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The instability of evaporating charged drops

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These studies constitute an extension of the work of Doyle, Moffett & Vonnegut (1964).

Experiments showed that the evaporation of a charged droplet of water, aniline or toluene supported in a vertical electric field is accompanied by no discernible loss of charge and a consequent increase in electrical pressure in the drop surface owing to the decrease in radius. When the relationship between drop charge Q and radius R becomes consistent with the Rayleigh criterion the drop disintegrates at its uppermost point, where the electrical pressure is a maximum, to eject about 25 % of its mass in the form of highly charged droplets. The measured relationship between the quantity of ejected charge, ΔQ, the mass loss, ΔM, and R was close to that derived theoretically from Rayleigh's equation, indicating that the value of ΔQ for a particular ΔM and R is approximately the theoretical minimum and that an individual drop may undergo a sequence of disintegrations, as was observed. The range of droplet radii studied by means of this technique was 30–200 μ.

The absence of disintegrations during the evaporation of much larger charged drops suspended from insulating fibres was attributed to corona discharge.

1. Introduction

Doyle *et al.* (1964) measured the strength of the electric field required to support evaporating charged droplets of various volatile liquids of initial radii ranging from 30 to 100 μ. Their observations indicated that as a charged droplet evaporates its surface density of charge increases until the electrical pressure attains a critical value, approximately consistent with the Rayleigh criterion, at which point the droplet disintegrates. The disintegration was accompanied by the ejection of one or more highly charged droplets, corresponding to a charge loss of about 30 % of the original value.

The present study constitutes an extension of the work of Doyle *et al.* The range of radii of droplets supported in an electric field was increased to 30–200 μ and further studies were made of the evaporation of larger charged drops of several millimetres diameter suspended from an insulating fibre. The technique of Doyle *et al.* did not permit continuous measurement of the charge and radius of the droplets before and after disintegration, nor was it possible to explore the relationships between the charge and mass loss and the droplet radii. An attempt was made in the present studies to remedy these deficiencies.

2. The disintegration experiments

The experimental arrangement, which embodies the principle of the classic oil-drop experiment of Millikan (1935), is shown in figure 1. The vertical electric field required to suspend charged droplets was applied between two parallel copper electrodes of side 36 cm and separation 11 cm by means of a continuously variable 600 V power supply. The voltage difference across the plates could be measured to within $\pm 1\%$ by means of an electrostatic voltmeter. Droplets were produced using the vibrating capillary device designed by Mason, Jayaratne & Woods (1963) and were charged by means of the application of a potential

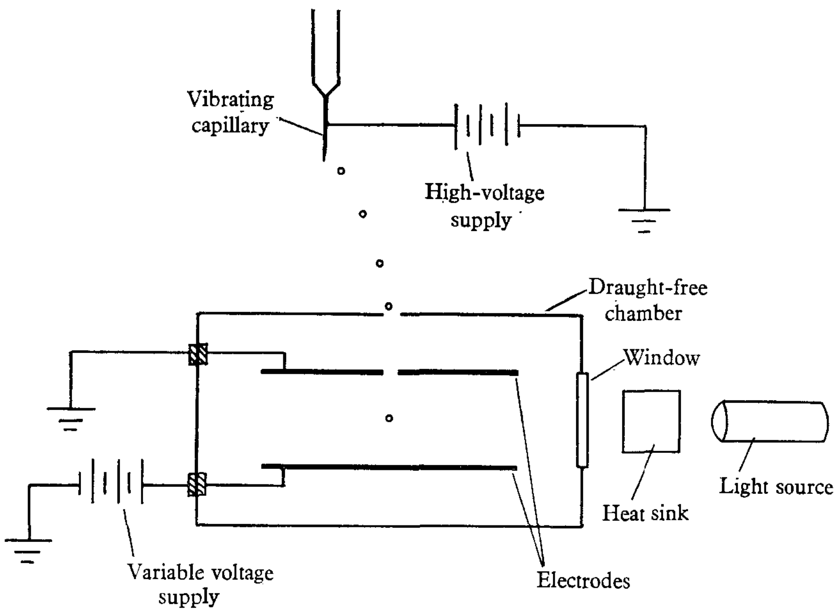

FIGURE 1. Apparatus for determining the behaviour of evaporating charged droplets.

from a continuously variable 6 kV supply to the stainless-steel capillary. Uniformly sized droplets of selected radii ranging from 30 to 200 μ could be produced by varying the capillary diameter or the flow rate through the droplet production device. The charged droplets were allowed to fall through about 1 m before passing through a small hole in the roof of a draught-free vessel in which the plates were situated. They then fell vertically to enter the field through a small slit in the upper electrode. Droplets suspended in the field were illuminated by means of a collimated light-beam which passed through a heat filter before entering the draught-free chamber in order to reduce convection. The droplets were observed through a window using a low power telescope. The electrodes and the interior walls of the chamber were painted black and the experiments were conducted inside a dark room in order to attain conditions of optimum visibility.

