

Systematic Charging of Water Droplets Produced by Break-up of Liquid Jets and Filaments

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The paper describes three separate experiments to elucidate the mechanisms responsible for the charging of droplets that result from the controlled break-up of small jets, threads and filaments of liquid. The results of these experiments are consistent with those of Iribarne and Mason on bursting bubbles and reveal that, in the absence of external fields and contact potentials, the droplets acquire systematic and consistent charges, the magnitude of which depend largely on the time for disruption and the concentration of the solution but not on the ionic species. The results find a reasonable quantitative explanation in terms of the two mechanisms proposed by Iribarne and Mason, one acting during formation of the jet and the other during the break-up stage, but both producing charge separation by the disruption of the electrical double layer that exists at the surface of a polar liquid.

Droplets produced during the atomization, splashing, disintegration and bubbling of liquid acquire an electric charge. This is of interest in many fields, e.g., the electrification of precipitating clouds, the production and stability of aerosols, the static charging of aeroplanes, the atomization of liquid fuels, etc. Random charging of droplets may arise during atomization because of local statistical fluctuations in the concentrations of positive and negative ions in the bulk liquid, but the charges on small droplets tend to be small and the aerosol as a whole to be neutral. Much larger and systematic charging may be induced either by external electric fields (as in thunderstorms), or by contact potentials between the liquid and the solid surfaces involved in the disintegration process. In polar liquids, where preferential orientation of molecules in the surface layers gives rise to an electrical double layer, break-up of the liquid into drops may involve rupture of this double layer and a separation of electric charge.

Such a process is likely to be particularly important in the atomization of water and aqueous solutions and has been invoked by Iribarne and Mason¹ (1967) to explain the electrification of droplets ejected during the bursting of gas bubbles in these media. Bursting of the bubble is followed by the formation of a tiny jet of water rising from the floor of the bubble cavity and this breaks up to form one or more drops. For pure water and very dilute solutions the droplets acquire a negative charge, the magnitude of which decreases rapidly with increasing concentration and becomes vanishingly small at concentrations greater than about 10^{-4} M. For more concentrated solutions the drops carry a small positive charge. The negative charging of the drops is explained by the dragging-up of a thin film of liquid from the inner surface of the bubble cavity which results in rupture of the electrical double layer at the air/water interface and a transfer of negative charge to the rising jet. Iribarne and Mason were able to account quantitatively for the observed negative

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charges by showing that, for a film of thickness δ , the surface density, σ , of the separated charge to be given by

$$\begin{aligned}\sigma &= -2\left(\frac{DRT}{500\pi}\right)^{\frac{1}{2}} C^{\frac{1}{2}} \exp\left[-\frac{1}{2} - \frac{10^8 C^{\frac{1}{2}} \delta}{3}\right] \\ &= -4.2 \times 10^4 C^{\frac{1}{2}} \exp\left[-(10^8 C^{\frac{1}{2}} \delta/3)\right],\end{aligned}\quad (1)$$

where D is the dielectric constant of water, C the molar concentration of the solution, T the absolute temperature and R the gas constant. This equation accounts well for the observed decrease of negative charging with increasing concentration and for the charge becoming undetectable at concentrations of about 10^{-4} M, if δ is of the order 1μ . The positive charging of the drops at high concentrations was attributed to the separation of charge during the break-up of an initially uncharged varicose jet, when the surface tension forces cause a differential flow of liquid from the contracting neck into the swelling regions which form the drops. With the negatively charged outer layers relatively immobile, the faster inner core of liquid carries a net positive space charge into the growing drops. Iribarne and Mason calculated this charging current to be roughly independent of the concentration of the solution and to be given by

$$i = 1.2 \times 10^{-7} e^{-\frac{1}{2}} \left(\frac{DRT}{500\pi}\right)^{\frac{1}{2}} \frac{\gamma b^3 (a-b)}{\eta a^3 (a+b)}, \quad (2)$$

where γ and η are respectively the surface tension and viscosity of the liquid, a the radius of the undisturbed cylindrical jet and b the amplitude of the sinusoidal disturbance causing the instability. Since during the formation and disintegration of the jet b/a varies from 0 to 1, the average current is

$$\bar{i} \simeq 1.0 \text{ e.s.u.},$$

which indicates an upper limit of order 10^{-5} e.s.u. for the charges on droplets produced by small jets disintegrating in times of order 10^{-5} sec.

