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Research Article

Determination of bubble size distribution in gas–liquid two-phase systems via an ultrasound-based method

Over the past few years, ultrasonic techniques are increasingly used to determine bubble size distribution in gas–liquid two-phase systems. However, the development of a precise and efficient measuring system is very challenging because bubbles are dynamic and unstable relative to time and space in a moving fluid, thus hindering an accurate validation of the measuring system. Therefore, this study examined the possibility of using artificial bubbles to establish an ultrasonic measuring system. The main concept for this study involved developing an ultrasound-based measuring system with the aid of a certain type of artificial bubbles. The developed system was subsequently adapted to the bypass pipeline of a propagator to demonstrate the reliability of this concept. The results indicated that the established system could measure a microbubble size distribution with a root mean squared error of validation that corresponded to 0.1243 % v/v. Additionally, the estimates of real bubble measurement agreed well with the reference method. Thus, this study demonstrated that the developed system could be a potential method to determine bubble size distributions in gas–liquid two-phase systems.

Keywords: Attenuation / Bubble size distribution / Optics / Process monitoring / Ultrasound

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1 Introduction

Determination of the bubble size distribution in a gas–liquid two-phase system plays an important role in chemical engineering, biochemical process, and food production [1–5]. This is because in certain processes in these areas, such as aerobic fermentation, gas is dispersed into liquid in an agitated vessel or bubble column in which gas–liquid mass transfer occurs. The transfer rate in turn is critical to control and monitor the dispersion process [1]. The gas–liquid mass transfer rate is derived by the liquid-side volumetric mass transfer coefficient that is related to the specific interfacial area. In order to obtain this value, it is necessary to consider gas volume fraction and this depends on a measure of the bubble size distribution [1, 6]. Furthermore, the determination of the bubble size distribution together with

other factors, such as bubble velocity, contributes to optimizing design and operation conditions for bioreactors and bubble columns since bubble size and the local distribution of bubble size play important roles in system hydrodynamics and process performance [7]. Conversely, with respect to food production, bubble size distribution is closely linked to structural and sensory properties of the final product [8]. However, the bubble size distribution strongly relies on production conditions including pressure, agitation speed, and temperature [9, 10]. Hence, it is very important to understand the relationship between these operating variables and bubble characteristics to modify final product quality. Therefore, it is necessary to consider information on bubble size distribution to control processes, production amount, and product quality in a gas–liquid two-phase system [5].

There are various approaches to determine a bubble size distribution. The most commonly used approaches are based on photographic [4, 5, 11, 12], electroresistivity [13], optic [2, 9, 14–16], and acoustic techniques [17–24]. However, such approaches have limited applicability due to changes in rates of bubble collapse and bubble concentrations with respect to time.

The photographic technique uses a camera to obtain bubble images in a container. The captured images are then analyzed

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Abbreviations: US, ultrasonic; HELOS, Helium-Neon Laser Optical System

using specific image-processing software to calculate the bubble size distribution. Result accuracy is highly dependent on image resolution, which typically relies on the quality of the applied digital camera. However, this method has two limitations. First, the camera must be placed outside the container to avoid contact with the fluid area, and thus the exterior of the container should also be transparent such that the camera can capture the images of the bubbles. Second, bubbles can be clustered and coalesced when bubble concentration within a container is too high. Thus, camera-captured images may not be sharp and could thereby be difficult to process. Thus, utilization of the photographic method is suitable only if the measurements are performed in a dilute suspension system [4]. The method is based on the electrorositivity principle, and despite the possibility of measuring high bubble concentration, the measurement of bubble distribution is inaccurate, because the sensor is inserted into the sample and this can interfere with the motion and behavior of the bubbles [13]. Typically, the laser refraction technique is applied to determine bubbles with a small size in a diameter range of 10–800 μm . This is only suitable for a small sampling volume and automatic data analysis [20].

The optical microscopy technique also allows measurements of the bubble size distribution [9]. This technique is most frequently used to measure smaller bubbles with a diameter ranging from 3 to 150 μm . Nevertheless, this technique relies on optical lens configuration, focus depth, and the image caption rate. Thus, an estimation of the size distribution relies on the measurement of the bubble size as well as bubble number, and a problem arises when fast-moving bubbles are present. Additionally, it is also difficult to automatically isolate overlapping bubbles [11].

