researchers for quite long have been in the pursuit of a more effective and cost-efficient technology.

Air, oxygen, and ozone microbubbles (MBs) have shown great potential for reducing the running cost and economising the wastewater treatment plants (WWTP) (Yamasaki *et al.* 2009). Micro- and nanobubbles have been reported to be effective in the processes of aeration, flotation and disinfection (Jyoti & Pandit 2001; Liu *et al.* 2016). Microbubbles are tiny bubbles with diameters in the range of 10–100 μm (Basso *et al.* 2018). The shared characteristics of these MBs, which make them unique is their longer residence time, slow buoyancy, self-pressuring effect, and large gas-liquid interfacial area as compared to the conventional macrobubbles (Hernandez-Alvarado *et al.* 2017).

MBs consist 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 the Young–Laplace equation (Takahashi *et al.* 2003). Bubbles smaller than the critical radius have low buoyancy forces and tend to slowly diffuse the gas present within and shrink while ascending slowly (Takahashi *et al.* 2003). Such bubbles finally collapse underneath the liquid surface, producing reactive free radicals. Conventional macrobubbles on the other hand, during coalescence develop sizes larger than the critical radius, rise-up quickly and explode on the liquid surface (Agarwal *et al.* 2011). Figure 1 shows the time-conditioned shrinkage mechanism of micro- and macrobubbles. The presence of MBs is known to influence the physico-chemical and mechanical properties of the bulk liquid. This change in liquid properties has been shown to be beneficial for further chemical reactions like oxidation.

2. MB GENERATION

The formation of bubbles in the water 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 cavitation. The driving mechanism of cavitation is a reduction in the static pressure of liquid below its vapour pressure, leading to vapour/gas-filled cavities in the liquid. The cavitation is unlike the bubble formation in boiling where the process is primarily driven by temperature change.

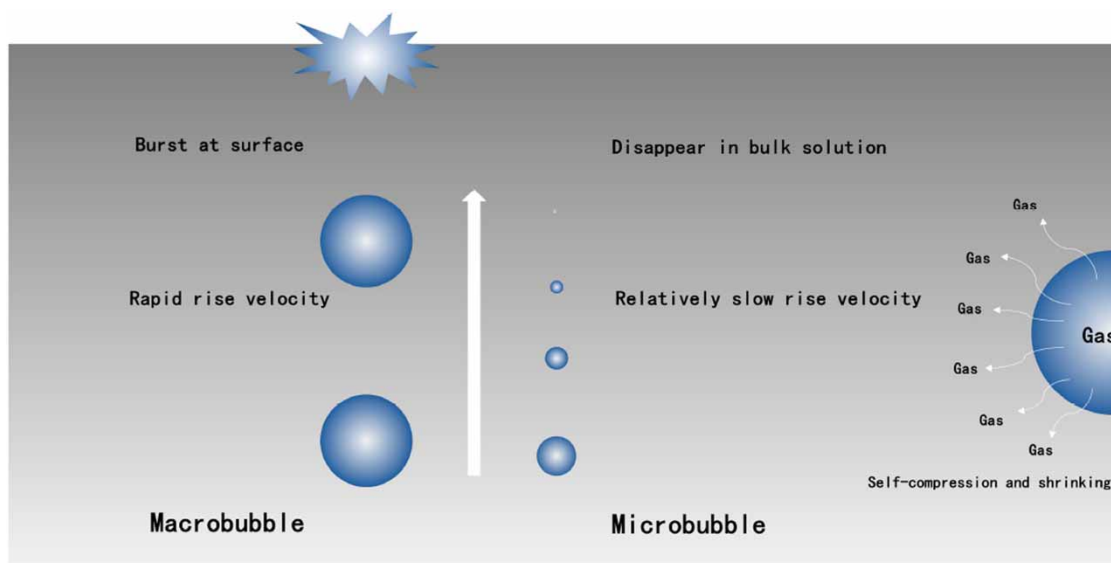


Figure 1 | Schematic diagram of macrobubbles and microbubbles (Xiong *et al.* 2019).

The microbubble generator (MBG) is chosen depending upon the application requirement, whether it is in production on a laboratory scale, for research purposes, actual field trials or at an industrial scale. The capacity and concentration of MBs production required for the treatment and the cost of treatment also decides the MBG employed.

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

1. Hydrodynamic cavitation (HC) – 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 costs are higher for this type when scaled at the industry level.
3. Particle cavitation – Particle cavitation is caused by a beam of elementary particles 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 wastewater treatment (Mancuso *et al.* 2020). Hydrodynamic and acoustic cavitation has shown to bring about the desired physico-chemical changes in the bulk liquid (Temesgen *et al.* 2017). When it comes to pilot- and large-scale processing, HC technology remains the first choice for MB formation in water treatment owing to high energy consumption and limited cavitation zone in the acoustic cavitation technique (Badve *et al.* 2013).

In water treatment processes (WTPs), 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, the MB is generated by the passage of air and water simultaneously through the Venturi tube. Pressurised fluid enters through the inlet and the velocity of the fluid increases at the cost of a decrease in static pressure in the narrow cross-section throat. This accelerated fluid generates cavitation and sucks in the gas creating microbubbles (Wilson *et al.* 2021). This type offers the benefit of lesser pump power, and compacted size.
- Orifice type generator – In this method, air is absorbed into the vacuum created by the movement of fluid through holes in the orifice plate. Unlike the Venturi, the orifice type generator generates intense cavitation conditions owing to immediate contraction and divergence sections.
- 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 (Parmar & Majumder 2013).

Compared with acoustic cavitation, HC appears to be the more practical and economical way to generate MBs at pilot and industrial scale and at higher rates using a simple centrifugal pump along with a flow confiner like Venturi tube, orifice plate, and throttling valves (Wang *et al.* 2021). For more information on the MB generation methods and bubble characterisation techniques, one can refer to recent articles (Sakr *et al.* 2021).

Some studies have reported the generation of significant reactive oxygen species or free radicals by HC as the MBs collapse. For example, Khuntia *et al.* (2015) 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 radicals at acidic pH than that in the alkaline medium. Zheng *et al.* (2015a, 2015b) ascribed the better degradation of biorefractory organic compounds by MB ozonation to the greater amount of unselective hydroxyl radicals produced using fluorescence detection. However, some other studies highlighted that the treatment process with MBs is most likely to be thermal based rather than free radical based (Bandala & Rodriguez-Narvaez 2019). There is debate over the mechanism behind the organic water pollutant degradation. Still, all these studies identified the feasibility of the HC in water treatment. Bandala & Rodriguez-Narvaez (2019) claimed that HC alone or coupled with other advanced oxidation techniques is a promising practice to remove organic contaminants like azo dye and antibiotics entering the water cycle. The limitation of the most advanced oxidation techniques to completely remove the complex organic matter in the water has led to the surge in exploring the effectiveness of HC or its coupling with other advanced techniques.

