

## Production of Monodisperse Sprays\*

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A technique for producing monodisperse sprays is described. The technique makes use of Rayleigh's criterion for the breakup of capillary jets by mechanically vibrating a number of uniform size capillary needles arranged in parallel. It was used to produce sprays in the size range of 290–950  $\mu$ , but in principle there is no reason why this range cannot be extended. The regime of spray sizes where drop coalescence is likely to take place is delineated, and methods of minimizing coalescence are described.

### I. INTRODUCTION

IN the course of studying detonations in heterogeneous systems (e. g., liquid fuel in gaseous oxygen),<sup>1</sup> the author developed a technique for producing monodisperse sprays in a 4 cm square, 4 m long tube. This technique is described here with the hope that it may prove useful to researchers in such fields as combustion, aerodynamics, meteorology, etc.

Schemes for producing uniform size drops have been attempted in the past, with apparently the most successful being that of Burgoyne and Cohen,<sup>2</sup> which is based on the Sinclair-LaMer aerosol generator,<sup>3</sup> and those of LaMer *et al.*<sup>4</sup> and Vonnegut and Neubaur.<sup>5</sup> The first two are based on the principle of condensation of a supersaturated gas and the latter on electrostatic atomization. In the first scheme, drop sizes of 7–50  $\mu$  were produced, whereas in the latter schemes drops of the order of 1  $\mu$  or less were obtained. Techniques for producing single streams of uniform drops were developed by several investigators such as Dimmock<sup>6,7</sup> (100–300  $\mu$  drops), Margarvey and Taylor<sup>8</sup> (500  $\mu$ –20 mm), Mason *et al.*<sup>9,10</sup> (30–1000  $\mu$ ) who used vibrating capillaries, Raynor and Haliburton<sup>11</sup> (50–700  $\mu$ ) whose device consisted of a rotating needle which scoops the same amount of water as it passes through a sheet of the liquid, and Wolf<sup>12</sup> (4–200  $\mu$ ) who used a vibrating reed which impinges through the surface of water held in a slit cut through a piece of sintered glass. The drop size obtainable by the last two methods is very

dependent on the penetration depth of the needle or reed. Furthermore, these methods are apparently limited to rather low frequencies (less than 150 cps). It was felt that extension of these devices to produce monodisperse sprays such as by making use of multineedles or reeds would prove difficult. On the other hand, extension of the vibrating capillary jet method was tried and found to be possible.

### II. THEORETICAL BACKGROUND

All vibrating capillary devices for producing single streams of uniform drops are based on Rayleigh's analysis of the instability of capillary jets,<sup>13</sup> which was made after the experimental observations of Savart<sup>14</sup> on the breakup of liquid jets. Rayleigh shows that the frequency  $f$  for maximum instability, which we will call optimum frequency, is related to the jet velocity  $u_j$  and diameter  $d_j$  by the following equation:

$$f = u_j / 4.058 d_j \quad (1)$$

When the jet is mechanically disturbed at such a frequency, drops of uniform size are formed. According to Schneider and Hendricks,<sup>15</sup> however, a range of frequencies such that  $3.5 d_j < \lambda < 7 d_j$ , where  $\lambda$  is the wavelength, can still result in uniform size drops. The drop size will depend on the frequency, as it can be easily shown from conservation of mass that

$$D/d_j = (1.5\lambda/d_j)^{1/3}, \quad (2)$$

where the drop mass is considered equivalent to that of a one wavelength long cylinder of the jet. It can also be shown that

$$\lambda/D = (\lambda^2/1.5d_j^2)^{1/3}. \quad (3)$$

Equations (2) and (3) give  $D/d_j = 1.89$  and  $\lambda/D = 2.38$  at the optimum frequency.

It should be pointed out that Rayleigh's analysis is effectively applicable to a stationary capillary column of an inviscid liquid. Weber<sup>16</sup> extended the analysis to include the effects of the liquid viscosity, and the velocity of the

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<sup>1</sup> E. K. Dabora, K. W. Ragland, and J. A. Nicholls, *Astronaut. Acta* **12**, 9 (1966).

