CHARGED PARTICLE RESEARCH LABORATORY REPORT NO. 1-82

ON-DEMAND PRODUCTION OF UNIFORM DT DROPLETS USING
PULSED ELECTROHYDRODYNAMIC SPRAYING

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Scientific Report Prepared for
Lawrence Livermore National Laboratory

by

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MASTER

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A technique suitable for on-demand production of uniform DT droplets is investigated using pulsed electrohydrodynamic (EHD) spraying. Liquid hydrogen is employed as the working liquid, into which charge is injected using a sharp tungsten needle raised to high voltage. By controlling this high voltage, the amount of charge injection required for disrupting the liquid surface into a smooth liquid jet of desired size is determined. For on-demand production of the liquid jet (which breaks up into uniform droplets), high voltage pulses of appropriate height and duration are applied to the charge injection electrode. Results obtained with liquid hydrogen and liquid nitrogen are presented. Considering the potential hazard and scarcity of tritium, the present technique may prove to be particularly useful when there is a need for filling ICF targets with a controlled amount of DT micropellets.

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1. INTRODUCTION

The need for refuelling fusion power plants has resulted in a number of machines for producing fusion fuel pellets.^{1,2} The requirements for refuelling necessitate the production of hundreds of small pellets (~100 μm dia) a second.

Foster, et al.³ mass-produced uniform droplets of liquid hydrogen through acoustically driven breakup of a liquid hydrogen jet. This method allowed one to vary the size of the droplets by changing the jet diameter. The frequency of generation of the droplets is controlled by the acoustic excitation to the jet. Foster's machine was used to study ablation rates by hydrogen pellets injected into a Tokamak-type reactor, but since the drops are produced continuously, he had to devise a gate valve mechanism which allowed single drops into the reactor, synchronously with its operation.⁴

In a modification of Foster's technique, Guttman⁵ produced hollow hydrogen droplets for proposed fuelling of laser fusion power plants. These drops were obtained when a superheated liquid hydrogen jet was cavitated by acoustic (vibrational) excitation.

Woosley⁶ produced charged droplets of liquid hydrogen in an entirely different manner: his method used the natural (unforced) breakup of an electrostatically sprayed jet. The jet forms due to charge injection into the liquid hydrogen. The charge injection electrode is a very sharp tungsten needle; the mechanism is either field emission (if the needle is negative) or field ionization (if the needle is positive). The jet that forms is charged with the same polarity as the potential applied to the needle. The jet is sprayed as long as a steady potential is applied to the needle.

All of the above methods for making fuel pellets produce the fuel continuously. The fuel that is easiest to ignite in a fusion reaction is a mixture of deuterium and tritium; tritium, in particular, is scarce, and one would like to have a way of shutting off the pellet production when fuel is not needed. Stopping the first two machines mentioned in the foregoing is inconvenient at best, because it involves interrupting the flow of liquid hydrogen (or DT mixture).

The electrostatic spraying of droplets,⁶ on the other hand, offers an obvious way of getting fuel only when it is wanted--by pulsing the voltage applied to the needle. Charged droplets have the added advantage that it is possible to accelerate, deflect, and position them using electric fields. In particular, instead of gaseous drag acceleration for injection into a Tokamak, as was done by Foster, et al.,⁴ one could use an electric field. Electrostatic acceleration could make possible higher velocities than gas acceleration.

Hydrogen and its isotopes are liquids only at very low temperatures, about 20 K. In addition, they are very good insulators, containing very low concentrations of free charge.⁷ This means that the conventional techniques of electrostatic spraying, which rely on the residual conductivity of the liquid, are no longer useful, and that a way of introducing charge into the liquid is needed. As was mentioned above, an electrode with a very sharp tip--one micron in diameter or less--at a potential of several kilovolts will produce a high enough field to inject charge even into insulating liquids. This technique was first used by Kim and Turnbull⁸ to spray Freon electrostatically.

The cryogenic nature of the hydrogen isotopes means that the apparatus must have a low heat influx from the room-temperature laboratory

surroundings. For example, a 1 W heat leak will evaporate about $100 \text{ cm}^3/\text{hr}$ of liquid hydrogen.⁹ The thermal insulation is obtained from a vacuum jacket, as in ordinary Dewar vessels. In addition to cooling down the apparatus, there is the problem of providing a high-voltage connection to the spraying site, for which a high-voltage, high vacuum and low temperature feed-through is required.

The apparatus to be described can produce charged liquid hydrogen drops in pulses instead of continuously. The charging is done by field emission (i.e., with the needle being negative) and the resulting droplets are negatively charged. The length of the spraying pulse can be varied, thus changing the quantity of liquid hydrogen sprayed. The injected current can be increased or decreased by raising or lowering the applied voltage. The sprayed droplets are ejected vertically; as a result, there is no liquid flow until the voltage is raised above a critical value, at which point a jet is formed.

Chapter Two contains a description of the pulsed, charged liquid hydrogen pellet generator. The experimental results and their analysis are in Chapter Three. The final chapter is for the concluding remarks and suggestions for future investigations.

