Factors affecting the atomization of saturated liquids

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FACTORS AFFECTING THE ATOMIZATION OF SATURATED LIQUIDS

BY

G. E. FLETCHER

A DOCTORAL THESIS SUBMITTED IN
PARTIAL FULFILMENT OF THE REQUIREMENTS FOR
THE AWARD OF
DOCTOR OF PHILOSOPHY OF THE LOUGHBOROUGH UNIVERSITY OF TECHNOLOGY.

DEPARTMENT OF CHEMICAL ENGINEERING
FEBRUARY, 1975

DIRECTOR OF RESEARCH
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SUPERVISOR
B. SCARLETT M. SC.

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ACKNOWLEDGMENTS

I would like to thank Riker Laboratories Limited, now part of the Minnesota Mining and Manufacturing Company, for financial support during this project.

I wish to thank also Professor D. C. Freshwater for provision of research facilities and B. Scarlett for supervision throughout the project.
SUMMARY

The atomization of a liquified gas propellant, as a means of dispersing a powdered drug or non-volatile solute, was investigated. Atomization was achieved by passing the propellant through a two-orifice nozzle assembly. A number of properties of the system were shown to be predictable with reasonable accuracy, in terms of the nozzle dimensions and thermodynamic properties of the propellant, together with minor empirical factors. The properties that could be predicted were the mass flow-rate, the pressure of the propellant in the expansion chamber between the two orifices, the quality, or mass fraction evaporated, of the propellant in the expansion chamber, and the initial velocity of the spray. By application of the principle of momentum conservation the axial velocity decay of the gaseous component of the resultant spray and to a certain extent the particulate component of the spray could also be predicted.

In addition to the above fundamental relationships, an empirical expression for the mass median diameter of the residual aerosol of a non-volatile solute dissolved in the propellant was determined.

Information thus obtained is of assistance in the optimisation of the design of liquified gas aerosol generators as a means of administering a drug by inhalation.
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1. INTRODUCTION

The purpose of the work described in this thesis was to investigate the operation of liquified-gas aerosol generators as a means of dispersing a powder and administering it into the deeper recesses of the respiratory tract; the powder of interest was a metered dose of a micronised drug. The size distribution of the drug necessary to achieve optimum administration is not known precisely, but it is generally agreed (8,25,42) that because of deposition by impaction and sedimentation the probability of particles of diameter above 3 to 5 μm reaching the required area of the lungs is low. Further there is a diameter of minimum deposition which represents particles too small for deposition by impaction or sedimentation but too large for deposition by the alternative means, which is diffusion; this diameter is about 1/2 μm. Thus a suitable size distribution may well be one in which the mass of the drug is concentrated in the above particle size range, 5 μm to 1/2 μm.

It is possible to produce the drug in powdered form, whose size distribution corresponds approximately to that given above. However, it is found that generally the size distribution of the drug is not reproduced in either that of the emitted spray or the residual aerosol. The latter distributions are generally coarser than that of the drug, and therefore efficiency of deposition is impaired. The reason that the emitted spray is coarser than the drug is that the spray is in the form of incompletely evaporated propellant droplets which tend to be larger than, and contain, the drug particles. The reason that the residual aerosol may be coarser than the drug is two-fold. Firstly, each emitted droplet may contain more than one drug particle; this tendency will depend on the size distribution of the spray droplets, and the concentration of the drug for a given drug size distribution. The second reason is that a non-volatile surface active agent is included in the formulation. Thus the residual
Aerosol will consist of one or more drug particles in a surfactant droplet; there may of course be surfactant droplets with no drug. The reason for including the surfactant in the formulation is to ensure that the drug does not coagulate prior to use, and to ensure smooth operation of the special metering valve. The relative concentrations of the drug and surfactant are typically 0.2% and 1% respectively.

A further point may be made regarding the position of deposition of the drug in the respiratory tract. This is that the velocity of the spray in the vicinity of the nozzle is much greater than that of inspired air in the region of the throat; the former is of the order of 30 m/sec, the latter of the order of 1.5 m/sec (35). This means that the probability of a particle impacting on the back of the throat is greatly enhanced. It follows therefore the velocities of the particles as well as their diameter are of interest in this study. A measure of the relative importance of the size and velocity of a particle is given by a dimensionless impaction parameter (\( \psi \)). This is defined as the ratio of the particle stop distance to the spray diameter, and the greater its value the greater is the probability of impaction. It is given by:

\[
\psi = \frac{1}{18} \frac{(\text{particle diameter})^2}{(\text{particle velocity})(\text{particle density})} \left( \frac{\text{fluid viscosity}}{\text{spray diameter}} \right)
\]

The relative importance of the various spray parameters, especially diameter which is raised to the power 2, is seen from this expression.

In principle an arrangement of baffles could be the solution to many of the problems associated with aerosol spray therapy. This is because a baffle could remove undesirably large particles and also destroy the momentum of the spray. However it is desirable that the aerosol generator should be inconspicuous and fast in operation; it should remain clean and not...
become coated with spray deposit. This means that baffles or a large adapter designed to permit attenuation of the spray are not acceptable.

It is clear from the above discussion that optimisation of the design of therapeutic aerosol generators is a multi-facet problem. Worthwhile areas of investigation could have included the study of the flow of fine suspensions through orifices, atomization of liquified gases and subsequent spray development, or the aerodynamics of particle flow in regions of restricted geometry. It was considered that the most profitable area of study would probably be that of atomization and subsequent spray development. This study was thus pursued. The first part was preliminary experimentation in which commercially produced metered-spray generators and equipment already available were used. From this work it was found that because of the transient nature of the metered spray, which was of short duration, its behaviour was not readily amenable to mathematical description. Therefore after the preliminary studies had been carried out a continuous-flow rig was built and further work carried out on this. Finally, information obtained from the continuous-flow rig was related where possible to the metered spray generator.

Throughout this thesis, halogenated hydrocarbon propellants (or refrigerants), for example propellant 12, are identified by an internationally adopted number coding system standardised by the American Society of Refrigerating Engineers in 1957 (Standard Code ASRE 34).

The A.S.R.E. number system for chemically saturated chlorofluorohydrocarbons is related to their chemical formulae in the
following way:—

Reading from right to left

First number = number of fluorine atoms
Second number = number of hydrogen atoms plus one
Third number = number of carbon atoms minus one

Thus, propellant 11 is \( \text{C}_3\text{Cl}_3\text{F} \) or trichloromonofluoromethane. Propellant 12 is \( \text{C}_2\text{Cl}_2\text{F}_2 \) or dichlorodichloromethane. Propellant 11\(_4\) is \( \text{C}_2\text{Cl}_2\text{F}_4 \) or dichlorotetrafluoromethane.

Mention may also be made of the use of mathematical symbols. Owing to the limitations of the typewriter, consistency in the mathematical symbol for multiplication was difficult to maintain. Thus "x" was used in the absence of the algebraic symbol "\( x \)" for distance, otherwise brackets or a point were used. Similar difficulty was experienced with the differential operator "d" and the algebraic symbol "\( \text{d} \)" for particle diameter. In the presence of the latter, a derivative was placed in brackets.
2. PRELIMINARY INVESTIGATIONS

2.1 Mechanism of atomization
2.2 Pressure and temperature measurement
2.3 Measurement of size and velocity of in-flight spray droplets
2.4 Measurement of size distribution of the residual aerosol
2.5 Conclusions

An initial survey of the literature indicated that there had been little investigation into the basic mechanisms of the atomization of the saturated liquids. However, much work had been done on the analysis of what is often the end product, an aerosol of fine solute particles.

In this work, preliminary experimentation was carried out with the purpose of obtaining a qualitative, and to a certain extent quantitative, concept of the mechanism of atomization. Also, the feasibility of various methods of particle size analysis as applied to aerosols was assessed. These tests were carried out in a laboratory maintained at a temperature of about 15°C to 20°C.

2.1 Preliminary investigations into the mechanism of atomization

Initial investigations were carried out on sprays generated by a metering device. This allowed 25mm$^3$ (or 50mm$^3$) of a liquified gas propellant to pass through a nozzle in a high-density polythene adapter, a section of which is shown in Figure 2.1. The propellant consisted of, by weight, 50% propellant 12, 25% propellant 11 and 25% propellant 114, into which was dissolved 1% by weight of the surface-active agent, sorbitan trioleate ("Span 85"). At 20°C the saturated vapour pressure of the propellant was approximately 300 kN/m$^2$ gauge (43 p.s.i.g.). Because the molecular weight of the sorbitan trioleate (962) is high compared with propellants 12, 11 and 114 (121, 137 and 171 respectively), the introduction of the solute in this concentration will have negligible effect on the SVP of the propellant.

In Figure 2.1, the metering chamber is shown filled with propellant which had entered through the groove in the plunger. The spray was
FIG. 2.1. CROSS SECTION OF A METERED-SPRAY GENERATOR.
produced by depressing the vial, whereupon the 0.58 mm (0.023 inch) orifice in the plunger entered the metering chamber. This allowed propellant to enter the expansion chamber and pass through the adapter nozzle into the atmosphere. At the same time, the groove was forced out of the metering chamber which was thus isolated from the propellant in the vial. In this way a metered dose was discharged which was independent of the duration of depression of the vial.

To investigate the mechanism of atomization, high speed cine photographs were taken with a "Fastax" rotating prism camera capable of 8,000 frames per second. Figure 2.2 shows the general arrangement. The metered-spray generator, previously described, was fitted into a solenoid activating mechanism. The solenoid was energised by an electrical pulse from the control unit of the "Fastax" camera. The spray was illuminated by a 7 kW xenon arc fitted with a cell of copper sulphate solution to act as a heat filter. In order to accelerate to the maximum framing speed of the camera and also have sufficient film to record the event, it was necessary to pass a 30 m roll of film through the camera. The film was then exposed in about 0.5 seconds. As the spray duration was about 0.1 seconds, it was necessary to discharge the metered spray after the passage of about half the roll of film. To obtain photographs, the xenon arc was switched on and the camera started. At the appropriate time a pulse from the camera control unit activated the solenoid. In this way, the light source, camera and spray were satisfactorily synchronised.

Three regions were photographed, representing three different stages in the atomization of the propellant and formation of the spray. They were: the expansion chamber and adapter nozzle; the region immediately downstream of the adapter nozzle; and the whole spray.

Initially, opaque polythene adapters only were available. In
FIG. 2.2. PHOTOGRAPHY OF A METERED SPRAY.
order to observe the propellant behaviour in the expansion chamber and nozzle, a section of the adapter was milled away until the expansion chamber and nozzle were revealed. The milled section was then replaced by a strip of perspex, 3.2 mm. (\(\frac{\text{\small{3}}}{\text{\small{8}}}\) inch) thick, which was bolted to the adapter. Eventually transparent tenite butyrate adapters were obtained, and in this case no milling was required.

The photographs in Figures 2.3 and 2.4 show prints of selected frames taken with reflected and transmitted light respectively, of the expansion chamber and orifice. The framing rate was about 5000 frames per second.

It was apparent from observation of these cine films that in the expansion chamber the propellant was partially vaporised and in the form of coarse ligaments. Their diameter was seen to become smaller as they increased speed to pass through the adapter nozzle. Movement through the nozzle, however, was too fast to be observed by this technique. Atomization appeared to be a two-stage process. Initial break-up took place in the expansion chamber followed by further break-up during passage through the adapter nozzle.

The prints in Figure 2.5 show the spray leaving the nozzle. Generally the spray was moving too fast for observations, and individual particles could not be resolved except for relatively large droplets at the periphery of the spray. The diameter of the largest of these peripheral droplets was about 100 \(\mu\text{m}\).

Figure 2.6 shows a more distant view of the spray. The vertical rod, which is 100mm from the nozzle, gives the scale of prints. From such films the build-up of the spray could be observed and the velocity of the spray-front could be measured. Individual particles within the spray
FIG. 2.3 HIGH SPEED CINE PHOTOGRAPHS OF THE EXPANSION CHAMBER AND NOZZLE.
FIG. 2.4 HIGH SPEED CINE PHOTOGRAPHS OF THE EXPANSION CHAMBER AND NOZZLE.
FIG. 2.5  HIGH SPEED CINE PHOTOGRAPHS OF THE FIRST FEW CENTIMETRES OF ASpray.
FIG. 2.6 HIGH SPEED CINE PHOTOGRAPHS OF THE DEVELOPMENT
OF A METERED SPRAY.
could not be resolved, however. A graph of the variation of spray front position with time is shown in Figure 2.7, while Figure 2.8 shows the variation of spray-front velocity with distance from the nozzle.

It is seen that, over the first few centimetres, the velocity of the spray-front remains essentially constant. Beyond this distance air is entrained into the spray by turbulent mixing and the spray-front velocity decays rapidly. Unfortunately it was not possible to measure velocities within the spray behind its front boundary.

Although no precise correlation between spray-front velocity and nozzle diameter was obtained, it is significant that the adapter with a 0.25 mm. (0.010 inch) nozzle generated a very low-velocity spray. This is to be expected on theoretical grounds as explained in Section 4.

From photographs of the complete spray, it was possible to measure the total volume of the spray. It was found that the latter was many times greater than the volume of vapour available from the metered volume of propellant. For example, the spray depicted in Figure 2.6 developed into a cone whose volume was approximately $70 \times 10^3 \text{ mm}^3$. Now the volume of vapour from 25 mm$^3$ of propellant is only $6 \times 10^3 \text{ mm}^3$. Therefore the resultant spray consisted of a surfactant aerosol in a gas phase comprising approximately 90% entrained air and 10% propellant vapour.

2.2 Pressure and temperature measurements within the expansion chamber

Atomization and velocity of discharge through a nozzle are intimately related to the pressure across the nozzle and to the liquid-to-gas ratio passing through it, the latter depending upon the propellant temperature. It was therefore desirable to measure the temperature and pressure within the expansion chamber.
FIG. 2.7. SPRAY-FRONT DEVELOPMENT OF A METERED SPRAY.
Fig. 2.8. Variation of spray front velocity with distance from the nozzle for metered sprays.
The time of duration of a metered spray is typically 0.1 seconds. Therefore it was essential that the instruments used to measure temperature and pressure had a fast response time, and that they were coupled to a fast recording instrument. Two fast-response recording instruments were available: a double-beam storage oscilloscope and an ultra-violet chart recorder. The U.V. recorder gave a permanent record and this was used. The arrangement is shown in Figure 2.9 and Figure 2.10.

A fine thermocouple was used to measure temperature. This was made by welding together, using capacity discharge, 0.12mm diameter (40s.w.g.) constantan and copper wires. It was then threaded through a small hole drilled through the expansion chamber of the adapter, and heat-sealed into position. The output was fed to the U.V. recorder via a reference thermocouple and a D.C. amplifier.

To measure the pressure within the expansion chamber, a quartz crystal piezo-electric pressure transducer was used. Its output was amplified with a charge amplifier and recorded on a second channel of the U.V. recorder. Initially the transducer was fitted directly into a drilled recess in the base of the adapter, with a small hole drilled through to the expansion chamber. It was found with this arrangement that the base-line of zero pressure rose during the spray. This was because the transducer had been cooled by the evaporating propellant, giving a reading in the same direction as that given by a positive pressure. This effect was partially overcome by smearing the surface of the transducer with silicon grease. However, no thermal effects were detectable during the discharge of the spray if the transducer was mounted in an adapter which increased the thermal capacity of the system. A disadvantage of the use of the adapter was that the volume of the expansion chamber was slightly increased.
FIG. 2.9. INSTANTANEOUS PRESSURE AND TEMPERATURE MEASUREMENT IN THE EXPANSION CHAMBER OF A METERED SPRAY GENERATOR.
FIG. 2.10  PRESSURE AND TEMPERATURE MEASUREMENT IN THE EXPANSION CHAMBER OF A METERED SPRAY GENERATOR.
The thermocouple and pressure transducer were calibrated as follows. The thermocouple, mounted on the metered spray generator, was placed in a Dewar flask containing distilled water, whose temperature was measured. By cooling the distilled water with ice, a number of calibration points were put onto the U.V. recording chart. The temperature could then be read from the chart to within 0.3 degrees Centigrade.

The pressure transducer was calibrated by applying a static pressure to it by means of a mercury manometer. Again calibration points were applied directly to the recording chart. A pressure pulse of about 100kN/m² could then be read to within ± 1kN/m².

Graphs showing the typical variation of pressure and temperature with time are shown in Figure 2.11. They indicate a rapid rise to maximum pressure and a simultaneous rapid fall to minimum temperature, followed by a slower return to ambient conditions. At no time during the discharge of the spray are steady-state conditions obtained.

Also included is the computed value of the saturated vapour pressure (S.V.P.) of the propellant. This was calculated from Raoult's Law, and is consequently subject to some error. For halogenated methanes, Raoult's Law generally gives a value of the S.V.P. below the measured value (7); the error is usually less than 10%. It would appear that towards the build-up to maximum pressure, the propellant is in a state approximating to thermodynamic equilibrium. However, during the latter part of the discharge, the pressure of the propellant is considerably lower than its S.V.P. During this part of the spray, therefore, the liquid propellant probably exists in a metastable state and the vapour in a superheated state.

2.3 Size and velocity measurements of spray droplets

It has been shown in the introductory remarks that for efficient
FIG. 2.11: TEMPERATURE AND PRESSURE MEASUREMENT WITHIN THE EXPANSION CHAMBER OF A METERED-DOSE AEROSOL GENERATOR.
operation of an aerosol generator employed for inhalation therapy, the velocity of the emitted droplets should be as low as possible and their size should be as small as possible. The size distribution of the emitted droplets is also a direct measure of the efficiency of atomization of a given combination of nozzle and propellant. It was considered necessary therefore to develop a method by which simultaneous velocity and size measurements of rapidly moving droplets could be made.

The results of the high-speed filming programme demonstrated that such measurements could not be obtained with a camera of the rotating prism type. Calculation showed that the exposure time must be no greater than one microsecond to give streak-free images, and that the interval between frames must be less than about ten microseconds in order that a particle on one frame could be identified on the next. The field must be magnified about ten times so that the image of the smallest resolvable particle would be bigger than the grain size of the film.

Equipment capable of framing rates of around \(10^6\) per second may well have provided a sequence of photographs on which individual particles were resolved. Instruments of this type, for example the Barr and Stroud C.P.5 camera incorporating a rotating mirror, or an image converter, were not readily available. The possibility of carrying out tests with the above equipment was discussed with A.W.R.E. (Aldermaston). They were of the opinion that the setting up of a high intensity source of illumination necessary for use with a high resolution optical system was not feasible. Indeed the C.P.5 camera was usually used for the photography of self-luminous events, such as explosions; otherwise the object was illuminated by a xenon "flash bomb". In view of the high cost of such equipment, the use of ultra high-speed cameras was not pursued.
The techniques so far considered for size and velocity analysis have relied on the formation of an image by a lens. Such a system has the inherent disadvantage that for high resolution the depth of field is limited, typically to about 300 μm. A number of reports had appeared in the literature (64,65,68,69) which described the use of pulsed laser holography as a means of measuring the size distribution of fog droplets. The depth of field was quoted as a few centimetres, and the resolution as 3 μm.

In view of the apparent advantages of this system, arrangements were made, again with A.W.R.E., to apply holographic particle sizing techniques to sprays generated by saturated liquids. An experimental arrangement is shown in Figure 2.12(a). This arrangement is not identical to that of A.W.R.E. but is similar to an equivalent system constructed, later, in the laboratory.

Holography of moving particles requires a coherent pulsed light source of extremely short pulse length. This was provided by a "Q" switched ruby laser which produced a pulse of 20 n·sec. Its beam was passed through a microscope objective lens by means of which it was focused and then spread. This served two purposes. Firstly, it spread the beam to a larger, more useful diameter. Secondly, the beam could be passed, at its focal point, through a spatial filter which improved its uniformity. The spatial filter consisted of a 10 μm aperture formed in thin aluminium foil by a previous discharge of the laser.

The spray was arranged so that it discharged between the laser and the photographic plate (Ilford R L). In addition, an array of three wires was placed at the intersection of the spray and laser beam, as shown; these were to facilitate reconstruction as explained later.

Holograms were formed by activating the solenoid which discharged
FIG. 2.12a. HOLOGRAPHY OF A METERED SPRAY.
FIG. 2.12b. RECONSTRUCTION OF A HOLOGRAM.
the spray and, after a predetermined time delay, pulsed the laser. Holograms would normally have consisted of a number of circular concentric interference patterns formed by the particles, and linear interference patterns formed by the wires. They are formed because an object in the path of a light beam diffracts some of the energy of the beam. This diffracted light interferes with the undiffracted, or reference beam and the resultant interference pattern, a hologram, may be recorded photographically. A hologram is shown in Figure 2.13. The interference patterns created by the wires are clearly seen. However, the concentric interference patterns typical of this type of holography of droplets were not present; instead the hologram resembled a schlieren photograph of the spray. There was insufficient time to determine the reason for this. However it seems likely that the resolution of this system was inadequate and the two largest wires, of diameter 100 \( \mu \text{m} \), were resolved while the spray particles, generally of diameter less than 50 \( \mu \text{m} \) were not. Resolution impairment was possibly caused by large refractive index gradients within the rapidly evaporating propellant.

In order to obtain an image of the original object, the hologram was reconstructed using apparatus shown in Figure 2.12(b). Light from a Helium-Neon gas laser was focused and spatially filtered. The expanding beam was passed through the hologram and on to a reflex camera, whose lens had been removed. The camera was moved along the laser axis until the focused images of the wires were seen. The film in the camera (Ilford Pan F) was then exposed and developed. A reconstruction of the hologram of 2.13 is shown in Figure 2.14. The wires are seen to reconstruct satisfactorily but for reasons given above, the droplets do not.

From the point of view of creating an acceptable hologram.
FIG. 2.13  HOLOGRAM OF A METERED SPRAY.

FIG. 2.14  RECONSTRUCTION OF THE ABOVE.
using the present apparatus, two methods were proposed. Firstly, the spray could be passed through a narrow slit to reduce the depth of vapour through which the laser beam must pass. Secondly, the spray could be directed into an atmosphere of propellant vapour, thus reducing evaporation and the magnitude of the refractive index gradients. Time did not allow either method to be tested, but in both cases the conditions would have been too artificial to be acceptable. Consequently, the use of holography was not pursued.

A survey of other methods of particle size and velocity analysis indicated that no system was ideal, although a number were likely to be reasonably satisfactory. It was essential to avoid the selection of a method whose commission was likely to occupy the major part of the time available for the project. It was decided therefore, to use a modification of the double-exposure photomicrographic technique first used by York and Stubbs (75) and more recently by Ramshaw (52). This is described in Section 5.

2.4 Measurement of the size distribution of the residual aerosol

One of the most important properties of a spray from a pharmaceutical point of view is the size distribution of the residual aerosol. In addition to being a measure of the efficiency of atomization of the spray generator, it is this parameter that determines, to a great extent, the part of the lungs on which the particles will be deposited. Particles which are too fine will, like smoke, be exhaled, and those too large will impact on the region of the upper respiratory tract. The optimum particle diameter was discussed in Section 1.

It was shown in Section 1 that for commercially used formulations, the residual aerosol would generally consist of drug particles in larger droplets of a liquid surfactant of very low volatility. Thus, size analysis of the residual aerosol would necessitate the analysis of relatively non-volatile
liquid particles. Many methods of size analysis of such particles have been described in the literature. A survey is given in Section 3, together with an assessment of their applicability.

Because of the polydisperse nature and high velocity of the emitted spray, special care must be taken to obtain a representative sample for the purpose of size analysis. It is especially important that there should be no discrimination against the larger particles, bearing in mind that the volume of one particle of diameter 20 μm is equal to that of 8000 particles of one μm diameter. For this reason the size distribution of the residual aerosol was obtained by optical microscope analysis of the droplets deposited on glass slides by sedimentation, as described below.

The spray was discharged ten times into a sedimentation cylinder whose dimensions were large compared with the dimensions of the spray. After replacement of a lid the aerosol was allowed to sediment for about fifteen hours onto cleared glass slides. In this way, no loss of larger particles resulted.

The number of discharges was chosen such that there was a reasonable number of droplets in the field of view of the microscope, but not so many that overlapping became a problem. The microscope counting procedure is described in Section 5.

Size analyses of two aerosols obtained from metered sprays are shown in Figure 2.15. One spray was generated by discharging 50 mm$^3$ of propellant through an adapter nozzle of diameter 0.25 mm. (0.010 inch). The second was generated by discharging 50 mm$^3$ of propellant through an adapter nozzle of diameter 0.58 mm. (0.023 inch).

The size distributions are plotted on log-probability graph paper. The graphs are approximately straight lines indicating that the size distributions are approximately log-normal. A better fit is obtained by plotting on Rosin-Rammler graph paper. This plot is shown
FIG. 2.15. SIZE DISTRIBUTION OF RESULTANT "SPAN 85" AEROSOL FROM A METERED SPRAY. [LOG-PROBABILITY PLOT]

PARTICLE DIAMETER μm

PERCENT BY VOLUME UNDERSIZE.

O.25 mm. ADAPTER ORIFICE

O.59 mm. ADAPTER ORIFICE.
in Figure 2.16. The size distributions may then be given more accurately by:

\[ \text{Fraction by volume oversize } = e^{-Ad^s} \]

where \( A \) and \( s \) are constants and \( d \) is the particle diameter.

The mass median diameters of the two aerosols were 4.9\( \mu \)m and 7.8\( \mu \)m for nozzle diameters of 0.25 mm and 0.58 mm respectively.

2.5 Conclusions

In this section, experiments have been described by means of which a qualitative concept of the atomization process within a metered-dose aerosol generator was obtained. Secondly, by testing a number of experimental techniques a guide to further experimentation was obtained by which a more quantitative description of the atomization process could be deduced. Thirdly, these preliminary experiments also enabled certain design criteria for an atomizing device to be obtained. These three aspects of the preliminary experimentation are summarised below.

Mechanism of atomization

Section 2.4 described the results of high-speed cine photography from which an insight into the atomization process within the expansion chamber was obtained. The propellant was seen to vaporise partially and break up into coarse ligaments which followed a turbulent path in the expansion chamber. The ligaments eventually passed through the adapter nozzle, possibly undergoing further break-up.

A complete evaluation of the process could not be made because the behaviour of the liquid/vapour mixture in the region of the nozzle and nozzle exit could not be seen. It was suspected that the ligaments became narrower as a result of their increase in speed on entering the nozzle, and that under the influence of surface tension forces broke down into shorter ligaments and droplets.
FIG. 2.16. SIZE DISTRIBUTION OF THE RESULTANT "SPAN 85" AEROSOL FROM A METERED SPRAY.
[Rosin–Rammler Plot.]
Support was given to this view by photomicrographs taken of the expansion chamber and adapter nozzle by the unit described in Section 5. One such photograph is reproduced in Figure 2.17. Although interpretation is not completely straightforward, the two-phase mixture in the nozzle is seen to be highly dispersed, with the liquid phase in the form of ligaments whose dimensions are considerably smaller than those of the ligaments entering the nozzle.

Further experimentation

The above constitutes a limited qualitative description of the atomization process. It was the purpose of the project, however, to obtain also a quantitative description of this process. This necessitated in-flight droplet measurements, together with measurements of temperature and pressure within the expansion chamber of the spray generator. These could then be related to the two fundamental parameters: the dimensions of the nozzle and the thermodynamic properties of the propellant. These preliminary experiments gave a guide as to how these measurements might be made.

It is shown in Section 2.3 that measurements of spray droplets in flight would require the construction of a double-flash photomicrographic system. Suitable apparatus was built and used in subsequent experimentation.

Also, measurements, described in Section 2.2, of the instantaneous temperature and pressure of the propellant in the expansion chamber indicated that satisfactory steady-state conditions were never reached during the discharge of the spray. Under these rapidly varying conditions it is unlikely that a relationship between properties of the spray and the spray generator would have been obtained. For these reasons a continuous-flow model, described in Section 5, was built and further experimentation carried out under steady-state conditions.
FIG. 2.17 PHOTOGRAPH OF THE DISCHARGE OF A METERED SPRAY.
Preliminary design criteria

Sections 2.1 and 2.4 gave information which would assist in the design of a spray generator suitable for inhalation therapy. It was shown in these sections that the smaller the diameter of the adapter nozzle, the finer the solute aerosol and also the slower the spray-front velocity. Therefore, in order to produce a suitable spray, it would seem that the adapter nozzle should be as small as possible. There are limits, however, to the fineness of the nozzle, for too small a nozzle diameter would lead to clogging when a powder suspension is dispersed through the device. Also the discharge time may become excessively long, but it is the former constraint that is considered to be the more serious.
3 LITERATURE SURVEY

Introduction

3.1 Atomization of saturated liquids
3.2 Particle size and velocity measurements
3.3 Flow of saturated liquids and two-phase fluids through nozzles
3.4 Conclusions

The areas of general interest in the study of inhalation sprays or aerosols were outlined in the Introduction, Section 1, while a preliminary study of the mechanism of atomization of such sprays was described in Section 2. It is the purpose of this section to review reported work which has a bearing on the various aspects of this study.
3.1 Atomization of saturated liquids.

The use of liquified gases as a spray generating agent grew rapidly during the Second World War. They were used mainly in the application of insecticides. The propellant then available was the halogenated hydrocarbon, dichlorodifluoromethane (propellant 12), whose saturated vapour pressure (S.V.P.) at room temperature (20°C) is 568 kN/m²(abs). It was atomized through capillary tubing. Owing to the relatively high S.V.P. of this propellant it had to be housed in strong metal containers, which were consequently inconvenient and expensive.

Because of the disadvantages of using a high pressure propellant, new propellants of lower pressure were sought and also efficient means of atomizing them. The lower pressures were achieved by the development of halogenated hydrocarbons, of low S.V.P., which were miscible with propellant 12. Such a compound is trichloromonofluoromethane, or propellant 11. Thus a continuous range of pressures was available, from 90 kN/m²(abs) the S.V.P. of propellant 11 at 20°C to 568 kN/m²(abs), as previously mentioned. Means of atomizing these low pressure propellants were sought. It was found that the capillary device was still satisfactory, but other means were investigated. In particular the two-orifice nozzle assembly for this use was developed in 1950 (76).

The operation, for continuous flow, of a two-orifice assembly was found to be as follows. The pressure drop across the first orifice caused vaporisation of the propellant within the expansion chamber. A two-phase mixture of propellant liquid and vapour then flowed through the second orifice. Because liquid only, tended to flow through the first (upstream) orifice only about one eighth of the overall pressure differential was dropped across this orifice. Thus a substantial fraction of the pressure differential was available at the second orifice. Atomization was considered to be a two-stage process. Initial break-up took place in the expansion
chamber as a result of the initial evaporation of the propellant. Further break-up took place as the resulting two-phase propellant passed through the second orifice.

Atomization through capillary tubes was similar. In this case however the process took place continuously over a few centimetres of the tubing.

Most of the work done on atomization through two-orifice nozzle assemblies has resulted in empirical relationships. These have usually been a correlation between the size distribution of the resultant aerosol of the active ingredient and various parameters such as nozzle dimensions, propellant pressure, proportion of propellant to active ingredient etc. Experimentation of a more fundamental nature has been performed on atomization through single-orifice devices however. Work on atomization through two-orifice nozzles and to a certain extent capillary tubing will be considered first.

Fulton et al (19) described the atomization of a formulation consisting of 43% propellant 11, 43% propellant 12 and 14% insecticide through capillary tubes. A size analysis was performed on the residual insecticide aerosol and the mass median diameter (M.M.D.) was related to capillary length and diameter. It was found that for a given capillary diameter, an optimum length produced a minimum M.M.D. However, over a range of capillary diameters from 0.34mm to 0.74mm this minimum M.M.D. was approximately constant and equal to 16μm.

Two-orifice nozzles were also tested. It was found that the most satisfactory spray was generated when the volume of the expansion chamber was 130mm$^3$ and the upstream and downstream orifice diameters were 0.38mm and 0.53mm respectively. In this case the criterion of efficiency was the mortality rate of house-flies.