DESIGN OF NEW EXPERIMENTS

In order to study in more detail the charging produced by the break-up of small jets, threads, and filaments of liquid, with control over their size and also the geometry, time scale, and mode of disintegration, we hoped to make use of the vibrating-needle device described by Mason, Jayaratne and Woods² which can be made to produce unstable threads of liquid that break up in a controllable and reproducible manner. Certainly the liquid filaments expelled from the tip of the needle can be made to produce drops of very uniform size and carrying highly reproducible charges. However, it was soon discovered that the magnitude and sign of these charges was largely determined by the existence of contact potentials of several 100 mV between the needle and the water, and although by neutralizing these with suitable counter potentials applied to the needle the charges separated during break-up of the liquid could be detected, priority was given to other techniques in which these complications were absent. The experiments now to be described were designed to measure the separation of charge accompanying the break-up of small volumes of water and aqueous solutions in the absence of external electric fields and to discriminate between the two mechanisms postulated by Iribarne and Mason, one being active during the formation of the jet and the other during its rupture.



FIG. 1.—A stream of water drops, each $150\ \mu$ in radius, impacts on a plane water surface to produce a series of small jets that break up to form a secondary stream of smaller droplets.

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CHARGING ASSOCIATED WITH PARTIAL COALESCENCE OF DROPS WITH A PLANE LIQUID SURFACE

EXPERIMENTAL PROCEDURE AND RESULTS

When a stream of uniform-sized drops of radius about $150\ \mu$ impinges on a plane, clean, liquid surface, within a narrow range of impact angles and velocities, partial coalescence takes place and uniform-sized secondary drops, smaller than the incident drops, are ejected from the surface. In this process,³ the secondary drops are pinched off from small jets of liquid rising from the surface as shown in fig. 1. Measurements of the charges on these secondary drops provides a convenient method of investigating the electrification associated with the disruption of liquid jets in the absence of solid surfaces.

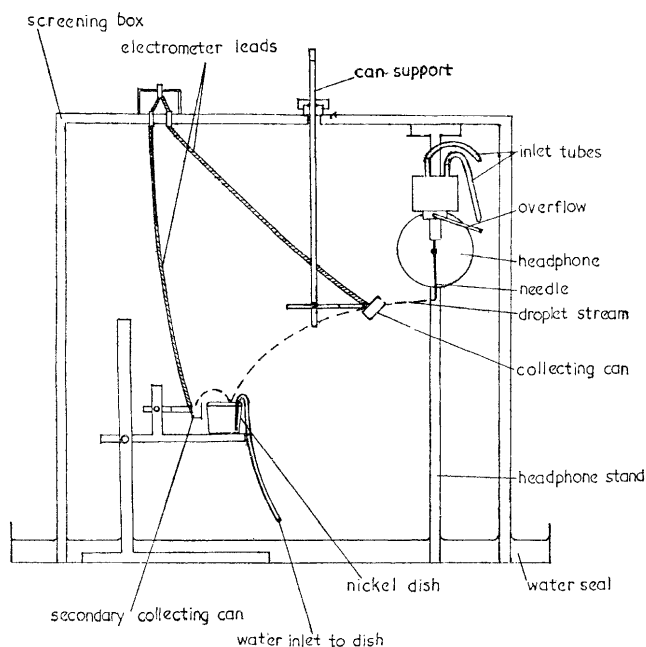


FIG. 2.—The apparatus used to measure the charges on the incident and secondary droplet streams shown in fig. 1.

The apparatus is shown diagrammatically in fig. 2. Streams of drops, of very uniform size, are produced by the modified version of the vibrating-capillary device described by Mason, Jayaratne and Woods.² Water is forced at a constant rate through a hypodermic needle attached to the centre of the steel diaphragm of an earphone. By adjusting the frequency of the audio-frequency oscillator connected to the energizing coil of the earphone electromagnet, the needle may be made to vibrate in a stable resonant mode so that the oscillating tip flings off a stream of drops of identical size. The size and rate of production of the drops are governed by the flow rate of the liquid, the needle diameter, its resonant frequency and its amplitude of vibration. In the present experimental arrangement the needle is fed with either very pure water, or with aqueous solutions whose concentration can be controlled by mixing pure water and concentrated solution in a small chamber and monitored by measuring their conductivity in a small cell containing platinum electrodes. All the apparatus and connecting tubes are carefully cleaned, and flushed for long periods with clean water both before and after assembly; the purest water supplied to the needle has a conductivity of $6 \times 10^{-8}\ \Omega^{-1}\text{ cm}^{-1}$. The vibrating needle is adjusted to give a stable

