Particle Production in Natural Seawater by Bubble Dissolution

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CANADIAN TECHNICAL REPORT OF

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PARTICLE PRODUCTION IN NATURAL SEAWATER BY BUBBLE DISSOLUTION

by

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ABSTRACT

    This study entails the design and development of equipment for the
production of organic particles by bubble dissolution, the collection of
the particles on filters, and the analysis of the aggregated organic substances
for carbon and nitrogen. The results suggest that the quantity and
character of the particulate matter produced by bubble dissolution are
complex functions of the prefilter pore size and of some unknown
seasonally dependent variation in the composition of particle forming
materials. The significance of bubble dissolution as a mechanism for the
production of particulate organic material is examined with regard to the
numbers of bubbles that might participate in the process in the marine
environment.

Johnson, Bruce, W.H. Sutcliffe, Jr. et R. Cooke, 1981.

Production de particules par dissolution de bulles dans l'eau

de mer.

RÉSUMÉ

Cette étude couvre la conception et la réalisation d'une

installation permettant de produire des particules de matière organique

par dissolution de bulles, de les recueillir sur un filtre, puis

de déterminer leur teneur en azote et en carbone. Les résultats

donnent à penser que la quantité et la nature des particules dépendent

de façon complexe de la dimension des pores du filtre et de certaines

variations saisonnières inconnues dans la composition des corps qui les

composent. On étudie la production de particules par dissolution de

bulles pour savoir dans quelle mesure ce processus intervient en

milieu marin.

Introduction

  The ubiquitous distribution of nonliving particulate organic material in the ocean has evoked considerable interest in the possible significance of this material as a source of food for zooplankton. What makes this concept especially attractive is the observation that particulate organic material is facilly generated by physical and chemical processes from the large reservoir of dissolved organic carbon in seawater.

  Early evidence of existence of amorphous organic particles came from Nishizawa et al (1954) who presented photographic evidence of aggregates greater than 1 mm in diameter. While these large visible aggregates may well represent a significant feature of particulate organic carbon in the ocean, (Aldredge 1979, Silver et al. 1978), the relative abundance of organic aggregates of all sizes including those of microscopic dimensions was not appreciated until the work of Riley (1963) in Long Island Sound. From his observations he concluded that, "....organic aggregates constitute a significant and often major part of the particulate matter in the sea".

  Evidence for the importance of organic particles as a foodstuff for organisms has come from several sources. Nonliving particulate organic material undoubtedly serves as a substrate for bacteria (Riley 1963) and its value in the sustainance of Artemia has also been demonstrated (Baylor and Sutcliffe 1963). While these organic particles may well be important for the maintenance of microzooplankton (Wangersky 1977), their use by nonselective feeders such as salps is probable (Aldredge 1979, Pomeroy and Diebel 1980).

  Numerous investigators have demonstrated both physical and chemical mechanisms for conversion of dissolved organic carbon to particulate form.

Baylor et al. (1962) and Sutcliffe et al. (1963) showed that particulate organic material could be produced by the bursting of bubbles. Wheeler (1975) demonstrated that particles could be produced by compressing surface films, and Sheldon et al. 1967 and Batoosingh et al. (1969) concluded that particles form "spontaneously" in seawater - a view that (as Wangersky notes 1977) seems to be supported by the experimental results of Ruckenstein and Nagaraizan (1975).

Johnson (1978) and Johnson and Cooke (1980) produced particles by dissolving bubbles, and examined the relationship between initial bubble size and particle size for such a mechanism. Their results show that a linear relationship between bubble size and particle size exists for dissolution in filtered seawater, but that no such correlation is apparent for bubble collapse in unfiltered seawater.

While the process of nonliving organic particle formation has been well described for dissolution of single bubbles in laboratory studies, the suitability of the process as a means of supplying particulate food in the ocean must be examined. It is the purpose of this work to investigate the significance of bubble dissolution for the formation of particulate material suitable as foodstuff for zooplankton.

Such an investigation entails:

1. The collection of particulate material produced by the process of bubble dissolution and analysis of this material for carbon and nitrogen content and other substances likely to be important as food materials for zooplankton.

2. The study of such particles by scanning electron microscope in an attempt to provide visual criteria for particle identification.

3. The estimation of oceanic production of foodstuff for zooplankton
    by the method of bubble dissolution.

Production of Particulate Material and Examination of its Chemical Composition

Experiments on the production of particulate organic carbon by bubble
bursting have typically used batches of water of less than 5 liters and
have bubbled through sintered glass frits with airflow rates of the order
of 1 liter/minute for periods of up to 24 hours (e.g. Batoosingh et al. 1968).
While these methods were shown to produce particulate organic material, they
are not well suited to the production of particles by bubble dissolution.

A number of criteria were examined for the development of a system
for particle generation from bubble dissolution. These criteria included
bubble size, particle generation efficiencies, amounts of particulate material
needed, and the potential of continuous flow versus batchwise operation.

The size of bubbles produced by forcing air through an orifice is
described by

$$\frac{\pi D^3 (\rho - \rho') g}{6} = \pi d \gamma$$

where D is bubble diameter, ρ and ρ' are respectively the densities of water
and air, g is gravitational acceleration, d is orifice size and γ is the
air water surface tension. For pore size of 0.5 - 1μm, corresponding
to the smallest pore commercially available sintered glass frits, bubbles
no smaller than 140μm in radius can be produced. Individual bubbles of
this size grow rather than dissolve if released from depths of several
meters (LeBlonde, 1969).

Increasing the ambient pressure of the water parcel provides increased
impetus for dissolution, reducing the time of bubble dissolution, and thus
the distance travelled. However, the pressures required to render this rise

distance manageable on a laboratory scale are of the order of several
atmospheres - pressures that are not safely or conveniently handled in
this type of experiment. Further complicating the problem of rise
distance, swarms of bubbles rise much more rapidly than do individual
bubbles by forcing convection on the water parcel, and thus the rise distance
becomes a strong function of bubble concentration as well as bubble size.
Because of the difficulty in dissolving large bubbles, and concern over
decreased efficiency of bubble scavenging as a function of bubble size
(Flint and Howarth, 1971, Cassel et al. 1975), considerable time and
effort were devoted to developing a bubble generator for production of
small bubbles. Because of the importance of small bubble generation in
these studies, the bubble generator, and its development and testing will
be described at length.

The Bubble Generator

Blanchard's analysis of the production of small bubbles by capillaries
(Blanchard, 1977), equally applicable to frits, reveals the nature of the
forces involved. As he concluded, a bubble is released when, "the bubble
buoyancy force equals the surface tension force that holds the bubble on
the tip". In order to produce small bubbles to examine enrichment of
bacteria in jet drops, Blanchard and Syzdek (1972) immersed the tip of a

capillary in a rotating tank of water and thus utilized drag forces instead of buoyancy forces to overcome the forces due to surface tension. Simularly drag forces can be utilized to produce large numbers of small bubbles with a frit. However, instead of using the rotating tank arrangement described by Blanchard and Syzdek, the fluid boundary layer that develops in flow along the surface of a frit can be most effectively controlled by restricting water flow to a narrow channel adjacent to the frit surface. After some analysis of the significant variables, the apparatus shown in figure 1 was assembled.

In this apparatus, a glass disc is maintained in position above the surface of a frit of the same diameter by a glass tube passing through the hole in the disc center. An O-ring in contact with a beaded region of the tube eliminates leaks while providing a small positive force for the disc against the frit. To maintain the relative positions of the frit and disc, the frit inlet tube is bent 180° into a position parallel to the disc inlet tube, and two glass tie rods are positioned in the gap.

In operation, water pumped through the tube in the center of the disc produces a separation of disc and frit and flows radially outward along the frit surface. Bubbles emerging from the frit are subjected to drag forces and are separated from the frit at sizes much smaller that if buoyancy were the only operating force.

With some simplifying assumptions, a model can be developed to predict bubble size as a function of water flow rate and disc and frit separation. These assumptions include:

1. The flow between the frit and disc is laminar and is fully developed over the entire frit surface.

2. Bubbles emerging from the frit pores experience significant force only from drag.

3. The gas to water flow rate is sufficiently small that the effects of coalescense are negligible and the properties of the fluid remain those of the entering water.

4. Fluid flow streamlines are not affected by interactions with bubbles upstream or affected by surface roughness of the frit. (To reduce surface roughness, the bubble-producing surface of the frit was polished before use with 400 grit waterproof abrasive paper).

Under these conditions, separation of the bubble at the frit occurs when a bubble radius is reached such that drag forces on the bubble euqals the surface tension force $\pi D\gamma$ as described by Blanchard (1977).

$$C_D \frac{\rho U_0^2 \, Ap}{2} = \pi D\gamma, \qquad\qquad 1.$$

where C_D is the drag coefficient, ρ is the fluid density, U_0 is the fluid velocity, Ap is the projected bubble area, γ is the gas-water surface tension, and D is the diameter of the frit pore. Because of the assumption of developed flow, the velocity varies across the fluid stream from zero at the frit and disc surfaces to a maximum at $S/2$, where S is the separation of disc and frit. Thus in the evaluation of the drag on an emerging bubble, the square of the velocity as a function of position relative to the frit surface can be integrated over the projected area of the bubble.

Batchelor (1967) presents an equation in two dimensions that describes fluid velocity U under a pressure gradient G between two fixed rigid planes:

$$U = \frac{G}{2\mu} (ys-y) \hspace{3cm} 2$$

where μ is the viscosity and y is the distance from one plane, in this case the frit. Rearrangement yields:

$$U = \frac{S^2 G}{8\mu} \left(\frac{4y}{S} - \frac{4y^2}{S^2} \right) \hspace{3cm} 3$$

At $y=0$ or $y=S$, U is zero, while at $y = \frac{S}{2}$, $U = \frac{S^2 G}{8\mu}$ which is evidently the maximum velocity. The average velocity can be determined by integrating the velocity, U, over S and dividing by S

$$U_{avg} = \frac{2}{S} \int_0^{S/2} \int_0^{} \frac{S^2 G}{8\mu} \left(\frac{4y}{S} - \frac{4y^2}{S^2} \right) dUdy \hspace{2cm} 4$$

yielding

$$U_{avg} = \frac{S^2 G}{12\mu} \hspace{3cm} 5$$

Comparing U_{avg} to U_{max} gives

$$U_{max} = 1.5 \, U_{avg} \hspace{3cm} 6$$

Here U_{avg} is merely

$$U_{avg} = \frac{U_{vol}}{2\pi LS} \hspace{3cm} 7$$

where U_{vol} is the volume flow rate of water, a know quantity, and L is the radial frit position. Thus the local velocity as a function of volume flow rate between the frit and disc is

$$U = \frac{3}{4\pi LS} U_{vol} \left(\frac{4y}{S} - \frac{4y^2}{S^2} \right) \hspace{2cm} 8$$

For a single bubble emerging from the frit, the result of U^2 taken over the projected bubble area can be found by converting to cylindrical coordinates, viz.

$$U^2 A_p = 2 \int_0^{\pi/2} \int_0^{2R\cos\theta} \int_0^{(\frac{3}{4\pi LS} U_{vol})^2 (\frac{16r^2}{S^2}\cos^2\theta - \frac{32r^3}{S^3}\cos^3\theta + \frac{16r^4}{S^4}\cos^4\theta)} dU^2 \, r \, dr \, d\theta \, , \qquad 9$$

with R being the bubble radius and r being the distance from the axis center at the surface of the frit.