York (76) reviewed the literature on the formation of sprays generated by liquified-gas propellants. Aerosol formation was divided into
four stages:—

1. primary atomization due to flashing of the propellant;
2. secondary atomization of droplets formed in stage 1, by impact with the atmosphere;
3. evaporation of the propellant and
4. entrainment of air into the spray.

It was concluded that, although much existing literature was applicable to the last three stages, no experimental work, of a fundamental nature, had been reported on the primary atomization of saturated liquids.

Lefebvre and Tregan (38) measured some of the factors affecting the size distribution of a kerosene aerosol generated from a solution of kerosene and propellant 11/12 solution. The particle size of the aerosol was measured by sedimentation on to a glass slide coated with magnesium oxide; many thousands of particles were counted. This method has a resolution of about 5 μm. Flash photography was used to give a qualitative assessment of the spray. However, no attempt was made to relate the variables to the mechanism of atomization.

Results are given for the variation of volume mean diameter (d_{43}) of the kerosene aerosol with:

(a) the temperature, over the range 15°C to 35°C,
(b) the ratio of propellant to kerosene,
(c) the fraction of propellant 12 in the propellant, and
(d) different nozzle designs.

The results are reproduced in Figs. 3.1(a), (b) and (c) and Table 3.1.

It is seen that the spray becomes finer with increase in temperature but that over the usual range of variation of ambient temperatures, spray size remains fairly constant. A low ratio of propellant to kerosene is seen to increase the size distribution dramatically below about 80% propellant. The composition of the propellant is also seen to affect the
FIG. 3.1 DEPENDENCE OF THE SIZE DISTRIBUTION OF KEROSENE AEROSOL ON VARIOUS PARAMETERS.

Standard conditions unless otherwise stated.
Propellant: 50% P12/50% P11 by weight.
Temperature: 20°C
Ratio of propellant: kerosene 85% : 15%
Nozzle: "Diffuseur Tourbillonmaur" $T_1$
Temperature = 20°C
Propellant = 50/50% by weight P11/P12
Propellant/Kerosene = 85/15% by weight.

Table 3.1: Effect of nozzle type on size distribution of a kerosene aerosol.

<table>
<thead>
<tr>
<th>NOZZLE</th>
<th>MEAN VALUE OF d_{43} (μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>&quot;Diffuseur Pinceau&quot;</td>
<td>71</td>
</tr>
<tr>
<td>&quot; Nuage</td>
<td>35</td>
</tr>
<tr>
<td>&quot; Tourbillonnair T_1</td>
<td>36.5</td>
</tr>
<tr>
<td>&quot; &quot; T_2</td>
<td>41</td>
</tr>
</tbody>
</table>

size distribution, with the higher pressure giving the finer spray. Nozzle type can also be seen to alter the size distribution considerably. Unfortunately no details of the nozzles were given.

Oxford Industrial Research and Development (48) reported the use of impinging jet nozzles for the atomization of a solution of liquified propellant and low volatile product. Atomization is shown to be caused by bubble growth within the impinging jets, aided by hydrodynamic and aerodynamic forces in a similar manner to the atomization of impinging water jets. The report indicates that vaporization in the expansion chamber should be avoided. It is claimed that the resulting spray contains 25% more particles in the 1 μm to 5 μm region.

Polli et al (51) used a cascade impactor to measure aerosol particle size from a metered spray generator containing propellant, powdered drug and surface-active agent. The study was designed to investigate
the influence of several formulation parameters on aerosol particle size. The results are shown in Tables 3.2 overleaf.

From Table 3.2 it is seen that the size distribution and concentration of the drug was found to have a marked effect on the aerosol size distribution; but that the surfactant concentration had minimal effect. Spray orifice diameter was found to have a marked effect, for diameters below about 0.6mm. The effects of vapour pressure and temperature were found to be similar to those found by Lefebvre and Tregan. A rise in temperature of about 20 deg.C from nominal room temperature was found roughly to halve the appropriate mean diameter of the spray. Similarly, doubling of the gauge pressure of the propellant was found roughly to halve the mean diameter; at pressures below about 140 kN/m$^2$ gauge however, the aerosol diameter was found again by both authors to increase rapidly. Polli found that high pressure of about 520 kN/m$^2$ gauge was required for an aerosol size of the same order as that of the original drug.

Tsetlin (67) measured the size distributions of kerosene and oil aerosols generated by a continuous spray. The aerosols were collected by sedimentation on to glass slides and mass medium diameters were measured by microscope counting. It was found that the M.M.D. was reduced by increasing the proportion of propellant and by increasing the temperature, this was in general agreement with the result of Tregan and Lefebvre, and Polli. Table 3.3 shows that the M.M.D. of the oil aerosol was considerable greater than that of the oil. This was attributed to the higher value of the viscosity of the oil (16.2cp) compared with that of the kerosene (1.27cp).
Standard conditions (unless otherwise stated):

- Propellant: 20% P12/80% P114
- Actuator diameter: 0.46 mm
- Ambient temperature: 24 °C
- Relative humidity: 30%

Concentration of:
- Drug: 0.143% by mass
- Surfactant: 0.2% by mass

<table>
<thead>
<tr>
<th>DRUG (µm)</th>
<th>AEROSOL PARTICLES (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.6</td>
<td>9.0</td>
</tr>
<tr>
<td>4.3</td>
<td>6.0</td>
</tr>
<tr>
<td>1.4</td>
<td>3.2</td>
</tr>
</tbody>
</table>

Concentration of 

<table>
<thead>
<tr>
<th>DRUG % BY MASS</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0175</td>
</tr>
<tr>
<td>0.143</td>
</tr>
<tr>
<td>0.286</td>
</tr>
</tbody>
</table>

SURFACTANT CONCENTRATION % BY MASS

| 0.0 | 4.6 |
| 0.2 | 3.2 |

Spray Orifice Diameter mm

| 0.76 | 11.0 |
| 0.61 | 11.0 |
| 0.46 | 3.2 |

<table>
<thead>
<tr>
<th>PROPELLANT</th>
<th>VAPOUR PRESSURE (kN/m² GAUGE)</th>
</tr>
</thead>
<tbody>
<tr>
<td>P114</td>
<td>110</td>
</tr>
<tr>
<td>20% P12/80% P114</td>
<td>214</td>
</tr>
<tr>
<td>P12</td>
<td>530</td>
</tr>
</tbody>
</table>

Temperature °C

<table>
<thead>
<tr>
<th>TEMPERATURE °C</th>
<th>PROPELLANT</th>
<th>VAPOUR PRESSURE (kN/m² GAUGE)</th>
</tr>
</thead>
<tbody>
<tr>
<td>24</td>
<td>P114</td>
<td>110</td>
</tr>
<tr>
<td>37</td>
<td>P114</td>
<td>206</td>
</tr>
<tr>
<td>24</td>
<td>20% P12/80% P114</td>
<td>213</td>
</tr>
<tr>
<td>37</td>
<td>&quot;</td>
<td>350</td>
</tr>
<tr>
<td>49</td>
<td>&quot;</td>
<td>516</td>
</tr>
<tr>
<td>5</td>
<td>P12</td>
<td>260</td>
</tr>
<tr>
<td>24</td>
<td>P12</td>
<td>530</td>
</tr>
</tbody>
</table>

**TABLE 3.2** THE INFLUENCE OF SEVERAL FORMULATION PARAMETERS ON PARTICLE SIZE.
The previous work has dealt with the atomization of saturated liquids passing through a two-orifice nozzle assembly. The work of Brown and York (6), who atomized saturated liquids through smooth single orifices will now be considered. The liquids atomized were water and, separately, propellant 11; they were at a temperature above their normal boiling point at atmospheric pressure, but sub-cooled, that is at a pressure greater than their S.V.P.

The atomization process was analysed from double flash photomicrographs taken of the jet just beyond the nozzle. Droplet size distributions were obtained from photomicrographs taken 150mm from the nozzle.

It was deduced that atomization was caused mainly by bubble growth.

<table>
<thead>
<tr>
<th>SOLUTE AND VISCOSITY</th>
<th>MMD OF SOLUTE AEROSOL</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kerosene: 1.27cp at 20°C</td>
<td>4.3 µm</td>
</tr>
<tr>
<td>Transformer oil: 16.2cp at 20°C</td>
<td>11.1 µm</td>
</tr>
</tbody>
</table>

**TABLE 3.3** EFFECT OF THE VISCOSITY OF THE SOLUTE ON THE SIZE DISTRIBUTION OF THE SOLUTE AEROSOL.
within the now superheated liquid jet. Atomization was considered to be influenced also by aerodynamic forces acting on the jet. The magnitude of these forces was determined by the value of the Weber number. This is:

\[ N_{\text{we}} = \frac{\rho_{\text{air}} u^2 D}{2 \sigma} \]

where \( \rho_{\text{air}} \) is the air density, 
\( u \) is the relative velocity between jet and air, 
\( D \) is the jet diameter, 
\( \sigma \) is the surface tension of the jet liquid.

The Weber number may be taken to be the ratio of the aerodynamic force tending to disrupt the jet, to the surface tension force holding it together.

For water, a correlation was obtained between the number mean droplet diameter, \( d_{10} \), 150mm from the nozzle, and the Weber number:

\[ d_{10} = \frac{1840 - 5.18 T(\text{°F})}{N_{\text{we}}} \]

Because of lack of data, no similar correlation was obtained for propellant 11 although atomization was considered to take place by the same mechanism.

Break-up of unsaturated liquids.

In Section 2 it was demonstrated that the spray was emitted from the final orifice in the form of liquid ligaments, droplets and vapour. Further it was shown that this liquid was, by this stage in the atomization process, no longer supersaturated. It is therefore relevant to consider the possibility of further atomization of this liquid beyond the nozzle.

One of the earliest studies of atomization was that by Rayleigh (53) who considered the break-up of a jet of non-viscous liquid issuing from a simple nozzle of diameter \( D \) at low pressure. The jet was shown to break up as a result of surface tension forces, into droplets whose diameter \( d \) is
given by: -
\[ d = 1.89 D. \]

This theory however does not account for smaller satellite droplets formed between the larger droplets.

Weber (70) modified the Rayleigh theory to include the effects of viscosity (\( \eta \)) and obtained the relationship:

\[ d = 1.88 D \left( f(\eta) \right)^{\frac{1}{3}}, \]

where

\[ f(\eta) = \left( 1 + \frac{3\eta}{\left( \frac{\rho_1}{\sigma} D \right)^{\frac{1}{3}}} \right)^{\frac{1}{3}}, \]

which gives the same result as the Rayleigh theory as \( \eta \) approaches 0.

Weber extended his analysis to include aerodynamic forces acting on the liquid jet surface. It was found that the behaviour of the liquid jet could be described in terms of the dimensionless number, now called the Weber number, defined earlier. Hinze (29) investigated the effect of the Weber number on the disintegration of a liquid jet. He found that for \( \text{We}_w \) less than 0.2 only the Rayleigh pinching effect was present. For \( \text{We}_w \) between 0.2 and 8 the jet became sinuous, breaking into segments. For values greater than 8 the action increased in violence and ligaments of fluid were torn from the jet.

The above analyses of Rayleigh and Weber applied to a liquid jet issuing from a nozzle, and are therefore of limited applicability to an emerging spray already consisting of ligaments and droplets. It may be more meaningful therefore to consider the possibility that these liquid particles may further break-up as a result of impact with the atmosphere or of impact with the spray ahead which in general is moving considerably more slowly as a result of entrainment of the atmosphere.
This further break-up is called secondary atomization. Criteria for secondary atomization have been determined by a number of people. All have found that secondary atomization occurred if a critical value of the Weber number was exceeded. Thus Lane (36), from experimental work with water droplets in air at high Reynolds number, derived a value for the critical Weber number of 4.1 for sudden application of the air stream, and 5.7 for steady application. Haas (27) gives a value of 5.6 from a study of mercury droplets suddenly exposed to high velocity air stream. Gordon (22) on theoretical grounds gives a value of 8. The implication of this criterion is considered later.

A vast amount of literature on atomization exists. It is mainly related to pressure atomization and pneumatic atomization of unsaturated liquids. The former bears little resemblance to the atomization of saturated liquids. The latter however may have some bearing on the later stages of saturated liquid atomization. This is because the propellant vapour could in some circumstances be instrumental in the atomization of the liquid propellant. It is useful, however, to compare these two methods with two-orifice saturated liquid atomization. This comparison is readily made from an extensive review of both pressure and pneumatic atomization by Dombrowski and Munday (11). An estimate of the volume-surface mean diameter \( d_{32} \) of a saturated liquid spray in the vicinity of the nozzle may be obtained from the size distribution of its residual aerosol by dividing by the cube root of the solute concentration. This assumes no coagulation of the spray. Application of this to the results of Polli, given earlier, produces a value of \( d_{32} \) of about 25 \( \mu \)m for the smaller orifices. Data given in Section 6 yield similar results. Pressure atomization of water from a swirl spray nozzle at the same pressure, of the order of 350 kN/m\(^2\) gauge, at approximately the same flow-rate, of the order of 1 gm/sec (obtained again from Section 6), is shown, from the above review
to produce a mean diameter $d_{32}$ of about 100 μm. As would be expected pressure atomization is considerably the less efficient. Prefilming pneumatic atomization would produce a mean diameter similar to that of a saturated liquid spray if the ratio by mass of gas to liquid was 2:1. As a typical saturated liquid spray, of modest propellant pressure, emits propellant as a two-phase fluid of the order of 10% vapour quality, it again follows that two-orifice saturated liquid atomization is comparatively efficient.

**Properties of gas jets and sprays.**

The behaviour of the spray in the atmosphere may now be considered. It was shown qualitatively in preliminary work that a metered spray was turbulent and furthermore the total volume of the resultant spray was considerably greater than the volume of propellant vapour; the spray therefore consisted mainly of entrained air. It is therefore relevant to consider published work on turbulent air jets, the effect of loading the jet with particulate matter, and the entrainment of air into sprays.

The behaviour of gaseous jets is well understood. Information on such jets is well documented (49, 66). This is considered in more detail in the next section. The behaviour of particulate matter within a jet is less well documented. Goldschmidt and Eskinazi (21) reviewed work on diffusion of particulate matter in turbulent flow and presented a theory for the diffusion of small particles in a plane air jet. Experimental data were obtained which were compared with the theory; good agreement was found. The data were obtained from measurements of the concentration flux and concentration of aerosol particles in the plane air jet. The concentration of the aerosol was very low, however, of the order of $10^{-7}$ by volume at the nozzle. It was found that the transverse distribution of concentration flux and concentration beyond about 30 nozzle diameters were approximately Gaussian. The axial distribution of concentration flux was inversely proportional to the downstream distance from the apparent origin of the
spray, while the axial distribution of concentration was inversely proportional to the square root of the distance.

Laats (34) measured the effect of loading an air jet with particulate matter whose mass flow-rate was of the order of that of the air. It was found that the divergence of the jet was reduced slightly and hence the attenuation of axial velocity was also slightly reduced.

Entrainment of air into jets may now be considered. Again for an air jet the theory is well established. For sprays Mayer and Ranz (40) presented a theoretical expression for entrainment of air, based on the assumption of uniform velocity across the spray.

Briffa and Dombrowski (3) measured, indirectly, the ratio of air entrainment into a flat spray obtained from the disintegration of a liquid sheet. They compared the results with theory based on momentum conservation. Reasonable agreement between experiment and theory was obtained. It was noted that their expression for the ratio of mass of entrained air to the spray liquid was similar to the analogous expression for an air jet. Benatt and Eisenklam (1) made direct measurements of the rate of entrainment of air into water sprays from swirl nozzles, using a porous pot technique. It was found that the mass rate of entrainment of air was approximately equal to that into an air jet if the spray angle was equal to that of the air jet.
3.2 Particle size and velocity measurements.

It has been demonstrated in Section 1 that the spray parameters of interest are the size and velocity distributions of the particles within the spray, and also the size distribution of the resultant aerosol of the pharmaceutically active ingredient. Because in general, within the spray, particle velocity varies with particle diameter, it is desirable, but not necessarily essential, to measure spray-particle size and velocity simultaneously.

In this sub-section methods of measurement of the size and velocity of rapidly moving particles will be reviewed. Also method of particle size measurement of the resultant aerosol will be summarised.

Double exposure photomicrography.

Double exposure photomicrography is probably the simplest technique for obtaining simultaneous size and velocity measurements of spray particles. It was used successfully by York and Stubbs (75), and others (52,17) for example. The usual design is a sub-microsecond spark as a light source and a high resolution lens arranged in the transmitted-light configuration. With this arrangement rapidly moving particles of diameter about 4 \( \mu \text{m} \) may be resolved at speeds of the order of 40 m/sec.

Particles smaller than 4 \( \mu \text{m} \) may be detected using dark field illumination, but this technique is not readily applicable to spray photography because of confusion caused by out-of-focus particles.

Rotating mirror photomicrographic system.

Ingebo (30,31) used a rotating mirror in a otherwise conventional photomicrographic system to measure the size and velocity distributions of ethanol particles in a rocket combustor. The use of the rotating mirror enabled rapidly moving particles to be photographed with a relatively long duration (8 \( \mu \text{sec} \)) spark light source. This duration was found necessary in order to provide sufficient light. From the optimum speed of rotation
of the mirror, the speed of the particles was determined; this ranged from 10 to 20 m/sec.

**Fluorescent Photography.**

The transmitted-light photomicrography technique described above gives good results providing a resolution limit of a few micrometres is acceptable. It has the disadvantage however that automatic scanning techniques for particle size analysis are not readily applicable (52) because of the presence of out-of-focus particles. To eliminate the problem of out-of-focus particles the illumination must be confined to a narrow sheet in a plane normal to the camera axis. Benson et al (2) tested this technique. They found that the intensity of the scattered light was low and that the particles were unevenly illuminated; highlights were created on one side of the particle, making size measurements difficult. These problems were overcome by dissolving an ultra-violet absorbing fluorescent dye into the spray-generating liquid, for example uranin in water. The spray droplets then became luminous, and as the induction and decay times of the dye were very short, of the order of 5 nsec, the droplets remained in focus while still emitting light. Consequently all droplets photographed were in focus and there was no bright background to reduce the contrast of the images of smaller particles.

Groeneweg et al (26) modified this technique by using a "Q"-switched ruby laser in a frequency doubling mode such that a 50 nsec pulse of ultra-violet (347 nm) was obtained. The authors state that with this system, resolved images of 10 μm particles travelling at 50 m/sec could be obtained. The lower limit of resolution is not given, but there seems no theoretical reason why smaller particles should not be photographed in this way. Further, the use of a double-pulsed laser system would enable velocities to be measured.

**Holography.**

Fraunhofer holography, as a means of particle size measurement was developed in 1966 (64, 65, 68, 69) initially to obtain the size distribution.
of the water droplets in fog. The resolution of the system was about $3 \mu m$ and the overall depth of field of the hologram was about $300 \mu m$. It was possible however, on reconstruction of the hologram to restrict the depth of field optically, and scan longitudinally through the total field of view. In this way the number of particles in focus in any one field could be restricted to a convenient number. The experimental arrangement of a single pulsed Fraunhofer holographic system was described in Section 2.

More recently Fourney et al (18) developed a double pulsed holography system with which size and velocity distributions of particles could be determined. Resolution was shown to be approximately $5 \mu m$.

It is considered that holography is a useful technique for particle size and velocity measurement, especially when a large depth of field is required. However, as explained in Section 2, the technique could not be applied to particles within a spray generated by a liquified gas because of loss of coherence of the laser beam as a result of its passage through the propellant vapour, and a dense field of particles.

The techniques so far discussed have been applicable to the determination of size and velocity distribution of rapidly moving particles. The determination of the size distribution of the residual aerosol may now be considered.

A large number of techniques are available for the determination of the size distribution of aerosols. However, as discussed in Section 2, it is necessary that the system should have a resolution of about $1 \mu m$ or less but not discriminate against particles of diameter up to about $50 \mu m$.

This will generally mean that no single method of size analysis is applicable. This is because either the method of sampling is discriminatory or the applicable size range is less than that required. For example the terminal velocity of a $50 \mu m$ particle is approximately $72 \text{ mm/sec}$. Thus any aerosol sizing method which relies on sampling from the aerosol will
inevitably discriminate against such particles. This disadvantage may be overcome by sampling twice, using two methods such that there is an overlap with respect to the diameter of particles sampled efficiently. Thus, using this principle Schönauer (58) for example, was able to measure the size distribution of an azulene medical spray whose size distribution ranged from approximately 0.1 μm to 30 μm. The coarser fraction, above 1.7 μm diameter, was sampled by sedimentation for 10 hours and analysed with an optical microscope. The finer fraction below 2.3 μm, was sampled with a thermal precipitator and analysed using an electron microscope using a standard replication technique together with knowledge of the "flattening factor" of the original flattened droplet on the electron microscope grid. The two distributions were readily combined to give the complete distribution.

Having thus eliminated the problem of the larger size fractions, a number of sizing techniques for the smaller fraction is available. For example, in addition to electron micrography mentioned above, the principle of light scattering from individual particles may be employed, using commercially available instruments (55), to determine particle size in the range 0.3 to 10 μm. Alternatively an electrical particle analyser (71) may be used to measure the size distribution of particles in the range 0.015 to 1.2 μm. Centrifugal sedimentation may be used to classify particles according to diameter. Compared with gravitational sedimentation, centrifugal deposition of sub-micron particles is quicker and more effective. An early device, the "conifuge" (56) graded particles in the size range 30 to 0.5 μm. A later version, the "aerosol spectrometer" (20), was applicable to particles in the size range 3 to 0.03 μm. Cascade impactors have been shown to classify particles successfully. An early four-stage version (39) had a lower deposition limit of approximately 0.7 μm. A later six-stage version (43) extended the lower limit to 0.3 μm. The upper limit in this case is determined by wall losses at the first orifice and, as before, sampling efficiency of large particles
from an aerosol. Wall losses were considered to become serious with particles above \( 50 \mu m \).
3.3 Flow of saturated liquids and two-phase fluids through nozzles.

The flow of saturated propellant through a two-orifice nozzle assembly of the type used in this work may be divided into two parts. The first is the flow through the first, upstream, orifice (short tube) when the propellant is in liquid form on entering the orifice. The second part is flow through the second orifice. In this case the propellant is in the form of an intimately dispersed two-phase fluid - the liquid propellant and its vapour. It is therefore relevant to review literature relating to these two types of flow.

3.3.1 The flow of a saturated liquid through an expansion device.

Early work on nozzle flow of saturated fluids, both liquids and vapours, assumed thermodynamic equilibrium throughout the flow. Thus saturated liquids were assumed to vaporise instantly on entering the nozzle as a result of the reduction in pressure, and vapours in general were assumed to condense. Using this model in which thermodynamic equilibrium was assumed, it was found that actual mass flow-rates were greater than the predicted values. Thus Stuart and Yarnell (61), for example, predicted mass flow-rates of saturated water through a two-orifice nozzle. They then compared their predictions (62) with experimentally determined values. It was found that the experimental values were considerably higher than predicted. For example the flow-rate of saturated water at 690 kN/m² abs was predicted to be $2.3 \times 10^3$ kg/m²/sec while the measured rate was $7.2 \times 10^3$ kg/m²/sec. The discrepancy was attributed to the existence of a metastable state, in which pressures were generally lower than the saturation levels. A further calculation was therefore performed in which metastability was included; it was assumed that there was no vaporisation during passage through the first orifice.
and that there was no change in vapour quality (mass fraction vaporised) during passage through the second orifice. It was further assumed that there was no change of specific volume or temperature through the second orifice. The predicted mass flow-rate, again for a saturation pressure of 690 kN/m², was $12 \times 10^3$ kg/m²/sec giving a discharge coefficient of 0.61 for the second, downstream, orifice. This, the authors considered acceptable. It would seem however that the assumption of constant specific volume through the second orifice was physically unrealistic; indeed it would result in adiabatic flow of decreasing entropy.

Pasqua (50) studied the flow of refrigerant (or propellant) 12, through orifices, short tubes and nozzles. For saturated liquid flow through orifices complete metastability was found, and the usual orifice formula could be used to predict the flow-rate.

For saturated liquid flow through short tubes it was found that the liquid immediately broke away from the wall of the tube and was surrounded by an annulus of vapour; the tube then displayed orifice characteristics as above. Below a critical downstream pressure the flow became mass limiting. This critical pressure $P_c$ was given by:

$$P_c = 0.37 \left( \frac{L}{D} \right)^{0.2} x \text{ (initial saturation pressure)}$$

where $L/D$ is the ratio of the length to diameter of the short tube.

For sub-cooled liquid flowing through short tubes, full tube flow was observed until the pressure fall resulted in a pressure at the vena contracta equal to the S.V.P. of the liquid; the flow then reverted to orifice flow as before.

For nozzle flow of saturated liquid, it was found again that the flow broke from the tube and a vapour annulus was formed. The flow could be
predicted by the conventional nozzle equation providing a coefficient of contraction was introduced, which was based on the length-to-diameter ratio of the nozzle.

Fauske and Min (16) studied the flow of saturated propellant through apertures and short tubes. Similar results to those of Pasqua were found. Thus for flow through apertures, the saturated liquid was found to behave as an unsaturated liquid. Short tubes behaved as apertures, with a discharge coefficient of 0.61, providing critical conditions were not reached. The critical point was defined in terms of a modified cavitation number:

$$\frac{2 \Delta P}{\rho_1 Q^2} A^2 \left(\frac{L}{D}\right),$$

where $\Delta P$ is the pressure difference across the tube, $A$ is the tube area, $\rho_1$ is the liquid density and $Q$ is the volume flow-rate. $L/D$ is again the length-to-diameter ratio of the tube.

Flow remained non-critical below a modified cavitation number of 10. Above a value of 14, the flow became critical or mass limiting. Between 10 and 14 there was a transition region in which both types of flow could take place.

3.3.2 The flow of two-phase fluid through an expansion device.

A vast amount of literature exists on two-phase flow. This is a result of work conducted mainly in connection with atomic power generation, in particular with the prediction of the rate of escape of coolant in the event of damage to the cooling system. Because of this, most of the work has been done on flow through pipes. One of the earliest reports of two-phase flow through nozzles, however, was that of Tangren et al (63) who
studied the flow of an air/water mixture through de Laval and convergent nozzles. A theoretical treatment was given. In this it was assumed that choking effects, analogous to those in single phase compressible flow, would be present for values of the ratio of downstream to upstream pressure below a critical value. Under conditions of choking it was assumed that the velocity of the fluid at the throat of a nozzle would be equal to the velocity of a pressure pulse through the fluid. On the basis of these assumptions the above critical pressure ratio was calculated for a number of values of the ratio of air to water. These were then compared with experimentally determined values. It was found that the measured critical pressure ratios varied from about 0.43 to 0.60 for initial values of the ratio by volume of air to water of 0.19 to 2.0. Predicted values of the pressure ratio were in error by no more than 13%.

The work of Tangren and other studies of single nozzle and twin-orifice nozzle assemblies were reviewed in a comprehensive survey by Smith in 1963 (59). The author of this survey recommended that for two-phase flow of low quality through short expansion devices, which is the type of flow to be expected through the downstream orifice of the twin orifice spray generator of the present work, a homogeneous metastable flow model should be used. This was the model used by Tangren.

Edmonds and Smith (12) studied mass limiting two-phase flow of propellant 11 in a long nozzle, a short nozzle and also a straight tube. Measurement of mass flow-rate and exit plane pressure were made for a constant initial pressure at different values of vapour quality and receiver pressure. It was found that as the receiver pressure was lowered, the pressure at the exit plane of the three devices became constant, but only when the receiver pressure was lower than the exit plane pressure by a substantial margin. In the case of the straight tube this margin was 30 kN/m²; for the long nozzle the margin was about 60 kN/m² and for the
short nozzle 70 kN/m$^2$. Further, the corresponding critical pressure ratios based on the receiver pressure were 0.56, 0.53 and 0.60 for the tube, long and short nozzles respectively. Based on exit plane pressures however the corresponding pressure ratios were 0.61, 0.69 and 0.82. The measured mass flow-rate in the straight tube was found to become constant for the above critical pressure ratio. In the case of the long nozzle the mass flow-rate continued to rise slowly as the receiver pressure was lowered below the critical, while for the short nozzle the mass flow continued to rise significantly as the receiver pressure was lowered.

Thus for the nozzles, choking effects were exhibited with respect to exit plane pressure but not to mass flow-rate. This behaviour thus contrasts with that of a single-phase compressible fluid. In spite of these apparent inconsistencies, the homogeneous metastable model mentioned above was found to predict reasonably well the mass flow-rate for a receiver pressure at or below critical. The so-called Fauske model (14) based on thermodynamic equilibrium and slip between phases, was found also to give a reasonable prediction of mass flow-rate.

**Prediction of sonic velocity.**

From the last paragraph it is seen that some doubt exists as to whether or not the mass flow-rate exhibits true choking effects. As choking effects are considered to occur when the fluid is moving at a sonic velocity in the throat of an expansion device, further insight into the concept of mass limiting flow could possibly be obtained by reviewing the literature on the prediction and measurement of the velocity of pressure pulses in two phase flow.

Gouse and Brown (23) reviewed the literature on the velocity of sound in two-phase mixtures, published up to 1965. It was concluded that in liquid/gas mixtures the velocity of sound was considerably lower than that in a single phase fluid. The minimum sonic velocity was generally only a few percent of the velocity of sound in the gas, and occurred
at a void fraction of 0.5. Agreement between the few experimental results available and predicted values for two-phase two component mixtures was shown to be good. For example the minimum value of the sonic velocity in water containing bubbles was predicted to be 20 m/sec, which was the value experimentally determined. For two-phase one-component mixtures the authors quoted results for a pressure pulse in boiling water. The predicted value for the minimum sonic velocity was 17 m/sec while the corresponding minimum pressure pulse velocity ranged from 15 m/sec to 24 m/sec.

Henry, Golmes and Fauske (28) produced expressions for, and determined experimentally, the velocity of pressure waves in gas/liquid mixtures. Five flow regimes were considered. They were bubble flow, stratified and annular flow, droplet dispersed flow and slug flow. The results were presented as the variation, with void fraction, of the ratio of the two-phase velocity to the sonic velocity in the gas. Good agreement between theory and experiment was demonstrated. For bubble flow, the flow regime of greatest relevance to this present work, the above ratio is given by:

\[
\frac{\text{Two-phase sonic velocity}}{\text{gaseous phase sonic velocity}} = \left( a^2 + \frac{a(1 - a) \rho_1}{\rho_g} \right)^{-\frac{1}{2}}
\]

where \( a \) is the void fraction and \( \rho_1, \rho_g \) are the densities of the liquid and gaseous components respectively.

Again minimum velocity was shown to occur at a void fraction of 0.5, and in the case of air in water at normal pressure and temperature this minimum sonic velocity was approximately 23 m/sec. This was in good agreement with their experimental value and also in fairly good agreement with the data quoted by Gouse and Brown above.

Moody (14) presented a pressure pulse model for two-phase critical flow and sonic velocity. From the paper and the subsequent discussion it
was concluded that, as demonstrated by the previous paper, the sonic velocity in a two-phase mixture was readily predictable. However doubt exists as to the predictability of critical velocity owing to difficulty in assessing conditions at the point of choking.
3.4 Conclusions from the literature.

It is apparent from a survey of the literature that fairly adequate empirical descriptions of the size distributions of pressure atomized and pneumatically atomized sprays were available. A theoretical basis for these descriptions however was generally lacking, although the primary break-up mechanisms appear to be reasonably well understood. In the case of atomization of saturated liquids, well established empirical relationships are lacking, although trends of variation of particle size distribution with such factors as temperature, propellant pressure and concentration, and the concentration and viscosity of the solute are available, although little attempt has been made to explain these trends theoretically.