2. EXPERIMENTAL APPARATUS

2.1 Introduction

As a preliminary step to working with liquid hydrogen, electrostatic spraying of liquid nitrogen was undertaken. Both pulsed and continuous spraying data were taken. The major differences between the behavior of nitrogen and hydrogen are in the surface tension, the density and, obviously, the boiling point. The surface tension (at the boiling point), density, and the boiling point of liquid hydrogen are 2.0×10^{-3} N/m,¹⁰ 70 kg/m^3 , and 20.4 K, respectively. The corresponding values for liquid nitrogen are 9.0×10^{-3} N/m,¹⁷ 810 kg/m^3 , and 77.3 K. The reason for the preliminary work with nitrogen was to refine the experimental technique, so that when electrostatic spraying of hydrogen was undertaken, results would be obtained more readily. The main advantages of liquid nitrogen are its cheapness and its ready availability.

The experimental apparatus for electrostatically spraying liquid hydrogen and liquid nitrogen consists of two principal parts: the cryogenic chambers, which are central to working with liquefied gases, and the associated electrical systems for producing electrostatic spraying and diagnosing the results. As shown in the block diagram in Fig. 1, the two cryogenic components are the spraying chamber and the hydrogen liquefier. For spraying liquid nitrogen the liquefier was not used, liquid nitrogen being available directly from a dewar, as is seen in the block diagram in Fig. 2, which shows the setup used for spraying liquid nitrogen. (Hereafter, all references to spraying will mention liquid hydrogen only; it is to

Figure 1. Block diagram of the system for pulsed electrostatic spraying of liquid hydrogen.

Figure 2. Block diagram of the system for spraying liquid nitrogen.

be understood that the same holds for nitrogen, unless otherwise noted.) However, liquid hydrogen is not generally available and the liquefier was constructed to produce liquid hydrogen in the laboratory, using liquid helium as the refrigerant. As noted in Ref. 11, liquid helium is much more common in research laboratories than is liquid hydrogen, mainly due to the safety considerations involved in the transportation of the latter. The detailed construction of the liquefier and the spraying chamber is described below. The liquid hydrogen is conveyed to the spraying chamber, where it collects in a reservoir cup. This cup contains the needle, which is the spraying site, and the reservoir nozzle from which the liquid is drawn for spraying. The configuration of electrode and liquid is unlike that used in Refs. 12 and 13. These workers conducted electrostatic spraying downward, that is, with a liquid reservoir above the spraying site. They had liquid flow out of a glass nozzle (in which the charge injection needle is located) in the absence of an applied voltage. After the application of a sufficiently high potential to the needle, a charged jet was formed, which increased the flow through the nozzle. Robinson¹² (and more recently, Kirkbride¹⁴) controlled the flow rate out of the reservoir while no voltage was applied to the needle. In the present experiment, however, there is no flow until a voltage is applied to the needle, to start the spraying. The needle projects upward out of a reservoir of the liquid to be sprayed. The level of the liquid is kept constant around the needle tip by putting the needle inside a small-bore glass tube. The lower end of the glass tube (which is immersed in liquid hydrogen)

is open, so that the tube fills to the top with liquid, which rises by capillary action. This arrangement ensures that disturbances in the liquid reservoir do not affect the liquid around the needle. It is very important to keep the tip of the needle under liquid at all times. The fields at the tip are high enough to cause charge injection into the liquid, so if the needle is exposed to gas, this will certainly cause a glow discharge which immediately dulls the tip and makes charge injection impossible.

When the capillary tube is placed with its opening facing down as in the previous work, a drop collects on the end, and if there is flow through the nozzle, drops periodically fall off. These drops will carry off some charge if there is some charge injection, but not enough for a jet to form. Consequently, there is a fluctuating current until the jet forms. In contrast, in the present experiment there is no such dripping mode, but rather intermittent jets are observed at potentials below those needed for a steady jet, and then a steady jet results as the voltage is increased.

2.2 Pulsed Electrostatic Hydrogen Spraying System

The electrostatic spraying system consists of two chambers: a heat exchanger-liquefier and the spraying chamber. The two are connected by a bayonet coupling for transferring liquid hydrogen from the liquefier to the spraying chamber. Both chambers are vacuum jacketed for thermal insulation. The bayonet coupling makes possible a more flexible design and simplifies disassembly of the system. The separate parts are described below.

2.2.1 Hydrogen liquefier

The heat exchanger used in this experiment is a counterflow heat exchanger of the Linde type¹⁴ similar to a design by Gijnsman and van Tuijl.¹⁵ This heat exchanger is designed to cool down room-temperature hydrogen gas with incoming liquid helium and to exhaust nearly room temperature helium gas. Thus the exiting and incoming fluids are not at widely different temperatures. This makes the best use of the cooling power of the helium. Compared to the sintered copper bead heat exchanger in Ref. 3, the fabrication is simplified.

