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System for Characterizing Aerosols from Bubbling Liquids

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The transfer of materials from the liquid phase to the gaseous phase by the bursting of gas bubbles at the liquid surface is of concern in many industrial and environmental applications, e.g., microorganism generation from wastewater treatment plants and health club whirlpools. Most previous studies have focused on single-bubble bursting, which is rarely found in actual situations. An experimental system has been developed that is large enough to accommodate multiple-bubble bursting and overcomes the difficulties of sampling and analysis of liquid droplets above the surface of a bubbling liquid. The system uses dynamic aerosol spectrometers to provide real-time information on the concentrations and size distributions of the liquid droplets generated and of the materials contained in the liquid droplets after their evaporation. Specially designed impactors are used to size classify the droplets so that the content of the droplets of various size ranges can be evaluated. Examples of measured droplet distributions show that the system is independent of the distance above the liquid surface at which the sample is taken. The geometric mean diameter of the droplets generated from 2.57-mm bubbles was found to be 2.0 μm with $\sigma_g = 1.96$. In one experiment with a suspension of 0.56-μm diameter latex spheres at a concentration of $10^9/\text{cm}^3$ in distilled water, 61% of the measured droplets contained latex particles. This represented a several hundredfold enrichment over the bulk concentration of particles.

INTRODUCTION AND BACKGROUND

Bubbling of air or other gases through a bulk liquid is a common phenomenon found in many natural and man-made processes. Bubbles rise to the surface of the liquid, come to rest and eventually burst. Two types of droplets are generated: 1) the more numerous, smaller film droplets, and 2) a few larger jet droplets. Film droplets are generated from the disintegration of the bubble-film cap into liquid ligaments which have a lacelike structure. The ligaments subsequently break into minute droplets (Dombrowski and Frazer, 1954). Their diameters range from less than 1 to 20 μm (Garner et al., 1954; Tomaides and Whitby, 1976) with half of them smaller than 10 μm (Blanchard and Syzdek, 1982). Jet droplets are produced from the breakup of the liquid jet rising upward from the collapsing bubble cavity. They can be as large as 10% of the bubble diameter (Blanchard and Syzdek,

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Aerosol Science and Technology 13:297–307

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1975), but only occur for bubbles less than 5 mm in diameter (Garner et al., 1954). As bubbles rise through a liquid, they scavenge particles and bacteria onto their surfaces. Studies on single bubble bursting have shown that both the resulting film and jet droplets may be enriched with dissolved and suspended materials from the liquid (Blanchard and Syzdek, 1982). These materials may remain airborne as residues after droplet evaporation. The resulting aerosol creates a potential human respiratory exposure to live bacteria and viruses as well as other sensitizing agents that are present in the bubbling liquid.

The generation of droplets from a bursting bubble and thus the transfer of materials from the liquid into the gas phase creates many industrial and environmental problems. Studies at several activated sludge wastewater treatment plants have shown that the atmospheric concentrations of bacterial indicators of fecal pollution are higher downwind than upwind from the aeration tanks (Randall and Ledbetter, 1966; Kenline and Scarpino, 1972; Fedorak and Westlake, 1980; Fannin et al., 1985). Also, Baron and Willeke (1986) measured the size distribution in the air above health club whirlpools. They concluded that the particles must be water droplets from the bursting of bubbles, and that the droplets were sufficiently large to contain *Legionella* bacteria and small enough to reach the human respiratory tract.

Nearly all previous studies on the generation and enrichment of bubble-induced droplets have concentrated on single-bubble bursting. However, for most natural or man-made liquid bubbling, multiple-bubble bursting is more common and it may differ from individual bubble bursting. For example, a large number of bubbles rising to the surface creates a convective upwelling plume of liquid resulting in vigorous bubbling at the liquid surface that is not present in single bubble experiments. Bubbles which have reached the surface may not burst immediately. They may randomly float around and may coalesce with adjacent bubbles into large hemispheres or may attach to each other and form a layer of rafts or foam patches. This certainly affects the size distribution and concentration of droplets generated from the bursting bubbles.

