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Electrohydrodynamic Atomisation of Liquids

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A critical analysis is given, with a list of 68 references, of the main problems facing the investigation and the application of the electrohydrodynamic atomisation of a liquid.

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I. INTRODUCTION

The production of highly dispersed monodisperse aerosols of similar charge is of great interest both scientifically and for several branches of industry. The vigorous attack on this problem has been due primarily to the use of charged disperse materials in the electronic–ionic industry and to the development of colloidal rocket propellents. Several processes are available for obtaining such aerosols, one of the most promising being the electrohydrodynamic atomisation of liquids, a particular case of electrostatic atomisation. The term "electrohydrodynamic atomisation of a liquid" appeared comparatively recently in certain foreign papers[1-3]. The customary term "electrostatic atomisation of a liquid" covers in essence a number of processes, which can be grouped under three main types of atomisation having characteristic fields of application.

1. Electrostatic atomisation with large mass flows of the liquids (up to ~ 50 $cm^3\ s^{-1}$), with the liquid fed under pressure, and with very high voltages (up to 100 kV and over). The process is accompanied by the formation of a coarsely dispersed polydisperse aerosol. This mode of atomisation has found its greatest application in industrial electrostatic painting.

2. Electrostatic atomisation with minute liquid flows (10^{-9}–10^{-3} $cm^3\ s^{-1}$) and relatively lower voltages (up to 10 kV) in a vacuum. Under such conditions many liquids (from pure dielectrics to molten metals) can be atomised to highly dispersed aerosols of particles having a high ratio of charge to mass ($\geqslant 10^3$ C kg^{-1}). The first account of the production of monodisperse aerosols by vacuum atomisation is that of Semple and Bollini[4]. This process has been applied in rocket technology to produce colloidal propellents and also in microelectronics in the manufacture of printed circuits.

3. Electrostatic atomisation under normal atmospheric conditions with small liquid flows (10^{-8}–10^{-4} $cm^3\ s^{-1}$) and with relatively low voltages on the liquid (3–8 kV). Under certain conditions this process yields a monodisperse aerosol from many liquids over a wide range of droplet sizes (0.1–1000 μm) and with charges on the drops comparable with the limit permitted by the Rayleigh relation for the stability of a charged spherical drop. This is the type of atomisation to which the term "electrohydrodynamic atomisation of a liquid" is mainly applied.

A characteristic feature of the electrohydrodynamic process is that the small mass flows of liquid and the relatively low voltages create identical conditions of formation for all droplets, merely by drops reaching the Rayleigh instability relation $Q^2 = 16\pi r^3\gamma$ (where Q is the surface charge of the drop, r the drop radius, and γ the surface tension of the liquid), which ensures the production of a monodisperse aerosol. This excludes the drawing-out of long continuous liquid filaments (with not too viscous liquids), which in the electric field would disintegrate into separate drops under the influence not only of Coulombic but also of hydrodynamic and gravitational forces, with consequent formation of a polydisperse aerosol. Production of a monodisperse aerosol in the electrohydrodynamic process is superficially analogous to the mechanism of its formation in disc atomisation with extremely small liquid flows.

A considerable number of papers on the investigation and the utilisation of the electrohydrodynamic process have recently been published both here and abroad. The only survey known to the Reviewers is that by Bailey[5]. However, his analysis of published information is insufficiently critical, and he ignores much published work, both recent and earlier, which would give an idea of the state of and prospects for research on the process. We now review and briefly analyse published results on this question.

The first mention of the effect of an electric charge on a liquid dates from 1745, when Bose[6] observed that the rate of cavitation of a liquid in a narrow capillary increases on application of an electric potential. This phenomenon was later explained by the action in a liquid drop of electric forces opposing those of surface tension. When the charge on the drop reaches a certain value, the electric forces become stronger than those of surface tension, and the drop becomes unstable. A criterion of the stability of a spherical drop of an ideally conducting liquid was first established by Rayleigh[7]. The stability of a charged spherical drop in an electric field was investigated by Zeleny[8-10], who evaluated the critical field strength at which the drop still remains spherical. In experimental studies of a corona discharge from a liquid apex[10] he observed that a liquid filament was drawn out from the narrow capillary, and then broke down into separate drops. This can be regarded as the first attempt to investigate the mechanism of the formation of an aerosol by the electrohydrodynamic atomisation of a liquid.