2.2.2 Construction of heat exchanger

A cross sectional diagram of the heat exchanger appears in Fig. 3, where the constructional details can be seen. The heat exchanger consists of two concentric cupronickel tubes, with a length of 11 feet and outer diameters, respectively, of $\frac{3}{16}$ and $\frac{1}{4}$ inch, coiled into a helix. The wall thickness of both tubes is 0.010 inch. Cupronickel was chosen in preference to stainless steel because of its ductility, which makes its bending easier, and because it is easy to make solder joints to it. The thermal conductivity of cupronickel is comparable to that of stainless steel (0.192 and $0.109 \text{ W cm}^{-1} \text{ K}^{-1}$, respectively between 20 and 300 K).¹¹ Before bending, the tubing must be annealed by heating it red hot with a hydrogen torch; this procedure makes the metal more ductile and prevents tearing of the tubes when they are bent into a helix. After annealing, the smaller O.D. tubing is pushed into the larger. The annular space between the outer and inner

Figure 3. Cross-sectional diagram of the hydrogen liquefier.

tubes is then filled with molten diphenylamine to prevent kinking during bending. The tubing is wrapped around a 2" O.D. mandrel in the form of a close-spaced helix. When the assembly is removed from the mandrel, the turns will spring apart slightly. Adjacent turns must not touch because they are at different temperatures, since there is a temperature drop along the heat exchanger. The diphenylamine is melted out of the helix by immersing it in hot water, and any residue is dissolved out with methanol. The inner and outer tubes are connected to the gas inlets and exhausts with 1/8 inch thin wall cupronickel tubing which is soft soldered into fittings. At the center of the helix is suspended a reservoir for the liquid hydrogen. This reservoir is of coaxial construction, also. It is made from copper tubing and commercial wrought copper fittings. The outside compartment contains the liquid hydrogen and the inner one the incoming cold helium. A copper-constantan thermocouple is soldered with Wood's metal to the outside wall of the reservoir. The temperature of the reservoir is monitored to make sure that the hydrogen does not freeze and block the flow through the heat exchanger. The thermocouple voltage is amplified and connected to a cold junction compensator, and is read on a 4 1/2 digit DVM. The liquid hydrogen comes out of a bayonet coupling at the bottom of the heat exchanger housing and is transferred to the spraying chamber. After the completion of all solder joints, the heat exchanger was leak checked with a helium mass spectrometer leak detector to prevent the possibility of having fine leaks which might

become larger at low temperatures because of different thermal expansion coefficients of the various parts. The entire heat exchanger is suspended from the top cover plate of the vacuum enclosure, making its removal for inspection very easy. Following the suggestion in Ref. 15, only a mechanical pump was used to evacuate the outside enclosure for a pressure of 10^{-3} torr. High vacuum for thermal insulation is automatically obtained as soon as parts of the cooling spirals cool to liquid helium temperatures since any residual gases will condense on the cold surfaces. The sole use of a mechanical pump, not accompanied by a diffusion pump, is another good reason to helium leak check the heat exchanger assembly, because if there should be a helium leak into the vacuum jacket, that helium would not be cryopumped.

2.3 Electrostatic Spraying Chamber

As shown in Fig. 4, the chamber in which the electrostatic spraying takes place was designed with double walls, insulated by a vacuum between them. Since it was anticipated that very high voltages (up to 20 kV) would be necessary for spraying liquid hydrogen, a reliable method was needed for introducing this high voltage into the inner chamber. A high-vacuum feedthrough mounted on the chamber wall would seem to be the obvious choice, but the number of this type of feedthrough are severely limited. Past experience had shown that a Kovar-glass soft-soldered feedthrough will crack and leak after thermal cycling. The only reasonable choice was, therefore, a ceramic-to-metal

Figure 4. Cross-sectional diagram of the chamber in which electrostatic spraying of liquid hydrogen and nitrogen is done.

seal which is mounted on a standard Varian UHV flange. This type of feedthrough has performed admirably, showing no signs of leaking at liquid hydrogen temperatures and at voltages up to 20 kV. In addition to these merits, the feedthrough is demountable, making removal of the inner chamber easy. The center conductor of the feedthrough projects into the inner chamber, and on its end is suspended a copper cup which holds the tungsten needle and acts as a reservoir for the liquid hydrogen. The cup is held onto the center conductor by a short length of tube which is a push fit onto the conductor rod. The liquid hydrogen flows down into the cup from an eighth-inch cupronickel tube which is connected to the bayonet coupling in the top plate of the spraying chamber enclosure. This tube is positioned so that the liquid will drip into the cup; if the cup is filled with liquid, the excess collects at the bottom of the chamber. The copper cup is thus at the same potential as the voltage connected to the feedthrough and has to have rounded edges to prevent electrical breakdown. The assembly holding the needle is screwed into the bottom of the cup. A detailed drawing appears in Fig. 5. The holder consists of two coaxial screws; the outer is a 3/8 inch long 4-40 machine screw and is drilled and tapped through the center for a 0-80 machine screw. The smaller screw positions the needle vertically. The larger screw is screwed into the bottom of the spraying cup and into its top end is glued a glass capillary tube, made from commercial 100 microliter micropipettes. This source of glass capillaries was chosen because the pipettes have extremely uniform diameter and wall thickness, making it easy to

Figure 5. Holder for the tungsten needle electrode.

replace them if they should break. The top end of the glass tube, at which spraying will take place, is necked down by heating with a hydrogen torch while the tube is turned on a glass lathe. The necked portion is scribed at a suitable inside diameter and is broken. Then the other end is broken off at a length which will allow the top to project out of the cup by 3 mm.