One aspect of the theoretical treatment of pressure and pneumatic atomization can be applied directly to atomization of saturated liquids. This is the criterion of secondary atomization which may be applied to determine the maximum droplet size in a spray. This criterion was shown to be that the droplets would further break up if the Weber number exceeded a value of about 5 for sudden application of the disrupting force. The Weber number for a particle diameter, \( d \), is given by:

\[
\frac{1}{2} \rho_s \frac{u^2}{\sigma} \cdot d
\]

For a pure, solute free, propellant the surface tension is about \( 12 \times 10^{-3} \text{ N/m} \) (12 dynes/cm). Assuming injection at 30 m/sec (from Section 6) into still air \( (\rho_g = 1.2 \text{ kg/m}^3) \), then:

\[
W_e = 0.45 \times 10^5 \cdot d > 5
\]

or \( d > 100 \mu\text{m} \).
The effect on the surface tension of a surface-active solute may next be considered. This has been discussed by Lange (37). It was shown that a surfactant in a concentration of about 1% or more could reduce the equilibrium surface tension by as much as 50%. However, it was also shown that when a new surface is formed, it is free from surfactant molecules, and considerable time is required for the surface tension to reach its equilibrium value. This time may be of the order of seconds. It is likely therefore that the value of the surface tension of newly formed droplets just outside the nozzle is little lower than that of the pure liquid propellant. In this case the above criterion, that only droplets above 100 μm dia. will be subjected to further break-up, would still be applicable. Now, photographs of sprays at the nozzle exit of efficient atomizers show little evidence of droplets larger than this. Consequently, it may be concluded tentatively that secondary atomization plays an insignificant role in the atomization of saturated liquids through two-orifice nozzles.

Velocity decay and entrainment of air into the spray may next be considered. These effects are intimately related in that it is the transfer of momentum to the entrained air that is the cause of the axial velocity decay. These effects are well documented for air jets, and application of the principle of conservation of momentum has enabled analogous expressions to be derived and verified for fan and swirl water sprays. It has been shown (40) that a general expression could be obtained to describe velocity decay and entrainment within a spray, providing initial, nozzle exit, conditions were assumed and simplification were made regarding the transverse distributions of velocity.

From the review of the literature it was decided that the following programme of work on sprays generated by passing a saturated liquid through a two-orifice nozzle should be followed. Firstly, an expression for the velocity decay would be developed, based on momentum conservation in terms...
of the fundamental parameters of the system. The initial momentum of the
spray, given in terms of mass flow-rate and initial spray velocity, would
be determined from the assumption of homogeneous metastable flow through
the downstream orifice. The validity of these predictions would be
assessed experimentally from measurements of pressure, temperature and
mass flow-rate of the propellant, and also from size and velocity data of
the spray; the latter would be obtained from double-exposure photomicrography.

The second part of the programme would be an attempt to extend or
consolidate existing knowledge regarding size distributions from such sprays;
if possible this information on the size distribution would be again
related to fundamental parameters of the system.
4. THEORY

Introduction

4.1 Axial distribution of the mean velocity within a spray.

4.2 Flow of a saturated liquid through a two-orifice nozzle.

Introduction

The formation of an aerosol spray by the passage of a saturated liquid through a two-orifice* nozzle assembly may be summarised as follows. Initially the propellant in the liquid phase, passes through the upstream orifice into the low pressure region of the expansion chamber. There flash evaporation creates a turbulent mixture of vapour and liquid ligaments. This two-phase fluid then discharges from the downstream orifice in the form of a turbulent jet, consisting of fine ligaments, droplets and vapour. The surrounding air is entrained, by turbulent mixing, into the jet which consequently diverges and slows down.

Within the jet, droplets initially formed at the exit of the downstream orifice may break up further as a result of surface tension forces or impact with the surrounding air; coalescence of droplets may also take place.

The final stage is the evaporation of the propellant. When only propellant is the spray-generating liquid, the result is a jet of propellant vapour and entrained air. In the case of a pharmaceutical spray whose propellant contains a solute or suspended powder, the jet also contains an aerosol of fine solute or powder particles.

The spray properties of interest have been discussed in the first section. There, it was shown that the properties of the spray most relevant to inhalation therapy were the velocity and size distribution of the spray particles. It was therefore considered desirable to be able to

* They resemble "orifices" although, as described later, they are "short tubes". 
predict these parameters theoretically.

It is apparent from the above description of spray formation that both of these parameters involve a number of related physical phenomena and that the theoretical description of either would be a multi-facet problem. The prediction of spray velocity requires the knowledge of the initial momentum of the spray together with the degree of entrainment of the surrounding air. The prediction of size distribution within the spray requires the prediction of the size distribution at the orifice together with knowledge of the degree of secondary atomization, coalescence and evaporation within the spray.

An investigation into the possibility of deriving a fundamental theory to describe the above two parameters together with experience gained from the preliminary work led to the following conclusions. The first was that the description should be confined to a continuous spray only. The reasons for this limitation were described in Section 2 where it was shown that the discharge of a metered spray because of its transient nature was not readily amenable to mathematical treatment, and would add undesirable complexity to the problem. The second was that it seemed likely that the various stages of the spray development, nozzle flow, jet development etc. could be predicted and that by combining these a complete description of the velocity profiles within the spray could be obtained in terms of the nozzle dimensions and the thermodynamic properties of the fluid with the need for the minima of assumptions and empirical factors. The third conclusion was that it would be unlikely that the particle size distribution could be similarly described. This is based on the experience reported by many authors, who found that particle size distribution could only be predicted empirically. Perhaps the best known example is that of Nukiyama and Tanasawa (46) who produced empirical expressions for the size distribution of pneumatically produced sprays. Consequently an empirical
approach was taken for this second part of the work.

It is the purpose of this Section therefore to develop an expression by means of which the velocity within the spray could be determined in terms of the fundamental properties of the system with resort to as few assumptions and empirical factors as possible. As transverse velocity profiles can usually be assumed to be Gaussian, as discussed in the literature, only the axial velocity profile need be considered. Thus, in the first part of the Section an expression for the axial velocity profile is developed in terms of the initial momentum of the spray. In the second part the initial momentum of the spray is determined in terms of the fundamental properties of the system. Therefore a relationship between these properties and the axial velocity profiles is established.
4.1 Axial distribution of mean velocity within a turbulent spray in terms of the initial momentum of the spray.

In order to develop expressions for velocity profiles within a turbulent spray, it is first useful to consider corresponding expressions for single-phase fluids, for example an air jet discharging into the atmosphere. Detailed qualitative and quantitative descriptions of air jets are given by many authors including Pai (49) and Townsend (66). A very brief summary of these properties will now be given.

A turbulent air jet discharging from a cylindrical nozzle into the atmosphere is shown diagrammatically in Fig. 4.1. The jet consists of a number of well defined regions. At the exit of the nozzle there is a region of potential flow which is non-turbulent. Surrounding this is a cylindrical, turbulent mixing layer. At about four diameters from the nozzle the potential flow disappears and the whole jet becomes turbulent, entraining the surrounding atmosphere. This is the transition region. Beyond this region the flow becomes fully turbulent, with an apparent origin downstream of the orifice. The initial momentum of the jet at the nozzle exit is of course preserved. The jet thus slows down as part of this initial momentum is transferred to the entrained atmosphere.

The axial velocity profile of a fully turbulent air jet from a cylindrical nozzle may be described in dimensionless form as (66):-

\[ \frac{u_a}{u_o} = \frac{5.2}{(x - z)/D} \quad \ldots \quad 4.1.1 \]

where \( u_a \) and \( u_o \) are the axial and exit velocity respectively; \( D \) is the nozzle diameter and \((x - z)\) is the distance downstream of the apparent origin of the spray; the latter is usually between 0.5 and 1.5 diameter downstream of the nozzle.

The transverse profile of velocity is approximately Gaussian. Thus:

\[ u/u_a = e^{-100(y/x)^2} \quad \ldots \quad 4.1.2 \]

where \( u \) is the longitudinal velocity at a distance \( y \) from the axis.
**Fig. 4.1** DEVELOPMENT OF A TURBULENT CIRCULAR AIR JET

**Fig. 4.2** SCALE DRAWING OF A CONTINUOUS SPRAY TAKEN FROM FLASH AND LONG EXPOSURE PHOTOGRAPHS. THE NOZZLE ASSEMBLY CONSISTED OF TWO THICKPLATE ORIFICES EACH OF DIAMETER 0.35mm
It follows from this equation that the divergence angle of the jet corresponding to the \(1/e\)-velocity locus is:

\[
\tan^{-1}\left(\frac{1}{100}\right), \text{ or } 5.8^\circ
\]

It also follows that the dimensionless transverse profile is independent of the distance downstream. This applies also to other properties of the jet, which is thus self-preserving.

The first equation, 4.1.1, can be readily derived from momentum conservation. Referring to Fig. 4.1, the initial momentum flux is:

\[
\dot{m} u_o = \frac{\pi}{4} D^2 \rho_{\text{air}} u_o^2
\]

The momentum of the jet is:

\[
\int_0^\infty 2\pi y \rho_{\text{air}} u_a^2 e^{-2(y/(x \tan a))^2} \, dy = \frac{\pi \rho_{\text{air}} u_a^2 x^2 \tan^2 a}{2}
\]

These two expressions may be equated providing the static pressure within the nozzle and the spray are equal. This requirement follows from the momentum equation. It would follow then that the surface integral of pressure over the control volume of Fig. 4.1 would be zero. Thus the momentum equation would reduce to:

Initial momentum flux = momentum flux at any transverse plane in the jet.

Therefore:

\[
\frac{\pi}{4} D^2 \rho_{\text{air}} u_o^2 = \frac{\pi}{2} \rho_{\text{air}} u_a^2 x^2 \tan^2 a
\]

or

\[
\frac{u_a}{u_o} = \frac{1}{2 \tan a} \frac{D}{x}
\]

From the constant of equation 4.1.1, it follows that:
\[
\frac{1}{2 \tan \alpha} = 5.2. \text{ Therefore } \alpha = 52^\circ, \text{ as before.}
\]

The above analysis applied to an air jet in which the pressure throughout the turbulent jet and that within the nozzle were essentially equal. Were these two pressures not equal then the momentum equation would need modification. Consider the case of an air jet issuing from a reservoir through a convergent nozzle when the downstream to upstream pressure ratio across the nozzle is below the critical. In this case the exit velocity will be sonic, and the pressure within the jet will be greater than ambient, being equal to:

\[
0.528 \times (\text{reservoir pressure}).
\]

On leaving the confines of the nozzle the jet expands through an oscillatory pattern of expansion and shock waves and its velocity rises above sonic. The velocity distribution of such a system was determined by Eggins and Jackson (13) using a laser doppler system for velocity measurement.

It is apparent therefore that in this case the effect of the excess nozzle pressure will have to be added to the simple momentum equation given above. Therefore to the initial momentum flux \( M u_0 \) must be added:

\[
(\text{nozzle area}) \times (\text{excess pressure}).
\]

In the case of two-phase flow, analogous situations may be found. This was discussed in the literature survey. Thus for two-phase flow through the downstream orifice (short tube) of a two-orifice nozzle assembly of the type used in this work, critical flow is expected if the expansion chamber pressure is greater than about 2 atmospheres absolute.

In Fig. 4.2, is shown the outline of an experimental spray for an expansion chamber pressure of about 360 kN/m² abs (approximately 3.6 atmospheres absolute). The outline was traced from a photograph of the spray taken at a distance of about 1m. The expansion region at the nozzle
exit is clearly seen, thus offering confirmation that choking effects are present in the downstream orifice.

Two situations may then be considered; firstly that relating to a relatively low expansion chamber pressure when no choking effects are present and secondly the situation with high expansion chamber pressure when choking effects must be taken into account. The non-choking situation will be considered first.

**Velocity profile within a spray not exhibiting choking effects**

In order to develop an expression for the velocity profile within such a spray the initial momentum of the two-phase propellant emerging from the nozzle may be equated to that of the spray downstream of the nozzle. The situation may be envisaged from Fig.4.2 providing the expansion region at the nozzle exit is ignored and the spray is assumed an apparent origin downstream of the nozzle. This initial rate of efflux of momentum of the emerging propellant is:

\[ \dot{M} u_0 \]

where \( u_0 \) is the exit velocity of the propellant or a weighted mean velocity of the propellant if the vapour and liquid phases do not emerge with the same velocity. This latter point is considered later.

At a distance \( x \) from the nozzle, part of this initial momentum has been transferred to entrained air, and the spray will have slowed down.

The total momentum flux within the spray is now that of the entrained air together with that of the propellant liquid and vapour. Therefore, assuming the vapour and entrained air have the same velocity, \( u_g \), the total momentum flux is:

\[
\int_0^\infty 2\pi y u_g^2 (1 - \frac{c_g}{\rho_g}) \rho_{air} \, dy + \int_0^\infty 2\pi y u_p^2 c_p \, dy + \int_0^\infty 2\pi y u_g^2 c_g \, dy \\
= \dot{M} u_0
\]

\[ \cdots \cdots \cdots \cdots \cdots \cdots 4.1.3 \]
In this equation \( u_p \) is a mean velocity of the liquid propellant (particles); \( c_g \) and \( c_p \) are, respectively, the concentration by mass of propellant vapour and particles in the spray, while \( \rho_g \) and \( \rho_{air} \) are respectively the densities of the propellant vapour and air.

Now from consideration of conservation of mass:

\[
\dot{M} = \int_0^{2\pi} \int_0^\infty u_p c_p \, dy + \int_0^{2\pi} \int_0^\infty u_g c_g \, dy .... 4.1.4
\]

It is necessary at this stage, to make assumptions about the distribution of the three variables \( u_g \), \( c_g \), and \( c_p \). It was shown by Reichardt(54) among others that within a jet of homogeneous gas, the transverse variation of longitudinal velocity is Gaussian, of the form:

\[
u_g = u_{ga} e^{-y^2/\sigma^2},
\]

where \( u_{ga} \) is the axial velocity and \( \sigma \) represents the distance from the axis where the velocity falls to \( u_{ga}/e \). It is shown experimentally in Section 6 that such a function represents also the variation of velocity within a spray generated by a liquified gas.

It has been shown also (49,21) that the concentrations of vapour and small particles also follow similar distribution. The vapour concentration \( c_g \) may therefore be expressed as:

\[
c_g = c_{ga} e^{-y^2/\phi^2}
\]

and similarly, the droplet concentration, \( c_p \), may be expressed as:

\[
c_p = c_{pa} e^{-y^2/\omega^2}
\]

where \( c_{ga} \) and \( c_{pa} \) are, respectively, the mass of propellant vapour and liquid propellant per unit volume of spray, measured on the axis of the spray.
Two further assumptions will be made. The first is that the particles and vapour have the same velocity; that is \( u_p = u_g \). The second is that transverse distributions of vapour concentration and the particle concentration are proportional to that of the longitudinal velocity; that is:- \( \phi = 1 \sigma \) and \( \omega = k \sigma \).

The first of these two assumptions appears rather drastic. Indeed in Section 6 it is demonstrated that beyond a few centimeters from the nozzle the particle velocity increases with the square of particle diameter, and for the larger particles of the spray, of diameter about 30 \( \mu m \), the particle velocity can be of the order of 50\% higher than that of the gas. However it can be shown that the contribution to the total momentum flux by the particles is generally small. This is because, as the spray develops the mass of entrained air greatly exceeds that of the initial propellant; further the droplets are evaporating rapidly, further reducing their contribution.

Consider the second assumption. Pai (49) quoting numerous authors, and Goldschmidt and Eskinazi (21), describing their own experimental work have shown that the widening of the three transverse profiles of velocity, vapour concentration and aerosol concentration, are each proportional to the downstream distance; consequently these three parameters \( \sigma, \phi \) and \( \omega \) may be considered to be mutually proportional. Further the three angles of divergence of those profiles are shown to be approximately equal. For example Pai gives the divergence angle for the 1/e – velocity locus as 5.7\(^\circ\) for a circular jet of carbon dioxide in air, while concentration divergence angle is 6.2\(^\circ\), that is, greater by 9\%. Corresponding figures for aerosol concentrations from (21) are not so straightforward. However the half-width of the velocity and aerosol concentration profiles were found to differ by only about 7\% at 70 nozzle diameters downstream, with the concentration distribution again being wider.

Therefore the values of \( k \) and \( l \) may be considered constant and
approximately equal to 1. It will be shown that the expression for the axial velocity is insensitive to variation of \( k \) and \( l \) about a value of 1.

Substitution of the Gaussian distribution into equations 4.1.3 and 4.1.4 and integrating gives:

\[
\dot{m} u_o = \pi u_{ga}^2 \sigma^2 \left( c_{pa} \frac{k^2}{2k^2 + 1} + c_{ga} \left( 1 - \frac{\rho_{air}}{\rho_g} \right) \frac{l^2}{2l^2 + 1} + \frac{\rho_{air}}{2} \right)
\]

and \( \dot{m} = u_{ga} \sigma^2 \left( c_{pa} \left( \frac{k^2}{2k^2 + 1} \right) + c_{ga} \left( \frac{l^2}{1 + l^2} \right) \right) \) ...

4.1.5

Inserting 4.1.6 into 4.1.5:

\[
\dot{m} u_o = \dot{m} u_{ga} \left( \frac{c_{pa} \left( \frac{k^2}{2k^2 + 1} \right) + c_{ga} \left( 1 - \frac{\rho_{air}}{\rho_g} \right) \frac{l^2}{2l^2 + 1}}{c_{pa} \frac{k^2}{k^2 + 1} + c_{ga} \frac{l^2}{2l^2 + 1}} \right)
\]

\[+ \frac{\pi}{2} \sigma^2 \rho_{air} u_{ga}^2 \] ...

4.1.7

This may be further simplified by considering the coefficient of \( \dot{m} u_{ga} \). Because of the symmetry with respect to \( l \) and \( k \) the coefficient will be insensitive to variation of these two terms about their approximate value of 1.0. Therefore this value of 1.0 may be substituted for \( l \) and \( k \). Further the numerical value of this coefficient is restricted to the rather narrow range from 2/3 to \( \frac{2}{3} \left( 1 - \frac{\rho_{air}}{\rho_g} \right) \); it will take the value of 2/3 close to the nozzle when \( c_{pa} \) is large compared with \( c_{ga} \) and it
will take the value $\frac{2}{3} (1 - \frac{\rho_{\text{air}}}{\rho_g})$ away from the nozzle as the propellant evaporates and $c_{pa}$ becomes small compared with $c_{ga}$.

Now, the density of the vapour comprising, by weight, 60% $P_{12}$ and 40% $P_{11}$ at atmospheric pressure and 20° C may be shown to be approximately 5.4 kg/m$^3$, therefore:-

$$\frac{2}{3} (1 - \frac{\rho_{\text{air}}}{\rho_g}) = \frac{2}{3} (1 - \frac{1.2}{5.4}) = 0.52$$

Providing the entrained air and propellant vapour are at about the same temperature then this value is largely independent of temperature. Thus the coefficient may vary from 0.67 to 0.52.

As the region of greatest interest will be at a distance downstream of the nozzle, the value of 0.52 will be taken.

Thus equation 4.1.7 reduces to:

$$\dot{M} u_o = 0.52 \dot{M} u_{ga} + \frac{\pi}{2} \sigma^2 \rho_{\text{air}} u_{ga}^2$$ \hspace{1cm} 4.1.8

For a conical spray diverging at a half-angle, $\sigma$ :-

$$\sigma = (x - z) \tan \sigma.$$ 

Therefore:-

$$\dot{M} u_o = 0.52 \dot{M} u_{ga} + \frac{\pi}{2} (x - z)^2 \tan^2 \alpha \rho_{\text{air}} u_{ga}^2$$ \hspace{1cm} 4.1.9

This equation may be solved for $u_{ga}$. Alternatively, $u_{ga}$ may be expressed more explicitly by rearranging thus:-

$$u_{ga} = \frac{1}{(x - z)\tan \alpha} \sqrt{\frac{2 \dot{M} u_o}{\pi \rho_{\text{air}}} (1 - 0.52 \frac{u_{ga}}{u_o})}$$
Now at a few centimetres downstream of the nozzle, \( u_{ga} \) falls considerably below \( u_o \) and with little loss in accuracy

\[
(1 - 0.52 \frac{u_{ga}}{u_o})^\frac{1}{2}
\]

may be put equal to

\[
(1 - 0.26 \frac{u_{ga}}{u_o}).
\]

Therefore:

\[
\frac{1}{(x - z) \tan \alpha} \sqrt{\pi \frac{2 \dot{M} u_o}{\rho_{air}}} \]

\[
= \frac{1}{1 + \frac{2.6}{(x - z) \tan \alpha} \sqrt{\frac{2 \dot{M}}{\pi u_o \rho_{air}}}}
\]

\[
\frac{1}{(x - z) \tan \alpha} \sqrt{\frac{2 \dot{M}}{\pi \rho_{air}}}
\]

\[\ldots\ 4.1.10\]

This equation may be further simplified in the case of a jet at low Mach number where compressibility effects can be ignored and at a distance of many centimetres downstream from the nozzle where the denominator becomes approximately equal to 1. Calling this denominator \( f \), and letting the term representing momentum flux \( \dot{M} u_o = J \) then:

\[
\frac{1}{f(x - z) \tan \alpha} \sqrt{\frac{2 J}{\pi \rho_{air}}}
\]

\[\ldots\ 4.1.11\]

where \( f \) is approximately 1.

Now, for the case of a fluid jet of a low Mach number, of less than about 0.3 say, flowing from a cylindrical nozzle or short tube, the momentum flux is given by:

\[
2 C_Q \cdot C_{vel} \cdot A \cdot (P_e - P_{amb})
\]

where \( C_Q \) is the discharge coefficient and \( C_{vel} \) the velocity coefficient. \( P_e - P_{amb} \) is the gauge pressure upstream.
of the nozzle or tube; in the case of a two-orifice (short tube) nozzle assembly, this pressure is the expansion chamber pressure. A is the area of the jet which is equal to the area of the nozzle or tube.

As the area of the jet is equal to the area of the nozzle then,

$$C_{vel} = C_{Q}$$

and the equation becomes:

$$J = 2C_{Q}^{2}A(P_{e} - P_{amb})$$ ... 4.1.12

Thus 4.1.11 becomes:

$$u_{ga} = \frac{1}{f(x - z)\tan\alpha} \sqrt{\frac{4C_{Q}^{2}A(P_{e} - P_{amb})}{\pi \rho_{air}}}$$ ... 4.1.13

**Velocity profile within a spray exhibiting choking effects**

Equation, 4.1.10 was developed for non choking-flow when the nozzle exit pressure would equal ambient; equation 4.1.13 was further restricted to Mach numbers less than about 0.3. The development of a corresponding equation for choking flow will now be considered.

It is apparent that the previous equation 4.1.10 can be used directly providing \( u_{o} \) can be replaced by an exit velocity term determined in a region where the static pressure is equal to atmospheric, but before any significant entrainment has taken place. An alternative approach using equation 4.1.10, as discussed earlier, is to add to the term giving momentum flux at the point of choking, the product:

\((\text{Orifice area})(\text{static pressure (gauge)} \text{ in the short tube at the point of choking})\)

as required by the momentum equation.
The term giving momentum flux at the point of choking is \( \dot{M} u_s \) where \( u_s \) is the velocity of a pressure pulse through the fluid, or the "sonic" velocity. Thus the modified momentum term becomes:

\[ \dot{M} u_s + A(P_c - P_{amb}) \]

where \( P_c \) is the absolute static pressure at the point of choking and the other terms are as before.

In the case of the application of the second equation, 4.1.13, to choking flow, it is found that errors become large as the Mach number approaches, and exceeds 1. For example, the momentum flux of air from a convergent nozzle with a parallel cylindrical exit section, which is just choked is, assuming ideal flow:

\[ J = 1.57 A(P_e - P_{amb}) \]

while the coefficient of \( A(P_e - P_{amb}) \) in equation 4.1.12 was 2.0. This is an error of 21% in \( J \) and 10% approximately in \( u_ga \).

However as equation 4.1.13 is very simple it will be retained as a rapid means of obtaining an approximate value of the spray velocity.

The application of equation 4.1.10, and its modification, to choking flow may now be considered in more detail.

In the case of the underexpanded air jet issuing from a convergent nozzle, described in the introduction of this sub-section, the former approach, that of determining \( u_o \) in a region of ambient pressure but before significant entrainment had taken place, would probably be inaccurate. This is because the oscillatory pattern of expansion and shock waves through which the jet returns to ambient pressure exists many nozzle diameters downstream of the nozzle; thus considerable entrainment may have taken place by the time atmospheric pressure is reached. The second approach, in the case of an air jet would be the
more suitable because the conditions within the nozzle of such a system are well established. The same is not true for two-phase flow however. In this case the behaviour of the two-phase propellant in the downstream orifice and just beyond the orifice exit is by no means completely predictable. However, from the two-phase flow literature a number of relevant conclusions can be made with considerable confidence. These are now considered. Firstly there is general agreement that the static pressure at the exit plane of a short tube is greater than ambient during mass limiting flow, and that this pressure falls to ambient at a few nozzle diameters from the exit plane. Secondly for fairly high quality flow, for example greater than about 20% steam in steam/water flow (33), this fall to ambient is accomplished by an initial excursion below ambient, a phenomenon similar to that of an air jet; for low quality flow, however, this effect is absent. Thirdly shock waves may be observed in high quality flow but are not present in low quality flow. This is attributed to the high momentum relaxation time of the liquid phase.

The application of equation 4.1.10 to the treatment of choking two-phase flow will be considered in the light of these three conclusions. From the point of view of producing a fundamental expression the modified equation, given again below, is the more satisfactory in that all the terms in the resultant expression are, in principle, predictable.

The expression is then:

\[
\frac{u_g}{a} = \frac{1}{r} \frac{1}{(x - z) \tan \alpha} \sqrt{\frac{2 \left( \dot{m} u_s + A (P_c - P_{amb}) \right)}{\pi \rho_{air}}}
\]

However as will be seen in the second part of this section, many assumptions are required to produce expressions for the sonic velocity and the critical
pressure. Consequently the former method would enable a more direct comparison between theory and practice. This method is considered applicable because in this case of low quality two-phase flow it is expected that there will be no oscillator pressure wave pattern; instead the pressure is expected to fall rapidly to ambient within a few nozzle diameters. Thus an exit velocity in a region of ambient pressure would be measurable before significant entrainment had taken place. Thus both methods will be considered, but for the purposes of direct comparison between theory and experimental results, equation 4.1.10 will be used where $M$ is the mass flow-rate and $u_0$ is the velocity of spray a few nozzle diameters downstream of the nozzle. This velocity will be denoted by $u_n$.

A further point may be mentioned. This is that the liquid and gas phases will be assumed to pass through the orifice with equal velocity. This assumption is not in accord with the findings of Fauske (15) who determined gas/liquid velocity ratios for pipe flow from direct measurement of void fraction. It is however, substantiated by data presented in Section 6. The Fauske findings would predict a gas/liquid velocity ratio of about 1.2. This is considered further in the next part of this section.

In order to reduce to a certain extent, the number of empirical factors in this analysis, the possibility of equating the divergence angle of the spray, which has been shown to consist mainly of air, to that of an air jet will be considered. Limited experimental support will be given in Section 6.
4.2 Flow of a saturated liquid through a two-orifice nozzle assembly

In the previous sub-section, the axial velocity profile of a spray was described by equation 4.1.10 in terms of the mass flow-rate of propellant and a discharge velocity determined at a point where the static pressure within the spray was equal to ambient. In the case of a non-choking flow this discharge velocity was simply the nozzle exit velocity. For choking flow, equation 4.1.10 is still applicable providing an initial velocity, \( u_n \), exists and can be predicted, such that static pressure has fallen to ambient before significant atmospheric entrainment has taken place. In the case of choking flow, however, the alternative equation, 4.1.14 may be used. From this it is seen that in order to describe the axial velocity profile theoretically it is necessary to predict, in addition to the mass flow-rate, the critical or choking velocity and the critical pressure. It is the purpose of this sub-section to predict these three parameters for a two orifice nozzle assembly. The method adopted is outlined below. Firstly an expression for mass flow-rate through this upstream orifice is determined in terms of non-fundamental parameters. Secondly an expression for the mass flow-rate through the downstream orifice is determined also in non-fundamental terms. On equating these two expressions for the mass flow-rates, an equation for the mass fraction of propellant evaporated is obtained in terms of fundamental parameters of the system. From this expression, the mass flow-rate through the nozzle assembly can be determined, also in terms of fundamental parameters of the system. In the course of these derivations, fundamental equations for the critical velocity and pressure are generated.

4.2.1 Mass flow-rate through the upstream orifice

The propellant entering the upstream orifice (short tube) is in liquid form. The question then arises as to whether the propellant remains in liquid form during its passage through this orifice, or whether it will evaporate as a result of the decrease of the pressure. A criterion by means of which this question may be answered was developed by Pasqua (50). This
was described in the literature survey and is discussed more fully in Section 6 where it is demonstrated that the propellant will indeed remain as a liquid during its passage through the first orifice. The Bernoulli equation may then be applied to this flow. This is given in BS 1042 as:

\[
\dot{m} = C_{Q1} \frac{\pi}{4} (D_1)^2 \sqrt{\frac{2 \rho_1 (P_i - P_e)}{1 - \left(\frac{D_1}{D_e}\right)^4}}
\]

In this equation \( \rho_1 \) is the density of the liquid propellant, \( P_i \) is the absolute pressure upstream of the orifice and \( P_e \) is the absolute pressure in the expansion chamber. \( C_{Q1} \) is the discharge coefficient of the orifice. \( D_1/D_e \) is the ratio of orifice diameter to supply tube diameter. The largest orifice used was 0.7 mm and the tube diameter was 3.2 mm, therefore, to within 0.2\%, \( 1 - \left(\frac{D_1}{D_e}\right)^4 \) may be considered equal to 1.

\[
\dot{m}_1 = C_{Q1} \left(\frac{\pi}{4}\right) (D_1)^2 \sqrt{\frac{2 \rho_1 (P_i - P_e)}}
\]

Therefore:

4.2.2 Mass flow-rate through the downstream orifice.

(a) with a low pressure differential across the orifice.

The flow of a dispersed two-phase fluid through an orifice is shown in Fig. 4.3. The propellant passes through the downstream orifice in the form of a liquid-vapour mixture. Unlike the previous case no single, relatively simple, well proven theory exists which describes the passage of a two phase fluid through a nozzle.

In order to obtain an equation for the mass flow-rate in a workable
Fig. 4.3  

FLOW OF A T wo-Phase FLUID THROUGH A SHORT TUBE

\[ X\text{-Sectional area} = A \]
\[ \text{Mean fluid density} = \bar{\rho} \]
form, a number of assumptions must be made. They are discussed below.

(a) The flow through the orifice is considered to be one dimensional and also isentropic, that is adiabatic and frictionless. Entrance and viscous losses that arise in practice are taken into account by the introduction of a discharge coefficient $c_Q$.

(b) The propellant in the orifice is assumed to be highly dispersed, with no large scale separation of the phases. Photographic evidence presented in Fig. 2.17 supports this view.

(c) There is no relative velocity between the liquid and gas phases. This assumption was discussed in more detail in the previous subsection. In Section 6 experimental evidence is presented in its favour.

(d) It is assumed that as the length of nozzle is short, typically 0.7 mm, then there is no mass-transfer between phases during the passage of the propellant through the orifice. This point has been discussed in the literature survey. Fauske ('15) has shown that good agreement between theory and experiment may be obtained, even with relatively long nozzles, by making this assumption. It follows that the fraction of propellant evaporated - sometimes called mass dryness fraction or quality ($q$), remains constant during the passage of the propellant through the orifice.

(e) For the same reasons expressed in (d) above it is assumed that there is no heat transfer between phases during transit through the orifice.

(f) The propellant vapour is considered to be a perfect gas.

