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Characterization of air submicrobubbles for water treatment under different generation conditions

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Micro-nanobubble (MNB) technology has been proven to be effective in water and wastewater treatment. Submicrobubbles (SMBs) are considered to be a subgroup of MNBs ranging from 1 to 10 µm with limited studies related to their fundamental properties. This study focused on the fundamental characteristics of SMBs and the effect of generation conditions such as temperature, aeration time, and water volume on their properties. SMBs were generated under high pressure using shear force and their size and distribution were measured using the dynamic light scattering method. Dissolved oxygen (DO) concentrations were monitored during and after the generation. The zeta potential of the generated bubbles was also measured to assess bubbles stability. SMBs with a median diameter of 2 µm persisted in water even after the generator was stopped, indicating the high longevity of SMBs in water. Regardless of the aeration time or water volume used, the zeta potential of SMBs was highly negative with average values ranged between − 28 and − 30 mV, indicating high stability in water. DO concentration increased by up to 1.5 folds within a few minutes of generation and slightly decreased over 1 h. Results demonstrate that air SMBs are stable with long lifespan and high DO concentration.

Keywords Submicrobubbles, Bubble properties, Bubble size distribution, Water treatment, Wastewater treatment, Aeration process

Micro-nanobubbles (MNBs) are micro/nanometer diameter-sized bubbles. The potential use of MNBs in several scientific and technological fields has gained attention in recent years, particularly in environmental engineering[1]. MNBs have proven to be a promising technology in applications such as groundwater remediation[2,3], wastewater treatment[4,5], surface water treatment[6], biochemical process enhancement[7], environmental pollution control[8], ecological restoration[9], food process[10], agricultural processes[11,12], aquaculture, and medical applications[13]. Specifically, MNBs have proven to be effective in water/wastewater treatment for the removal of organics[14,15] and oils[16], disinfection and sterilization processes[17,18], and surface cleaning[19]. In addition, MNBs have been applied in various processes, including flotation[20], aeration[21], and membrane processes[22].

The increase in MNB application in water treatment is related to the following advantages, including (1) reduced chemical usage; (2) significant potential for cost reduction in operations and design[23]; (3) low cost, convenience, and environmental friendliness for cleaning of conducting surfaces[24]; (4) slow rise velocity; (5) high mass-transfer efficiency; (6) high specific surface area; (7) generation of free radicals; and (8) longevity[4,23]. In contrast, conventional macrobubbles (millibubbles) rise rapidly to the liquid surface and burst at the air–liquid interface. Therefore, macrobubbles have different physicochemical properties than small-sized bubbles[25].

In several studies, the unique characteristics of MNBs enabled them to outperform conventional macrobubbles. Nam et al. (2019) showed that microbubble (MB) ozonation is superior to conventional macrobubble ozonation in terms of generating higher concentrations of hydroxyl radicals and ozone in aqueous solutions[26]. Another study reported better performance for the degradation of bio-refractory organic compounds for MB ozonation compared to macrobubble ozonation[27]. Sun et al. (2020) showed that the reaction rate constant of MB ozonation in removing petroleum hydrocarbons from oily sludge was twice that of macrobubble ozonation[28].

The efficiency of MNBs in treating polluted effluents in terms of biochemical oxygen demand (BOD) and chemical oxygen demand (COD) was evaluated in a systematic review and meta-analysis[29]. The diameters of the MNBs used ranged from 0.01 to 70 µm. The review concluded that MNBs could remove BOD_5 and COD

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at efficiencies ranging from 68% to 100%. In addition, the review found that MNBs removed up to 99.9% of inorganic and microbiological pollutants. Several studies have investigated the effect of using MNBs to reduce solids or pollutants in different water sources. All studies demonstrated high efficiencies of NBs or MBs in the reduction of total dissolved solids (TDS)[30], turbidity, BOD, and COD[31].

Air MNBs have been utilized in several studies to enhance treatment process performance. For example, Li et al. (2022) confirmed that air MNBs provide enhanced normalized fluxes of natural organic matter during the ultrafiltration separation of feeds containing various contaminants[32]. Under similar experimental conditions, the results with MNBs were significantly better than those obtained without MNBs. Air MNBs have been successfully applied in reverse osmosis desalination processes to improve membrane performance, and control gypsum scaling[22].

Air MNBs have been used in biological wastewater treatment to improve aeration process. Ahmadi et al. (2022) determined that a nanobubble (NB) aeration system could significantly enhance the measured treatment efficacy parameters for a sequential batch reactor[33]. NB and MB aeration also enhanced gas mass transfer compared with conventional aeration in the activated sludge process. Therefore, they could be considered efficient upgrades to the current activated sludge process using conventional macrobubble aeration. This is reflected in the reduced energy consumption owing to the improvement in oxygen transfer, as well as easier organic and nutrient removal[21]. A recent study confirmed that MNBs are a great choice for treating blackening and odorization in rivers when combining activated sludge and biofilms. They demonstrated that MNB aeration was better than macrobubble aeration, with a 12-fold higher oxygen-transfer efficiency and 52.6% lower oxygen decline rate[34]. In addition, MNB aeration resulted in 50% less energy consumption compared to macrobubbles and showed higher oxygenation performance[35]. Therefore, MNB aeration has greater potential for biological wastewater treatment than macrobubble aeration. Furthermore, the oxygen-transfer efficiency was improved by decreasing the bubble size.

Air MNBs have shown potential for use in various wastewater treatments, either alone or in combination with other processes. One study applied MNBs and activated hydrogen peroxide to study the degradation of tetracycline in wastewater. The degradation rate of tetracycline hydrochloride reached 92.43%, and the main reactive oxygen radical was •OH, followed by HO_2•/•O_2-[36]. Another study applied MNB aeration to enhance the efficiency of Rhodamine B degradation during heat-activated persulfate oxidation. The MNBs successfully accelerated the reaction rate and increased the DO concentration. In addition, the combined system stably generated the radicals •SO_4^- and •OH, enhancing Rhodamine B degradation[37].

Because the size of the bubbles plays a crucial role in their fundamental properties, different sizes will lead to different properties, which will be reflected in the treatment. Although some researchers describe MNBs as small-sized bubbles with diameters on the nano- and micrometer scale[1], others specify MNBs as bubbles in the range of 200 nm to 10 μm[25,38]. The MNBs generated in this research will be referred to as submicrobubbles (SMBs), which lie in the range of 1–10 μm[23].

Most of previous studies on MNBs characterization in water focused on MBs (10–100 μm) or NBs (< 1000 nm), whereas research on bubbles in the 1–10 μm range (SMBs) is limited. Temesgen et al. (2017)[23] highlighted that bubbles with diameters ranging from 10 to 100 μm burst in liquids and take minutes to rise to the surface. This behavior differs from bubbles ≤ 1 μm, which swell, burst in the liquid, and take hours or even weeks to rise to the surface due to Brownian motion. Temesgen et al. (2017)[23] indicated that the properties of SMBs lie between those of MBs and NBs. MBs are suitable for several applications including dissolved air flotation especially within the range of 30–50 μm because their micro-size promotes the adhesion of bubbles to water particles and gives suitable bubble collision efficiency. NBs within the range 700–900 nm are close to being stationary in water making them suitable for several applications such as aeration and advanced oxidation processes due to increased mass transfer rates because mass transfer depends on bubbles surface area and rising velocity[39]. As a result, it is expected that SMBs, with their size lying between MBs and NBs, will remain for a reasonable time, enabling efficient treatment. SMBs also have large interfacial area, slow rising velocity and high gas solubility for enhanced aeration process. In addition, when using SMBs it is expected to avoid problems related to over-aeration or very fast rising velocity that can affect water treatment process. Although some studies used air SMBs in their treatment process[4], and some studies examined some of SMBs characteristics[40]; up to our knowledge, the investigation of SMBs basic properties in relation to their experimental conditions such as aeration time and water volume remains underexplored. This study focuses solely on, the under-investigated, SMBs properties to understand their behavior and identify their suitable applications in water and wastewater treatment.

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The main aim of this research is to investigate the properties of air SMBs generated using shear force and high pressure under different experimental conditions. The investigated properties include longevity, stability, and DO concentration levels, and the experimental conditions include aeration time and water volume. To achieve this aim, this paper investigates the impact of the experimental conditions on (1) the size and distribution, (2) the longevity and DO concentration, and (3) the stability of SMBs. Accordingly, the research aims to fill in the gap in bubble characteristics between NBs and MBs. Future applications in water and wastewater treatment are also considered. Subsequently, the zeta potential, which plays a crucial role in bubble stability, was measured. In addition, DO enhancement was studied for future applications of submicrosized bubbles. Finally, the main challenges encountered during bubble generation are highlighted.

