

# Micro–nanobubble technology and water-related application

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## ABSTRACT

Currently, there is a growing demand for water treatment technologies considering global environmental challenges such as degradation and depletion of water resources. Micro- and nanobubble (MNB) technology and its application for wastewater treatment has emerged as a problem-solving alternative for such challenges. This paper reviews the important studies on water treatment in the areas of MNBs and discusses their fundamental properties, such as bubble stability (as tiny entities in water solutions), generation methods, and various chemical and physical features. The paper further overviews the current status of MNB application in water treatment processes such as flotation, aeration, and disinfection and its uses in various sectors, including agriculture, aquaculture, medical, and industry. Based on this review, studies regarding MNBs' basic properties, generation, and application are identified and recognized for future research. This study concludes that despite the promising role of MNBs in water-related application, the current status of research has not reached its true potential. Specifically, there is a need to enhance MNB application at a broader scale.

**Key words** | application, micro–nanobubbles, water treatment

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## INTRODUCTION

Global economic development cannot be separated from water resources. Currently, in many developing countries, water resources are facing extreme challenges such as water scarcity, imbalances in the water distribution and production model, low efficiency of water use, droughts, and other environmental problems (Khan *et al.* 2020). Further, the increasing amount of wastewater caused by rapid urbanization and industrialization has significantly increased the challenges of both water

availability and quality (Kivaisi 2001). Therefore, the treatment and recycling of wastewater are increasingly needed to ensure the sufficient availability of water. Generally, biological methods, such as activated sludge, have been used to treat the pollutants from both industrial and domestic waste water (Terasaka *et al.* 2011). However, such methods have disadvantages such as high energy costs and result in an abundant amount of solid waste, creating an additional cost to dispose of the produced waste (Orel *et al.* 2017). Therefore, there is dire need to develop a water treatment technology that can effectively address the increasing challenges of water scarcity sustainably.

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In such a scenario, micro- and nanobubbles (MNBs) have emerged as a useful technology to be used in water treatment. MNBs are tiny bubbles with diameters of nanometres and micrometres having several unique physical properties that make them useful for water treatments (Xiaoli *et al.* 2017). For instance, their unique property of having large surface area enables an efficient mass transfer process between the liquid and gas phases, which helps to facilitate chemical reactions (Bouaifi *et al.* 2001). Such processes lead to the collapse of the MNB, which produces shock waves in the water, generating OH radicals as a result (Khuntia *et al.* 2015). The water treatment processes, such as electroflotation and dissolved air flotation, generally make use of MNBs (Miettinen *et al.* 2010). In recent years, the use of such methods has been widely adopted for decontamination of domestic and industrial water treatment, due to the higher bioactivity of MNBs (Kulkarni & Joshi 2005). Further, due to the enormous feasibility, MNB-based water treatments are being applied in various sectors, including industry and agriculture (Serizawa 2017).

During recent years, research on MNBs for water-related applications has significantly increased, considering their feasibility as a sustainable technology for water treatment (Tsai *et al.* 2007). Despite a wide range of studies and experimental evidence, the use and application of MNBs for water treatment is still limited. This study, in this regard, is designed to present an overview of existing literature on micro-nanobubbles and their use in water-related applications. This study will provide insights into recent developments and the status of research on MNBs and highlight future trends regarding the use of MNBs.

The paper is divided into seven sections, beginning with the introduction as the first section. The second section explains the basic properties of MNBs. The third section discusses the MNB generation mechanism and overviews the development timeline of MNBs. Similarly, major water treatment processes and MNB use and applications in various sectors are discussed in the fourth and fifth sections, respectively. Sections six and seven provide future scope and conclusions, respectively.

## FUNDAMENTAL PROPERTIES OF MNBs

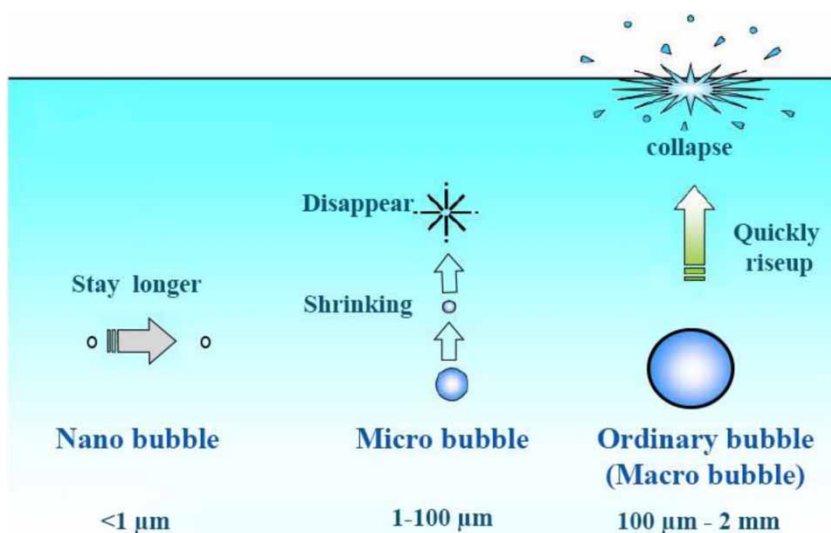
Generally, water bubbles can be categorized into three major types, i.e., ordinary or macrobubbles, microbubbles

(MBs), and nanobubbles (NBs). The diameter of macrobubbles ranges from 100  $\mu\text{m}$  to 2 mm. These bubbles quickly rise to the surface of a liquid and collapse. While microbubbles are smaller than macro-bubbles, with a diameter range of 1  $\mu\text{m}$ –100  $\mu\text{m}$  (Azevedo *et al.* 2019), these bubbles shrink in the water and then dissolve into it.