Recently, increasing attention is focused on the use of ultrasonic techniques to measure bubble size distribution. In contrast to other existing techniques, an ultrasound-based measuring system is capable of rapid and continuous sampling, which is advantageous for samples that contain fast-moving bubbles [25]. Furthermore, it is not necessary for an ultrasonic sensor to be inserted into the sample and touch sample during measurements. Thus, the interference of the motion and behavior of bubbles can be avoided. Additionally, ultrasonic techniques can be applied in an optically opaque sample in the gas–liquid as well as gas–liquid–solid system, and therefore has a much wider application range [26]. Hence, several studies have used ultrasonic techniques to determine bubble size distribution for inline process monitoring [27–29]. Most of these studies involved experiments under various laboratory conditions that typically consisted of the following three parts: a bubble generation system to obtain a bubbly fluid, an acoustic-based system for bubble distribution measurements, and a reference system to verify acoustic results. The use of the ultrasound to determine the bubble size distribution was comprehensively reviewed by extant research [17]. The theory of measuring bubble size distribution relies on different arrangements of ultrasonic sensors and includes the following three relationships between bubble population density and the acoustic quantities: backscattering, attenuation, and dispersion. The aforementioned relationship can be used to estimate bubble population density, $n(a)$, with different models by measuring the ultrasonic velocity and attenuation coefficient.

A multifrequency backscatter system was used to measure bubble size distributions for investigating air–see interaction processes [30]. The system included six frequencies of acoustic sensors in the range of 29–400 kHz. The estimation reliability was limited by the void fraction that was less than 10^{-5} .

Measurements using the attenuation relationship are broadly accepted and used by several studies to estimate bubble size distribution [23, 31, 32]. Studies indicate that there is a good level of agreement between photographically and acoustically determined bubble size distributions. The use of numerical techniques can enhance measurement accuracy and avoid errors caused by the approximation method. An iterative approach to calculate an exact bubble size distribution was investigated in a previous study [21]. The approach combined a resonant approximation model and an integral model such that complex mathematical procedures could be avoided.

A research group (DYNAFLOW) developed a commercially available acoustic-based instrument, namely the Acoustic Bubble Spectrometer® (ABS) [29]. Bubbles were generated using either an electrolysis method or the method of injecting air through porous tubes. The obtained acoustic results were compared with microphotography results. The developed technique also solved the ill-posed integral equations using the constrained optimization technique. However, it was difficult to obtain an accurate estimate of bubble size distribution. Larger errors (greater than 10 times the number of over estimates or underestimates) could therefore occur due to the mismatch between the model and measurement conditions [23]. Conversely, the bubble size distribution is dynamic in terms of time and space in a moving fluid environment, and it is difficult to obtain static bubbles that are of importance in analyzing and validating the investigated ultrasonic (US) method [28].

Therefore, this study proposed a new method to obtain static bubble samples. In the proposed method, real bubbles were simulated by artificial bubbles known as 3M Microbubbles™, which corresponded to air-filled bubbles with a shell of certain waterproof material and a radius range of 5–87 μm . This product was not studied to-date. However, the present study hypothesized that this product was suitable to simulate real bubbles in an ultrasound-based measuring system.

The objectives of this study include the following: first, the development of an ultrasound-based measuring system with multiple frequencies to estimate a laboratory-scale bubble size distribution; second, obtaining a method of simulating bubbly fluid by using an artificial product with similar characteristics and providing the required bubble size distribution with a constant experimental setup; third, testing the established ultrasonic measuring system in a bubbly fluid to verify the feasibility of the developed system.

2 Materials and methods

2.1 Sample preparation

Artificial bubbles with a predetermined size distribution range are required to perform the new experimental method to detect bubble size distribution. This was accomplished using a type of commercial hollow glass spheres, namely 3M™ Glass

Figure 1. Photograph of the microbubble-water mixture with a concentration of 0.1 %v/v as captured by a digital microscope (Keyence VHX-2000 with a VH-Z500R Zoom lens).

Table 1. Volume fraction of the samples using K1 microbubbles

Volume concentration (%v/v)	Microbubble mass (g)	Mass of total mixture (g)
0.1	0.0625	498.084
0.2	0.1250	497.648
0.3	0.1875	497.212
0.4	0.2500	496.776

Bubbles K1. These microbubbles have a radius in the range of 5–87 μm and a mean radius of 30–35 μm . This product consisted of 90% air and was stabilized by a soda-lime-borosilicate glass wall. There are two advantages of using these microbubbles. First, they are stable and water resistant. Second, they are of low density (0.125 g/cm^3) with approximately 90% of air, and thereby behave more like the air bubbles than other products such as ultrasound contrast agents and other glass bubbles. Figure 1 presents an image captured using a digital microscope (Keyence VHX-2000 with a VH-Z500R Zoom lens) to visualize the structure of the microbubbles. In the photograph, it was clearly observed that the microbubbles were uniform and had a spherical shape. The inside of each microsphere was hollow and filled with air such that the middle part of each microsphere was white in the captured image.