It follows from assumptions (d), (e) and (f) that during passage through the orifice the propellant vapour obeys the isentropic gas
equation:--

\[ \frac{P}{\rho_g^\gamma} = \text{constant} \]

where \( \gamma \) is the ratio of specific heats at constant pressure and volume for the propellant vapour.

Referring to Fig. 4.3 a differential element of fluid is in the process of passing through the orifice from the expansion chamber at an absolute pressure of \( P_e \) to the orifice exit where the pressure is \( P_0 \).

Assuming no irreversibility then the exit velocity \( (u_0) \) is obtained by integrating the one-dimensional Euler equation.

\[
\frac{u_0^2}{2} = \int_{P_e}^{P_{\text{amb}}} \frac{dP}{\bar{\rho}} \quad \ldots \quad 4.2.2
\]

where \( \bar{\rho} \) is the mean density of the two-phase propellant.

Now:

\[
\frac{1}{\bar{\rho}} = \frac{q}{\rho_g} + \frac{1-q}{\rho_l} \quad \ldots \quad 4.2.3
\]

Therefore:

\[
\frac{u_0^2}{2} = \frac{1-q}{\rho_l} (P_e - P_{\text{amb}}) - \int_{P_e}^{P_{\text{amb}}} \frac{q}{\rho_g} dP \quad \ldots \quad 4.2.4
\]

It was shown above that during the passage through the orifice \( P \) and \( \rho_g \) are related by the equation: \( \frac{P}{\rho_g^\gamma} = \text{constant} \), or

\[ \frac{P}{\rho_g^\gamma} = \text{constant} \]
\[ \frac{p}{p_e} = \frac{p_e}{\rho_{gs}} \]

where the subscript "e" refers to the expansion chamber.

\[ \therefore \]

4.2.5

\[ p_{amb} \]

Therefore: \[
\frac{u_0^2}{2} = \frac{(1 - \gamma)}{\rho_1} (p_e - p_{amb}) - q \int_p \frac{1}{\rho_{ge}} \frac{1}{p} \frac{1}{\gamma} dp
\]

\[ \therefore \]

4.2.6

\[ \therefore \]

4.2.7

\[ \frac{u_0^2}{2} = \frac{(1 - \gamma)}{\rho_1} (p_e - p_{amb}) + q \frac{p_e}{\rho_{ge}(1 - \gamma)} (1 - \frac{1}{\gamma}) \left( p_e - p_{amb} \right) \]

\[ \therefore \]

4.2.8

The mass flow-rate is given by:

\[ \dot{m}_2 = C_{Q_2} \frac{\pi}{4} (D_2)^2 \rho_0 \frac{u_0}{2} \]

where \( \rho_0 \) is the density of the two-phase fluid at the orifice exit.

4.2.3 Mass flow-rate through the downstream orifice.

(b) with a high pressure differential across the orifice.

The derivation of the expression for the exit velocity and mass flow-rate of a two-phase fluid (equations 4.2.7 and 4.2.8) did not include any restriction on the magnitude of the ratio of the orifice exit pressure to the expansion chamber pressure \( \left( \frac{p_o}{p_e} \right) \) or an upper limit to the value
of the exit velocity $u_0$. As discussed earlier, however, on lowering the value of the pressure ratio across a nozzle discharging a compressible fluid, the exit velocity $u_0$ reaches a maximum, which is the velocity of a pressure pulse through the fluid. Ideally, further reduction of downstream pressure therefore would not increase the velocity, or mass flow-rate through the nozzle. Under these conditions the flow is described as critical, choked, or mass-limiting.

A number of authors have shown that choking flow occurs when liquid and vapour mixtures are discharged through nozzles, providing the ratio of the downstream to upstream pressure is less than about 0.5, (for example (63,12)).

It is considered that choking phenomena must be taken into account when describing the flow through the downstream orifice (short tube). This is because the pressure of the propellant in the expansion chamber during continuous flow discharge is typically 300 to 380 kN/m$^2$ and the absolute pressure ratio across the orifice is therefore 0.33 to 0.26. These values are considerably lower than the generally accepted critical pressure ratio of about 0.5, indicating that choking flow conditions should prevail within the downstream orifice. Although mass flow-rates at varying ambient pressures were not made, it is considered that the existence of mass limiting flow was confirmed by the rapid expansion at the nozzle exit of the jet discussed in sub-section 4.1.

Referring to Fig.4.3, the pressure ratio $P_o/P_e$ is considered to be small and consequently mass limiting flow conditions are considered to prevail. The maximum velocity in the orifice is the velocity of a pressure pulse in the two-phase mixture ($u_s$).

An expression for velocity $u_s$ of a wave or pressure pulse through an isotropic homogeneous elastic medium is: $\frac{1}{u_s^2} = -\frac{dP}{dP}$
Accepting the assumptions discussed previously then:

\[ \frac{1}{u_s^2} = \frac{d}{dP} \left( \bar{\rho} \right) \] \hspace{1cm} \ldots 4.2.9

Therefore:

\[ \frac{1}{u_s^2} = \frac{d}{dP} \left( \frac{\rho_1}{q \rho_1 + (1 - q) \rho_g} \right) \] \hspace{1cm} \ldots 4.2.10

From 4.2.5

\[ \rho_g \propto \frac{1}{P^\gamma} \]

Therefore it may be shown that:

\[ u_s^2 = \frac{\gamma q P}{\rho_g} \left( 1 + \frac{1 - q}{q} \frac{\rho_g}{\rho_1} \right)^2 \] \hspace{1cm} \ldots 4.2.11

Assuming that the maximum velocity of the spray, under choking conditions, is equal to the velocity of a pressure pulse through the two-phase fluid, then this maximum velocity is given by \( u_s \). Denoting, by a subscript \( c \), the value of a variable at the point of choking then:

\[ u_s^2 = \frac{\gamma c P_c}{\rho_g} \left( 1 + \frac{1 - q_c}{q_c} \frac{\rho_g}{\rho_1} \right)^2 \] \hspace{1cm} \ldots 4.2.12

The critical mass flow-rate through the downstream orifice is then given by:

\[ \dot{m}_2 = C_{q2} \left( \frac{\pi}{4} \right) (D_2)^2 \left( \bar{P}_o \right) u_s \] \hspace{1cm} \ldots 4.2.13

where \( C_{q2} \) is the discharge coefficient of the downstream orifice.
of diameter \( D_2 \), and \( \bar{\rho}_c \) is the mean density of the propellant at the point of choking.

Now:-
\[
\left( \frac{\bar{\rho}_c u_s}{\gamma_0} \right)^2 = \frac{\frac{1}{2} \frac{\rho_{gc}^2}{\rho_1}}{ \left( \frac{\rho_1}{\rho_1} + (1 - q_c) \frac{\rho_{gc}}{\rho_{gc}} \right)^2} \times \frac{\gamma_0 \frac{q}{\rho_0} \frac{P_c}{\rho_{gc}}}{\left( 1 + \left( \frac{1 - q_c}{q_c} \right) \frac{\rho_{gc}}{\rho_1} \right)^2} = \frac{\gamma_0 \frac{\rho_{gc} P_c}{q_c}}{q}
\]

Therefore:
\[
\dot{M} = \frac{C Q_2}{4} \left( \frac{\gamma_0 \frac{\rho_{gc} P_c}{q_c}}{q_c} \right)^{\frac{1}{2}} \cdot 4.2.14
\]

Equations 4.2.13 and 4.2.14 contain parameters which must be evaluated at the point of choking, generally within the orifice, where these values are generally unknown. It is essential, therefore, that these equations are expressed in terms of variables evaluated under expansion chamber conditions which are more readily predicted. The variables in question are the specific heat ratio \( \gamma_c \), the quality of the propellant \( q_c \), the density of the propellant vapour \( \rho_{gc} \) and the critical pressure \( P_c \). They are each considered below. The specific heat ratio \( \gamma_c \) is a slowly varying function and an average value for the point of choking may be found. This value is determined in Section 6. The subscript \( c \) will thus be ignored.

Secondly consider the quality of the propellant, \( q_c \). This is assumed to remain constant during passage of the propellant through the downstream orifice. Therefore \( q_a = q_c = q \). Thirdly \( \rho_{gc} \) is assumed to be related to the pressure by the expression \( \rho_{gc} \propto P_c^\gamma \). It remains therefore only to relate the value of the critical pressure in the orifice \( P_c \) to the value of the pressure in the expansion chamber \( P_e \). This may be done (63) by determining a "critical pressure ratio" \( r_c = \frac{P_c}{P_e} \), whose value is shown to be
approximately constant; then \( P_c = r_c \rho_e \). The critical pressure cannot be equated to the value of the ambient pressure because, as discussed previously, the pressure in the jet during critical flow is generally greater than the ambient pressure.

The critical pressure may be found, however, by allowing \( P_{amb} \) in equation 4.2.7, the expression for the exit velocity in non-choking flow, to fall to the critical pressure. Then, the exit velocity, \( u_o \) of equation 4.2.7 becomes equal to the critical velocity \( u_s \) and the pressure in the jet is then just equal to the external pressure. Equating the expression for \( u_s^2 \) (equation 4.2.12) to \( u_o^2 \) (equation 4.2.7), gives:

\[
\frac{\gamma q P_c}{\rho_{gc}} \left( 1 + \left( \frac{1 - q}{q} \right) \frac{\rho_{gc}}{\rho_1} \right)^2
\]

\[
= 2 \left( \frac{1 - q}{\rho_1} \right) (\rho_e - P_c) + q \frac{P_e}{\rho_{gc}} \left( \frac{1}{1 - \gamma} \right)
\]

\[
= \frac{1}{\gamma} \left( \frac{P_e}{P_c} - \frac{1}{1 - \gamma} \right)
\]

\[
\cdots 4.2.15
\]

Now:

\[
\rho_{gc} = \frac{\rho_{gc}}{P_c} = \frac{1}{\gamma}
\]

where \( r_c \) is the critical pressure ratio \( P_c / P_e \).

By defining \( X_c = \left( \frac{1 - q}{1 - q} \right) \frac{\rho_1}{\rho_{gc}} \)

\[
\cdots 4.2.16
\]

where \( X_c \) represents the ratio of the volume of vapour to the volume of liquid at the point of choking, equation 4.2.15 may be reduced to:
\[\gamma x_c (1 + \frac{1}{x_c})^2 = 2 \left( \frac{1 - 1}{r_c} - 1 \right) + x_c \left( \frac{1 - 1}{}\right) \]

... 4.2.17

The value of \(\gamma\) is shown in Section 6 to be 1.17. Therefore, substituting for \(\gamma\), equation 4.2.17 may be solved graphically to obtain the variation of \(r_c\) with \(x_c\).

A graph of \(r_c\) against \(x_c\) is shown in Fig. 4.4. In practice \(x_c\) varies between 2 and 4. Therefore \(r_c\) varies between 0.47 and 0.54 and to first approximation may be considered to be constant.

Two expressions have now been derived for the mass flow-rate through each orifice. For steady flow \(\dot{M}_1 = \dot{M}_2\). From this relationship an expression for the temperature fall across the upstream orifice, \(\Delta T_1\), may be derived in terms of the fundamental parameters of the system. From the expression for \(\Delta T_1\), equations for the vapour quality, \(q\) and the pressure in the expansion chamber \(P_e\) are readily obtainable.

Now, \(\dot{M}_1 = C_{Q1} \left( \frac{\pi}{4} \right) D_1^2 \left( 2 \rho_1 (P_1 - P_2) \right)\), and using the equation for the mass limiting flow-rate for the downstream orifice (4.2.14):

\[\dot{M}_2 = C_{Q2} \left( \frac{\pi}{4} \right) D_2^2 \left( \frac{\gamma P_c}{q} \frac{P_{go}}{q} \right)\]

Therefore:
\[C_{Q1} \left( \frac{\pi}{4} \right) D_1^2 \left( 2 \rho_1 (P_1 - P_2) \right)^{\frac{1}{2}} = C_{Q2} \left( \frac{\pi}{4} \right) D_2^2 \left( \frac{\gamma P_c}{q} \frac{P_{go}}{q} \right)^{\frac{1}{2}}\]

... 4.2.18

In order to obtain the temperature fall \(\Delta T_1\), explicitly from this equation, a number of simplifications are necessary. These may be made by
FIG. 4.4. VARIATION OF CRITICAL PRESSURE RATIO \( r_c \) WITH THE RATIO BY VOLUME OF VAPOUR TO LIQUID \( X_c \)
considering in more detail the behaviour of the propellant
in the process of discharging through the nozzle assembly. Firstly
consider a relationship between the density of the propellant vapour, \( \rho_{ge} \),
in the expansion chamber and the expansion chamber pressure, \( P_e \). It is
shown in Section 6 that for a nozzle assembly whose orifices are of
approximately the same diameter, the temperature fall across the first
orifice is less than about 6 deg C. Thus to a first approximation the
temperature in the expansion chamber may be considered to be constant
providing the diameter of the second orifice is not more than about twice
that of the first. Therefore the density of the propellant vapour in
the expansion chamber may be taken to be proportional to the pressure
within the expansion chamber (\( P_e \)).

Therefore:\
\[
\rho_{ge} = C_1 P_e \quad \ldots \ldots 4.2.19
\]

where \( C_1 \) is determined for a temperature of about 16\(^\circ\) C.

Now, from before,
\[
\rho_{gc} = \rho_{ge} \left( \frac{P_c}{P_e} \right)^{\frac{1}{\gamma}}
\]

and also:\
\[
P_c = r_o P_e
\]

where \( r_o \) is approximately constant.

Therefore
\[
\rho_{gc} = C_1 P_e \left( r_o \frac{P_e}{P_e} \right)^{\frac{1}{\gamma}}
\]

or
\[
\rho_{gc} = C_1 P_e r_o \frac{1}{\gamma} \quad \ldots \ldots 4.2.20
\]

The above restriction on the ratio of orifice diameters or temperature
variation, does not detract from the value of this theory because, in
commercial practice, the diameters are usually approximately equal.

Maintaining this restriction on temperature variation, a further
simplification may be made. This is that mass fraction of propellant evaporated \( q \), is proportional to the temperature fall across the first orifice, \( \Delta T_1 \). Therefore:

\[ q = c_2 \Delta T_1 \]  
\[ \ldots \ldots 4.2.21 \]

In this equation \( c_2 \) is determined over the range 14°C to 20°C.

A third simplification will now be made. This is to express the pressure fall across the first orifice \( \Delta P_1 \) in terms of the temperature fall \( \Delta T_1 \), again assuming a restricted range of expansion chamber temperature. If conditions of thermodynamic equilibrium exist in the expansion chamber then \( P_e = SVP_{Te} \). Also if the propellant above the first orifice is saturated then \( P_i = SVP_{Ti} \). Therefore:

\[ \Delta P_1 = P_i - P_e = SVP_{Ti} - SVP_{Te} \]

For a small change in temperature the change in SVP is proportional to that temperature change. Therefore:

\[ \Delta P_1 = c_3 \Delta T_1 \]  
\[ \ldots \ldots 4.2.22 \]

This applies for equilibrium above and below the first orifice. If the propellant above the first orifice is sub-cooled by a head of propellant above the nozzle, as is usually the case, then :-

\[ P_i - P_e = \Delta P_1 = c_3 \Delta T_1 + \Delta P_{ex} \]  
\[ \ldots \ldots 4.2.23 \]

where \( \Delta P_{ex} \) is the pressure due to this head.

The simplified expression for \( \rho_{gc} \), \( q \) and \( P_i - P_e \) may now be substituted into equation 4.2.18. Therefore:-
\[ c_{q1} (D_1)^2 \sqrt{2 \rho_1 (c_3 \Delta T_1 + \Delta P_{ex})} \]
\[ = c_{q2} (D_2)^2 \sqrt{\frac{\gamma c_1}{c_2} \Delta T_1} \left( \frac{\gamma + 1}{\rho c_3 \Delta T_1 - \Delta P_{ex}} \right) \]

A value of \( \Delta T_1 \) corresponding to saturation conditions upstream of the first orifice may then be obtained by letting \( \Delta P_{ex} = 0 \) in equation 4.2.24.

Therefore:
\[ \Delta T_1 = \frac{P_1}{c_3 + \frac{1}{2\gamma} \left( \frac{c_{q1}}{c_{q2}} \right) \left( \frac{D_1}{D_2} \right)^2 \sqrt{\frac{2 \rho_1 c_2 c_3}{\gamma c_4}}} \]

Metastability

In obtaining equation 4.2.25, it was assumed that equilibrium conditions prevailed in the expansion chamber.

Fig. 6.20 shows the variation of measured pressure in the expansion chamber \( P_e \) with S.V.P. obtained from the measured temperature \( T_e \).

It is apparent that, in general, the propellant in the expansion chamber is not in thermodynamic equilibrium but in a metastable condition at a pressure lower than the S.V.P. corresponding to its temperature. However, as can be seen from Fig. 6.20, when the diameters of both orifices are less than about 0.5 mm and when the diameter of the second orifice is less than or approximately equal to that of the first then metastability effects are negligible; in these cases equilibrium may be assumed.

Metastability in the expansion chamber is obviously a measure of the
difference between the rate of escape of vapour through the second orifice and the rate of production of vapour by propellant in the expansion chamber.

Now, the rate of escape of vapour through the second orifice is relatively high if the quality of the vapour \( q \) is high. Further, the rate of production of vapour will be low if the residence time of the propellant in the expansion chamber is short. Therefore, an elementary model of metastability would indicate that metastability effects might be proportional to \( q \) and inversely proportional to some power of the residence time in the expansion chamber, the latter given by:

\[
\frac{\bar{P}}{\dot{M}} \times \text{(expansion chamber volume)}
\]

It will be shown in Section 6 that generally terms such as quality \( q \) and mass flow-rate \( \dot{M} \) can be expressed very simply in terms of the orifice diameters. Thus \( q \) is found to be proportional to \( (D_2 / D_1)^2 \) and \( \dot{M} \) to \( (D_1 D_2) \). It may be expected therefore that metastability defined as:

\[
\Delta P_{\text{meta}} = \text{SVP}_{T_e} - P_e,
\]

may be similarly expressed.

An empirical correlation between \( \Delta P_{\text{meta}} \) and \( D_1, D_2 \) was obtained from the data shown in Fig.6.20. The relationship is:

\[
\Delta P_{\text{meta}} = 1.80 \times 10^3 \text{(SVP at } T_e) \times \frac{D_2^4}{D_1^{2.5}}
\]

where \( D_1, D_2 \) are in metre.

This is plotted, together with the experimental data, in the Figure.

Metastability may now be incorporated in the expression for \( \Delta T_1 \).

Substituting \( \text{SVP}_{T_e} - \Delta P_{\text{meta}} \) for \( P_e \) and \( \text{SVP}_{T_i} + \Delta P_{\text{ex}} \) for \( P_i \), equation 4.2.23
becomes:

\[ P_i - P_e = C_3 \Delta T_1 + \Delta P_{ex} + \Delta P_{meta} \]

and therefore:

\[ c_{q1} (D_1)^2 \frac{1}{2} \rho_1 (C_3 \Delta T_1 + \Delta P_{ex} + \Delta P_{meta}) \]

\[ = c_{q2}(D_2)^2 \sqrt{\frac{\gamma C_1}{C_2 T_1}} \frac{\gamma + 1}{r_c \gamma^2} \left( P_i - c_3 \Delta T_1 - \Delta P_{ex} - \Delta P_{meta} \right) \]

\[ \ldots 4.2.28 \]

Or, letting \( \Delta P_{ex} \) become zero, as before,

\[ \frac{1}{r_c \gamma^2} \frac{c_{q1}(D_1)^2}{c_{q2}(D_2)^2} \sqrt{2 \rho_1 (C_3 \Delta T_1 + \Delta P_{meta})} \frac{c_2 \Delta T_1}{\gamma C_1} \]

\[ = P_i - C_3 \Delta T_1 - \Delta P_{meta} \]

\[ \ldots 4.2.29 \]

where \( P_i \) is now the S.V.P. of the propellant at \( T_1 \).

By substituting equation 4.2.27 into 4.2.29, \( \Delta T_1 \) may be obtained in terms of \( D_1, D_2, P_i \) and constants obtainable from thermodynamic data. However, equation 4.2.29 may be simplified by noting that for the range of nozzles tested in these experiments \( \Delta P_{meta} \) is always less than about a half of \( C_3 \Delta T_1 \). Therefore:

\[ \sqrt{C_3 \Delta T + \Delta P_{meta}} = \sqrt{C_3 \Delta T_1} \left( 1 + \frac{1}{2} \frac{\Delta P_{meta}}{C_3 \Delta T_1} \right) \]

This is accurate to within at least 3\%, while for nozzle assemblies with
\[ D_1 \geq D_2 \text{ the error is considerably less.} \]

Therefore:

\[
\frac{1}{r_c} \frac{1}{2y} \left( \frac{C_{Q1}}{C_{Q2}} \right) \frac{D_1^2}{D_2} \sqrt{2} \frac{C_3 C_2}{\gamma C_1} \left( \Delta T_1 + \frac{1}{2} \frac{\Delta P_{\text{meta}}}{C_3} \right)
\]

\[ = P_i - C_3 \Delta T_1 - \Delta P_{\text{meta}} \quad \ldots \ldots \text{4.2.30} \]

Therefore:

\[
\Delta T_1 = \frac{P_i - \Delta P_{\text{meta}}}{\left( 1 + \frac{1}{2r_c} \frac{1}{2y} \left( \frac{C_{Q1}}{C_{Q2}} \right) \frac{D_1^2}{D_2} \sqrt{2} \frac{\rho_1 C_3}{\gamma C_1} \right)}
\]

\[
\quad + \frac{1}{r_c} \frac{1}{2y} \left( \frac{C_{Q1}}{C_{Q2}} \right) \frac{D_1^2}{D_2} \sqrt{2} \frac{\rho_1 C_3 C_2}{\gamma C_1}
\]

\[ \ldots \ldots \text{4.2.31} \]

where \( P_i \) is now the S.V.P. at \( T_i \) and \( \Delta P_{\text{meta}} \) may be obtained from equation 4.2.27. It may be noted that \( P_i \) is much greater than the term containing \( \Delta P_{\text{meta}} \); the latter is thus a small correction for the effect of metastability on the temperature fall.

The application of this equation to the determination of mass flow-rate and expansion chamber pressure is demonstrated in Section 6.
The purpose of the experimental programme was two-fold; firstly, to provide experimental evidence in support of the theoretical expressions given in Section 4; secondly, to provide experimental data relating to continuous sprays from which empirical expressions may be developed.

The apparatus was designed to permit the study of a continuous spray. This was generated by the flow, through a two-orifice nozzle, of a saturated liquid at high pressure containing a surface-active solute of low volatility.

Particle size and velocity distributions of droplets at selected points within the spray could be measured together with other parameters relevant to spray behaviour. These were: the pressure and temperature of the propellant up-stream of, and inside, the nozzle assembly; the rate of flow of propellant; and the size distribution of the residual surfactant aerosol.

5.1 Experimental Apparatus

The continuous flow apparatus is shown diagramatically in Fig. 5.1 and photographically in Fig. 5.3. Details are given in Figs 5.2 to 5.7. A brass bottle supplied saturated liquid propellant via a flexible feed-pipe to a nozzle assembly mounted on a support capable of movement in two dimensions. The spray emerging from the nozzle could be photographed by a double-exposure photomicrographic unit. The spray could also be collected in a sedimentation cylinder (not shown). Temperatures and pressures at the various positions indicated could be measured.
PRESSURE I GAUGES.

PROPELLANT: 60% $P_1$, 40% $P_\Pi$ BY WEIGHT.

SOLUTE: 1% "SPAN 85" BY WEIGHT.

COPPER CONSTANTAN THERMOCOUPLES.

NOZZLE ASSEMBLY ON ADJUSTABLE MOUNTING.

DOUBLE SPARK UNIT.

INTERVAL TIMER.

DIGITAL VOLTMETER AND DATA LOGGER.

FIG. 5.1 DIAGRAM OF THE APPARATUS.
FIG. 5.2. NOZZLE ASSEMBLY.
FIG. 5.3 GENERAL VIEW OF THE EXPERIMENTAL APPARATUS.
FIG. 5.4  TWO-DIMENSIONAL MOVEMENT.
FIG. 5.5  NOZZLE ASSEMBLY.
FIG. 5.6 PHOTOMICROGRAPHIC UNIT.
FIG. 5.7. DOUBLE-SPARK UNIT AND CAMERA.
For these measurements copper-constantan thermocouples and Bourdon pressure gauges respectively, were used.

5.1.1. Feed-pipe and associated instrumentation

The flexible feed-pipe consisted of a polythene tube of 6.4mm. (¼ inch) inside diameter. It was connected at the upper end to the propellant container via a tap. At the lower end, the pipe was connected to a "T"-piece to the side of which was fitted a Bourdon pressure gauge. Also in the feed-path, below this "T"-piece, was a section of copper tube containing a copper-constantan thermocouple. The lower end of this section was connected to the nozzle assembly via a tap and a second "T" piece, whose side-arm was normally blanked off. This second "T" piece was used as a means of indicating the axis of the spray, as described in sub-section 5.3.

In order to avoid cavitation in the propellant and to ensure that a single-phase fluid was fed to the first orifice, the bottle was placed about 500mm above the nozzle; also constrictions in the feed pipe were avoided.

The outputs of the thermocouples in the feed-pipe and in the nozzle were read by the digital voltmeter (D.V.M.) of a data-logging unit which scanned each thermocouple in turn and punched the value of the thermal e.m.f. The resolution of the D.V.M. was 1 microvolt and as the sensitivity of a copper-constantan thermocouple is approximately 43 microvolt per degree centigrade, the temperature could be measured, ideally, to within 0.023 degrees C. In practice however, the "zero" of the D.V.M. drifted by a few microvolts and the practical temperature resolution was about 0.1 deg C.

5.1.2. The two-orifice nozzle assembly

The nozzle assembly is shown in Figs. 5.2 and 5.5. It consisted of two orifices separated by an expansion chamber. These were assembled
together with a 150mm long perspex tube, in a brass cylinder. The nozzles, which were made of perspex, were transparent and dismountable. This arrangement had three advantages. Firstly, it was possible to observe the two-phase fluid flow in the expansion chamber. Secondly, it was possible to ensure that bubbles, formed in the liquid approaching the first orifice at the start of spraying, had discharged from the system before measurements were taken. Thirdly, different orifice combinations could quickly be formed, and blockages easily removed.

Two difficulties were associated with the use of perspex in contact with the propellant. Firstly, according to the manufacturer's literature, perspex is sparingly soluble in halogenated hydrocarbons. Secondly, perspex tends to "craze" along stress lines. To test the effect of solubility, a nozzle was made up and the orifice diameters were measured with a microscope. Propellant was sprayed through the nozzle for many hours during preliminary trials. Afterwards, the diameters were again measured. No change in diameter was detected; therefore the effect of solubility was ignored.

Crazing along stress lines was a more serious problem. After a few minutes of spraying, cracks would appear in the perspex tube leading to the nozzle and in the perspex of the expansion chamber; the orifice sections, however, were unaffected. These difficulties were overcome by annealing and coating with Araldite embedding resin as explained below.

Nozzle manufacture

The nozzles were made as follows. A 150mm (6") perspex tube, of 12.7mm (½") outside diameter (O.D.) and 3.2mm (⅜") inside diameter (I.D.) was faced on a lathe, and an "O"-ring groove cut in one end. A second perspex tube 12.7mm (½") long was machined similarly with "O"-ring grooves cut in both ends. Each piece of perspex was annealed at 90°C.
in an oven for about half a day. Two small holes were then drilled through one side of the 12.7mm length of perspex. Through one hole, a copper-constantan thermocouple was inserted and, after degreasing, was sealed with resin. The other hole was tapped with a 5 B.A. thread and into it was screwed a hollow copper tube, similarly threaded; this was also sealed with resin and thus served as a pressure tapping. This 12.7mm length of perspex became the expansion chamber in which both temperature and pressure could be measured. A thermocouple was similarly inserted in the 150mm tube, within about 6mm of one end. Similar 150mm and 12.7mm perspex tubes, with no pressure tappings or thermocouple tappings, were also made.

These perspex tubes were then degreased and a thin film of resin allowed to run down the centre of each and also over the machined faces. Excess resin was removed and the tubes were again placed in an oven at 90°C in order to harden the resin and further anneal the perspex. This process prevented the crazing referred to above.

The orifice sections were then made from 1.6mm (1/16") perspex sheet, using a high-speed watchmaker's lathe as follows. A disc of perspex 1.6mm (1/16") thick and about 25mm in diameter, was cemented by its outer edge to a perspex mandril inserted in the lathe chuck. A drill of appropriate diameter was held in a pin-chuck inserted in the tail-stock of the lathe. The drill was used to cut a fine hole in the perspex. The centre of the disc was then faced down until the thickness of the perspex surrounding the hole was approximately equal to the diameter of the hole. The walls of the drilled hole (orifice) were then polished by rotating the perspex disc about a thread of cotton impregnated with perspex polish. In order that the orifice walls should remain parallel during polishing, the thread was positioned along the lathe axis. This was done by inserting one end of the thread
in the pin-chuck of the tail-stock and the other end, over an aligned "V" guide, to a weight. After about 5 minutes polishing, first with coarse then with fine polish, the disc was removed from the mandril. This was done by turning the disc down to 12.7mm (½") diameter. Thus an orifice section of the required diameter was obtained. The orifice was then inspected on a stereoscopic microscope and its diameter and thickness measured at a number of positions. Fifteen orifice sections, whose dimensions are given in Appendix A1, were similarly manufactured.

Finally, the brass tube was manufactured, conventionally, on a lathe, and the nozzles could then be assembled when required, as indicated in Fig. 5.5.

During the early part of this project, the nozzles were stuck together using resin as seen in Fig. 5.4, but this arrangement proved rather unsatisfactory as a blockage of the inner orifice was difficult to remove.

**Two-dimensional nozzle movement**

The emerging spray was axially symmetrical. Therefore photographic data, representative of the whole spray could be obtained by photography along the spray axis and in one plane containing this axis. To do this without disturbing the accurate alignment of the optical system it was necessary to mount the nozzle such that movement relative to the camera was possible. Therefore the nozzle was clamped to a mounting which permitted vertical and horizontal movement in the plane normal to the axis of the optical system. This is shown in Fig. 5.4. It consisted of a 150mm square x 12mm thick duralumin block which could be moved vertically, in guides, by a screw thread. Mounted on this block was a smaller block, also guided, which was capable of horizontal movement normal to the camera axis. The nozzle holder, which was mounted on this smaller block, consisted of two 12mm thick plates with a vertical groove milled into
the inner surface of the outer plate. The nozzle was clamped in this groove.

5.1.3. Double-exposure photomicrographic unit

To obtain particle size and particle velocity measurements within a spray, double-exposure and single-exposure photographs were taken with a double-exposure photomicrographic unit. A specification for such a system was obtained as follows. It was considered necessary to obtain streak-free images of all particles resolved by the optical system. Also, for velocity measurements, it was considered essential that the double images produced should be close enough together for easy identification, but sufficiently far apart for accurate measurement of their displacement.

Now the practical resolution limit of the photo-micrographic unit, as explained later, was a few microns and maximum velocities within the sprays were known, from preliminary experimentation, to be about 40 metres per second. Therefore the required period of exposure, which is given approximately by:

\[
\frac{\text{diameter of smallest resolved particle}}{\text{particle velocity}}
\]

was of the order of 0.2 microseconds.

The minimum time interval between exposures was considered to be that which gave an image displacement of 10 times the diameter of the smallest resolved particle. This minimum interval was therefore in the order of two microseconds, although in practice an interval of 10 microseconds proved to be more suitable.

To provide, conveniently, exposures of such duration, a double-pulse spark unit was used. A suitable unit was manufactured by Lunartron Electronics Limited. To the basic unit were added, as explained below, an interval timer and an F/1 condenser lens. A photographic record was
obtained with a camera system, shown, together with the spark unit in Fig. 5.7.