In contrast, NBs are extremely small gas bubbles that have several unique physical properties that make them very different from normal bubbles. Generally, NBs range <1  $\mu\text{m}$  in diameter (Azevedo *et al.* 2019); however, the NBs that are <100 nm in diameter randomly drift in what is termed Brownian Motion and with a lower buoyancy can remain suspended in liquids for an extended period of time (Figure 1) and have the ability to change the typical characteristics of water.

Despite subsequent studies, there are still controversies over NBs' size and their existence in solutions under atmospheric conditions (Agarwal *et al.* 2011). According to classical thermodynamics theory, NBs cannot exist or be stable due to the limitation of radius curvature (Ljunggren & Eriksson 1997; Ushikubo *et al.* 2010). For instance, the small radius of curvature gives NBs a higher internal pressure relative to the external pressure, which leads to rapid dissolution (Holmberg *et al.* 2003). For example, an NB with radius 100 nm (atmospheric pressure in the surrounding water =  $10^5 \text{ N m}^{-2}$  and surface tension = 72 mN/m) gives an internal pressure of 1.5 MPa (Attard 2013). The basic theory of this phenomenon is that high internal pressure contained in the NB does not achieve balance with the atmosphere, which would inevitably lead to the bubbles bursting in a very short time (Ljunggren & Eriksson 1997).

Lou *et al.* (2000) first reported the existence of NBs as bright and stable spheres on a flat solid surface, through AFM (atomic force microscopy). However, in recent years various other methods have also been adopted for NB investigation, such as neutron reflectometry, X-ray reflectivity and quartz crystal microbalance (Zhang 2008). One of the unique properties of NBs is stated to be their higher longevity, which gives them a larger contact angle and longer existence time in the liquids (Lou *et al.* 2000; Takahashi *et al.* 2007; Ying *et al.* 2013).



**Figure 1** | Working of MNBs (Takahashi *et al.* 2007; Azevedo *et al.* 2019).

### Zeta potential of MNBs

Zeta potential is a physical characteristic used to measure the magnitude of the attraction between particles and bubbles or electrostatic repulsion. It is an important property that determines the longevity of MNBs in a colloidal system (Jia *et al.* 2013). For the calculation of zeta potential, the Smolochowski equation is used (Han & Dockko 1998; Ushikubo *et al.* 2010). Generally, MBs and NBs have a negative charge in the pH range of 2–12, depending on the kind of gas introduced, and the zeta potential ranges from –50 to –40 mV and –30 to –20 mV at a neutral pH. The negative zeta potential value is determined by the excess of hydroxyl ions (OH<sup>–</sup>) relative to hydrogen ions (H<sup>+</sup>) at the gas–water interface (Ohgaki *et al.* 2010). The charging mechanism of NBs is also associated with the preferential adsorption of hydroxyl ions in the electrolytic solution. Further, the stability of the NBs is highly dependent on hydrogen bonding at the water–gas interface, due to which the NB has an impermeable, kinetically stable surface making it more diffusion-resistant (Wang *et al.* 2013).

### Mass transfer properties

In various gas–liquid phase operations, the efficiency of the process is generally determined by the gas to liquid transfer

rate; hence mass transfer is a system's critical property (Wilkinson & van Dierendonck 1990). The mass transfer efficiency relies on the bubbles' size distribution, rising velocity, gas–liquid hydrodynamics, coalescence, and break-up surface-to-volume ratio, and physical properties (Bouaifi *et al.* 2001). According to gas absorption two-film theory, the mass transfer rate of two phases is determined by the coefficient of liquid–gas mass transfer, surface area to volume ratio, and concentration gradient within these phases (Whitman 1962; Bouaifi *et al.* 2001). The unsteady-state method is used to calculate the gas–liquid mass transmission rate (Terasaka *et al.* 2011). Studies show various correlation approaches for the calculation of the mass transfer coefficient of MBs based on various mathematical models. Bredwell & Worden (1998) proposed a dynamic approach, which stated that the mass transfer coefficient is associated with factors such as the reduction in bubble size, which he numerically solved with a single MB of size 25 μm. Further, Ying *et al.* (2013) also developed a model in order to enhance the mass transfer rate, which was associated with the transfer rate of carbon dioxide based on the bubble size. Subsequently, a stirred reactor was used by Li *et al.* (2016), who examined the mass transfer process using a 50 μm MB. They found that bulk liquid circulation by the MB generator enhanced the bulk liquid's temperature, affecting the gas solubility, specifically for a

smaller-volume bulk liquid. They calculated the mass transfer rate employing a temperature correlation factor at 20 °C.