stream of drops of radius about $150\ \mu$, the charges on the drops being neutralized by applying a small potential to the needle. The droplet-stream is collected in a small insulated metal can connected to a sensitive electrometer and the potential of the needle adjusted until the electrometer records zero current; in this way the average charge per drop can be reduced to less than 10^{-8} e.s.u. The uncharged drops are then directed on to the plane surface of a liquid contained in a small earthed nickel dish, the height of which is adjusted until partial coalescence occurs, the secondary, ejected droplets being collected in another insulated can also connected to a vibrating-reed electrometer. The sizes, the angles and velocities of impact of the drops can be determined from photographs such as fig. 1, and the average charge on a secondary drop determined by dividing the current recorded by the electrometer by the frequency of droplet production, i.e., the vibration frequency of the needle. The apparatus is carefully screened against stray electric fields and placed in a sealed, earthed metal box filled with clean, filtered air.

Using highly purified water and solutions of sodium chloride, thorium nitrate and ammonium chloride, the charges on the secondary drops were determined as a function of the solute concentration. The impacting drops were allowed to land either on a stationary surface of pure water from which the fragments of impacting drops were removed periodically with a directed stream of nitrogen, or upon a continually renewed (slightly overflowing) surface of a solution having the same concentration as that composing the drops. A few experiments were made in which the solute concentrations of the drops remained

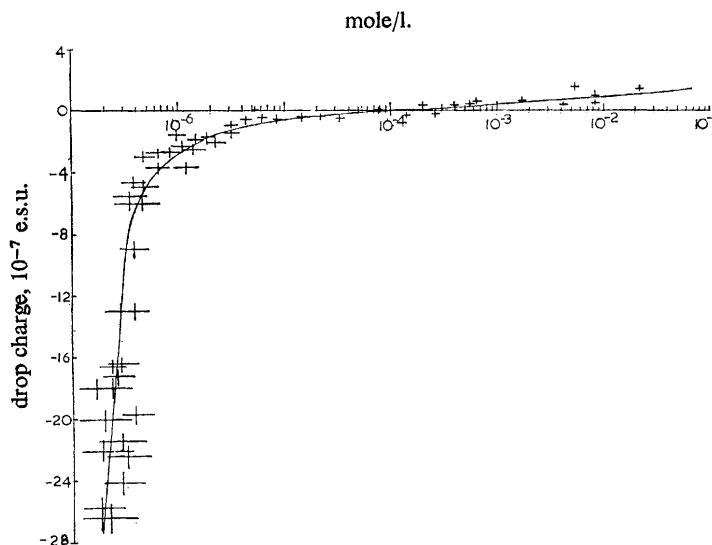


FIG. 3.—The charges carried by secondary droplets of radius $65\ \mu$ produced by the impact of $180\ \mu$ droplets of sodium chloride solution on a plane surface of the same solution as a function of the solute concentration.

constant while that of the liquid target was changed. Fig. 3 shows the results obtained for drops of sodium chloride solution of radius $180\ \mu$, impacting on a surface of the same solute concentration at an angle of 50° to the horizontal, with a velocity of 140 cm sec^{-1} , and producing secondary drops of radius $65\ \mu$ ejected at 35 cm sec^{-1} . With pure water, the secondary droplets acquire negative charges of about 3×10^{-6} e.s.u.; but with more concentrated solutions the charges decrease, become zero at concentrations of about 10^{-4} M, and thereafter reverse so that the droplets acquire small positive charges of order 10^{-7} e.s.u., which increase only slowly as the concentration is raised towards 0.1 M. The fact that the experimental curves for solutions of thorium nitrate and ammonium chloride are similar in both shape and actual magnitude to those for sodium chloride suggests that the electrification is not sensitive to the ionic species. Varying the concentration of the solution forming

the target has little effect compared with altering that of the incident drops. Moreover, the difference obtained with stationary and moving surfaces are small except for drops of very dilute solutions ($C < 10^{-6}$ M), which acquire larger negative charges from a moving surface.