Several technical difficulties impede the sampling and analysis of liquid droplets. Liquid droplets are difficult to collect and retain in their original form for later sizing and counting. They may change in size due to evaporation or condensation. The coating of collection slides with magnesium oxide (MgO) or gelatin (May, 1950, 1961) is the most frequently used technique for the study of film and jet droplets. In this technique, a glass slide, coated with a thin layer of MgO or gelatin, is held above a bursting bubble and the imprints on the MgO or gelatin layers created by droplets impacting on the slide are measured under a microscope and are then corrected by the spread factor (the ratio of drop size to imprint size) to obtain the equivalent volume diameter of the actual droplet. Blanchard and Syzdek (1975) developed an electrostatic induction technique to enhance the collection of droplets on the slide and made it possible to collect film droplets separately from jet droplets for size distribution determinations. It appears that this technique works well with the few, large jet droplets but the size measurement and counting become tedious and time consuming for the numerous small film droplets. Since the time to analyze a single test condition may take several days, it is too time-consuming to conduct any extensive parametric study. The technique also has limitations and questionable accuracy when dealing with small droplet sizes.

To overcome the limitations and difficulties of previously used techniques, a laboratory-scale experimental system has been developed in which the liquid-to-gas transfer of materials from bubbling liquids is studied by use of advanced instrumentation which makes real-time measurements of the bubble-induced aerosols. The system measures in-situ the concentrations and size distributions of the liquid droplets and of the residues in droplets of various size ranges. The effects of several operational parameters can be studied with this system, e.g., liquid properties and compositions, bubble size, bubble rise distance, and superficial gas velocity. These parameters may affect the generation and the characteristics of the bubble-induced aerosols. The results gained from studies will provide information for the development of control measures and the reassessment of the chosen operational parameters so that aerosolization of materials in liquid can be minimized.

The system was tested using latex particles to approximate bacteria during the aerosolization of these microorganisms from liquid suspension (Wangwongwatana, 1989). The results of some of these tests are included below to demonstrate the capabilities of the system.

EXPERIMENTAL SYSTEM AND METHOD

Figure 1 shows a schematic diagram of the overall experimental system which was developed for characterizing aerosols from bubbling liquids. The system consists of three major components: 1) bubbling tank, 2) liquid droplet sampling and analysis component, and 3) droplet residue sampling and analysis component. Details of each component are described in the following sections.

Bubbling Tank, Bubble Generation, and Sizing

The bubbling tank holds a study liquid through which gas bubbles generated from gas diffusers rise to the liquid surface, burst and generate droplets. The tank is made of $\frac{3}{8}$-in.-thick acrylic plastic and has a square cross section of 35 × 35 cm. The cross-sectional area was chosen to be large relative to the individual bubble size. The tank can be modified to a cylindrical shape by inserting a circular plastic section. Our tank is 76 cm high, but can be increased through the addition of extension sections at the top of the tank so that the liquid depth, i.e., the bubble rise distance, can be varied. There are seven rows of seven circular holes at the bottom of the tank to accommodate a maximum of 49 gas diffuser heads. A removable acrylic plastic hood is sealed to the top of the tank with a gasket around the inside edge of the hood. A slightly positive pressure is maintained inside the top enclosure to prevent contamination of the study aerosols by the room air. Study aerosols are extracted through two sampling ports symmetrically located on opposite sides of the hood: one for sizing liquid droplets, the other for sizing the droplet residues.

To determine the effect of bubble size on droplet formation, it was necessary to generate a large number of uniform-size gas bubbles of desired diameters. Gas diffusers made up of capillary tubes were found to reproducibly generate bubbles of uniform size and constant generation rate (Cheng and Lemlich, 1983). Gas diffusers were fabricated using 1.3-cm-long capillary tubes. The tubes were inserted into a 3.5-cm diameter plastic cap whose inside surface was lined with a rubber pad to prevent gas leakage. Each cap was attached to a small glass funnel which served as a gas distribution chamber. The inside diameter of the capillary tubes are 0.2, 1.0, and 1.5 mm. The bubbles generated by these tubes have been labeled in Table 1 as fine, medium, and coarse diffusers, respectively. The air to the diffusers was filtered and humidified to saturation as indicated in Figure 1. The flow rate was controlled to give the specific volumetric flow rates per diffuser head indicated in Table 1.