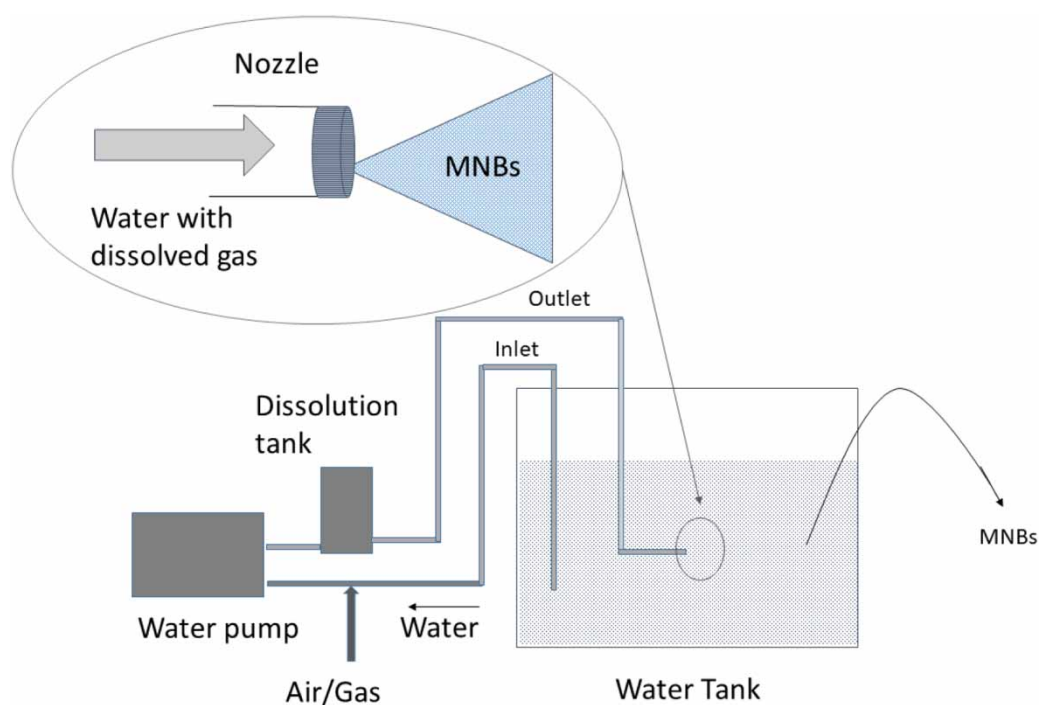
## MNB GENERATION AND TECHNOLOGY

### MNB generation mechanism

MNBs are generated on the surface of hydrophobic particles (Fan et al. 2010a, 2010b). Depending on internal or external factors, the formation of MNBs can be induced in several possible ways (Demangeat 2015). Generally, water bubbles are generated by dissolving gas with pressure and releasing gas while reducing pressure bubbles, which is considered a conventional method. In this method, the device is mainly composed of a water pump, air compressor, and air tank. The water pump provides a certain pressure to send the circulating water to the dissolved gas tank, and the air compressor presses the air into the dissolved gas tank. The high-pressure gas–water mixing state formed in the dissolved

gas tank makes the gas supersaturated and dissolved, and then the gas is precipitated out of the water in the form of MNBs by sudden decompression (Deng et al. 2014). Figure 2 shows a schematic diagram of the commonly used MNB generator (Takahashi et al. 2007). A pump circulates water in a transparent acrylic tank through a gas-dissolution tank and MNB generating nozzle. The air is injected into the circulating water on the suction side of the pump and is dissolved by a high-pressure system. A gaseous phase of MNB is then produced from the water supersaturated with air due to the pressure reduction at the nozzle. There are variations in the methods related to MNB generation, and a comparison of various bubble-generation principles and apparatus is shown in Table 1 (Xiong et al. 2016).

NBs are often formed when a homogeneous liquid phase undergoes a phase change due to a sudden pressure drop below a critical value, known as cavitation (Wu et al. 2012). Usually, cavitation is formed by the passage of ultrasonic waves or changes in high pressure in a running fluid (hydrodynamic cavitation) (Agarwal et al. 2011; Demangeat 2015). NBs can also be generated by ultrasonication (Kim