Materials and methods

A framework for characterizing and investigating air SMB properties is illustrated in Fig. 1 and detailed in the subsequent sections.

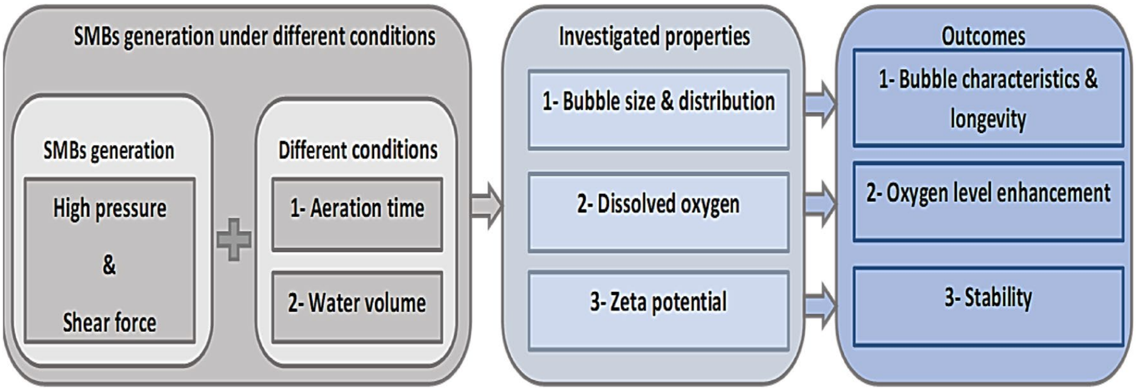


Fig. 1. Overview of applied experiments and investigations.

SMB generation

SMBs were generated in ultrapure water (18.2 MΩ cm) that was produced using a Milli-Q Direct 8 water purification system (Merck) and was analyzed before bubble generation to avoid conflicting results. Considerably small particles of ≤ 10 nm were excluded from the analysis. Bubbles were produced using Newmantech MNB generator, developed in 2021, equipped with a self-priming pump and pressure vessel. The generator technology relies on the shear force to form small gas bubbles. A nozzle was conncected to the outlet pipe of the generator at the bottom of the aeration tank. Air was introduced into the generator, and its flow rate was adjusted using a flowmeter while water was pumped from the water tank. In the generator, the gas-water mixture was introduced at high pressure, and then the mixture passed through the nozzle in the water tank. The mixture of water and air was introduced to the generator at a pressure of 0.42–0.44 MPa. The volumes of water used were 10, 15, and 20 L, and the gas flow rate ranged from 0.2 to 0.4 L/min. The experiments were conducted at room temperature with an initial water temperature of 18 ± 0.5 °C.

Bubble size, size distribution, and zeta potential

A Nanotrac Wave II Nanoparticle Size Analyzer (Microtrac MRB) was utilized in this research. The analyzer is capable of directly measuring particle size, electrophoretic mobility, and the resulting zeta potential (ζ), which relies on dynamic light scattering (DLS).

The analysis was adjusted for the particles as air with a refractive index of 1. Each experiment was conducted at least three times. Using the Analyzer, particle size, size distribution, and zeta potential of the samples were measured using DLS. The bubble size distribution figures provide a summary of the bubble size measurements. The bubble size distribution curves were compared to understand the behavior of the bubbles after the generation had ceased.

The ζ measurements ranged from − 200 to + 200 mV. The zeta polarity is determined during the ζ measurement, where + or − indicates a positive or negative polarity, respectively. The limit of the particle-size determination is in the range of 0.8 nm to 6.5 µm. The median diameter or 50th percentile was calculated from measurements of the average particle size.

DO concentration and temperature

Temperature and DO concentration were measured using a DO meter (HACH HQ 40 d multi-probe meter) at certain intervals. The meter can measure the DO concentration in the range from 0.1 to 20 mg/L and the temperature in the range from 0 to 60 °C.

Experimental procedures

SMBs were generated and the generator was kept for 5 min after reaching stability. The water tank was covered during bubble generation. Water samples were collected at intervals of 1, 3, and 5 min. Samples were collected for different aeration times and water volumes. The sample cell was cleaned and dried before sample measurements. The samples were analyzed using a particle size analyzer; each sample was measured three times, and the average of the readings was reported. Various samples were collected for DO measurements. These samples in a glass container were sealed to prevent DO loss at room temperature. The DO concentration was measured at different aeration times, water volumes and after 30 and 60 min to examine DO longevity. The temperature was monitored during bubble generation. The samples were characterized using a particle size analyzer at the specific time intervals. The samples were then kept sealed for 1 h to determine SMBs lifespan. The zeta potential was measured using a particle size analyzer. This can be summarized as (1) bubble generation using a generator with DO and temperature monitoring, (2) bubble characterization for size, size distribution, and zeta potential, and (3) longevity check for bubble size, distribution, and DO measurements. Experiments were repeated for different aeration times and water volumes. The experimental set up is shown in Fig. 2.

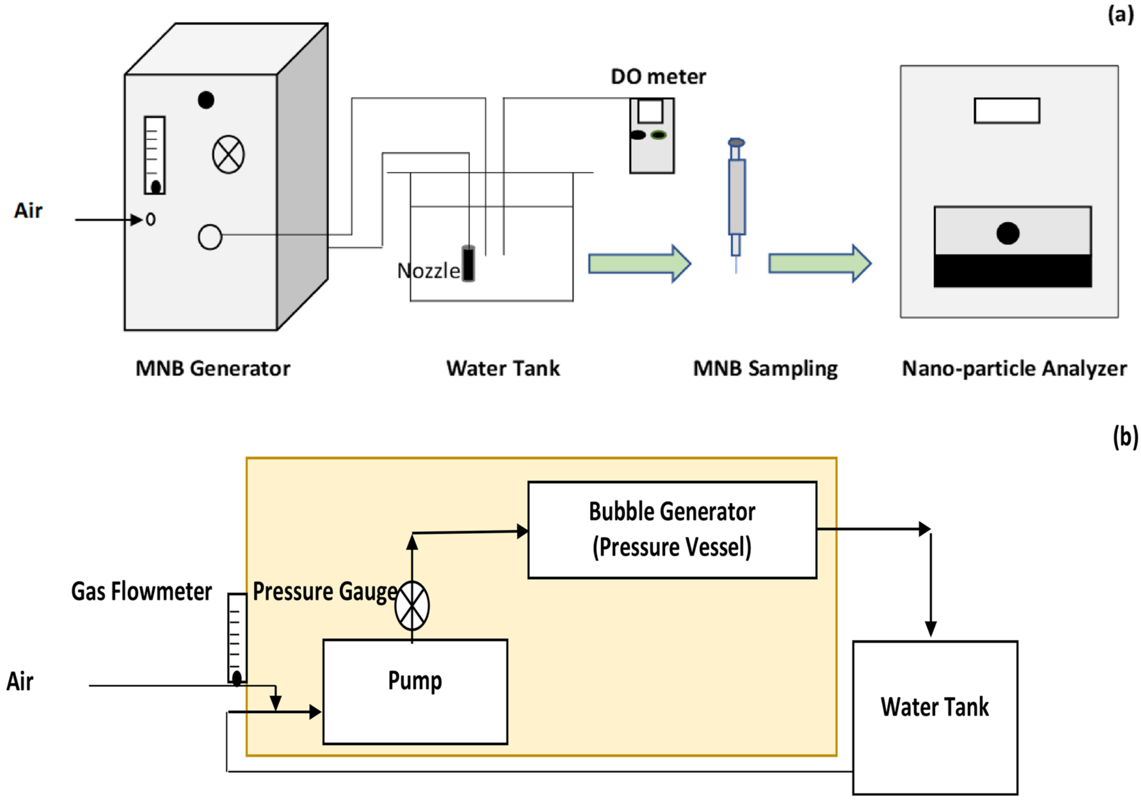


Fig. 2. (**a**) Experimental procedure for bubble generation and analysis. (**b**) Principle of generation.