The ever-increasing interest in MB technology 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). Although studies pertaining to

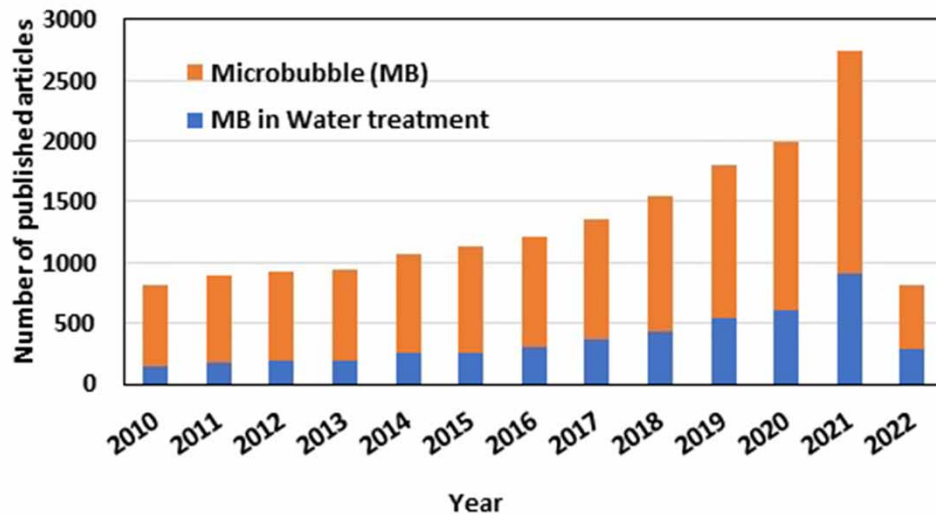


Figure 2 | Publications related to microbubbles and their application in water treatment over the years. Source: Elsevier.

microbubbles in bulk liquid have been followed over a long period but still ambiguity exists regarding the mechanism of its operation and real-world application of this technology (John *et al.* 2022a).

Previous reports have focussed on generation techniques, operational conditions employed, bubble size and distribution measurements, and characterisation techniques. However, limited efforts are directed towards studying the application gaps of MBs/NBs in water treatment technology. Few reports have 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 WTPs.

3. APPLICATION OF MICROBUBBLES IN WATER TREATMENT

3.1. Aeration

Environment friendly biological treatment methods have been the preferred treatment of organic wastewater. The metabolism of microorganisms is used in biological treatments like bio-activated sludge, biofilm, and membrane bioreactors to degrade 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 (Drewnowski *et al.* 2019). Microbubbles with long retention times and high gas mass-transfer efficiency are conducive to energy conservation and cost reduction in WWTP. Microbubbles make it possible to diffuse oxygen more effectively in aerobic wastewater treatment.

A novel aeration system was proposed for wastewater treatment employing an MBG 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 MBG and typical gas distributors. The specific power consumption of the MGBs 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 for MGBs as they aid in the downsizing of aeration tanks and reduce the residence period of wastewater (Terasaka *et al.* 2011).

Microbubble technology is believed to support biological aerobic wastewater treatment owing to its high oxygen mass-transfer rates, but few studies have exposed the negative effects of microbubble technology on the mixed liquor properties of activated sludge during microbubble aeration. The high shear force generated during MB generation was found to break-down sludge flocs, reducing the microbe population available for oxidation of organic matter (Liu *et al.* 2012a). To tackle this

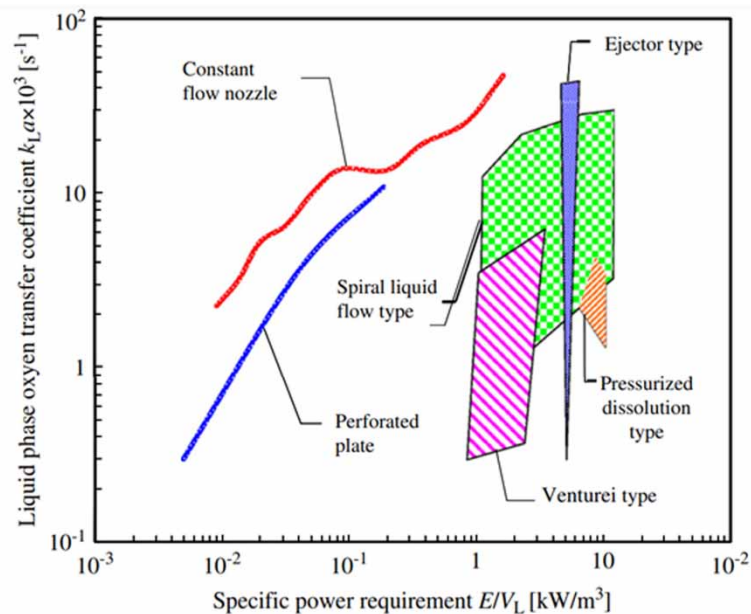


Figure 3 | Specific power requirement of various aerators to dissolve oxygen into water (Terasaka *et al.* 2011).

problem Budhijanto *et al.* (2015) proposed to combine MBGs with an attached growth aerobic system. The successful application of an MBG as the aerator with higher soluble chemical oxygen demand (SCOD) removal efficiency in the low gas flow rates is seen. They emphasised the careful design and selection of the MBG configuration and its relative position (if more than one MBG is used) in the reactor to avoid bubble coalescence. Lei *et al.* (2016) investigated the effect of microbubble aeration on the biofilm formation. The synthetic municipal wastewater was treated in a fixed bed biofilm reactor using microbubbles generated from the 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/(m² d) with a COD removal efficiency as high as 91.7%.

Much research has also been carried out to study the effect of aeration by MBs on the degradation of organic matter, seed germination and growth. A visible difference in aerial vegetative plant growth is seen with MBs and macrobubble aeration (Park & Kurata 2009). MB aeration has also been known to boost the growth of white shrimps *Litopenaeus vannamei* and biofloc (Lim *et al.* 2021). 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.

3.2. 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 wastewater treatment. The major steps leading to the separation are adsorption of gas bubbles on suspended particles forming bubble-particle aggregates. As formed aggregates are lighter, they rise to the water surface and can then be separated. Conventional air-flotation is limited by the bubble-particle interaction or collision and so can separate only a narrow particle size range. Several studies have shown that reducing the size of bubbles helped in increasing the overall efficiency of flotation separation by promoting bubble-particle collision and attachment (Moosai & Dawe 2003). Microbubble technology has been used to enhance the flotation process (Vion 2007). Experimental measurements have supported the application of smaller MBs to increase the mass-transfer coefficient in flotation and aeration process (Suwartha *et al.* 2020).