The bubble sizes in deionized distilled water have been measured by still-picture photography (Kintner et al., 1961; Wangwongwatana, 1989). The shape of the photographed bubbles is approximated by an oblate spheroidal whose minor axis is in the direction of bubble motion. The diameter

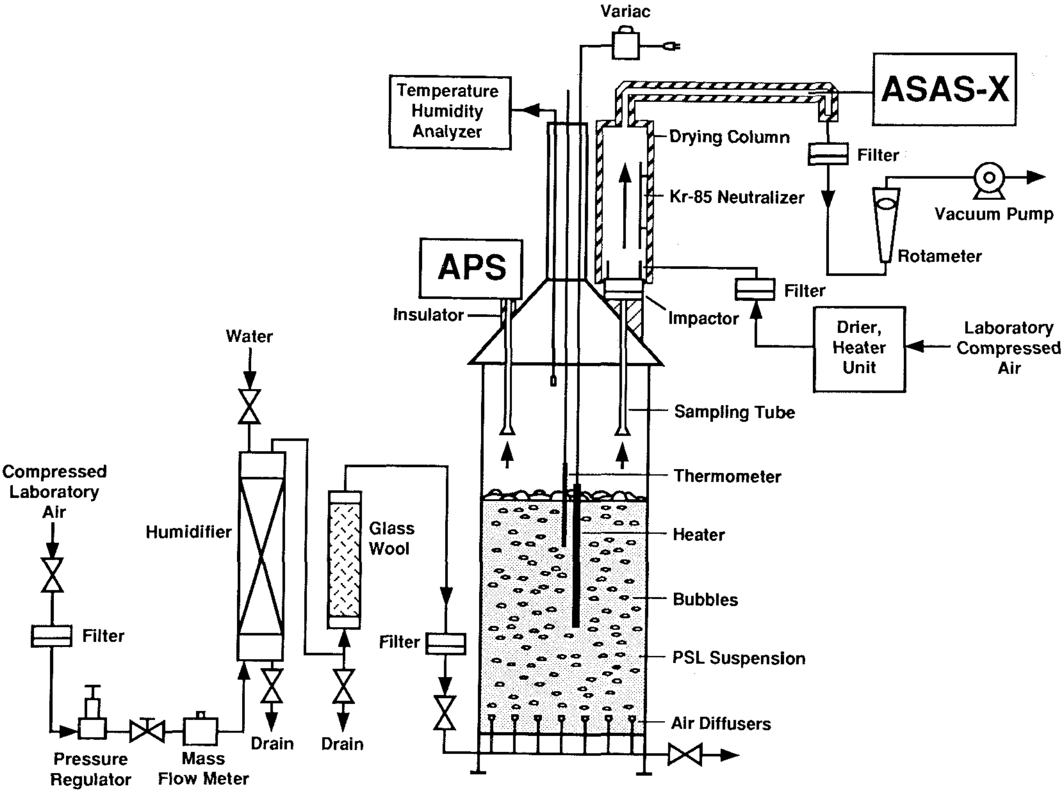


FIGURE 1. Schematic diagram of the system for characterizing aerosols from bubbling liquids. APS, aerodynamic particle sizer; ASAS-X, active scattering aerosol spectrometer; PSL, polystyrene latex spheres.

was chosen to equal the projected area diameter (D_a), which is defined as the diameter of a circle having an area equivalent to the area of the bubble cross-section perpendicular to its minor axis. The equivalent volume diameter (D_v) has also been calculated by equating the volume of a sphere to the volume of each oblate spheroid. The equivalent volume diameter is defined as the diameter of a sphere having the same volume as that of the bubble.

The characteristics of the three diffusers are summarized in Table 1. The fine diffuser produces bubbles with a count median diam-

TABLE 1. Characteristics of Capillary-Tube Gas Diffusers and Their Generated Bubble Sizes in Deionized Distilled Water

Bubble	Capillary ID (mm)	No. of Tubes per Head	Air Flow per Head (cm^3/min)	Projected Area Diameter Mean (mm)	Projected Area Diameter CMD (mm)	Projected Area Diameter σ_g	Equivalent Volume Diameter Mean (mm)	Equivalent Volume Diameter CMD (mm)	Equivalent Volume Diameter σ_g
Fine	0.2	21	387	3.21	3.23	1.15	2.59	2.57	1.13
Medium	1.0	4	700	8.55	8.45	1.11	6.48	6.45	1.1
Coarse	1.5	1	429	13.1	14.5	1.2	9.9	10.2	1.14

Abbreviations: ID, inside diameter; CMD, count median diameter; σ_g, geometric standard deviation.