Integration yields

$$U^2 A_p = \frac{U_{vol}^2 R^4}{2L^2 S^4 \pi} \left(\frac{45}{2} - 63 \frac{R}{S} + \frac{189}{4} \frac{R^2}{S^2} \right) \qquad 10$$

and the   drag on an emerging bubble is then

$$F_D = C_D \, \rho \left(\frac{U_{vol}^2 R^4}{4L^2 S^4 \pi} \right) \left(\frac{45}{2} - 63 \frac{R}{S} + \frac{189}{4} \frac{R^2}{S^2} \right). \qquad 11$$

Bubble separation occurs when

$$C_D \, \rho \left(\frac{U_{vol}^2 R^4}{4L^2 S^4 \pi} \right) \left(\frac{45}{2} - 63 \frac{R}{S} + \frac{189}{4} \frac{R^2}{S^2} \right) = \pi D \gamma . \qquad 12$$

Thus a bubble is swept from the frit when a radius is reached such that the dynamic separating force due to drag equals the retention force due to surface tension.

Assuming that the gas flow rate is constant over the frit surface, the rate of bubble generation N as a function of radial frit

position and total gas flow rate W can be determined by the expression

$$N = \frac{3}{\pi R^3} \int_{L_1}^{L_2} \int_0^{\pi/2} \int_0^{\pi L_w^2 \frac{W}{2}} dz d\theta L dL \ , \tag{13}$$

or

$$N = \int_{L_1}^{L_2} \frac{3 WL}{2\pi R^3 L_w^2} dL \ , \tag{14}$$

where L_w is the radius of the bubble generating surface of the frit, Z is the axis at the frit center normal to the plane of the frit surface and R is the radius of bubble at separation for position L on the frit.

However, from 12,

$$L = \frac{C_D \rho U_{vol}^2 R^4}{4 S^4 \pi^2 D\gamma} \left(\frac{45}{2} - 63 \frac{R}{S} + \frac{189}{4} \frac{R^2}{S^2}\right)^{1/2} \tag{15}$$

and thus N can be expressed in terms of R alone as

$$N = \frac{3C_D W\rho U_{vol}^2}{8 L_w^2 S^4 \pi^3 D\gamma} \int_{R_1}^{R_2} 45 - \frac{315}{2} \frac{R}{S} + \frac{567}{4} \frac{R^2}{S^2} dR \tag{16}$$

The result can then be expressed as

$$N = \frac{3C_D W\rho U_{vol}^2}{8 L_w^2 S^4 \pi^3 D\gamma} \left[45R - \frac{315}{4} \frac{R^2}{S} + \frac{567}{12} \frac{R^3}{S^2} \right]_{R_1}^{R_2} . \tag{17}$$

The value of the drag coefficient for bubble separation from the frit can be determined through use of equation 1,

$$C_D = \frac{\pi D \gamma}{A_p \rho U_o^2 / 2} \qquad\qquad 18$$

Since the Reynolds Number,

$$N_{Re} = \frac{2 R U_o}{\nu} \qquad\qquad 19$$

Where ν is the kinematic viscosity then

$$C_D = \frac{1}{N_{Re}^2} \frac{8 D \gamma}{\rho \nu^2} \qquad\qquad 20$$

and

$$C_D N_{Re}^2 = \frac{8 D \gamma}{\rho \nu^2}$$

Since ρ, γ, and ν are properties of the water and D, the frit pore diameter is known within narrow limits, then the value of $C_D N_{Re}^2$ is also known. It is interesting to note that for constant frit pore size, $C_D N_{Re}^2$ is independent of the frit geometry, the bubble size, and the water velocity, and that a unique value of the drag coefficient is predicted from a C_D versus N_{Re} curve.

Thus for given separation of frit and disc and for a given flow rate of water, the range of bubble size produced by the bubble generating

apparatus can be calculated from 12. Equation 17 can then be utilized to determine the numbers of bubbles generated for the known bubble size range, or for any part of that range.

Experimental

The apparatus for the experiment can be seen in figure 2. Seawater was pumped with a peristaltic pump from a reservoir, passed through a partially filled 2 liter round bottomed flask to damp surges, and introduced into the disc centered tube of the bubble generator. Gas (He in this case, although the type of gas is largely immaterial) was introduced into the glass tube leading to a 3.2 cm diameter glass frit of 4-5.5 µm pore size. Before each experiment the water flow rate was measured by holding the bubble generator over a volumetric flask and noting the time required for filling. The gas flow rate was found by maintaining the bubble generator in a water reservoir and determining the rate of water displacement by the gas in an inverted water filled beaker. As the flow-forced separation of disc and frit varied according to flow rate, the separation was measured with calibrated shim stock under the conditions of flow for each experiment.

To begin an experiment the bubble generator was operated for several minutes in a beaker of sea water to insure a steady state. It then was introduced into a reservoir of seawater in which there was a seawater-filled plexiglass cell similar to that described by Cipriano (1980). This plexiglass cell of 1 x 6 cm rectangular cross section was inverted

with its open end slightly below the reservoir surface while the main portion, sealed at the upper end, protruded above the air water interface. The generator attitude was maintained such that emerging bubbles were injected directly into the seawater filled cell, and photographs were made of a region very near the point of emergence of bubbles from the generator to insure that the source population and not an aged population was being photographed. Illumination was provided by strobe lamps facing the three accessible edges of the cell. The lens magnification was ½ x and the film and method of development are those described in Johnson and Cooke (1979). Bubble images were measured directly from the film by microscope with the aid of an ocular micrometer with from 200 to 400 bubbles measured per distribution. With these methods, bubbles as small as 12 μm in radius were measurable.

Results and Discussion

The distribution of bubbles produced by the generator with a gas flow rate of 8 cm^3/min and water flow rates of 243, 500 and 776 cm^3/min appear in the histograms of figures 3a-c respectively. As can be seen in these distributions, virtually all of the bubbles produced under the flow conditions described are less than 100 μm in radius with a lower limit of about 15 μm and a range that narrows with increasing water flow. In the narrowest distribution, that for 776 cm^3/min water flow, the range is represented by radii of about 15 to 50 μm with an average radius of only 25 μm.

Included in figure 3 are the range of sizes and the shapes of the distributions predicted by equations 12 and 17 utilizing a drag coefficient of 1.0. The value of the drag coefficient can not be taken from curves of C_D versus N_{Re} that are determined for resistance of solid spheres, because unaged bubbles possess mobile interfaces (Levich, 1962), and thus velocity at the bubble surface does not go to zero. Energy is dissipated by gas motion in the bubble interior, and drag experienced by an unaged bubble is less than if the interface were rigid. As surface active material accumulates at the interface the bubble assumes a more rigid character and in times of the order of seconds (Detwiler and Blanchard, 1978) behaves as a solid sphere.

For the frit used in these experiments, limits on the value of the drag coefficient can be determined for the average pore size, D, for fluid sphere behavior (Moore, 1963) and for solid sphere resistance (Batchelor, 1967) and are respectively 0.5 and 2.0. An intermediate value of 1 for C_D was found to give a very good fit to the data although the sensitivity of the model to the value of the drag coefficient is such that over the range of C_D of 0.5 to 1.5 the predictions of the model do not materially change. Such a drag coefficient, intermediate between that expected for solid and fluid sphere behavior, was found also by Detwiler and Blanchard (1978) and Tedesco and Blanchard (1979) when they examined the characteristics of rise for both aged and newly formed bubbles.

The size ranges and distribution shapes predicted by the model are seen to be quite good, with only a small fraction of the bubbles in any of the distributions falling outside of the regions predicted. As equation 20

predicts, a single value of the drag coefficient seems to apply over the wide range of conditions of water flow rate and bubble size of the experiments.

The validity of the assumptions upon which the model is based can be examined. The Reynolds number of the channel, $\frac{SU_{avg}}{\nu}$, where ν is the kinematic viscosity, has a minimum of about 40 that corresponds to a water flow of 243 cm^3 min^{-1} at the edge of the frit. A maximum Reynolds number of about 500 can be determined for the conditions of 776 cm^3 min^{-1} water flow at the point of introduction of flow to the channel between frit and disc. Thus if 2100 represents the Reynolds number above which transition to turbulent flow is possible, the flow conditions that obtained when the bubble generator was operated are well within the laminar regime.

The equation describing the development of flow between two flat plates (Schlichting, 1934) can be used to examine the assumption that developed flow over the region between frit and disc exists. In this treatment, the flow is nearly fully developed when

$$\frac{L}{S} = .04N_{Re},$$

where L is distance from the front edge of the plate and S is the separation of the plates. For a Reynolds number of 500, the calculated value of L is 0.35 cm. This solution describes nearly fully developed flow, or a boundary layer that reaches almost to S/2, but bubbles in this transition region are sheared from the frit at diameters of closer to S/5, and thus for emerging bubbles flow is effectively developed at much less than 0.35 cm. While the fluid velocity profile in the transition region is not exactly that of developed flow, the assumption of developed flow over the entire frit surface would seem to be a reasonable approximation.

The best proof of the validity of the assumptions made in developing
equations 12 and 17 lies in the fit of the predicted range and distribution
shapes to the data. However, at higher ratios of gas flow to water flow,
coalescense seems to become important as demonstrated for a gas flow of
41 cm^3 min^{-1}, figure 4a-c. The absence of a term for the gas flow rate
in equation 12 means that the ranges of the distributions for 41 cm^3 min^{-1}
gas flow should, at about the same water flow rates, be nearly the same
as those in figure 3a-c for 8 cm^3 min^{-1} gas flow. This is obviously not
true, with the greater distribution ranges at the higher gas flow rate
being ascribable to bubble coalescence and perhaps to deflection of flow
streamlines by bubbles emerging in upstream flow. While populations of
bubbles produced under conditions that violate the assumptions of the
model may not be predictable from the theory and must be determined
empirically, they will undoubtedly be useful in oceanographic studies
of bubble bursting and scavenging.

System Design

An important consideration in the design of a system for particle production is that sufficient amounts of material be generated for chemical analysis. While amounts of the order of tens of micrograms are adequate for C-H-N determinations, the generation of as large amounts of material as possible is desirable for other types of analysis. Batoosingh et al. (1968) found that a maximum of about $71 \pm 70\mu g$ C/ℓ could be produced in a single batchwise bubbling operation and that this production occurred at a diminishing rate over the 24 hour bubbling period. Assuming that comparable amounts of material in a liter of seawater can be converted to particulate form by bubble dissolution and that this conversion similarly occurs with diminished facility as a function of time, some important design criteria can be established. Since particle production occurs with greatest efficiency at the commencement of bubbling, the rate of particle production for a given reaction vessel and gas flow rate in batch operation increases with the volume of water treated. This means that the amount of particulate material generated as a function of time is positively correlated with the frequency of renewal of water in the vessel in batch operation at constant bubbling rate. Thus as the frequency of batch renewal is increased, a continuous flow system is approximated and is evidently the most efficient means of employing a reaction vessel to produce particulate material by bubbling.

Prefiltration

The procedure for particle production by bubble dissolution entails prefiltration of the seawater sample prior to bubbling, introduction and dissolution of bubbles, and harvesting of the resulting particles with a collection filter. Further, if some samples are bubbled and others are

treated as methods controls, the contribution of particles from extraneous sources is apparent, and the production of particles from bubble dissolution can be isolated.

Prefiltration ensures that particles already present in the seawater sample and of dimensions larger than the collection filter pore size do not dominate the feature under examination. Indeed, there is even some indication that prefiltration is essential to new particle formation (Batoosingh et al. 1968).