Results and discussion

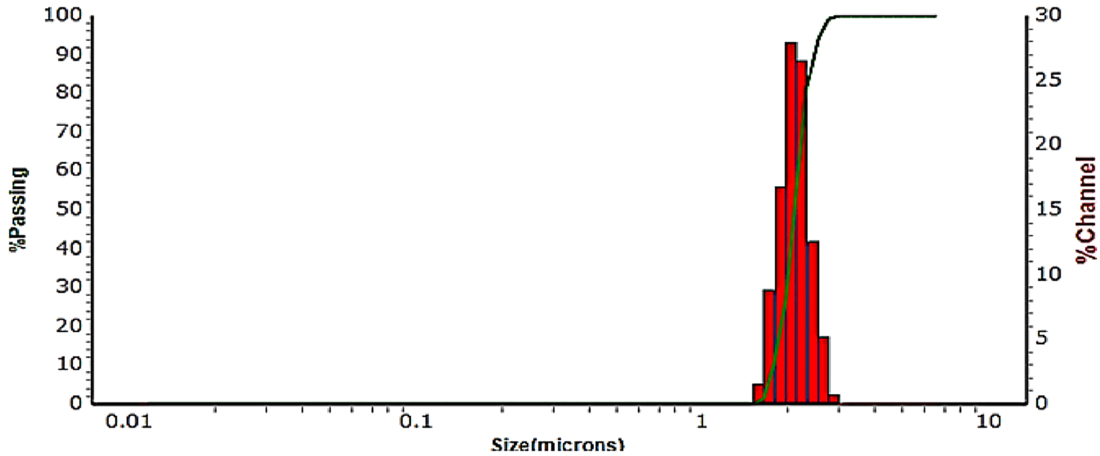
Bubble size and size distribution

The bubble size distribution of air SMBs using 15 and 20 L of water over 5 min of continuous aeration was analyzed. Samples were collected at different times to determine the effect of aeration time on bubble size. Overall, the generated size of the bubble was analyzed and found to fall in the range of 1.5 to 3 µm with a median diameter of ≈ 2 µm using the two volumes with standard deviations of 0.2498 and 0.246 as shown in Fig. 3. The generated bubble sizes at different times and water volumes are shown in Fig. 4. The generated bubbles sizes for both volumes did not change at the three selected times even when increasing aeration time or volume; however, the peak of the bubble size distribution curve increased as the aeration time increased up to 3 min. At 5 min, the peak slightly decreased. The temperature of the water increased by increasing the aeration time to 5 min due to the increase in the temperature of the generator. The average temperature increase for the two volumes used over 5 min of continuous aeration is shown in Fig. 5. The temperature increase was higher in smaller volumes than in larger ones.

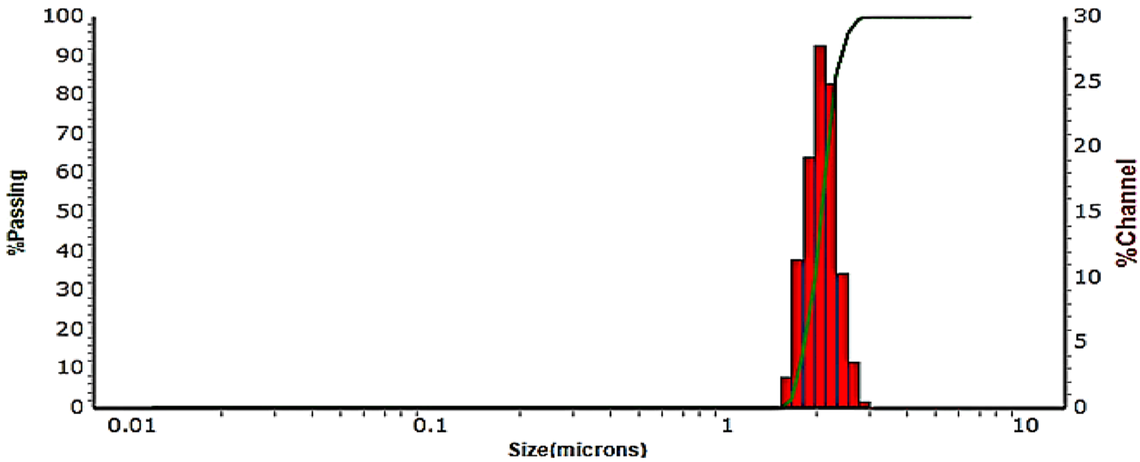
When the bubbles were generated in the smaller water volume, 10 L, the generated bubble size fluctuated; probably because water is circulated more times than the case of larger volumes. With more water circulation, temperature increases and, consequently, the size of the bubbles is affected. Accordingly, producing a desired specific size range for the bubbles was a challenge for the small volumes. Therefore, a different range of bubble sizes is detected at the three-time intervals. The range of bubble sizes detected was between 200 nm and 2 µm—for all aeration times. Therefore, a wide range of bubbles was detected but not a specific size. The varying bubble sizes generated using 10 L samples are shown in Fig. 6. Consequently, the detection of other specific properties of the bubbles for this volume captures a wide range of bubble sizes (MNBs) and not the target-specific range. As the aim of the study was to characterize submicrobubbles within the range of 1–10 µm, other properties were excluded from the analysis.

Bubbles longevity and distribution

The sealed samples were checked individually after 30 and 60 min. Figure 7 shows the size distribution of the bubbles generated in 15 L of water for 1, 3, and 5 min of generation. From this figure, it is evident that bubbles with aproximatly the same size were observed after 30 min, with nearly the same size distribution. Specifically, most bubbles remained stable for at least 30 min in all samples of all aeration times. Although some bubbles were still present after 60 min, their size distribution fluctuated. The stability of the bubbles after 60 min depends largely on aeration time. For example, 1 min aeration showed nearly the same bubble size even after 60 min, with a shorter peak bubble-size distribution (Fig. 7a). In contrast, 3 and 5 min aeration showed mostly different bubble sizes after 60 min without a specific trend (Fig. 7b and c, respectively). Specifically, some bubbles had smaller sizes over time while others merged or had larger size. The smaller bubbles that appeared after 1 h can



(a)



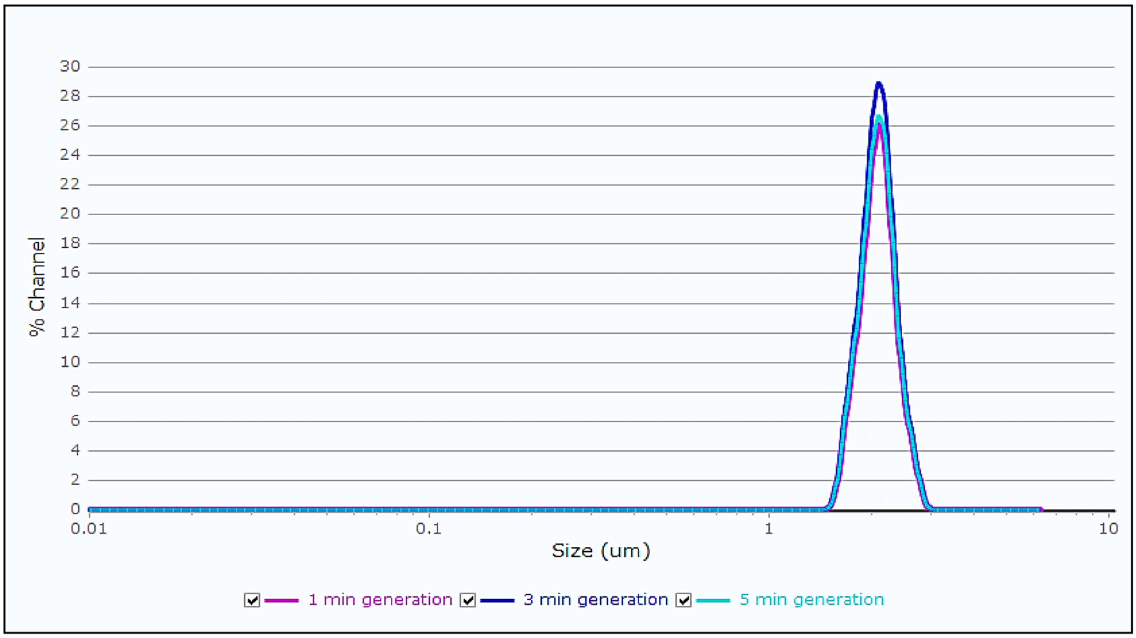
(b)

Fig. 3. Generated bubble size at 3 min in water volumes of (**a**) 15 L and (**b**) 20 L.

result from gas diffusion over time. The formation of larger bubbles, on the other hand, could be attributed to the decrease in the concentration of bubbles over time as well as the variation in water temperature with increasing aeration time when using 15 L of water. Another explanation could be the merging of bubbles because of using a smaller water volume.