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. Dissolved air flotation (DAF) with MBs and NBs has been 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 emulsions. The conditioning of oil flocs with NBs 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 (Etchepare *et al.* 2017a). The same authors have extended the DAF technology for the removal of Fe^{3+} precipitates and $\text{Fe}(\text{OH})_3$ 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 (Etchepare *et al.* 2017b).

The separation of contaminants from oil-containing restaurant wastewater has been undertaken by the novel microbubble air flotation. It was reported that the highest oil removal efficiency was achieved when the microbubbles are of similar size as the 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 (Zheng *et al.* 2015a, 2015b). Liu *et al.* (2012b) reported a cost-effective and efficient ozone MB application in the coagulation-MB flotation process for the treatment of coke wastewater 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 (Liu *et al.* 2012b).

A superior microbial degradation and flotation separation has been suggested with microbubbles for wastewater treatment from the beverage industry. The study has supported the exploitation of bubble surface adsorption and flotation methods for the removal of high nitrogen-containing dissolved organic matter (DOM) from wastewater. As compared to macrobubbles, microbubbles have been shown to support the growth of aerobic bacteria and accelerate the degradation of DOM (Fukami *et al.* 2021).

Recently, an effective, and flotation simulation method has been proposed for the mixture optimisation between MBs and NBs in flotation arrangement. The laboratory-scale experimental study confirmed the deterioration effect of only NBs present to the water quality due to the long stagnation time of NB aggregate. However, the same study argued the improvement in removal efficiency of fine particles as small as 25 μm of nylon, polyvinyl chloride (PVC) and kaolin by hybrid bubbles (MB and NB). NBs help MBs to aggregate whereas MBs increase the removal efficiency of NB aggregates in the flotation process (Kim *et al.* 2020). It is envisaged that with the use of competitive microbubble technology in the flotation plants, the otherwise fraught field of flotation or physical separation is making a strong comeback as compared to large settling tanks set-up. The microbubble flotation technique is flourishing with clarification of lightly laden wastewater on the grounds of energy saving, compactness and ease of operation.

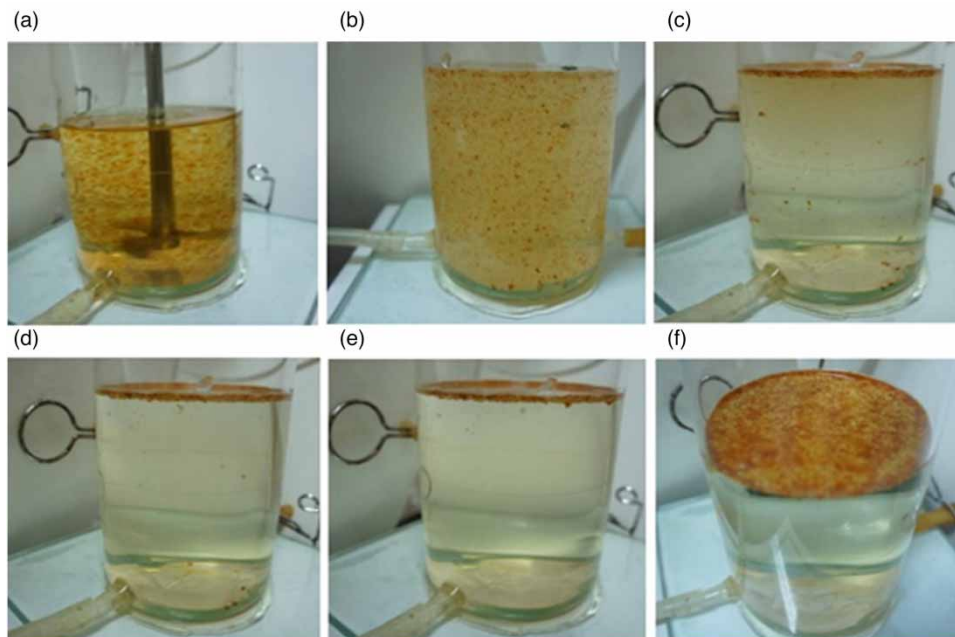


Figure 4 | Photographs of different stages of $\text{Fe}(\text{OH})_3$ precipitation, before and after flotation with MBs and NBs. (a) Precipitation; (b) injection of MBs and NBs; (c) flotation at 30 s; (d) flotation at 1 min; (e) flotation at 5 min; (f) floated precipitates and treated water. Conditions $[\text{Fe}^{3+}]_{\text{feed}} = 30 \text{ mg/L}$; $\text{pH} = 7$; saturation time = 30 min (Etchepare *et al.* 2017b).

3.3. Degradation of organic and inorganic pollutants

Microbubble technology can enhance the oxidation and remediation to degrade 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 WTPs like breakdown of refractory organic matters and disinfection. Ozonation is the preferred choice as compared to chlorination because of no chemical residuals after process completion, whereas chlorine leaves 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 the OMB system. The TOC and DMSO removal profiles (Figure 5) at different pH depict the huge difference in the removal rates by the OMB and OMLB systems 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⁻ to degrade the reaction intermediates produced during the ozonation of DMSO (Jabesa & Ghosh 2021).