**Figure 2** | Schematic diagram of the MNB generator. Adopted and modified from Takahashi et al. (2007).

**Table 1** | Advantages and disadvantages of commonly used MNB generation methods (Xiong et al. 2016)

	Method	Principle	Advantage	Disadvantage
Dissolving–releasing gas	Dissolving gas with pressure and releasing gas while reducing pressure	Dissolve the gas in water at high pressure and form a supersaturation state, then enter the gas to reduce pressure and form a large number of fine bubbles. The size and strength of the bubbles depend on the release conditions, including surface water tension.	At high pressure, the gas is highly soluble, so the number of bubbles is large and more bubbles rise stably.	The efficiency is low since the whole process is continuous.
	Dissolving gas with pressure and releasing gas with impeller	Fine bubbles are produced directly by the gas dispersed by the impeller, or in combination with the pressurized gas, performing three processes of mixing gas and water, releasing gas and gas dissolving.	Simple principle, efficiently produced in the combination of gas–water mixtures, more dissolution and release of the gas into the pump.	The process is complex, usually forms large bubbles (>50 µm).
Dispersing gas method	High-speed rotational flow	The gas–water mixture flows into the circulating hollow, forming a pressurized gas. The gas cuts into ultra-fine bubbles and rotates as the water passes.	Generally high quality and efficient.	The flow path is difficult to design and produce.
	Flow-path section change	The flow cross-section slowly decreases and then quickly increases; the water collapses violently and vortices are formed. Repeating the process leads to stronger turbulence and finer bubbles.	A large flow path makes it easy to repair and less likely to block up.	Difficult to adjust oxygen content during streaming significant changes in conditions.
	Fine porous materials	The pressure gas forms fine bubbles through the strength of small porous materials.	The quality of the bubbles mainly depends on the porous material. The method is simple.	Stuck frequently.

et al. 2000; Oeffinger & Wheatley 2004), and chemical reactions such as electrolysis (Wu et al. 2012).

The Venturi-type generator is widely used to generate NBs by the hydrodynamic cavitation mechanism (Agarwal et al. 2011; Ahmadi & Darban 2013). The Venturi system consists of three main parts, i.e., input flow, tubular, and tapered output flow (Parmar & Majumder 2013). The reduction of pressure in the Venturi tube can be attained by increasing the speed of the fluid in the converging areas of the pipe of narrow diameter (Fan et al. 2010a). In the Venturi-type generator system, liquid and gas are transmitted simultaneously through the Venturi tube to generate the bubbles (Parmar & Majumder 2013). When the pressurized liquid is injected into the tubular part, the flow of fluid into the cylindrical throat becomes higher, while the pressure becomes lower than the input section, leading to cavitation (Fan et al. 2010a).

NBs can be obtained in the wider size range (within several hundred nanometres). Ahmadi & Darban (2013) produced NBs of 130–545 nm through a Venturi tube from the hydrodynamic cavitation mechanism. Wu et al. (2012) also created NBs with a size of less than 500 nm on the basis of hydrodynamic cavitation. Cho et al. (2005) used a palladium electrode and generated NBs with an average diameter of 300–500 nm from ultrasonication. Oeffinger & Wheatley (2004) generated NBs with an average diameter of 450–700 nm from ultrasonication using octa-fluoro propane gas.

### Effects of various operational conditions on bubble size

The literature shows that the distribution and size of MNBs depend on the system design and various operational conditions. For example, MBs' fraction and size are generally



associated with the pressure changes across the nozzle system. The more the pressure, the smaller the bubble size due to the increase in air density; however, the size of MBs remains constant at a pressure above 3.5 atm (de Rijk 1994), while the size of the NBs is mainly determined by conditions such as the size and the type of hose (Wu et al. 2017), pressure (Kim et al. 2012) and sonic power (Cho et al. 2005). In addition, different types of gases influence the existence time of NBs, for example, the presence of NBs can be detected for days when pure oxygen is used to generate the bubbles, while in the case of air bubbles, the existence time is only for less than 1 h (Ushikubo et al. 2010). For macrobubbles, using a gas of higher molecular weight enhances the density and hence results in smaller bubbles (Kulkarni & Joshi 2005).

Similarly, in terms of chemical properties, studies have compared the impact of electrolyte solutions (with NaCl, and other salt ions) and frothers (chemical additives) on sea-water along with impeller speed and airflow rate (O'Connor et al. 1990). Results revealed that a low impeller speed (4.2 m/s) or a high gas velocity (0.5 cm/s) generated an 850  $\mu\text{m}$ -size bubble of sea salt solution, which was higher than the sizes generated by the solution of frothers (600–700  $\mu\text{m}$ ). Similarly, regarding the comparison on a single impeller speed and gas flow rate, the interaction of salts and frothers has also been explained (Sovechles et al.

2016), which showed that, without the frother, bubbles generated in sea-salt solution and 0.5 M NaCl were relatively smaller than in deionized water. Concerning the concentration range of frothers, the study reported that by adding a small amount of frother (<10 ppm), the NaCl solution (0.5 M) generated relatively larger-size bubbles, while the electrolyte solutions above the concentration of 10 ppm produced bubbles with similar size (Sovechles et al. 2016).

Further, studies also compared cases considering the ionic strength of the electrolyte solution with regard to the critical coalescence concentration (Castro et al. 2013; Sovechles et al. 2016). Results showed that the solution of sea salt (0.7 ionic strength) generated larger bubbles relative to the NaCl solution (0.5 ionic strength). In the comparison of NaCl solution and sea-salt solution, the size of the bubbles increased due to the interaction of salt ions existing in the sea-salt solution (Sovechles et al. 2016). Many studies considered NaCl solution as a base with other salt ions (KCl etc.) to evaluate their impact on the size of the bubbles individually, which shows the other salt ions had no significant difference in the bubble size (Quinn et al. 2014; Sovechles & Waters 2015; Sovechles et al. 2016).

