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Production of charged monodisperse water droplets by electrical dispersion

D. V. AHIRE and A. K. KAMRA

Indian Institute of Tropical Meteorology, Pune

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सार – विद्युत परिक्षेपण तकनीक से उच्च आवेशी एक समान जल बिन्दुओं के उत्पादन संबंधी प्रयोग के प्रेक्षणों का विवरण दिया गया है यह देखा गया है कि जब पानी रिसता है तो विभव बढ़ाने पर बंद का आकार कम हो जाता है और बूंदों की उत्पादन-बारंबारता बढ़ जाती है यह भी देखा गया कि अधिक बाह्य व्यासों वाली केशिकाओं का न्यूनतम छिड़कावी विभव ज्यादा होता है । छिड़काव या स्प्रे की क्रिया में जल बिन्दुओं की समकण परिक्षेपी धारा उत्पन्न करने के लिए आवश्यक विभव बहुत ही क्रांतिक स्थिति में होता है और उसमें जरा-सा परिवर्तन हो जाने से बूंदों का आकार एक समान नहीं रह पाता । इस क्रांतिक विभव पर बूंदों का आकार मुख्यत: प्रभावदर पर आश्रित देखा गया है। बूंदों के विशिष्ट आवेश क प्रेक्षित मानों की सैद्धान्तिक गणना करके उनसे तुलना की गई है । यह रेले द्वारा गणित अस्थिरता सीमा के आधे से कुछ ज्यादा है अतिसूक्ष्म समकण परिक्षेपी कणों की धूमिका उत्पन्न करने के लिए आवश्यक विभव केशनलिका की मोटाई के साथ बढ़ती हुई प्राप्त हुई ।

ABSTRACT. Observations of an experiment for production of highly charged uniform water droplets by the electrical dispersion technique are described. It is observed that in dripping mode, the drop size decreases and frequency of drop production increases as the applied potential is increased. Minimum spraying potential is observed to be higher for capillaries of larger outer diameters. In spraying mode the voltage required to produce monodisperse streams of water droplets is very critical and a slight variation in it produces nonuniformity in drop sizes. At this critical voltage the dropsize is found to depend mainly on the flow rate. Measured values of specific charge on the droplets are compared to the theoretically calculated ones and found to be little more than half of the Rayleigh's limit of instability. The voltage required to produce a mist of very fine monodisperse particles is found to increase with the thickness of the capillary tube.

1. Introduction

Several laboratory simulation experiments to study the microphysical processes responsible for the growth of drop in clouds require the generation of drops or a stream of drops of uniform size. Although several mechanical methods are known for generating such drops, electrical atomization of liquids provides a convenient method to generate a stream of drops of uniform size and charge with sufficient accuracy. The electrical atomization of liquids is, basically, a process by which a liquid volume is broken up into small charged particles as a result of electrostatic pressures overcoming the surface tension pressures. Bailey (1974) provides a review of the theoretical and experimental aspects of the problem. Most of the experiments in the available literature, have been carried out with liquids other than water (*e.g., see* Hendricks 1962; Hogan and Hendricks 1965; Hendricks *et al.* 1964; Jones and Thong 1971; Thong and Weinberg 1971); main interest of many of these investigators being in the combustion of liquid particulate suspensions.

Vonnegut and Neubauer (1952) investigated the production of monodisperse liquid particles by applying voltages of 5 to 10 kV to water placed in small glass capillaries. Bollini and Sample (1970) and Sample and Bollini (1972) also used water as the working fluid and investigated the problem quantitatively. However, in their experiments, as in most of the other experiments, they placed, an earthed electrode very close to the capillary tip and studied the problem in terms of the electric field at the tip of the capillary.

The present study is an extension of the work reported by Vonnegut and Neubauer (1952). All earthed bodies are kept at sufficiently large distances away from the capillary tip so that their presence has no appreciable effect on the drop production, and the phenomenon is studied in terms of the electric potential. Various parameters such as drop size, the frequency of drop production (number of drops produced per second), electric charge per drop, the net electric current carried by a stream (spray) of droplets, and specific charge are studied in relation with the applied voltage, flow rate and capillary size. The phenomenon of mist production, and the variation of voltage required to produce mist with capillary-size are also studied quantitatively.