II. MECHANISM OF THE FORMATION OF AN AEROSOL BY THE ELECTROHYDRODYNAMIC ATOMISATION OF A LIQUID

Experimental results indicating the possibility of obtaining a highly dispersed monodisperse aerosol by the electrohydrodynamic method were first published in 1952–1953 by Vonnegut and Neubauer[11,12]. They atomised such liquids as water, sugar solution, lubricating oil, alcohol, suspensions of fine inorganic colorants in poorly conducting liquids, etc., using both direct and alternating voltages of 6–10 kV. The degree of monodispersity and the size of the droplets were assessed from the presence of bright colours in the higher-order Tyndall spectra. Vonnegut and Neubauer determined experimentally for several liquids the upper conductivity limit at which a highly dispersed monodisperse aerosol could still be formed, and stated that a monodisperse aerosol can be obtained even from more highly conducting liquids, but considerably larger drops (of diameter $\geqslant 25$ μm) were then produced. However, these results were not related to the conditions and are of limited value. The authors stated also that a monodisperse aerosol could not be obtained at negative potentials, but offered no explanation. Nor did they attempt to explain the formation of uniform particles, i.e. the mechanism of formation of a monodisperse list, but nevertheless took[11,12] the first step in a theoretical examination of the break up of the initial volume of charged liquid, by adopting an energetic approach to the problem. They put forward the hypothesis of the minimum energy of the system, on the basis that a charged spherical drop (or other finite volume of liquid) tends to a state in which the total energy of the system (the surface energy and the energy of Coulombic interaction of surface charges) is minimal. Such an energy state should correspond to the ensemble of droplets formed when a primary drop breaks up. Examination of the equation for the total energy of the system enables the equilibrium specific charge on the droplets to be determined. For many authors this hypothesis served as starting point in a theoretical investigation of the electrohydrodynamic process[13–15].

The problem of the mechanism by which aerosol particles—in particular a monodisperse aerosol—are formed is undoubtedly basic for an understanding of the physics of the electrohydrodynamic process. It is not surprising, therefore, that many investigators have been concerned with this question. Thus it was suggested[16] that, at the critical potential for the start of fine atomisation (i.e. formation of a monodisperse aerosol), the electric forces of repulsion exceed the forces of surface tension, and the main filament stretching out from the capillary breaks down into a multitude of symmetrically arranged fine filaments, which become unstable and immediately disintegrate into very fine charged drops. Other workers[17,18] have expressed a similar view on the mechanism. The paper on the electrohydrodynamic production of uniform emulsions using an earthed liquid counter-electrode gives photographs of the atomisation process, on which liquid filaments emerging from the capillary can be seen[18]. However, the fairly long exposure in photographing such a rapid process makes unconvincing the authors' statement that aerosol formation occurs by the rupture of filaments extending from the capillary. Furthermore, the comparatively large masses of liquid consumed in the experiments, the conclusion that a highly dispersed monodisperse aerosol can be obtained from non-polar liquids (such as tetrachloromethane), and finally the fact that it was not an aerosol but an emulsion formed by immersing the particles in a liquid that was investigated—in general cast doubt on the reliability of these results.

Some confusion has hitherto existed in many publications concerned in some measure with the mechanism of electrohydrodynamic atomisation and in particular with the formation of a monodisperse aerosol by this process. Thus electrostatic atomisation with large mass flows of liquid has been investigated theoretically and experimentally[19–25]. The high rate of flow, like high viscosity, results in the formation of a stable and relatively long (ten times the drop size) liquid filament, whose subsequent behaviour in the electric field (bending, cleavage into segments, and breakdown of the latter into individual drops) is governed to comparable extents by electrostatic forces and by hydrodynamic and gravitational forces. Yet the results of these investigations have been compared with those obtained for the electrohydrodynamic atomisation of a liquid. Results for atomisation with large mass flows and with very small flows of liquid—the latter involving purely electrohydrodynamic atomisation—should be discussed separately. It is significant that workers who used the latter régime[11,16,17] concluded that a stage in which a liquid filament extends from the tip of the capillary is essential to the formation of a highly dispersed monodisperse aerosol.