<sup>2</sup> J. H. Burgoyne and L. Cohen, *J. Colloid Sci.* **8**, 364 (1953).

<sup>3</sup> D. Sinclair and V. K. LaMer, *Chem. Rev.* **44**, 245 (1949).

<sup>4</sup> V. K. LaMer, E. C. Y. Inn, and I. B. Wilson, *J. Colloid Sci.* **5**, 471 (1950).

<sup>5</sup> B. Vonnegut and R. Neubaur, *J. Colloid Sci.* **7**, 616 (1952).

<sup>6</sup> N. A. Dimmock, *Nature* **166**, 686 (1950).

<sup>7</sup> N. A. Dimmock, "The Controlled Production of Streams of Identical Droplets," National Gas Turbine Establishment, Memo M 115, (1961).

<sup>8</sup> R. H. Margarvey and B. W. Taylor, *Rev. Sci. Instr.* **27**, 944 (1956).

<sup>9</sup> B. J. Mason, P. W. Jayaratne, and J. D. Woods, *J. Sci. Instr.* **40**, 247 (1963).

<sup>10</sup> B. J. Mason and J. L. Brownscombe, *J. Sci. Instr.* **41**, 258 (1964).

<sup>11</sup> A. C. Raynor and W. Haliburton, *Rev. Sci. Instr.* **26**, 1124 (1955).

<sup>12</sup> W. R. Wolf, *Rev. Sci. Instr.* **32**, 1124 (1961).

<sup>13</sup> J. W. S. Rayleigh, *Proc. London Math. Soc.* **10**, 4 (1878).

<sup>14</sup> F. Savart, *Ann. Chimie* **53**, 337 (1833).

<sup>15</sup> J. M. Schneider and C. D. Hendricks, *Rev. Sci. Instr.* **35**, 1349 (1964).

<sup>16</sup> C. Weber, *Z. angew. Math. und Mech.* **11**, 136 (1931).

jet. However, Crane *et al.*<sup>17</sup> show that if the velocity is low (less than 10 m/sec) these effects can be neglected.

When a liquid is forced through a small hole or a capillary tube, it is noticed that, if the liquid head is small, the liquid forms a small drop which is attached to the exit surface by the action of surface tension. The drop grows, and when it reaches a size such that its weight can overcome the surface tension, it detaches from the surface and falls. Drops formed this way are usually large. Now if the liquid head is increased, there is a pressure level at which a jet of a diameter equal to the exit diameter is formed. The jet usually breaks into irregular size drops at a short distance away from the exit. However, if forced oscillations at frequencies compatible with Rayleigh's analysis are imposed on the exit plane, the drops become regular.

The velocity of a smooth jet exiting from capillary tubes is found by Lindblad and Schneider<sup>18</sup> to be dependent on the surface tension  $\sigma$ , the density  $\rho$ , of the liquid, and the diameter of the jet as follows:

$$u_j^2 \geq 8\sigma/\rho d_j \tag{4}$$

i. e., a capillary jet must have a certain minimum velocity,  $u_{min}$ , before it can be established. Experimentally, we found that actual minimum velocities are lower by 25-35% than this relation indicates as the capillary tube diameter was varied from 0.15-0.5 mm.

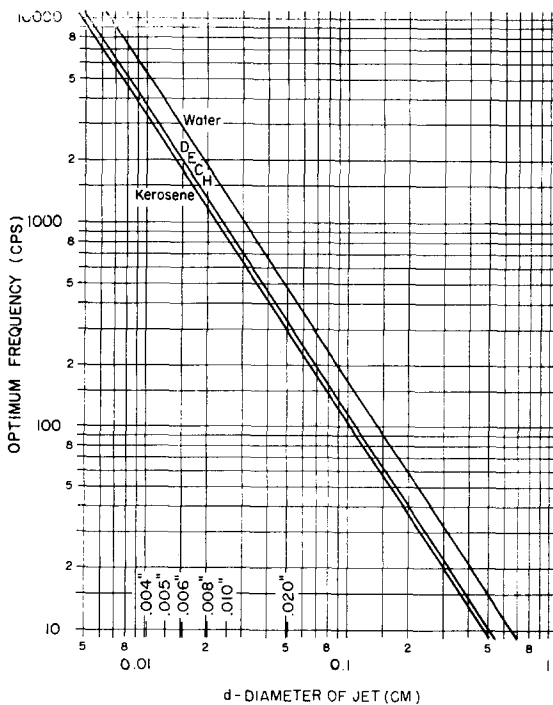


FIG. 1. Optimum vibration frequency for capillary liquid jets at minimum velocity.