SMBs remained stable and detectable after 30 min. After that, shorter peaks of different sizes were detected. Apparently, generating bubbles under high pressure helps maintaining their stability for a longer period. This was also confirmed by Wan et al. (2001), who reported that the generation of MBs under high pressures reduces their dissolution[41]. Different studies have reported various observations regarding bubble size changes over time. Li et al. (2014) indicated MNBs less than 50–65 µm shrink over time, in contrast to macrobubbles which enlarge over time[1]. Shekhar et al. (2013) mentioned that micro-sized bubbles shrink over time owing to gas diffusion[42].A recent study on SMBs confirmed that their size increased from 4.5 to 7 µm over 2 h [43]. Another study succeeded in maintaining stable air bubbles, using surfactants, with diameters between 1 and 2 µm for up to 5 h[44]. By contrast, a study examining the stability of nanosized bubbles showed that the bubble size unexpectedly increased with time[45]. Zhou et al. (2023)[46] studied the stability of air bubbles with sizes of 1–10 µm using shear force under high pressure based on the intensity of the light scattering. They concluded that the observed high intensity of water treated using air could be attributed to a more stable entity.

In conclusion, it is challenging to predict the general duration of bubbles longevity in pure water[43]. In addition, the behavior of bubbles after generation is influenced by different factors, such as (1) a decrease in bubble size due to high initial water depth[47] and high internal pressure, which promotes gas diffusion from areas of high partial-pressure inside the bubbles to areas of low partial-pressure in the surrounding medium[48,49]. (2) Bubble size can increase due to bubble movement caused by Brownian motion, leading to bubble coalescence, a decrease in the zeta potential over time[50], and a variation in the temperature of the water such that increasing the temperature decreases the water surface tension, leading to an increase in bubble size fluctuation[51]. According to Yang et al. (2023), MBs and NBs show all types of size changes: cluster, collapse, and shrinkage[52].



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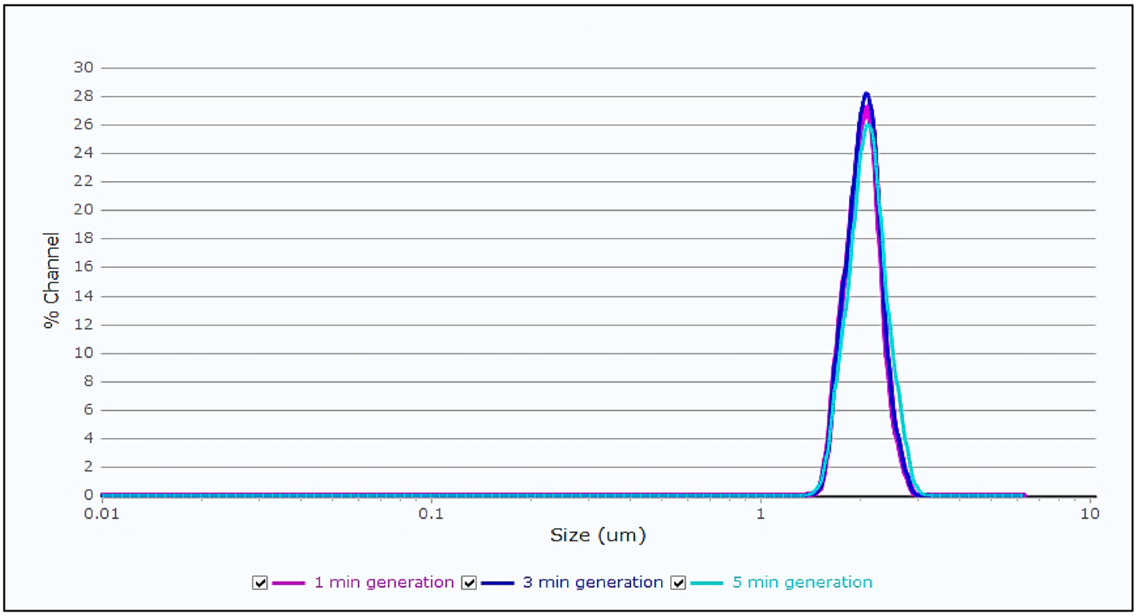


(b)

Fig. 4. Generated bubble size distribution at 1, 3, and 5 min using water volumes of (**a**) 15 L and (**b**) 20 L.

The size distribution of air SMBs using 20-L of water is shown in Fig. 8. This larger water volume used here resulted in a relatively smaller rise in temperature. An increase in water volume indicates a higher initial water depth with more space for bubbles to be generated in the water. Similar to the 15-L water volume, bubbles of the same size were detected after 30 min, while changes were observed at 1 h. In contrast to the 15-L water volume, a longer aeration time (5 min) showed better consistency with time than a shorter time (1 min). This means that for higher volumes, a longer aeration time was required with careful monitoring of the temperature variation. After 60 min, the bubbles became smaller for 1 min aeration (Fig. 8a), while the 3- and 5-min aeration samples contained bubbles of nearly the same sizes, with shorter distribution peaks. In some cases, smaller bubble sizes reaching ≈ 1.6 µm with shorter peaks were observed (Fig. 8b-Run 7 and 8) and (Fig. 8c-Run 1,3,5 and 6). In contrast to the 15-L sample, the sizes of the bubbles did not increase with time. Therefore, after 1 h, either the bubbles remained similar in size, with shorter peaks or became smaller in size and reached 1.6 µm. Larger bubbles were not detected. This can be explained by the lower disturbance associated with the larger volumes used, which prevents bubble coalescence/merging.

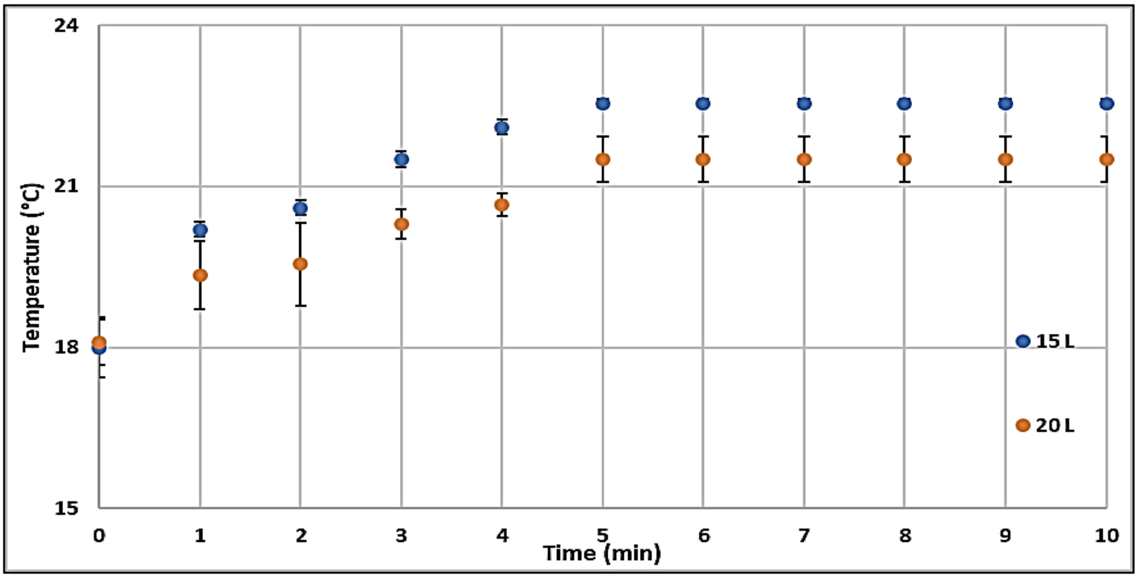


Fig. 5. Variations of water temperature with time for different water volumes.