The experimental procedure was to produce charged drops in the manner described and to allow them to enter the region between the plates. The lightsource was switched on and the field was adjusted until a droplet was suspended within it, whereupon the droplet supply was switched off. The selected droplet was maintained between the plates by continuous adjustment of the applied potential difference and all other droplets initially residing between the plates quickly drifted out of the field of view since their charge-to-mass ratio was different from that required for levitation. The charge and radius of the drop remaining in the field were recorded at frequent intervals by alternatively recording the magnitude of the field E_s required to maintain the droplet at a constant horizontal level and the time τ taken for the droplet to fall in a weaker field of known strength E_m between two horizontal lines of vertical separation $L = 6$ cm drawn on the viewing window.

When a spherical droplet of radius R carrying a charge Q is maintained at a constant horizontal level in a field of strength E_s we can write, to a reasonably high degree of accuracy,

$$\tfrac{4}{3}\pi R^3 \rho g = E_s Q, \tag{1}$$

where ρ is the density of the droplet and g is the gravitational acceleration.

The velocity of fall $V\,(= L\tau^{-1})$ of the droplet between the two horizontal lines is related to the prevailing field strength E_m by means of the equation, accurate to about 1 % for liquid drops,

$$\tfrac{4}{3}\pi R^3 \rho g = 6\pi\eta R V + E_m Q, \tag{2}$$

where η is the viscosity of the air.

The radius of the droplet can be expressed by means of (1) and (2) as

$$R^2 = \frac{9\eta V}{2g\rho}\left(\frac{E_s}{E_s - E_m}\right) \tag{3}$$

and the corresponding value of the droplet charge Q can be deduced by direct substitution of the value of R obtained from (3) into (1). The successive measurements of E_s and τ therefore permit values of Q and R to be recorded continuously throughout the period of observation. This continuous method for determining the values of the radius and charge of the evaporating droplet possesses several advantages over that adopted by Doyle *et al.* The earlier workers determined the radius of a test droplet by measuring the size of the stain produced on impregnated filter paper upon which it was collected. This method yields only a single value of R and neglects the continuous change in drop radius produced by evaporation and the even more pronounced reduction produced by disintegration. Their values of R and Q, which is deduced from R, were therefore possibly quite inaccurate in many cases, whereas in the present studies continuous measurement of these parameters prevented serious errors in these values from arising.

This procedure for determining R and Q was repeated continually throughout the period of observation of a suspended charged droplet. If an explosion occurred the field was quickly raised in order to maintain the suspension of the residual droplet and the measurements of τ and E_s were continued. After intensive practice it became possible to suspend a droplet through a succession of disintegrations before it was removed from the field; on several occasions five explosions of a single drop were recorded. Measurements were made for charged droplets of water, aniline and toluene.

As observed by Doyle *et al.* the field required to support a charged droplet before disintegration occurred had to be decreased gradually and continuously