The sample was prepared by mixing microbubbles with distilled water to produce a mixture with a required concentration. Table 1 shows the volume fractions of the samples. Distilled water was first heated by using a heating plate until the water was boiled for approximately 5 min. After the water was cooled to room temperature ($22 \pm 1^\circ\text{C}$), a certain amount of the microbubbles was mixed to reach the desired volume fraction. The sample mixture was then kept in a water bath to maintain the experimental temperature. The experiments were conducted to measure the attenuation coefficient of the frequency domain and ultrasonic velocity of time domain. The samples were measured under volume concentrations of 0.1, 0.2, 0.3, and 0.4 %v/v.

2.2 Ultrasonic measurement

2.2.1 Microbubble measurement

The measurements of microbubble size distribution were performed using an experimental setup that was composed of the following two parts: an ultrasonic measuring system and a sample circulation system. Figure 2A shows the complete experimental setup. The ultrasonic measuring system was equipped with an in-house developed ultrasound generation device that was composed of a personal computer, a microcontroller, an analog–digital converter (Transio A52, MOXA[®]), and a power supply box. The developed system utilized an ultrasonic transmission technique that employed two pairs of ultrasonic transducers (first pair: in-house built, frequency range: 500 kHz transducers; second pair: UP-DW, frequency range: 2–200 kHz; GEOTRON[®], Germany). A PT-100 sensor was used to measure the average temperature of fluid because an ultrasound is sensitive to temperature. The rectangular impulses of 30 V amplitude and 900 ns length were sent to the ultrasound-transmitter when the ultrasonic sensors were connected with the microcontroller that transmitted ultrasound signals to the sample. The ultrasound signals propagating through the sample were preamplified (10 dB) prior to being received by the ultrasound receiver. Finally, the received ultrasound signals passed through the analog–digital converter and were then stored in the computer. The sampling frequency of the stored ultrasonic data corresponded to 50 MHz.

The ultrasonic measuring system was controlled by a software that was developed in-house (Virtual Expert[®]). This software served as the interface that could realize real-time visualization of both ultrasound and temperature signals. Additionally, the software was used to input the aforementioned ultrasonic measurement operation parameters including excitation voltage, sampling frequency, impulse duration, and gain of preamplifier to generate an ideal ultrasound signal. This software also allowed simultaneous data acquisition of both temperature and ultrasound signals.

First, measurements using distilled water were performed. The aim of these experiments included obtaining a reference ultrasound data and the reference distance. The reference distance corresponded to the accurate distance between the ultrasound transmitter and the ultrasound receiver, and the value of this distance was required later to estimate the attenuation coefficient and ultrasonic velocity.

In the case of the 500 kHz transducers developed in-house that do not have matching layers, the reference distance was calculated according to the following equation:

$$d = USV \cdot t_1 \quad (1)$$

where USV denotes the ultrasonic velocity in pure water that can be calculated using an equation used in a previous study [33], and t_1 denotes the time of taken by an ultrasonic signal to propagate only once across the path between two transducers and is determined by calculating the first zero crossing point prior to the first rising peak as denoted by the red circle marked in Fig. 2B.

Thus, it was necessary to calculate the reference distance in the case of the commercial UP-DW transducers, which have matching layers with unknown thicknesses. The reference distance for

Figure 2. (A) Illustration of the experimental setup. Circulation of a microbubble–water mixture in the circulation system is performed through the circulating route marked with the dashed lines. In this manner, the sample mixture flows through the measuring tube via the route 11→13a→12→13b→7→13c→11 in order to remain homogenous. The measuring tube is a Plexiglas cylinder cell with a volume of 196.3 mL. (B) Estimation of the TOF in a signal in which the red point denotes the first zero crossing point.

Figure 3. (A) Schematic diagram of the experimental configuration for real bubble measurements. The ultrasound-based measurement configurations. Two transducers are embedded in a cylindrical bypass pipeline. (B) Illustrations of the experimental configuration of the photographic imaging system.

the UP-DW transducers was estimated based on the following equation:

$$d = \frac{1}{2} \text{USV} (t_2 - t_1) \quad (2)$$

The sample was maintained homogeneously using the sample circulation system that was composed of a magnetic stirrer and a peristaltic pump.

Following the completion of the reference distance measurements, the microbubble–water mixture was distributed and collected into three glass bottles for reference measurements using a laser diffraction instrument known as the Helium-Neon Laser Optical System (“HELOS”) particle size analyzer (HELOS/KR, Sympatec GmbH; HELOS: Helium-Neon Laser Optical System) to determine the reference data of microbubble size distribution.

2.2.2 Real bubble measurement

In order to validate the performance of the developed ultrasonic system, real bubble measurements were performed in a pipeline that was equipped with a gas injector (“TurboAir”; Fa. ESAU & HUBER GmbH), which used an air supplier as a yeast propagation tank. After supplying the air, the bubbles were generated by the nozzle of the gas injector and were then mixed with

water to obtain the bubbly fluid. The experimental configuration was equipped with ultrasonic and photographic capabilities as shown in Fig. 3.