THE CHARGING MECHANISM

As with bursting bubbles, the charging of the ejected drops is likely to be closely related to their origin and motion, which can be inferred from detailed examination of sequences of photographs such as those of fig. 1. The incident drop, on hitting the water surface, flattens, forms a crater beneath it, and oscillates between a flattened and elongated shape as indicated in fig. 4. The air film between the drop and the

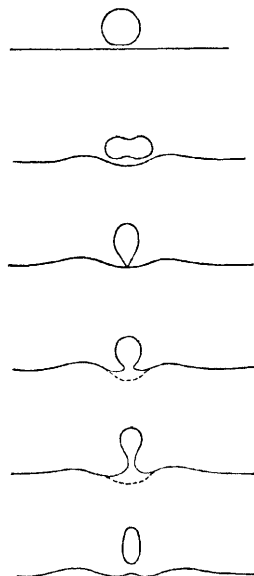


FIG. 4.—Sketches to illustrate successive stages in the production of secondary droplets by the impact and partial coalescence of a larger droplet on a plane liquid surface.

water surfaces thins, finally ruptures, and the liquid composing the drop begins to drain through the expanding bridge into the bulk liquid beneath. Completion of this process would lead to complete coalescence of the drop but, in the transition regime of partial coalescence, the surface tension in attempting to reduce the overall surface area, forces the residue of the drop upwards to form a tiny jet that drags the surface of the underlying liquid up with it. The depressed liquid surface in returning to its equilibrium position overshoots and, on its second downward journey, pulls out the tiny jet which breaks up to form a secondary drop that appears to bounce off the surface. Tests with chemical tracers show that the secondary drops are formed almost entirely from the liquid composing the incident drops, little, if any, of the liquid from the target surface being carried up into the jet. This is consistent with the finding that the charge on the secondary drops depends only on the solute content of the incident drops and not on that of the target. The manner in which the charges on the rebounding drops varies with the concentration of the solution composing the incident drops is similar to that reported by Iribarne and Mason¹ for the drops ejected from the small jets of bursting bubbles. However, the magnitudes of the charges in the present experiments are much smaller than those measured by these workers. Whereas drops of radius $10\ \mu$ ejected from nitrogen bubbles of radius $100\ \mu$ bursting in pure water carried charges of order 10^{-3} e.s.u.,

fig. 3 shows maximum charges of only about 10^{-6} e.s.u. According to eqn. (1), the net charge separated by shearing of the electrical double layer decreases rapidly with increasing thickness δ of the film. Iribarne and Mason estimated that, in their experiments, the thickness of the film dragged up from the inner surface of the bubble cavity by the rising jet was about 1/30th the radius of the ejected drop. If, in the present experiments, we assume the film to be collected over an area equal to the cross-section of the incident drop, the negative charge on the ejected drop would be given by

$$q = \pi R^2 \sigma = 4.2\pi 10^4 R^2 C^{\frac{1}{2}} \exp(-10^8 C^{\frac{1}{2}} \delta/3). \quad (3)$$

The experimental results, such as those of fig. 3, give for $R = 150 \mu$ and $C = 10^{-6}$ M, $q = 4 \times 10^{-7}$ e.s.u., and substitution of these values in eqn. (3) gives $\delta = 3\mu$. Since the radius of the ejected drop is $r \simeq R/2 = 75 \mu$, we arrive at a value of $\delta \simeq r/25$ in this case.

The inference is that the negative electrification of the drops produced by the break-up of tiny jets formed during the bursting of bubbles and the impact and partial coalescence of drops on a liquid surface is of similar origin, although the dynamics are different in the two cases. The small positive charges that are observed in both cases at concentrations in excess of about 10^{-4} M may also be attributed to a common cause, i.e., to the separation of charge by the flow of liquid from the contracting to the swelling parts of the breaking jet.

CHARGE SEPARATION DURING EXTENSION AND RUPTURE OF A LIQUID THREAD

EXPERIMENTAL TECHNIQUE AND PROCEDURE

A narrow capillary tube, just touching the surface of liquid contained in a small nickel-plated dish connected to an electrometer, is withdrawn at a predetermined speed to produce a narrow thread of liquid which elongates until it ruptures. The charge separated during rupture of this single thread of liquid is then measured by the sensitive electrometer, and its sign and magnitude determined as functions of the rate of extension of the thread, the diameter of the capillary tube, and the concentration of solute in the water.

The capillary tube, held in a small chuck and connected through a vertical rod to a lever arm held in tension by a vertical spring, is allowed just to *touch* the liquid surface because movement of the capillary tip *through* the liquid produces unwanted charges from contact potentials. On releasing the lever, the capillary is retracted at a speed governed by the tension in the spring. The vertical velocity of the capillary at the time of rupture of the thread was determined by measuring the time taken for the tip to travel 2 mm with an electrical make-break circuit containing a rapid galvanometer and a photographic recorder provided with time-markers at 0.01 sec intervals. Having obtained a calibration curve of capillary velocity against tension of the retaining spring, it was possible to predetermine the former by adjusting the latter, and to vary the rate of extension of the liquid thread from about 50 to 500 cm sec⁻¹.