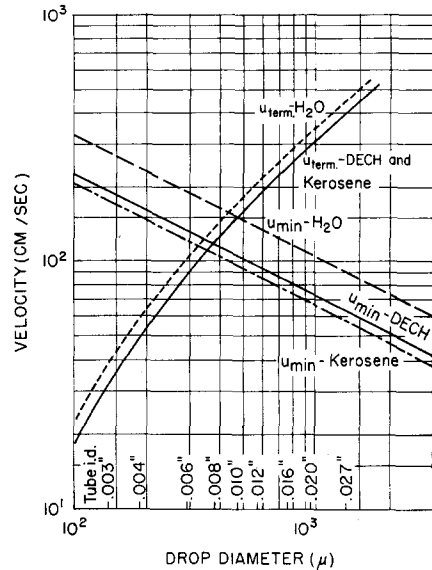


FIG. 2. Terminal and minimum velocities of liquid drops in oxygen at NTP.

It is useful, however, to evaluate the optimum frequency at the minimum possible velocity. This is done by combining Eqs. (1) and (4) to obtain

$$f = 0.627(\sigma/\rho d_j^3)^{1/2} \tag{5}$$

which is plotted in Fig. 1 for three liquids: water, kerosene, and diethylcyclohexane (DECH).

The terminal velocity of the generated drops imposes some restrictions on the design of the spray generator if collisions between successive drops are to be avoided. Figure 2 shows the terminal velocity of water, kerosene, and DECH drops falling in quiescent oxygen at NTP. Shown on the same plot are the initial drop velocities, which are assumed equal to the corresponding minimum jet velocities figured at condition of optimum frequency. It can be seen that for each liquid, there is a certain size tube which would result in a terminal velocity equal to the minimum velocity. When larger tubes are used, the terminal velocity is greater than the initial drop velocity, so that the distance between drops increases as they fall. For smaller tubes, the distance between drops tends to decrease, resulting in collisions and coalescence.

### III. APPARATUS

A modification of the method of Margarvey and Taylor<sup>8</sup> was used for monodisperse spray generation. A schematic diagram of the system is shown in Fig. 3. The liquid is forced into the generator chamber, which has a bleedoff to eliminate air bubbles. The generator consists of a cylindrical chamber (4.3 cm i. d.  $\times$  2 cm long) fitted on its lower end with a plate having the desired number of capillary tubings of the desired size. To insure uniform velocities of the issuing jets, needles of the same length

<sup>17</sup> L. Crane, S. Birch, and P. D. McCormack, Brit. J. Appl. Phys. 15, 743 (1964).  
<sup>18</sup> N. R. Lindblad and J. M. Schneider, J. Sci. Instr. 42, 635 (1965).

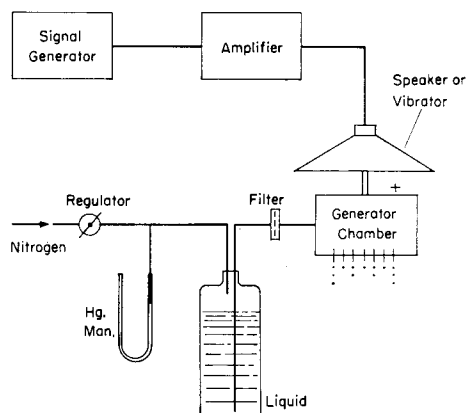


FIG. 3. Schematic of spray generator system.

are used. Needle plates with large needles (o. d.  $\geq 0.5$  mm) are made by inserting the tubings through predrilled holes in the plate and epoxying them to the plate. For smaller needles, an easy and inexpensive way which avoids the drilling of small holes in the plate is used to attach the needles to the plate. Short pieces of tubes whose inside diameter is large enough to accommodate the needles are first epoxyed to the plate and then the needles are inserted and epoxyed. In both cases the needles are made longer than the plate thickness with protrusions from either of its sides. The protrusions from the inside are helpful in eliminating the accumulation of any particles at the tube inlet that would result in clogging, whereas those from the outside are found helpful in eliminating any creeping of the liquid from the needles to the bottom plate surface, which usually causes irregular dripping.