As shown in Fig. 3, the bubble size distribution measurements using both methods were performed at the same position, which was located 20 cm away from the outlet of gas injector in a pipeline. Specifically, the central part of the experiment position was composed of two glass walls with a viewing area of $26 \times 26 \text{ mm}^2$ and could therefore be used by the photographic system to capture the bubble image. Air was dispersed into water with an aeration rate of 1.8 L/h to generate the bubbles. Due to maximal frame rate of the photographic imaging system, the air bubbles were generated at this relatively low aeration rate to make the air volume sufficiently small such that the bubble shape could be detected clearly by the camera system within the viewing area. All experiments are conducted at a temperature of $16 \pm 0.5^\circ\text{C}$.

As shown in Fig. 3A, an ultrasound transducer UP-DW set was employed in measurements using an ultrasound. Two sensor connection parts were used to fix the transducers that were embedded in the pipeline. The experiment that used water without bubbles was first conducted to measure the distance between the two transducers.

In measurements using the photographic technique, the photographic imaging system was equipped with an industrial

camera (BASLER aviator, 2 MP sensor, frame rate 65 fps, a 35 mm high-resolution lens), a computer, and light resources. The camera was placed in front of the water sight glass with a distance of 18 cm. The size of the imaged area (i.e., the area of the sight water glass) corresponded to 5.3 cm². The light resource was positioned on the opposite side of the camera and behind the glass wall to produce a high-quality bubble image in which the bubble outlines were easy to detect, and there was a high contrast between the water background and the bubbles. The experimental configuration is shown in Fig. 3B. The calibration of the photographic imaging system was performed by placing a ruler at the image location to obtain the image pixel scale. Following the completion of the experiment, the captured images were analyzed with an in-house developed image processing software. The contour of each bubble was created to obtain the bubble area. The average bubble diameter was then calculated using the above-obtained image pixel scale. It was assumed that each bubble had a circular form.

2.3 Principle of ultrasonic bubble sizing

Ultrasound energy is reduced when ultrasound waves propagate through liquid containing bubbles. The energy attenuation can be attributed to the following three mechanisms: reflection, scattering, and absorption. Part of the energy reduction is caused by reflection and scattering. The reflection mechanism is dominant if the size of the bubbles exceeds the sound wave, and this implies that energy loss is mainly caused by reflection. The scattering is dominant when the bubble size is much smaller than the sound wavelength, and thus energy loss is mainly due to the scattering. Another part of the ultrasound attenuation is caused by the energy absorption in bubbles [34]. Absorption and scattering can be quantified by the following two values: scattering cross-sections (denoted as σ_s) and the absorption cross-section (denoted as σ_a) [17] as follows:

$$\sigma_s = \frac{I_s}{I_p} \quad (3)$$

$$\sigma_a = \frac{I_a}{I_p} \quad (4)$$

where the I_s and I_a denote the total power of scattering and absorption, respectively, and I_p denotes incident wave intensity. The excess acoustical cross-section (denoted as σ_e) represents the aggregation of scattering and absorption effectiveness, which can be defined as follows:

$$\sigma_e = \sigma_s + \sigma_a \quad (5)$$

A model based on the relationship between the total extinction cross-section (denoted as S_e) of a bubbly fluid and the sound excess attenuation (denoted as α_e) can be used to estimate the bubble size [17] as follows:

$$\alpha_e = 4.34 S_e = 4.34 \int_0^\infty \sigma_e n(a) da \quad (6)$$

The ultrasonic through-transmission technique is required for the measurements to obtain the sound excess attenuation data. The received ultrasonic signals are used to determine the attenuation coefficient (α_e) of the ultrasonic signal propagating

through the bubbly fluid. Equation (6) is also expressed as a Fredholm integral equation of the first type [32]. This equation is considered as the Fredholm integral model.

However, a universal method for the inversion of the Fredholm integral model has not been found to date [23]. Hence, a solution based on resonant bubble approximation (RBA) theory was proposed and developed. The Fredholm integral model can be simplified based on assumptions suggested by [18, 35]. Based on their assumptions, only the bubbles with a resonant frequency equal to the incident ultrasound frequency contribute to the attenuation. The resonant approximation model is obtained to directly estimate the bubble density population [17] as given by the following expression:

$$n(a_R) = \frac{\alpha_e \delta_{Rr}}{8.68 \pi^2 a_R^3} \quad (7)$$

where δ_{Rr} denotes radiation damping coefficient at frequency f_R , α_e denotes attenuation coefficient, $n(a_R)$ denotes density population function per unit volume of bubbles with radius a_R that are resonated by the frequency f_R as defined by Eq. (8) given below:

$$f_R = \frac{1}{2\pi a_R} \sqrt{\frac{1}{\rho} \left[3\kappa \left(P_0 + \frac{2\sigma}{a_R} \right) - \frac{2\sigma}{a_R} \right]} \quad (8)$$

where $\kappa = C_p/C_v$ denotes ratio of the specific heat capacity, C_p denotes specific heat capacity at constant pressure, C_v denotes specific heat capacity at constant volume of the gas, P_0 denotes hydrostatic pressure, ρ denotes density of the bubble-free fluid, and σ denotes surface tension [36].