**Double-spark unit**

The double-spark unit consisted of two spark gaps on the same optical axis with a system of condenser lenses arranged so that each spark appeared to originate from the same position. The spark duration was about 0.2 microsecond.

The spark discharges were initiated by two H.T. trigger units. A delay unit was included which enabled the front spark to be triggered a pre-set interval of time after the rear spark. The magnitude of this interval, which was set by a dial, was reproducible to ± 1%. The minimum time interval between sparks on the standard spark unit was 20 microseconds. In order to obtain shorter intervals, a timing capacitor was replaced in the delay unit. In this way, time intervals down to two microseconds were achieved.

It was necessary to calibrate the delay unit. This was done with an oscilloscope as follows. Two insulated wires, placed close to each spark gap, were fed to the "Y" input of the oscilloscope. When the spark unit was operated, each spark generated an induced e.m.f. in the corresponding wire. The first spark triggered the oscilloscope; thus both pulses were conveniently displayed on the screen. Measurement of the distance, on a screen, between the two pulses enabled the time interval between them to be determined with an accuracy of about 0.2 microseconds. The interval was then adjusted to the nominal value on the dial of the delay unit.

When photographs were taken, this time interval was continuously monitored using a Racal interval timer which was started and stopped by the inductive pick-up system previously described. The resolution of the timer was relatively long, however, (1 microsecond), and it was
therefore used only to ensure that the spark unit had operated satisfactorily. For example, occasionally the second spark would trigger prematurely, in which case the interval timer would record an interval shorter than that set on the flash unit.

**Camera System**

Images of the droplets of the spray were formed with a lens and recorded with a camera. Transmitted-light illumination was used; consequently silhouette images were obtained. The image-forming lens was a high resolution copying lens capable of resolving 500 lines per millimetre. Its focal length was 76mm (3") and its maximum relative aperture was F/4. The lens was designed to operate at a magnification of ten times with green light of wavelength 5461 ±100Å.

The 76mm focal length allowed photomicrography of the spray without interference to the latter, whilst a magnification of ten times ensured that the image of the smallest resolvable particle was much bigger than the grain structure of readily-available fine-grained film.

The magnification of ten times was achieved by mounting the lens on a reversing ring, such that the short conjugate was facing the field of view; the camera was then mounted on an extension tube 840mm long. Part of the extension tube was a bellows system on a rack and pinion mounting. By means of this, the magnification could be adjusted precisely and the camera system could be focused by longitudinal movement. To provide light of the appropriate wavelength a green Wratten filter was included in the optical path, as shown in Fig. 5.7.

The camera was of the reflex type, fitted with a focal plane shutter.

**Alignment of the flash unit and the camera**

It was necessary to align the flash unit and the camera to ensure that each spark illuminated the field of view equally and evenly and
also to ensure optimum resolution of the optical system.

This was done by a series of adjustments.

First, the width of each spark gap was reduced such that, with no trigger, sparking just did not occur. This ensured a maximum concentration of optical energy. The centre of each gap was then adjusted to coincide with the axis of the optical system.

Secondly, the three condenser lenses were adjusted longitudinally to a distance of twice their focal length from their appropriate "object". This arrangement produced an unmagnified image of each spark located between the external condenser lens and the camera lens.

Thirdly, the camera was aligned so as to focus on these two superimposed images. However, inevitable slight lateral and longitudinal misalignment of the two sparks was now magnified by the camera. This was corrected for by further fine adjustment of the condenser lens between the two spark gaps.

The above configuration resulted in equal illumination by both sparks but, because of the rectangular nature of the sparks, the photographic film would not have been evenly exposed. This problem was remedied by moving the external condenser lens back towards the flash unit. The images of the sparks were then formed nearer to the camera lens, which consequently was not focused on them. This resulted in an evenly illuminated field of view and an evenly exposed photographic film.

The last adjustment was the positioning of the bulb, situated at the back of the flash unit, such that its image coincided with those of the sparks. Henceforth adjustments could be simply made by alignment with the image of the bulb.

With the system accurately aligned its resolution was optimum. This was because the condenser lens, of large relative aperture
provided a wide angle cone of light at the object plane striking the camera lens. This angle was greater than that subtended by the aperture of the camera lens at the object plane. This meant that the resolution of the system was defined by the aperture of the camera lens and not reduced by an illuminating system of too narrow a cone angle.

Choice of lens aperture and photographic film

a) Lens aperture

It was necessary to select carefully the aperture of the image-forming lens. A large aperture would have collected much light, given good resolution and given a desirably small depth of field. However contrast between particles and the background would have been poor. This is because, with transmitted light illumination, a particle contrasts with the background illumination primarily because it scatters light. Now, particles larger than the wavelength of light scatter the light mainly in the forward direction. Consequently, the larger the aperture the more the scattered light is collected and the poorer is the contrast.

It was found experimentally that with relative apertures of F/4 (the largest available) and F/5.6, contrast was too poor to allow easy identification of pairs of small particles. It was found that F/8 was optimum. The resolution of the photomicrographic unit at this aperture setting, as determined from the lens specification, was about 250 lines per millimetre.

b) Photographic film

The photographic film was selected as follows. At ten times magnification, it was necessary that the light from the flash unit should "expose" the film adequately and that the grain size should be smaller than the smallest resolvable particle; also the "contrast" of the image should be high. Tests showed that Ilford film Type "Fan F",...
With a resolution of 100 lines per millimetre, met these requirements. The film was developed at 20°C for 3 minutes in Kodak "D8" developer.

It was found, however, that with only the green filter in the optical path, the film tended to be over-exposed, resulting in poor contrast between droplet image and background. To ensure optimum exposure, neutral density (N.D.) optical filters were inserted in the optical path between the spark source and the spray. The correct value of the neutral density filter was determined as follows. With no spray, spark photographs were taken with different values of neutral density filter in the optical path. The density of each developed film was measured on a densitometer. A graph (the "H-D" curve) of film density against the value of the N.D. filter (proportional to \( \log \) (exposure)) was then plotted. From this graph the value of the density of the filter corresponding to its linear section, representing optimum exposure, was then established.

The filters required, in addition to green, in the absence of spray, are given below.

<table>
<thead>
<tr>
<th>F-Number</th>
<th>Number of Exposures</th>
<th>N.D. Filter required</th>
</tr>
</thead>
<tbody>
<tr>
<td>8</td>
<td>Single</td>
<td>0</td>
</tr>
<tr>
<td>8</td>
<td>Double</td>
<td>0.3</td>
</tr>
<tr>
<td>5.6</td>
<td>Single</td>
<td>0.3</td>
</tr>
<tr>
<td>5.6</td>
<td>Double</td>
<td>0.6</td>
</tr>
</tbody>
</table>

In the presence of spray, however, the light entering the camera was attenuated and in most cases no filter was required.

The contrast of the film is given by the slope of the linear section of the H-D curve. This value was 3.7 which represents a film of fairly high contrast.
5.1.4 Constant temperature enclosure

The saturated vapour pressure of the propellant mixture varied by about 10 kN/m² (1.5 p.s.i.) for a change of temperature of one degree C. It was essential, therefore, that the propellant temperature was closely controlled.

The experimental apparatus was contained in a laboratory held at a temperature constant to within about ± 3 deg.C by thermostatically controlled electric heaters. The section housing the propellant cylinder and nozzles was held constant to within about ± 0.5 deg.C. by an aluminium radiation shield and controlled by a contact thermometer. Heat was distributed uniformly by a fan at the top of the apparatus.

Variation in propellant temperature resulting from change of ambient temperature was thus minimised. It remained only to ensure that the temperature of the propellant in the container did not drop significantly during spraying as a result of evaporative cooling. The permissible duration of spraying was estimated from the heat balance given below.

Typical mass flow-rate = \(3 \times 10^{-3}\) kg/sec

Mean density of P 11/12 solution = \(1.4 \times 10^3\) kg/m²

" " " vapour = 6 kg/m²

Mass of propellant in the container = 2 kg

Specific heat of the solution = 0.92 kJ/kg/deg C

Latent heat of vaporisation = 170 kJ/kg

Therefore in time \(t\) seconds, the volume of vapour produced to replace the discharged propellant = \(\frac{3 \times 10^{-3}t}{1.4 \times 10^{-3}}\) m³

Therefore the mass of the vapour produced = \(\frac{3 \times 10^{-3}t}{1.4 \times 10^{-3}}\) 6 kg

Therefore \(\frac{3 \times 10^{-3}t}{1.4 \times 10^{-3}}\) 6 x 170 kJ = 2 x .92 x (allowed temperature drop)
If the temperature drop was to be less than 0.2 deg C, say, the spray duration must be less than 200 seconds - providing the propellant container is almost full. However, as the duration of the spray was typically 30 sec, temperature fall during spraying could be neglected until the container was almost empty.

5.1.5 Sedimentation apparatus

In order to obtain a size analysis of the residual surfactant spray, particles of the spray were collected on microscope slides placed at the bottom of a cylinder of large dimensions referred to in Section 2. It consisted of a 600mm deep polythene container of diameter 400mm equipped with a close fitting lid. Before each analysis 20 glass microscope slides were cleaned in chromic acid, dried and placed at the bottom of the container.

5.2 Calibration

5.2.1 The magnification of photomicrographic unit

The copying lens magnified nominally ten times. The precise magnification was determined by photographing a ruled graticule, placed accurately at the plane of focus of the image-forming lens.

5.2.2 The depth of field of the photomicrographic unit

In order to determine the depth of field of the photomicrographic unit, polydispersed aluminium powder, of mean diameter about 30μm, was photographed. The powder was first sprayed on to a microscope slide, which was then positioned in the plane of focus of the image-forming lens, but at an angle of about 80° to the optical axis of the camera system. Single-flash photomicrographs were taken at varying lens apertures, and prints of an overall magnification of 50 times were obtained. Because the microscope slide was not normal to the optical axis of the camera, a central band of particles only was in focus.
The width of this band was measured from the prints. The depth of field was then obtained trigonometrically from the width of this band. A graph of depth of field for a given lens aperture is given in Fig. 5.8.

5.2.3 Pressure gauges

During the course of the experimental programme two pressure gauges of the Bourdon type (0 - 345 kN/m² or 50 p.s.i.g.) were used. Each had a 150mm (6") dial and could be read to within ± 1.4 kN/m² (0.2 p.s.i.). They were calibrated, using a mercury manometer, in an orientation identical to that in use.

5.2.4 Thermocouples

The reference thermocouple was placed in a Dewer flask of crushed melting ice, frozen from distilled water. The thermocouple to be calibrated was placed in a second Dewer flask containing distilled water and a "Gold Line" thermometer graduated to 0.1 deg.C. The calibration of this thermometer was checked at 0°C and 100°C.

The output of the thermocouples used on the continuous flow apparatus were fed directly to the digital voltmeter, and a calibration curve drawn. The resolution was 0.1 deg.C.

5.3 Experimental programme

As stated earlier, the purpose of the experimental programme was to provide verification of theoretical expressions developed previously and also to obtain empirical relationships relevant to inhalation therapy.

The theoretical expressions were developed to enable spray characteristics to be determined in terms of the nozzle dimensions and the thermodynamic properties of the propellant only. It was found however that additionally three empirical factors were required; they were the degree of metastability of the propellant in the expansion chamber, the divergence angle and the position of the apparent origin of the spray. The first part
FIG. 5.8. VARIATION OF DEPTH OF FIELD OF THE CAMERA WITH APERTURE SETTING.
of the experimental programme, therefore, included the measurement of the three empirical factors. This information together with further data constituted a means by which the theoretical expressions could be verified.

The second part of the programme was the measurement of size distributions of the resultant surfactant aerosols generated by different nozzle combinations.

It is appropriate, here, to review the derived expressions and explain their relevance.

Initially, equations for the mass flow-rates through both upstream and downstream nozzles were derived. By combining those two equations, the temperature fall (and pressure fall) across the first orifice in terms of the dimensions of the nozzle and the thermodynamic properties of the propellant only were derived. It was necessary however to include, in some cases, an empirical expression for the degree of metastability of the propellant in the expansion chamber. From these expressions the mass flow-rate could be determined, also in terms of fundamental parameters.

It was then possible to derive an equation for the exit velocity of the spray from the nozzle. From this exit velocity, together with the previously derived mass flow-rate and the empirically determined spray origin and divergence angle, the velocity of the gaseous component of the spray at a given point in the spray could be calculated. The velocity of the particulate component of the spray could be determined from standard particle drag theory.

The above is necessarily an over-simplification and further details are given in Section 6.

The experimental programme and the expression to which each part of the programme relates are listed in the chart following.
## Chart Showing the Programme of Experimental Work

<table>
<thead>
<tr>
<th>Experimental Work</th>
<th>Application</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>First part</strong></td>
<td></td>
</tr>
<tr>
<td>1. Measurement of the temperature and pressure of the propellant during discharge.</td>
<td>From these measurements an empirical expression for the degree of metastability of the propellant in the expansion chamber was determined. Also the theoretical expression for the temperature fall across the first orifice could be verified.</td>
</tr>
<tr>
<td>2. Measurement of the mass flow-rate of the propellant during discharge.</td>
<td>Mass flow-rate measurements were used to verify expressions for the discharge of a saturated liquid through a two-orifice nozzle assembly.</td>
</tr>
<tr>
<td>3. Photomicrography of the spray</td>
<td>Double-exposure photomicrographs were used to determine the axial velocity profile of the spray, including exit velocity, which was then compared with theory. Transverse profiles of velocity and the variation of particle velocity with particle size were also determined.</td>
</tr>
<tr>
<td>4. Photography of the complete spray.</td>
<td>From photographs of the complete spray, the position of apparent origin of the spray was measured.</td>
</tr>
<tr>
<td><strong>Second part</strong></td>
<td></td>
</tr>
<tr>
<td>5. Measurement of the size distribution of the resultant solute aerosol.</td>
<td>This information is relevant to aerosol inhalation therapy. Although no theoretical expression for the size distribution was developed with which to compare the results, an empirical relationship was determined.</td>
</tr>
</tbody>
</table>
5.4 Experimental procedure

The propellant mixture used throughout consisted of, by weight, 60% of propellant 12, 40% of propellant 11. Into this was dissolved 1% by weight of the (liquid) surface-active agent sorbitan trioleate ("Span 85").

The propellant bottles, described in Sub-Section 5.1, were filled with propellant using a "cold-fill" technique in which both bottle and propellant were first cooled in an isopropyl alcohol (IPA) refrigerator. The propellant was then poured directly into the bottle. Atmospheric water vapour, which may have condensed on to the inside of the bottle, was thus prevented from doing so by a blanket of IPA vapour covering the refrigerator.

After each bottle had been filled, the lid was screwed on and then vapour was vented to remove any air from the bottle. This method had an advantage over pressure injection filling, in that the amount of air finally dissolved in the propellant was kept to a minimum.

Before each run, the apparatus was allowed sufficient time to acquire a steady temperature, nominally 20°C. Immediately before the run, the bottle was shaken to ensure that the propellant and surfactant were thoroughly mixed.

Each part of the experimental programme, given in the chart above, will now be considered.

5.4.1 Measurement of the temperature and pressure in the expansion chamber of the nozzle

Measurements of the temperature and pressure in the expansion chamber were made for the nine nozzle combinations given in Section 6. For each nozzle combination the pressure and temperature of the propellant in the feed-pipe was checked and recorded before spraying, to ensure that correct thermal conditions had been acquired. This was done by reading, respectively, pressure gauge 1 and thermocouple 1,
situated as shown in Fig. 5.1. The spray was then discharged and the data-logger started.

During spray discharge, the e.m.f.'s of the thermocouples 1, 2 and 3 were recorded automatically by the data logger, whilst the steady-state readings of pressure gauges 1 and 2 were recorded manually. It was found that pressure gauge 1 quickly became steady at a value slightly below its initial (no spray) value. Pressure gauge 2, however, rose quickly from zero to an intermediate value but then continued to rise slowly, reaching steady-state conditions after about 10 to 15 seconds. It was this steady-state reading of gauge 2 that was recorded.

The temperatures of the propellant either side of the first orifice were obtained from a print-out of the data-logger tape. A typical graph, showing the variation of these temperatures with time, is shown in Fig. 5.9. The steady-state reading of thermocouple 3 was recorded.

5.4.2 Mass flow rate

The apparatus was allowed to reach the steady temperature of the constant-temperature enclosure. The spray generator, that is container, feed-pipe and nozzle, was then transferred to a stand placed on a direct-read-out balance. The configuration of the spray generator on the stand was maintained the same as that on the spray rig, except that, for convenience, the nozzle was positioned to spray horizontally.

The spray was then started and when steady-state conditions had been reached, the time taken for 20.0gm to discharge, was measured with a stop-watch. Three such readings were taken, and the mean calculated.

Afterwards, the spray generator was returned to the constant-temperature enclosure, the nozzle changed, and the procedure repeated.

5.4.3 Photomicrography

Double-exposure and, for reference purposes, single-exposure
FIG. 5.9. TEMPERATURE OF THE PROPELLANT IN THE NOZZLE ASSEMBLY.
photomicrographs were taken of the emerging spray. The double-exposure photographs were taken to determine:

a) the exit velocity of the spray from the nozzle.
b) the longitudinal velocity along the spray axis,
and c) the transverse profile of longitudinal velocity in a plane containing the spray axis.

The nozzle combinations used and the parts of the spray which were photographed are given in Section 6.

From the double-exposure photographs taken at a given point in the spray, an accurate value of the velocity of particles at that point was required. However as a spray is in general a turbulent system, velocities measured at a given position but at different times, vary considerably. Therefore, a time-average must be obtained. In order to get an accurate time-average, a number of photographs must be taken at a given point, this number depending on the accuracy required and the degree of turbulence within the spray.

To estimate the number of photographs required, let it be assumed that there is to be a 90% probability that the sample time-averaged velocity, obtained from N photographs, should be within 10% of the long-term time-averaged velocity ($\bar{u}$). Then, assuming a normal distribution of instantaneous velocities (with standard deviation $s$), 1.6 standard errors of the distribution of time-averaged velocities (45) should be less than or equal to 10% of the time-averaged velocity.

Therefore: $1.6s / \sqrt{N} \leq (10/100) \bar{u}$

or, $N \geq (16)^2 (s/\bar{u})^2$

Here, $s/\bar{u}$ is the relative turbulence, an estimate of which would enable $N$ to be calculated. Initial experimentation showed that the relative turbulence,
although varying with position in the spray, was typically 0.2, a figure in agreement with reported values (66).

Insertion of this figure into the above equation leads to a value of ten for the required number of double-exposure photographs. However, the time required to take and analyse this number of photographs at many positions within a large number of sprays would have been prohibitive. Consequently, this criterion was relaxed to an extent depending on the situation, as discussed in Section 6. Inevitably, a larger error in the estimate of the average velocity was therefore incurred.

That so many photographs are required to obtain an accurate value of the average velocity within a turbulent system is seen to be a serious disadvantage of this method of velocity measurement. For example, to ensure a 90% probability of incurring an error of no greater than 1% would require the analysis of about 1000 double-exposure photographs.

Method

The part of the spray first to be photographed was usually an axial point close to the exit of the nozzle. The latter was first lowered and adjusted transversally until it appeared in the field of view of the camera. So that the camera could be focused precisely on the spray axis, the latter was first defined by a jet of water. To do this, the water was injected, at moderate pressure, through the "T" piece provided in the feed-pipe. It then emerged as a continuous jet along the spray axis, providing a suitable object on which to focus the camera.

The propellant was then discharged. When bubbles were seen to have cleared from the perspex tube above the first orifice, and steady-state conditions had been reached, photographs were taken of the spray.

To photograph other axial positions within the spray, the nozzle was raised relative to the camera. It was necessary, after raising
the nozzle, to check that the spray axis still passed through the object plane of the camera and use was again made of the water jet.

To obtain photographs of off-axis positions in the spray, the nozzle was moved transversely relative to the camera.

5.4.4 Photography of the complete spray

In order to obtain a direct measure of the apparent position of the origin of the spray, photographs of the complete spray were taken. By setting a 35mm. camera about 1 metre from the spray, which was illuminated with flood lights, photographs of 500mm length of the spray were obtained. From these, the position of the spray origin could be measured. Half-microsecond flash photographs were also taken.

5.4.5 Size analysis of the resultant aerosol of surface-active solute.

Twenty microscope slides were cleaned, dried and placed at the bottom of the sedimentation cylinder which was placed beside the spray rig. After the latter had acquired a steady temperature, the nozzle was removed from its mounting and the spray discharged. After steady-state conditions within the nozzle had been reached, the spray was directed into the sedimentation cylinder for about two seconds. This length of time was found to be optimum; a shorter spraying time resulted in too few droplets on the slides, whilst a longer time resulted in overlapping of the droplets.

The nozzle was then directed away from the column and spraying stopped. The lid of the sedimentation cylinder was then replaced. The resultant aerosol was allowed to sediment for fifteen hours on to the twenty glass slides which were then removed from the cylinder.

The size distribution was obtained by microscope analysis using a technique first described by May (39). The procedure is described briefly below.
Firstly, the spread factor or the droplets was obtained. This is the ratio of flattened droplet diameter \( D_p \) to spherical droplet diameter \( d \). The size distribution of the flattened droplets was then determined, using conventional techniques. Finally the size distribution of the original aerosol was determined by application of the spread factor.

To determine the spread factor, the microscope was placed near a window. A droplet on one of the glass slides was selected and its diameter measured. The condenser of the microscope was removed and the plain illuminating mirror of the microscope was adjusted until light from the window illuminated the droplet. The specimen stage was lowered until the image of the window could be seen through the microscope. The flattened droplet, now acting as a plano-convex lens had focused an image of the window (effectively at infinity) in its focal plane. The distance the stage was lowered, as measured from the vertical fine-adjustment drum, was the focal length \( F \) of the plano-convex lens. Measurements on about ten droplets were made, covering the size range found in the aerosol.

The mean value of \( F/D_p \) was obtained, and from the graph reproduced in Fig. 5.10 the angle of contact was found and hence the mean value of the spread factor \( D/d \). The relative standard deviation in the measured values of the spread factor was 4%.

Having determined the spread factor, it was then necessary to measure the size distribution of the flattened droplets on the twenty glass slides. This was done by a procedure based on that given in B.S.3406(5).

**Applicability of this method**

It has been shown by Whitby et al (72) that aerosols of the type discussed in this thesis contain large numbers of sub-micron particles. It was also evident that after the fifteen hours sedimentation, a large
FIG. 5.10. DETERMINATION OF THE "SPREAD FACTOR"
FOR A FLATTENED DROPLET.
number of particles remained in the sedimentation chamber. This could be demonstrated by directing a beam of light from a microscope lamp through the chamber. A vast number of particles were observed in the beam. Application of Stoke's Law indicated that the diameters of these droplets were less than 0.7 μm.

It would have been meaningless, therefore, to calculate a mean diameter which was sensitive to the large number of sub-micron particles. However, it was shown earlier that the information required from the size distributions was the fraction by volume of particles below a particle diameter of approximately 5 μm. For the aerosols of interest, generated from saturated liquids, the fraction by volume in the sub-micron region will be negligible compared with that below 5 μm. Therefore the size distributions will be meaningful, despite the omission of the sub-micron particles.
EXPERIMENTAL RESULTS AND COMPARISON WITH THEORY

6.1 Determination of the values of the constants $C_1$, $C_2$, $C_3$ and $\gamma$.

6.2 Measurement of mass flow-rate and comparison with theoretical expressions for the flow-rate through a single orifice. Determination of discharge coefficients.

6.3 Measurement of the temperature fall, $\Delta T_1$ across the first orifice of the nozzle assembly and comparison with theory.

6.4 Experimental verification of the expression for the mass flow-rate through a two-orifice nozzle assembly.

6.5 Measurement of the relative velocity between liquid and gas phases at the exit of the nozzle.

6.6 Measurement of the exit velocity of the spray and comparison with theory.

6.7 Measurement of the transverse profile of longitudinal velocity within the spray.

6.8 Measurement of the divergence angle of the spray.

6.9 Measurement of the longitudinal profile of longitudinal velocity within the spray and comparison with theory.

6.10 Measurement of the variation of particle velocity within a spray with particle size, and comparison with standard theory.

6.11 Determination of the size analyses of the resultant surfactant aerosols and their variation with nozzle dimensions.

6.12 Determination of the degree of metastability of the propellant in the expansion chamber.

In Section 4 an expression was produced by which the longitudinal velocity profile within a continuous spray could be predicted. A double-flash photomicrograph of such a spray is shown in Fig. 6.1. In this section the quantitative predictions of each step in the development of the above expression are presented and comparisons are made with the experimental results. The verification
Magnification = 100
Time interval between flashes = 10 microsec.

FIG. 6.1 DOUBLE-FLASH PHOTOMICROGRAPH OF AEROSOL SPRAY.
of a number of assumptions made in Section 4 is also given. Also presented are size analyses of the resultant aerosol obtained from a continuous spray, the propellant of which contained 1% by weight of a non-volatile solute. In this latter case, however, no theoretical prediction of size distribution is given.

Before the predictions of theory can be presented, however, it is first necessary to calculate, from thermodynamic data, a number of numerical factors used in the theoretical expressions referred to above. The value of these factors was essentially constant over the range of experimental conditions. Therefore, although these factors were not determined experimentally, experimental results were used to specify the applicable conditions; consequently their determination has been delayed until this section.

There are four such "constants" as explained below.

6.1 Determination of the values of the constants $C_1$, $C_2$, $C_3$ and $\gamma$ used in Section 4

6.1.1 Determination of $C_1$

$C_1$ was defined by equation 4.2.19 as:

$$C_1 = \frac{\rho_{ge}}{P_e}$$

where $\rho_{ge}$ is the density of the propellant vapour in the expansion chamber and $P_e$ is the corresponding absolute pressure.

The situation is shown in Fig. 6.2. $C_1$, of course, does not maintain a constant value as it will vary with both expansion chamber temperature and pressure. However it is seen from Appendix 1.3 that, for a constant propellant temperature of 20°C, the expansion chamber temperature of the nozzle assemblies, with one exception, varied over the rather narrow range of about 14°C to 18°C, a mean of 16°C, while the expansion
Liquid solution of propellant 12/11 (60/40% by weight) and 1% by weight "Span 85"

Pressure \( (P_1) \) is slightly greater than the saturated vapour pressure (SVP) of the solution.

Temperature \( (T_1) = 20^\circ C \)

Temperature = \( T_e \).
Pressure \( (P_e) \) is generally less than the S.V.P. of the solution at \( T_e \).
Propellant vapour 91% by weight propellant 12.

Propellant solution: composition essentially unaltered

**Fig. 6.2** FLOW OF SATURATED PROPELLANT THROUGH TWO-ORIFICE NOZZLE ASSEMBLY
chamber pressure varied over the range 330 to 400 kN/m². Further, as is shown later, the propellant in the expansion chamber is generally in a metastable state and that consequently the propellant vapour is superheated. The vapour will therefore approximate to a perfect gas and \( c_1 \) will vary only very slowly with expansion chamber pressure, a mean value of which is 360 kN/m². It follows therefore that, with a propellant of constant initial temperature and vapour pressure, \( c_1 \) is approximately constant for the range of nozzle assemblies tested. The deviation from this constant value is considered later.

In order to determine \( c_1 \), it is first necessary to determine the relative proportions of propellant 12 and 11 in their vapour. As the propellant consists of 60% by weight of the very volatile component propellant 12, and 40% by weight of the much less volatile component propellant 11, the vapour will therefore consist largely of propellant 12. Now, for a temperature drop of a few degrees centigrade across the first orifice, the fraction of propellant evaporated is about 5% and the composition of the remaining liquid may be considered unaltered. By application of the Law of Partial Pressures and Raoult's Law, or otherwise (30) it may be shown that the vapour comprises about 91% by weight of propellant 12 and 9% of propellant 11. The corresponding mole fractions are 0.92 and 0.08 respectively.

This proportion is then independent of temperature drop, providing the latter is small.

The partial pressures of propellants 12 and 11 at an overall pressure of 360 kN/m² abs. are, therefore, 330 kN/m² and 30 kN/m² respectively. From thermodynamic tables (30), the mass of propellant 12 per unit volume of propellant 12/11 vapour is, at 16°C, 18.0 kN/m³ and that of propellant 11 is 1.80 kN/m³. Therefore the density of the mixture is 19.8 kN/m³.
Therefore, \( c_1 = \frac{19.8 \text{ kg/m}^2}{3.60 \times 10^9 \text{ N/m}^2} \)

or \( c_1 = 5.50 \times 10^{-5} \text{ kg/J} \)

Direct application of a nomograph (30), which is applicable strictly to saturated vapour only, gives a value of \( c_1 \) of \( 5.56 \times 10^{-5} \text{ kg/J} \).

Further this nomograph indicated that over the range of nozzles tested, the variation of \( c_1 \) was less than 2%; this may be considered to be a measure of the error in the present case.

6.1.2 Determination of \( c_2 \)

Equation 4.2.21 defined \( c_2 \) as

\[
 c_2 = \frac{q}{\Delta T_1}
\]

where \( q \) is the mass fraction of propellant evaporated and \( \Delta T_1 \) is the temperature drop across the first orifice.

\( c_2 \) may be evaluated by considering an enthalpy balance across the first orifice by letting the enthalpy of propellant before the first orifice \( (H_i) \) equal the enthalpy of vapour and propellant in the expansion chamber \( (H_e) \).

Referring again to Fig.6.2, the enthalpy, \( H_i \), of unit mass of liquid propellant above the first orifice at 20°C, assuming an ideal solution, is given by:

\[
 H_i = 0.60 \text{ (specific enthalpy (h) of saturated P 12)} + 0.40 \text{ (specific enthalpy (h) of saturated P 11)}
\]

Here, the effect on the enthalpy of the difference between the SVP of each component and the SVP of the solution. Also the kinetic energy of two-phase propellant in the expansion chamber will be assumed to be negligible.

The enthalpy of the liquid and vapour in the expansion chamber is given by:

\[
 H_i = \frac{c_1}{c_2}
\]
\[ H_e = (1 - q) \left( 0.6(\text{h of saturated liquid } P_{12}) + 0.4(\text{h of saturated liquid } P_{11}) \right) + q \left( 0.91(\text{h of } P_{12} \text{ vapour at its partial pressure}) + 0.09(\text{h of } P_{11} \text{ vapour at its partial pressure}) \right). \]

Here \( q \) is the fraction of propellant by mass evaporated.

In this equation the partial pressures of propellant 12 and propellant 11 are not known precisely because the degree of metastability is not known precisely. However, the vapour is superheated and approximates to a perfect gas. Therefore, over a wide range of pressures the variation of the enthalpy may be considered negligible. For example the variation of enthalpy of propellant 12 at a few Centigrade degrees superheat is about 0.8% over the range 270 to 350 kN/m\(^2\) abs. The values of the specific enthalpy of propellant 12 and propellant 11 may then be calculated using a typical value of about 350 kN/m\(^2\) abs. for the expansion chamber pressure.

By equating \( H_i \) and \( H_e \), and inserting thermodynamic data, the equation reduces to:

\[ H_e = (0.6 \times 23.591 + 0.4 \times 21.61) \times 2.326 \text{ kJ/kg} \]

\[ = H_i \]

\[ = (1-q) (0.6 \times 22.221 + 0.4 \times 20.37) \times 2.326 \]

\[ + q(0.91 \times 85.09 + 0.09 \times 101.1) \times 2.326 \text{ kJ/kg} \]

for a 3.33 deg C fall in temperature.

Therefore \( q = 0.0203 \) per 3.33 deg C temperature fall,

or \( C_2 = 0.0061 \) per deg C.

The accuracy of this value depends on that of the technical data which is assumed to be accurate to the number of significant figures quoted when differences are taken. However, the assumption was made that the kinetic energy of the propellant in the expansion chamber was negligible. Now, from photographs taken of the expansion chamber, shown in Section 2, it is apparent that all the kinetic energy is not
recovered. It is relevant, therefore, to consider the effect on \( C_2 \) of this loss of thermal energy due to the non-recovery of all the kinetic energy. In the worst case, when no kinetic energy is recovered, the error in \( C_2 \) is estimated to be 0.7\%, and therefore this source of error is considered negligible.