Two equally successful methods of vibrating the liquid jets are used. In one method the chamber is vibrated as a whole, in the other a thin diaphragm—brass shim stock  $\sim 0.076$  mm thick—which replaces the top end plate of the cylindrical chamber is mechanically vibrated. A vibrator or a speaker coil driven by a variable signal generator is used to control the frequency and amplitude of the vibration. Spray observation is made with a Strobflash unit and all photographs of the spray are made with back lighting from a spark source of  $\sim 0.2$   $\mu$ sec duration. Collimated light is used to obtain a large depth of focus.

#### IV. EXPERIMENTAL OBSERVATIONS

The production of monodisperse sprays with drop sizes of the order of  $700 \mu$  in diameter presented little difficulty. Figure 4, for example, shows nine streams of  $750 \mu$  drops obtained from 9 needles arranged in an array of  $3 \times 3$ , 1 cm apart. (The light source and camera are oriented at such an angle that all streams could be photographed.) The uniformity of the drops and the accelerating effect predicted from Fig. 2 are evident. Observation of the spray at distances larger than 60 cm below the generator indicated that the drops remained uniform, but the dis-

tances between drops became random. This can be attributed to the wakes and the flow fields formed by the drops with the resulting irregular convective flow of the gaseous medium. In this respect, there is evidence both experimental<sup>19</sup> and theoretical<sup>20</sup> that a row of spheres falling in a viscous fluid behaves in such a manner that the spheres do not fall along vertical lines, but can overtake, rotate around, and transpose each other.

The spray shown in Fig 4 is unconfined by a tube. It is found that if the spray is confined the spreading of the spray as it falls can result in the wetting of tube walls. However, this wetting is negligible if the spray flow duration is equivalent to the transit time of the drops down the tube. Figure 5 shows a confined  $950 \mu$  spray observed at 2 m below the generator. The needles were arranged in an array of  $5 \times 5$ , 0.5 cm apart. The photograph was taken at 2 sec after the liquid flow was started. It can be seen that the drop size uniformity is excellent; however, the distance between drops is random and some wall wetting occurs, as evidenced by the long, dark streaks. In studying several photographs similar to that of Fig. 5, it is found that the number density of the drops is somewhat lower than expected. Aside from possible loss of drops by wall wetting, there is the possibility that the absolute vertical velocity of the drops can become higher than the terminal velocity of the drops because of the convective flow of the gaseous

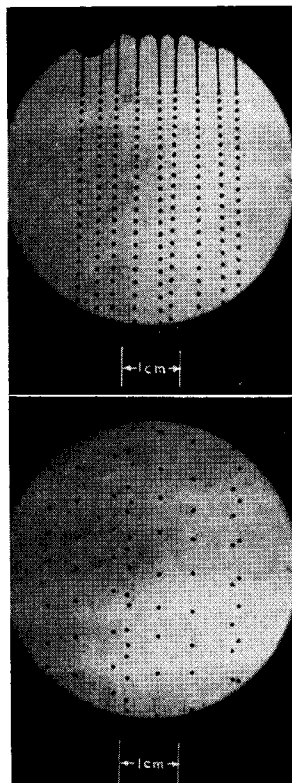
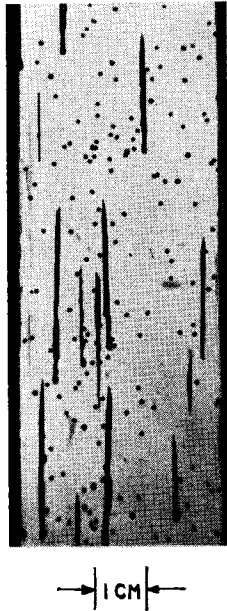


FIG. 4. Monodisperse DECH spray in air produced by  $9 \times 0.04$  cm i. d. needles ( $f=460$  cps). Top: at generator head; bottom: 60 cm below generator.