Methods Controls

Unbubbled controls provide a means of evaluating the contribution of particles from sources other than bubbles to the particulate material obtained on the collection filter. The importance of these controls in particle production experiments has been demonstrated by Menzel (1966) and Batoosingh et al. (1968) who found that a significant percentage and sometimes the total POC on the collection filter could be explained by the control.

There are several apparent sources of the particulate material that appears on the methods control filter. Normally in particle production experiments the prefilter and particle collection filter are of the same or nearly the same pore size, however, as Sheldon and Sutcliffe (1969) and Sheldon (1972) have shown filters are not effective for size class separations. The problem, as they point out, is that a filter not only retains all particles larger than a certain size (not necessarily the nominal pore size), but also retains a percentage of the particles below that critical size. Thus a significant amount of material that passes through the prefilter collects on a downstream filter of comparable pore size.

"Spontaneous" generation of particles is a demonstrated feature of particle dynamics in seawater (Sheldon et al. 1967) and is a source of particulate material collected on the methods control filter. As Sheldon et al. found, particles form spontaneously and rapidly after filtration. While the contribution from this effect may be small for experiments of short duration, too little data exist for adequate evaluation. Some experiments on particle generation have been conducted with the results of immediate refiltration used as a control that is compared to the results of filtration after as much as 24 hours of bubbling. Obviously any time dependent changes that occur in the particle spectrum, including spontaneous formation, would not be accounted for in the control, but would be attributed to bubble activity.

Another contribution to particulate material collected on the methods control filter comes from contamination that is introduced into the seawater sample following prefiltration. Except for introduction of contaminants via the air supply for bubbling, such contamination is equally likely to appear in methods control samples and would be apparent from statistical analysis.

A final contribution of material to the method control filter comes from adsorption of organic material. However, the relative effect of this adsorption decreases with increasing volume of sample (Gordon and Sutcliffe 1974).

Several design criteria are apparent from this examination of the sources of contribution of material to the control filter. These include:

1. Control samples are essential for an evaluation of experiments
   on particle production. These controls must be methods controls that

undergo all of the experimental procedure except for bubbling. Only from a comparison of bubbled samples and unbubbled method controls can the real particle production from bubbling be evaluated.

2. The relative contribution of adsorption of organic material on the filters as well as most kinds of random contamination decrease with increasing volume of water treated.

3. The effect of spontaneous formation of particulate material after prefiltration, along with alterations in the particle spectrum that occur from biological processes can be reduced by minimizing the period of sample treatment.

From the above arguments it appears that a continuous flow system represents the most efficient system in terms of rate of production of particulate material by bubble dissolution, and that minimization of the affects of spontaneous particle formation, biological processes, adsorption and random contamination can be affected by processing large volumes of water over reasonably short time periods.

The Apparatus (A Laboratory Langmuir Cell)

With the aforementioned design considerations in mind, the apparatus of figure 5 was assembled. This flow through system consisted of a 20 cm diameter prefilter holder, a vessel of 2" inside diameter made of Kimax tempered glass tubing, a teflon and glass outflow system with a 47 mm millipore inline filter holder for particle collection, a Cole Parmer peristaltic pump for recirculation and an all glass bubble generator. To minimize sources of contamination all materials that were used in the apparatus were glass and

teflon with the exception of the prefilter holder made of PVC, the collection

filter holder made of polypropylene, and the silicon tubing of the peristaltic

pump. Those components of the system that were not made of glass or teflon

were aged for a minimum of two weeks in seawater, and thoroughly rinsed in

distilled water before being incorporated into the system. Air was supplied

from a small compressor and was passed through a tube of activated charcoal

and then through a millipore filter of $0.22\mu m$ before entering the bubble

generator.

In operation, water from the Bedford Institute seawater facility flows

through the prefilter and into the Kimax glass tubing used as a bubbling

vessel. As this water passes downward through the vessel, bubbles from

the bubble generator are entrained and dissolve in transit to the outflow

ports. At the outflow port the flow divides, with one stream passing

through the particle collection filter, and the other being recirculated,

first through the pump, and then back into the vessel through the bubble

generator.

The recirculation system permits operation under a greater range of

flow conditions and attendant residence times of water in the system.

Water flow through the column must be sufficiently fast that bubbles

emerging from the generator are entrained, but must be slow enough that these

bubbles dissolve before reaching the outflow port. These requirements are

achieved through control of water flow from the prefilter and from the

recirculation pump, with the relative contributions of each determining

the residence time of water in the column, i.e. decrease of water flow into

the system through the prefilter and out through the collection filter

provides an increase of residence time of water in the system, but must be

accompanied by an increase in recirculation rate to ensure bubble entrainment. The control of water flow into and out of the system is accomplished with the adjustable clamps that are located on the outflow lines, while the pump controls are used to establish the recirculation rate.

The control of bubble dissolution rate in such a system can only be affected through application of pressure. This is because the impetus for bubble dissolution comes from the difference in fugacities (or under these conditions pressures) of the gases dissolved in the water and the gases within the bubbles. With the gases dissolved in surface seawater being very nearly in equilibrium with the atmosphere, bubble dissolution requires the imposition of pressures above atmospheric.

Thus the thermodynamic and kinetic control of bubble dissolution depends upon the magnitude of the difference in partial pressures of the gases within the bubble, and those dissolved in the water. The control of pressure is accomplished through adjustment of the three way valve on the inlet line, thereby establishing how much of the 80 psi seawater line pressure is applied to the flow through system.

Experimental Procedure

Filters to be used for collection of generated particles were precombusted for four hours, with those of silver being heated at 400°C and those composed of glass fiber heated at 450°C. All precombusted filters, as well as blank filters and collection filters were kept in Steripak plastic petri dishes or in Pyrex petri dishes that were precombusted in a manner similar to that used for glass fiber filters. No difference in values of carbon on blank filters was found to occur as a result of the type of petri dish that was used.

Because of the somewhat arbitrary oceanographic definition of particulate material that specifies retention by a 0.45μm filter, removal of "particles" that are already present in the water, prior to bubbling, requires prefiltration through a filter of pore size no larger than 0.45μm. Gelman A/E glass fiber filters with a nominal pore size of 0.33μm were chosen for the prefilter because of the less than 0.45μm pore size requirement and because they are available in sheet form, 20.3 x 25.4 cm. This large size was necessary because the prefilter holder was designed to be as large as practicable (20 cm in diameter) to provide adequate filtering capacity for the total water flow volume.

Particle collection filters were primarily GFC filters with a nominal pore size of 1.2μm or Flotronics 0.8μm silver filters. These filters were chosen because their particle retention characteristics have been well described, (Sheldon and Sutcliffe 1969, Sheldon 1972), and because their pore sizes are compatible with the type of prefilter that was used, i.e. there should be little overlap in the characteristic curves that describe the % particle retention versus particle diameter for the collection filter and prefilter. This means that a large percentage of the particles that pass through the prefilter would also pass through the collection filter and particles formed by bubble dissolution would constitute the major source of particles on the collection filter. According to Sheldon (1972) nearly 100% of particles of 0.33μm pass through a GFC filter and more than 90% through a 0.8μm Flotronics silver filter. While the characteristics of the Gelman A/E filter used as a prefilter are not known other than the nominal pore size of 0.33μm, the performance of this filter should be similar to that of other glass fiber filters. If this assumption is correct,

then the overlap in the retention curves of the prefilter and the collection filter would be little, and particle formation from bubble dissolution should be the major contributor of particles on the collection filter. These considerations are essential if chemical analysis of material on the collection filter is to be made, because retention of particles other than those of interest, i.e., those from particle dissolution, would obfuscate the results of the analysis.

Prior to the beginning of an experiment, the bubble generator and filter holders were cleaned in strong detergent solution, and these along with the column, tubing, and other components of the system were rinsed thoroughly in distilled water. Following this rinse the appropriate filters were placed in the prefilter and collection filter holders and then the seawater valve was opened and the column was filled. For this filling, a vent was opened at the top of the column to allow air to escape, and the water outflow lines were clamped to prohibit water loss. When the column was full, the air vent was closed, and the clamps on the collection filter bypass line was loosened to permit a water flow of 500 mℓ/min. Thus the column was rinsed, and conditioned with seawater for 15 minutes, with no water having passed through the collection filter.

To begin an experiment the recirculation pump was turned on, providing a recirculated water flow of 500 mℓ/min through the bubble generator. In the samples in which bubbles were to be dissolved, air was introduced at 4 mℓ/minute into the gas inlet tube of the bubble generator. As the water in which the first of the bubbles had dissolved reached the outflow port, the water outflow of 500 mℓ/min was changed from the bypass line to the collection filter line, thus permitting the collection of particles produced by bubble

dissolution. The water that passed through the collection filter was stored in a 26 liter bucket to ensure that the amount of water that had been treated was accurately known.

At least two filters for blanks, a particle laden filter from bubble dissolution, and a methods control filter were collected for each experiment. The filters for blanks were secured in the filter holder, removed, and saved for analysis. The methods control filter was subjected, as nearly as possible, to the same procedures in handling, and experimental treatment as was the filter through which bubbled water passed, with the exception that no bubbles were introduced. Such a methods control ensures that the results of filter analysis can be evaluated and contribution of particles from bubble dissolution ascertained.

All filters were sealed in petri dishes as they were removed from the collection filter holder, and immediately placed in a freezer. Before analysis 150 $\mu\ell$ of 1 N HCL was added to each filter, and with lids canted slightly (a position maintained with tape) to allow water escape, the petri dishes were placed in a dessicator under vacuum. When the filters were dry, they were run in a Perkin Elmer model 240B C-H-N analyzer.

Results and Discussion

The results of experiments in which entering seawater was prefiltered through a single Gelman A/E filter are shown in Table 1. The date of the experiment, the type of collection filter, and the amount of water treated are included in the table, as are the results of carbon and nitrogen analysis.

As can be seen from the results of the carbon analysis, there seems to be little difference in the amount of carbon per liter for water in which bubbles had dissolved, and that from the unbubbled method controls. The

results of the nitrogen analysis are similar, with little difference in PON being apparent between the method control and the sample in which bubbles had dissolved. However, the ratio of moles of carbon to moles of nitrogen is more interesting, with this C/N ratio being greater in each sample pair for the sample in which bubbles were dissolved. In a paired T test the significance fell on better than the 95% confidence level. While with more data it might be demonstrated that particulate carbon is produced on a statistically significant level, the important conclusion for this study is that insufficient particle production occurred for analysis of the material.

Actually this failure to produce appreciable amounts of particulate material under the conditions of these experiments is not surprising. Batoosingh et al. (1968) found that pore size of the prefilter determined the degree of success of particle production by bubbling. They found that particles could not be produced even with 24 hours of bubbling in water that had been prefiltered through a 0.22μm filter, and they described their particle production results as "completely insignificant" for bubbling of water prefiltered through a 0.45μm filter. Thus it is not surprising that measurable amounts of particulate matter could not be produced by bubble dissolution after prefiltration through the Gelman A/E filter of 0.33μm pore size.

What is apparent from an examination of the absolute amounts of carbon on the filters, both particle collection and method control, is the necessity of the method control approach in particle production experiments. Without the methods control filters for comparison, the 7 to 10 μg C/ℓ that was collected on the particle collection filters would be attributed to particles produced by bubble dissolution.