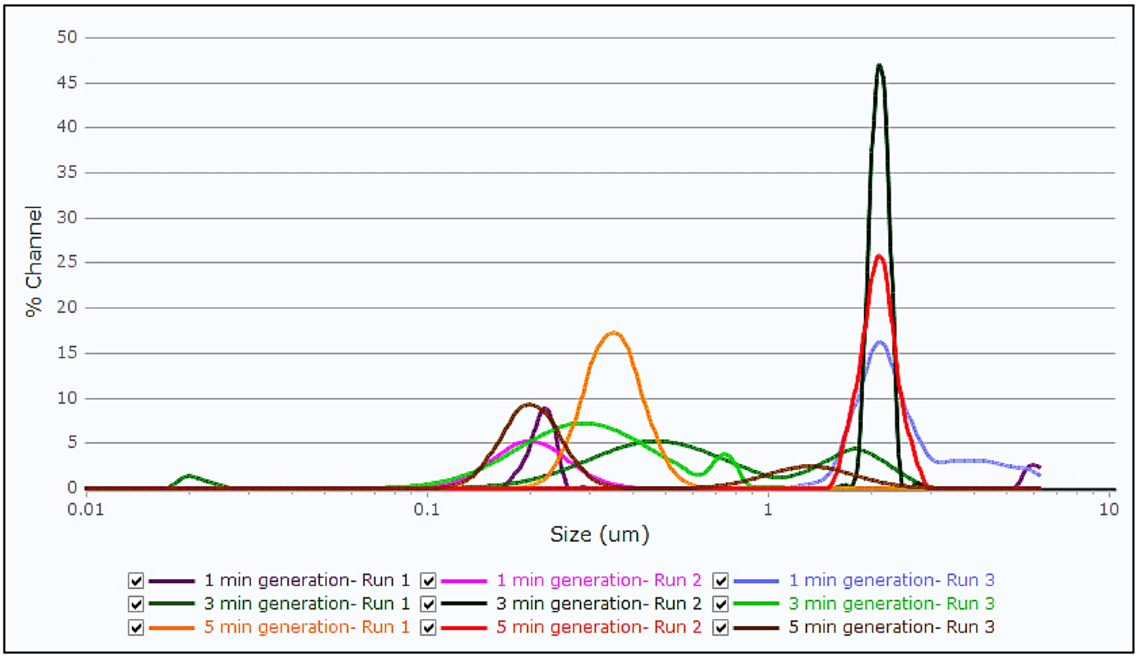
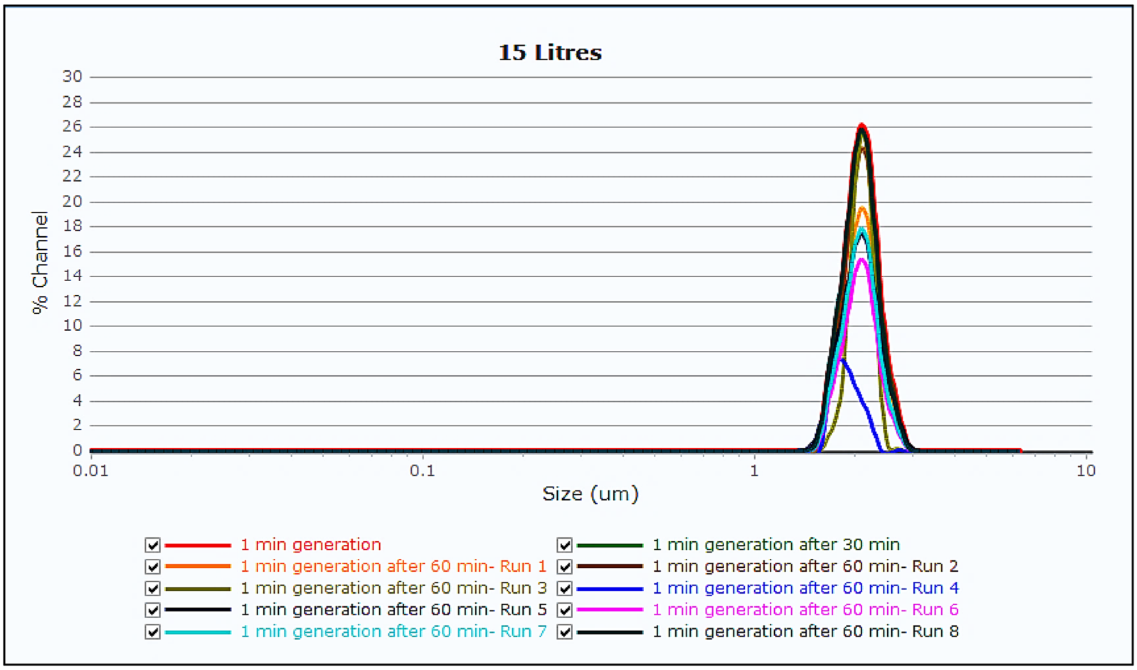


Fig. 6. Bubble size range obtained using 10 L of water.

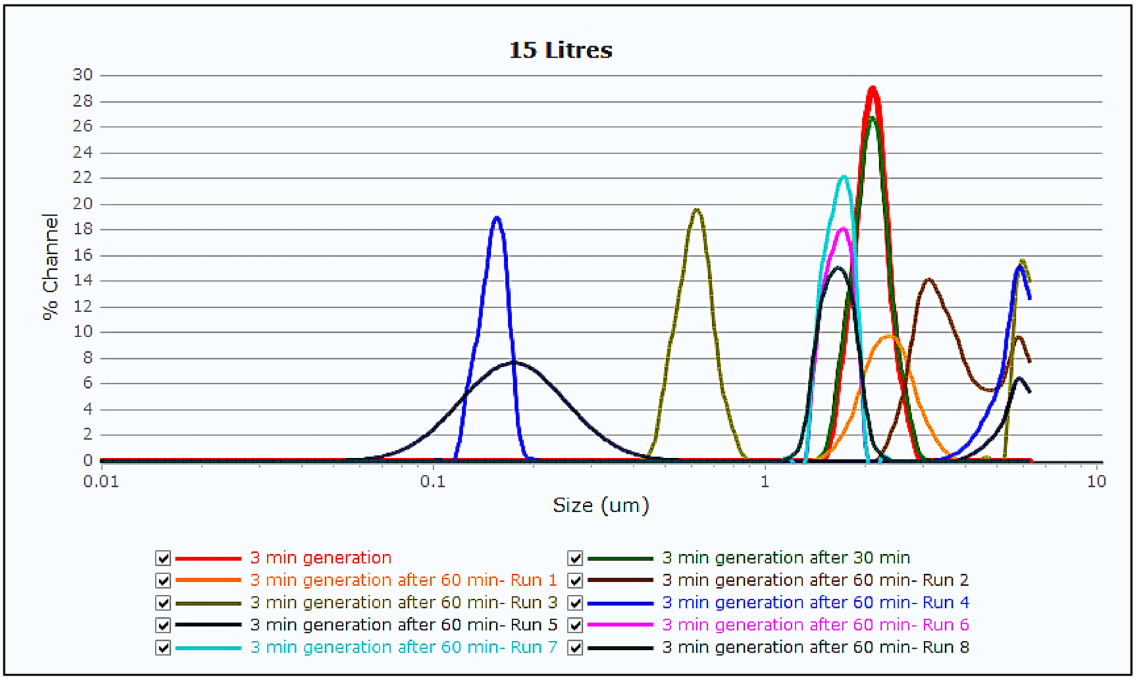
The rising velocity of the bubbles inside the liquid can be expressed using Stokes' law, which states that the rising velocity is directly proportional to the size of the bubble[48]. It should be mentioned that bubbles > 7–8 μm follow Stokes' law, while bubbles < 7–8 μm rise more slowly than the calculated values[53]. This implies that the rising velocity is slow when the bubble diameter is smaller. The retention time can also be affected by the depth of the water. Consequently, more bubbles remain in the water when bigger volumes are used.