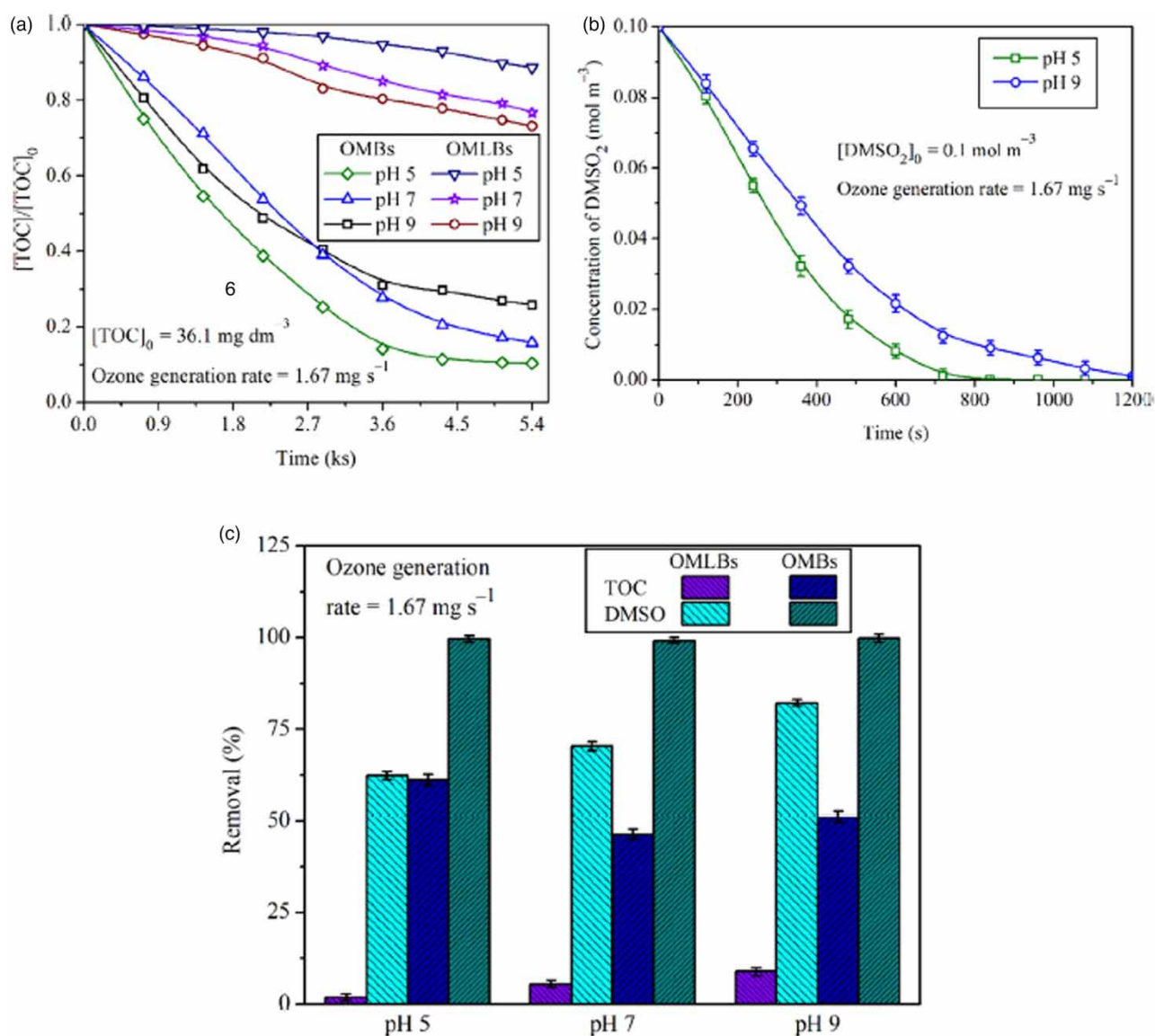


Figure 5 | (a) TOC removal profiles at different pH, (b) oxidation of DMSO in water by the OMBs, and (c) comparison of the OMB and OMLB processes in terms of DMSO and TOC removal efficiencies at 2.16 ks of ozonation time (Jabesa & Ghosh 2021).

A pilot-scale ozone MB treatment with the possibility of scale up to a large treatment plant is given by Ryskie *et al.* (2020). 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 decreased the removal efficiency with ozone MBs (Ryskie *et al.* 2020). A successful demonstration of complete degradation and mineralisation of butylated hydroxy toluene (BHT) pollutant in water by ozone MBs is given by Achar *et al.* (2020). As compared to conventional ozonation, ozone MBs produced a 1.3–19 times enhancement in ozone gas mass transfer. The BHT removal rate was found to depend on the initial BHT concentration. A 60 s treatment with ozone MB resulted in complete removal of 0.34 and 0.45 μM BHT, however, 77% is removed when the 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 concentrations which consumes OH^\cdot radicals (Achar *et al.* 2020).

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 (John *et al.* 2022b). One study emphasised the increase in the concentration of ozone in the interfacial region for the enhancement of degradation of organic pollutants like phenol. A mathematical model-based simulation study further supported their claim by showing a steep ozone concentration gradient in the liquid film of the MBs as shown in Figure 7 (Wu *et al.* 2019).

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 contributed to enhanced degradation of organic contaminant like Atrazine (Liu *et al.* 2020). 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 the DEP micropollutant as compared to the direct oxidation with molecular ozone (Jabesa & Ghosh 2016). Microbubble technology 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. The hydrogen microbubble reactor performed better as compared to a 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 (Eamrat *et al.* 2020).

For 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 the combination of HC and H_2O_2 (40%) was found to be significantly higher than the HC alone (7.9%) (Thanekar & Gogate 2019). Microbubble ozonation has been effective as a pre-treatment in a peat water treatment plant to remove carcinogenic disinfection by-products (DBPs) like trihalomethanes (THMs) and haloacetic acids (HAAs)

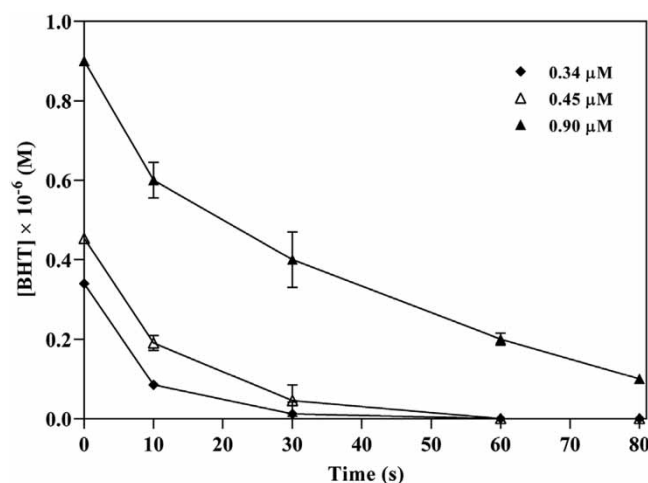


Figure 6 | Effect of BHT concentrations on OMB treatment at pH 7 (mean \pm SD, $n = 3$). Initial ozone concentration was 0.27 mM (Achar *et al.* 2020).