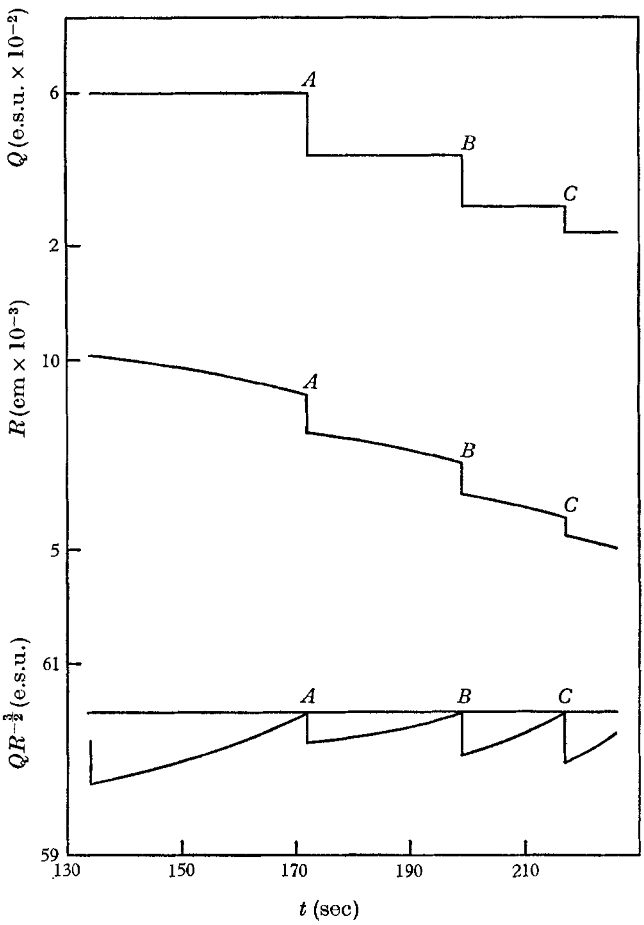

FIGURE 2. The variation with time, t, of the charge, Q, the radius, R, and the parameter $QR^{-\frac{3}{2}}$ for an evaporating water droplet. The horizontal line represents the theoretical curve obtained from Rayleigh's criterion.

as the evaporation proceeded. Evaporation therefore produces an increase in the charge-to-mass ratio of the droplet. When a disintegration occurred the droplet was observed to fall quickly and the field strength had to be abruptly increased in order to retain the droplet between the plates. A disintegration is therefore accompanied by a decrease in the charge-to-mass ratio of the droplet. Careful observation showed that the disintegration always occurred at the uppermost part of the droplet. This finding is consistent with the theoretical predictions since the electrical pressure is greatest at this point, where the electric field produced in the droplet surface by the charge Q is reinforced by that due to the external field. The rapid fall of the droplet after disintegration is therefore a consequence both of the sudden decrease of the charge-to-mass ratio and the law of conservation of momentum.

Figure 2 shows the variation with time of the charge and radius of a charged droplet of water evaporating in the region between the plates. Each disintegration is accompanied by a loss of about 25 % of the charge residing on the droplet. It is seen that no measurable loss of charge occurs in the intervals between successive explosions but that R decreases continuously in these periods. Figure 2 also demonstrates that the electrical pressure in the quiescent periods increases until the value predicted by the Rayleigh criterion is obtained when an explosion occurs which produces a sudden decrease in the charge-to-mass ratio of the droplet.

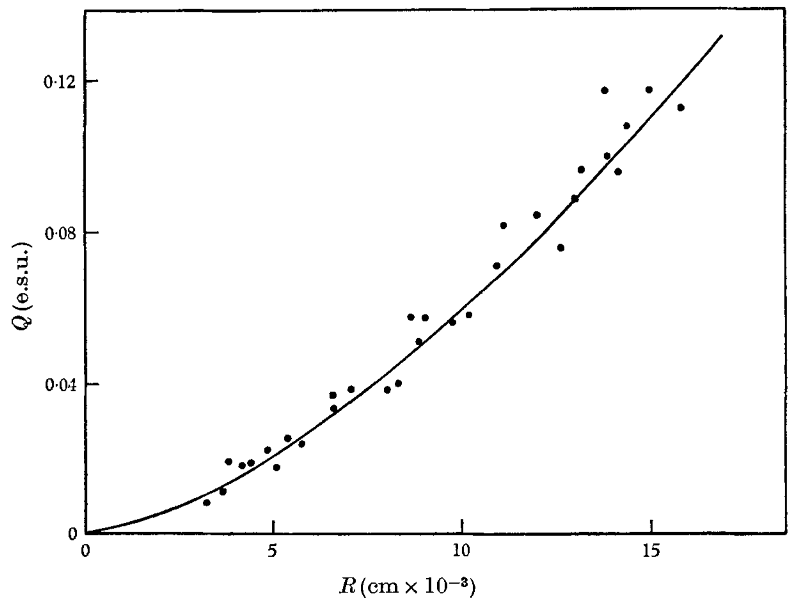

FIGURE 3. The relationship between the radius, R, of an evaporating droplet of water and its charge Q. ●, experimental points; ——, theoretical curve.

Figures 3–5 illustrate the excellent agreement found to exist between the theoretical and experimental relationships between Q and R at the disintegration point for charged droplets of water, aniline and toluene respectively.