6.1.3 Determination of \( C_3 \)

\( C_3 \) was defined in Section 4 as the slope, at about 18\(^\circ\)C, of the curve relating the SVP of the propellant to its temperature. This slope is readily obtained by applying the Clapeyron equation to a two-component system. This application is valid if the two components have physically similar properties which is the case for most halogenated hydrocarbon aerosol propellants. The following equation then results:

\[
-\frac{1}{T} \lambda \frac{dT}{dP} + \bar{v}(\text{vapour}) \frac{dP}{dT} = 0,
\]

where \( \lambda \) is the specific latent heat of the two-component solution and \( \bar{v} \) is the specific volume of the vapour phase.

By assuming that the vapour behaves as a perfect gas, \( \bar{v}(\text{vapour}) \) may be put equal to \( \frac{RT}{(\text{SVP})} \). Substitution into the above equation and integration gives, assuming \( \lambda \) to be constant over a small range of temperatures,:

\[
\ln(\text{SVP}) = \frac{\lambda}{RT} + \text{constant}.
\]

Therefore over a small range of temperatures a plot of \( \ln(\text{SVP}) \) against \( \frac{1}{T} \) will be straight.

The value of \( \frac{dP}{dT} \), or \( C_3 \), is given by:

\[
C_3 = \frac{P}{T^2} \times \text{(slope of the plot of } \ln(\text{SVP}) \text{ against } \frac{1}{T} \text{)}.
\]

From data supplied by Imperial Smelting (30) a straight line plot of \( \ln(\text{SVP}) \) against \( \frac{1}{T} \) was obtained, as shown in Fig. 6.3. Its slope is \( 2.33 \times 10^3 \) K.
Fig. 6.3 The Clapeyron equation applied to a solution of propellants 12 & 11 (60% - 40% by weight)
Now, at 18°C the S.V.P. is 370 kN/m².

Therefore \( C_3 = \frac{370}{(291)^2} \times 2.33 \times 10^3 = 10.1 \text{ kN/m}^2/\text{deg C} \).

The inherent accuracy of this figure again depends on the accuracy of the technical data and is thus expected to be adequate. However, errors occur if the temperature varies about a mean other than 18°C. For example, \( C_3 \) determined at 16°C is 9.70 kN/m²/deg C, an error of 4%.

In such a case, therefore, it would be desirable to use the new value for \( C_3 \).

6.1.4  Determination of \( \gamma \)

In equation 4.2.11 it was shown that the value of \( \gamma \), the ratio of the specific heats of the propellant 12/11 vapour was required at the point of choking. It has been assumed that at the point of choking the following conditions prevail:

1. The pressure is given by:
   \[ \frac{P_c}{P_e} = \left( \frac{r_c}{0.52} \right) \times \text{(expansion chamber pressure)} \]

2. The fractions by weight of propellants 12 and 11 at the point of choking are equal to those within the expansion chamber. This is because it is assumed that no mass (or heat) transfer takes place during the passage of the propellant through the orifice. These fractions are approximately 0.9 and 0.1 respectively.

3. The temperature ratio \( \frac{T_c}{T_a} = \left( \frac{r_c}{0.52} \right) \frac{\gamma - 1}{\gamma} \).

By definition:
\[ \gamma = \frac{C_P}{C_V} = \frac{0.9 C_P(P12) + 0.1 C_P(P11)}{0.9 C_V(P11) + 0.1 C_V(P11)} \]

It is therefore necessary to determine values of \( C_P \) and \( C_V \) under conditions defined above.

As \( \gamma \) is a slowly varying function, typical values of critical
conditions may be used. A typical value of the expansion chamber pressure is 350 kN/m$^2$ abs. Therefore the pressure at the point of choking is $0.52 \times 350$ or 180 kN/m$^2$. A typical value of the expansion chamber is 17°C. Therefore the temperature of the vapour at the point of choking is given by:

$$T_c = 290 \left( 0.52 \right)^{\gamma-1} \gamma$$

As the value of $\gamma$ is required before $T_c$ can be determined, an iterative mathematical process is necessary. The last stage of this is that an approximate value for $\gamma$ of 1.16 gives a value of -9°C for the temperature of the vapour at the point of choking.

By definition:

$$C_p = \left( \frac{\delta H}{\delta T} \right)_p$$

and

$$C_v = T \left( \frac{\delta S}{\delta T} \right)_V$$

or, more conveniently

$$C_v = C_p - T \left( \frac{\delta S}{\delta T} \right)_p \left( \frac{\delta S}{\delta V} \right)_T \right)$$

From thermodynamic tables for propellants 12 and 11 the following may be obtained:

For propellant 12 under the conditions stated:

$$C_p = 0.605 \text{ kJ/kg/deg C}$$

$$C_p - C_v = 0.089 \quad \text{"}$$

Therefore

$$C_v = 0.516 \quad \text{"}$$

For propellant 11 under the same conditions:

$$C_p = 0.531 \text{ kJ/kg/deg C}$$

$$C_p - C_v = 0.05 \quad \text{"}$$

Therefore

$$C_v = 0.468 \quad \text{"}$$

Therefore

$$\frac{0.9 \times 0.605 + 0.1 \times 0.531}{0.9 \times 0.516 + 0.1 \times 0.468} , \quad \frac{0.9 \times 0.605 + 0.1 \times 0.531}{0.9 \times 0.516 + 0.1 \times 0.468} ,$$

or 1.17.

The accuracy of this value is dependant on the confidence with which
the conditions at the point of choking are defined. An error arises if the assumption that no heat transfer takes place during the passage of the propellant through the orifice is wrong. In the worst case of rapid heat transfer, such that the gas remains at expansion chamber temperature during the passage through the orifice, $\gamma$ would take a value of 1.15.
6.2 Measurement of mass flow rate and comparison with theoretical expressions for the flow rate through a single orifice. 

Determination of the discharge coefficients of the upstream and downstream orifices of the nozzle assembly, \( C_{Q1} \) and \( C_{Q2} \) respectively.

In Section 6.1, four quasi-constant parameters were determined from thermodynamic data. As explained in the introduction, it is now possible to present experimental verification for the various steps in the development of a fundamental expression for velocity profiles within a spray. The first step in this development was to equate an expression for the mass flow-rate through the upstream orifice to that for the downstream orifice, thus producing an equation for the temperature fall \( \Delta T_1 \) across the first orifice. It is therefore first necessary to verify each of these two expressions for the mass flow-rate. This may be done by following standard fluid dynamics practice which is to demonstrate that over a wide range of conditions the discharge coefficient of the orifice is constant and approximately equal to 0.83, a value determined independently in Appendix 3.

6.2.1 Determination of the discharge coefficient of the upstream orifice, \( C_{Q1} \)

It is anticipated that the upstream orifice passes the propellant in the liquid phase. This follows partly from Pasqua's criterion (50) described in Section 2, which states that a saturated liquid, discharging through a short tube, will not exhibit choking effects providing the downstream pressure is greater than:

\[
0.37 \times \text{(initial saturation pressure)} \left( \frac{\text{Length of tube}}{\text{Dia. of tube}} \right) 0.2
\]

Now, in this work the initial saturation pressure was 400 kN/m\(^2\) abs. and the length of each orifice was approximately equal to its diameter. Therefore, for a downstream (expansion chamber) pressure of greater than
0.37 x 400 or 150 kN/m² abs. no choking effects are expected. Reference to Appendix 1.3 shows that in all cases the expansion chamber pressure greatly exceeded 150 kN/m² abs.; the lowest value was 244 kN/m² abs. Further, the propellant at the first orifice is sub-cooled by a half-metre head of propellant. Consequently, it is considered that the standard expression for the discharge of a single-phase incompressible fluid may be used to define $C_{Q1}$. This was given in equation 4.2.1 as:

$$
\dot{m}_1 = C_{Q1} \left( \frac{\pi}{4} \right) \left( D_1 \right)^2 \sqrt{\frac{2 \rho_1 (P_i - P_e)}{(P_i - P_e)}}
$$

If this equation is valid then a plot of $\dot{m}_1$ against $\sqrt{(P_i - P_e)}$ for all nozzle assemblies will be linear, and equal to $C_{Q1} \sqrt{\frac{2 \rho_1}{\pi}}$. Fig. 6.4 shows the above plot obtained from data given in Appendices 1.2 and 1.3.

It can be seen that the plot is reasonably linear, with a slope of 41.4 (kg/m³)¹⁄². As the density of the propellant, $\rho_1$, is $1.40 \times 10^3$ kg/m³, $C_{Q1} = 0.78$. Further, as the plot is linear, support is given to the assumption that the propellant remains a liquid during its passage through the upstream orifice.

The possible error associated with these data points may next be considered. The most likely source of error is that due to non-uniformity along the diameter of the orifices. In the worst case this could cause an error of about 4% in the determination of the area of the orifice. The two other sources of error, those associated with the measurement of the mass flow rate ($\dot{m}$) and the pressure differential across the first orifice ($\Delta P_1$), are generally smaller than 4%, as is seen from Appendices 1.2 and 1.3. This is not the case however when the pressure differential is small. Thus the departure from linearity of the data point for nozzle 0.65 x 0.35 is attributed to this latter source of error. The other data points are within about 4% of the regression line.
FIG. 6.4. DISCHARGE OF PROPELLANT THROUGH THE UPSTREAM ORIFICE.
The linearity of the regression line and the good agreement between the measured discharge coefficient and the expected value is taken to confirm the applicability of the above expression (equation 4.2.1).

6.2.2 Determination of the discharge coefficient of the downstream orifice, \( C_{Q2} \)

It has been shown that unlike the upstream orifice, the downstream orifice discharges a two-phase fluid - the liquid propellant and its vapour. In Section 4 two expressions were developed for the discharge of such a fluid, one for a low pressure differential across the orifice and the other for a large pressure differential which would be expected to create choking or mass-limiting effects within the orifice. A probable criterion for mass-limiting flow is that the ratio of downstream (ambient) pressure to upstream (expansion chamber) pressure should be less than about 0.5. Reference to Appendix 1.3 shows that in all cases this ratio is less than 0.52, the largest value being 0.42. It is therefore to be expected that the second expression, which includes the effects of mass-limiting flow, would be applicable. This expression was given by equation 4.2.14 as:

\[
\dot{m}_2 = C_{Q2} \left( \frac{\pi}{4} \right) (D_2)^2 \left( \frac{\gamma \rho_c P_c}{q} \right)^{\frac{1}{2}},
\]

in which the variables are as defined in Section 4 and refer to the point of choking.

Now it was shown subsequently that the following substitutions could be made in order to transform the equation into a more amenable form:

\[
q = C_2 \Delta T_1, \\
P_c = r_c P_e,
\]
\[ p_{ge} = p_{ge} \left( \frac{p_c}{p_e} \right)^{\frac{1}{\gamma}}, \text{ and} \]
\[ p_{ge} = C_1 p_e, \text{ and that therefore,} \]
\[ p_{ge} = C_1 p_e \frac{1}{\gamma} \]

Substitution into the above gives:

\[ \frac{\dot{M}_2}{(\pi/4)(D_2)^2} = C_{Q2} \left( \frac{\gamma C_1}{C_2} \right)^{\frac{1}{2}} r_c^{\frac{\gamma + 1}{2\gamma}} \frac{p_e}{\sqrt{\Delta T_1}} \]

The coefficient of \( C_{Q2} p_e / \sqrt{\Delta T_1} \) on the right hand side of the above is essentially constant over the range of operating conditions. Therefore a linear relationship between \( \dot{M}/((\pi/4)D_2^2) \) and \( p_e / \sqrt{\Delta T_1} \), and a value of \( C_{Q2} \) again approximately equal to 0.8 would be taken as support for the applicability of the above expression. Fig. 6.5 shows this relationship. It is seen that of the nine data points, six fall on a reasonably straight line while three depart significantly from this line. Of these three, the data from nozzle 650 x 350 could be in error because the value of the temperature differential \( (\Delta T_1) \) is small. This is not the case however with the other two. A possible source of error in the data from these two nozzles is that the physical conditions within the nozzle were not typical of the conditions for which the values of \( C_1, C_2, C_3 \) and \( \gamma \) were determined (this, of course, applies to any nozzle). However, both nozzles have orifice pairs which are of similar diameter and consequently the conditions within those nozzles are fairly typical. Perhaps in view of the number of assumptions made in the development of the mass-limiting flow rate equation, and its simplicity in describing the mass flow-rate per unit area of two-phase flow in terms of a pressure (that within the expansion chamber), a temperature differential (that across the first orifice) and a quasi-constant factor, then the correlation between theory and experimental
FIG. 6.5. DISCHARGE OF PROPELLANT THROUGH THE DOWNSTREAM ORIFICE. [FOR CHOKING FLOW]
data may be considered satisfactory. Further discussion of the applicability of the mass-limiting flow-rate equation is given in later sub-sections.

Accepting, tentatively, that this equation describes reasonably well the mass flow-rate of a two-phase fluid, then from the regression line of Fig. 6.5 a value for the discharge coefficient of the orifice may be determined. This is:

\[ C_{Q2} = 0.78. \]

This is in good agreement with that obtained from single-phase flow, thus further supporting to the applicability of equation 4.2.14.

It is instructive to compare, with experimental flow-rate data, not only the prediction of mass-limiting flow theory (equation 4.2.14) but also that of non mass-limiting flow theory. This was expressed in equation 4.2.8 as follows:

\[ \dot{m}_2 = C_{Q2} \left( \pi/4 \right) \frac{D_2^2}{\rho_0} u_0, \]

where \( \rho_0 \) is the density of the two-phase fluid at the nozzle exit and \( u_0 \) is the corresponding velocity.

\( \rho_0 \) is given by:

\[ \frac{1}{\rho_0} = \frac{q}{\rho_{go}} + \frac{1 - q}{\rho_1}, \]

and \( u_0 \) is given by:

\[ \frac{u_0^2}{2} = \frac{1 - q}{\rho_1} (P_e - P_{amb}) + \frac{q \cdot \rho_0}{\rho_{ge}} \frac{1}{\gamma} \left( P_e \frac{1}{\gamma} - \frac{1}{\gamma} - P_{amb} \frac{1}{\gamma} \right), \]

where the variables are as defined in Section 4.

As with the previous equation substitutions may be made. These are:

\[ q = C_2 \Delta T_1, \quad \rho_{ge} = C_1 P_e \quad \text{and} \]

\[ \rho_{go} = \rho_{ge} \left( \frac{P_{amb}}{P_e} \right)^{1/\gamma} = C_1 P_e^{1 - \frac{1}{\gamma}} P_{amb}^{1/\gamma}. \]
Equation 4.2.8 then reduces to:

\[
\frac{\dot{m}_2}{(\pi/4) D_2^2} = \frac{c_{Q2} \sqrt{2}}{c_2 \Delta T_1} \left( \frac{1}{c_1 P_e^{1-\frac{1}{\gamma}} P_{amb}^{\frac{1}{\gamma}}} + \frac{1 - c_2 \Delta T_1}{\rho_1} \right)
\]

This equation again includes only two variables. They are the expansion chamber pressure, \(P_e\), and the temperature fall across the first orifice, \(\Delta T_1\). The other parameters are either constant or quasi-constant. However, the equation is seen to be more complex than that incorporating mass-limiting flow.

Fig 6.6 shows a plot of the mass flow-rate per unit area against the theoretical expression given by equation 4.2.8 above. It is seen that, rather surprisingly, agreement is better than in the previous case—in particular the data point for nozzle 650 x 350 now falls on the regression line. The discharge coefficient is 0.87 which is a little higher than the expected value for this type of orifice, however.

Comparison between the predictions of choking and non-choking flow theory are further discussed in sub-section 6.7.
FIG. 6.6. DISCHARGE OF PROPELLANT THROUGH THE DOWNSTREAM ORIFICE.

THEORETICAL VALUE ASSUMING NO CHOKING EFFECTS.
6.3 Measurement of the temperature fall across the first orifice 
\((\Delta T_1)\) and comparison with theory.

In the previous section, experimental support for expressions 
giving the mass flow-rate through the upstream and the downstream 
orifices was presented. In Section 4, these two expressions had been 
equated, and this equation had produced an expression for the temperature 
fall \((\Delta T_1)\) across the first orifice. The evaluation of this temperature 
fall was then used to determine the mass flow-rate through the two-orifice 
nozzle assembly and the exit velocity of the spray. For this evaluation the 
nozzle dimensions and thermodynamic data together with a simple empirical 
expression for the metastability of the propellant in the expansion chamber were 
required. These two parameters, the mass flow-rate and exit velocity, 
were then used in the determination of velocity profiles within the 
spray. It is therefore desirable, as the next step, to verify, 
eperimentally the expression for \(\Delta T_1\), which is given by equation 4.2.31.

This equation however related to standard conditions. These were that 
the propellant above the first orifice should be saturated and at a 
temperature of \(20^\circ C\). Experimentally it was impossible to achieve such 
conditions precisely because, firstly, it was necessary to sub-cool 
slightly the propellant above the first orifice. This was to prevent 
spurious bubbling in the feed pipe. Sub-cooling was achieved by placing the 
propellant container about 500mm above the nozzle. Secondly, the temperature 
tended to fluctuate by about \(\pm\) \(\pm\) \(\frac{1}{2}\) deg C about the required \(20^\circ C\).

In order to compare the theoretical predictions with experimentally 
determined values it is therefore necessary to reduce the experimental 
values to standard conditions. This may be done by firstly considering 
the effect on \(\Delta T_1\), of lowering the propellant cylinder such that the 
pressure of the propellant above the first orifice \((P_i)\) becomes equal 
to the S.V.P. of the propellant \((SVP_{T_1})\). Then the excess pressure
Fig. 6.7 Effect of increase in height of the propellant cylinder on the flow through a twin-orifice nozzle.
(P - SVP, Ti), or ΔPex becomes zero. Having brought this excess pressure to zero, it is then necessary to bring the initial temperature to the standard 20°C.

The reduction of the excess pressure to zero will first be considered. The situation is shown semi-quantitatively in Fig. 6.7. This figure shows that as the initial pressure is raised above the saturation pressure, the pressure differential, (ΔT1) falls. Consequently the corrected temperature differential, that is temperature differential corresponding to an initial pressure equal to the saturation pressure, will be greater than the measured value.

Fig. 6.7 was developed for a twin orifice nozzle assembly whose orifice diameters are each approximately 0.34 mm. The development was as follows. The initial conditions, corresponding to the "actual operating point" in the figure were taken from Appendices 1.2, and 1.3 for the 320 x 350 nozzle. For simplicity, metastability was ignored. The basic relationship is that between the mass flow-rate through the first orifice (M1) and the corresponding differential across it (ΔP1). This is:

\[ \dot{M}_1 \propto \sqrt{\Delta P_1} \]

This parabola was first plotted. Further, from equation 4.2.14, tentatively verified in the last sub-section, the mass flow-rate through the second orifice (M2) was taken to be proportional to the expansion chamber pressure (P2) and inversely proportional to the square root of the temperature differential across the first orifice (ΔT1). Now it is seen that for two-phase flow through the expansion chamber, the expansion chamber pressure remains approximately constant. Therefore, ignoring variation in the latter:

\[ \dot{M}_2 \propto \frac{1}{\sqrt{\Delta T_1}} \]
Assuming saturation condition in the expansion chamber, that is ignoring metastability, the relationship between the expansion chamber pressure and the mass flow-rate may be determined from the above relationship, and from:

\[ P_e = \left( SVP_{Ti} \right) - C_3 \left( \Delta T_1 \right). \]

Therefore \( P_e = \left( SVP_{Ti} \right) - \text{(constant)} \frac{C_3}{N^2} \).

The constant was found from initial conditions and then this latter equation was plotted.

Thirdly the initial pressure \( (P_i) \) was plotted. This is given by the sum of the expansion chamber pressure plus the pressure differential across the first orifice. Thus:

\[ P_i = P_e + \Delta P_i. \]

At high initial pressures, the propellant will, of course, flow through the nozzle without vaporising. The onset of this condition is when the expansion chamber pressure becomes equal to the saturation pressure. Further for twin orifices the pressure fall across each orifice \( (\Delta P) \) is the same.

Therefore:

\[ P_i = 2\Delta P + P_{amb}. \]

and \( P_e = SVP = P_i - \Delta P = \Delta P + P_{amb} \)

or \( \Delta P = SVP - P_{amb} \)

This last equation defines the point, on the \( \Delta P \) curve, at which the liquid-only flow zone starts. Conventional nozzle flow theory may then be used to plot lines at these higher mass flow-rate, as shown.

It may be noted that the theoretical \( P_e \) curve, given by

\[ P_e = \left( SVP_{Ti} \right) - \text{(constant)} \frac{C_3}{N^2} \], can reach the S.V.P. line only when
becomes infinite. However it must cross the S.V.P. line at the onset of liquid-only flow, that is, at finite mass flow-rate. The reason for these conflicting requirements is that the expression for mass limiting flow

\[ \dot{M}_2 \propto \frac{1}{\sqrt{\Delta T_1}} \]

predicts a flow-rate approaching infinity as the temperature fall (\( \Delta T_1 \)) approaches zero, that is, as the onset of liquid-only flow approaches. Therefore according to this model, liquid-only flow cannot occur. In practical terms the large mass limiting flow-rate could not be sustained by the modest pressure differential (\( P_e - P_{amb} \)) across the second orifice and therefore the theory fails at very low values of the temperature fall (\( \Delta T_1 \)). Consequently the part of the \( P_e \) curve for which \( \Delta T_1 \) was very low, was obtained by interpolation between the position relating to the onset of liquid-only flow and the theoretical curve at high values of \( \Delta T_1 \), as shown.

From Fig. 6.7 an assessment of the magnitude of the required correction may be made. It is seen that the operating point is very close to the ideal point and that the required correction is small. It is also seen that operating point is far removed from the region of very low \( \Delta T_1 \) where the theory is considered to be inapplicable.

The required correction may now therefore be derived analytically. In this analysis the fractional change in the absolute value of the expansion chamber pressure, which will be very small, is considered to be negligible. Also, metastability is included but its magnitude is considered to remain constant.

Now from equations developed in Section 4,

\[ \dot{M}_1 (\text{expt.}) = \dot{M} (\text{expt.}) = \left( \frac{\pi}{4} \right) C_1 (D_t)^2 \sqrt{2 f_1 (C_3 \Delta T_1 + \Delta P_{ex} + \Delta P_{meta})}, \]
where, $\Delta P_{\text{meta}} = \text{SVP}_{Te} - P_e$, as before.

Now, if $T_1 = 20^\circ C$, then denoting by $\dot{\mu}^*$ the corrected value of a parameter and letting $\Delta P_{\text{ex}}$ become 0:

$$\dot{\mu}_1^* = \left(\frac{\pi}{4}\right) c_{q1} (d_1)^2 \sqrt{2} \rho_1 (c_3 \Delta T_1^* + \Delta P_{\text{meta}}),$$

where it is assumed that $\Delta P_{\text{meta}}$ has remained constant.

Now, $\frac{\dot{\mu}_1^*}{\dot{\mu}} = \sqrt{\frac{c_3 \Delta T_1^* + \Delta P_{\text{meta}}}{\Delta P_1}}$.

Also, $\dot{\mu}_2 (\text{expt.}) = \dot{\mu} (\text{expt.})$

$$= \left(\frac{\pi}{4}\right) c_{q2} (d_2)^2 \frac{\gamma + 1}{r_c^2 \gamma} P_e \sqrt{\frac{\gamma c_1}{c_2 (\Delta T_1)}}$$

and $\dot{\mu}_2^* = \left(\frac{\pi}{4}\right) c_{q2} (d_2)^2 \frac{\gamma + 1}{r_c^2 \gamma} P_e \sqrt{\frac{\gamma c_1}{c_2 (\Delta T_1^*)}}$, where it is assumed that $P_e$ has remained constant.

Therefore $\frac{\dot{\mu}_2^*}{\dot{\mu}} = \sqrt{\frac{\Delta T_1}{\Delta T_1^*}}$.

Now $\dot{\mu}_1^* = \dot{\mu}_2^*$.

Therefore $\frac{c_3 (\Delta T_1^*) + \Delta P_{\text{meta}}}{\Delta P_1} = \frac{\Delta T_1}{\Delta T_1^*}$.

Therefore $\Delta T_1^* = \frac{-\Delta P_{\text{meta}} + \sqrt{(\Delta P_{\text{meta}})^2 + 4c_3 (\Delta T_1)(\Delta P_1)}}{2c_3}$. 


In situations where saturation conditions prevail in the expansion chamber,
\[ \Delta P_{\text{meta}} = 0 \]
and
\[ \Delta T_1^* = \sqrt{\frac{(\Delta T_1) \Delta P_1}{c_3}} \]

The above analysis produced an expression for the temperature difference across the first orifice \( \Delta T_1^* \), corrected for sub-cooling of the propellant above the first orifice. In that analysis it was assumed that the temperature was at the standard 20°C. Generally, this will not be the case and a further correction is required. The situation is shown, again semi-quantitatively, in Fig. 6.8 where it is assumed that the excess pressure is zero and metastability is negligible. In this case it is seen that for a temperature higher than 20°C the measured temperature difference is greater than the reduced value and vice versa.

Fig 6.8 was derived in similar manner to Fig. 6.7. First, the standard relationship between mass flow-rate \( \dot{M} \) and the pressure drop across the first orifice \( \Delta P_1 \) was plotted. Secondly, the relationship between mass flow-rate and expansion chamber pressure \( P_e \) was plotted. This relationship, \( P_e \propto \dot{M}^2 \), was derived as follows. Assuming saturation conditions in the expansion chamber then :-
\[ \Delta P_1 = c_3 \Delta T_1 \]
Now, \( \dot{M} = \dot{M}_1 \propto \sqrt{\Delta P_1} \),
and, as before, \( \dot{M} = \dot{M}_2 \propto \frac{P_e}{\sqrt{\Delta T_1}} \).
Therefore \( P_e \propto \dot{M}^2 \).
Finally, the initial pressure, \( P_i \), was plotted. This is given by :-
\[ P_i = P_e + \Delta P_1 \].
FIG. 6.8. EFFECT OF INCREASE IN THE TEMPERATURE OF THE PROPELLANT ON THE FLOW THROUGH A TWIN-ORIFICE NOZZLE.
From the completed graph, it is seen that again the operating point is very close to the ideal operating point and that the correction is small.

Quantitatively this second correction to $\Delta T_1$ is readily obtained from equation 4.2.31. It is:

\[
\frac{\Delta T_1^*}{\Delta T_1} = \frac{\text{SVP at } 20^\circ \text{C} - \text{constant } x (\Delta P_{\text{meta}})}{\text{SVP at } T_i - \text{constant } x (\Delta P_{\text{meta}})}
\]

The term constant $x (\Delta P_{\text{meta}})$ is small compared with the S.V.P. Therefore, providing $\Delta P_{\text{meta}}$ remains approximately constant, this equation simplifies to:

\[
\frac{\Delta T_1^*}{\Delta T_1} = \frac{\text{SVP at } 20^\circ \text{C}}{\text{SVP at } T_i}
\]

Therefore the total correction is:

\[
\Delta T_1^* = \sqrt{\frac{\Delta T_1 \Delta P_i}{C_3}} \cdot \frac{\text{SVP}_{20}}{\text{SVP}_{T_i}}
\]

Appendix 1.3 shows the temperature drop across the first orifice, $\Delta T_1$, for different nozzle combinations. Also given is the corrected value, $\Delta T_1^*$. These data may now be compared with the predicting equation, 4.2.31, which was derived in Section 4. This equation is:

\[
\Delta T_1 = \frac{P_i - \Delta P_{\text{meta}} (1 + \frac{D_1}{D_2})^2}{C_3} \cdot \frac{D_1^2}{(1 + 2B\frac{D_1}{D_2})^2}
\]
where

\[ B = \frac{1}{\frac{\gamma + 1}{2} \cdot \frac{C_{q1}}{C_{q2}}} \sqrt{\frac{2\rho_1 c_2}{\gamma c_1 c_3}} \]

= 4.93 in this context.

Other symbols are defined in Section 4.

Before plotting this equation it is first desirable to choose a convenient abscissa. This may be done by considering the relative magnitude of the various terms in the above equation for \( \Delta T_1 \). These were discussed in Section 4 where it was shown to a first approximation the term containing \( \Delta P_{\text{meta}} \) is small compared with \( P_1 \). Also \( 2B \left( \frac{D_1}{D_2} \right)^2 \) is large compared with 1 for small values of \( \left( \frac{D_2}{D_1} \right)^2 \). Therefore an approximate version of the equation is:

\[ \Delta T_1 \approx \frac{P_1}{2B C_1} \left( \frac{D_2}{D_1} \right)^2 \]

Thus \( \left( \frac{D_2}{D_1} \right)^2 \) is a suitable choice of abscissa.

Fig 6.9 shows the full equation plotted against \( \left( \frac{D_2}{D_1} \right)^2 \), with \( D_2 \) as a parameter. Also shown are the experimental values of the temperature differential \( \Delta T_1 \), together with the corrected value \( \Delta T_1^* \); these two values are plotted in pairs vertically in line.

It is seen that agreement between experimental and predicted values is generally good, and that for values of \( \left( \frac{D_2}{D_1} \right)^2 \) less than about 2, the temperature drop is proportional to \( \left( \frac{D_2}{D_1} \right)^2 \), where the constant of proportionality is marginally dependant on the value of \( D_2 \).

It is also apparent that the correction to \( \Delta T_1 \) is generally small and that correlation between theory and experiment is not significantly improved by its introduction. A similar argument applies to mass flow-rate. For this reason no corrections will be made to the mass flow-rate data considered in the next sub-section.
KEY.

NOMINAL VALUES OF \( D_2 \)

- O. 35 mm.
- △ 0.45 mm.
- □ 0.70 mm.

- UNCORRECTED DATA.

THEORETICAL CURVES FOR GIVEN NOMINAL VALUES OF \( D_2 \):

- SMALL

- O. 35 mm
- O. 45 mm
- O. 70 mm

FIG. 6.9. TEMPERATURE FALL ACROSS THE UPSTREAM ORIFICE.
6.4 Experimental verification of the expression for the mass flow-rate through a two-orifice nozzle assembly.

In the previous sub-section experimental support for the theoretical expression for the temperature drop across the first orifice $\Delta T_1$ was given. This expression is required to predict the mass flow-rate and exit velocity of the spray in terms of fundamental data. In this sub-section predicted mass flow-rates will be compared with theory while the exit velocity of the spray will be considered in a later sub-section, 6.6.

A fundamental expression for the mass flow-rate may be obtained by considering flow through the first (upstream) orifice. The equation for this flow was shown in sub-section 6.2.1. to be:

$$\dot{m} = C_{Q_1} \left( \frac{\pi}{4} \right) D_1^2 \sqrt{2 \rho_1 (P_i - P_e)}$$

In that sub-section a value of 0.78 was derived for $C_{Q_1}$.

Now if the propellant, both above the first orifice and in the expansion chamber is saturated, then:

$$P_i - P_e = C_3 \Delta T_1$$

as discussed previously.

Therefore

$$\dot{m} = 0.78 \left( \frac{\pi}{4} \right) D_1^2 \sqrt{2 \rho_1 C_3 \Delta T_1}$$

where $\Delta T_1$ is given by equation 4.2.31.