Experiments were made with capillaries consisting of stainless steel hypodermic needles, 0.25 mm diam., with their ends ground flat, and with drawn capillaries of Pyrex with ground and polished ends, all of which were carefully cleaned and stored in sealed tubes of clean water before use. The water or aqueous solutions in the reservoir were supplied from the apparatus described and all the same precautions for cleaning and screening the apparatus were also taken. The absorption of carbon dioxide from the air was not thought to be important since a measurement could be made within a few seconds of turning off the supply of fresh liquid to the capillary, but its effects were noticeable if the liquid surface was allowed to age for a few minutes.

The separation of charge accompanying the rupture of the liquid thread was measured by noting the change in potential recorded by the vibrating-reed electrometer connected to the reservoir of liquid. The input capacitance of the system was reduced to a minimum to obtain maximum sensitivity. With an input impedance of $10^{13} \Omega$ and an effective input capacitance of 1.6 pF , a change in input potential of 1 mV corresponded to a charge of $5 \times 10^{-6} \text{ e.s.u.}$ Charges as low as $1 \times 10^{-6} \text{ e.s.u.}$ could be detected but random noise and pick-up made accurate measurements below $5 \times 10^{-6} \text{ e.s.u.}$ difficult.

RESULTS

In order to assess the reproducibility of the results, a series of measurements was made with one particular capillary withdrawn repeatedly at the same speed from a $1.5 \times 10^{-7} \text{ M}$ solution of sodium chloride. The results are shown in the histogram of fig. 5; half of the 40 individual measurements lie within 5 % of the mean value,

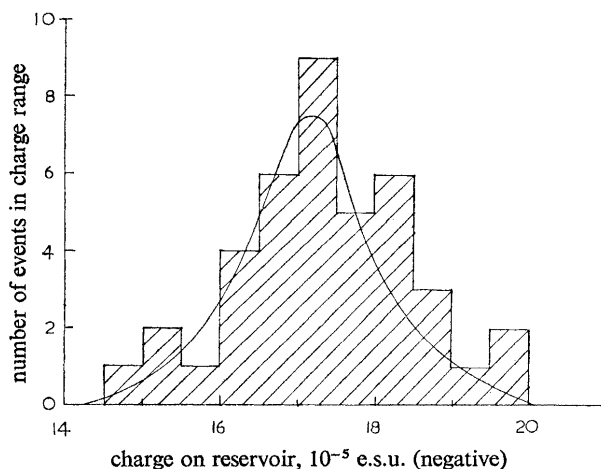


FIG. 5.—The charges separated by the rupture of a liquid filament produced by the withdrawal of a capillary tube from a dilute NaCl solution. The histogram shows the results of 40 repetitive trials.

the random error in each individual measurement being about $\pm 10 \%$. The manner in which the separated charges varied with the velocity of the capillary tip for solutions of various concentrations are shown in fig. 6. Each point represents the mean of usually three individual measurements except for the small charges associated with the most concentrated solutions, where each point is the mean of at least six readings. The results obtained with steel and glass capillaries were similar indicating that charges could not be attributed to contact potentials between the needle and the liquid. For capillary speeds of less than about 250 cm sec^{-1} the reservoir becomes negatively charged, the magnitude of the charge being independent of the capillary diameter and of the velocity if this is $< 150 \text{ cm sec}^{-1}$. But, at higher speeds, the negative charging decreases in magnitude and eventually reverses, the positive charging now increasing with increasing speed, and roughly in proportion to the diameter of the capillary. In both regimes the magnitude of the charge separation decreases as the concentration of the solution is raised, except for the very weak positive charging of dilute solutions.

DISCUSSION

Again it seems that these results can be explained in terms of the two charging mechanisms encountered during the formation and rupture of a small jet of liquid—the negative charging produced by the dragging up of the surface layers of liquid,