DO concentration

The changes in DO concentration of the SMBs, using different aeration times and water volumes, are shown in Fig. 9. This figure shows that the initial concentration of the DO for pure water was 8.66 ± 0.16 mg/L. The same figure shows DO concentration increased when aeration time was increased. The peak value of the DO was higher when 20 L of water was used. This is mainly related to the higher temperature increase in smaller volumes. Also, the higher depth of water leads to a longer retention time. In addition, it is evident that, for all



(a)

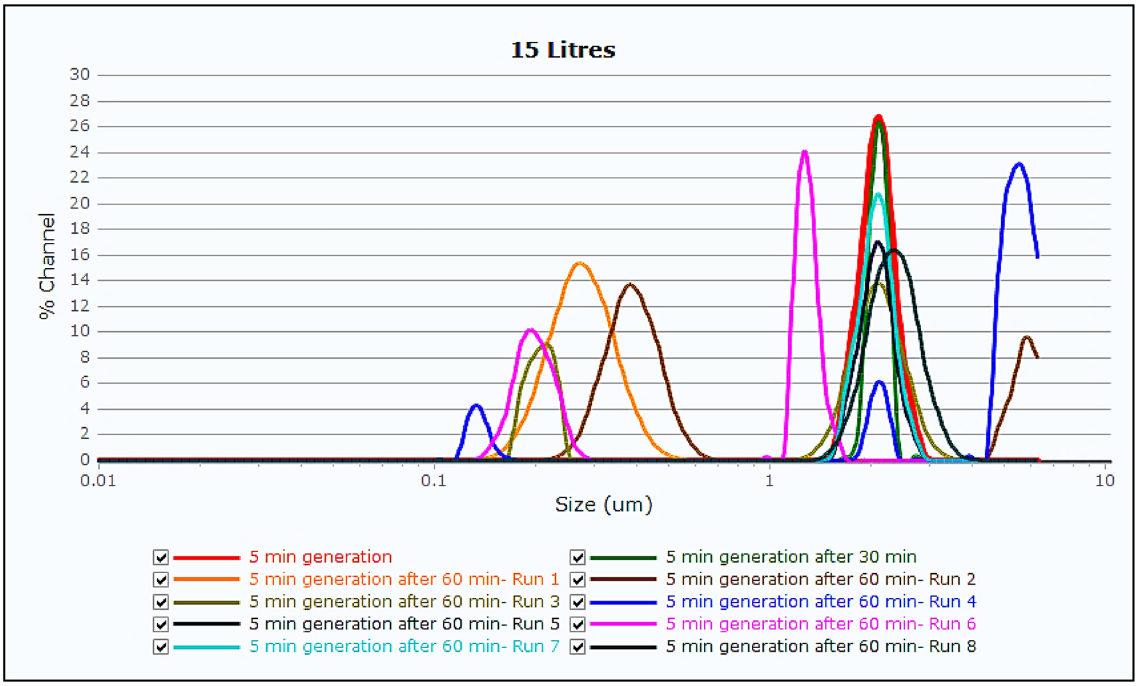


(b)

Fig. 7. Size distribution of bubbles generated at 15-L after (**a**) 1 min of generation and its detection at 30 and 60 min, (**b**) 3 min of generation and its detection at 30 and 60 min, (**c**) 5 min of generation and its detection at 30 and 60.

aeration times and volumes, the DO concentrations remained higher than the initial concentration after the generator was stopped for the monitoring period.

The average DO concentrations were 12.84 and 12.66 mg/L after 5 min of aerating 20 and 15 L of water, respectively. After the generator was stopped, the DO concentrations began to decrease, with the greatest decrease occurring directly after the generator stopped in the first 5 min. The DO then decreased gradually for 1 h. Even with the DO decrease, the values were higher for larger volumes than for smaller volumes. Although the temperature increased by 3.5–4.5 °C during the first 5 min of aeration in the two water volumes used, a



(c)

Fig. 7. (continued)

DO concentration increase was still noticed. This means that a small increase in temperature did not affect the increase in DO for the same aeration time.

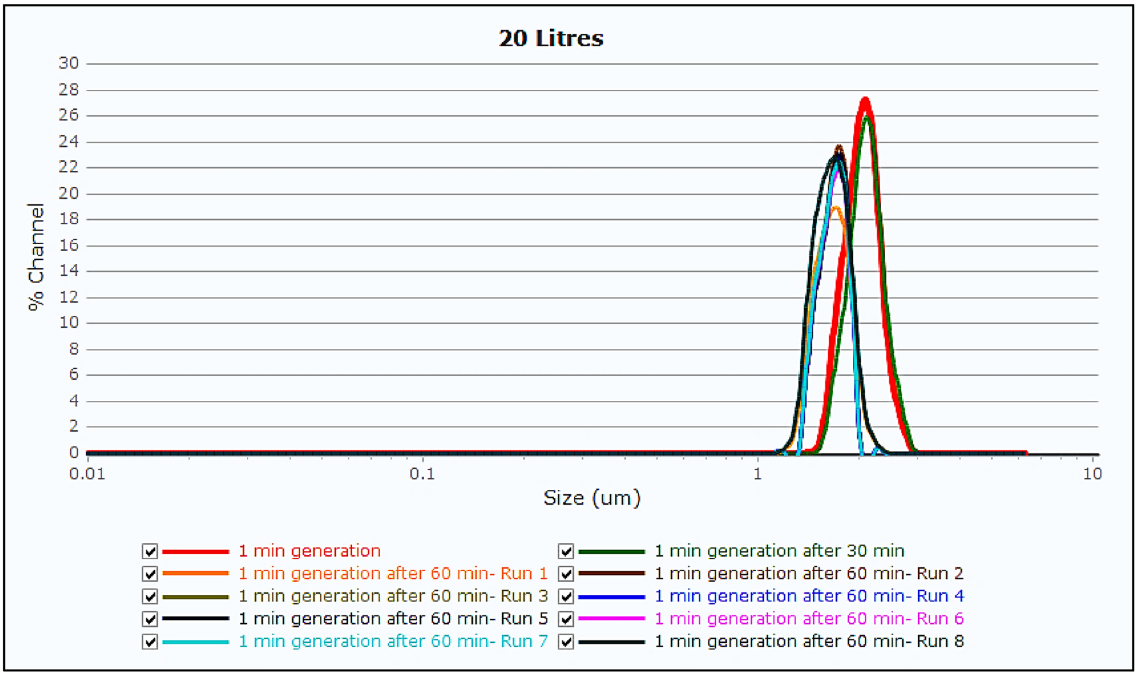
In conclusion, within only a few minutes, the DO increased by 4.2 mg/L. The highest increase in DO concentration always occurred during the first minute of aeration for all aeration times and volumes and was concomitant with the minimum increase in temperature. Overall, the highest reduction in DO concentration after the generator was stopped occurred during the first 5 min. The average decrease in DO did not exceed 0.8 mg/L during the 1 h monitoring time.Different factors can affect the DO concentration. According to Ushikubo et al. (2010), as time passes, the oxygen inside the bubble dissolves in water and then diffuses into the atmosphere[54]. The stability of bubbles can be extended when the initial DO concentrations are higher because the time for oxygen diffusion will be longer.

Zeta potential

Zeta potential (ζ) is an important parameter for studying MNBs because it indicates surface charge measurements that affect stability. This also determines the longevity of the bubbles. The zeta potential is a measure of the magnitude of electrostatic repulsion[4]. Therefore, the higher the absolute value of ζ, the higher the stability of the bubbles. On the other hand, lower absolute values of ζindicates instability of the bubbles, which will result in bubble coalescence[55]. Different factors affect the value of ζ in air bubbles, including (1) pH of the water: higher pH indicate more negative ζ; (2) temperature of the water; and (3) type and concentration of the surfactant, which affects the intensity of ζ and the nature of bubble surface charges[56].

The measured ζ for air SMBs showed that their average values ranged from − 28 to − 30 mV under pH of 6.6 ± 0.2, as shown in Fig. 10. All the zeta potentials, measured for the three aeration times, were negative. It was shown that the zeta potential did not depend on the aeration time, as all measured values were in the same range, with minimal variation. The ζ was also measured using two different volumes of water (15 and 20 L) and had the same range of values. Therefore, the values of ζ depend neither on the aeration time nor the volume used, but rather depends on the size of the bubble formed. As the generated bubbles had nearly the same size among all aeration times and volumes, the measured values of ζ had the same range. This is in agreement with other findings reported in the literature[56,57]. However, it should be highlighted here that under different operating conditions and factors, the absolute value of ζ could change.