2.4 Data analysis of microbubbles

The data processing procedure is shown in Fig. 4. The input data includes the ultrasound signal, temperature data, and reference

Figure 4. Block diagram of the algorithm for calibration and validation.

Table 2. Parameters used in the prediction.

Parameter	Symbols	Unit	Value	Reference
Hydrostatic pressure	P_0	dyne/cm ²	1.02×10^6	Calculated
Depth	h	m	0.1	Measured
Surface tension	σ	dyne/cm	72.56	Calculated
Density of water (21°C)	ρ	g/cm ³	0.9979	Calculated
Ratio of specific heat (air)	κ	–	1.4	[17]
Polynomial coefficient	b_0	–	-6.342457×10^{-1}	[36]
	b_1	–	1.55834×10^{-2}	[36]
	b_2	–	-4.86179×10^{-5}	[36]
	b_3	–	4.930897×10^{-8}	[36]

results measured by a “HELOS” particle size analyzer. The data preprocessing includes three steps. First, ultrasound signals are filtered with a Butterworth fourth-order low-pass filter and a Butterworth fourth band-pass filter to remove the signal noise. The second step involves removing the digital component offset (such that the mean power of the signal is zero) of the ultrasound data. Discrete values of an ultrasound signal are averaged to obtain a mean value, which is then subtracted from each discrete value in this signal. The aim of the third step involves obtaining a bubble size density distribution $q_3(x)$ with a resolution $dx = 1 \mu\text{m}$. This step is necessary because raw microbubble size distribution data is in a cumulative form $Q_3(x)$ with an inhomogeneous resolution, which varies from 1.6 to 24 μm . These values do not satisfy the size interval (specified as 1 μm) in the theoretical model. Nevertheless, those intervals are determined by the reference measuring instrument, and they cannot be configured. Therefore, it is necessary to use a mathematical method to normalize the microbubble size interval measured by the US method and the reference method.

The data of ultrasonic experiments are then divided into two sets to compare the model performance. A set is referred to as the calibration set, which is used to obtain the new model adapted to artificial bubbles in the simulation study. The other set is termed as the validation set and is used to validate the feasibility of the new model. Each set contains 10 signals. The first received signal impulse propagating a single path between two transducers is selected for further data processing as shown in Fig. 2B. The data of the first arriving impulse was selected and subsequently transformed to the frequency domain using a fast Fourier transformation (FFT) analysis.

Effective frequencies $f(i)$ ($i = 1, 2, \dots, n$) were obtained from the frequency spectrum. The discrete frequency values $f(i)$ within this range were used as input ultrasound frequencies. With respect to each frequency value, a value of the attenuation coefficient was calculated in conjunction with a value of the resonance bubble radius. The attenuation coefficient (denoted as $\alpha_{e,i}$) at $f(i)$ is calculated by the following expression:

$$\alpha_{e,i} = \frac{1}{d} 10 \log_{10} \frac{I_{0,i}}{I_i} \quad (9)$$

where $\alpha_{e,i}$ has a unit dB/m, d denotes the distance between two transducers, and $I_{0,i}$ and I_i refers to the power intensity of the fluid without or with artificial bubbles, respectively.

The bubble resonance radii (denoted as a_R) are calculated using Equation (8). The hydrostatic pressure is calculated by the following expression:

$$P_0 = P_{atm} + \rho gh \quad (10)$$

where P_{atm} denotes the atmospheric pressure, g denotes gravitational acceleration, and h denotes immersion depth of the measuring tube in water bath. Additionally, ρ denotes the density of water, which is calculated by Equation (11) [37]:

$$\rho = b_0 + b_1 T + b_2 T^2 + b_3 T^3 \quad (11)$$

where T denotes the temperature in °C. The related parameters used in Equations (1)–(11) are listed in Table 2.

According to the attenuation approximation model of Medwin (MR model), the attenuation coefficient in decibels per meter of the bubbly fluid is expressed as follows:

$$\alpha_{e,i} = \frac{8.68 \cdot \pi^2 \cdot a_i^3 \cdot n(a_i)}{\delta_{Rr,i}} \quad (12)$$

where $n(a_i)$ denotes the bubble number density distribution over the range $a_i + da$. Conventionally, da is defined as 1 μm [18,24] per cubic meter and $\delta_{Rr,i}$ is defined as follows:

$$\delta_{Rr,i} = k_{R,i} a_i \quad (13)$$

where $k_{R,i}$ denotes the wave number of water at the resonance frequency f_i .

