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Charges on Jet Drops Produced by Bursting Bubbles

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The sizes and charges of jet drops produced by bursting air bubbles are examined experimentally. The variation in charge for bubble lifetimes of up to 200 sec in NaCl and KCl solutions is reported. Jet drops refer to drops produced by a jet rising from a cavity left in a liquid surface by a bursting bubble; bubble lifetime is the interval during which a bubble is in motion between generation and rupture. With other variables held constant, drop charge shows a characteristic variation of a rise to a maximum followed by a decrease to an asymptotic value, as bubble lifetime is increased. The influences of various parameters on the characteristic behavior are discussed.

INTRODUCTION

Numerous investigations of the electric charges carried by liquid drops have been conducted, involving drops produced by a variety of methods. That some water drops produced by bursting bubbles carry net charges has long been known (1). When a bubble bursts, a jet is formed at the bottom of the cavity left in the surface. Depending upon the size of the bubble and the fluid properties, the tip of this jet may break into one or more drops before collapsing. In his initial examination of such drops, Blanchard (2) observed a dependence of drop charge upon bubble lifetime, i.e., the length of time a bubble was in motion relative to a liquid. A subsequent investigation (3) led to the conclusion that the charges on drops produced by bubbles of identical size exhibited a reproducible variation with bubble lifetime. The motivation behind Blanchard's work was interest in a possible mechanism for maintenance of the air-earth electrical current over the world ocean.[5] As a consequence, the majority of his data are for short bubble lifetimes (10 sec or less) with sea water as the test liquid, although some data for lifetimes of up to 120 sec were presented. The present experimental investigation, also employing air bubbles, used test liquids of known composition and extended the range of bubble lifetime to approximately 200 sec. Its purpose was to gain additional information about what Loeb (1) has called a newly found asymmetric spray or bubble electrification mechanism.

EXPERIMENTAL FACILITIES AND PROCEDURES

Bubbles were generated continuously by drawn glass capillary tubing nozzles located in the lower portion of the glass column shown in Fig. 1. The bubble air supply passed through both a particle filter and a CO_2 absorbent cartridge, the latter necessitated by the sensitivity of the electrical conductivity of aqueous solutions to absorbed CO_2. The mean volume of a bubble produced by any nozzle was obtained by measurement of the displacement of the meniscus in a sealed length of precision bore glass tubing as a function of the number of bubbles trapped therein.

The glass column was one element in a flow loop that also contained a pump and a glass distilling column wound with electrical heating tapes. A glass sheathed thermocouple was located immediately upstream from the column. The loop materials used (glass, Teflon, Tygon tubing, Hypalon, poly-

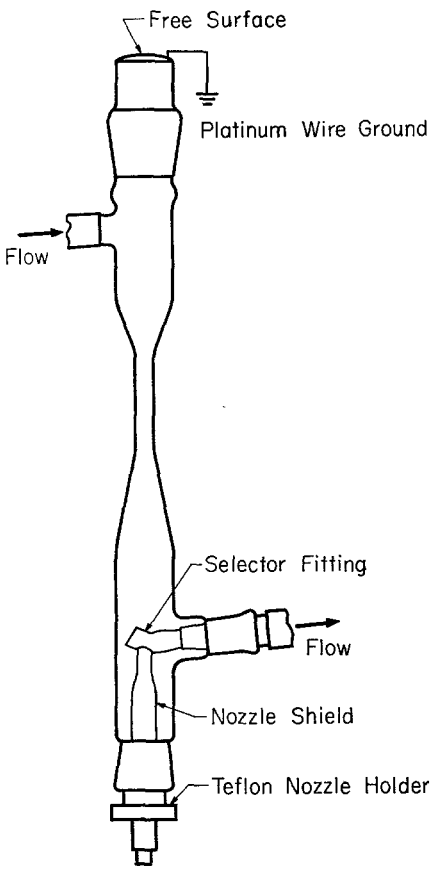

FIG. 1 Bubble aging column.

ethylene, and platinum), coupled with stringent cleaning techniques, minimized contamination of test liquids. The specific conductance of high purity water sealed in the loop for periods of up to 40 hours changed negligibly.