If metastability is significant then the equation becomes:

$$\dot{m} = 0.78 \left( \frac{\pi}{4} \right) D_1^2 \sqrt{2 \rho_1 (C_3 \Delta T_1 + \Delta P_{meta})}$$

Here $\Delta P_{meta}$ may be obtained from the empirical equation, 4.2.27.
This last equation may be further developed by substituting the expression (equation 4.2.31) for $\Delta T_1$

Therefore

$$\dot{m} = 0.78 \left(\frac{\pi}{4}\right) D_1^2 \sqrt{2 \gamma \rho_1} \frac{C_3}{C_0} \left(\frac{P_1 - \Delta P_{meta}(1 + B(D_1/D_2))}{C_3 + 2BC_3 \frac{D_1}{D_2}}\right)^{2}$$

where again

$$B = \frac{1}{2 \gamma \left(\frac{\gamma + 1}{\gamma - 1}\right)} \sqrt{\frac{C_{91}}{C_{92}}} \left(\frac{2 \gamma}{\gamma - 1}\right)$$

$$= 4.93$$

Other symbols are as defined in Section 4.

Therefore:

$$\dot{m} = 0.78 \left(\frac{\pi}{4}\right) \sqrt{2 \gamma \rho_1} D_1^2 \frac{C_3}{C_0} \left(\frac{P_1 - \Delta P_{meta}(1 + B(D_1/D_2))}{C_3 + 2BC_3 \frac{D_1}{D_2}}\right)^{2}$$

$$= 0.78 \left(\frac{\pi}{4}\right) \sqrt{2 \gamma \rho_1 D_1 D_2} \frac{P_1 + \Delta P_{meta} \left(\frac{D_1}{D_2}\right)}{(2B + \frac{D_2}{D_1})^2}$$

It is seen that the two terms containing $\Delta P_{meta}$ have partly cancelled. Further for most nozzles the term $\Delta P_{meta} B(D_1/D_2)^2$ is small compared with $P_1$. Therefore, ignoring initially, the metastability term, the mass flow-rate $\dot{m}$ may be plotted linearly against $D_1 D_2$ with $D_2/D_1$ as parameter.

Fig. 6.10 shows the above equation plotted, with the parameter $D_2/D_1 = 0.1$ and 2, thus covering the range of orifice diameter ratios.
THEORETICAL CURVES FOR THE THREE GIVEN VALUES OF $D_2/D_1$:

**KEY**
- **D2/D1**
- **SMALL**
- **1**
- **2**
- **--- DENOTES METASTABILITY CORRECTION.**

**FIG. 6.10. MASS FLOW RATE THROUGH A TWO-ORIFICE NOZZLE.**
used in this work. Experimental data are also plotted. In this case, however, for reasons given in the previous sub-section, no correction was made for sub-cooling or a non-standard initial temperature. For four of the nozzles the assumption of negligible metastability was not valid and a correction to the simplified equation was made; this is shown as a dashed line related to the relevant data point.

It is apparent from Fig. 6.10 that generally, the linear relationship between mass flow-rate (\( \dot{m} \)) and the product of the diameters of the two orifices of the nozzle assembly \((D_1D_2)\) is borne out by the data. Further the predicted trend of a reducing mass flow-rate with increasing orifice diameter ratio \(D_2/D_2\) is also supported. However, it is seen that the slope of the theoretical prediction is slightly lower than required by the data when the ratio \(D_2/D_1\) is 0 or 1; an increase of slope by 7% would produce a better correlation. This would require a discharge coefficient of 0.84, instead of the 0.78 determined in sub-section 6.2.1.

When the ratio \(D_2/D_1\) is approximately 2, however, agreement between data and the simplified equation (in which metastability is ignored) is good. The necessary metastability correction impairs the agreement. However, because only two data points with \(D_2/D_1\) equal to 2 are available, this finding should perhaps be treated with caution.
6.5 Measurement of the velocity ratio between liquid and gas phases at the exit of the nozzle assembly.

In the last section, it was shown that mass flow-rate through a two-orifice nozzle assembly could be adequately predicted in terms of fundamental data and an empirical expression for the metastability of the propellant in the expansion chamber. The mass flow-rate and the exit velocity have been shown to be the two main parameters required to describe velocity profiles within a spray. Having shown that mass flow-rate can be adequately predicted it is then necessary to show that exit velocity can also be adequately predicted. However before the exit velocity is considered, it is appropriate to verify an important assumption made in the theoretical development in Section 4. This is that during the passage of two-phase propellant through the downstream orifice, the ratio of the velocity of the liquid propellant to that of its vapour is unity. If this ratio, sometimes called the "slip" factor, were to depart significantly from unity then the development of theoretical expressions in Section 4 would require significant modification. Experimental evidence is now presented in support of this assumption.

The velocity ratio between phases was determined by analysing double-flash photographs taken of the spray at the nozzle housing exit. The analysis consisted of the measurement of the displacement and diameters of pairs of particles. The velocity of the gaseous phase was obtained by measurement of the velocity of very small particles.

For this method to be effective therefore, it was desirable to obtain the displacement of particles below about 10μm in diameter. In order that pairs of particles of this diameter could be identified against a background crowded with other particles, a flash interval to about 2 or 3 microseconds was necessary. However, with such a small time interval
and because of the limitations of the resolution of the oscilloscope and interval timer, absolute velocity measurements were rather inaccurate. However velocity ratio measurements were satisfactory.

A total of 16 double flash photographs were analysed. From each photograph, the distance between all identifiable pairs of particles was measured, together with the particle diameter. The data were classified according to particle size. The mean displacement for each class of particle diameter was then calculated; this was a spatial mean displacement. From these mean values the overall mean over a number of photographs or the temporal mean, was calculated for each diameter range for a given nozzle. Also calculated were the standard deviation and standard error of the mean. Thus, three statistical indices were determined for each nozzle for a nominally constant flash interval. Statistical indices were calculated for the temporal data rather than the spatial data because the former, being the mean of data from several photographs were considered the more reliable.

The results are shown in Table 6.1, overleaf. The displacement between the particles is shown as a "nominal" velocity, that is, displacement divided by nominal flash interval. It is seen that there is negligible variation in particle velocity with increasing particle size, up to a particle diameter of about 50 μm, the maximum observed.
<table>
<thead>
<tr>
<th>NOZZLE</th>
<th>DISTANCE FROM NOZZLE EXIT</th>
<th>NUMBER OF PHOTOGRAPHS</th>
<th>NUMBER OF PARTICLES</th>
<th>MEAN &quot;NOMINAL&quot; VELOCITY m/sec</th>
<th>STANDARD DEVIATION m/sec</th>
<th>STANDARD ERROR OF MEAN m/sec</th>
<th>IN THE GIVEN PARTICLE SIZE RANGE (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>320 x 350</td>
<td>0 mm  2.4 µsec</td>
<td>3</td>
<td>23</td>
<td>5</td>
<td>29</td>
<td>2</td>
<td>1</td>
</tr>
<tr>
<td>320 x 350</td>
<td>5 mm  2.4 µsec</td>
<td>4</td>
<td>40</td>
<td>2</td>
<td>26</td>
<td>25</td>
<td>2</td>
</tr>
<tr>
<td>320 x 450</td>
<td>0 mm  3.5 µsec</td>
<td>4</td>
<td>18</td>
<td>2</td>
<td>38</td>
<td>4.0</td>
<td>1</td>
</tr>
<tr>
<td>450 x 450</td>
<td>5 mm  3.5 µsec</td>
<td>4</td>
<td>52</td>
<td>4</td>
<td>32</td>
<td>4.5</td>
<td>1</td>
</tr>
<tr>
<td>680 x 700</td>
<td>0 mm  2.5 µsec</td>
<td>1</td>
<td>15</td>
<td>3</td>
<td>31</td>
<td>33</td>
<td>2</td>
</tr>
</tbody>
</table>

*Nominal* velocity = distance between image pairs
nominal flash interval

TABLE 6.1  DETERMINATION OF VELOCITY RATIO BETWEEN LIQUID AND VAPOUR PHASES:
VARIATION OF EXIT VELOCITY WITH PARTICLE SIZE.
6.6 Measurement of the exit velocity of the spray and comparison with theory.

Having shown, in the last sub-section, that the concept of a single exit velocity independent of particle size, is applicable to a spray, it is appropriate to compare experimental values of this exit velocity with theoretical predictions. Having done this, then the two main parameters required to describe the velocity profiles within the spray — mass flow-rate, and exit velocity, will have been considered.

Two expressions from which the exit velocity of the spray may be derived were given in Section 4. The first was applicable to non-choking flow (equation 4.2.7) and gave the exit velocity, $u_0$, directly. The second was applicable to choking flow (equation 4.2.12) and gave an expression for the velocity of a pressure pulse $u_p$ through the two-phase propellant at the point of choking within the downstream orifice. The exit velocity was then assumed to be equal to this pressure pulse velocity.

However it was demonstrated in Section 4 that the static pressure within the nozzle, because of choking, was higher than ambient, and that within a few nozzle diameters downstream the static pressure had returned to ambient with a consequent rise in the velocity of the jet. In practice it was possible to photograph the spray only after all, or a substantial part of this expansion had taken place. This was because of the masking effect of the nozzle housing. Consequently the measured jet velocity was expected to be somewhat higher than the sonic velocity, $u_s$. This increase in velocity may be determined theoretically by comparing the initial total momentum of the jet with that of the expanded jet. The initial momentum of the jet was shown to be; assuming, in the first instance no orifice entry losses:—

$$\dot{M} u_s + A_2 (P_c - P_{amb})$$

where $\dot{M}$ is then the mass flow-rate, $A_2$ the downstream orifice area.
and \( P_c - P_{amb} \) is the gauge critical pressure.

The momentum of the expanded jet, assuming no significant entrainment has taken place, is:

\[
\dot{M} u_n
\]

where \( u_n \) is the higher, expanded jet velocity.

Therefore

\[
\dot{M} u_n = \dot{M} u + \frac{A_2}{\dot{M}} (P_c - P_{amb})
\]

The measured exit velocity may now be compared with \( u_n \) above. As before a comparison with the non-choking exit velocity \( u_0 \) will also be given.

In order to determine the theoretical value for \( u_n \), the value of the sonic velocity \( "u_s" \) must first be determined. This was given equation 4.2.12 as:

\[
u_n = \left( \frac{\gamma q P_c^{\frac{1}{2}}}{g_c} \right) \left( 1 + \frac{1 - \frac{\rho_{gc}}{\rho_1}}{q_{gc}} \right),\]

where the parameters were defined in Section 4.

The variables are to be determined at the point of choking.

Therefore, as before, substitutions may be conveniently made as follows:

\[
P_c = r_c P_e,
\]

\[
\rho_{gc} = \rho_{ge} (r_c)^\frac{1}{\gamma} = C_1 P_e (r_c)^\frac{1}{\gamma},
\]

and \( q = c_2 \Delta T_1 \)

Therefore the equation becomes:

\[
u_n = \left( \frac{\gamma c_2 \Delta T_1 r_c P_e^{\frac{1}{2}}}{C_1 P_e r_c^{\frac{1}{\gamma}}} \right) \left( 1 + \frac{(1 - c_2 \Delta T_1) C_1 P_e (r_c)^{\frac{1}{\gamma}}}{c_2 \Delta T_1 \rho_1} \right)
\]
Inserting values of \( r_1, c_1, c_2 r_0 \) and \( \gamma \) determined previously then:

\[
\mathbf{u}_s = 10.9 \left( \Delta T_1 \right)^{\frac{1}{2}} \left( 1 + (1 - 0.0061 \Delta T_1) \right) 3.65 \times 10^{-6} \frac{P_e}{\Delta T_1} \text{ m/s,}
\]

where \( \Delta T_1 \) is in deg. C and \( P_e \) in N/m² absolute.

The theoretical, non-choking, expression for the exit velocity may now be considered. This was given in equation 4.2.7 as:

\[
\frac{u_0^2}{2} = \frac{1 - \frac{q}{\rho_1} (\rho_e - \rho_{amb})}{\rho_1} + \frac{q \frac{1}{\gamma} P_e}{\rho_{ge} \left( 1 - \frac{1}{\gamma} \right)} (P_e^{1 - \frac{1}{\gamma}} - \rho_{amb}^{1 - \frac{1}{\gamma}})
\]

or

\[
u_0 = \sqrt{2 \left[ \frac{\left( 1 - 0.0061 \Delta T_1 \right) (P_e - \rho_{amb}) + 7.67 \times 10^2 \Delta T_1 \left( 1 - \frac{\rho_{amb}}{P_e} \right)^{1 - \frac{1}{\gamma}}}{1.41 \times 10^{10}} \right]^{\frac{1}{2}}}
\]

The two equations for the exit velocity \( u_n \) and \( u_o \) may now be compared with exit velocity data. In order that the comparison be direct, measured values of the two variables, the temperature fall across the first orifice \( \Delta T_1 \) and the expansion chamber pressure \( P_e \), were inserted into the equations rather than the values obtained from theoretical predictions. However, it may be shown that the difference between the theoretical curve derived from fundamental data and that derived from empirical data, for each of the two-expressions for the exit velocity will be small. This is because the methods of predicting the two variables, \( \Delta T_1 \) and \( P_e \) (indirectly), have been shown to be satisfactory.

The values of \( u_n \) and \( u_o \) are compared with measured exit velocity data in Table 6.2, overleaf. It is seen from this Table that the data were obtained from two sources, from spray velocity profile measurements to be discussed in sub-section 6.9 and also from the "slip" factor velocity measurements given in the previous sub-section. As previously stated these latter measurements gave accurate values for the velocity ratios, the parameter then required, but inaccurate values for absolute velocity. The
<table>
<thead>
<tr>
<th>NOZZLE</th>
<th>CALCULATED &quot;SONIC&quot; VELOCITY ((u_s^\text{calc})) m/sec</th>
<th>(u_n) (theoretical) (= u_s + \frac{A_2(r_0^2P_0 - P_\text{amb})}{M}) m/sec</th>
<th>CALCULATED VELOCITY FOR NON CHOKE FLOW (u_s) m/sec</th>
<th>MEASURED EXIT VELOCITY ((u_n)) When Static Pressure=Ambient STANDARD DEVIATION, STANDARD ERROR OF THE MEAN (m/sec)</th>
<th>SOURCE OF DATA VELOCITY PROFILE (Sec. 6.9)</th>
<th>VELOCITY RATIO (sec. 6.5)*</th>
<th>VELOCITY COEFFICIENT (= \frac{u_n}{u_s + A_2(r_0^2P_0 - P_\text{amb})} \frac{1}{M})</th>
</tr>
</thead>
<tbody>
<tr>
<td>320 x 350</td>
<td>27.3</td>
<td>37</td>
<td>35</td>
<td>28, 4, 1.8</td>
<td>26.5</td>
<td>2, 1.4</td>
<td>0.76</td>
</tr>
<tr>
<td>320 x 450</td>
<td>30.4</td>
<td>40</td>
<td>39.5</td>
<td>-</td>
<td>38</td>
<td>2</td>
<td>0.95 * (from velocity ratio)</td>
</tr>
<tr>
<td>320 x 700</td>
<td>39</td>
<td>47</td>
<td>47</td>
<td>30</td>
<td>-</td>
<td>-</td>
<td>0.64</td>
</tr>
<tr>
<td>450 x 450</td>
<td>26.4</td>
<td>35.2</td>
<td>33.5</td>
<td>29.5, 1, 0.7</td>
<td>31</td>
<td>3</td>
<td>0.83</td>
</tr>
<tr>
<td>II</td>
<td>25.4</td>
<td>36</td>
<td>31</td>
<td>28, 1</td>
<td>-</td>
<td>-</td>
<td>0.78</td>
</tr>
<tr>
<td>(estimated from data of nozzle 680 x 700)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>III</td>
<td>not available</td>
<td></td>
<td></td>
<td>27.3, 2.5, 1.5</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**Table 6.2**

COMPARISON BETWEEN THE MEASURED EXIT VELOCITY OF THE SPRAY AND CALCULATED VALUES

*Nominal values only

**See text**
error is estimated to be of the order of 20%. Data from the velocity profile measurements were of adequate accuracy but the number of velocity profiles measured was limited to five. It is because only a small number of such measurements were available that "slip" factor data, which would otherwise have been rejected because of its inaccuracy, were included.

From Table 6.2 it is seen that the velocity coefficients, each given by the ratio of the measured exit velocity, \( u_n \), (from velocity profile data) to the theoretical value of \( u_n \), are within the range 0.64 to 0.83. These values are comparable with the value of 0.83 for the discharge coefficient of those short tubes given in Appendix 3 for air flow, and with 0.78 given in sub-section 6.2.2 for two-phase flow. As the flow regime through a short tube is that of full tube flow, then the velocity coefficient would be expected to be equal to the discharge coefficient. That such an agreement, albeit only approximate, is obtained is taken as further support for the theoretical description of two-phase flow through the downstream orifice.

It is useful to compare the value of the exit velocity for non-choking flow, \( u_0 \), with the sonic velocity, \( u_s \). This comparison is made in Fig. 6.11 where equations 4.2.7, giving \( u_0 \) and 4.2.12 giving \( u_s \) are plotted against downstream to upstream orifice diameter ratio \( D_2/D_1 \) with \( D_2 \) as parameter. The equations were plotted as previously described, from empirical values of \( \Delta T_1 \) and \( P_e \) for each nozzle. However for those areas where no nozzle data were available, and where interpolation was not possible, the theory was used to extrapolate the curve from an adjacent data point. Such situations are denoted by plain points as shown. The two equations may now be compared. It is seen that over the range of orifice diameter ratios tested, from about 0.5 to about 2, the two equations predict velocities within about 20% of each other, with the sonic velocity the lower as would be expected. Further, over this range, the sonic velocity varies over the rather narrow range from
FIG. 6.11. COMPARISON BETWEEN SONIC VELOCITY AND NON-CHOKING EXIT VELOCITY.
KEY TO FIG. 6.11

○ Theoretical value of exit or sonic velocity for $D_2 = 350 \, \mu m$
△ " " " " " $D_2 = 450 \, \mu m$
□ " " " " " $D_2 = 700 \, \mu m$, (nominal)

Expansion chamber conditions determined from experimental data.

Extrapolated or interpolated theoretical values.

○ Experimental value of the exit velocity, $D_2 = 350 \, \mu m$
△ determined at the nozzle housing, after $D_2 = 450 \, \mu m$
□ expansion of the jet. $D_2 = 700 \, \mu m$, (nominal)
25 m/sec. to 40 m/sec. while the non-choking velocity varies from 25 m/sec to 47 m/sec.

Beyond this range of orifice diameter ratios, for values of $D_2/D_1$ less than 0.5, the conventional, non mass limiting, equation predicts the lower velocity. The reason for this reversal, as described previously, is that at these low values of orifice diameter ratio, $D_2/D_1$, the fraction of propellant evaporated is very low, the mean density of the propellant approaches that of the liquid, and the expansion chamber pressure is no longer capable of sustaining the high "sonic" discharge velocity. Thus for a value of $D_2/D_1$ less than 0.5 the conventional equation must be used. The limiting velocity as $D_2/D_1$ approaches 0 is that of the discharge of liquid propellant of density 1.4 kg/m$^3$ sustained by a pressure differential equal to:

$$(\text{the initial pressure, } 390 \text{ kN/m}^2 \text{ - ambient pressure}).$$

This discharge velocity is 20 m/sec.

It is also apparent that the two curves will cross not only at low values of $D_2/D_1$, but also at a value of $D_2/D_1$ greater than 2. This is because, for high levels of $D_2/D_1$, the temperature fall, and hence the pressure fall, across the first orifice will be high; consequently the expansion chamber pressure will be low. Again, therefore, the expansion chamber pressure would not be able to sustain the high sonic discharge velocity.

Values of the measured exit velocity, $u_n$, are also plotted in Fig. 6.11. Comparisons between measured and theoretical values were discussed previously.
6.7 Experimental determination of the transverse profile of longitudinal velocity within the spray.

The main parameters in the equation describing the longitudinal profiles of velocity within a spray have been discussed. These were the mass flow-rate and the exit velocity. Before the applicability of the velocity profile equation can be tested directly, however, two further assumptions made in Section 4 will be considered, and the measurement of a minor empirical parameter will be described. The first of the two assumptions is that the transverse profile of longitudinal velocity of the gaseous component of the spray was Gaussian and the second is that the divergence angle of the spray was approximately equal to that of an air jet; the minor empirical factor is the position of the apparent origin of the spray. The first assumption is considered in this sub-section, the second, together with the position of the spray origin, is considered in the next sub-section.

The assumption of a Gaussian transverse profile of longitudinal gaseous velocity scarcely needs experimental verification as there are many reports in the literature (49, 21, 34 for example) which show that the transverse distributions of velocity within single phase jets, liquid sprays and dust-laden gas jets are Gaussian. However, experimental determination of this transverse profile not only verifies the assumption but also enables an assessment to be made of the divergence angle of the jet.

Because a substantial amount of evidence already exists to vindicate the first assumption only one such profile was measured. This was for a distance of 80 mm from nozzle III; the profile is shown in Fig. 6.12. It was developed from measurements of the diameter and velocity of aerosol particles, as described later. Also plotted in Fig. 6.12 is
FIG. 6.12 TRANSVERSE PROFILE OF LONGITUDINAL VELOCITY.

\[ u_g = e^{-11000 \gamma^2} \]
the Gaussian function:

\[ u_g = 11 e^{-1000y^2} \text{ m/sec.}, \]

where \( u_g \) is the velocity of the gaseous component and \( y \) (in metre) is the off-axis distance.

It is seen that agreement between the data and this function is reasonably good except at distances from the axis of greater than about 20mm. Beyond this distance the spray velocity is of the order of 0.5 m/sec. while the Gaussian distribution function predicts 0.14 m/sec. and \( 0.5 \times 10^{-3} \) m/sec at off-axis distances of 20mm and 30mm respectively. The reason for this is presumably that the measured velocity of about 0.5 m/sec., which is vertically downwards, is the sedimentation velocity of the aerosol cloud at the periphery of the spray. A velocity of this magnitude is to be expected for the sedimentation of such a cloud. This is because the terminal velocity of 25μm particles, for example, is about 0.25 m/sec and a cloud of such particles would fall at a higher velocity depending on its concentration. Thus a cloud containing a range of particle sizes up to about 40 μm, as is the case with these sprays, may well fall at 0.5 m/sec. with the larger particles falling considerably faster. This relative distribution of velocities is found to be the case as is demonstrated in the latter part of this sub-section.

The data points in Fig. 6.12 appear to be distributed slightly asymmetrically about the ordinate. This is probably due to an error in the setting of the nozzle relative to the camera. As explained in Section 5, this alignment was achieved by forcing water through the nozzle and then by focusing on the water jet. An off-set of about 2mm, which is sufficient to cause the asymmetry, would be difficult to eliminate at a distance of 80mm. Also it is possible that the water jet and spray do not have a common axis.

The development of Fig. 6.12 may now be considered. Each data point,
representing the velocity of the gaseous component of the spray at a given off-axis distance, was obtained as follows. At the given point in the spray a number of double flash photographs were taken as explained in Section 5. Each photograph was analysed and a distribution of spatial mean particle velocity against particle size was obtained. The overall temporal mean velocity of each class of particle size, \(<5\mu m, 5-10\mu m\) etc. was then found by averaging the spatial mean velocities. This distribution, of temporal mean velocity against particle size which is generally parabolic as discussed in sub-section 6.10, was plotted and extrapolated to zero particle diameter. The velocity thus obtained was taken to be that of the gaseous phase. Such distributions are shown in Fig. 6.13.

The standard deviation and the standard error of the mean for each data point are given in Figs. 6.12 and 6.13, providing detail would not have been obscured. As these statistical indices relate to the temporal variation in particle velocity the standard deviation is thus a measure of the turbulence at that point within the spray. The standard error of the mean, as usual, is approximately equal to the 68% confidence limit.

As the standard deviation is a measure of turbulence, then the relative turbulence within the spray is readily determined. It is instructive to compare the relative turbulence within the spray to that within an air jet. Relative turbulence is defined as:

\[
\text{RMS turbulence} = \frac{\text{RMS turbulence}}{\text{mean velocity}}
\]

In the case of the spray it may be obtained by dividing the standard deviation, discussed above, by the mean velocity at that point. For the air jet, values of relative turbulence are well documented, and there is very good agreement between authors. Thus Townsend (66) quoting many authors gives the variation of relative turbulence across a turbulent air jet in the self-preserving region of the jet, that is, at a distance of more than about 15 nozzle diameters downstream. This variation is
FIG. 6.13. TRANSVERSE VARIATION OF LONGITUDINAL VELOCITY WITHIN A SPRAY.

KEY.

- CENTRE.
- RIGHT (mm)
  - ◼ 6
  - ▼ 10
  - △ 15
  - ◼ 20
  - ◼ 25
  - ● 30

LEFT (mm)

- △ 5
- ◆ 10
- ◼ 20
- ● 30

PARTICLE DIAMETER.

PARTICLE VELOCITY (m sec⁻¹)
shown in Fig 6.14, where the relative turbulence is plotted as a function of ratio of local mean velocity to axial velocity. Also plotted for comparison are the values of relative turbulence, as defined above, for the spray. It is seen that in the central region of the jet:

$$0.5 \leq \text{(ratio of local mean to axial velocity)} \leq 1.0,$$

the relative turbulence of the spray is very close to that of the air jet. At the outer regions of the spray, agreement is not so good.

It is concluded therefore that, on the basis of data from this one nozzle, the transverse distribution of longitudinal velocity is Gaussian, in the developed region of the spray, and that the turbulence properties of the spray are similar to those of an air jet.
FIG. 6-14. COMPARISON BETWEEN THE TURBULENCE OF AN AIR JET AND THAT OF AN EXPERIMENTAL SPRAY.
6.8 Measurement of the divergence angle of the spray, and the determination of the position of its apparent origin.

Two assumptions made in the theoretical development in Section 4, have now been verified. A third, that the divergence angle of the spray was approximately equal to that of an air jet, may now be considered. A related parameter, the position of the apparent origin of the spray will also be considered.

6.8.1 Measurement of the divergence angle of the spray.

It was shown in Section 2 that the developed spray consisted mainly of entrained air. It is therefore relevant to compare experimentally-determined spray angles with the spray angle for a circular turbulent air jet. This angle is given by Townsend (66), again quoting numerous references, as 4.8° for the half-angle of air jets, determined for the locus of points where the mean off-axis longitudinal velocity has fallen to half the mean axial longitudinal velocity. This value may be converted to the half-angle corresponding to 1/e of the axial velocity, the angle used in Section 4, by assuming a Gaussian lateral distribution for longitudinal velocity; thus, this 1/e-velocity half-angle of an air jet is 5.8°.

The effect of loading the air jet with an aerosol may be considered. Laats (34), for example, has shown that loading an air jet with ash reduced the spread of the jet; this consequently resulted in a reduced longitudinal attenuation of the jet. It was found however that a mass flow-rate of ash equal to the initial mass flow-rate of the air reduced the half-velocity divergence half-angle by only a very small angle. This was from 4.85° (a 1/e-velocity angle of 5.85°) to 4.3° (a 1/e-velocity angle of 5.2°). This applied at a distance downstream of 15 to 20 nozzle diameters. At this downstream distance the mean ratio, by mass, of ash to air is estimated to be approximately 0.2. This situation can
now be related to the present case. This same ratio, by mass, of liquid aerosol to entrained air would be obtained, for a non-evaporating propellant at a distance of about 160mm from the 680 x 700 nozzle assembly and about 80mm from the 320 x 350 nozzle assembly. Further, the propellant is evaporating and therefore the aerosol-to-gas ratio would be considerably less than the value of 0.2 quoted above. It would not seem unreasonable to assume, therefore, that beyond a few centimetres downstream of the nozzle, the liquid aerosol content will have negligible effect on the divergence angle. As the vapour concentration rapidly diminishes also, the divergence half-angle of the spray will be taken as that of an air jet, which is 5.8°, for the 1/e-velocity locus, beyond the first few centimetres.

Limited experimental support for this assumption may be obtained from the data given in the previous sub-section. There, it was seen that at a distance of 80mm from the nozzle the spray half-width, corresponding to 1/e of the axial velocity was given implicitly by:

\[
\frac{1}{e} = e^{-11000y^2},
\]
\[
\text{or } y = 9.5\text{mm}
\]

The average divergence angle over the first 80mm of the spray may now be determined providing the position of the apparent origin of the spray is known. With air jets this origin is downstream of the nozzle. With the spray generators in these experiments the spray-origin appeared to be upstream of the nozzle; this was because of the rapid expansion immediately downstream of the nozzle, as described earlier. An assessment of the position of the apparent origin of the spray was obtained as described in 6.6.2. The result is quoted here. For nozzle assemblies whose orifice diameters were approximately equal, the origin of the spray was estimated to be about 50mm upstream of the second orifice. For nozzle assemblies such as nozzle III for which the ratio of the second orifice diameter to that of the first was about 1.3, the corresponding upstream distance was approximately 25mm.
The divergence half-angle, corresponding to the $1/e$-velocity locus is therefore given by :-

$$\theta = \tan^{-1} \left( \frac{9.5}{(80 + 25)} \right) = 5.2^\circ.$$ 

This figure is in agreement with the value given by Laats for an ash-laden air-jet. That the two values coincide is fortuitous. This is because of the physical differences between the developing spray and the laden air-jet, and further because of the uncertainty in the value of the effective diameter of the spray, 9.5mm in this case, as was seen in the previous sub-section, and also because of doubt about the accuracy of the origin upstream of the spray, 25mm in this case, as is demonstrated in 6.8.2, below.

This agreement however does give limited support to the initial assumption that, beyond a few centimetres downstream of the nozzle, the effect of the aerosol and vapour content on the divergence angle of what is essentially an air-jet may be neglected.

6.8.2 Assessment of the position of the spray origin.

A description of the assessment of the position of the spray origin may now be given. This was obtained by taking photographs of the flood-lit spray at a distance of about 1 metre. The position of the origin of the spray was then determined by tracing the outline of the spray, from the projected image, and extrapolating upstream to the apparent origin. Because of the nebulous nature of the edge of the spray, this method is not precise. Further it is doubtful whether the origin, as determined by this method, is equivalent to the origin required by the mathematical development of Section 4, this latter being the apex of the cone which is the loci of points where the spray velocity has fallen to $1/e$ of the axial value. The most satisfactory method of
measuring spray-angle, therefore, would have been to have repeated the transverse scan, described in 6.7, at many distances from the origin. However in the absence of such data, the results of the photographic method will be assumed to give an approximate assessment of the position of the origin. Table 6.3 gives these results for a number of nozzle assemblies.

<table>
<thead>
<tr>
<th>NOZZLE</th>
<th>DISTANCE OF SPRAY ORIGIN UPSTREAM OF FINAL ORIFICE (z) ± STANDARD DEVIATION (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>320 x 350</td>
<td>26 ± 2</td>
</tr>
<tr>
<td>450 x 450</td>
<td>29 ± 1</td>
</tr>
<tr>
<td>680 x 700</td>
<td>37 ± 6</td>
</tr>
<tr>
<td>320 x 450</td>
<td>26 ± 4</td>
</tr>
<tr>
<td>320 x 700</td>
<td>10 ± 1</td>
</tr>
</tbody>
</table>

**TABLE 6.3** ESTIMATION OF THE POSITION OF THE SPRAY ORIGIN, OBTAINED BY MACROPHOTOGRAPHY OF THE SPRAY.

It is seen that the position of the apparent origin of the spray varies from 26mm to 37mm upstream of the nozzle for orifice pairs of comparable diameter, while for the 320 x 700 nozzle assembly the spray origin was 10mm upstream. That this latter value should be significantly smaller than the others is to be expected. This is because such a nozzle combination generates a relatively low expansion chamber pressure \((243 \text{ kN/m}^2)\). Thus the excess pressure in the jet at the nozzle exit, which is responsible for the movement upstream of the spray origin, as explained in Section 4, is as follows:

\[
r_c \times P_e - P_{\text{amb}} = (r_c 243 - 101) \text{ kN/m}^2 \quad \text{or, for } r_c = 0.5,
\]

\[
= 21 \text{ kN/m}^2.
\]
This value of 21 kN/m² may be compared with a typical value, for nominally twin orifice nozzles, of:

\[(366 \times r_0 - 101) \text{ kN/m}^2\], or 82 kN/m², which is four times larger.