The measured values of ζ here were consistent with those of other studies. A recent study, for example, showed that the ζ values of air MNBs in ultrapure water were negative and ranged between − 10 and − 30 mV although under different operating conditions. It was concluded in that study that the aeration rate affects ζ values such that the higher the aeration rate, the lower the potential charge. In their measurement of ζ, they were mainly focused on nanosized-range bubbles. It should be mentioned here that after the preparation of MNB solution, they used NaCl as a background electrolyte before running the zeta potential measurement[58]. Nirmalkar et al. (2018) showed that ζ value of air NBs in pure water was about − 28 mV[59]. Montazeri et al. (2023) found that ζ value was − 19.48 ± 1.89 mV for air NBs in deionized water[60]. Graciaa et al. (1995) showed that ζ at the air–deionized water interface is − 65 mV[61]. Jadhav et al. (2021) showed that bulk NBs in pure water had a ζ value of



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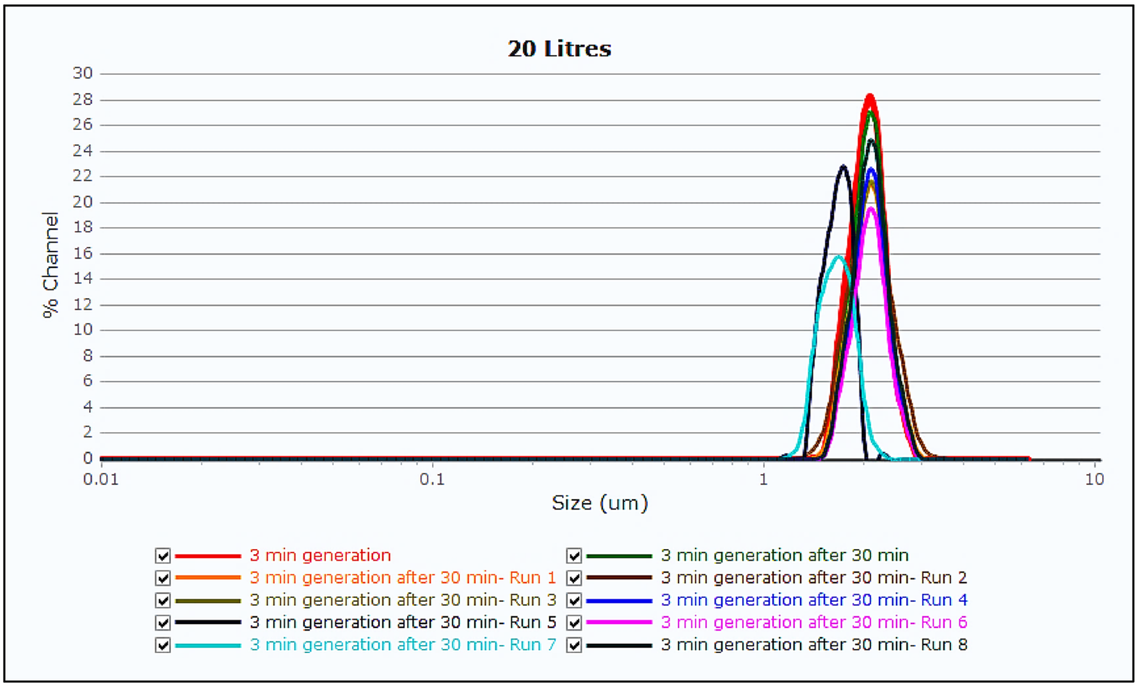
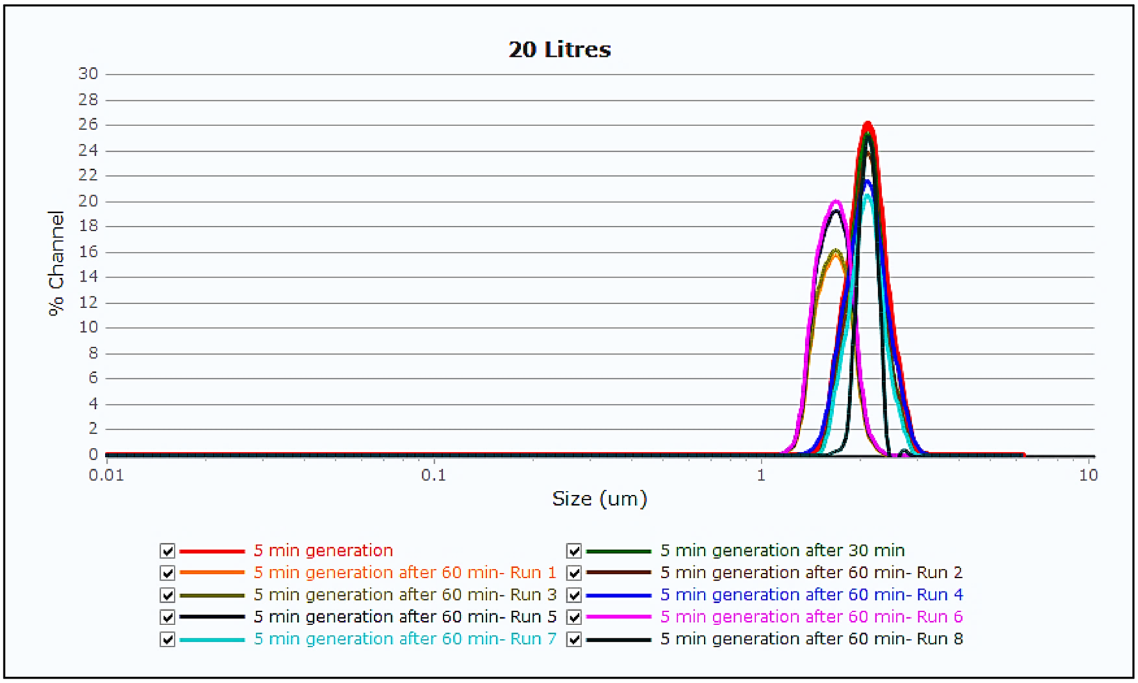


(b)

Fig. 8. Size distribution of bubbles generated at 20-L: (**a**) 1 min of generation and its detection at 30 and 60 min, (**b**) 3 min of generation and its detection at 30 and 60 min, (**c**) 5 min of generation and its detection at 30 and 60 min.

– 25 mV[62]. Air bubbles in the nano- and microsized range had negative ζ values in ultrapure water and deionized water, with absolute values of 17–20 mV and 15 mV, respectively[53,54,63]. However, Takahashi (2005) showed that the zeta potential of MBs is – 35 mV regardless of their size in distilled water with dissolved ambient CO_2[64]. Zhou et al. (2021) found that air NBs in deionized water had long-term stability over 60 d; in their study, the zeta potential of the air NBs reached – 32 mV[65].



(c)

Fig. 8. (continued)

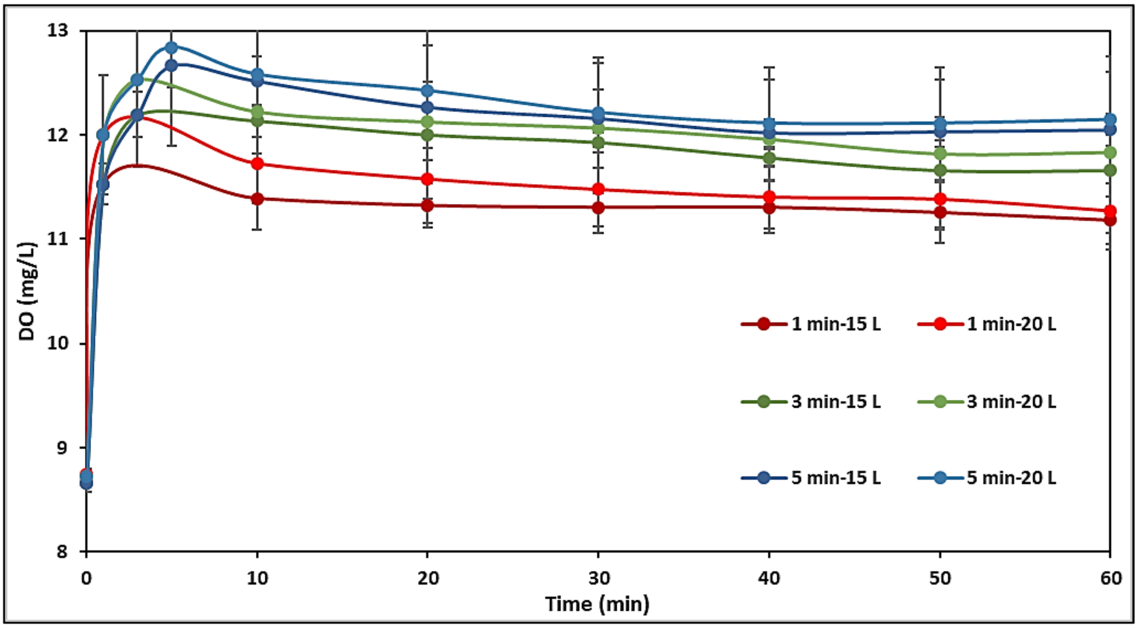


Fig. 9. DO concentration of SMBs at different aeration times and volumes and their longevity after generator stoppage.