The tungsten needle is made from 12 mil tungsten wire, which is pushed into a 12 mil ID stainless steel tube which, in turn, is a push fit into a drilling in the C-80 machine screw. The needle is made by etching the tungsten wire in 0.5 M sodium hydroxide solution with an AC voltage of 2-5 volts. The etching procedure is as follows: after the tungsten wire is mounted into the screw, it is etched to a diameter of 1-2 mils; then the wire and screw are inserted into the 4-40 screw and the thinned wire is carefully threaded into the capillary tube from which it projects by about 1 cm. The final sharpening is done with the wire inside the capillary. The needle tip is examined under a microscope periodically during the etching, and when the tip is too small to be resolved with the microscope, etching is stopped. The needle is retracted into the capillary so that the tip is 25 microns below the plane of the capillary opening. This distance assures that the tip will be covered by liquid and will not cause a discharge which destroys the tip. After etching is complete, the needle and capillary are screwed into position into the copper cup. The cup is then pushed onto the end of the feedthrough center connector.

The bottom of the spraying chamber is sealed off with a removable flange. As mentioned above, the spraying chamber may be required to hold liquid hydrogen. Thus, the seal must not leak at this low temperature--20 K--and must be easily made and broken. An indium wire compressed between two flat surfaces is a very reliable seal under these conditions. The seal is a .040 inch indium wire ring gasket. The cover flange is made of stainless steel to minimize leaks from thermal expansion and contraction. The indium is held in place on the flange by a shallow groove, .025 inch wide x .015 inch deep. Access to the copper cup is through this flange opening.

There are two windows on the inner chamber and two more on the outer vacuum jacket. The inner set of windows gets very cold, as mentioned above, and accordingly, it is sealed with indium gaskets. The window seat and holddown flange are both machined from stainless steel and are soft soldered to the chamber wall. The windows are made large enough to be able to photograph the spraying.

2.4 Electrical Apparatus

Since both pulsed and continuous spraying was done, there are slight differences in the equipment used. Common to both types of spraying was the high-voltage source which is a Hipotronics 0-50 kV DC power supply. For continuous spraying, the power supply was connected directly to the Varian high-voltage feedthrough. The voltage is then increased until spraying is observed while viewing through the microscope. The voltage is read from the meter on the power supply. Since the needle is connected directly to the voltage source, the tip voltage is known. The

sprayed droplets are collected on a flat plate which is above the needle at a distance of 13 mm. This distance proved to be small enough so that the drops would be collected, but far enough from the needle top to prevent arcing from the tip to the plate.

Previously, a distance of 3 mm had been tried, which resulted in an arc between the needle and plate and destroyed both needle and capillary. The collection plate is connected to a Keithley 410 micro-micro ammeter which measured the current carried by the sprayed drops. This location for a current measurement places the ammeter at a low voltage instead of at the power supply voltage, as would have been the case if the ammeter had been inserted between the power supply and the needle. The connection between the plate and the ammeter is made by a wire which is fed through a 1/8 inch stainless steel tube running from the inner chamber to the top flange of the vacuum jacket. The seal between wire and tube is made by running Hysol 3x gray epoxy into the tube for a distance of about 3 mm. The cable leading to the micro-micro ammeter is double shielded coax to minimize extraneous signal pickup. When no voltage is applied, the typical currents measured are less than 1×10^{-11} A. The currents due to the collected droplets range from 10^{-9} to 10^{-7} A, thus ensuring that the desired current is not masked by noise from external sources. Since the voltage drop across the micro ammeter is only one volt, and one terminal is grounded, the charge collection plate is essentially at ground potential compared to the several kilovolts that are on the needle. As a result, charged drops which leave the needle are attracted to the plate and deposit their charge on it. The collection efficiency is good (i.e., no charge is lost) as

can be seen from the voltage-current plots in Chapter 3. The current is proportional to the square root of the voltage, as in Ref. 16. Had some charge been lost, it is doubtful whether a proportionality would have been found in a consistent manner and from different runs.

For the pulsed electrostatic spraying, a generator of rectangular variable-duration high-voltage pulses was needed. After investigation, a thyratron circuit was rejected because it did not produce long enough pulses and the pulse shape was exponential rather than rectangular. In addition, the maximum voltage rating of the 5C22 thyratron is 16 kV which is rather less than the desired 20 kV. The thyratron circuit basically discharges a capacitor to produce a negative going spike. The discharge time constant of the capacitor is determined by the capacitance in the circuit and the holding current of the thyratron.

The method used in this experiment to generate high voltage pulses was to drive a high-voltage vacuum relay with a pulse generator. The relay is a single-pole, double-throw type with one stationary contact connected to ground and the other, also stationary, to the high voltage power supply. The center contact is normally connected to ground, but when energized, it makes contact with the high voltage. Breakdown between the high voltage and ground contacts is prevented by placing them in high vacuum inside the glass relay envelope. By applying a pulse to the relay coil, one gets a single high-voltage pulse which starts at ground and goes to the power supply voltage and back to ground. The pulse rise time is fast because the voltage rises to the power supply voltage when the relay contacts close, which occurs in a very short time. Measured rise times were about 1 microsecond. On

"make," there is no contact chatter. On "break," the contact did bounce for about 200 microseconds. The relay coil is a 24-volt DC, 60-ohm unit. It was driven by a power transistor in series with the coil and a 24-volt battery. A positive trigger pulse at the transistor base saturates the transistor and lets current flow through the relay coil. The transistor trigger pulse is obtained from a Tektronix 161 waveform generator, from the "gate" output. The gate width is continuously adjustable from 10 s to 0.1 msec. The relay pulse is shorter than the gate pulse because of the mechanical inertia of the contact. The shortest pulse available from the relay is 2 milliseconds wide. Because the relay pulse is somewhat shorter than the gate pulse, the length of the pulse applied to the needle was measured with a Tektronix P6015 1000x probe and the width was read from a storage oscilloscope trace. The 1000x probe has an input impedance of 100 megohms. At 10kV it draws 10 μ A, large compared to the typical currents of 10^{-8} A measured for spraying.