In only one paper[26] has the mechanism of the formation of a highly dispersed aerosol by electrohydrodynamic atomisation been regarded as the detachment of single drops from a liquid apex at the tip of the capillary. However, no experimental data were given to support this mechanism. High-speed photomicrographs of the breakaway of a drop and its movement near the capillary showed[27] large drops under a "dropping" atomisation régime with an exposure time lasting milliseconds, so that it is impossible from this paper to assess the formation and the behaviour of the highly dispersed aerosol obtained by electrohydrodynamic atomisation.

The production of monodisperse droplets of size $r = 5–20$ μm and the formation of an aerosol jet during the electrohydrodynamic atomisation of various liquids (dibutyl phthalate, ethanol, and their 50% solution) have been investigated[28] by high-speed photomicrography using a powerful pulsed laser (20 MW for 35 ns). The experiments showed that formation of a monodisperse mist in this process results from the emission of single droplets from a liquid apex or from the tip of a small liquid cylinder: it is not produced by the stretching out and rupture of a liquid filament, as had been stated earlier[11,16–18,29]. The picture of the process is analogous to the formation of a monodisperse aerosol by disc atomisation with a small supply of liquid.

One of the main problems in the physics of the electrohydrodynamic atomisation of a liquid is to test experimentally the Rayleigh instability relation for a charged spherical drop, as well as to investigate theoretically and experimentally the charges on aerosol particles obtained by this process. A charged electrically conducting spherical drop should retain its size and shape until $Q^2 = 16\pi r^3\gamma$. This relation has been confirmed theoretically[30]. Several workers[31–33] undertook experimental verification by similar methods, consisting in suspending a charged spherical drop in a vertical electric field and observing the moment at which a number of fine droplets were expelled from the surface of the initial drop. This phenomenon was observed in 1926 by Macky[34] in experiments on drops of water placed in a strong electric field. More recently Doyle et al.[31] made a similar experiment on volatile drops. During evaporation the ratio of the surface

charge to the mass of the drop increases, and when the Rayleigh relation is reached a cloud of daughter droplets is emitted. These workers found that the charge on the drop at the instant of disruption of its stability agreed quite well with that calculated from the Rayleigh relation.

A second important result from this work was that the charges on the daughter droplets were very roughly measured as half the Rayleigh value, or at least did not exceed the latter. This work[31] was later developed by Abbas and Latham[33]. The application of an original method[35] to measure the size and charge of drops (30–200 μm) of n-octanol obtained by electrohydrodynamic atomisation confirmed also that the drop becomes unstable on attaining the Rayleigh relation and that the ratio of mass and charge lost by the drop also corresponded to this relation. Schweizer and Hanson[32] have shown with almost the same measuring apparatus that the Rayleigh relation is satisfied to at worst 4%, while the losses of mass and charge by the initial drop as a consequence of the emission of daughter droplets are respectively 23% and 5%.

A series of theoretical and experimental studies of the subdivision of a charged drop in an electric field were regarded by Ryce et al.[1,36-39] as applying to electrohydrodynamic atomisation. They were related to the mechanism of the formation of a monodisperse aerosol during atomisation, since they solved the problem of the symmetrical and unsymmetrical splitting of the initial drop as a function of the closeness of its approach to the Rayleigh relation. A theoretical solution was based on the hypothesis of minimum energy[11] with the postulates[36-38] that (i) the resultant droplets do not interact, (ii) the initial drop is able to divide when $Q^2 < 16\pi r^3\gamma$, and (iii) conduction by the surrounding medium is isotropic, i.e. division of the drop is unaffected by the space charge. The solution leads to the conclusion that drops are able to undergo symmetrical cleavage when $Q^2 > 16\pi r^3\gamma$, and unsymmetrical cleavage when $Q^2 \leq 16\pi r^3\gamma$. The experimental model involved a large drop of water (d = 2 mm) with addition of a surface-active agent in stearin oil. Since the behaviour of a liquid drop in a gaseous medium differs significantly from its behaviour in a liquid, such a model is quite obviously too remote from the actual process of electrohydrodynamic atomisation. Thus the simplifying postulates in the theoretical solution and the evident incompatibility of the experimental model with the electrohydrodynamic atomisation of a liquid diminish the value of these studies for determining the true mechanism.