### An earlier overview study on MNBs

Figure 3 presents major development events and important studies in the field of MNBs. In 1950 Epstein Plesset's

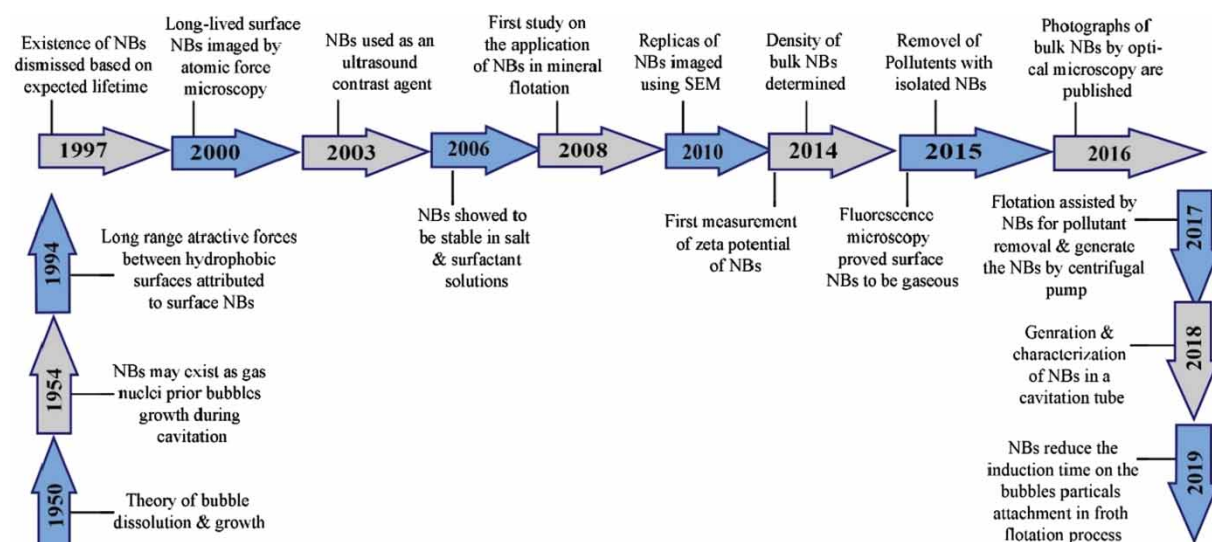


Figure 3 | A chronological overview of research and development of MNB application.

theory was developed (duration of a single bubble based on its radius and saturation) (Plesset & Sadhal 1982). In 1954 it was suggested that organic films stabilized small units of gas ( $<1\text{ }\mu\text{m}$ ), serving as the nucleus of microbubble growth during cavitation (Zatakis 1954). In 1994 long-range forces were measured between water-based surfaces (Parker et al. 1994). In 2000 surface NBs were recorded using atomic force microscopy (AFM) (Lou et al. 2000). In 2003 MNBs were reported as an ultrasonic contrast agent (Dube et al. 2003). In 2006 the effect of surfactants and salt on the size and shape of surface NBs was revealed to be negligible (Zhang et al. 2006). In 2008 the application of MNBs in froth flotation was studied (Fan & Tao 2008). In 2010, MNBs were scanned by cryo-SEM (Ohgaki et al. 2010). In 2014, the relative mass density of MNB to the solvent was calculated with a micro-resonator (Kobayashi et al. 2014). In 2015 researchers used a concurrent combination of fluorescence microscopy and various imaging methods to show the gaseous surface of NBs. In the year 2016, developing of MNB photographs was done by optical microscopy (Azevedo et al. 2016). In 2017 flotation was assisted by NBs for pollutant removal and a second study generated NBs by a centrifugal pump (Oliveira et al. 2017). In 2019 the froth flotation process NBs reduced the induction time on bubbles-particles (Tao & Sobhy 2019).

## MNB APPLICATION IN WATER TREATMENT

The literature shows that MNBs have been useful in methods of water treatment, aeration (Tasaki et al. 2009; Yamasaki et al. 2010), disinfection, and flotation (Jyoti & Pandit 2001; Mezule et al. 2009; Kim 2010). According to studies, the basic areas of MNB applications in water treatment are to reduce the size of the system structures, operating time, the operating costs of processing plants, and increasing efficiency in eliminating water pollutants.

### Aeration process

Aeration is the process of introducing or penetrating oxygen into the water substance. In water treatment, aeration has an important role in the supply of oxygen, which is an essential element for biochemical substrate reactions and aquatic life.