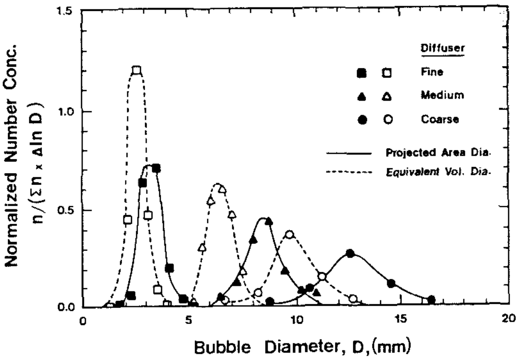


FIGURE 2. Normalized number size distribution of bubbles generated from fine, medium, and coarse diffusers. The bubble sizes are characterized by projected area diameter and equivalent volume diameter.

eter (CMD) of 2.57 mm in equivalent volume diameter. The bubble CMDs are 6.45 mm for the medium diffuser, and 10.2 mm for the coarse diffuser. The bubble sizes are quite uniform for all three diffusers as indicated by geometric standard deviations (σ_g) of less than 1.15. Figure 2 shows the number distribution of the bubbles characterized by projected area diameter and equivalent volume diameter. Each bubble size distribution is approximately normal.

Sampling and Analysis of Liquid Droplets

Several technical difficulties impede the sampling and analysis of liquid droplets. Because of their instability in shape, liquid droplets cannot be collected on a substrate, such as a filter, in their original form for subsequent sizing and counting. They also may change in size, in space and time due to either evaporation or condensation. Therefore, in-situ sampling and real-time analysis are needed to obtain rapid, reliable, and representative measurements.

The droplet concentration and size distribution in the system are measured by an aerodynamic particle sizer (model APS 3300, TSI Inc., St. Paul, MN). The APS is suitable for measuring liquid droplets because of its high-resolution, fast measuring process, and short path to the sensing volume. If the air surrounding the droplets is kept saturated with the vapor, the change in droplet size will be minimized. With the APS, aerosols are sampled in-situ and are analyzed rapidly by a microcomputer. This instrument has been used successfully by Baron and Willeke (1986) to investigate the size distribution of water droplets above health-club whirlpools.

The APS measures the aerodynamic particle diameter by accelerating the sampled particles through a nozzle and measuring the time-of-flight between two intercepting laser beams. Acceleration through the nozzle may cause distortion of the liquid droplets from a sphere to an oblate spheroid. This alters their acceleration rate and thus, their apparent aerodynamic diameter. Baron (1986) used a laser imaging system to photograph accelerated liquid droplets and found negligible distortion of water droplets in the 20–100-μm range. The distortion was observed only in low surface tension droplets. Even for these droplets, the distortion decreased markedly with decrease in particle size. Therefore, distortion is unlikely to occur for the smaller water droplets (0.5–20 μm) in the present experiments.

The APS was size-calibrated with monodisperse latex particles in the range of 0.312–20 μm. Flow calibration of the inlet was accomplished with a bubble meter. The original APS software from the manufacturer (TSI Inc.) has been modified to extend the APS measuring range from 0.5–15 μm to 0.5–20 μm. About 0.8 μm has been used as the lower detection diameter because the APS is not able to detect particles less than that size with 100% efficiency. This extended range covers the larger film droplets. The larger jet droplets are caught by the foam above the liquid or only reach a height of a few centimeters before falling back into the liquid under the influence of gravity.

The lowest aeration rate used in our experiments was 8 L/min which is only 3 L/min more than the total sampling rate of the APS. The inlet of the APS was therefore modified so that the sample aerosol and the sheath air enter the APS inlet separately. The APS was turned upside-down above one of the two sampling ports on top of the bubbling tank hood. Inversion of the APS does not affect its measurement accuracy because the acceleration in the nozzle is so much greater than that due to gravity. A sampling probe connected to the APS aerosol inlet was inserted through the port (Figure 1) so that only 1 L/min of aerosol from inside the tank was introduced directly into the inner nozzle of the APS. Varying lengths of probe tubing made no apparent difference to the measured concentration, so sampling line losses were assumed to be negligible. Sheath air at 4 L/min was drawn from the room. The sheath air was saturated with vapor of the liquid bubbling in the tank prior to entering the outer nozzle of the APS to prevent evaporation of liquid droplets in the APS sensing volume.