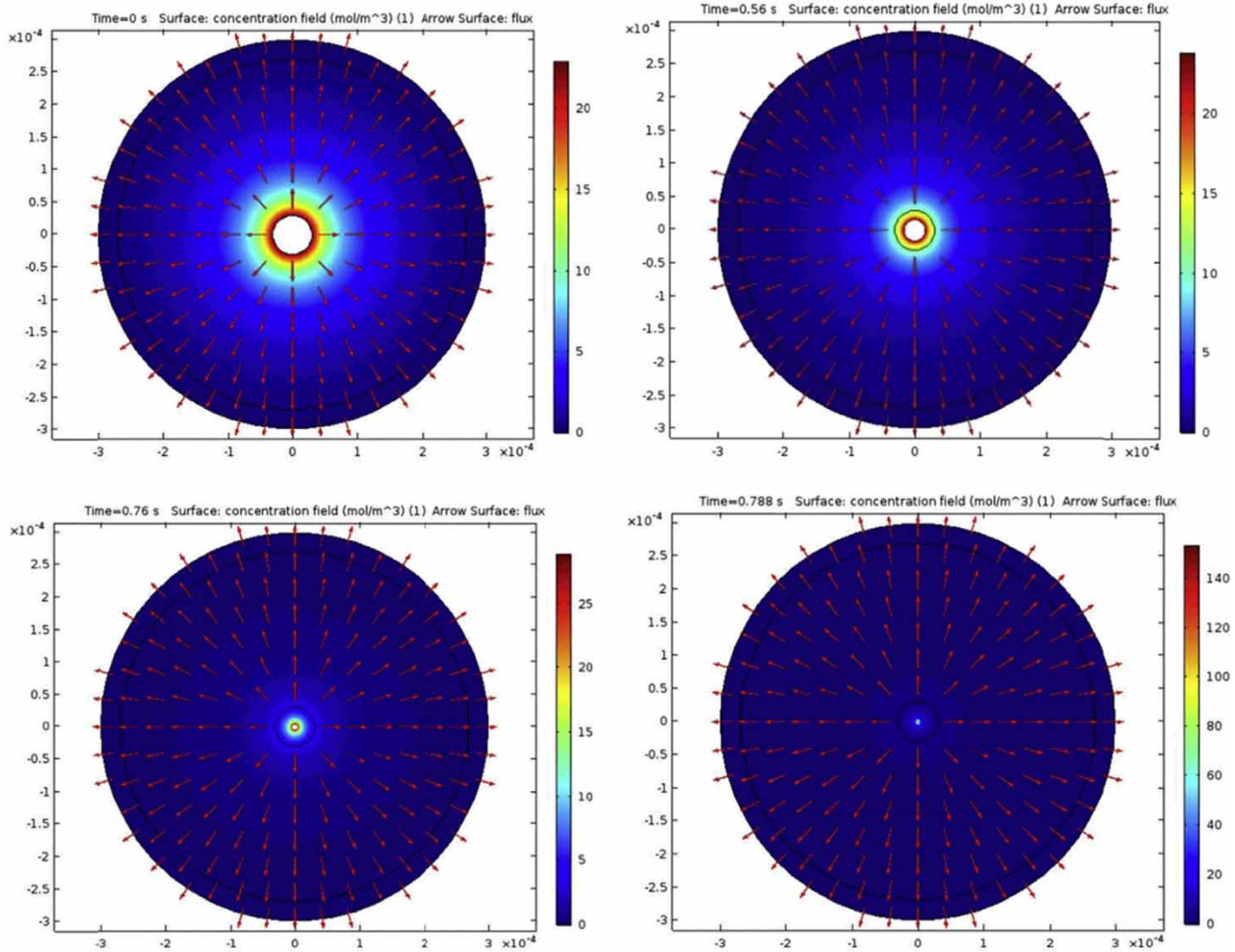


Figure 7 | The change in bubble concentration at the bubble surface during the bubble contraction process (Wu *et al.* 2019).

formed during chlorination. The microbubble pre-ozonation treatment at pH 7 for 30 min decreased the concentration of THM4 to 33.73 ± 0.40 $\mu\text{g/L}$ and that of HAA5 to 49.89 ± 0.09 $\mu\text{g/L}$ acceptable as per USEPA standard (Qadafi *et al.* 2020).

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 the pharmaceutical compound was enhanced with the higher solubilisation rate of O₃ and OH⁻ and enhanced gas mass transfer, which is related to the smaller size of the microbubbles. The humic acid and temperature were found to have inhibitory and facilitatory effects, respectively, on the degradation efficiency (Lee *et al.* 2019).

A catalytic exhibition of microbubble ozonation for simulated printing and dyeing wastewater (methylene blue) is reported by Nkudede *et al.* (2021). A drastic increase in COD removal efficiency of up to 93.5% and fast decolourisation within 10 min time is recorded at high pH. To summarise, the implementation of microbubble technology 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 as represented in Table 1.

3.4. Disinfection

Chlorine is one of the most used chemical disinfectants 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 form reactive oxygen species that can help in disintegrating the

Table 1 | Summary of microbubble applications

MBG/Bubble property	Water source	Target pollutant	Result	Reference
Pressurised dissolution/ decompression	Synthetic initial DEP concentration 0.18 mol m^{-3}	Diethyl phthalate (DEP)	<ul style="list-style-type: none"> At pH 7, 97% of TOC removal efficiency achieved Complete mineralisation at higher pH 	Jabesa & Ghosh (2016)
200 L Pilot-scale ozone microbubble	Synthetic effluent and real mining effluent	Ammonia nitrogen	<ul style="list-style-type: none"> Removal efficiency of $\text{NH}_3\text{-N}$ with ozone MB was more at pH 9 than at pH 11 in batch and continuous flow testing 	Ryskie <i>et al.</i> (2020)
Ozone microbubble $14.64 \pm$ $2.08 \mu\text{m}$ diameter	Synthetic initial BHT stock solution $0.90 \mu\text{M}$	Butylated hydroxy toluene (BHT)	<ul style="list-style-type: none"> 1.3–19-fold improvement in ozone mass transfer OH^\cdot being dominant (82%) oxidation species 	Achar <i>et al.</i> (2020)
Hydrogen microbubbles ($25 \pm 13 \mu\text{m}$) from microbubble generator with oscillating mesh	Synthetic ground water	Nitrogen	<ul style="list-style-type: none"> Microbubble enhanced biodegradation process achieving 99% nitrogen removal efficiency 	Eamrat <i>et al.</i> (2020)
Ozone microbubbles with average size of $30 \mu\text{m}$ in semi-batch mode	Synthetic water	Phenol nitrobenzene	<ul style="list-style-type: none"> 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 <i>et al.</i> (2019)
Ozone microbubble using rotating flow/vortex diffuser	Peat water from Riau peatland, Indonesia	Haloacetic acids (HAAs), trihalomethanes (THMs)	<ul style="list-style-type: none"> Primary treatments reduce THM but not HAA Microbubble pre-ozonation reduced HAA in all pH conditions except alkaline pH 	Qadafi <i>et al.</i> (2020)
Ozone microbubbles by pressurised dissolution	Synthetic water	Atrazine	<ul style="list-style-type: none"> Microbubble ozonation enhanced degradation at all pH Self-decomposition of ozone supported by accumulation of OH^\cdot – on gas–liquid interface 	Liu <i>et al.</i> (2020)
Ozone microbubble	Simulated printing and dyeing waste water	Methylene blue	<ul style="list-style-type: none"> 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 <i>et al.</i> (2021)

bacteria. But this technique is not of practical importance because of the allied cost factor. However, HC 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 less ozone dose as compared to conventional ozonation systems (Sumikura *et al.* 2007). In 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 microbubble technology with high inlet ozone concentrations (Zhang *et al.* 2013).

A meaningful insight into the disinfection mechanism of both Gram-positive and Gram-negative bacteria are provided by Jain *et al.* (2019) 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 bacterium *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 a value close to 98% (Jain *et al.* 2019).