With no liquid flow down through the column, the stream of bubbles, produced at frequencies of 0.5/sec or less, passed through the selector fitting and proceeded upward. For any flow rate above some critical value dependent upon bubble size the bubbles were carried along with the flow and out of the column. The selector fitting and the restricted section near the middle of the column were sized so that, for any size of bubble in the range covered, the flow rate required to hold a bubble below the restricted section was well above the corresponding critical value.

During testing, the circulation rate was adjusted to a value capable of holding a bubble within or below the restricted section. When it was desired to remove a bubble from the escaping stream, the pump was shut off long enough to allow formation and travel of a single bubble into the region above the selector fitting. Resumption of pumping held the bubble for the desired period of time, after which the flow rate was reduced to a value slightly above the critical rate, allowing only the trapped bubble to rise to the free surface indicated in Fig. 1. Both the interval of no flow and the lapse of time prior to flow reduction were controlled by adjustable time delay relays.

Test solutions were prepared using both triple distilled sterile water and demineralized water. No significant difference in charge data was found. Specific conductances of each test solution were within 1% of reported values (4). Mixed solutions were stored in Pyrex serum bottles. Thorough aeration of test solutions both prior to and during testing was accomplished by means of gas dispersion tubes in the reservoir bottle and in the base of the heat exchanger, preventing the dissolution of small bubbles during aging.

During testing, liquid from the elevated reservoir flowed continuously into the loop, causing an outflow from the top of the aging column. This outflow maintained a curved meniscus at the top of the column, the value of which is discussed below.

Drop sizes and drop charge-to-mass ratios were measured separately. The size of drop produced by some specified combination of test liquid, temperature, and bubble size was determined by trapping successive drops in a liquid matrix of mineral oil and vaseline, as described by May (5). Over the 200 sec range examined, bubble lifetime was not found to influence drop size. At 23°C the drops were quite uniform in size, with over 95% of the drops having diameters within $\pm 2\%$ of their mean values.

Drop charge-to-mass ratios were determined by a method reminiscent of the Milliken oil drop experiments, in that it involved the suspension of a drop between horizontal, parallel plates by means of an electrical field. To understand the present

application of this method it is necessary to review some aspects of jet drop behavior. When produced, jet drops are ejected in a direction normal to the surface. The height obtained by the topmost drop is repeated quite closely by successive bubbles of the same size, over the range of bubble sizes of present interest. The curved meniscus maintained at the top of the aging column caused any bubble which did not burst immediately upon reaching the surface to migrate toward a fixed point on the meniscus. This resulted in drops being ejected along essentially the same vertical path in the majority of cases. Such behavior, coupled with the reproducibility of ejection heights, made possible the method utilized. This is substantially the same method used by Blanchard.

The parallel plate assembly consisted of two 12 cm diameter brass disks, each pierced by a 4 mm diameter hole at center. The plate spacing of 2 cm was maintained by suspending the upper plate from the top of an enclosure, upon the base of which the lower plate rested. In use, the entire enclosure was positioned horizontally so that the drop path passed through the hole in the lower plate, and vertically so that the topmost drop reached its maximum height between the plates.

With the use of the same delay settings in the pump control circuit, bubbles aged for essentially the same period of time were allowed to reach the surface and burst. Drop motion in the electrical field was observed with a long working distance 6X microscope containing a reticule scale. In order to minimize the effect of evaporation upon drop mass, a jet of dry air was used to blow out from between the plates any drop which was not balanced within a few seconds. Normally, no more than three or four drops would have to be produced before the field strength required to suspend a drop was found. When this procedure was followed it was noted that renewed drop motion, attributed to evaporation, did not begin for several seconds.

For infinite parallel plates without holes the potential gradient is everywhere equal to the ratio of potential difference to plate spacing. Departures from this value for the pair of disks utilized were determined by numerical solutions of the governing Laplace equation. In the center 60% of the plate gap the actual field strength was found to lie between 1.00 and 0.98 times the infinite plate value. This range was incorporated as an uncertainty, along with uncertainties in applied voltage and plate gap, into the subsequent presentation of results.

Additional details concerning material in this section are available elsewhere (6).