Sampling and Analysis of Droplet Residues

The principle of the droplet residue measurement is to size classify the droplets, evaporate liquid from the droplets and then measure the concentration and size distribution of the remaining residues. This is accomplished by a multiple component system (Figure 1) consisting of a specially designed single-stage impactor, a drying and charge neutralization column, and an active scattering aerosol spectrometer (model ASAS-X, PMS Inc., Boulder, CO). This system was suspended above the second sampling port. The sampling probe was identical to the one used with the APS. One liter per minute of aerosol was drawn from inside the tank through the impactor which allowed droplets smaller than its cutoff diameter to pass into the drying column. One liter per minute of dried (< 2% relative humidity) and heated (50°C) aerosol-free dilution air was introduced into the drying column near the bottom and mixed with the sample aerosol to evaporate the liquid droplets. Evaporation time in this column for a 10-μm water droplet was 0.15 s according to a calculation procedure by Houghton (1933). The actual average residence time was calculated to be 42 s, enough to ensure complete evaporation of all droplets down to residue particles.

A 10 mCi ^{85}Kr radioactive source was placed inside the drying column for charge neutralization, thus decreasing particle losses in the sampling system due to electrostatics. The droplet residues at the top of the drying column were sampled by the ASAS-X for concentration and size analysis. By using single-stage impactors of different cutoff diameters ahead of the ASAS-X, the concentration and size distribution of residues from droplets of various size ranges was determined.

The ASAS-X is a single-particle optical counter utilizing a laser as a light source and gives in situ and real-time display of particle size distributions. It measures optical scattering diameters of particles between 0.09 and 3.0 μm and was calibrated with monodispersed latex particles in the range of 0.109–2.02 μm. These calibration particles had the same reported refractive index as the latex particles used in the bubbling suspension. Software programs, developed by Holton (1986), were used to analyze the computer acquired data for particle concentration and size distribution. Other single-particle optical counters could have been used as well.

Several single-hole, single-stage impactors having different cut-off diameters were fabricated (Wangwongwatana, 1989). They were designed according to the criteria and procedures recommended by Marple and Willeke (1976) and Willeke (1978). The inlet section and two extension rings of a plastic 37-mm conventional filter cassette were used as the impactor body, as schematically shown in Figure 3. An insert, with a single round hole, made of acrylic plastic served as the impactor nozzle and was oper-

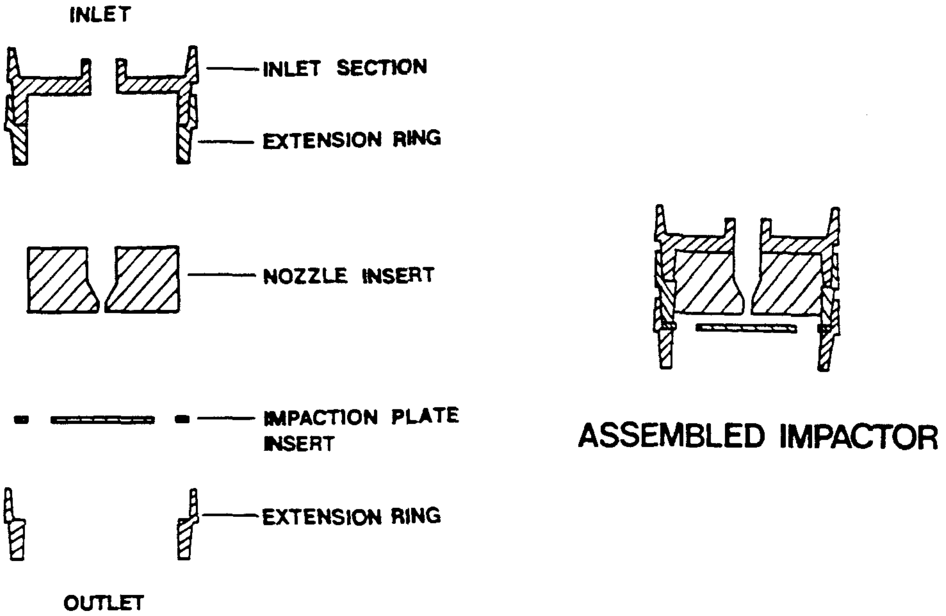


FIGURE 3. Schematic diagram of the single-hole single-stage impactor utilizing an inlet section and two extension rings of the plastic 37-mm conventional filter cassette as the impactor body.

ated at 1 L/min. The assembled impactor was inserted into the inlet of the drying column.