A faster and greater disinfection outcome was suggested for *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 membranes 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 by OMB induced leakage and coagulation of intracellular components following on to lysis of spores and final cell death (Tamaki *et al.* 2018). Another case study of domestic wastewater treatment in Carhuaz, Peru highlighted the role of air-ozone MBs/NBs in reducing faecal and total coliform counts. The application of MB/NB treatment on the wastewater with initial faecal coliform count of 130,000 CFU/100 mL achieved a reduction of up to 100 CFU/100 mL (99.92%) (Cruz & Flores 2017).

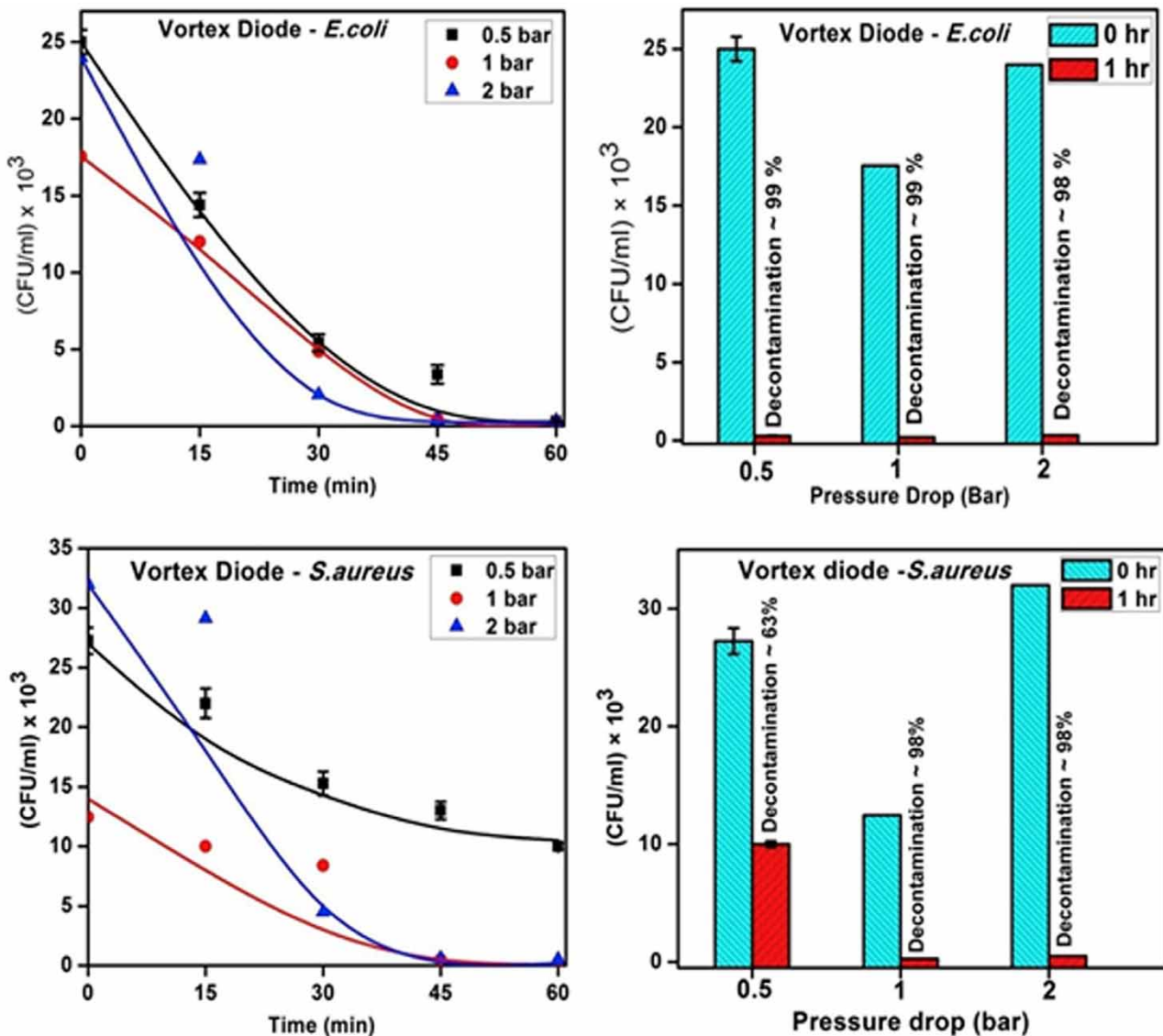


Figure 8 | Effect of pressure: disinfection of *E. coli* and *S. aureus* by vortex diode (Jain *et al.* 2019).

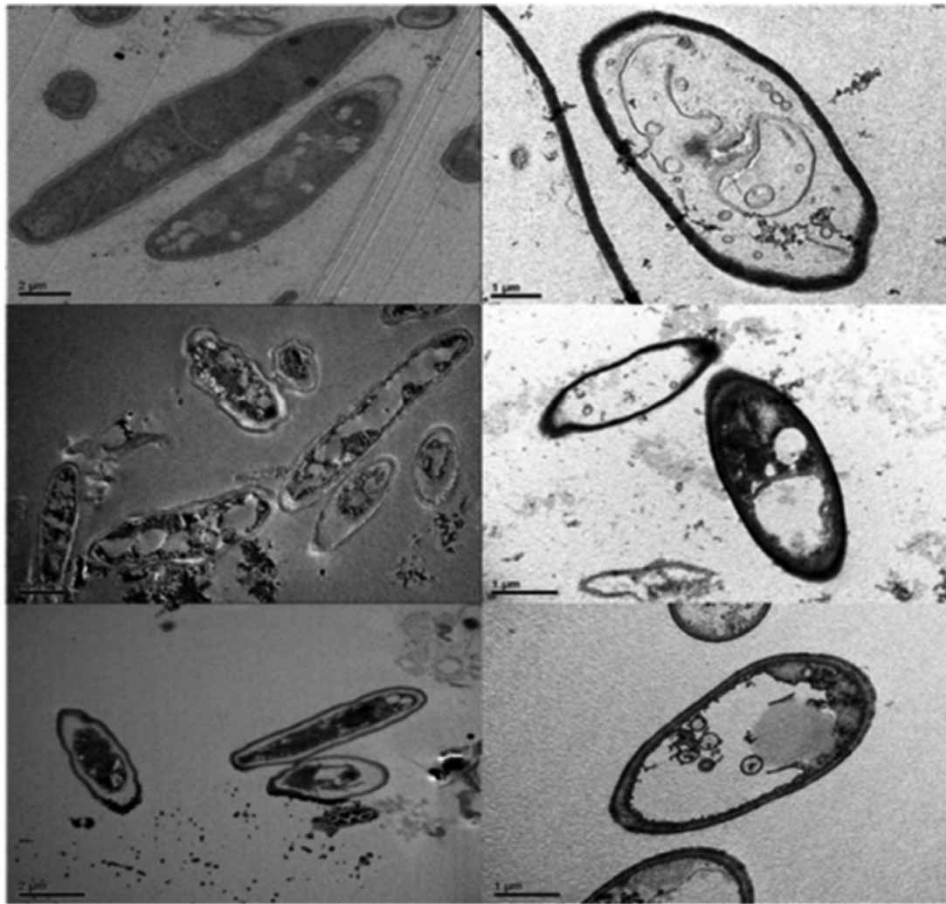


Figure 9 | TEM images of *F. oxysporum* f.sp. *melonis* spores. Top: Non-treated, Middle: After treatment with O3MMB for 180 s, Bottom: After treatment with O3MB for 180 s (Tamaki *et al.* 2013).