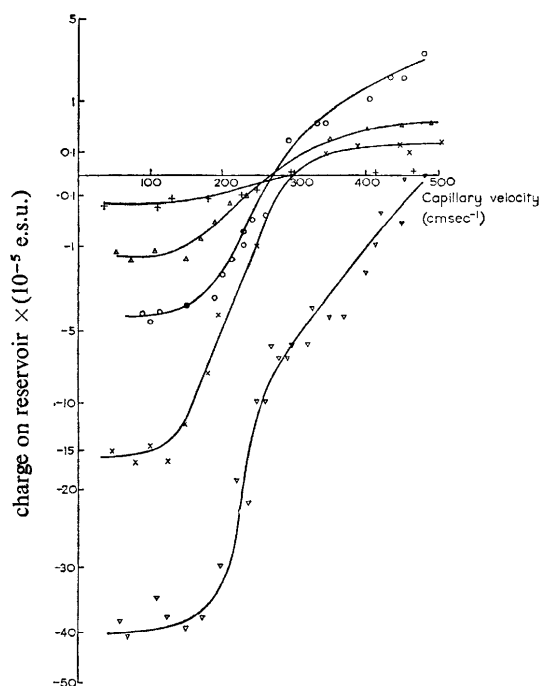


FIG. 6.—The charges separated by the rupture of threads of NaCl solutions as a function of the rate of extension of the thread and the concentration of the solution.

∇ , 6.0×10^{-8} M; \times , 1.5×10^{-7} M; \circ , 3.0×10^{-7} M; \triangle , 5.0×10^{-5} M; $+$, 9.0×10^{-4} M.

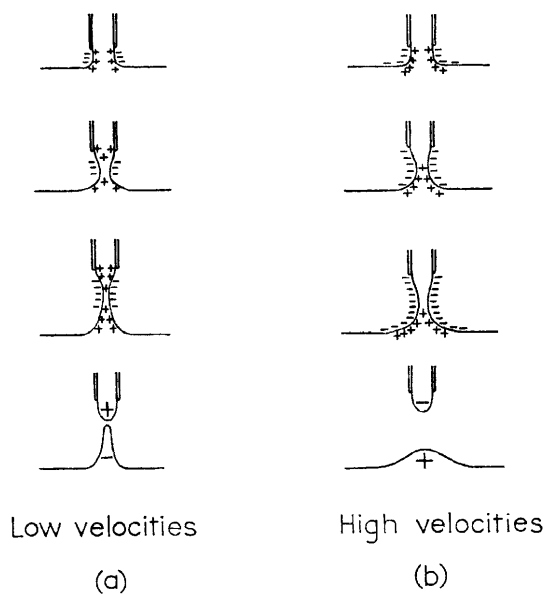


FIG. 7.—Sketches to illustrate the mechanism of charge separation during the extension and rupture of threads of polar liquids.

and that produced by differential flow within the varicose jet. Withdrawal of the capillary tube from a liquid surface draws out a filament which narrows in the centre as shown in fig. 7, and the pressure differences set up by surface tension cause liquid to flow from the neck into the extremities with a mean velocity given approximately by

$$\bar{u} = (1/4\pi)\gamma b/\eta a, \quad (4)$$

where γ and η are respectively the surface tension and viscosity of the liquid, a the radius of the equivalent undistorted cylindrical thread, and $(a-b)$ is the radius of the constriction. If $b/a = \frac{1}{2}$, $\bar{u} = 150 \text{ cm sec}^{-1}$ for water.

If the capillary is withdrawn and the thread elongates at speeds much greater than the mean flow velocity \bar{u} , the charging produced by the dragging-up of the surface of the liquid is likely to dominate over that arising from differential flow inside the thread. One might then expect the thread to acquire a negative charge and, after rupture, leave a positive charge on the reservoir as indicated in fig. 7(b). In this case, the results are consistent with eqn. (1) in that the charge separation is proportional to the capillary diameter, i.e., proportional to the area of the surface film, and in that it decreases with increasing concentration of the solution.

On the other hand, if the capillary is withdrawn slowly, positive charging of the extremities of the filament by differential internal flow may well dominate. In this case, if the filament ruptures and leaves behind the greater part of its volume attached to the bulk liquid, this should acquire a net negative charge as indicated in fig. 7(a). Iribarne and Mason calculated the current flowing through the neck of the varicose thread to be of order 1 e.s.u., this being independent of the initial diameter of the filament and of the concentration of the solution if this is low. Since the rupture times of the filament in the present experiments are about 1 msec, the measured charges of order 10^{-4} e.s.u. shown in fig. 7 for dilute solutions may be reconciled with the theory if one allows for the fact that rupture will not usually ensure complete segregation of the charge separated during the extension of the thread. In the more concentrated and highly conducting solutions, leakage currents along the thread will tend to neutralize the separated charges and so account for the smaller charges that are measured in these circumstances.