The criterion deduced by Rayleigh (1882) for instability can be re-expressed in terms of the mass M of the droplet by the equation

$$Q^2 = 12TM/\rho, \tag{4}$$

where T is the surface tension. Therefore, if the relationship between the charge loss ΔQ and the mass loss ΔM associated with a disintegration is such as to reduce the energy level of the droplet by a minimal amount and therefore to yield a residual droplet of radius r on the verge of disintegration we can write

$$\frac{\Delta Q}{\Delta M} = \left(\frac{9T}{\pi\rho^2}\right)^{\frac{1}{2}} \left(\frac{1}{R^{\frac{3}{2}} + r^{\frac{3}{2}}}\right). \tag{5}$$

Equation (5) thus provides an expression for the minimum loss of charge ΔQ of a droplet of initial radius R which disintegrates to eject a mass ΔM and produce a residual droplet of radius r. Figure 6 provides a comparison between the measured relationship between ΔQ, ΔM and R and that computed from equation (5) on the basis of the observation that about 80 % of the droplets lost between

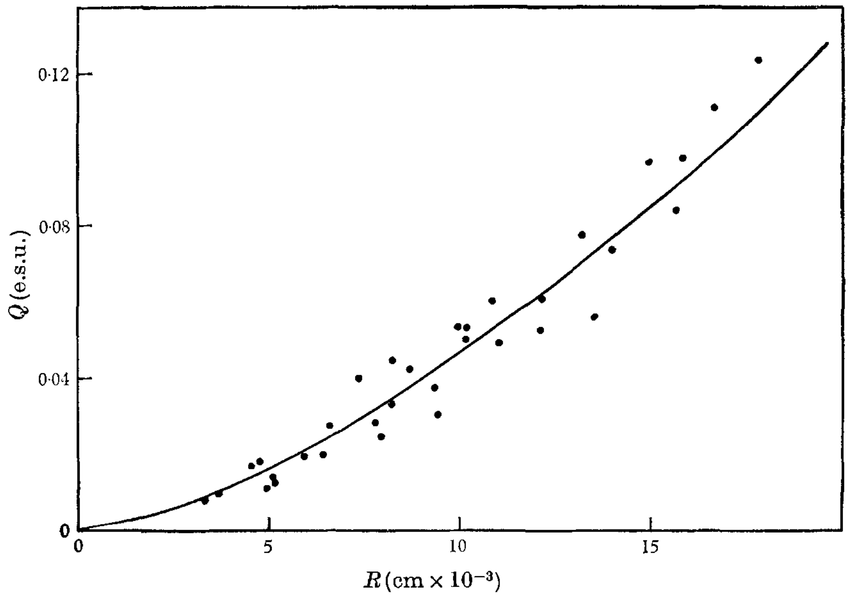

FIGURE 4. The relationship between the radius, R, of an evaporating droplet of aniline and its charge Q. ●, experimental points; ——, theoretical curve.

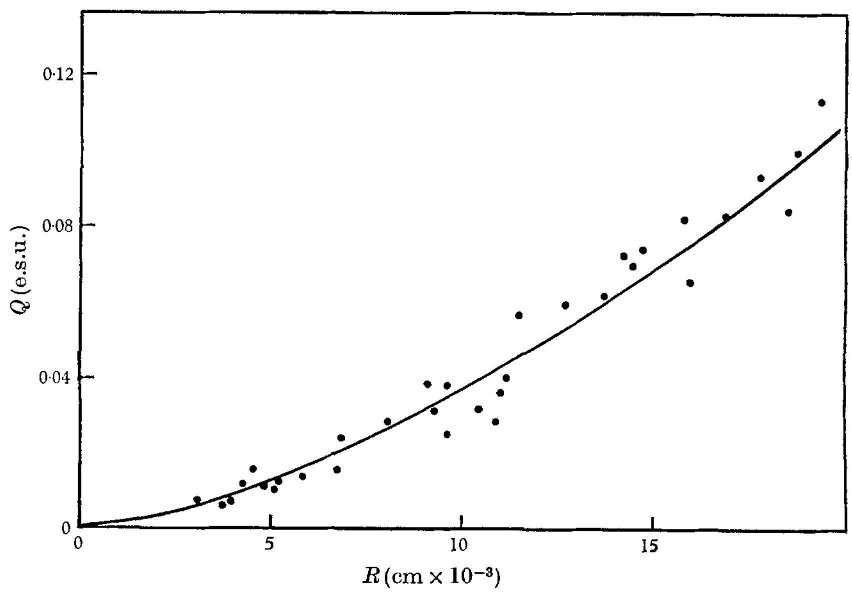

FIGURE 5. The relationship between the radius, R, of an evaporating droplet of toluene and its charge Q. ●, experimental points; —— theoretical curve.