Although many investigators have adopted the minimum energy hypothesis for the theoretical assessment of atomisation results, some authors reject it completely. Thus Krohn[2] obtained droplet charges exceeding half the Rayleigh value for the atomisation of Wood's alloy. On the basis of these experimental results and an analysis of the work of Vonnegut, Ryce, Hendricks, and Doyle[31], who employed the hypothesis, he asserts that the minimum energy principle cannot be used to obtain quantitative results and that the droplet charges close to the equilibrium value (i.e. half the Rayleigh charge) found by Hendricks and his coworkers are obviously due to the poor conductance of the liquids employed. He publishes his experimental results, but does not give the method for measuring the parameters of the aerosol obtained, so that it is difficult to judge the reliability of these results.

Until recently the magnitude of the charge on the droplets obtained by electrohydrodynamic atomisation has remained an open question. The most interesting work has been done by Hendricks[40], Pfeifer[3,41], and Semple and Bollini[42]. Experiments by Hendricks et al. on the atomisation of octoil and glycerol showed that the individual charges on drops measured by means of a Faraday cage and a flight-path charge spectrometer agreed quite satisfactorily with the theoretical equilibrium charges and never exceeded them. Semple and Bollini used distilled water as working liquid in their experiments, but accomplished the process under a so called "dropping" régime with monodisperse drops of size 140–420 μm formed with a frequency of 350–950 s^{-1}. As in the above work[40] the charges on these drops were quite close to the equilibrium values, but somewhat exceeded the latter. This result was obviously due to errors of technique, e.g. in the measurement of drop parameters consequent on the difficulty of allowing for evaporation of the drops. The Reviewers have recently shown[43] that the individual charges on monodisperse droplets (5–15 μm) of dibutyl phthalate—a liquid of high boiling point, which eliminates effects due to vaporisation and condensation—are close to the calculated equilibrium value, and at least do not exceed the latter.

Thus, in view of the absence of an exact theory of the electrohydrodynamic atomisation of a liquid, the minimum energy hypothesis can be regarded for the present as a convenient basis for examining (with quite good approximation) qualitative aspects of the phenomenon.

III. POSSIBILITY OF QUANTITATIVE ASSESSMENT OF THE PROCESS

The publication discussed above were devoted to a qualitative examination of the underlying physics of electrohydrodynamic atomisation, but a new line of research—a quantitative appraisal of the possibilities of the method, the relation between the process parameters and the characteristics of the resulting aerosol, and hence the range of possible variation of the process parameters with the aim of controlling the aerosol characteristics—was required for the successful application of the process in colloidal rocket propellents. A physical model for the process must, of course, be chosen before the process can be described quantitatively. We shall therefore consider a few models suggested by different authors.

As mentioned above, the model of a charged spherical drop in an electric field suggested by Ryce is too far from the actual process of electrohydrodynamic atomisation. A model involving periodic variation of the form of a liquid hemispheroid under the influence of an electric field at the tip of a capillary was suggested by de Schon and Carson[44]. Assuming a Poiseuille flow of liquid in the capillary, these authors obtained the dependence of the form factor of the liquid hemispheroid on the time and the parameters of the process—the internal radius and length of the capillary, the height of the hydrostatic column of working liquid, the viscosity of the liquid, and the electric potential applied to the liquid—but neither the electrical conductivity of the liquid nor the size of the droplets obtained was considered. Furthermore, the whole work related to pulsating generation of the aerosol, so that the model cannot be used to describe the production of a monodisperse aerosol, since a pulsating régime does not yield monodispersity[43].

Semple and Bollini[42] describe the production of a collimated beam of monodisperse water drops carrying charges of the same sign. The experimental results gave the dependence of the process on the capillary diameter, the hydrostatic pressure of liquid, and the applied voltage. However, large drops (d = 140–420 μm) were generated, when not only Coulombic forces but also gravity played a significant part ("dropping" régime), so that the model cannot be extended to electrohydrodynamic atomisation.