2.5 Photographic Technique and Speed Measurement of the Charged Droplets.

The spraying is observed through a Bausch and Lomb zoom stereomicroscope which has a maximum magnification of 70x. Illumination is provided by a General Radio xenon strobe lamp. During visual observation, the strobe is triggered by a GR oscillator at a frequency comfortable for viewing. For taking photomicrographs, the strobe is flashed once, and is triggered either manually or by a pulse generator, as explained later. The microscope is equipped with a camera adapter and the magnification is 7.5 times the zoom setting. This combination

of camera and zoom microscope is very convenient because it allows one to change the magnification on the photograph at will. In addition, the depth of field can be controlled by changing the magnification.

The speed of the charged droplets was measured by taking double-exposure photographs. The speed measurement is valuable because it gives a value for the flow rate of the liquid being sprayed, and when used along with the current measurement, gives the charge on the droplets. The double exposure photographs were taken by flashing two strobe lamps at a known interval and adjusting their intensity to give a distinct image for each flash. The images of the moving droplets should not be too widely separated because this makes it difficult to identify the images of consecutive droplets in a stream.

A block diagram of this arrangement used to trigger the strobe lamps is shown in Fig. 6. The Tektronix R116 pulse generator is in the "single delayed pulse" mode and is triggered manually by a pushbutton, and delivers a short pretrigger pulse to trigger the oscilloscope. The scope sweep starts, generating a coincident positive gate pulse which is used to trigger the first strobe lamp. The second strobe lamp is triggered by the delayed pulse from the R116 generator. The two strobe lamp flashes are combined by a beamsplitter, which also directs part of the light into a photodiode. The photodiode signal is displayed on the scope, giving a check on whether both strobes fired and on their intensities. The spacing between the light flashes is controlled by the delay setting of the pulse generator. Because of the delays in triggering the strobe lamp, the spacing between light flashes is slightly less than the delay between pretriggering and delayed pulses. This dif-

Figure 6. Block diagram of the electrical apparatus used to make double-exposure photographs of charged droplets.

ference is $0.5 \mu\text{s}$ and is insignificant for delays over $20 \mu\text{s}$ (i.e., the flash spacing can be taken to be equal to the pulse generator delay setting).

3. EXPERIMENTAL RESULTS

3.1 Experimental Procedure

The procedure for spraying liquid hydrogen is as follows. The vacuum jackets of the liquefier and the spraying chamber are evacuated with a 500 l/min mechanical pump. The pressure in the vacuum jacket before cooldown is at the low-pressure limit of the pump, about 5×10^{-4} torr. The helium line of the liquefier is flushed with helium gas by flowing helium into the exhaust port and exhausting it at the inlet. Flushing is done to make sure that there is no air or water vapor in the tubes carrying cold helium vapor and/or liquid helium. These impurities would freeze at the low temperatures involved, blocking flow through the heat exchanger, and requiring an interruption of the experiment to unblock the passages involved. Similarly, the tubes carrying gaseous and liquid hydrogen, and the spraying chamber are both flushed with ultra-pure hydrogen. The spraying chamber is evacuated to a pressure of ~ 20 mtorr, then filled with hydrogen gas; the procedure is repeated, and hydrogen is finally flowed through the chamber and out of the exhaust.

After the completion of purging of the low-temperature fluid paths, cooldown of the heat exchanger is begun. The liquid helium dewar is pressurized to 4 psig to send liquid helium through the liquefier. The helium exhaust is throttled with a valve, to make sure that the hydrogen passages in the heat exchanger do not freeze. At the same time, hydrogen gas is allowed to flow through the heat exchanger and the spraying chamber. This hydrogen is exhausted to the atmosphere. The pressure of hydrogen entering the liquefier is regulated to 3 psig. Flow is measured with a Dwyer flowmeter calibrated for air, and it is controlled with a needle valve upstream from the flowmeter. During

cooldown, the temperature of the hydrogen reservoir is closely monitored, to ensure that the heat exchanger tubes are not blocked by frozen hydrogen. One sign that this has happened is that the hydrogen reservoir temperature drops below the boiling point of hydrogen, but no liquid comes out. When the frozen hydrogen melts (and subsequently evaporates), the pressure in the hydrogen lines can rise drastically, and has to be relieved.

Once liquid hydrogen has been obtained, the flow of helium gas can be varied slightly to either cool or warm the liquid in the reservoir. The liquid hydrogen flows downward out of the liquefier and into the reservoir cup in the spraying chamber. It collects in the cup, boiling until the cup cools to liquid hydrogen temperature. After equilibrium has been reached, the evaporation rate of hydrogen in the cup is less, and the excess incoming hydrogen simply spills out of the cup onto the bottom of the spraying chamber.