Harmful algal blooms (HAB) consisting of algae and cyanobacteria because of their more frequent occurrence in water bodies like lakes and rivers are becoming an epidemic. The presence of these organisms in water results in decreases in dissolved oxygen content, penetration of light and the exchange of other gases which is harmful for the aquatic life. 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 (Seridou & Kalogerakis 2021). Hydrodynamic cavitation producing MBs and NBs is believed to be a feasible approach suggested for eliminating surface blooms and associated toxins. A pilot study on Lake Neatahwanta, New York, USA with field blooms suggested a 50% reduction in cyanobacteria chlorophyll after 72 h of microbubble treatment by HC. The higher decline percentage of 80% was reported with additional treatment with peroxides (Shaw 2020). A study by Thomas *et al.* (2019) showed the potential of MBs induced by small, inexpensive nozzles and water circulation systems 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 HC 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 walls and the absence of gas vacuoles, which is known to initiate the secondary production of free radicals (Thomas *et al.* 2019).

3.5. Real wastewater treatment

MBs have been utilised to treat both synthetic and real wastewater and have shown significant pollutant removal efficiency. Real wastewater is a complex mixture of different kinds of contaminants or pollutants such as hydrocarbons, pathogens, synthetic organic chemicals, toxic metals at the same time. The mixture of these contaminants has adverse effects on the treatment efficiency and complicates the treatment process. To ensure that improvements realised in pure water laboratory tests using MB system transverse across to more complex water matrices having organic matter, more experimental tests on real wastewater are to be carried out. For instance, air microbubbles have exhibited significant results in the treatment of dyeing wastewater. Compared to the conventional air bubbles, MBs have been shown to reduce the coagulant doses and treatment time and enhance the pre-treatment efficacy of coagulation–flotation. The removal efficiency of COD, colour and oil of 89, 72 and 99%, respectively, is achieved with coagulation microbubble flotation. After the coagulation microbubble flotation, the biodegradability index of wastewater also increased from 0.290 to 0.363 (Liu *et al.* 2010). In yet another study, the rate of decolourisation and organic reduction of practical textile wastewater was found to be much faster with MB ozonation. The COD removal efficiency with MB system was increased by 20% compared to the macrobubbles (Chu *et al.* 2008). An advanced treatment process with the novel combination of microbubble catalytic ozonation and biological process was successfully applied to treat real bio-treated coal chemical wastewater. The authors successfully demonstrated that catalytic microbubble ozonation results in 32.16% COD removal efficiency, consuming 1.38 mg of ozone per mg of influent COD removed with an ozone utilisation efficiency of 98%. Further biological treatment resulted in additional removal of biodegradable COD to 60.82% and release of inorganic nitrogen $\text{NH}_3\text{-N}$ (Liu *et al.* 2018).

Another experiment examined the combination of MB and ultraviolet light irradiation to treat the river water containing refractory component from secondary effluent. This combination approach significantly improved the biodegradability of the water with increase in the amount of OH^\cdot by almost 2–6 times compared with the conventional ozonation system. The enhanced degradation rate of refractory organics was seen with COD and UV, UV removal rates reaching as high as 37.50%, 81.15% and 94.74%, respectively (Gao *et al.* 2019).

4. 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. The reviewed data evidence a strong base for improvement through the implementation of microbubble system in ozonation, however, it must be emphasised that these data take into account the results of laboratory trials. The generation of ozone is portrayed as an energy demanding process with an energy requirement of 3.3–16 kWh/kg O_3 which covers 50–75% of the cost of total ozonation process.

Zhang *et al.* (2018) has reported a gas utilisation efficiency of 96% with 51 μm microbubble in a 32 cm deep tank. The enhanced gas utilisation suggests that a less amount of ozone is required to achieve an intended residual ozone concentration. So, microbubbles act to decrease the operational cost through decrease in the ozone input dose and minimized requirement of expensive off-gas destruction systems. Additionally, enhanced pollutant removal rates can offer opportunities for smaller dimensions of contact tanks and in this way reducing the capital investment and carbon footprint of the plant. Based on the minimum and maximum mass-transfer coefficient enhancement observed with microbubbles, Nam *et al.* (2021) and Zhang *et al.* (2018) have suggested the possibility of reducing the reactor volume by 16 and 81%, respectively, compared to systems using conventional-sized bubbles. However, no experimental work to date has normalised the reactor volume and tank depth when using microbubble systems.

Encouraging are findings of reduced reactor volume with microbubble generators in ozonation; and reducing the capital cost of ozonation and operational cost in areas including reduced chemical consumption and off-gas destruction makes the full-scale microbubble ozonation a feasible option. While there has been convincing data on enhanced gas utilisation and mass transfer using microbubbles, it has not been realised in practical situations as most of the experiments to date have utilised relatively shallow water depths compared with the commercial ozonation contactors with 3–7 m deep tanks.

Although the laboratory-scale set up does not give an accurate idea of energetics of upscaling, it can provide a suggestion on the probable trend. An economic efficiency study was carried out by Andinet *et al.* (2016) to assess the running cost–benefit of supplying air or oxygen microbubbles for aeration of 20 min. 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 was suggested to improve the natural water bottom area by oxygen MBs. An alleviation in membrane fouling is seen in the vacuum membrane distillation desalination process by MB aeration. Inclusion of MB aeration is merely contributing $2.8 \pm 0.3\%$ 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 (Ye *et al.* 2019).

A low-cost, efficient mineralisation of total petroleum hydrocarbon (TPH) is achieved with microbubble ozonation by synergising oxidation and flotation phenomena. A 70.9% reduction in TPH was observed in 120 min of microbubble treatment with 0.27 gO₃/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 3.44×10^6 British pound sterling (GBP)/m³ TPH. This is much less when compared to other conventional oxidation processes (Sun *et al.* 2020). 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 H₂O₂ 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 m³ of wastewater containing 5 ppm of methyl orange was estimated to be 1707, 626 and 80 GBP/m³ for HC only, HC + H₂O₂, HC + H₂O₂ + metal system, respectively (Innocenzi *et al.* 2019).