20 and 30 % of their original mass on explosion. Values of $\Delta Q/\Delta M$ were therefore computed from (5) using the approximation $r = 0{\cdot}92R$. It is seen from the figure that the loss of charge accompanying the ejection of material from a disintegrating droplet is close to the minimum required to reduce the electrical pressure in the drop surface below the critical value calculated from the Rayleigh criterion. It is therefore not surprising that several explosions of the same drop were observed to occur within a short interval of time. Similar experimental curves to that illustrated in figure 6 were obtained with droplets of aniline and toluene.

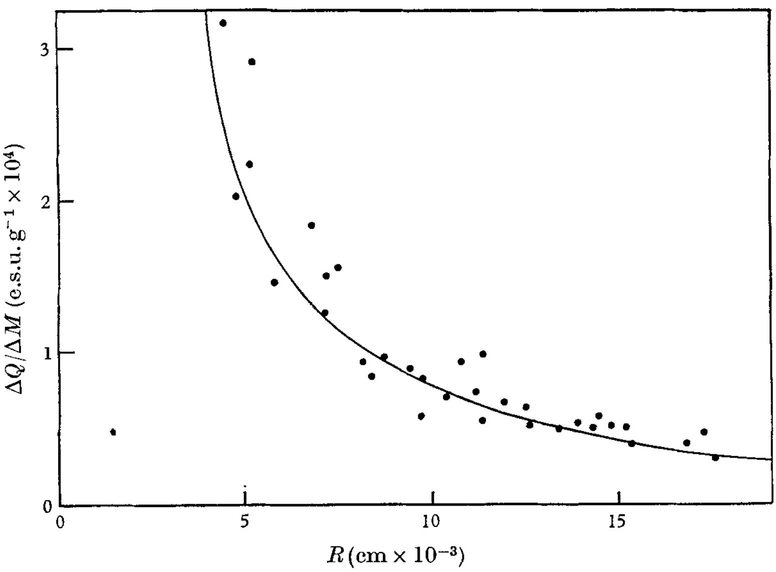

FIGURE 6. The variation of the ratio of the charge ejected, ΔQ, to the mass loss ΔM, accompanying the disintegration of an evaporating water droplet with the initial drop radius, R, at the instability point. ●, experimental points; ——, theoretical curve.

In a separate study highly charged water drops of radii ranging from 0·10 to 0·15 cm were suspended from an insulating fibre and ventilated with a stream of dry nitrogen at a temperature of 95 °C. The drops evaporated to zero radius within 1 min, but although subsidiary experiments had demonstrated that the charge leakage occurring in this period from a drop of constant radius was less than 10 %, no explosions were observed. Although it is possible that the presence of the fibre may have inhibited disintegration in these experiments, the probable explanation for the absence of explosions is that the radial field surrounding these large drops extends to a sufficient distance from the drop to permit stray electrons entering the field to be accelerated to sufficiently high energies to produce ionization of air molecules with which they collide, thereby providing conducting channels for removal of a large fraction of the charge residing on the drop.

3. Discussion

The experiments described in the previous section have demonstrated that the criterion for loss of charge of evaporating droplets of radii less than about 200 μ is physical disintegration of the droplets accompanied by the ejection of highly charged tiny droplets. This disintegration has been shown to occur when the relationship between the charge on the droplet and its radius is consistent with that predicted by the Rayleigh criterion. For larger droplets the assumption of sphericity on which Rayleigh's equation is based will not be valid and more accurate values for the disintegration criteria can be obtained from the equation of Abbas, Azad & Latham (1967) which is applicable to non-spherical drops. Experiments have also established that the mechanism of charge removal from evaporating drops of radii greater than 0·1 cm is probably corona discharge. The threshold of radius separating these two processes of charge transfer must therefore lie between 200 and 1000 μ.

As mentioned by Doyle *et al.* the disintegration of evaporating charged droplets will occur in the atmosphere at the edges of electrified clouds and at the surface of lakes and oceans (Blanchard 1959). The contribution of this phenomenon to the global electrical budget is almost certainly negligible, but it may provide significant modifications to localized droplet populations. In order to assess its efficacy in this respect it will be necessary to perform further experiments, probably involving high-speed microphotography, in which measurement is made of the quantities and size spectra of the tiny droplets ejected during the process of disintegration.

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