While less than 10 µg C/ℓ was found on the methods control filters of these experiments, 18 to 36 µg C/ℓ was reported by Menzel (1966) for the results of immediate refiltration, and typically greater than 25 µg C/ℓ was found by Batoosingh et al. (1968) for filtration of unbubbled controls. This material on the control filters in the various experiments is probably due to the aforementioned inefficiency of filters for size class separations, the adsorbtion of surface active organics onto filters and the "spontaneous" generation of organic particles.

In a series of experiments that were designed to reduce the amount of extraneous particulate material, i.e., that not produced by bubble dissolution, two A/E prefilters were placed in series. At the same time to test the possibility that some size difference might exist between the particles produced by bubble dissolution and those comprising the background population, a Whatman GFD collection filter, nominal pore size of 2.5µm, was placed in a filter holder upstream and in series with the GFC collection filter. All other conditions were the same as those previously described for the single A/E prefilter experiments.

As can be seen in Table 2, the difference per liter between POC in the sample in which bubbles were dissolved and the methods control was again not significant. Again, the failure to produce particulate material from bubble dissolution might be attributed to the fine pore prefilter. Further, the sum of the amounts of carbon on the two filters, GFC and GFD, for each experiment shows that the amount of extraneous material per liter was not reduced.

In order to test the assumption that pore size is the significant variable for particle production by bubble dissolution, a GFD filter (2.5µm nominal pore size) was employed as a prefilter. Since GFD filters were not available in sheet form to fit the 20 cm prefilter holder, 4.7 cm filter holders were

employed instead. Rapid particle loading of the small diameter filters
necessitated the use of two filter holders in parallel and a third downstream
in series. When one of these prefilters in parallel became loaded with
particles, it was isolated with clamps, and the filter was replaced. The
downstream filter in series ensured the removal of any particles that were
introduced during filter changes upstream. Because of the larger pore size
of the prefilter in these experiments, a GFD filter was also used for particle
collection. Except for these differences in filters, all other conditions of
the experiment were as previously described.

From the results of these experiments, Table 3, it is apparent that
measurable amounts of filterable organic carbon and nitrogen were generated
by bubble dissolution. These amounts of POC produced ranged from a low of
0.59 μg/ℓ to a high of 3.36 μg/ℓ and averaged 1.95 μg C/ℓ, while filterable
organic nitrogen increased an average of 1.33 μg/liter.

While this 1.95 μg of carbon per liter produced by bubble dissolution
seems low relative to the yields obtained by Batoosingh et al 1968, other
criteria must be examined. In the experiment reported by Batoosingh et al.
in which an adequate methods control was performed (experiment 7) they reported
an average production of 118 $\overset{+}{-}$ 70 μg C/ℓ, or an average increase of 71 $\overset{+}{-}$ 70 μg C/ℓ.
In their experiments they bubbled 4 liters of seawater for 24 hours at a gas
flow rate of 1 liter per minute, for a total of 1,440 liters of air, while for
the flow through system the water residence time was only 8 minutes and only
8 mℓ of air were injected per liter of seawater.

The importance in the lower gas input lies, if not in a closer approach
to real ocean conditions, in a considerably reduced potential for contamination
via the air supply. However, the most important conclusion to be drawn from
the comparison of the two experiments is that different effects are probably
being measured, even beyond the obvious difference between the processes of

bubble bursting and bubble dissolution.

In the 24 hour bubble bursting experiments of Batoosingh et al. chemical and biological processes probably occur in response to the changes in the particle size spectrum, temperature, and physical regime of turbulence. The bubble dissolution experiments with an 8 minute water residence time and very little temperature change may well approximate the instantaneous potential for particle formation. This conclusion follows from the conditions of the experiment.

With an air flow rate of 4 mℓ/minute and water flow through the bubble generator of 500 mℓ/minute the bubble spectrum of figure 3b applies to these bubble dissolution experiments. The dissolution of a bubble of average size from this distribution should produce a particle of at least 3.5μm according to the curve of particle diameter versus initial bubble size (Johnson and Cooke 1980). If this is true then the dissolution of the 2.2×10^8 bubbles comprising the 8 cm^3 of air injected per liter of seawater should produce about $8 \times 10^{-4} cm^3$ of particulate material or assuming a density of 1.0 for the particulate material produced, 0.8 mg of particles per liter of seawater bubbled. If 50% of that were organic matter (Heisbrunner and Wangersky 1976) of which 50% was carbon, then 200 μg of carbon might be produced per liter of seawater by the bubbles dissolved.

A comparison of the 200 μg C/ℓ that might potentially be produced and the 1.95 μg C/liter observed must be explained by either a paucity of particle forming material that is instantaneously available, or to a low efficiency of scavenging by bubbles. Which of these explanations is valid is difficult to assess. However, it is known that small bubbles rising in seawater become coated with surface active materials in times of the order of seconds (Detwiler and Blanchard 1978) thus suggesting that scavenging efficiencies of bubbles are reasonably high.

From the results of these bubble dissolution experiments certain aspects of particle formation in seawater are suggested. Perhaps most important is the observation that particle formation is limited by either the instantaneous amount of particle forming material present, or by some bound on the efficiency of conversion of this material to particulate form. The second conclusion is that pore size of the prefilter is an important parameter in particle formation studies and is a result of this study and of those of Batoosingh et al. 1968. The final and perhaps most interesting observation is that some fractionation of carbon relative to nitrogen is possible as a result of particle formation from bubble dissolution, and in these experiments was manifest as a somewhat higher molar ratio of C/N for single prefiltration with a Gelman A/E filter. It is not clear however whether this enrichment of carbon relative to nitrogen is due to some aspect of size class unique to the condition of single Gelman A/E prefiltration, or whether the result is seasonal. The results of Table 2 do not show the same enrichment of carbon relative to nitrogen, and yet the conditions of the experiment differed only in that a double A/E prefilter was used instead of the single filter used for the experiments of Table 1. Thus it seems probable that the enrichments of carbon seen in Table 1 are seasonal and reflect some aspect of winter DOC, while the samples of Table 2, taken in June and July, show no systematic enrichments.

Visual Criteria for Particle Identification

The primary purpose in seeking visual criteria for distinguishing particles produced by bubble dissolution from other marine particles is that of providing a means of assessing the significance of the process by the examination of naturally occurring particles.

Aged, naturally occurring particles generally appear with phytoplankton, fecal matter, silt, and unidentifiable detritus imbedded in a yellow to brown amorphous matrix. These aggregates have been reported to be of microns to meters in size (e.g., Riley 1963, Nishizawa et al. 1954) and to be a general feature of the world oceans.

Reported sources of these aggregates have included whole or fragmented appendicularian housings (Aldredge 1979) and salp feces (Pomeroy and Diebel, 1980) as well as physical chemical conversion of dissolved organic material to particulate form. After formation, regardless of the source, particles are colonized by bacteria, probably grow by acretion of new material and aggregate with other particles. Thus the sources of aged particles or aggregates cannot generally be determined on the basis of visual criteria.

Some basis for distinguishing newly formed particles of bubble dissolution origin from those produced by bubble bursting might be determined from an examination of the mechanisms involved. In bubble bursting a segment of the bubble surface film is presumed to accompany the downward jet of water and remain as an organic particle, while in bubble dissolution the surface film is seen to collapse, folding as in insoluable monolayer collapse and remaining in particulate form.

Particles that have been filtered from samples of seawater in which large numbers of bubbles had burst are seen to be thin and uniformly translucent, probably a single bubble film thickness (a molecular multilayer perhaps) and sharp edged, probably as the result of shredding of the surface film during the violent bursting process (Riley 1963). Particles produced by bubble dissolution are also generally translucent, but are less uniformly so than are those from bubble bursting. This is because

the surface film folding that accompanies dissolution provides at least two bubble film thickness. Further, the particle from bubble dissolution is generally rounded on the edges and of a more three dimensional character. A comparison of the two types of particles can be seen in Figure 6 in which a flake that had adsorbed to the surface of a 60μm bubble was seen to form an aggregate with the particle that was produced when the bubble dissolved. The particle that resulted from bubble dissolution appears in the upper half of the aggregate while the flake is apparent in the lower half.

Particles produced by bubble dissolution were examined by scanning electron microscope in an attempt to provide a better basis for visual assessment. To ensure the formation of large particles for accurate identification, large bubbles, of the order of several hundred microns in radius were dissolved against a microscope cover glass that was suspended in seawater. This seawater had been partially degassed under vacuum to promote bubble dissolution and was filtered through a GFC filter prior to introducing the coverslip and bubbles.

After bubble dissolution the coverslip with particles was rinsed in distilled water, mounted directly on the conductive SEM stub (particle side up), and dessicated. The resulting SEM photographs appear in Figure 7.

While the particles were seen to be derived from the dissolved bubbles and are of characteristic appearances, one artifact of their mode of formation against the coverslip is apparent. The large size of the bubbles that were dissolved gave them a buoyancy that resulted in distortion and attendant increased contact area against the glass. When the bubble dissolved, portions of

the surface film "wetted" the glass and were torn from the main body of the particle. The segments of the film that adhered to the glass are visible as small pieces around the main particle perimeter. Some distortion of the main particles, manifest as more angular dimensions, may have resulted from the film adhesion to the glass. However, the small particles that were separated from the main particle provide an indication of the thickness of a single layer bubble surface film, probably similar to a flake produced by bubble bursting. In contrast, the main particle results from a multiply folded bubble surface film and thus appears less transparent and more three dimensional in character.

While newly formed particles produced by bubble dissolution might be identifiable in a sample of naturally occurring particles, the basis for identification would certainly rapidly disappear upon particle aging.

An Estimate of the Production of Foodstuff for Zooplankton

The failure in these experiments to produce large amounts of particulate material by bubble dissolution does not diminish the estimation of the probable significance of the process for marine particle production. However, this estimation becomes difficult with a strict adherence to the definition of particle that requires retention by a filter of 0.45μm pore size. If the operational definition of particle is required, then the results of Tables 1 and 2 apply (0.33μm prefilter pore size) and any estimate of particle production must be based upon a knowledge of which is limiting, particle producing material or dissolving bubbles. The results of Table 1 show some enrichment of carbon relative to nitrogen, thereby suggesting that some particle production has occurred, but production

that is apparently lost in the random error of the experiments. It
must be remembered that the bubble dissolution study was conducted to
produce particulate material for analysis, and that the 8 minute residence
time of seawater in the column provides no information on the rate of
generation of particle forming material in seawater.

Estimates of the significance of particle production by bubble
dissolution can be more easily made if based upon the particle production
results of GFD prefiltered water (Table 3). With 1.95 µg C/ℓ produced in
8 minutes of bubbling effort, and the results of Batoosingh et al. 1968
to suggest that much more than this is available over 24 hours of bubbling,
then the rate of particle production in the ocean is certainly limited by
the numbers of bubbles that participate in the process of bubble dissolution.

An estimate of the numbers of bubbles involved in the process of
bubble dissolution has been made, (Johnson 1978) and particle input from
bubble dissolution was calculated to be 2g/m^2/year at the rate of input
of sea state 3 and 10g/m^2/year at sea state five.

However, estimates based upon world ocean conditions might be made
on the basis of new information on bubble populations in a breaking wave
(Cypriano and Blanchard in press). They found approximately 45 bubbles
of 50 to 150µm in radius being injected per cm^2 per second in their model wave.
This result is determined by assuming that the entire tank surface represents
the "white water" associated with the model wave (a conservative assumption),
and thus the 2 x 10^4 bubbles/sec that reach the tank surface can be divided
by the tank surface area to obtain the bubble injection rate.