The collection efficiency curve of each impactor was determined experimentally by use of the APS. The procedure was similar to that described by Jones et al. (1983). Polydisperse corn oil aerosols were passed into an aerosol chamber and mixed with dilution air. The challenge aerosol size distribution in the chamber was first measured by the APS. The impactor was then connected to the inner nozzle of the APS and the size distribution of aerosols penetrating the impactor was measured. The collection efficiency curve of the impactor was obtained by dividing the latter size distribution by the former. The 50% cutoff diameter (D_{50}) of the impactor was then determined from the collection efficiency curve. The curves for the three impactors are shown in Figure 4. Table 2 summarizes the characteristics of these impactors and their D_{50}'s which were determined experimentally and theoretically.

EXAMPLES OF EXPERIMENTS AND RESULTS

Experiments were carried out to demonstrate the usefulness of the system. Relatively monodisperse polystyrene latex (PSL) particles were mixed into deionized distilled water at a concentration of $10^9/\text{cm}^3$. The number median diameter of the PSL particles was 0.59 μm with $\sigma_g = 1.14$ (measured by scanning electron microscopy).

The aqueous phase of the stock PSL normally contains small amounts of surfactants which helps in keeping the individual PSL particles uniformly dispersed. Presence of surfactants results in two undesirable effects: 1) foaming at the surface of the aerated suspension, and 2) formation of extraneous particles or increase in size of the airborne PSL particles after droplet evaporation. In the preparation of the PSL suspension, dissolved surfactants and other water-soluble materials were removed from the aqueous phase by diafiltration or serum replacement (Labib and Robertson, 1978). Thus, the la-

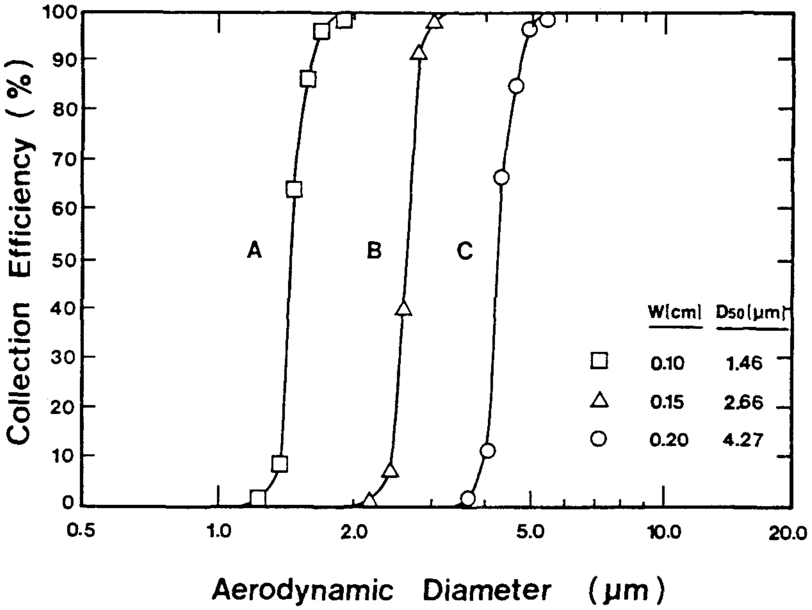


FIGURE 4. Collection efficiency curves of the three single-hole single-stage impactors. W, nozzle diameter; D_{50}, 50% cutoff diameter.

tex serum was replaced gradually with deionized distilled water. The cleaned PSL solution was then diluted with deionized distilled water to the desired concentration.