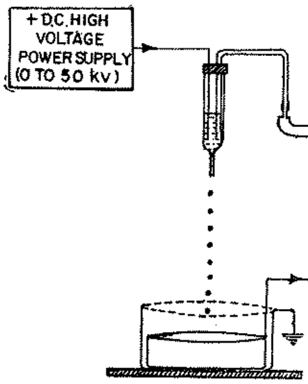

Fig. 1. Experimental arrangement for the production of water drops

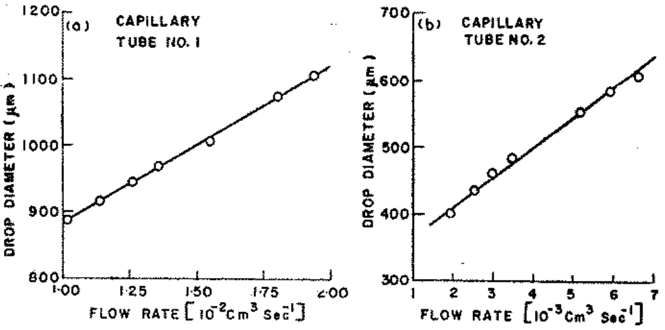

Fig. 2. Monodisperse drop diameter *vs* flow rate at Vc= 6.4 kV for capillary No. 1 and at Vc=8.0 kV for capillary No. 2

2. Experimental technique

A glass tube drawn down to a capillary having a diameter of the order of a few tenths of a millimeter is filled with distilled water (Fig. 1). The flow rate in the glass capillary can be controlled with a cock provided between the glass capillary tube and a glass bottle containing distilled water. A source of variable high positive d. c. voltage (0 to 50 kV) is connected to the distilled water in the glass capillary by a wire placed in the tube.

A copper cylinder of diameter 16 cm and height 6 cm shielded with another grounded aluminium cylinder of diameter 20 cm and height 11 cm is used for collecting the drops falling down from the tip of the capillary. The copper cylinder is placed 45 cm below the tip of the capillary on a teflon sheet and in connected to a Keithley electrometer (Model 610 C). Ground of the d. c. source, the aluminium cylinder, and the ground terminal of the electrometer are connected to a common ground.

The flow rate is so adjusted that water, with no potential applied, slowly comes out of the capillary and forms drops of a few millimeters in diameter which fall off the tip at the rate of a few per minute. In this case, the drop grows until its weight overcomes the net vertical component of the surface tension force. At this point the water close to the capillary forms a neck, which eventually ruptures and allows the main part of the drop to fall from the capillary. When the d. c. voltage is applied to the water in the capillary, the electrical force acting on the drop is in the same direction as the gravitational force; thus the weight of the drop necessary to cause its separation from the capillary tip is reduced. Therefore, the effect of the applied voltage is to reduce the size of the falling drops and to increase their frequency of formation. At low potentials, this mode of drop formation is called the dripping mode.

As the voltage is raised to about 5 kV and above, a sudden transition from the dripping mode to the high frequency spraying mode occurs. This transition is characterized by a sharp decrease in the drop diameter, and a sharp increase in the frequency of drop formation. Once the spraying is established, the residual meniscus which remains attached to the capillary tip after droplet emission is usually larger than the emitted droplet, whereas in the dripping mode this residual meniscus is so small it appears as though the drops are pulled off directly from the capillary tip.

Different techniques are used for the drop size measurements. For drops having diameters of 1 mm or above, the weighing method is used. About twenty-five drops are collected in a small glass container having a close fitting top and weighted in a microbalance having a sensitivity of 0.1 mg. Size of the droplets of about a millimeter or less in diameter, is measured by catching them in a shallow dish of high quality machine oil (Victre A-69). The droplet sizes are then measured by using a microscope with a graticule in its eye-piece. Size of the droplets of the order of 100 μm or less in diameter, is measured by the MgO coated glass slide method.

Since the presence of any earthed object close to the tip of the capillary influenced the drop production, a minimum distance of 40 cm between the capillary tip and any of the earthed objects or the observers own body was always kept in all these measurements. In all the experiments reported here, the earthed metal cylinder is placed 40 cm below the capillary tip.

Electric charge on each individual drop or the net current carried by a stream of droplets is measured by collecting them in the measuring cylinder. In dripping mode, electric charge on each drop is measured separately. Electric current is then determined by the relation :

$$i = nq \tag{1}$$

where n is the number of drops produced per second and q, the charge on each drop. n is found from the time (measured with a stopwatch) required to collect twenty five drops. If the volume of each drop is V, then the volume flow rate, V_f, of water through the capillary is,

$$V_f = nV \tag{2}$$

Thus, the specific charge defined as the charge per unit mass is

$$\frac{q}{m} = \frac{i}{V_f\,\rho} \tag{3}$$

where m is the mass of each drop and ρ, the density of liquid (for distilled water, ρ=1 gm cm^{-3}).