The most noteworthy papers are those by Hogan and Hendricks[40] and by Pfeifer and Hendricks[41]. In the latter[41] theoretical and experimental investigation of the dependence of the specific charge of the droplets obtained by electrohydrodynamic atomisation on the process parameters the starting point was the minimum energy hypothesis and the authors' model of the mechanism of the emission of droplets of a dielectric liquid from the tip of a capillary in a strong electric field. Both régimes with and those without allowance for the effect of the space charge in the region of atomisation were considered. A formula was obtained[41] for determining the specific charge on the particles from the parameters of the liquid and the process (viscosity, dielectric constant, surface tension, electric potential on the liquid, mass consumption of liquid, geometry of the capillary, and distance from capillary to counter-electrode). Experiments on the atomisation of glycerol yielded an empirical formula for the specific charge on the droplets, which agreed quite well with the theoretical formula. In the deduction of the latter, however, the radius of the liquid tip was equated to that of the capillary in the expression for the strength of the electric field between a liquid point and a plane, which is possible only in the limiting case of an ideal dielectric. In the other limiting case (an ideally conducting liquid) the radius of the tip must obviously be equated to that of the droplet emitted from the liquid tip. This would introduce a correction into the proposed[41] formula.

IV. ROLE OF THE ELECTRIC POTENTIAL AND THE CORONA. NEUTRALISATION OF THE CHARGES ON DROPLETS

Among the main characteristics of electrohydrodynamic atomisation are the initial atomisation potential and the critical potential. The former corresponds to the appearance of instability in the liquid meniscus and the start of atomisation. The critical potential represents the development of the process with increase in the voltage applied to the capillary, and the transition to steady emission of droplets from the liquid point. It has been shown theoretically and experimentally[45,46] that with rise in potential the liquid meniscus is stretched to a cone having a vertical angle of $\sim 100°$, after which the surface breaks down with the emergence of a liquid filament or with the detachment of droplets from the tip of the cone. The Reviewers have been unable to confirm a theoretical formula obtained[40] for the initial atomisation potential. This formula obviously represents only a particular case of atomisation, and disregards a variety of process parameters and properties of the atomised liquid.

The most interesting characteristic of the generation of a monodisperse aerosol by electrohydrodynamic atomisation is the critical potential[30,43], corresponding to the transition from pulsating generation with formation of a polydisperse aerosol to steady generation of a monodisperse aerosol. The nature of the critical potential can be explained by the variation in the conductivity of the liquid under the influence of the electric field[41], but it is difficult at present to evaluate this potential theoretically.

Since electrohydrodynamic atomisation takes place in strong electric fields, it is accompanied under certain conditions by the development of a corona discharge from the liquid point or liquid filament. Two main problems—the effect of the corona on atomisation as a function of the sign of the applied potential and its effect on the atomisability of the liquid—have been examined in the many studies[2-11,29,35,46-48] of such a discharge from a liquid drop and its effect on electrohydrodynamic atomisation. All experiments on the effect of a corona have been made with potentials of at least 4 kV. A highly dispersed monodisperse aerosol can be generated electrohydrodynamically at the relatively low potentials of 3–4 kV.[28] The process then occurs in the absence of a corona, whether the potential on the liquid is positive or negative.

The liquid droplets formed by electrohydrodynamic atomisation carry a very high charge, approximately half the Rayleigh value, and therefore possess high electric mobility. The rate of electrostatic leakage is then so large that a stable cloud cannot be produced from such an aerosol, while it is difficult to control its transport and formation of an aerosol flux, which entail large losses on the walls of the tubes. In some cases, therefore, the charge and hence the mobility of the droplets must be lowered, sometimes removed completely, or more precisely the aerosol must be brought to an equilibrium charge distribution. An aerosol bearing charges all of the same sign can be neutralised by electrostatic coagulation with oppositely charged particles in the presence or in the absence of an electric field. Such coagulation is more intense only from a certain critical field E_{cr}, which increases with increase in the individual charges on the coagulating particles[49]. Since the individual droplet charges obtained electrohydrodynamically are extremely large, it is pointless in this case to apply an external field to accelerate coagulation.