Numerous studies have been carried out dealing with the impact of aeration processes on wastewater and biological water treatment, and groundwater recovery. One of the key objectives of many studies was to enhance the efficiency of the factors that affect the speed of mass transfer. In common aerobic systems, dissolved oxygen is an important feature of tackling such inefficiency (Tang et al. 2016). In such systems, the mass transmission amount of oxygen is very important as most of the contact equipment uses diffusers or mechanical aerators that require huge maintenance costs and electrical input (El-Zahaby & El-Gendy 2016). Therefore, studies have focused more on the optimization of conventional bubbles and aerator design to improve mass transfer aeration. However, research on the use of high mass transfer of bubbles is not well established on an industrial scale (Li et al. 2016). Weber & Agblevor (2005) described the characteristics of the transfer rate of gas-liquid mass using MBs for aeration in the stirred-tank reactors and concluded that MB aeration is better suited to bioreactors. They further studied the effect of MB aeration on *Trichoderma reesei*'s fermentation (a filamentous fungus and mesophilic), which is dependent on the limited rate of oxygen mass-transmission. This study showed that the concentration of dissolved oxygen was higher than the concentration at a lower agitation rate due to the use of MB aeration. Further, the concentration of cellular mass increased swiftly during the rapid growth phase, with an increase from 0.1 to 0.18 g/LH compared with conventional bubbling.

Liu et al. (2015) reported that using NBs in aerated water, the germination of the seed was improved compared with ordinary water. Likewise, Park & Kurata (2009) studied the growth of lettuce (*Lactuca sativa*) by using MB aeration and found that the dry and fresh bulk of appropriately aerated MB lettuce was 1.7 and 2.1 times greater than with macrobubble aeration. In these studies, the researchers hypothesized that higher germination and growth rates were associated with the specific high surface area of MNBs and the greater attraction capability of positive ions. Besides, this oxygen MNBs have 126 times more mass transfer efficiency, and three times more dissolved oxygen (DO) compared with air micro-NBs. Increased mass transmission in the bubble interface further increases the efficiency of oxygen transmission and leads to longer

stagnation of MBs in the water. In wastewater treatment with the use of MNBs, Temesgen (2017) conducted a study on the use of NBs for the degradation of aerobic waste. The results showed that the rate of use of oxygen and the volume transfer rate in the synthetic aerated NB treatment plants was almost twice as high as with conventional air bubbles. The degradation time of organic waste production in NB-aerated units was less than half compared with the conventional systems. Lastly, the decay and growth rate in the NB aerated unit was also much faster than the conventional system (Temesgen 2017). Due to such properties, there is further need for experimental studies in this area.

### Flotation process

Flotation has also been a major separation process in the area of water purification (Hopper & McCowen 1952). The most specific substances that need to be removed from flotation are dust, chemicals, organic matter, metal ions, and oils (Rubio *et al.* 2002). The efficiency of the separation process is closely linked with the size of bubbles, for instance, Dockko & Han (2004) discussed the probability of improving the flotation efficiency, by primarily changing the characteristics of the bubbles, i.e., the size of the surface, and particle properties. In flotation, NBs and MBs are commonly used to remove pollutants from the water with greater efficiency. A subsequent number of experimental studies has made evident the efficiency of this method in terms of collecting bubbles–particles (Miettinen *et al.* 2010). Ahmed & Jameson (1985) found that the speed of flotation was highly dependent on the size of the bubbles as speed increased 100 times with the reduction of bubble size from 655 to 75  $\mu\text{m}$ .

In addition, Yoon (1993) indicated that the reduction in the size of the bubble in flotation is strongly correlated with an increased possibility of the tiny particles colliding with the small bubbles, which consequently lead to an increase in separation efficiency. In addition to particle sizes and small bubbles, surface charges also have a significant role in flotation (Collins & Jameson 1976; Bui *et al.* 2015). Positively charged MBs are expected to effectively isolate algae from the water, with a rate of 90% elimination of cells and 92% reduction in chlorophyll (Teixeira *et al.* 2010). Concerning organic substances, i.e., dissolved and organic carbon

and aliphatic or aromatic mixtures, the MBs could achieve an elimination rate of more than 30% (Takahashi *et al.* 2007; Bui *et al.* 2015).

Sumikura also investigated that the NBs expanded the area of flotation particles and increased the hydrophobicity of surface particles leading to enhanced efficiency of flotation (Sumikura *et al.* 2007). It was found that NB flotation (negatively charged) is a cost-effective process for the mechanical and chemical treatment of wastewater (Tsai *et al.* 2007). MNB experiments recorded a 40% increase in the efficiency of wastewater treatment, with about 95% removal of solid waste and silica. Besides this, the cost and performance of flotation technology using NBs are significantly lower than in normal processes (Han & Dockko 1998; Haarhoff & Edzwald 2001; Han *et al.* 2006).