Additionally, the resonant wave number of bubble-free liquid is defined as the ratio of angular velocity given as: $\omega_i = 2\pi f_i$, and the USV denotes the wave propagation velocity in pure water [33] as follows:

$$k_{R,i} = \frac{\omega_i}{USV} = \frac{2\pi \cdot f_i}{USV} \quad (14)$$

By combining Equations (12)–(14), the bubble number density distribution per unit volume can be written as follows:

$$n(a_i) = \frac{\alpha_{e,i} \cdot f_i}{4.34 \cdot USV \cdot \pi \cdot a_i^2} \quad (15)$$

It is necessary to convert the $n(a_i)$ into volume percent to compare the results measured by the “HELOS” particle size analyzer instrument. Since the volume concentration of the measured mixture and the total microbubble volume are known, the conversion is obtained by using Equation (16) as follows:

$$M(a_i) = \frac{n(a_i) \cdot da \cdot \frac{4}{3} \pi \cdot a_i^3}{C} 100\% \quad (16)$$

where $M(a_i)$ denotes the microbubble volume frequency of size a_i over a radius range of $1 \mu\text{m}$, and C denotes the microbubble volume concentration. By combining Equation (15), the bubble size distribution can be expressed as follows:

$$M(a_i) = \frac{2\alpha_{e,i} \cdot f_i \cdot a_i \cdot da}{6.51 \cdot \text{USV} \cdot C} 100\% \quad (17)$$

An attempt was made to calculate theoretical attenuation result using Equation (18) based on the assumptions of Medwin and Commander, which is termed as the RBA model [32], as given below:

$$\text{RBA}(a_i) = \frac{n(a_i)}{4.6 \times 10^{-6} \cdot f_i^3} \quad (18)$$

A calibration factor term is added into the MR model to establish a new model adjusted to the microbubbles. The calibration factor (denoted as $\text{Corr}(k)$) is a function that is related to the mixture concentration and the mean experimental temperature as follows:

$$\text{RBA}(a_{ik}) = \text{Corr}(k) \cdot \alpha_{e,ik}, \quad (i, k = 1, 2, \dots, n) \quad (19)$$

$$\text{Corr}(k) = \beta_0 + \beta_1 \cdot T(k) + \beta_2 \cdot C(k) \quad (20)$$

where β_i denotes regression coefficients, $T(k)$ denotes mean experimental temperature, and $C(k)$ denotes mixture concentration.

The calibration factor can be expressed in the form of a matrix as follows:

$$\begin{bmatrix} \text{Corr}_1 \\ \text{Corr}_2 \\ \vdots \\ \text{Corr}_{k-1} \\ \text{Corr}_k \end{bmatrix} = \begin{bmatrix} 1 & T_1 & C_1 \\ 1 & T_2 & C_2 \\ \vdots & \vdots & \vdots \\ 1 & T_{k-1} & C_{k-1} \\ 1 & T_k & C_k \end{bmatrix} [\beta_0 \beta_1 \beta_2] \quad (21)$$

Then, $\text{Corr}(k)$ is combined with Equation (18). A new model of estimation of the bubble size distribution is expressed as follows:

$$M(a_i) \% = \frac{2\alpha_{e,i} \cdot f_i \cdot a_i \cdot da}{6.51 \cdot \text{USV} \cdot C} \cdot \text{Corr}(k) \cdot 100\% \quad (22)$$

In order to evaluate performance of the modified model, the root mean squared error of calibration (RMSEC) is used [38] as follows:

$$\text{RMSEC} = \sqrt{\frac{\sum_{i=1}^n (M(a_i) - \hat{M}(a_i)_c)^2}{n}} \quad (23)$$

where $M(a_i)$ denotes the reference distribution and $\hat{M}(a_i)_c$ denotes the estimation of the bubble size distribution using the calibration set.

It is necessary to validate the new model to avoid the overfitting issue. Thus, the validation data set is employed to verify the new model. Its feasibility is evaluated by the root mean squared error of validation (RMSEV), which uses the following Equation (24) [38]. This value is used to indicate the measurement accuracy.

$$\text{RMSEV} = \sqrt{\frac{\sum_{i=1}^n (M(a_i) - \hat{M}(a_i)_v)^2}{n}} \quad (24)$$

In the case of real bubble measurement, the bubble size distribution is analyzed in the form of “average bubble number of a given bubble radius” within the imaged volume (denoted as V), which corresponds to 35 mL (imaged area $\times$ distance between two transducers). The bubble size estimation is expressed as given below:

$$n(a_i) = \frac{V\alpha_{e,i}f_i}{4.34\text{USV}\pi a_i^2} \quad (25)$$

3 Results and discussion

The effective frequency range of transducers is obtained through analysis of the frequency spectrum as shown in Fig. 5. The effective frequencies of the UP-DW broadband transducer correspond to the peak values of the total frequency spectrum as shown in Fig. 5A. In the case of the 500 kHz transducers, the effective frequency range is denoted by a red line as shown in Fig. 5B. The detectable bubble radii were calculated under the experimental conditions using Equation (8). Therefore, the detectable bubble size at temperature $21 \pm 0.5^\circ\text{C}$ is approximately in the range of $6.7\text{--}23 \mu\text{m}$.