TABLE 1

Measured values of drop-diameter, frequency of the drops production, electric charge on each drop at different voltages for capillary No. 1. Electric current and specific charge are calculated from Eqns. (1) and (3)

(Flow rate$=1.02\times10^{-2}$ cm^3/sec, Conductivity of the distilled water $=3.1\times10^{-4}$ mho/metre, Temperature range$=30°C_{max}$ —$20°C_{min}$)

+D.C. voltage (kV)	Diameter of the drop (mm) (*d*)	Frequency of the drops production, (*n*)	Electric charge per drop (x10^{-10} coul)	Electric current (x10^{-10} amp)	Specific charge (x10^{-5} coul kg^{-1}
0.0	2.98	0.74	—	—	—
1.0	2.96	0.75	0.92	0.69	0.68
2.0	2.89	0.80	1.70	1.36	1.35
3.0	2.78	0.91	2.33	2.12	2.08
4.0	2.63	1.10	2.95	3.16	3.00
5.0	2.37	1.47	3.10	4.60	4.50
5.5	2.08	2.00	3.10	6.20	6.60

In spraying mode, the net current being carried by the streams of droplets and reaching the measuring cylinder, is measured. Electric charge on each drop, specific charge etc., can then be calculated from Eqns. (1) to (3).

3. Results

All the experiments reported here are performed in open air using distilled water as the working fluid. The electrical conductivity of distilled water used is 3.1×10^{-4} mhos m^{-1}. Most of the data are taken with the following two glass capillary tubes :

(*i*) Glass capillary tube No. 1 of inner diameter 0.28 mm and outer diameter 0.71 mm.

(*ii*) Glass capillary tube No. 2 of inner diameter 0.17 mm and outer diameter 0.82 mm.

Glass capillaries are clamped vertically as shown in Fig. 1. For measurements in dripping mode, the flow rate is kept constant and measurements of the drop-diameter, frequency of drops production and electric charge on each drop are made at different voltages. Electric current and specific charge are calculated from the measured values of electric charge on each drop and the flow rate. The results are given in Tables 1 and 2.

The dropsize decreases and the frequency of drops production increases with increase in the applied voltage; the variations being nonlinear and more rapid at higher voltages. Irrespective of the decrease in dropsize, electric charge on each drop increases as the applied voltage is increased upto 5 kV. Between 5 and 5.5 kV, the electric charge on each drop remains almost constant and it decreases with any further increase in voltage. But the electric current and specific charge on a drop always increase with increase in the applied voltage.

TABLE 2

Measured values of drop-diameter, frequency of the drops production, electric charge on each drop at different voltages for capillary No. 2. Electric current and specific charge are calculated from Eqns. (1) and (3)

(Flow rate$=3\times10^{-3}$ cm^3/sec., Conductivity of the distilled water $=3.1\times10^{-4}$ mho/metre, Temperature range$=30°C_{max}-20°C_{min}$).

+D.C. voltage (kV)	Diameter of the drop (mm) (*d*)	Frequency of the drops production, (*n*)	Electric charge on each drop (x10^{-10}coul)	Electric current (x10^{-10} amp)	Specific charge (x10^{-5}coul kg^{-1})
0.0	3.11	0.19	—	—	—
1.0	3.10	0.20	1.08	0.22	0.69
2.0	3.04	0.21	2.05	0.43	1.40
3.0	2.90	0.24	2.80	0.67	2.20
4.0	2.71	0.30	3.35	1.00	3.20
5.0	2.41	0.42	3.60	1.51	4.92
5.5	2.18	0.57	3.60	2.05	6.65
6.0	1.90	0.87	3.20	2.78	8.93

At the end point of the dripping mode (~5 kV and above) the water meniscus at the capillary tip becomes electrohydrodynamically unstable, and electrical spraying of droplets from the meniscus starts. The potential at which the transition from the dripping mode to the spraying mode takes place is called the minimum spraying potential (V_{min}). In Table 3, measured values of V_{min} for glass capillary No. 1 and 2, and four other capillaries are compared with the calculated ones from the expression derived by Hendricks *et al.* (1964), *viz.*,

$$V_{min} = 300\sqrt{20\,\gamma\,\pi\,a} \tag{4}$$

where γ is the surface tension of the liquid used (for distilled water $\gamma=72$ dynes cm^{-1}) and a is the radius of the capillary. The measured values of V_{min} are generally found to be more than the calculated ones for the reasons discussed by Bailey (1974). Observations with capillaries of almost same inner diameters but having different outer diameters [Table 3(B)] indicate that V_{min} increases with increase in the outer diameter of the capillary.