Air was used to generate bubbles in the liquid. Compressed air was passed through an absolute filter and was then humidified to saturation in a vertical column filled with Raschig rings and deionized distilled water. The saturated air was then filtered again before entering the tank through capillary gas diffusers. The temperature of the PSL suspension was maintained at 25°C. The air above the bubbling PSL suspension was maintained at a slightly supersaturated condition (100.15% relative humidity) and was

TABLE 2. Characteristics and Cutoff Diameters of the Three Single-Hole Single-Stage Impactors

	Impactor		
	A	B	C
Nozzle diameter (W), cm	0.10	0.15	0.20
Jet-to-plate distance (S), cm	0.11	0.33	0.42
Nozzle throat length (T), cm	0.15	0.25	0.35
S/W	1.12	2.16	2.06
T/W	1.47	1.64	1.73
Avg. throat velocity, cm/s	2040	918	520
Reynolds number (Re)	1380	926	697
Estimated pressure drop, mmHg	25.6	5.2	1.7
Cutoff diameter (D_{50}), μm			
Theoretical	1.33	2.49	3.89
Calibration by APS	1.46	2.66	4.28
Calibration/theoretical	1.10	1.07	1.10

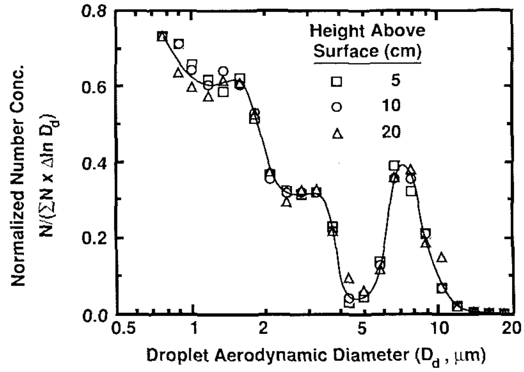


FIGURE 5. Number size distributions of droplets at three different heights, i.e., 5, 10, and 20 cm above the surface of the bubbling PSL suspension: bubble equivalent volume diameter D_v = 10.2 mm, superficial gas velocity, V_g = 11 cm/min, bubble rise distance, BRD = 45 cm, and bulk PSL particle concentration, $C_{PSL} = 10^9/\text{cm}^3$.

monitored continuously by a humidity analyzer (model 911 Dew-All, EG&G, Inc.). A slightly supersaturated condition was required to prevent the water droplets from changing in size due to evaporation or condensation. The supersaturated condition was established inside the tank by reducing the room temperature to less than 25°C.

Typical droplet size distributions are shown in Figure 5. In these experiments, aerosols were sampled at three different heights above the liquid surface. As seen, there was no observable change in the droplet size distribution due to evaporation or condensation. The distribution in this figure as well as in Figure 6 indicates the complex nature of the bubble breakup process.

Figures 6 and 7 show typical droplet and residue sizes obtained by the two measurement systems for the same conditions in the aeration tank. The APS and the ASAS-X systems sampled simultaneously at 15 cm above the surface of the bubbling suspension. As seen in Figure 6, the large majority of the APS-measured droplets were smaller than 6 μm.

The ASAS-X measured size distributions of PSL particle residues remaining after droplet evaporation are shown in Figure 7.

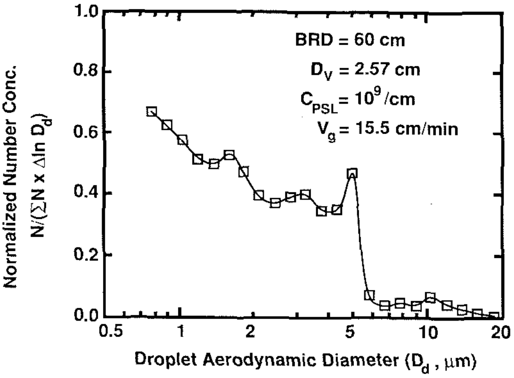
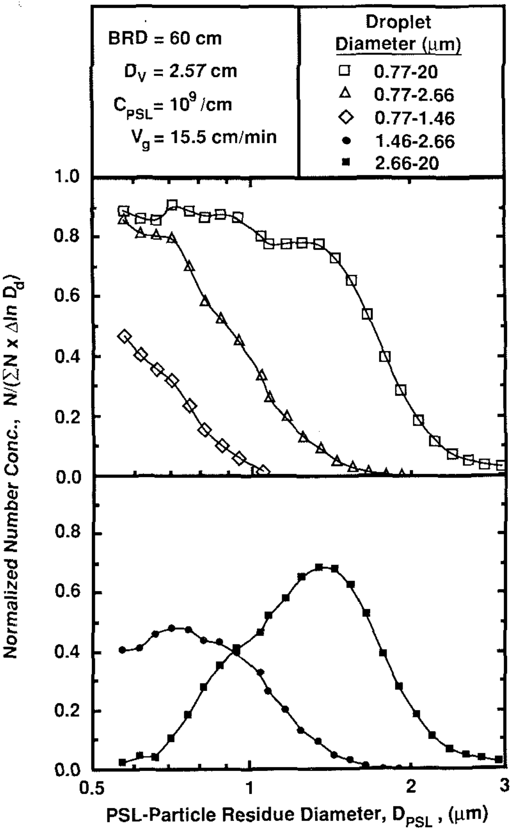