The capillary tube fills with liquid, which may bubble if the liquid in the cup is boiling. When the hydrogen and its surroundings are cold enough, there is a stationary liquid surface covering the tungsten needle. Voltage is now applied to the needle while observing it through the microscope. It is noticed that, at voltages too low to initiate a jet, the surface of the liquid is pulled up into a convex shape because of the electrostatic force on the surface. The needle injects some charge, but not enough to cause spraying. As the voltage is increased, brief unsteady jets are formed. Finally, with a high enough voltage (and a correspondingly higher injected current), a steady, continuous jet is formed. This jet can now be photographed using either a single flash from the strobe lamp, or a double flash, for speed measurement.

The procedure for a single flash photograph is as follows: the camera shutter is opened manually, the strobe is flashed once (again manually), and the shutter is closed. The collected current at the time of the photograph and the needle voltage are both recorded. For a speed measurement (double flash photograph), the procedure is almost as simple: referring to Figure 6, Chapter 2, the two strobes are flashed in sequence by triggering the Tektronix pulse generator. The shutter is again held open manually. Even though the film is exposed while the shutter is open, so little extraneous light enters the microscope when the strobe is not on that there is no fogging of the film.

For obtaining pulsed spraying, the high voltage power supply is connected to the high voltage relay mentioned in Chapter 2, and thence to the needle. The relay is closed and opened by a single pulse from the Tektronix 161 waveform generator. The waveform generator is triggered manually, but it may be operated repetitively. The upper frequency limit is set by the repetition rate of the relay, which is 50 contact closings/sec. In the present experiment, only single pulses were desired. The strobe flash for photographing pulsed spraying is synchronized with the spraying pulse by using an oscilloscope probe to detect the high-voltage pulse, trigger the oscilloscope, and then use the scope "gate" output to fire the strobe lamp. The "gate" pulse is a positive going pulse coincident with the start of the scope sweep waveform.

3.2 Current-Voltage Characteristics of Continuous Electrostatic Spraying

The current collected by the plate and the spraying voltage were plotted for both liquid nitrogen and liquid hydrogen continuous jets.

As noted by Halpern and Gomer,¹⁶ the current injected into these liquids by field emission is space-charge limited for current greater than 10^{-10} A. This means that the voltage and current are no longer related by the Fowler-Nordheim equation, which gives an exponential dependence of current on voltage; rather, the current follows a power law dependence on the voltage. Since all of the collected currents are larger than this, it was assumed that the emission in the present experiment is also space-charge limited. The voltage-current relation for space-charge limited field emission is¹⁶

$$V = V_{\text{vac}} + 2 \left(\frac{8R}{3\epsilon\epsilon_0} \right)^{1/2} I^{1/2}$$

where R = needle tip radius

α = solid angle into which emission takes place

ϵ = dielectric constant of the liquid

u = ion mobility

V_{vac} = voltage necessary to obtain I in a vacuum

Thus, it is seen that for space-charge limited emission, the plot of V vs. $I^{1/2}$ should be a straight line with intercept V_{vac} . Note that the slope of the line is proportional to the square root of the tip radius, so that different tip sizes will, in general, give different slopes for the voltage-current plot.

The square root of the collected current as a function of the voltage applied to the needle is plotted in the graph in Figure 7, for two separate runs with liquid nitrogen. In Figure 8, the corresponding data for continuous liquid hydrogen spraying are plotted. The slopes of the two lines for nitrogen are 5.3×10^4 kV/A^{1/2} and 4.5 kV/A^{1/2}; for the hydrogen plot, the slope is 5.1×10^4 kV/A^{1/2}. Halpern estimates the mobilities in H_2 at 21 K and N_2 at 77 K to be 3×10^{-2} cm²/V-s and

Figure 7. Plot of V vs $I^{1/2}$ for continuous spraying of liquid nitrogen at 77 K. Data from two experimental runs are plotted.

Figure 8. Plot of V vs $I^{1/2}$ for continuous spraying of liquid hydrogen at 21 K.

$8 \times 10^{-3} \text{ cm}^2/\text{V-s}$, respectively. Using these values, the ratio (hydrogen plot slope/nitrogen plot slope) is .5, if the emitter tip radius is the same. In the measured data the ratio of the slopes is close to 1, and in addition, the slope of the hydrogen plot is higher. Obviously, the tip radius in this case was larger than when nitrogen was sprayed.

The usefulness of collecting data on current-voltage characteristics is that this eliminates one variable--either voltage or current--from consideration. Experimentally, one controls the voltage, which then determines the injected current. On the other hand, the current is a more desirable parameter to measure, because it determines the specific charge on the electrified jet.

3.3 Field Emission Spraying Data

A series of photographs showing the course of an experimental run as the injected current is increased, is shown in Fig. 9, for continuous spraying of liquid nitrogen. In a, the injected current is too low to drive a jet, and the surface of the liquid is merely pulled up by the electric field, as mentioned in Chapter 2. In Fig. 9b, we see a smooth jet which is characteristic of operation with a not-too high injected current. In Fig. 9c, the current has been increased, and the jet is breaking into drops sooner than in photograph 9b. Figure 9d shows (at higher magnification) a highly charged jet with a large instability tending to move the jet laterally, as described by Taylor¹⁷. The current increases from $1.2 \times 10^{-8} \text{ A}$ in photo 9b, to $6.0 \times 10^{-8} \text{ A}$ in photo Figure 9d.