As reported by Bailey (1974), in majority of cases electrostatic atomizatoin generates a polydisperse spray with a considerable distribution in droplet size. It consists of main streams of droplets accompanied with some smaller streams of droplets going in different directions. However, at some critical value of the applied voltage, V_C, in the spraying mode, it is possible to obtain a monodisperse spray of droplets. In such a case, the droplets are collinear in trajectory and the smaller streams of droplets disappear. The dropsize in the monodisperse spray is a function of both, the capillary diameter and flow rate. It decreases with decrease in the capillary diameter and/or flow rate. Values of V_c for glass capillaries No. 1 and 2 are found to be 6.4 and 8.0 kV respectively and these are practically the same for positive

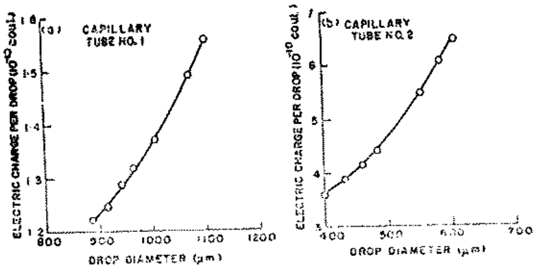

Fig. 3 (a–b). Electric charge per drop *vs* monodisperse drop diameter at V_C=6.4 kV for capillary No. 1 and at V_C=8.0 kV for capillary No. 2;

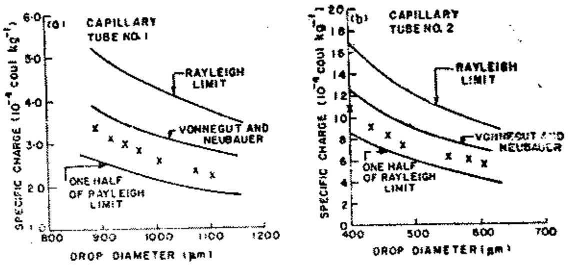

Fig. 4. Specific charge *vs* drop diameter at V_C=6.4 kV for capillary No. 1 and at V_C=8.0 kV for capillary No. 2

and negative voltages. V_C depends mainly on the characteristics of the glass capillary; increasing with the increase in the outer diameter of the capillary. At voltages slightly less or more than V_C, little non-uniformity in the dropsizes is observed. But the mean dropsize is always found to decrease with increase in the applied voltage.

At this critical value of the applied voltage V_C, dropsize and electric current are measured for different flow rates. Electric charge on each drop, frequency of drop production and specific charge are calculated from the measured values of electric current and flow rate. Results for the glass capillaries No. 1 and 2 are shown in Figs. 2, 3 and 4. Dropsize and the electric charge on each drop (and hence the electric current) increases with increase in the flow rate for both capillaries. But the specific charge decreases with increase in the flow rate (dropsize). In Fig. 4 the measured values of specific charge are compared with the calculated ones from the expression derived by Vonnegut and Neubauer (1952)

$$\frac{q}{m} = \frac{3\sqrt{3}\,(\gamma\epsilon_0)^{\frac{1}{2}}}{\rho\, r^{3/2}} \qquad (5)$$

where, ϵ_0 is the permitivity of free space, and r is the radius of the drop.

Rayleigh's (1882) criterion for instability of a charged drop is

$$\frac{q^2}{64\,\pi^2\,\epsilon_0\, r^3} > \gamma \ \text{(MKS units)} \qquad (6)$$

From this the maximum specific charge that a drop can have, can be derived as

$$\frac{q}{m} = \frac{6\,(\gamma\,\epsilon_0)^{\frac{1}{2}}}{\rho\, r^{3/2}} \qquad (7)$$

Curves showing the Rayleigh's limit of specific charge, and half of the Rayleigh's limit, calculated from Eqn. (7), are also drawn in Fig. 4. The measured values of specific charge are found to be less than the calculated ones from Eqns. (5) and (7). This is expected from the fact that some of the droplets about a micrometer in diameter are occasionally observed to leave the capillary tip at relatively high velocities in directions such that they may not reach the measuring cylinder. These droplets may be highly charged. Loss of charge due to corona is also likely at such high voltages. However, it is interesting to note that, in agreement with the results of Sample and Bollini (1972), most of our experimental data points lie close to the line of half of the Rayleigh's limit for specific charge.

4. Production of mist

When glass capillary tubes are clamped horizontally, hydrostatic pressure is almost negligible and normally water does not come out of capillary unless some voltage is applied. When the voltage is increased then at some high value of voltage generally greater than 5 kV, water starts coming out of the capillary in the form of a mist of very fine particles along with some smaller streams of droplets. It is possible to produce mist alone without the streams. The particle size in the mist is of the order of a micrometer or so. When the mist is illuminated with a beam of parallel light then sometimes it is coloured red, green and orange depending on the angle of observation. When the voltage is raised beyond that required to produce the mist, the smaller streams appear again.