### Disinfection process

Pollutant oxidation and pathogen disinfection using ozone is a promising method for purifying wastewater (Zhang *et al.* 2013; Khuntia *et al.* 2015). Studies show that gas bubbles of ozone due to their powerful disinfection ability successfully treat water even with short contact time and low concentration. The effectiveness of this method has frequently increased its application for chemical-resistant spore-forming bacteria (e.g., *Cryptosporidium parvum* and *Bacillus subtilis*) (Kim *et al.* 2004; Aydogan & Gurol 2006). Further, MBs make this process more efficient, as kinetic disinfection demonstrates a faster decrease rate of *Escherichia coli* (bacteria type) (99% reduction), with a relatively small water tank and a lesser amount of ozone (for applying MBs) than the normal ozone disinfection process (Sumikura *et al.* 2007). Another experiment to stop the multiplication of *E. coli* resulted in a 75% reduction within three minutes using 490 W/L energy (Mezule *et al.* 2009). Similarly, hydrodynamic cavitation results from various experiments proved the efficiency of such a non-reagent method as MNBs for water disinfection (Jyoti & Pandit 2003).

Studies further showed that MBs eliminate *Bacillus subtilis*, and result in higher log reduction (e.g., reducing by 5 log by 140 mg/L, reducing by 1.6 log by 110 mg/L and reducing by 0.3 log by 40 mg/L in a period of two minutes (Zhang *et al.* 2013)). Further experiments on bacterial deactivation showed that the small-sized bubbles' burst force



determined the efficiency of water disinfection (Kim 2010). Chen (2009), during application of disinfection of swimming pools, observed a steep decline of pathogens. Specifically, in this experiment, two rotation structures attached the reservoir with the bathtub, while using NBs of size 10–20 nm. The experiment revealed that a higher burst of NBs resulted in better disinfection compared with other methods such as an ultrasonic vibrator.

Moreover, the advanced oxidation process (AOP) has the potential to convert most organic compounds into  $\text{CO}_2$ , which is achieved either by the direct reaction of molecular  $\text{O}_3$  (Wilderer 2010; Khuntia *et al.* 2015) or by the indirect reaction of oxidation of the hydroxyl radical formed by  $\text{O}_3$ . Notably, the AOP-based  $\text{O}_3$  systems dominated by radicals such as hydroxyl radicals (OH) have shown improved water purification efficiency (Lucas *et al.* 2009; Wilderer 2010). In such structures, compound oxidation of OH is highly reactive to many organic pollutants due to greater oxidation potential (2.80 V) compared with direct oxidation (2.07 V) (Lucas *et al.* 2010). However, reaction mechanisms need to be improved for the overall effectiveness of disinfection.

The literature shows that the ozone-based AOP process is significantly associated with water properties such as alkalinity, type of biological substance, pH, and temperature (Glaze *et al.* 1993; Siddiqui & Amy 1993; Tang *et al.* 2005). For instance, water pH is a critical factor for the decomposition of  $\text{O}_3$  (Tomiya *et al.* 1985; Sehested *et al.* 1991; von Gunten 2003). Although MBs increase the amounts of OH and  $\text{O}_3$ , which improves disinfection efficiency, no research has been conducted on the generation of by-products during disinfection. Likewise, more work is required for the case of NBs. More work is also required concerning the impact of MNBs on the efficiency of  $\text{O}_3$  use, and reactor optimization and temperature reduction effect.

## APPLICATION OF MNBS IN VARIOUS SECTORS

During recent years, due to the promising features of MNBs, their application has been extended to various fields such as agriculture, medical, industry, aquaculture and domestic use. Table 2 provides an overview of the application of MNB technology in various fields.

## FUTURE TRENDS

Due to unique characteristics, micro-nanobubbles have attracted more and more attention in recent years. Until now, the mechanisms of MNB generation, i.e., the generation of bubbles that displace air, creating bubbles that form the air, and producing bubbles using electrolysis, have been studied. The use of MNB generators in mineral buoyancy, water treatment, reducing the resistance of ships, drug administration, and thermal jet printing are widely observed (Deng *et al.* 2014). However, there are certain areas where the trends of MNB application seem more promising.

For instance, due to the enhanced time-cost efficiency and lower chemical consumption, the application of MNB has great potential in environmental sustainability in addition to water treatment. The distinctive properties such as higher mass transfer rate, collision efficiency, lower rising velocity, customized surface charge, and radical generation give MNBs a more promising role in future techniques of water treatment such as disinfection and flotation.

In such treatment, high time-cost efficiency is predicted in terms of higher organic pollutant separation, flotation, aeration efficiency for biological treatments, and advanced oxidation using OH radicals. The chemical-free radical generation property has vast potential in ozone-based MNB uses for the oxidation process. Further, the higher bursting energy of MNBs and higher aeration efficiency also have subsequent potential in terms of reducing membrane fouling and sludge formation in membrane bio-reactors.

In addition, future research on MNB technology may also focus on the optimization and automation of MNB generators to customize the MNB generation mechanism in various sectors, including industry and agriculture. For instance, improved smart-technologies-based automation and control may facilitate the precise application of MNB-based water treatment in agriculture and fulfill the growing demand for water treatment at various scales.

Lastly, despite the vast potential of MNBs as a smart, efficient, and cost-effective treatment technology, its application is still limited to the laboratory. Hence future research and projects need more focus on its development and application at the agricultural and industrial scales.