RESULTS AND CONCLUSIONS

At 23°C, over a bubble diameter range of from 200μ to 600μ, for demineralized water, 5000 ppm NaCl, 50,000 ppm NaCl, and 5000 ppm KCl solutions, the topmost drop diameter was found to be equal to 0.0941 ± 0.002 times the bubble diameter. The lack of dependence upon solution is attributed to the comparatively small differences in surface tension, density, and viscosity among the four solutions. The values used by Blanchard for sea water yield a coefficient of 0.096.

Figure 2 shows the data obtained for one test solution, and Table I summarizes the results of a number of cases. No contribution due to variation in drop size has been included in the uncertainties. Bubble lifetime refers only to the interval during which a bubble was in motion with respect to the liquid, the period between release from a nozzle and arrival at the surface. Blanchard, on the basis of extensive testing over short bubble lifetimes, concluded that the time required for bubble growth at a nozzle, as well as the surface lifetime, did not influence drop charges. Nothing noted during the present investigations indicated otherwise. Since the bubble lifetime of each bubble generated during testing was measured individually, the width of the uncertainty line is a reasonable measure of the uncertainties in measurement.

All of the cases studied in the present investigation yielded the same characteristic variation in drop charge with increasing bubble lifetime. As shown in Fig. 2, drop charge passed through a maximum followed by a decrease to an asymptotic value. Blanchard (3) reported the results of three sea-

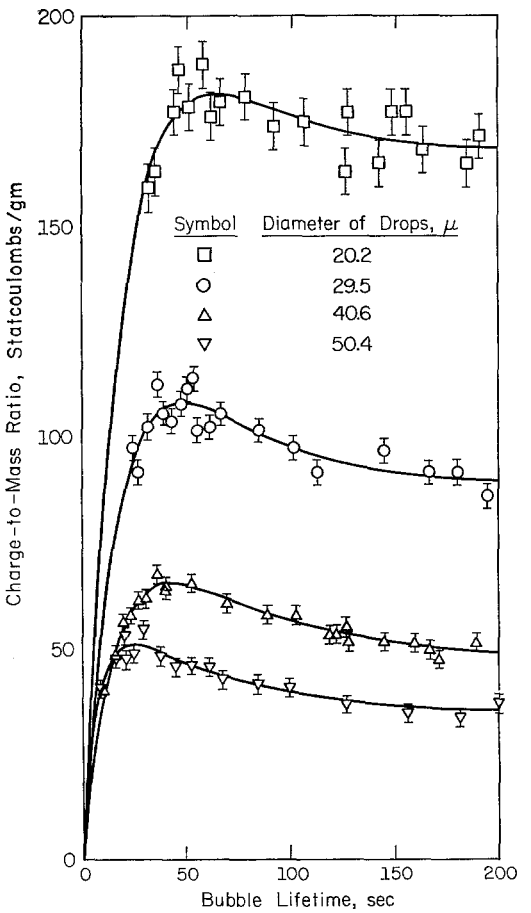

FIG. 2. Charge-to-mass ratios for jet drops from a 5000 ppm NaCl solution at 23°C.

water tests for bubble lifetimes up to 120 sec. In two of these three, cases 10 and 11 in Table I, the same characteristic behavior was found. The third case is discussed below in connection with certain trends observed in the data.

Since one of the objectives of this investigation was to examine liquids of known composition, the minimum salt concentration tested was 5,000 ppm, with the aim of masking impurities introduced by the water and by the loop itself. The satisfactory agreement between cases 1 and 2 in Table I supports the conclusion that the results presented here are the consequence of the materials added to the solvent, rather than of contaminating factors. The two sets of data were collected approximately six weeks apart, during which interval certain of the loop components were replaced; in addition, the loop was cleaned several times.

Values in Table I were obtained by least squares fits with an equation of the form

$$Q = a_1(e^{b_1 t} - 1) + a_2(e^{b_2 t} - 1), \quad [1]$$

where Q is the charge-to-mass ratio and t is the bubble lifetime.

This form is suggested both by the data and by the work of Alty (7) concerning the time variation of the net charge on bubbles in comparatively pure water. This equation agreed with his data when the constants were adjusted in an empirical manner. Although the model from which this equation was obtained has been questioned (8) and only some of Alty's observations have been verified by other investigations (9), the dependence of drop charge upon bubble lifetime indeed suggests that the origin of drop charge may lie in some phenomenon similar to that suggested by Alty.