The initial bubble size analysis demonstrated that it was not possible to directly apply the MR model for the estimation using artificial bubbles. The estimated size distributions measured by the ultrasonic system were approximately four orders of magnitude lower than that by the reference method as shown in Fig. 6A. The black curve indicates the results obtained from the reference method, while the red color denotes the estimated results using the original MR model. The different structures of the real bubble and the artificial bubble could lead to an underestimation. When compared with the real bubble, the used microbubble has a shell, and this may result in other forms of energy losses that are not included in the theoretical model. The problem arising from the shell was also studied and solved by extending the theoretical model [39].

The correction factor was then added to the MR model to develop a new model to adapt the artificial microbubbles. First, the ratio of the measured and predicted attenuation coefficients was calculated to obtain a vector denoted as Corr . The average temperatures of the calibration experiment data were calculated and denoted as a vector T , and the sample mixture concentrations were calculated and denoted as a vector C . Multiple linear regression analysis was used to obtain the coefficient (denoted as β_i) for each transducer set.

The corrected bubble size distributions were calculated using the new model modified by the correction factor. Figure 6B shows a comparison of the calibration measurement of 0.2 v/v using the US method and the reference method (HELOS).

Figure 6B presents the results using the calibration data set. As shown in the figure, the established new model eliminated the three-order magnitude underestimation. Therefore, it indicated that the correction factor associated with the MR model was available to estimate the size distribution of microbubbles in the artificial bubbly fluid. The obtained results using the US method approximated those using the reference method. The most accurate calibration experiment resulted in an accuracy of 0.1197 v/v (RMSEC) under a concentration of 0.2 v/v .

Figure 5. (A) Frequency spectrum of the UP-DW broadband transducer. (B) Frequency spectrum of the 500 kHz transducer.

Figure 6. (A) Comparison of the results using the ultrasonic (US) method and the results using the laser diffraction method (HELOS). The values measured by the ultrasonic system are approximately four orders of magnitude lower than that obtained using the reference method. (B) Bubble size distribution (volume fraction) using the calibration data set. (C) Bubble size distribution (volume fraction) using the validation data set. Specifically, “+” denotes measurement using the US method, and “•” denotes the reference measurement using the laser diffraction method.

The new model was then validated using the validation data set. The average temperature of each experiment and the corresponding concentration were inserted to the new model to calculate the microbubble size distribution. The most accurate validation experiment resulted in an accuracy of 0.1243 % v/v (RMSEV) under a sample concentration of 0.2 % v/v as shown in Fig. 6C. This demonstrates that the results using ultrasound-based system agreed well with those using the reference method.

With respect to the measurements of the real bubble size distribution, the generated bubbles were first measured by the photographic imaging system. In order to analyze the bubble number and bubble size, the contour of each bubble was created using an in-house developed image processing software known as “ImgPro” as shown in Fig. 7. Approximately 140–186 bubbles were counted in each captured image. The resultant bubble size was in the range of 23–1593 μm . Given the camera resolution and

Figure 7. Photographs of the bubbles in the bypass pipeline of a yeast propagation tank. (A) Prior to image processing, and (B) after image processing.

frame rate, it was difficult to detect the small bubbles. Therefore, only the results for a bubble with a $23\ \mu\text{m}$ radius could be compared with the results measured by the photographic imaging system.

The mean number of the bubbles using the photographic method corresponded to 17 per measured volume, while that using the US method corresponded to 28 per measured volume. This demonstrates that the size distribution in this radius class agreed well with the result using the photographic method. The following aspects could be observed based on the experiment involving real bubbles. First, the ultrasound-based measuring system could determine the distribution of the bubbles with a radius of approximately $23\ \mu\text{m}$ in a relatively more accurate manner. A deviation of 11 bubbles per measured volume was obtained when the results of the two methods were compared. The deviation could result from measuring the time intervals of both methods. The time interval between two ultrasound signals was 20 s, while each photograph was captured every 0.015 s. Evidently, the bubble distribution spectra (number of bubbles in an imaged area of a given size) changed significantly between one measurement and the next. Second, it was difficult to capture the bubbles located beyond the focus area of camera. Consequently, it was not possible to avoid underestimating the number of bubbles.