In comparison, two photographs of continuous liquid hydrogen spraying

Figure 9. Continuous electrostatic spraying of liquid nitrogen. Photo a) shows the displaced liquid surface with an injected current which is too small to drive a jet. In b) and c), we see a smooth 18 μm diameter jet. Photo d) shows a high current 30 μm diameter jet with a large lateral instability. Magnification is 22.5x in a), b), c), and 30x in d).

a)

b) $V=8.25\text{kV}$, $I=1.25\times 10^{-8}\text{A}$ c) $V=12\text{kV}$, $I=3.9\times 10^{-8}\text{A}$ d) $V=15\text{kV}$, $I=6.0\times 10^{-8}\text{A}$

are shown in Fig. 10. The voltage is 4 kV, which is significantly lower than the voltage needed to spray liquid nitrogen. The jets shown are shorter than liquid nitrogen jets, because of the lower surface tension of hydrogen. The jet size variation is due to a pressure fluctuation in the capillary tube, from a bubble forming.

Pulsed spraying of liquid nitrogen is shown in Fig. 11. The electrode voltage increases from 15 kV to 21 kV. A jet was first observed at 14 kV. The minimum pulse width at 14 kV was 15 ms, decreasing to 10 ms at 21 kV. The photographs in Fig. 11 show pulsed spraying at pulse lengths close to, but not equal to the minimum length. The pulsed jets that are formed are highly perturbed, no doubt because of their transient nature. The jets are beginning to break up into drops. The wavelength of the disturbance is about five times the jet diameter, which is close to the Rayleigh wavelength for the most rapidly growing disturbance on a liquid jet.¹⁸

By contrast, pulsed liquid hydrogen jets are much smoother and break up into uniform charged drops. These jets are shown in Figure 12. The pulse width was kept constant at 18 ms in this series of photographs, and the voltage was varied. As with continuous spraying, typical voltages required for jet formation are lower than those for liquid nitrogen. The thinner jets are again believed to be caused by a sudden pressure decrease in the capillary tube because of the collapse of a bubble. The pulsed hydrogen jets and the drops produced from them are identical to the continuously sprayed jets, and exhibit none of the turbulence observed in pulsed nitrogen jets.

The two photographs in Fig. 13 show a smooth liquid nitrogen

Figure 10. Continuous spraying of liquid hydrogen. Photo a) shows a 29 μm diameter jet, and b) shows a 13 μm diameter jet, both at the same voltage. The jet diameter in b) is reduced due to a lowering of the flow rate following the collapse of a bubble in the glass nozzle. Magnification = 37.5x.

a) $V=4\text{kV}$, $I=1.7 \times 10^{-8}\text{A}$.

b) $V=4\text{kV}$

a) $V = 15\text{kV}$, $t_p = 18\text{ms}$

b) $V = 16\text{kV}$, $t_p = 18\text{ms}$

c) $V = 18\text{kV}$, $t_p = 18\text{ms}$

d) $V = 21\text{kV}$, $t_p = 12.5\text{ ms}$

Figure 11. Pulsed spraying of liquid nitrogen at short pulse durations. Magnification = 37.5x.

a) $V=4.7\text{kV}$, $19\text{ }\mu\text{m}$ jet dia.

b) $V=4.7\text{kV}$, $27\text{ }\mu\text{m}$ jet dia.

c) $V=5.5\text{kV}$, $24\text{ }\mu\text{m}$ jet dia.

d) $V=7.5\text{kV}$, $56\text{ }\mu\text{m}$ jet dia.

Figure 12. Pulsed spraying of liquid hydrogen. Pulse width = 18ms. Magnification=37.5x.

a) LN_2 , $V=13.2\text{kV}$, $27\mu\text{m}$ jet dia.

b) LH_2 , $42\mu\text{m}$ jet dia.

Figure 13. Liquid nitrogen and liquid hydrogen jets.
Magnification=37.5x.

jet at higher magnification, and a liquid hydrogen jet breaking into drops. Both jets are continuous. The quality of liquid nitrogen droplets at a point downstream from the end of the jet is seen in the photographs in Fig. 14. The drops are not all identical because more than one wavelength exists on the nitrogen jet, producing different drop sizes upon breakup.

3.4 Specific Charge Measurement

For obtaining the charge carried by the jet, the speed of the charged droplets resulting from jet breakup was measured. Two double-exposure photographs of charged droplets are shown in Fig. 15. The spacing between the drops increases with increasing distance from the end of the jet for two reasons: the drops repel one another because they are charged with the same polarity, thus increasing interdrop distance, and because of the acceleration by the electric field which is between the liquid surface and the charge collection plate. The surface charge on the droplets is calculated from the injected current and the flowrate. The droplet speed in photograph (a) increases from 2.0 to 2.8 m/s as the drop travels from the end of the jet, at the bottom of the picture, to the top of the picture, which is about 1 mm above the end of the jet. The droplet speeds are almost the same in photograph (b). The speeds were calculated from the measured drop spacing in the two exposures and the known time interval between the *strobe flashes*. The surface charge carried by the jet is related to the current by

a) $V=11.3\text{kV}$, $I=1.25\times 10^{-8}\text{A}$

b) $V=13.3\text{kV}$, $I=1.4\times 10^{-8}\text{A}$

Figure 14. Charged liquid nitrogen droplets resulting from the breakup of a jet. Magnification=45x.

a) 50 μ s flash spacing, jet speed=1.3m/s,
37 μ m jet dia.

b) 30 μ s flash spacing, jet speed=1.7m/s,
29 μ m jet dia.