This 45 bubbles/cm^2/sec of size 50 to 150µm represents 4.5 x 10^5
bubbles/m^2/sec injected by a breaking wave. If 3.5% of the ocean

is covered by whitecaps (Blanchard 1975) then about 5×10^{11} bubbles/m^2/year are injected for the 50 - 150μm size range. Since a particle of 15μm in diameter or $1.8 \times 10^{-9} cm^3$ (assuming spherical form) is produced by the average size bubble, in the size range of interest (Johnson and Cooke 1980), then only 1 bubble in 225 of those injected in this size range need dissolve to account for 4 cm^3 of particulate matter or $\sim$ 1gC/m^2/year. Because the depth that a bubble of 100μm in radius needs to be injected to dissolve before reaching the air-sea interface is only 0.75 meters at 0°C (deeper at higher temperatures), the significance of particle production by bubble dissolution might be great indeed.

The quality of particles from bubble dissolution as food for zooplankton is difficult to assess, but some information is available in Tables 1 and 3. If the enrichment of carbon seen in Table 1 demonstrates a general feature of particle production in winter, then one must conclude that particles produced at that time are deficient in nitrogen, and might well be composed of carbohydrates or lipids. The particles produced in the GFD prefiltered water, Table 3, show no systematic enrichment of carbon or nitrogen, thus it is probable that the particles produced by bubble dissolution possess a molar C/N ratio similar to the bulk material on the filter. This ratio ranges from 5.01 to 6.45, and thus, all are below the 6.7 Redfield ratio, making them relatively rich in nitrogen. From these arguments it would appear that particles produced in the winter by bubble dissolution are nitrogen deficient while those produced in the summer in these experiments reflect the ambient particle composition, and are richer in nitrogen.

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TABLE 1 Single A/E Prefilter (0.33μ)

Date	Collection Filter Type	S.W.Volume Treated Liters	POC With Bubbles μg	POC Without Bubbles μg	Difference Per Liter μg	Control Carbon Per Liter μg	PON With Bubbles μg	PON Without Bubbles μg	Molar Ratio With Bubbles C/N	Molar Ratio Without Bubbles C/N
23/10/79	GFC (1.2μ)	28	301	240	2.9	8.9	52	49	6.77	5.73
31/10/79	GFC	42	395	380	0.12	9.0	66	66	7.00	6.74
20/12/79	0.8μmAg	65	176	180	-0.06	2.7	21	24	9.81	8.78
08/03/80	0.8μmAg	18	129	127	0.11	7.0	19	19	7.94	7.82
10/03/80	GFC	36	287	256	0.86	7.1	39	40	8.61	7.49
12/03/80	GFC	36	305	359	-1.50	10.0	53	63	6.73	6.67

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TABLE 2   Double A/E Prefilter (0.33µ)

Date	Collection Filter Type	S.W. Volume Treated Liters	POC With Bubbles µg	POC Without Bubbles µg	Difference Per Liter µg	Control Carbon Per Liter µg	PON With Bubbles µg	PON Without Bubbles µg	Molar Ratio With Bubbles C/N	Molar Ratio Without Bubbles C/N
29/06/80	GFC (1.2µ)	52	268	307	-0.75	5.9	54	70	5.81	5.12
	GFD (2.5µ)	52	201	259	-1.12	5.0	40	39	5.88	7.77
02/07/80	GFC	52	290	277	0.25	5.3	60	55	5.66	5.89
	GFD	52	241	231	0.19	4.4	35	38	8.06	7.11
03/07/80	GFC	52	293	306	-0.25	5.9	62	61	5.53	5.87
	GFD	52	211	204	0.13	3.9	39	36	6.33	6.63

TABLE 3 GFD Prefilter (2.5μ)

Date	Collection Filter Type	S.W.Volume Treated Liters	POC With Bubbles μg	POC Without Bubbles μg	Difference Per Liter μg	Control Carbon Per Liter μg	PON With Bubbles μg	PON Without Bubbles μg	Molar Ratio With Bubbles C/N	Molar Ratio Without Bubbles C/N
18/04/80	GFD	36	429	308	3.36	8.6	80	59	6.27	6.11
25/04/80	GFD	36	480	459	0.59	11.6	91	83	6.17	6.47
28/06/80	GFD	36	353	320	1.27	12.3	64	52	6.45	7.20
14/07/80	GFD	36	771	688	2.59	19.1	180	174	5.01	4.63
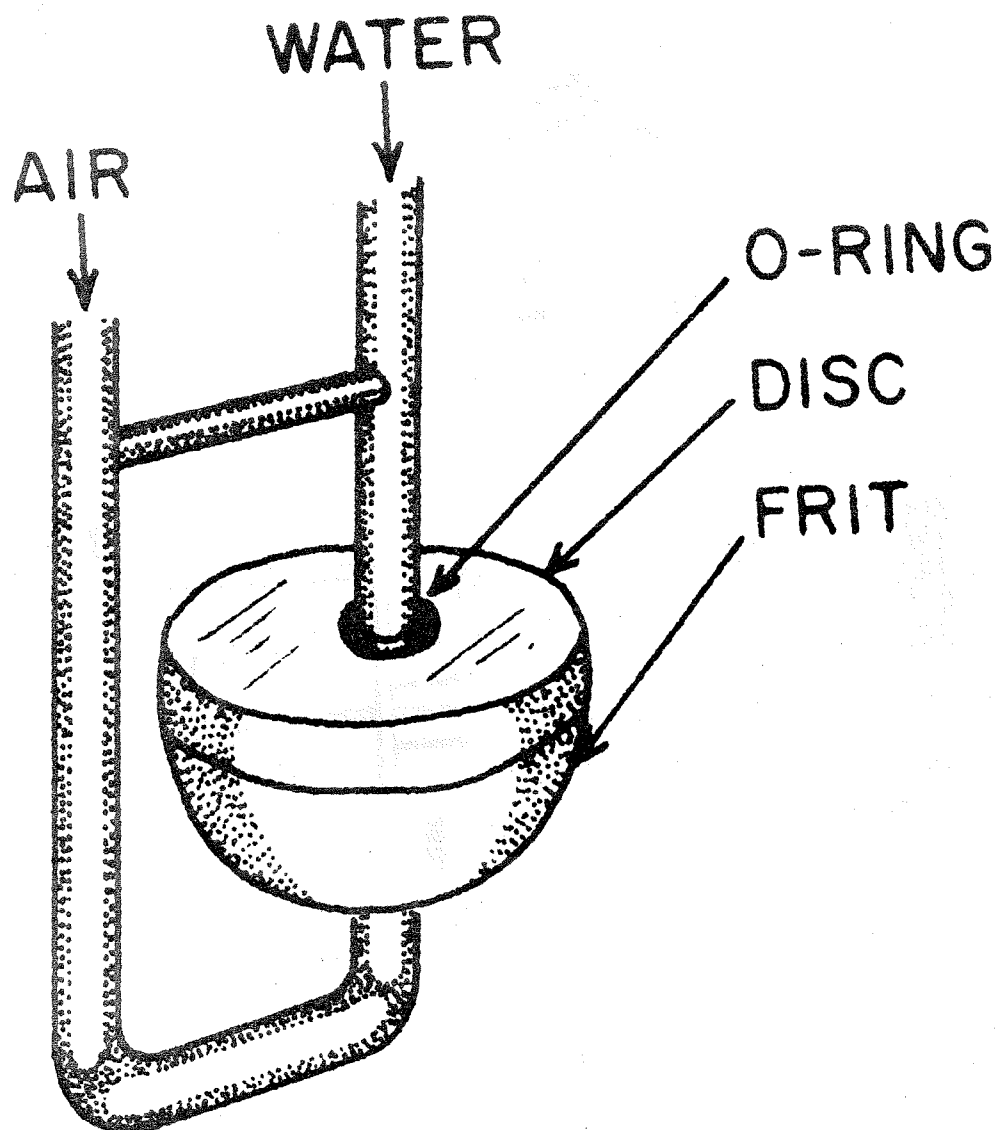
Figure 1:

   Bubble generating device consisting of a 3.2 cm. diameter frit of
4 to 5.5 μm pore size and a glass disc with o-ring and center tube.
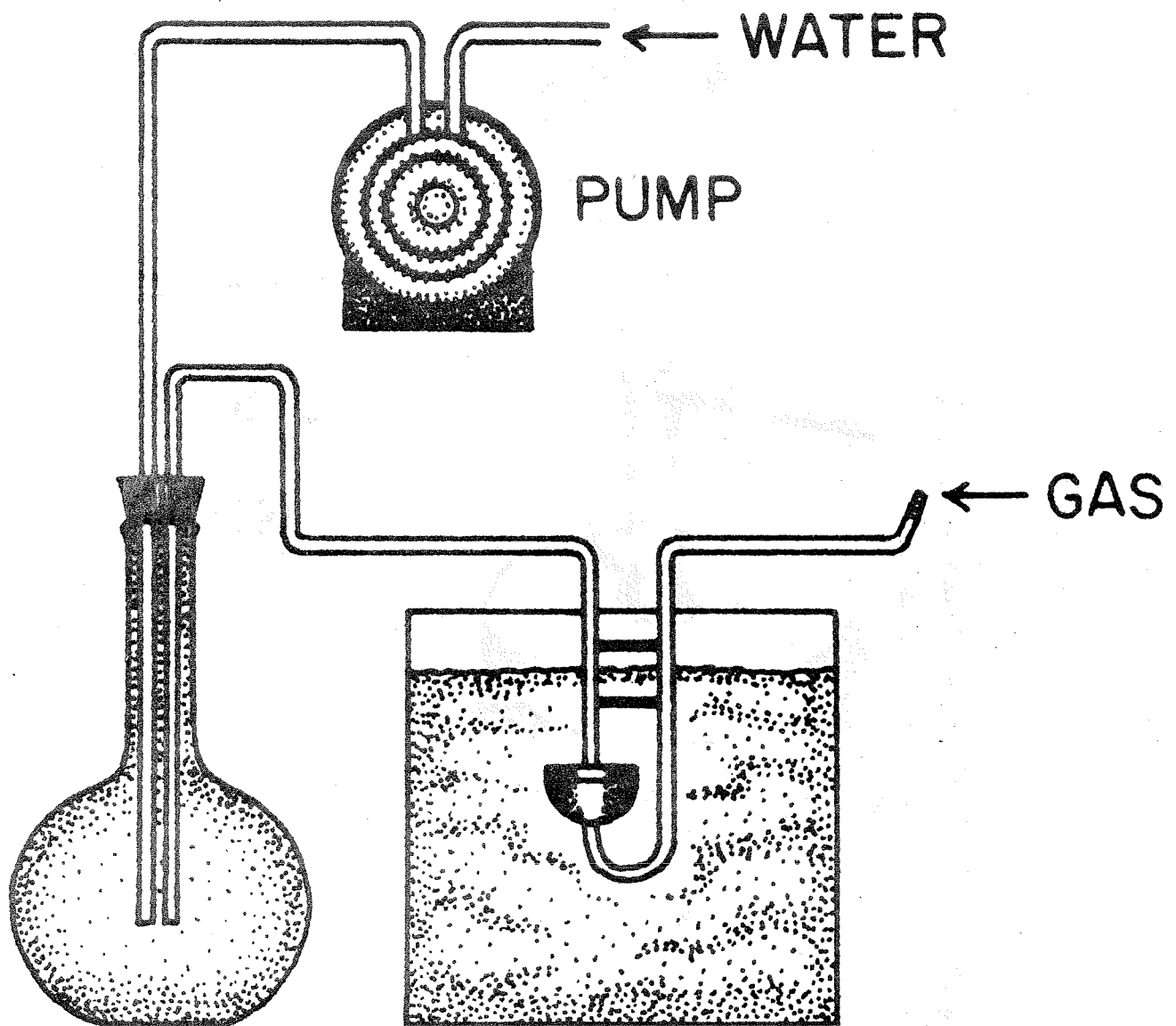
Figure 2:

Experimental apparatus for examining populations of bubbles from bubble generating device. (The plexiglass cell is not shown here).
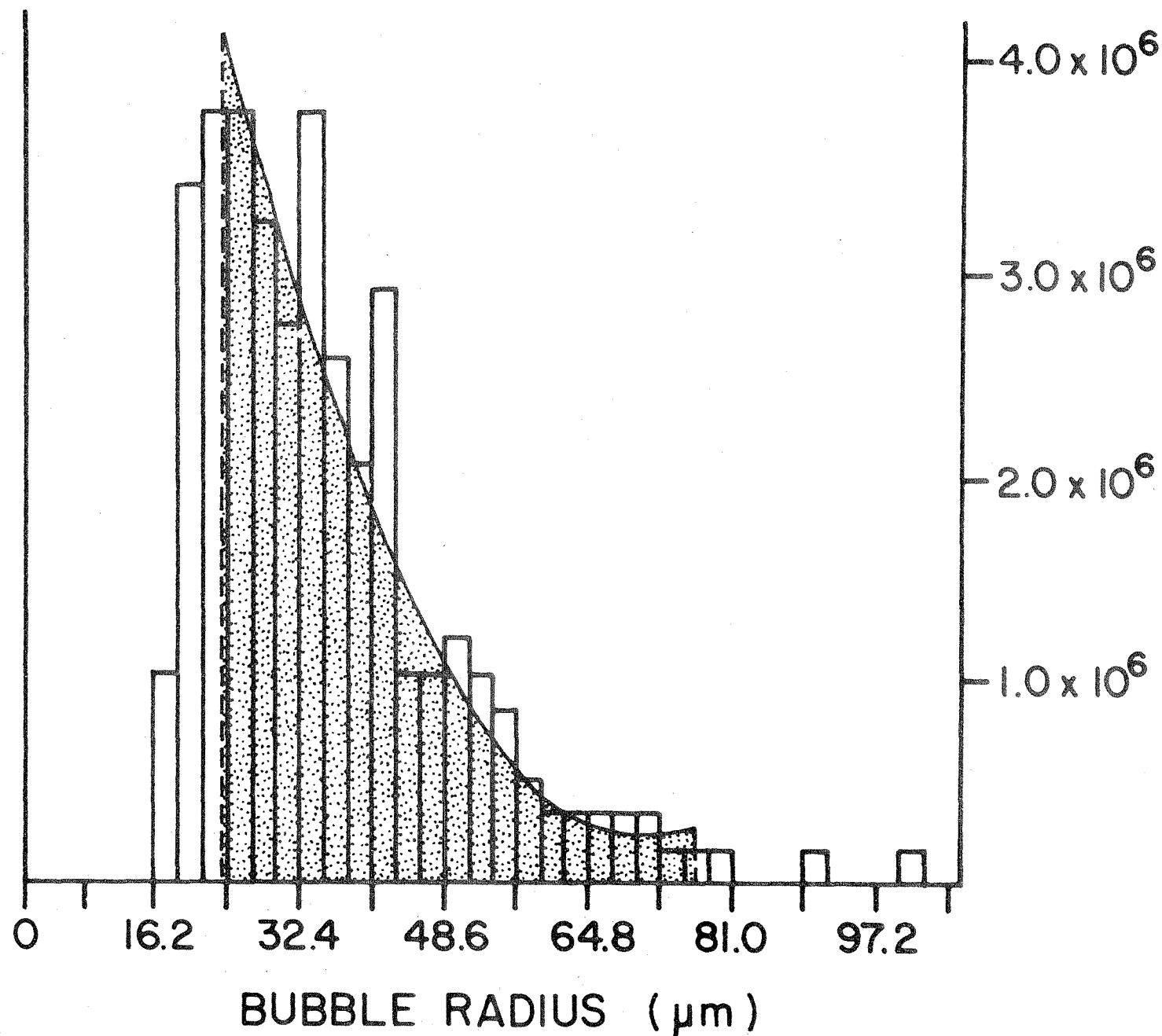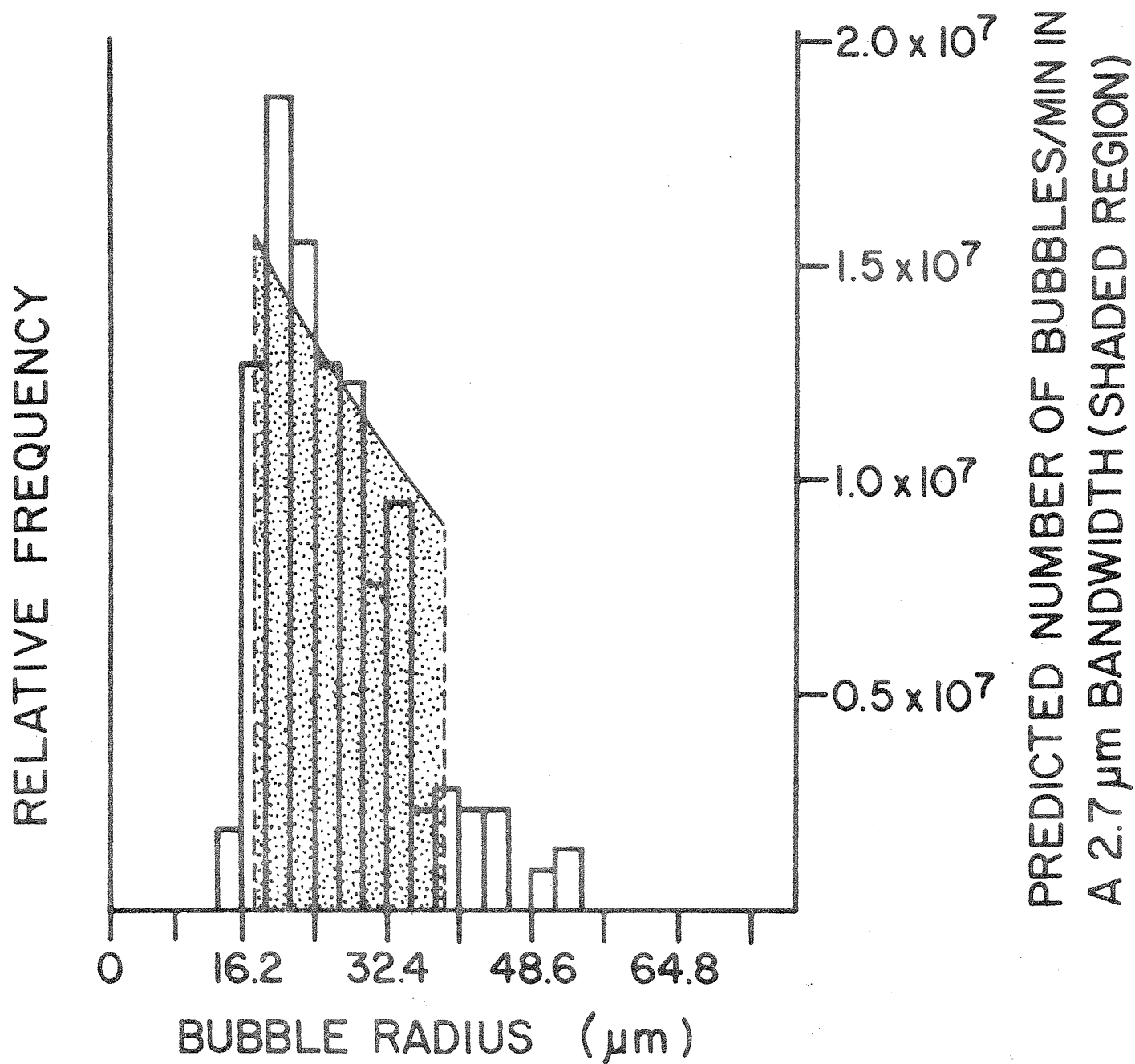
Figure 3a:

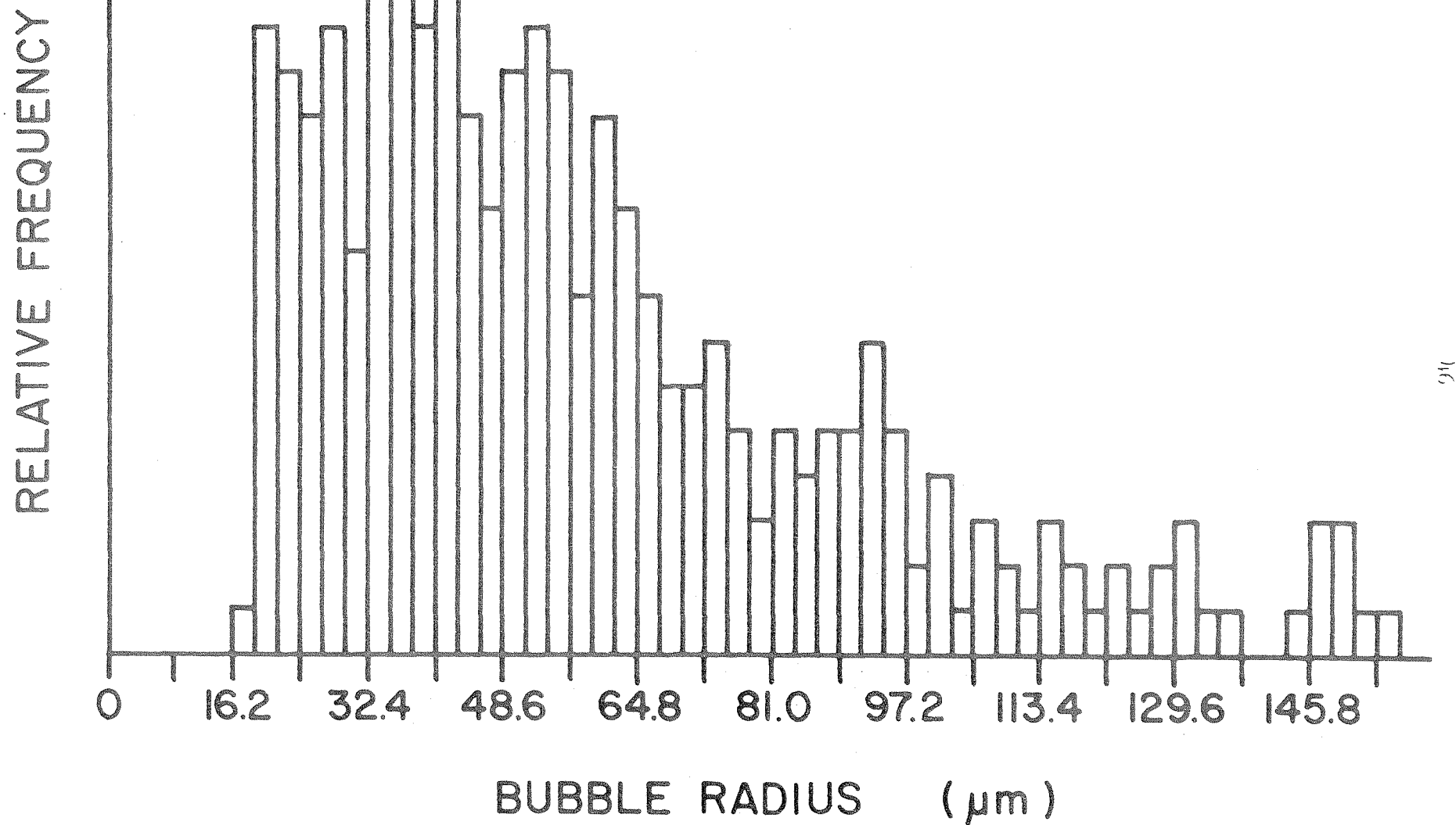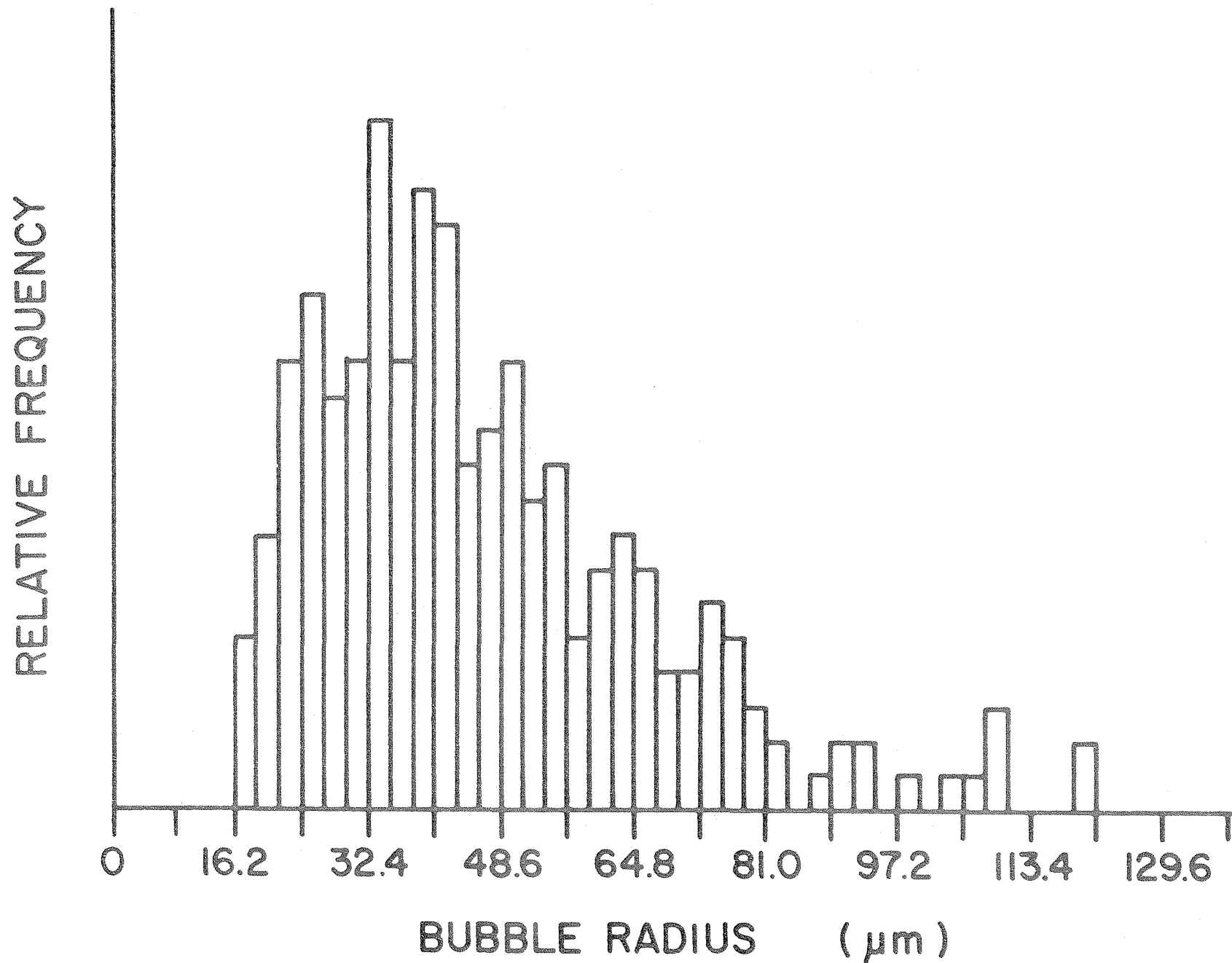Histogram of bubbles produced by the generator with a gas flow rate of 8 cm^3/min and a water flow rate of 243 cm^3/min. The scaling factors for positioning the curves predicted by the model relative to the ordinate were chosen to provide the best basis for comparison of shape with the observed distributions.
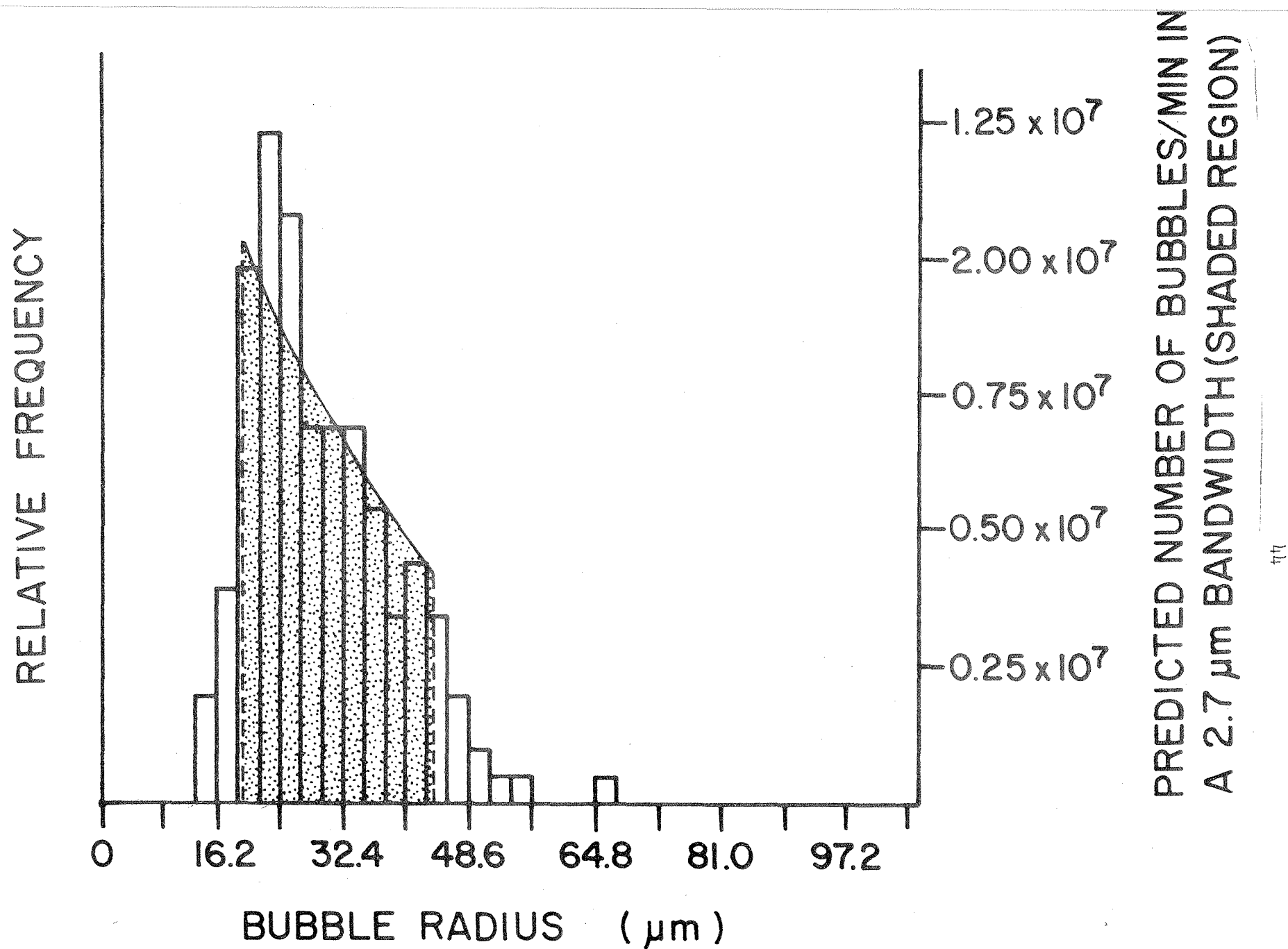
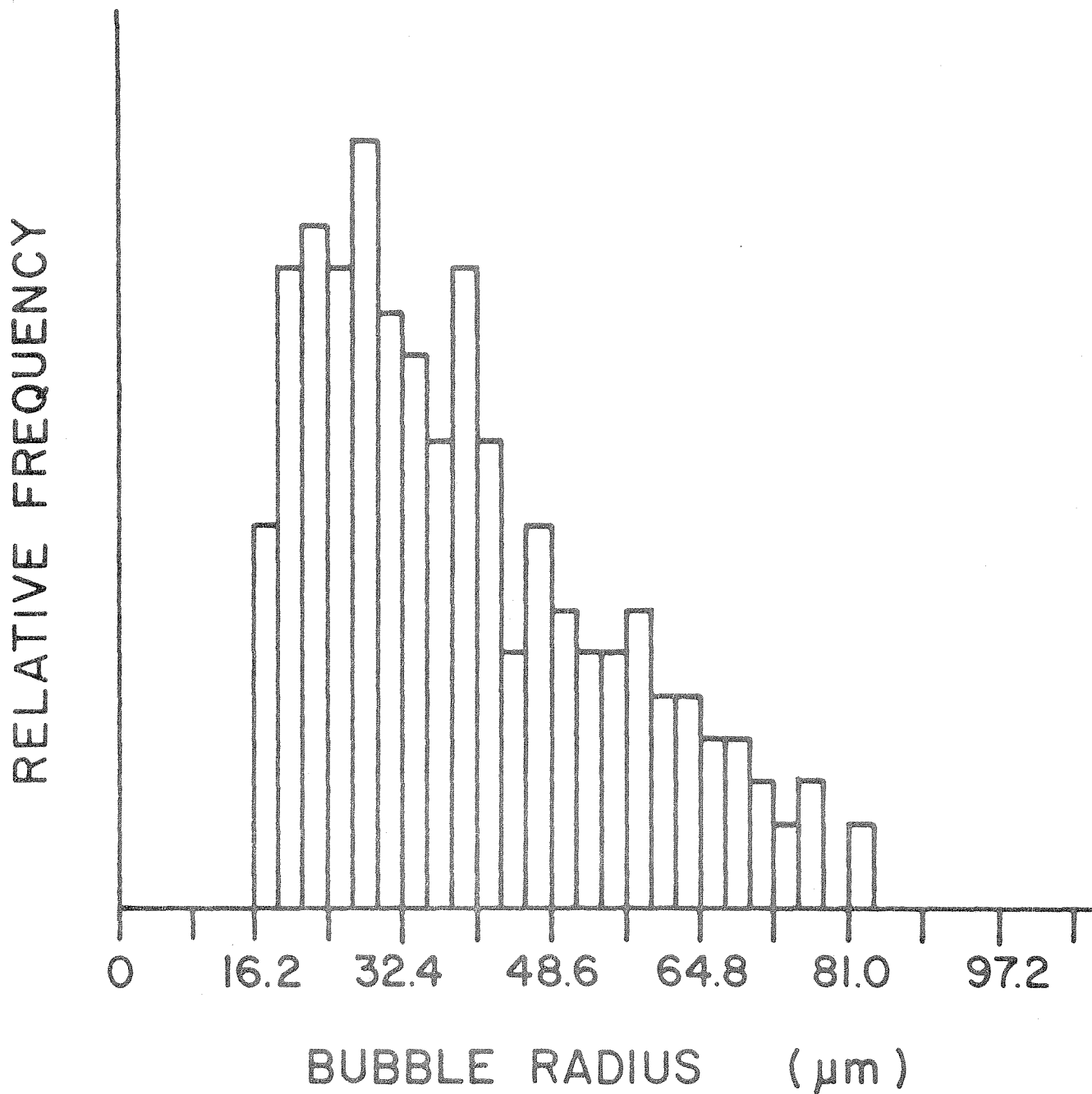Figure 3b: Histogram of bubbles produced by the generator with a gas flow rate of 8 cm^3/min and a water flow rate of 500 cm^3/min.