<sup>19</sup> K. O. L. F. Jayaweera, B. J. Mason, and G. W. Slack, *J. Fluid Mech.* **20**, 121 (1964).

<sup>20</sup> L. M. Hocking, *J. Fluid Mech.* **20**, 129 (1964).

FIG. 5. Confined monodisperse DECH spray in oxygen produced by  $25 \times 0.05$  cm i. d. needles ( $f=300$  cps, 200 cm below generator).



medium. Such a possibility can account for the discrepancy in the number density.

In experimenting with sprays of lower than  $750 \mu$  drops, it is found that coalescence of the drops becomes important. Careful observation indicated that coalescence took place mostly between drops from the same stream, rather than between drops from adjacent streams. The reason for this is believed to be the fact that the drops are following in the wake of their predecessors. The result is that one drop travels faster because of the wake of its preceding one, whereas the succeeding drop, being now at a larger distance from its predecessor, may not be affected by the wake, and may travel at its normal velocity. Hence pairing, which was observed experimentally, can take place, and the effect can be intensified until coalescence takes place.

In order to avoid or minimize coalescence, we have experimented with electrically charging the drops. For instance, one drop generator used to effect this is as follows: The needle plate had ten equally spaced  $0.015$  cm i. d. needles placed on a  $1.5$  cm diam circle. A brass plate  $0.16$  cm thick which has ten  $0.28$  cm holes located so that they are concentric with the needles is placed at about  $0.12$  cm away from the tip of the needles. This charging plate is insulated from the needles, and is held at a different potential from them by a dc power supply. Each jet and its corresponding hole in the plate form two elements of a condenser, so that an electric charge is induced in the jet. As the drops form, they carry this charge. The principle involved in this manner of charging the drops was used by Schneider<sup>15</sup> and Sweet.<sup>21</sup>

It was found that in order to prevent any coalescence, the voltage used (for water, in the above configuration,  $250$  V was adequate) must be high enough that a reasonable

lateral motion of the drops takes place at a distance where coalescence is expected to start in the absence of charging. Visual observation with a Strobflash indicated that no coalescence was taking place at a  $2$  m distance from the needles. However, a disadvantage of this approach is that the spray continues to diverge due to the mutual repulsive forces between drops, and hence the number density becomes variable. Although such a system may prove useful for some applications, it was not suitable for ours.

To avoid the spreading out of the spray, opposite charges were induced in alternate streams. This was effected by using a charging plate made of Plexiglas in which the holes were lined with a conducting material. Five alternating holes were connected to the positive side of a dc power supply and the other five were connected to the negative of a similar power supply. The negative of the first and the positive of the second were connected to the needles. The idea behind this technique is that the spray as a whole would have no net charge and therefore would not spread out. The fact that two adjacent streams are separated by a distance much larger than that between drops of the same stream results in attractive forces much weaker than repulsive forces initially, and therefore the probability of coalescence of drops from adjacent streams is decreased. The results obtained by this method when water was used did show a decided improvement over the uncharged sprays as far as coalescence is concerned. However, the method was abandoned because of the extremely high voltages that would have been necessary, if, as was required in our application, hydrocarbon fluids whose conductivity is orders of magnitude lower than water had to be used.

An equally effective method in minimizing coalescence was realized by providing a gaseous flow around the liquid jets through holes concentric with needles. The idea behind this is that the gaseous flow creates disturbances adequate enough to prevent the drops from the same stream in following the trajectory of their predecessors. The optimum amount of gaseous flow is arrived at by experimentation, and usually results in a very gentle stream. For a needle configuration shown in Fig. 6, Fig. 7 shows  $290 \mu$  DECH spray close to the generator