Some of the previously mentioned values are presented in Table 1 with respect to bubbles classification. These values were compared to those obtained in this study. The values could differ from one experiment to another, depending on the overall experimental conditions. This is common among all experiments in which charged surfaces are present, whereas the absolute value depends more on the experimental conditions. Regardless of the absolute value of ζ, the charged MNBs will be repelling each other, thus preventing bubble coalescence. This helps to retain bubbles in the solution for a longer time. Negatively charged bubbles, with their longevity in water, assist in the treatment process. In addition, high DO contents in water facilitate processes such as

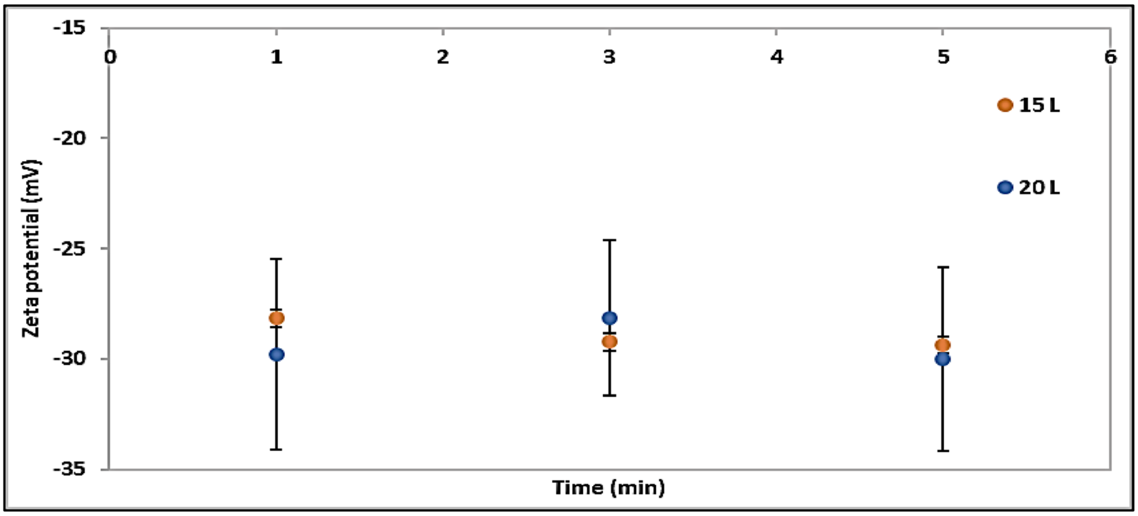


Fig. 10. Zeta potential of SMBs at different aeration times and volumes.

Bubble classification	Water type	Aeration time (min)	Gas flow rate	T (°C)	pH	ζ measurement	ζ (mV)	Bubble generation method	Ref.
NBs	Ultrapure water (0.1mM NaCl)	0 to 10	0 to 80 mL/min	25	…	Zeta potential analyzer (Omni, Brookhaven, USA)	−10 to − 30	Hydrodynamic cavitation	[58]
NBs	Pure water	…	150–300 mL/min	…	6.5	ZEN5600 ZetaSizer Nano ZSP instrument (Malvern instruments)	−28	Hydrodynamic cavitation in a high-pressure microfluidic device	[59]
NBs	Deionized water	60	170 L/min	20	6.7	BI-SCP disposable plastic cuvettes and a BI-ZEL electrode assembly from Brookhaven Instruments Corporation	− 19.48 ± 1.89	Venturi which generates MBs then cavitation cylinder which generates NBs	[60]
NBs	Ultrapure water	30	1 L/min	20	6.7	Zetasizer Nano ZSP instrument (ZEN5600, Malvern-UK).	−25	High-shear rotor-stator device	[62]
NBs	Ultrapure water	30–40	0.1 L/min	20	6.1 -6.4	Zeta Potential Analyzer (Zeecom, Microtech Co. Ltd., Japan).	−17 to − 20	High pressure in a pressurized tank then depressurization of the gas-supersaturated water led to the nucleation of the bubbles	[54,63]
SMBs	Ultrapure water	1,3,5	0.2–0.4 L/min	18 ± 0.5	6.6 ± 0.2	Nanotrac Wave II analyzer (Microtrac MRB)	−28 to − 30	High pressure and shear force	This study
MBs	Deionized water	15	240 mL/min	…	…	DelsaNano C zeta potential analyzer (Beckman Coulter, Inc., US).	−15	Spiral liquid flow type	[53]
MBs	Distilled water (dissolved ambient CO_2)	…	…	…	5.8	Calculated	−35	Microbubble aerator using water pump, centrifugal force and shearing force	[64]

Table 1. Zeta potential of different experiments using different analyzers and generation methods.

aeration, which require a high oxygen content. Accordingly, the use of MBs/SMBs/NBs is more beneficial than conventional processes.

The physical mechanism of bulk NB stability has been explained by Zhang et al. (2020)[66]. Charge enrichment is caused by the significant affinity of negative charges for the NB interface, which results in electric field energy that gives rise to a local minimum in the free-energy cost of bubble production and thermodynamic metastability of the charged NBs. A size-dependent force is generated mechanically by excess surface charges. This force serves as a restoring force when the NB is thermodynamically disturbed from its equilibrium state and balances the Laplace pressure. Further studies need to be conducted on the physical mechanism of the stability of bulk MNBs using different bubble size ranges.

Special characteristics and challenges of bubble generation

Aeration is a common process used to treat wastewater. Recently, MNBs have shown several advantages that can enhance conventional aeration processes. The unique characteristics of SMBs have been confirmed, including (1) the longevity of the generated bubbles after stopping the generation, as shown by the size distribution for air SMBs with a specific refractive index of air; (2) an increase in DO contents that remained high, even after the generator was stopped, in contrast to macrobubbles; and (3) high values of negative zeta potential measured with SMBs. Charged surfaces help avoid bubble coalescence. Ushikubo et al. (2010) related these evidences to the stability of bubbles[54].However, it should be mentioned that a change in the experimental conditions can cause changes in the bubble size, distribution, longevity, repeatability, and other characteristics. Therefore, the generation of MNBs requires careful examination of all experimental conditions to ensure the repeatability of the results, as presented below.

The experimental conditions, including water volume and temperature, should be examined when studying bubble generation in terms of size, distribution, and longevity. With the same flow rate, a smaller volume of water was associated with a higher temperature. According to Park et al. (2020), temperature changes have an impact on the life of a bubble[67]. For a larger volume of water, bubbles exhibited more stable behavior. Consequently, avoiding a rapid temperature increase by carefully choosing the suitable volume of water can assist in maintaining stable bubbles. The selection of a suitable bubble generator size based on the water volume to be treated will help in achieving the required results.

Moreover, selecting a suitable aeration time is required, especially when a longer aeration time is needed when using a larger water volume. It should be mentioned that other experimental conditions could influence the behavior of the bubbles formed as mentioned previously in the literature including type of gas used (plays an important role in the repeatability of the results[68]), gas flow rate (an important factor that affects the bubble size[69]), generation methods, measurement techniques, as well as the purity, temperature, and pressure of the water that affect the stability and lifespan of bubbles[43].

Conclusion

The generation of air SMBs under high pressure and shear forces was confirmed using a DLS method particle analyzer. Bubble generation was conducted using different aeration times (1, 3, and 5 min) and water volumes (15 and 20 L). The results showed that (1) SMBs of the same size were generated under all aeration times and volumes used; however, this was more evident for 20 L. (2) Increasing the aeration time when using a larger water volume, and vice versa, helped producing stable bubbles. (3) SMBs have distinctive properties that make them more stable in water. Specifically, the small size and charged surface of SMBs enable them to remain present in water for longer periods even when using short aeration times. (4) The DO concentration increased by 4.2 mg/L of its initial value within a few minutes of aeration using 20-L of water volume. An increase in water volume from 15 to 20 L resulted in higher DO concentrations from 12.66 mg/L to 12.84 mg/L after 5 min of continuous aeration. By increasing the aeration time, the DO continued to increase, but after the generator was stopped, a gradual decrease in the DO concentration was observed. After 1 h, the average DO concentration decreased by a maximum of 0.8 mg/L. (5) The zeta potential was highly negative under all aeration times and water volumes. However, its absolute value did not depend on aeration time or water volume, but it depended on the size of the bubble formed. These results indicate directions for future investigations, highlighting the promise of SMB in the aeration process as a valuable treatment method.

Data availability

The datasets used and/or analysed during the current study available from the corresponding author on reasonable request.

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Declarations

Competing interests

The authors declare no competing interests.

Additional information

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