Figure 3c:

Histogram of bubbles produced by the generator with a gas flow rate of 8 cm³/min and a water flow rate of 776 cm³/min.

Figure 4a: Histogram of bubbles produced by the generator with a gas flow rate of 41 cm^3/min and a water flow rate of 183 cm^3/min.

Figure 4b: Histogram of bubbles produced by the generator with a gas flow rate of 41 cm^3/min and a water flow rate of 475 cm^3/min.

Figure 4c:

Histogram of bubbles produced by the generator with a gas flow rate of 41 cm³/min and a water flow rate of 776 cm³/min.
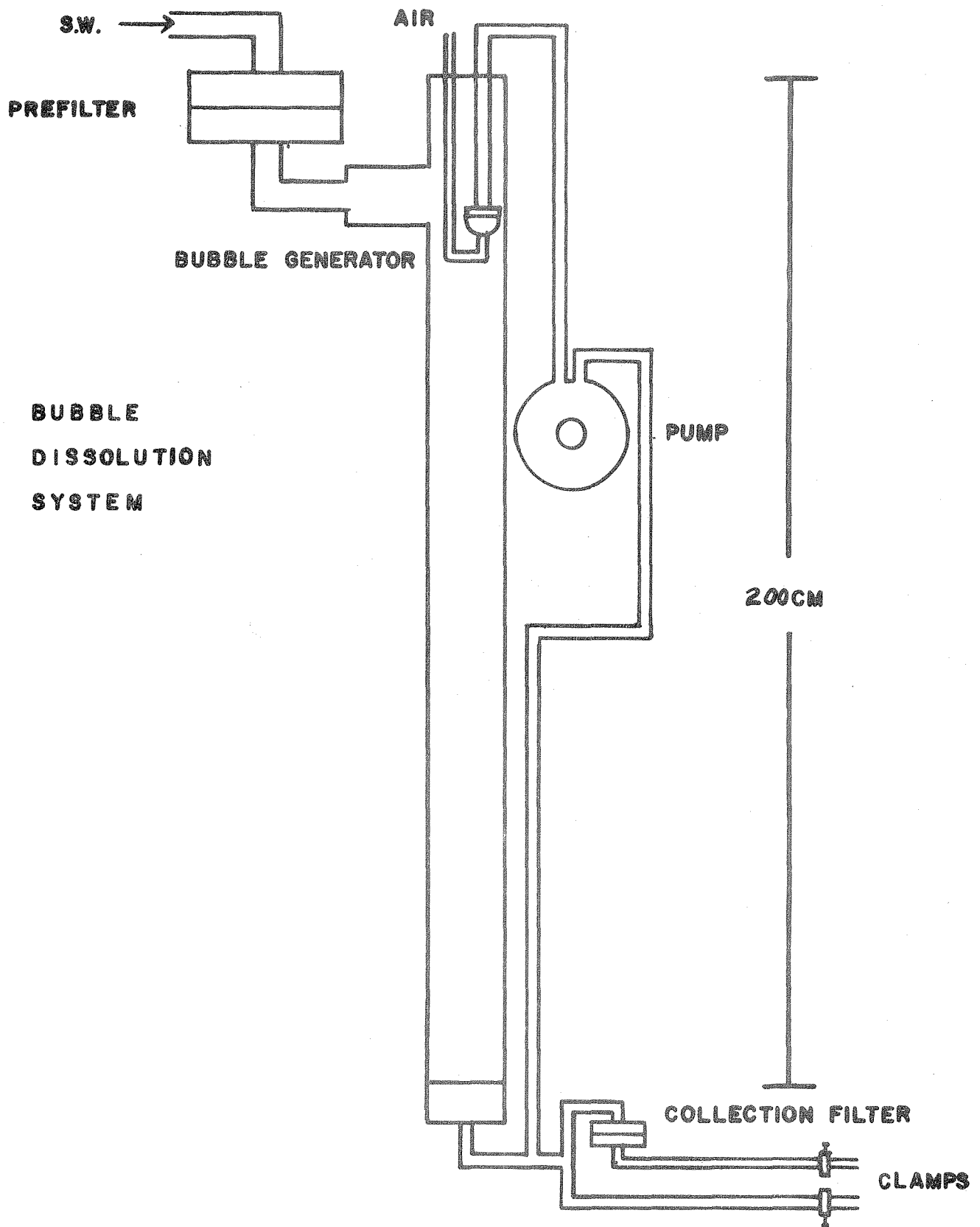
Figure 5:   Apparatus for particle generation and collection
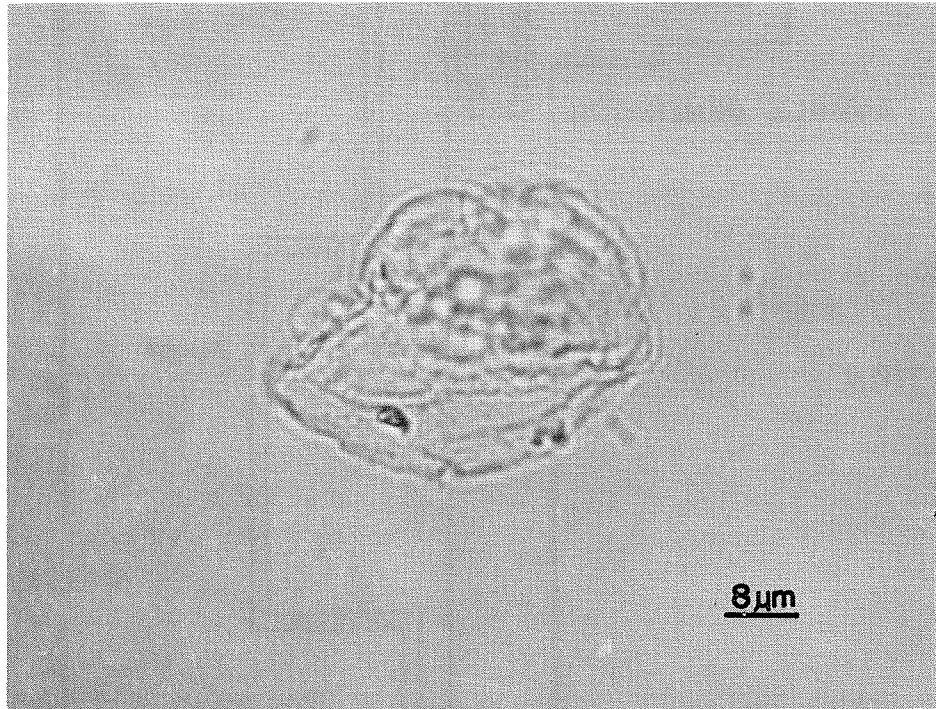
Figure 6:

Aggregate composed of a particle produced by bubble dissolution
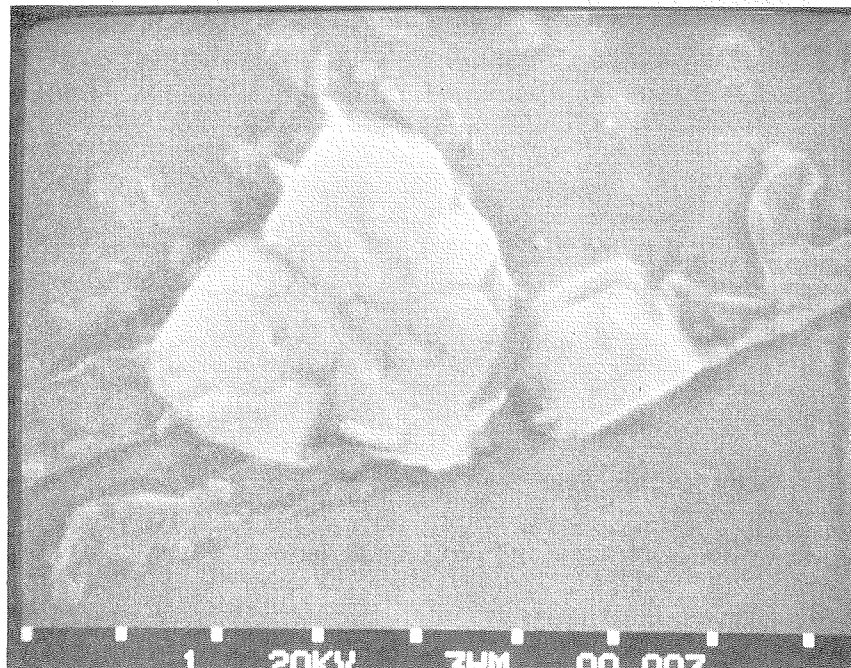(upper portion) and a flake that was scavenged by the bubble
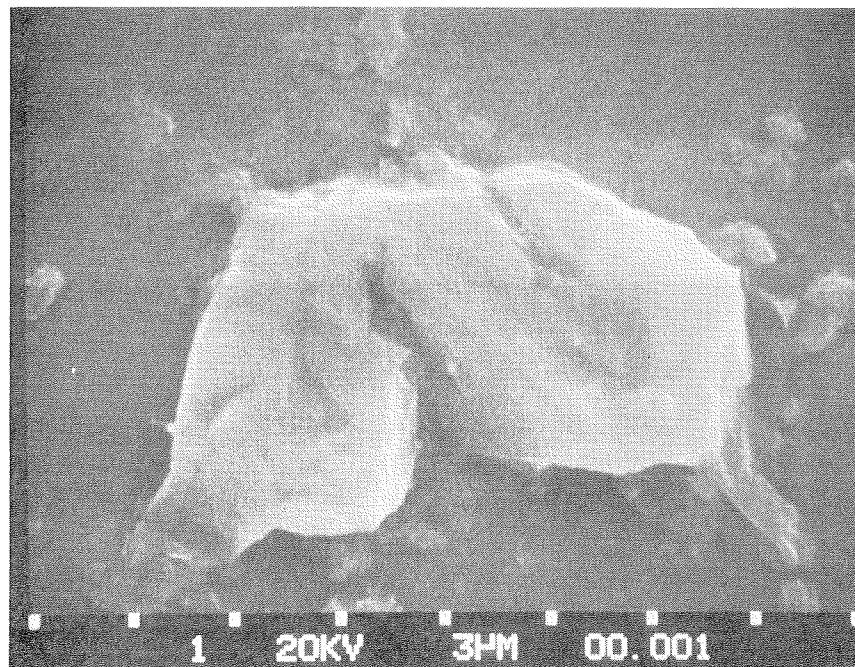before dissolution.

Figure 7:

SEM photographs of particles produced by bubble dissolution.