**Table 2** | Application of MNBs in different sectors

Area	Application
Agriculture	<p>MNB-treated water in agriculture improves the soil's physiological and biological conditions by encouraging aerobic microorganisms, which improves soil particle structure, water absorption and oxygen dissolution, levels of the rhizosphere, microbial species and phosphate and urease, which positively impact plant growth (Takahashi <i>et al.</i> 2007; Teixeira <i>et al.</i> 2010).</p> <p>NB water reduces CH<sub>4</sub> emission and arsenic dissolution through an oxidative shift of the redox conditions in the flooded soil (Minamikawa <i>et al.</i> 2015).</p> <p>MNBs accelerate metabolism in animal and plant species (Liu <i>et al.</i> 2013a; Ebina <i>et al.</i> 2013).</p> <p>MNB use in hydroponics solutions for cleaning and sterilization of irrigation water (ozone bubbles) (Takahashi <i>et al.</i> 2007).</p> <p>The high oxygen content and permeability of MNBs promotes plant root growth, enhances nutrient absorption, shortens the growth cycle and hence improves economic efficiency (Nakano <i>et al.</i> 2009; Inatsu <i>et al.</i> 2011).</p> <p>MNB application through a drip kills bacteria, removes harmful substances and odors from water, and improves freshness and taste and yield of fruits and vegetables (Takahashi <i>et al.</i> 2007).</p> <p>MNB use for biological and weed control (i.e., facilitating Triopsidae growth, which stops weed growth in rice fields and also decreases chemical and fertilizer usage) (Serizawa 2017).</p> <p>MNB water improves the rate of seed germination (Liu <i>et al.</i> 2013b).</p>
Aquaculture and fisheries	<p>MNBs improves blood flow and branchial respiration of fish (Serizawa 2017).</p> <p>MNBs can be applied to the purification of sludge at the sea bottom (the air is supplied to sludge in the form of nanobubbles, which can recover the poor oxygen condition at the sea bottom, activate marine life and decompose organic substances) (Serizawa 2017).</p> <p>MNB-treated water application on aquatic plants and fisheries significantly increases growth by improving nutrient uptake (Cho <i>et al.</i> 2019).</p>
Cellular biological	MNBs are used in fermentation (Marui 2013).
Medical	<p>MNBs are used to diagnose tumors in the human body by ultrasonic imaging (Yin <i>et al.</i> 2012).</p> <p>MNBs are used for curing cancer patients in different forms and treatment methods (Lukianova-Hleb <i>et al.</i> 2014; Orel <i>et al.</i> 2017).</p> <p>MNBs are used in dental treatment and the conditions that affect the teeth and gums (insertion, repair, and extraction of teeth) (Hayakumo <i>et al.</i> 2014; Gulafsha &amp; Anuroopa 2019).</p> <p>MNBs are used for the detection of malaria (Kreft &amp; Jetz 2007; Rebelo <i>et al.</i> 2016).</p> <p>MNBs are used in genetics (drug-oxygen-gene delivery nexus) (Cavalli <i>et al.</i> 2009; Thakur <i>et al.</i> 2019).</p>
Industry energy systems	<p>MNBs eliminate mixed oil and carbon from water and provide economic benefits for water reuse (dissolving air with polyelectrolytes with hydrogen MNBs) (Tansel &amp; Pascual 2011).</p> <p>MNBs are used in solar energy (solar vapor nanobubble generation as a result of the complex interaction of several phenomena that occur at the nanoscale and can be used in a variety of applications, i.e., solar steam energy NB generation) (Polman 2013).</p>
Domestic uses	<p>Oxygen produced by ozone decomposition generates ions that can oxidize pollutants in drinking water (Batagoda <i>et al.</i> 2019).</p> <p>MNBs are used in laundry and tableware. Their use in swimming pools, showers, and bathtubs for strong antioxidant effects have subsequent health benefits such as removal of aging skin, deep-rooted dust particles, bacteria, and chemical residue, which accelerate skin metabolism, and blood circulation (Nessbert 2017; Nikusystec 2019).</p>

## CONCLUSION

MNB is a promising technology, and its application has spread significantly over the last decade to a wider range of sectors covering energy, environment, industry, agriculture, and aquaculture. This review article mainly presents the existing context and the state of research on MNB use in various areas of water

treatment applications such as in flotation technology, disinfection, aeration, and advanced oxidation. The application of MNB technology, especially in agriculture and aquaculture, is highly promising with abundant opportunities. Although the current status of research and practice is making significant progress gradually, development is steady. For instance, most of the literature available on MNB application is based on

water-related applications in major primary sectors such as agriculture and aquaculture; however, expansion is needed for sectors such as medical science.

Further literature shows ambiguity in terms of fundamental properties, such as stability of bulk NBs, which have not been well explained until now. Only a few experimental studies deal with the stability and longevity of NBs, with no considerable consensus. The studies have yet to decide whether properties such as separation and stability follow scientific guidelines (classical thermodynamic principles). Although progress has been made and several hypotheses have been proposed explaining the long-term stability of NBs, none of them describe these experimental observations.

Lastly, this study seeks further investigation regarding improvements in performance techniques, bubble growth, and size under various physical and chemical conditions, bubble generation methods, and automated optimization.

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