Thanekar *et al.* (2020) compared the cavitation yield and operation cost of different types of treatments for degradation of dimethoate pollutant based on power consumption. The HC treatment alone was found to cost 8.5 GBP/m³ 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 1.2 GBP/m³ with a cavitation yield of 20.1×10^{-5} mg/J (Thanekar *et al.* 2020). In a similar study, the synergistic combination of HC and advanced oxidation processes (AOP) was applied for the treatment of industrial wastewater on 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 322.38 GBP/m³ for electricity and additive (Joshi & Gogate 2019).

An economical and suitable large-scale commercial operation of the HC/chlorine dioxide (ClO₂) composite process is proposed for the treatment of explosive-containing wastewater. The cost calculations and cavitation rate comparison suggest the reduced treatment cost of 8.77 GBP/m³ for HC/ClO₂ treatment as compared with 16.33 GBP/m³ for single cavitation along with an enhanced cavitation rate of 26.78×10^{-3} mg/J for the former (Wang *et al.* 2020). Table 2 shows the cavitation yield and operational costs of different treatment approaches based on energy requirement. The operational costs based on energy

Table 2 | Cavitation yield and operational cost based on energy for different treatment approaches

Contaminant	Treatment	Extent of degradation (%) COD/TOC	Cavitation yield (mg/J)	Energy required (KWh)	^a Total operational cost based on energy (GBP/m ³)	Reference
Total petroleum hydrocarbon (TPH)	O ₃ microbubble (0.27 g O ₃ /gTS)	70.9			3.44×10^6	Sun <i>et al.</i> (2020)
Dimethoate	HC (slit venturi)	14.63	2.95×10^{-5}	0.094	8.5	Thanekar <i>et al.</i> (2020)
	HC + UV	30.8	4.24×10^{-5}	0.065	5.9	
	HC + H ₂ O ₂	72.5	14.6×10^{-5}	0.019	1.7	
	HC + Fenton	100	20.1×10^{-5}	0.013	1.2	
Methyl orange	HC (Venturi tube)	53.69	4.90×10^{-7}	1.44	1,707.1	Innocenzi <i>et al.</i> (2019)
	HC + H ₂ O ₂	53.11	5.47×10^{-7}	1.56	626.08	
	HC + H ₂ O ₂ + metal	90	1.2×10^{-6}	0.20	79.98	
2,4,6-Triamino - 1,3,5-trinitrobenzene (TATB)	HC (orifice)	13.59	5.52×10^{-3}	0.2721	16.33	Wang <i>et al.</i> (2020)
	ClO ₂	20.38	0	0	5.4	
	HC + ClO ₂	65.9	26.78×10^{-3}	0.0561	8.77	
Complex industrial wastewater	HC + O ₂ + Fenton (orifice)	63	47×10^{-3}	2,084.2 kWh/m ³	322.38	Joshi & Gogate (2019)

Note: ^aCNY = 0.12 GBP, Rs = 0.010 GBP, Euro = 0.86 GBP, US\$ = 0.81 GBP.

Table 3 | Comparison of cavitation yield and cost-effectiveness of various processes

Process	Time required for 75% COD reduction (min)	Cavitation yield (mg/l)	Energy consumption for 75% COD reduction (kwh/m ³)	^a Total cost (GBP/m ³)
HC	360	0.005	32.05	1.814
HC + H ₂ O ₂ (2 g/L)	180	0.013	16.02	1.094
HC + H ₂ O ₂ (5 g/L)	50	0.04	4.27	0.713
HC + H ₂ O ₂ (7 g/L)	55	0.03	4.80	0.940
HC + O ₃ (3 g/h)	75	0.19	29.17	1.652
HC + H ₂ O ₂ (5 g/L) + O ₃ (3 g/h)	30	0.34	11.67	1.134

Note: ^aUS\$ = 0.81 GBP.

use data are converted to similar base units for comparative purpose. All costs were adjusted to the 2022 GBP. So, it could be concluded that combination treatment approaches help in cutting down the cost of the treatment.

Mukherjee *et al.* (2020) 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 H₂O₂ 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 H₂O₂ 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 and is summarised in Table 3. All costs were adjusted to the 2022 GBP. Energetic and cost calculations support HC + H₂O₂ (5 g/L) as the most feasible treatment since the cost of this process is the least with comparable cavitation yield as HC + H₂O₂ + O₃ (Mukherjee *et al.* 2020).

A novel idea has been put forward by Ranade *et al.* (2021) to present a multi-scale modelling of HC devices for degradation of pollutants at four different scales (scaling of approximately 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.

Any specific conclusions cannot be drawn as to which hybrid technology is the best 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 an other type of pollutant. 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.

5. CHALLENGES IN IMPLEMENTATION AT INDUSTRIAL SCALE

- Although much research and laboratory studies have shown the immense potential of microbubble technology 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 are the need of the hour to evaluate the efficiency of the process and to obtain working knowledge and experience.
- Simulation studies on MB generation and reaction with contaminants are limited. Simulation aids in understanding the microbubble-assisted processes without carrying out the experiments. Simulations can be done before upscaling the process and can significantly cut the design and operation costs if used for optimisation of the process.
- 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 design of novel systems retro-fitted with microbubble technology.
- The majority of the published literature has concentrated on mass-transfer efficiency of microbubble technology, but very less importance is being given to the heat transfer. MBGs during their course of operation are certainly generating heat (rise of the temperature of the medium), which could be prominent in several reaction mechanisms and can reduce the energetics of the treatment. This area can be further explored.

6. CONCLUSIONS

We identified the significant relevance of microbubbles in water treatment technology. The broad application of microbubble technology 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 oxidation from free radical production. The microbubbles have been shown to selectively degrade harmful algae, leaving behind beneficial ones. The results of implementation of microbubble technology in aeration and ozonation overcoming the limitations of low gas utilisation efficiency and higher oxidant dosages of conventional methods have been predominantly encouraging. Many studies have stressed the role of microbubble technology in downsizing the treatment plant and reducing the operation costs of WWTPs along with increased contaminant removal efficacy. However, evidenced data from experiments correspond to the microbubble treatment in shallow water depths which may not be the same in the real scenario of WWTPs.

The cost and energy consumption analysis are indicative of the potency of microbubble technology combined with AOPs to replace the existing expensive processes in water treatment. However, the most effective treatment process and combination varies for different pollutants. More industrial-scale studies are recommended for bridging the gaps between laboratory-scale research and implementation at larger scales. A comprehensive assessment of degradation performance, energy efficiency and economic evaluation is to be carried out on real wastewater with different types and concentrations of pollutants to establish an optimised and distributed water treatment system. It is suggested that forthcoming studies should explore in detail the impact of treated water depth along with a reasonable and effective synergetic index of the microbubble/AOP hybrid system to establish a standardised approach for the comparison and clarification of best wastewater treatment strategy.

ACKNOWLEDGEMENTS

This project has received funding from the ‘THYME project’ programme.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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First received 17 May 2022; accepted in revised form 5 October 2022. Available online 12 October 2022