CHARGE SEPARATION DURING BREAK-UP OF UNCHARGED DROPS IN AIR

EXPERIMENTAL METHOD

An almost ideal method of detecting and measuring any charge that might be separated during the break-up of a liquid filament due to differential internal flow would be to arrange for an *initially uncharged* filament to break up freely in a field-free space and to measure the charges on the individual fragments. Such an experiment became feasible when careful observation of the vibrating-needle device through a telescope in stroboscopic illumination revealed that the tip throws off relatively large drops joined temporarily to the needle by a long narrow thread of liquid, which subsequently detaches itself and then breaks up into two or three very small droplets. Since this mode of break-up can be made reproducible, the secondary droplets separate into homogeneous coherent streams that can be collected in a small metal can connected to a sensitive electrometer and their average charge measured. In the ideal case, in which the thread would carry no initial detectable charge, the measured charges on the individual fragments could be attributed entirely to the disruption process. In practice, the threads usually carry a significant initial charge unless special steps are taken to neutralize it but, even so, this can be distinguished from the charge q produced during disruption as follows. Suppose that the narrow thread, carrying an initial charge Q breaks up into two similar sized drops of radii r_1 and r_2 . Then assuming that Q is divided between the drops in proportion to their capacitances, i.e., to their radii, the resultant

charges on the two drops will be

$$q_1 = r_1 Q / (r_1 + r_2) + q; \quad q_2 = r_2 Q / (r_1 + r_2) - q,$$

hence

$$q = (r_2 q_1 - r_1 q_2) / (r_1 + r_2) \quad (5)$$

and if all the quantities on the right-hand side of eqn. (5) are determined, the charge q separated during break-up can be deduced.

Another technique was to neutralize the initial charge on the filament by catching the two streams of secondary droplets in different collectors, connecting the latter in parallel, and adjusting the potential applied to the needle until the electrometer gave a zero reading. By moving away each can in turn, (taking care to produce no significant changes in capacity of the system), it was possible to check that the currents carried by the two streams were equal and opposite and thus obtain a measure of the separated charge q .

In a third method, it was possible to adjust slightly the amplitude of vibration of the needle tip so that, without producing any significant change in the mode of formation of the filament, it was possible to make the secondary streams collide and coalesce to form a single stream. This procedure would affect the separated charge but not the original charge Q . The potential on the needle could then be adjusted until the combined streams appeared unchanged, i.e., $Q = 0$. When the streams were again separated by a small change in amplitude, they were found to carry equal and opposite charges, which were however, a function of the amplitude.

A detailed comparison of these three methods was made for drops of 5×10^{-6} M sodium chloride solution produced at the rate of 325/sec. The currents carried by the two streams of drops having radii 21 ± 2 and $30 \pm 2 \mu$ were measured separately as the potential applied to the needle was increased. With the needle earthed, the smaller drops carried an average charge of $+2.1 \times 10^{-6}$ and the larger drops -4.2×10^{-6} e.s.u. Substitution of these values in eqn. (5) yields a value of 2.9×10^{-6} e.s.u. for the charge transferred during break-up, with the smaller drop receiving the positive charge. The charges on the two streams were equal and opposite when a potential of 160 mV was applied to the needle, their magnitude then being 2.6×10^{-6} e.s.u. In the third method, the combined stream showed no charge when a potential of 165 mV was applied to the needle. When the streams were allowed to separate again and the amplitude of the vibration restored to its initial value, the average charge on the smaller positively-charged drops was 3.0×10^{-6} e.s.u. Thus within the estimated experimental error of $\pm 10\%$, all three methods gave results that were in good agreement. Consequently most measurements were made with the second method, which was the most convenient and obviated the need for accurate and tedious measurements of droplet size.

RESULTS

Filaments of liquid could be made to disintegrate into two droplets in such a reproducible mode that the charges separated on secondary droplets of radius 23 ± 2 and $37 \pm 2 \mu$ could be measured with solutions of differing concentration. The results are shown in fig. 8 in which the smaller droplets are shown to carry a positive charge. The charge separated for a fixed concentration is not greatly influenced by slight variations in the mode of break-up provided that the ratio of the diameters of the two secondary drops does not exceed about 2. It was also possible to make the thread break up into three droplets, the two outer ones having radii $30-40 \mu$ and the middle one, formed from the thinner central part of the thread, being about 20μ in radius. In this case the outer drops were found to carry a positive charge and the centre drop a compensating negative charge. Again the magnitudes of the charges decreased with increasing concentration of the solution in much the same manner as indicated in fig. 8. Similar results to those of fig. 8 were also obtained with solutions of thorium nitrate, ammonium chloride and dilute hydrofluoric acid,

again a strong indication that the charge separation is not very sensitive to the ionic species.