An examination of the values of V_{ms} (voltage at which only mist is produced) observed for seventeen different glass capillary tubes indicated that V_{ms} depends upon the outer diameter of the capillary as well. To confirm this, capillaries with almost same inner diameters but having different outer diameters are selected. The results obtained with such capillaries are shown in Table 4. V_{ms} increases with increase in the outer diameter of the capillary.

5. Discussion

A cylinderical charged liquid jet coming out of a capillary, disintegrates under the combined effect of electrical, molecular and hydrostatic forces. Changes in the nature of its disintegration as the applied voltage is increased, suggest that the mechanisms responsible for the jet disintegration may be different in different voltage ranges. For example, when the voltage is initially increased from zero, specific charge on the drops increases as the applied voltage is increased. However, this increase in specific charge is very sharp when the transition from the dripping mode to the spraying mode occurs. Again, the voltage at which monodisperse spray of droplets

TABLE 3

Experimental and calculated minimum spraying potentials for different capillary sizes

	Capillary diameter (mm)		V_{min} (kV)	
	O.D.	I.D.	Experimental	Calculated
(a)	0.82	0.28	5.8	3.83
	0.71	0.17	6.5	4.10
(b)	0.62	0.13	5.8	3.60
	0.78	0.13	7.3	4.00
	1.18	0.13	7.6	4.90
	1.30	0.12	7.7	5.15

TABLE 4

Variation of the voltage required to produce mist (V_{ms}) with capillary sizes

	Capillary diameter (mm)		Voltage to produce mist (kV) V_{ms}
	O.D.	I.D.	
(a)	0.61	0.13	7.00
	0.91	0.13	8.25
	1.18	0.13	9.50
	1.30	0.12	9.75
(b)	0.65	0.23	8.15
	0.90	0.25	9.25
	1.04	0.23	10.00
	1.10	0.24	10.00
	1.31	0.23	10.10
	1.36	0.25	10.45

can be obtained, is very critical. In contrast to the result of Jones and Thong (1971) who report a narrow range for V_C, we observed that even much smaller changes in V_C, produced non-uniformity in dropsizes. This may be either because of different liquids being used in the two experiments or differences in the experimental set-ups of Jones and Thong and that of ours.

Phenomenon of mist production seems to be much different. Earlier studies (Vonnegut and Neubauer 1952 and Drozin 1955) show that with water, mist can be produced only when (*i*) Applied voltage is positive, (*ii*) Magnitude of the applied voltage has some critical value, (*iii*) Flow rates are small, and (*iv*) Conductivity of the water is low. Our observations show that the value of voltage at which mist is produced depends on the outer diameter of the capillary. This voltage, V_{ms}, increases as the outer diameter of the capillary tube increases. Further we observed that V_C also increases with the increase in outer diameter of the capillary tube. These observations suggest the possible role of the adhesive force of attraction between the water and glass as water comes out and spreads at the face of the capillary tip, in drop production.

Production of mist, as mentioned earlier, is possible only when the flow rates are kept small, *e.g.*, when the capillary diameter is very small or the capillary is mounted horizontally. In such cases, water usually does not come out of the capillary unless some voltage is applied. It should be interesting to know what would be the size and charge of the drop if water instead of coming out as mist, comes out of the capillary tip as a single drop. To get an estimate of this, we extrapolated the curves of Fig. 2 to get the diameter of drop which will come out of the capillary if the flow rates are reduced to zero. These were found to be 630μm and 312.5μm for capillary No. 1 and 2 respectively. Then, for these drop diameters we extrapolated the curves of Fig. 3 to get the values of charge these drops would carry. These were found to be 1.02×10^{-10}C and 3.4×10^{-11}C for drops from capillaries No. 1 and 2 respectively. These values of drop diameters and the charge on them give $q^2/64\pi^2r^3 = 59.6\times10^{-3}$ and 54.1×10^{-3} newton metre^{-1} which are not far from the value of $\gamma=72\times10^{-3}$ newton metre^{-1} considering the possible errors involved in extrapolation. Further, the ratio of the diameters of these drops to the diameters of the respective capillaries were found to be 2.25 and 1.82. These values are not much different from the observed values (Jones and Thong 1971 and Huebner 1969) of the ratio of drop diameter to the jet diameter for Rayleigh's theory of the stability of the capillary jets. These results indicate that the hemispherical tip of the water jet just emerging from the capillary tip under the applied potential only, will be carrying charge very close to the limit of instability, and is likely to disintegrate into smaller drops.

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