Several studies of the neutralisation of droplets have used gaseous ions from a corona discharger or a thermal ioniser. Thus the corona discharge with a point–plane electrode system was used[18] to neutralise a cloud of droplets of a polymer solution bearing charges of the same sign. An earthed grid was placed between the corona-displaying point and the capillary to eliminate the effect of the field due to the corona on the field at the end of the capillary. A thermal ioniser—an incandescent tungsten wire—was used[50] to neutralise a flux of charged particles of a water–glycerol mixture, representing the working medium of a colloidal propellent. In this case electrostatic coagulation takes place in the absence of a field.

V. PRODUCTION OF MONOMOBILE AEROSOLS

The need to produce monomobile aerosols—having charges of like sign with a narrow particle distribution function with respect to electrical mobility—arises in many fields of aerosol research as well as in certain special applications of electronic–ionic technology. There is no sharp boundary between mono- and poly-mobile aerosols, as in the case of mono- and poly-disperse aerosols. What aerosol is regarded as monomobile depends on specific requirements. Application possibilities for monomobile and monodisperse aerosols are analogous—calibration of charged-particle counters and spectrometers, investigation of physical processes in charged aerosols, directed deposition of particles in an electric field, etc. Monomobile aerosols located in different mobility ranges are usually obtained by different methods, the chief of which are outlined below.

1. The charging of monodisperse[51] aerosols in the field of a corona discharge (shock charging)[52,53] or in an ionising atmosphere in the absence of an external field (diffusion charging)[54,55]. Many dischargers suitable for the generation of monomobile aerosols have been described[56-62]. Measurement of the particle mobility distribution function

by means of an instrument of high resolving power is the most interesting[62]. Charging by gaseous ions will yield monomobile particles over a fairly wide range of mobilities (0.05–0.0002) with a comparatively high aerosol concentration.

2. The vibrating capillary method with application of a potential to the liquid or induction of charge on the surface of the liquid at the outlet from the capillary. Coarsely dispersed monomobile particles (d = 120 μm) were obtained[63], but no quantitative data were given on their mobility.

3. Preparative spectrometry of charged aerosols. Strictly monomobile latex particles were obtained[64], bearing one or more elementary charges.

4. Electrohydrodynamic atomisation to produce a monodisperse aerosol. This provides a very wide range of particle sizes (~ 0.2–700 μm). The individual charges on the particles are very large, around half the Rayleigh limit. The only reported[43] measurement of the mobility of such particles, in which a modified UT 7003 aspiration ion-counter was applied to a monodisperse dibutyl phthalate aerosol of particle size r = 5–20 μm, showed that a monodisperse aerosol obtained by electrohydrodynamic atomisation is also extremely monomobile, with the particles of this size range having mobilities of ~ 0.2–0.5 $cm^2 V^{-1} s^{-1}$.

VI. APPLICATION OF THE ELECTROHYDRODYNAMIC ATOMISATION OF LIQUIDS

The possibility of using electrohydrodynamic atomisation to obtain monomobile aerosols having like charges from many liquids—including solutions of organic and inorganic substances, volatile and non-volatile liquids, suspensions, and even melts—guarantees a wide range of applications for the method. Discussion is already proceeding on the possibility of using the process in such fields as colloidal rocket propellents[20,40,41,50], microencapsulation[65], microdosage of very toxic substances[66], preparation of emulsions[17,67], demonstration of electrophysical experiments[68], and, in combination with hydraulic[22–24] and disc[25] atomisation, in electrostatic painting. This method shows great promise for (*a*) obtaining very homogeneous, thin coatings from solutions of polymers, oligomers, and also salts and electrolytes, (*b*) calibrating aspiration counters and ion spectrometers, flow-type charge spectrometers, aerosol classifiers, counters of condensation nuclei, photoelectric aerosol counters, and other aerosol instruments, and (*c*) obtaining and investigating individual polymer molecules by the highly dispersed monodisperse atomisation of extremely weak solutions of the polymers.

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