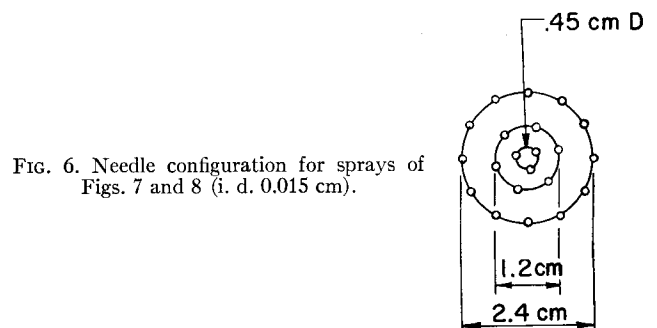


FIG. 6. Needle configuration for sprays of Figs. 7 and 8 (i. d.  $0.015$  cm).

<sup>21</sup> R. G. Sweet, Rev. Sci. Instr. 36, 131 (1965).

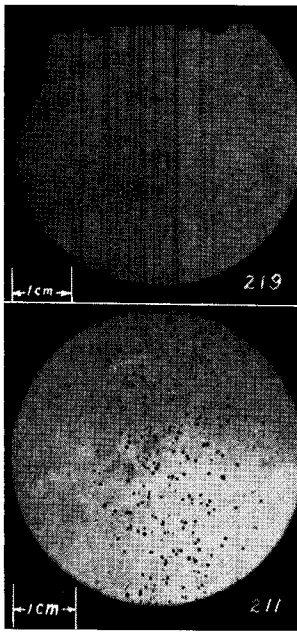


FIG. 7. 290  $\mu$  DECH spray without gaseous flow ( $f=2000$  cps). Top: at generator head, bottom: 60 cm below generator.

head and 60 cm below it when no gaseous flow was used. At this distance it is evident that considerable coalescence has taken place. On the other hand, Fig. 8 shows the same spray with some gaseous flow. The drop streams

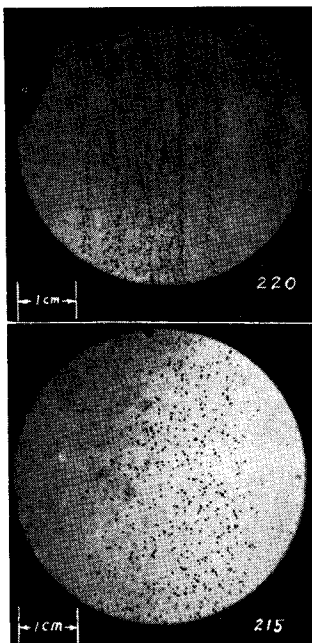


FIG. 8. 290  $\mu$  DECH spray with gaseous flow ( $f=2000$  cps). Top: at generator head, bottom: 60 cm below generator.

are seen to be disturbed at a short distance below the generator with the disturbance being decidedly effective in minimizing coalescence at 60 cm below the generator. Thus a nearly monodisperse spray is obtainable by this technique.

## V. REMARKS

Although the ideas for the above technique have not been followed exhaustively, in principle we see no reason why it could not be used to obtain smaller than 290  $\mu$  sprays. The limitations of course would be the availability of small uniform size tubings and the pressure requirements to force the fluid at its minimum velocity which for laminar flow would be inversely proportional to  $d_j^{5/2}$ .

We should mention that the orderly, simultaneous breakup of liquid jets from multiparallel needles requires a great deal of patience before it can be achieved, as the jets are extremely sensitive to disturbances within the chamber. Bubbles in the generator head, as well as outside disturbances, are found to affect the quality of the spray. The needles must provide equal impedance to the flow so that equal flow velocities are achieved. Dirt accumulation in one tube can cause irregular jet breakup. Sometimes satellite drops are formed, but they could be eliminated by minor adjustment in either flow velocity, vibration frequency, and/or amplitude. It is also observed in some cases that coalescence can be intensified if the drop generator is driven at a harmonic of the natural frequency of the confining tube. Therefore proper care to avoid such a situation is recommended.

The extent of the field of uniform drops depends on many factors, such as drop sizes desired, distances between needles, the confining tube, etc. To study all these factors exhaustively would be very time consuming. However, with the above guidelines, persons interested in monodisperse sprays can reduce the time to develop a system suitable to their particular application.

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