4 Concluding remarks

In this study, an ultrasound-based measuring system was developed to measure the bubble size distribution in a gas–liquid two-phase system under laboratory conditions. The experimental method was much more effective than previous methods due to improvements performed using a simulated bubbly fluid. The ultrasonic velocity in bubble-free water and the attenuation coefficient in microbubble–water mixture were measured using two sets of ultrasonic transducers, namely a 500 kHz pair and a broadband transducer pair, which covered bubble radii in the range of $6\text{--}23\ \mu\text{m}$. The measured ultrasonic data were inverted to estimate the bubble size distribution by using a new model based on the resonant attenuation approximation model derived by Medwin. With respect to the measurements

using the simulated bubbly fluid, this ultrasound-based measuring system obtained accuracy in the form of RMSEV that corresponded to 0.1243 % v/v. With respect to the measurements of the bubble size in the bypass pipeline, the feasibility of the established ultrasonic measuring system was also proven by comparing it with the results from the photographic method. A deviation of 11 bubbles per measured volume was obtained as the differences in the sizes that were detected in the photographic method.

In conclusion, this study demonstrated that the proposed method could be potentially used to establish an ultrasound-based system that enabled the determination of the bubble size distribution in gas–liquid two-phase systems. However, the present study should be considered as an initial step in this direction. It is necessary to conduct further investigations to improve the measuring robustness of the developed system and extend the application area. The following aspects should be considered for future studies. First, given the determination limitation of the gas volume fraction (1.13 %), it is necessary to perform further investigations to consider the upper limit of the attenuation method to decrease the measurement error and improve measuring system stability. Second, although the newly modified model can estimate the bubble size distribution, there is still a need to improve the sensibility and stability of the newly modified model, which can be adapted to measurements in the gas–liquid two-phase systems as well as in the gas–liquid–solid three-phase systems. Third, the radial vibration effect of the receiver can lead to a certain amount of underestimation and overestimation of the attenuation coefficient in conjunction with the microbubble size distribution. It is necessary to consider two aspects with respect to the attenuation coefficient measurement. First, accuracy depends on sound propagation distance measurement. The findings indicated that when the distance between two transducers corresponded to 30 mm, a measuring error of 0.03 mm could result in 0.1 % uncertainty of the attenuation coefficient. Conversely, the S/N ratio of the electronic device can also result in the variance of the attenuation coefficient measurements. Thus, it is suggested that the ultrasonic transducers should be optimized in further experiments. Additionally, the uncertainty of attenuation coefficient measurements should be less than 2.5 % [40].

Practical application

In this study, a novel method was proposed, and an ultrasound-based monitoring system was developed to determine bubble size distribution in gas–liquid two-phase systems based on the ultrasound-based method. The key concept of the method involved the use of artificial bubbles to simulate a bubbly fluid environment to rapidly measure real bubble size distribution in practical application cases. Simulation results in the study demonstrated that the method of using the simulated bubbly fluid could also be used to calibrate real measuring systems. Additionally, results revealed that the developed measuring system could be potentially used for inline monitoring of the bubble size distribution in gas–liquid two-phase systems.

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Nomenclature

ABS		Acoustic Bubble Spectrometer®
a_r	[μm]	Bubble resonant radius
ADC		Analog-to-digital converter
dB		Decibel
C_p	[J/K]	Heat capacity at constant pressure
C_v	[J/K]	Heat capacity at constant volume
d	[m]	Distance
f_R	[Hz]	Resonance frequency
FFT		Fast fourier transformation
HELOS		Helium-Neon Laser Optical System
I_0	[W/m ²]	Signal power intensity of bubble-free fluid
k		Wave number
k_R		Wave number of water at the resonance frequency f_R
MR-model		Resonant bubble approximation of Medwin
$N(a)$	[m ⁻³]	Bubble number function
$n(a)$	[m ⁻⁴]	Bubble population density
Pt		Platinum
P_{atm}	[Pa]	Atmospheric pressure
P_0	[Pa]	Hydrostatic pressure
RMSEV		Root mean square error of validation
RMSEC		Root mean square error of calibration
RBA-model		resonant bubble approximation model
$Q_3(R)$		particle size cumulative volume function
$q_3(R)$		particle size density volume function
S_e		total extinction cross section
T	[°C]	temperature
TOF	[s]	Time-of-flight
USV	[m/s]	ultrasonic velocity
UP-DW		GEOTRON® transducer production name
US		Ultrasound

Greek symbols

κ		Heat capacity ratio for real gas
K_g	[W/m·K]	Thermal conductivity of gas
ρ	[g/cm ³]	Density
ω	[rad/s]	Angular velocity
α	[dB/m]	Attenuation coefficient
α_e	[dB/m]	Excess attenuation coefficient
σ	[dyne/cm]	Surface tension
σ_e		Scattering cross section
δ_{Rr}		Damping constant at resonance

Indices

atm	atmospheric
g	gas
e	excess
R	resonance

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