Figure 15. Double-exposure photographs of liquid hydrogen droplets. The closely spaced drops are the images of a single drop exposed twice. Magnification=37.5x.

$$\sigma_s = \frac{I}{2\pi r_j v_j} \text{ coulombs/cm}^2$$

where r_j = jet radius

v_j = jet velocity

From the Rayleigh formula for the most unstable wavelength for jet breakup, $\lambda = 10r_j$, we get the average size of the drops on jet breakup:

$$\frac{4}{3}\pi r_d^3 = r_j^2 (10r_j) \cdot \pi$$

$$r_d = 1.95 r_j$$

The subscript d refers to the quantities pertaining to droplets.

This relationship is also observed experimentally in the photographs, so that we may take the droplet radius to be twice the jet radius.

Then the charge per drop is

$$q = 2\pi r_j \cdot (10r_j) \sigma_s = 10\pi r_j^2 \sigma_s$$

or, in terms of the measured quantities,

$$q = \frac{10Ir_j}{v_j}$$

The measured specific charges for liquid hydrogen droplets are summarized below:

r_j (μm)	v_j (m/s)	I (A)	q (coul)	q_{max} (coul)
20	2	7×10^{-9}	7.0×10^{-13}	9.3×10^{-13}
16	5	3×10^{-8}	9.6×10^{-13}	6.6×10^{-13}
17	6	4×10^{-8}	1.1×10^{-12}	8.9×10^{-13}

The observed jet diameters ranged from a low of 13 μm to a maximum of 56 μm , with the majority of jets being in the 25-40 μm range.

As noted above, the droplet spacing is not uniform after they become detached from the end of the jet. The spacing increases, and it was decided to plot the square of the droplet speed against the distance from the end of the jet. If the droplets are uniformly accelerated by a constant electric field, the plot should give a straight line. The plots for four different jet diameters are shown in Fig. 16. The smaller droplets (resulting from smaller diameter jets) are seen to be travelling faster than the larger droplets.

Curiously, the jet diameter seemed to stay almost constant as the injected current was increased over a large range (four to six times the minimum current needed for spraying). Previous theoretical analyses^{12, 13} had all indicated that the jet radius should vary as $I^{-1/3}$, for a constant flow rate. These analyses had not included the effects of electrical force on the charged liquid surface exposed at the capillary opening. In this experiment, it was observed that there is a significant force exerted on the liquid surface, as shown in Fig. 9(a). The effect of this upward force when a jet is being sprayed is to increase the flow rate. Robinson's expression for the jet radius¹² contains the flow rate as the most important parameter in determining the jet radius:

$$r_j = \frac{\rho \mu}{4\pi^2 r_t} \frac{Q^{5/6}}{V^{1/6} I^{1/3}}$$

where ρ = density of fluid

μ = ion mobility

r_t = electrode tip radius

Q = volume flow rate

V = electrode voltage

I = injected current

Figure 16. Plot of v^2 of droplets vs distance from the end of the jet for liquid hydrogen.

The observed insensitivity of jet radius to the injected current could be explained by a smaller increase in the flow rate with increased current. Since the flow rate enters in the expression for jet radius as the $5/6$ power, while the current enters only as the $-1/6$ power, one can see that the flow rate would have to increase only slowly, in order to offset a much larger increase in the injected current, and, thus, keep the jet radius constant.

4. CONCLUSION

The experimental work presented here has shown the possibility of producing uniform charged liquid hydrogen drops in short pulses on demand. The quality of pulse-sprayed drops is essentially the same as that of continuously sprayed ones. The minimum spraying pulse width is 8 ms at the lowest voltage that could drive a jet. The droplet radii range from 13 to 55 μm in diameter. At high voltages, corresponding to high injected currents, continuously sprayed drops are charged to the Rayleigh limit, within experimental error.

In common with previous investigations, it was found that the flow rate of the liquid hydrogen is an important parameter in determining the jet diameter and thus the droplet size. However, the expected jet diameter decrease with increased injection current was not observed. In the present experiment the flow through the nozzle is due entirely to charge injection, because spraying is directed upward; in previous work the nozzle faced downward and there was a pressure-driven flow rate even in the absence of charge injection. The configuration described here should enable one to evaluate the influence of the electrohydrodynamic effects only, isolated from pressure effects. Future experiments should investigate whether a restriction on the nozzle exit diameter would produce a jet size reduction with increasing current, because of the flow rate restriction. A new feature of this experiment is that there is no liquid flow for voltages below a critical value, at which point a jet is sprayed. Thus, there is no dribbling or intermittent jet mode as reported earlier.¹³

The current-voltage relationship for continuously sprayed jets can be fitted by a square-law dependence of the current on the applied voltage, indicating that space-charge limited emission takes place. This result enables one to eliminate the voltage from the electrical equations describing steady-state spraying. Robinson's analysis¹² failed to reveal such a dependence; instead, he fitted his current-voltage relation to the Fowler-Nordheim equation for field emission.

The present experimental apparatus could be improved by adding cooling to the interior enclosure in which spraying is done. In this way, one could avoid the thermal fluctuations encountered in spraying liquid hydrogen. The cooling can be readily implemented by soldering copper tubing coils to the chamber walls and running cold helium gas through.

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