Experiments with non-polar liquids such as benzene and carbon tetrachloride yielded the important result that no detectable charging was produced. The droplets

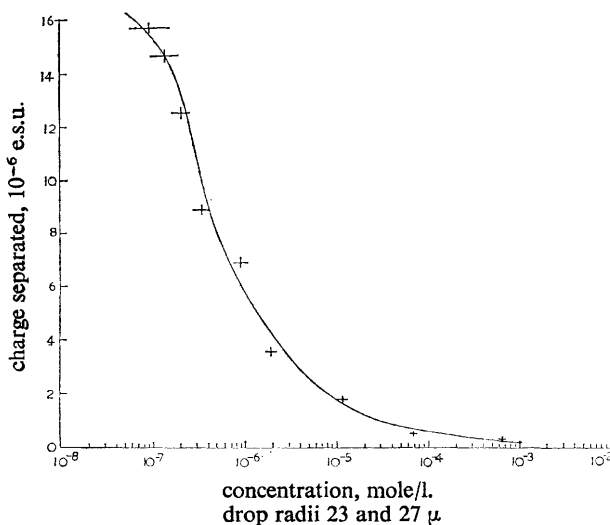


FIG. 8.—The positive charge carried on the smaller droplet produced by the break-up of a thread of NaCl solution into two droplets of radius 23 and 27 μ , as a function of the solute concentration.

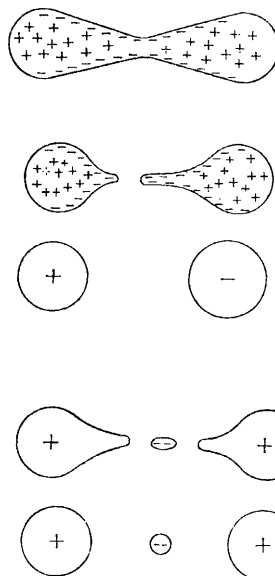


FIG. 9.—Sketches to illustrate charge separation during the break-up of threads of a polar liquid into two and three droplets.

produced from paraffin carried small but highly variable charges which probably arose from the presence of impurities. The fact that the polar nature of the liquid seems an essential feature of the charging process provides strong support for the view that the electrification results from disruption of an electrical double layer formed as a result of the preferential orientation of polar molecules at the liquid/air interface.

The observations on threads producing three droplets, with the centre one negatively charged, provide perhaps the most convincing evidence for Iribarne and Mason's theory that in an unstable varicose filament the liquid flowing from the contracting neck into the swelling extremities carries a net positive space charge.

The measured charges of about 1.0×10^{-5} e.s.u. on drops of dilute solutions ($C = 10^{-7}$ M) are consistent with the results of a calculation based on eqn. (2) and Castleman's⁴ (1931) formula for the break-up time τ of a jet, viz.,

$$\tau = a^{\frac{3}{2}} \log(a/b), \quad (6)$$

where a is the initial radius and b the amplitude of the initial disturbance, and which predicts an upper limit of 3.3×10^{-5} e.s.u. for the charge separated during the rupture of the thread of radius 10μ . The charge actually separated during rupture of the thread will be usually less than the maximum possible charge. A thread, breaking symmetrically into two exactly equal drops, would produce no separation of charge; the fact that two slightly unequal drops are usually formed with the larger one acquiring a negative charge may be a result of the latter acquiring the major part of the central connecting thread as depicted in fig. 9. Again the lower charges associated with more concentrated solutions may be explained by appreciable leakage of charge back through the more highly conducting thread.

CONCLUDING REMARKS

The results described in this paper and those of Iribarne and Mason on bursting bubbles reveal that the break-up of small jets and threads of water and aqueous solutions, in the absence of external fields and contact potentials, produces systematic and consistent charging of the resulting drops. The magnitude of the charges separated during disruption depends largely on the time taken and upon the concentration of the solution but not on the ionic species. The results find a reasonable quantitative explanation in terms of the two mechanisms proposed by Iribarne and Mason, one acting during formation of the jet and the other during the break-up stage, but both producing charge separation by the disruption of the electrical double layer that exists at the surface of a polar liquid.

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³ O. W. Jayaratne and B. J. Mason, *Proc. Roy. Soc. A*, 1964, **280**, 545.

⁴ R. A. Castleman, *Bur. Stand. J. Res.*, 1931, **6**, 369.