Some general trends, most easily seen in Table I, have been noted. For all solutions tested, increasing drop size showed a decreasing charge-to-mass ratio. To avoid the possibility of arcing between the plates, the maximum field strength employed was 10,000 volts/cm. In such a field, jet drops of 70 μ in diameter and larger could not be held, although their fall back down through the field was retarded. Thus, over a bubble lifetime range from 7 to 150 sec, drops of these sizes, produced from a 5,000 ppm NaCl solution, possessed charge-to-mass ratios of less than 20 statcoulombs/gm. The inability of such a field to measurably alter the ejection height of a 200 μ drop produced from the same solution indicates a charge-to-mass ratio of less than 70 statcoulombs/gm for such a drop.

Blanchard reported charge values for a solution of known composition (50,000 ppm NaCl) for times of 10 sec or less, but his measurements for greater times were restricted to filtered sea water of unknown composition. From such measurements as are available for comparison, results of the present investigation lie somewhere between 5% and 10% below these given previously. From cases 3 and 6 in Table I it would ap-

TABLE I

SUMMARY OF JET DROP CHARGE INFORMATION

Test solution	Case no.	d_b[a]	d_d[a]	Q_{max}[b]	Time of Q_{max} (sec)	$\frac{Q_{asy}}{Q_{max}}$[b]
5,000 ppm NaCl in distilled water	1	536	51.4	51.0	19	0.48
5,000 ppm NaCl in demin. water	2	531	50.4	51.1	24	0.66
	3	423	40.6	65.2	42	0.72
	4	320	29.5	108.0	48	0.82
	5	217	20.2	181.2	63	0.92
50,000 ppm NaCl in demin. water	6	419	39.0	65.2	42	0.62
	7	362	34.7	81.6	50	0.77
5,000 ppm KCl in demin. water	8	430	39.6	42.6	51	0.67
	9	353	32.2	53.6	69	0.74
Seawater[c]	10	379[d]	36.4	53.3	20	0.59
	11	321[d]	30.8	77.7	24	0.75

[a] Expressed in microns, d_b is the average bubble diameter and d_d is the average drop diameter.

[b] Q_{max} refers to the maximum charge-to-mass ratio, while Q_{asy} is the long-time value; Q_{max} is expressed in statcoulombs/gm.

[c] Blanchard, D. C., "Progress in Oceanography," Vol. I, p. 71. Macmillan, New York, 1963.

[d] Estimated value.

pear that a change in NaCl concentration is of little importance. Limited data for a 25,000 ppm NaCl solution also showed no difference. Iribarne and Mason (10) have studied the effects of solute concentration upon the average charge on drops produced by bursting bubbles. In this investigation bubbles were generated at a fixed depth below a free surface, so that bubbles of a given size were all aged for the same length of time. Since their measurement technique determined the average charge carried by all drops produced, and not just that of the topmost drop, no meaningful comparisons of charge magnitude can be made. The data presented for average drop charge as a function of NaCl concentration, however, exhibited an insensitivity to concentration over the range considered in the present investigation.

The time at which the maximum charging occurred decreased with increasing drop size, as did the ratio of the long-time value of Q to its maximum. The actual values of the above time and this ratio are not well defined, with the latter being particularly sensitive to the removal of even a single long-time point. It is felt, however, that sufficient evidence exists to establish the trends. For a 20.2 μ diameter drop, Blanchard's results show what appears to be a relatively smooth approach to a maximum Q with no subsequent dropoff, reinforcing the conclusions of the present investigation.

Cases 6–9 indicate no fundamentally different behavior between NaCl and KCL solutions. For drops of nearly the same size, the latter solution yielded charges approximately 35% lower, whereas maximum charges occurred at somewhat greater times. Cases 10 and 11 also display no anomalous behavior.

Tests at significantly higher temperatures indicated a deterioration in the uniformity of drop size. At 50°C a range of $\pm 10\%$ was required to include 95% of the measured drop diameters, compared with the $\pm 2\%$ range at 23°C; at least a portion of this increase was due to variation in bubble size. In addition, nozzle stoppage increased significantly. The information collected indicated nothing beyond a decrease in charge-to-mass ratios of approximately 10%.

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