FIGURE 6. Normalized number concentration size distribution of droplets generated from fine bubbles.

FIGURE 7. Number size distribution of ASAS-X measured PSL particle residues remaining from droplets of five impactor-selected size ranges after their evaporation. The droplet number size distribution is shown in Figure 6.



The upper part of the figure shows the size distributions obtained by the aerosol spectrometer when the system is operated without an impactor (top curve) or with impactors that have D_{50}'s of 1.46 and 2.66 μm, respectively. The lower part of the figure shows the data obtained when these curves are subtracted from each other: the top curve is the residue size distribution in droplets larger than 2.66 μm, the bottom curve is for droplets in the 1.46–2.66-μm size range. For the entire droplet size range, the PSL particle residues were found to be as large as 3 μm with a geometric mean diameter of 1 μm and $\sigma_g = 1.45$ (count median diameter of PSL particles in bulk suspension = 0.59 μm and $\sigma_g = 1.14$). The residue size indicates that some droplets contain more than one PSL particles, an observation confirmed by electron micrographs of PSL particle residues collected on a polycarbonate filter. The micrographs show PSL particle residues as aggregates of different numbers of PSL particles.

A term called "residue-to-droplet ratio" is used to indicate the fraction of droplets that contain PSL particles. It is the ratio of the number concentration of PSL particle residues from droplets of a given size range to the number concentration of droplets in that size range. For the entire droplet size range, 61% of the generated droplets contain at least one PSL particle. In the droplet size range 2.66 μm or larger, 97% of the droplets contain one or more PSL particles. In the 1.46–2.66-μm range, 68% contain PSL particles, and in droplets smaller than 1.46 μm, only 24% contain one or more PSL particles. The airborne droplets have clearly been enriched with particles due to the bubble scavenging process mentioned above. For example, based on the bulk concentration of latex particles, approximately one out of every 250 droplets of 2 μm diameter is expected to contain a residue particle as opposed to the more than one in two observed.

CONCLUSIONS

The experimental system developed in this study can be used to investigate the transfer of materials from the liquid phase to the gaseous phase by multiple-bubble bursting at the surface of the bubbling liquid. A number of operational parameters, such as bubble size, superficial gas velocity, bubble rise distance, and the compositions of the liquid can be changed. Uniform bubbles of several sizes are obtained with capillary-tube gas diffusers. The system provides real-time information on concentrations and size distributions of the liquid droplets and of the materials remaining from droplets of various size ranges after their evaporation. The fraction of the generated droplets which contains the study material (residue-to-droplet ratio) can be determined. The system was employed to characterize the bubble-induced aerosols from a simulated activated sludge aeration tank and examples of these results are presented. However, with modifications, the system could be used to simulate other processes involving gas bubbling in a liquid, such as fermenters, health club whirlpools and other wastewater aeration tanks. The liquid in these systems will contain dissolved solids affecting the surface tension of the liquid suspension. This in turn will cause changes in the bubble size and shape, the particle collection efficiency of the bubbles, the droplet formation from the bubble film, and hence, the size of the residual particles in the final aerosol. Further studies will help elucidate these changes.

DISCLAIMER

Mention of product or company name does not constitute endorsement by the National Institute for Occupational Safety and Health.

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