As the movement upstream of the spray origin depends on the excess pressure in the jet, it was anticipated that the apparent origins of the three nominally twin-orifice nozzles would be the same. This may possibly be the case, and the differences shown in Table 6.3 may merely reflect a deficiency in the method of measurement. It would be expected, for example, that as the mass flow-rate increased, with increasing nozzle diameter, the apparent width of the spray, as recorded photographically, would increase. This is because, at a given off-axis distance, the concentration of spray particles would be larger for the larger mass flow-rate. As the density of the image on the photographic film depends among other things, on this concentration, the apparent dimensions of the spray would appear to increase with increasing nozzle diameter, although geometrically they had remained constant. In the absence of further data a mean value of 30mm will therefore be taken as the distance upstream of the apparent origin of the spray for twin orifice nozzles. This distance will be considered to diminish as the ratio of the diameters of second to first orifices rises. Thus for nozzle \( \text{III} \), for which the position of the spray origin was required earlier, the origin will be taken as 25mm upstream; this figure is similar to that given in the Table for nozzle 320 x 450 which has a similar diameter ratio.

It is apparent that this photographic method of determination of the distance upstream of the apparent origin of the spray is very unsatisfactory. However as this distance may be regarded as being merely a correction to the longitudinal dimension of the jet, it is considered that such a treatment is justified.
6.9 Measurement of the longitudinal profile of longitudinal component of axial velocity of the gaseous component within the spray, and comparison with theory.

All the steps in the development of the equation (4.1.10) for the longitudinal profile of the axial longitudinal velocity of the gaseous component \( u_{ga} \) within a spray have now been considered. This equation, which is applicable beyond a few centimetres downstream of the nozzle, is:

\[
\frac{1}{(x + z) \tan \theta} \sqrt{\frac{2}{\pi}} \frac{\dot{M} u_n}{\rho_{air}}
\]

where

- \( x \) is the distance downstream of the nozzle,
- \( z \) is the distance upstream of the nozzle of the apparent origin of the spray
- \( \alpha \) is the half-angle of the spray corresponding to the \( 1/e \) - velocity locus,
- \( \dot{M} \) is the mass flow-rate,
- \( u_n \) is the velocity of the expanded jet, in the case of choking flow, measured before significant entrainment has taken place. For non-choking flow this is the exit velocity \( u_0 \).
- \( \rho_{air} \) is the density of the entrained air.

The denominator, \( f \), is given by:

\[
f = 1 + \frac{0.26}{(x + z) \tan \theta} \sqrt{\frac{2}{\pi}} \frac{\dot{M}}{u_n \rho_{air}}
\]

In this application, \( f \), is approximately equal to 1.0 a few centimetres beyond the nozzle.

It would be convenient to be able to reduce this equation to a dimensionless form, applicable to all self-propelled aerosol generators.
This form would most conveniently be: \[ \frac{u_{g1}}{u_n} \ll \frac{1}{x'} \], where \( x' \) is a hitherto undefined dimensionless form of \( (x + z) \). The equation for the velocity profile would then be similar to that of an air jet.

To a limited extent, this may be done by invoking the linear relationship, determined in sub-section 6.4, between the mass flow-rate (\( \dot{M} \)) and the product of the diameters of the two orifices \( (D_1D_2) \). Thus:

\[ \dot{M} = \frac{G}{\sqrt[4]{4}} (D_1D_2), \]

where \( G \) is therefore the mass flow-rate divided by the geometric mean area of the two orifices, and is thus analogous to the "mass velocity" for a single orifice nozzle.

Substitution for \( \dot{M} \), division by \( u_n \) and calling \( x' = (x + z)\sqrt{\frac{D_1D_2}{D_1}} \) gives for the above equation (4.1.10):

\[
\frac{u_{g1}}{u_n} = \sqrt{\frac{\frac{G}{\sqrt{2u_n\tan^2\alpha}}}{x' + .26 \sqrt{\frac{\frac{G}{2u_n\tan^2\alpha \rho_{air}}}{\sqrt{\frac{2u_n\tan\alpha \rho_{air}}}}}}}
\]

where the second term of the denominator is generally much smaller than \( x' \), the first term.

The extent to which the above is of the desired form, \( \frac{u_{g1}}{u_n} \ll \frac{1}{x'} \), may now be considered. Assuming \( \alpha \) to remain constant, this is seen to depend on the characteristics of \( G, u_n \) and \( z \). For a given propellant at constant temperature, \( G \) may be considered to be constant, as previously stated, although in Fig. 6.20, it was seen to vary slightly with the diameter ratio of the second to first orifice. However, as \( G \) appears as the half power, this slight variation will be ignored for the present purpose. Consider now the "exit" velocity, \( u_n \). This has been
shown, both theoretically and experimentally, to be constant and approximately equal to 29 m/sec. for nozzle assemblies with orifices of nominally equal diameters. Also for nominally equal diameters, $z$ was shown to be constant; its value was found to be 30 mm. Therefore the above inverse proportionality between $u_{ga}/u_n$ and $x'$ can be considered applicable to twin-orifice nozzles assemblies at a fixed ambient temperature and discharging a given propellant.

The applicability of the inverse proportionality to nozzle assemblies with dissimilar orifice diameters may now be considered. In this case theory predicts that the exit velocity ($u_n$) will not be equal to the above 29 m/sec; also, $z$, the distance upstream of the apparent spray origin does not remain constant. Consider firstly variation in $u_n$. Reference to Table 6.2 and Fig. 6.11 shows as previously discussed, that the predicted velocity, assuming choking flow, varies over the range of 25 m/sec to about 47 m/sec; this applies to the range of orifice diameter ratios ($D_2/D_1$) from about 0.5 to 2.0. The data, however, although not extensive, shows that this range may be narrower than indicated above. Therefore, as the half power, of $u_n$ appears in the equation, theory predicts that the assumption of a constant $u_n$ for the above range of orifice diameters would lead to an error of about 20% in the determination of $u_{ga}/u_n$. Empirically this error could be considerably less if the limited variation of exit velocity suggested by the exit velocity data were substantiated. Even so an error of this magnitude would seem excessive. Consider now the variation in the position of the apparent spray origin, $z$. This total variation was shown to be as great as 30 mm, and could therefore be large in comparison with downstream distances of up to 300 mm, say.

There seems to be justification therefore for considering that the equation for the longitudinal axial velocity of the gaseous component of a spray:

$$u_{ga}/u_n = \sqrt{\frac{G}{2 u_n \tan^2 \alpha \rho_{air} \frac{G}{x'} + .26 \sqrt{2 u_n \tan^2 \alpha \rho_{air}}}}$$
is applicable to all the nozzles tested in this work. For the special case of nominally twin orifice nozzles, however, the equation may be simplified by inserting experimentally determined value for $G$, $u_n$, $\rho_{air}$ and $\tan^2\alpha$ as discussed previously. The equation then becomes:

$$\frac{u_{ga}}{u_n} = \frac{111}{x' + 28.8}$$

where $x'$ is generally much greater than 28.8.

These equations may now be compared with experimental data. To do this, longitudinal profiles of the longitudinal velocity of the gaseous component of the spray were obtained for a number of nozzles. They were obtained in a similar manner to the transverse profiles described in sub-section 6.7. Mention may be made, however of the determination of the exit velocity, $u_n$, a parameter not required for 6.7. This was obtained from velocity data taken at the exit of the nozzle housing. These data are given in reduced form in Appendix 1.4. The exit velocity was obtained by taking the mean exit velocity over all the particle sizes for which data was available. Non-availability of data in any particle size range would not invalidate the result because particles of all sizes have been shown to emerge with approximately the same mean velocity. In cases where velocity data at the exit of the nozzle housing were unavailable, either because particle image pairs were not identifiable or otherwise, then $u_n$ was obtained from velocity data obtained 5mm from the nozzle housing or at 10mm in the case of a large diameter orifice because in this case velocity decay over 10mm is small. Exit velocities thus determined are given overleaf. The longitudinal velocity profiles referred to above are plotted together with the theoretical values in Fig. 6.15 (a) and (b). As the orifice diameter of each nozzle assembly were approximately equal it was found that the simplified equation :-

$$\frac{u_{ga}}{u_n} = \frac{111}{x' + 28.8}$$

only, adequately provided theoretical values.
### EXIT VELOCITY ($u_n$)

<table>
<thead>
<tr>
<th>NOZZLE</th>
<th>MEAN m/sec</th>
<th>STANDARD DEVIATION m/sec</th>
<th>STANDARD ERROR OF MEAN m/sec</th>
</tr>
</thead>
<tbody>
<tr>
<td>320 x 350</td>
<td>28</td>
<td>4</td>
<td>1.8</td>
</tr>
</tbody>
</table>
|               |            |                          | **Taken from velocity data at**
|               |            |                          | the nozzle exit and 5mm
downstream.                  |
| 450 x 450     | 29.5       | 1                        | .7                           |
|               |            |                          | **Taken from the velocity data at**
|               |            |                          | the nozzle exit and 5mm
downstream.                  |
| 320 x 700     | 30         |                          | **Taken from the velocity data at**
| (estimated-too few data for reliable assessment) | | | 0mm and 5mm downstream. |
| Nozzle II     | 28         | 1                        | .5                           |
|               |            |                          | **Taken from the maximum velocity**
|               |            |                          | obtained at 10mm downstream. The
|               |            |                          | standard deviation and standard error
|               |            |                          | of the mean are probably underestimated. |
| Nozzle III    | 27.3       | 2.5                      | 1.5                          |
|               |            |                          | **Taken from velocity data at**
|               |            |                          | 5mm downstream.            |
| METERED SPRAY | 20         | -                        | -                            |
| 500 x 200     |            |                          | **Taken from the velocity of**
|               |            |                          | large particles at 10mm.   |

**TABLE 6.4**

**DETERMINATION OF THE EXIT VELOCITY OF THE PROPELLANT FROM SEVERAL NOZZLES.**
FIG. 6.15a LONGITUDINAL PROFILE OF LONGITUDINAL VELOCITY.
FIG. 6.15b. LONGITUDINAL PROFILE OF LONGITUDINAL VELOCITY.

RATIO: EXIT VELOCITY / AXIAL VELOCITY.

DIMENSIONLESS DISTANCE FROM THE NOZZLE \( \left( \frac{x+z}{\sqrt{D_1 D_2}} \right) \)

KEY:
AS FOR FIG. 6.15a.
Also plotted for comparison purposes is the profile of an air jet and that from a metered spray; these are discussed later.

It is seen that generally the data are scattered about the theoretical plot, although the siting of the two extreme experimental points at large values of \( x' \), \((x + z) / \sqrt{D_1 D_2} \), suggest that the theory is predicting too high a value. The statistical significance of the difference between the theoretical curve and these two data points will now be considered. Had each data point been obtained from a large number of photographs then the probability that the difference between data and theory was not a chance occurrence could have been calculated conventionally. This would be done by first calculating the standard error of the mean. From this, a 95% confidence interval would be determined using the Normal distribution. If the theoretical curve were then to lie outside this confidence limit, the difference could have occurred by chance only once in twenty times. Therefore the difference would be considered to be "probably significant" in the statistical sense. However, because of the large amount of analysis required for each experimental point, the number of double-flash photographs taken for each of these two points was reduced to two as seen in Appendix 1.4. The same statistical procedure as that suggested above may, of course, still be used. However, because of the small sample of photographs, the corresponding confidence interval, generated from the standard error of the mean and the "t" distribution (instead of the Normal distribution), would be very large. Thus a modest, but otherwise significant difference between theory and data would be shown to be insignificant. However a second approach is possible. This is to invoke the known statistical properties of the turbulence within the spray. In sub-section 6.7, these were shown to be similar to those of an air jet - in particular the relative turbulence of both air jets and sprays was shown to be about 0.2, for axial positions. Therefore the
standard deviation associated with a theoretical point is:

\[(0.2) \text{ (predicted velocity ratio)}\].

If therefore any individual velocity measurement, of the four which make up these two data points, were to lie beyond 2.0 standard deviations from the theoretical point, the probability of a chance occurrence is 5% and the difference is, again "probably significant".

Although individual velocity measurement are not given in Fig. 6.15, it is seen that, in both cases, the mean value lies within ±2.0 standard deviations of the theoretical curve. Further, reference to the original data revealed that two of the four data points lie well within the range of ±2.0 standard deviations while the other two lie just within this range. This tendency is reflected in the values of the standard deviation given for each data point. It seems therefore that the theory could be predicting too high a value, but this difference can not be shown to be statistically significant.

The point representing nozzle 320 x 350 at \(x' = 330 \text{ (or } x = 80 \text{mm)}\), \(\frac{u_a}{u_0} = 0.5\) is considered to be anomalous. The value of 0.5 (from App. 1.4) for \(\frac{u_a}{u_0}\) was a mean of 0.67 and 0.36. As the difference between these two values is about four times greater than the expected standard deviation, the higher figure was ignored and the lower figure only plotted.

It is concluded tentatively therefore, in the absence of further data, the equation 4.1.10 predicts reasonably well the axial longitudinal velocity profile within a continuous spray, beyond a few centimeters from the nozzle.
Mention may now be made of the data from a metered spray. This was included in order that the applicability of equation 4.1.10 to such sprays may be tested. The data were obtained from the metering device, described in Section 2, which discharged 50 mm$^3$ of propellant through a plunger orifice of 0.50 mm diameter and an adapter orifice of 0.25 mm diameter. The spray was directed through the sample volume of the double-flash unit, which was triggered from a thermocouple placed just beyond the sample volume. Thus photographs were automatically taken during the discharge of the spray, but at an undefined part of that discharge.

The data are again shown in Fig. 6.15. It is seen that the theoretical curve applicable to continuous sprays does not predict velocities of metered sprays. This was to be expected because neither the mass flow-rate nor the exit velocity are equal to those of a continuous spray passing through a similar pair of orifices. However if the appropriate values of these two parameters are inserted into the equation (4.1.10) then a more applicable theoretical curve, may be obtained. The evaluation of the two parameters, mass flow-rate and exit velocity of the metered spray may now be described; they were determined as follows. The mean mass flow-rate was obtained by determining the discharge time of the spray acoustically. This was done by placing a microphone close to the spray. The noise caused by the discharging propellant was then displayed on a storage oscilloscope. The mean mass flow-rate was then:

\[
\text{weight of metered propellant} \quad \text{discharge time}
\]

The weight of the metered propellant was \(0.07 \times 10^{-3}\) kg (taking data from Appendix 1.4) and the mean discharge time was \(0.66 \pm 0.1\) sec. Therefore the mean mass flow-rate was \(0.11 \times 10^{-3}\) kg/sec. This is seen to be considerably lower than the value of approximately \(0.83 \times 10^{-3}\) kg/sec.
determined from sub-section 6.4, for a continuous spray through a similar two-orifice nozzle. This overall mean discharge rate of $0.11 \times 10^{-3}$ kg/sec is perhaps an unsuitable mean to use in this instance. This is because, the majority of the propellant would be discharged at a higher rate than the overall mean. This follows from the shape of the expansion chamber pressure profile given in Section 2. It is expected therefore that a better estimate of discharge rate is perhaps somewhat higher than that given above; this is considered later. The value of the exit velocity $u_0$ may now be considered. This was obtained from Appendix 1.4 and was taken as 20 m/sec, the velocity of large particles close to the nozzle; this may be compared with about 29 m/sec, the exit velocity of continuous sprays.

Assuming a constant spray angle, the theory applicable to continuous sprays may now be modified for this 0.50 x 0.25 diameter orifice, metering device by multiplication by the factor:-

$$\sqrt{\frac{\text{Mass flow-rate for metered spray}}{\text{Mass flow-rate for continuous spray}}} \times \frac{\text{Exit velocity for cont. spray}}{\text{Exit velocity for metered spray}}$$

$$\approx \sqrt{\frac{0.11}{0.85}} \times \frac{29}{20} = 0.44$$

Multiplication of the theoretical curve relating to a continuous spray by 0.44 produces a curve shown as a dotted line in Fig.6.15. This curve is lower than, but more closely related to the velocity data from a metered spray, beyond a few centimetres from the nozzle. A better correlation for this data would be obtained if a higher value for the mean mass flow-rate were used, as discussed earlier. A more precise comparison however would require information on the instantaneous values of mass flow-rate and exit velocity of the spray. It does seem however
that this theoretical approach could yield meaningful predictions, given expressions for the above two parameters.

It is of interest to compare the continuous and metered spray data with that of an air jet, also shown in Fig. 6.15. It is seen that for given nozzle configuration, velocity decay is very rapid compared with either of the liquid-spray decay rates. In principle this suggests that a powder aerosol of lower velocity, would be more readily obtained if the powder is dispersed by an air or propellant vapour jet, of similar initial velocity, rather than by atomization of a liquid.
6.10 **Measurement of the variation of particle velocity within a spray with particle size, and comparison with standard theory.**

In the last sub-section an expression for the longitudinal velocity profile of the gaseous component of the spray was given. However the pharmaceutical properties of the spray are determined by the behaviour of its liquid or solid component. It is therefore desirable to predict also the velocity profiles of particles within the spray.

Figs. 6.16 (a) and (b) show longitudinal velocity profiles plotted from the data given in Appendix 1.5. For each nozzle two profiles are plotted, one for the velocity of the gaseous component and the other for particles whose instantaneous diameter is 30\(\mu\)m to 35\(\mu\)m. It may be noted that this latter profile does not represent the velocity decay of a 30 - 35\(\mu\)m particle because in general the particles are evaporating.

In principle two methods are available for predicting the velocity of a particle at a given position in the spray. Firstly, using an iterative mathematical process, the drag on a particle could be calculated and the total reduction in velocity along its trajectory may be found. This method suffers the disadvantage that the variation in particle diameter, due to evaporation and possibly coagulation, is generally unknown, and consequently large errors would appear in the results of the calculation. This method will therefore not be pursued.

A second approach is to consider the situation at a given point in the spray. In this case the drag on the particle created by the relative velocity between the particle and the spray, may be equated to the mass of the particle multiplied by its deceleration.

Therefore:

\[
\text{Drag} = m_p \cdot u_p \cdot \left( \frac{du_p}{dx} \right)
\]

where \(m_p\) is the mass of the particle, \(u_p\) and \(\frac{du_p}{dx}\) are the
FIG. 6.16a. AXIAL VELOCITY PROFILE.

KEY.

NOZZLE:

COMPONENT.

○ 320 x 350 GAS.
○ 320 x 350 35µ PARTICLE.
△ 450 x 450 GAS.
△ 450 x 450 35µ PARTICLE.

AXIAL VELOCITY, m sec⁻¹

DISTANCE FROM NOZZLE mm.
FIG. 6.16 b. AXIAL VELOCITY PROFILE.

KEY.

NOZZLE COMPONENT.

▼ III 370 x 500 GAS.
▼ III 370 x 500 35µ PARTICLE.
♦ II 690 x 700 GAS.
♦ II 690 x 700 35µ PARTICLE.
× METERED 500 x 250 GAS.
× METERED 500 x 250 35µ PARTICLE.

AXIAL VELOCITY, m sec⁻¹

DISTANCE FROM NOZZLE mm.
particle velocity and its gradient respectively.

This equation follows from the general equation of particle motion in a turbulent flow. This is given by Soo (60), quoting a number of authors, as:

$$\frac{\pi}{6} \frac{d}{d\tau} \frac{3}{\rho} \left( \frac{du}{d\tau} \right) =$$

$$3 \pi \eta \frac{d}{d\tau} \rho u_{rel} - \frac{\pi}{6} \frac{d}{d\tau} \rho \left( \frac{du_{rel}}{d\tau} \right) + \text{"Basset integral"}$$

The term on the left of the above equation represents the inertia force, (mass × acceleration). The terms on the right are:

1. The drag force, given in this case by the Stokes equation.
2. The force due to the pressure gradient.
3. The force required to accelerate the apparent mass of the particle relative to the fluid.
4. The "Basset integral". This constitutes an instantaneous flow resistance, and takes into account the effect of the deviation in the flow pattern from steady state.

It may be shown that these latter terms are negligible if the density of the fluid phase is much less than that of the particle, as is the case with spray particles. Therefore this general equation may be reduced to 6.10.1, as stated previously.

The drag term in equation 6.10.1 may be given by the Stokes equation when the particles are small. This is because the relative velocities of these particles is low, and thus the Reynolds number of the particle is also low. With large particles, whose relative velocities are high the general drag equation must be used. The case of small particles will
be considered first.

In the Stokes region, equation 6.10.1 becomes

$$3\pi\eta d_p u_{rel} = \frac{\pi}{6} d_p^3 \rho_p \cdot \frac{du}{dx}$$

where $\eta$ is the viscosity of the gaseous phase, $u_{rel}$ is the relative velocity between particle and gas, $d_p$ the particle diameter and $\rho_p$ the particle density.

Therefore:

$$u_{rel} = \frac{\frac{\rho_p}{18\eta} d_p^2}{u} \cdot \frac{du}{dx}$$

For small particles, at a given position, $u_p$ and $(du_p/dx)$ are approximately equal to $u_g$ and $(du_g/dx)$ respectively, which are the corresponding values for the gaseous phase. Therefore the slope of the plot of the relative velocity ($u_{rel}$) against the square of the particle diameter ($d_p^2$) should be equal to $\frac{\rho_p u_g}{18} \cdot \frac{du_g}{dx}$ for small particles.

Consider each term of this expression for the slope. The particle density, $\rho_p$, is known. Also $u_g$ and $(du_g/dx)$ can be determined from equation 4.1.10. This was reduced in the previous sub-section to:

$$u_g = u_n \cdot \frac{111}{x' + 29},$$

where $u_n$ is the nozzle exit velocity of the propellant,

and $x' = \frac{x + z}{\sqrt{D_1 D_2}}$,

where $z$ is the distance upstream of the nozzle of the apparent origin of the spray and $D_1$, $D_2$ are the diameters of the orifices of the nozzle assembly.

Therefore:

$$\frac{du}{dx} = -u_n \cdot \frac{111}{(x' + 29)^2} \cdot \frac{1}{\sqrt{D_1 D_2}},$$

It remains for $\eta$, the value of the viscosity of the gaseous phase of the spray, to be determined. The use of a semi-empirical expression by Wilke (108) reveals that for sprays of this nature, beyond a few
centimeters from the nozzle, the viscosity of the vapour phase will approximate to that of air. This is because the mole fraction of the propellant relative to entrained air is very low at this distance. For example at a distance of 80mm from nozzle 450 x 450, the viscosity of the gaseous phase (at 20° C) is calculated to be 1.76 \cdot 10^{-5} \text{ kg/m/sec}, while that of air is 1.8 \cdot 10^{-5} \text{ kg/m/sec}.

Therefore 6.10.3 becomes:

\[
\frac{d^2}{x' + 29} \approx \frac{1.2 \cdot 10^4}{(x' + 29)^3 \sqrt{D_1 D_2}} \ldots 6.10.4
\]

which is applicable for small particles.

The following numerical values may be inserted:

\[
\begin{align*}
\rho_p &= 1.4 \cdot 10^3 \text{ kg/m}^3 \\
\eta &= 1.8 \cdot 10^5 \text{ kg/m/sec} \\
\nu_\infty &= 29 \text{ m/sec}.
\end{align*}
\]

Then

\[
\frac{d^2}{x' + 29} \approx \frac{0.44 \cdot 10^{14} \text{ m/sec}}{1.4 \cdot 10^3 \text{ kg/m}^3} \ldots 6.10.5
\]

This equation may be compared with relative velocity data given in Appendix 1,4. This comparison is made in Fig. 6.17 (a) and (b) for a number of sprays at distances of 40mm and 80mm and 160mm from the nozzle. It is seen that the above equation, 6.10.5, generally represents the proportionality between relative velocity and the square of the particle diameter, for particle diameters up to 30 to 40 \mu m, the maximum size generally observable. It is seen however that the slope of the theoretical curve may be in error by as much as 50% in some cases. However although the error in the prediction of relative velocity of the particles may be high, in most cases the errors in the prediction of the absolute velocity
FIG. 6.17a. PREDICTION OF THE RELATIVE VELOCITY BETWEEN PARTICLE AND GASEOUS COMPONENT OF A SPRAY.
FIG. 6.17b. PREDICTION OF THE RELATIVE VELOCITY BETWEEN PARTICLE AND GASEOUS COMPONENT OF A SPRAY.
will be less, providing of course that the predicting equation 4.1.10 for the velocity of the gaseous component is accurate.

Examining Fig. 6.17 in more detail, it is seen that the agreement between theory and data is best at the 80mm range (Fig. 6.17 (a)). At 160mm and 120mm (Fig. 6.17 (b)) relative velocity measurements tend to be unreliable because the differences in mean velocity between gas and particle are generally less than the level of turbulence. Therefore many more double-flash photographs than those taken would be necessary in order to achieve the required accuracy. At 40mm (Fig. 6.17 (b)) data for the three smaller nozzles, 320 x 350, III and 450 x 450 correlate reasonably well with the theoretical curve, but the data for the large nozzle, III, (not plotted) show no correlation. This is because the equation predicts relative velocities which would require particles above about 12μm travelling at a velocity greater than the exit velocity. Thus for large nozzles, of diameter about 0.7mm, the equation, 6.10.5, is applicable for distances no closer to the nozzle than about 80mm. For smaller nozzles, up to about 0.5mm diameter, this limit is about 40mm.

The limitations of the above equation with respect to nozzle dimensions and distance from the nozzle have been considered. Limitation regarding particle size may now be considered. It was stated earlier that this equation was limited to small particles. The upper limit of the particle size for which the equation is applicable will now be considered. There are two factors restricting the upper range of particle size. The first is that as the size increases, the relative velocity and hence the Reynolds number of the particle increases. Consequently the Stokes equation no longer gives the drag force accurately and the latter would be seriously underestimated. The second factor is that as the relative velocity increased then the approximation :-

\[ u_p \left( \frac{du}{dx} \right) \neq u_g \left( \frac{du}{dx} \right) \]
loses its validity.

Consider first the effect of an increasing Reynolds number of the drag force. Stokes equation is applicable for a particle Reynolds number of less than 0.1, or if a 10% error can be tolerated, less than about 1. Therefore:

\[ \frac{u_{rel}}{d_p} \frac{d}{ho} \frac{g}{\eta} < 1. \]

Incorporating again the assumption that the gaseous component of the spray is mainly air, then:

\[ u_{rel} \frac{d}{p} < \left( \frac{\eta}{\rho} \right)_{air}. \]

or \[ u_{rel} \frac{d}{p} < \frac{1.8 \times 10^{-5}}{1.2} = 1.5 \times 10^{-5} \text{ m}^2/\text{sec} \]

Thus, the maximum values of the relative velocity for an error in the drag equation of no greater than 10% are approximately:

- 0.3 m/sec for a 50 \( \mu \text{m} \) particle
- 0.75 m/sec for a 20 \( \mu \text{m} \) particle
- 1.5 m/sec for a 10 \( \mu \text{m} \) particle

It is seen, from Fig. 17 that for particles above about 15 to 20 \( \mu \text{m} \), the above criterion is exceeded, except at a distance beyond about 160 mm from the nozzle. This is considered later.

Consider the second factor limiting the validity of equation 6.10.5. This was that \( u_p (du_p/dx) \) was no longer approximately equal to \( u_g (du_g/dx) \).

Reference to Fig. 6.16, and Fig. 6.16 (a) especially, reveals that for 35 \( \mu \text{m} \) particles, for example, the slope \( (du_p/dx) \) is approximately equal to or only slightly greater than that of the gas, beyond about 50 mm from the nozzle but that the particle velocity, \( u_p \), may be of the order of 50% greater than that of the gas, \( u_g \). Thus 35 \( \mu \text{m} \) particles may give rise to errors of the order of 50%. Consequently the maximum size of particle for which the error would be again 10%, say, will be similar to that
determined from the Reynolds number criterion.

An explanation is required, therefore, as to why the relationship between $u_{rel}$ and $d_p^2$ remains linear at particle diameters greater than the limit of 15 - 20 μm determined above.

Reference to equation 6.10.3:–

$$u_{rel} = \frac{d^2_p}{\rho_p} \frac{1}{\eta} u_p \left( \frac{du_p}{dx} \right),$$

offers some explanation as to why the linear relationship is maintained.

For large particles, both the acceleration term $u_p \left( \frac{du_p}{dx} \right)$ and the drag component increase simultaneously (but not necessarily proportionately).

As these two terms appear as numerator and denominator respectively in the equation, it is possible that linearity could be maintained because, over a limited range of particle sizes, their ratio could remain approximately constant.

An example may be given. Consider a 35 μm particle at 80 mm from nozzle 450 x 450. From Fig.6.16 (a) the deceleration terms for particle and vapour are:

$$u_p \left( \frac{du_p}{dx} \right) = -(17 \text{ m/sec}) (150 \text{ /sec}) = -2500 \text{ m/sec}^2,$$

and

$$u_g \left( \frac{du_g}{dx} \right) = -(9 \text{ m/sec}) (130 \text{ /sec}) = -1170 \text{ m/sec}^2.$$  

Therefore the acceleration term is greater than that given by the approximation, $u_g \left( \frac{du_g}{dx} \right)$, by a factor of $\frac{-2500}{-1170}$ or 2.1.

Now $u_{rel} = 8 \text{ m/sec}$. Therefore the particle Reynolds number, again assuming that the spray consists mainly of entrained air, is approximately:

$$\frac{u_{rel}}{d_p} \frac{\rho_{air}}{\eta_{air}} \approx \frac{1}{18}.$$
Reference to the Standard Drag curve (60), assuming this to be applicable, reveals that the drag is greater than that given by Stokes equation by a factor of 1.8. This may be compared with the factor of 2.1 for the acceleration ratio, previously discussed. Therefore the constant of proportionality, between the relative velocity and the square of the particle diameter for 35 μm particles increases by only \( \frac{2.1}{1.8} \) or 17%. Thus in this case the two errors — although large — compensate to a considerable extent.

It is therefore concluded that the velocity of particles relative to the gaseous phase within a spray may be determined approximately using equation 6.10.2 for particles of diameters up to about 10 μm. Further, for the range of nozzles tested in this work, there is empirical evidence that this upper size limit may be extended to about 30 or 40 μm. This equation applies to distances up to about 40mm from the nozzle, for small nozzles of less than about 0.5mm diameter; for nozzles of diameter about 0.7mm the equation is valid up to about 80mm.
6.11 Determination of the size distribution of the resultant surfactant aerosols and their variation with particle size.

The size distribution of the active ingredient of a spray is of general interest in aerosol technology. In the case of inhalation therapy this size distribution among other things determines the effectiveness of penetration of the aerosol into the lungs. In this context the most important parameter regarding the size distribution is then the fraction by volume between a certain size range; this size range being that which reaches, and is retained in, the lower recesses of the lungs. Although it is by no means certain what the precise limits of this range are, there appears to be general agreement that particles above about 5 μm diameter will not reach the alveoli, and those of about 0.5 μm in diameter will not be retained. Thus the relevant diameter range is about 0.5 to 5 μm. In the case of aerosols generated by atomisation of liquids, and by most other methods, this lower limit is of little importance because the fraction by volume below this diameter is usually negligible. Thus the fraction between 0.5 and 5 μm may be obtained approximately by measuring the fraction below 5 μm.

Size analyses were obtained of the resultant surfactant aerosol generated by the atomisation of a solution of propellant and 1% by weight of the surfactant "span" (or "arlasel") 85. The propellant again consisted of 60/40% by weight of propellant 12 and 11 respectively. The concentration of surfactant, 1% by weight, is typical of that used in commercial practice.

The size analyses are shown in Figs. 18(a) and (b), as fraction by volume below a given particle diameter against that diameter. The original data are given in Appendix 1.5. Table 6.5 shows mass median diameter (MMD), volume-surface mean or Sauter mean diameter (SMD) and the fraction by volume below 5 μm particle diameter.
FIG. 6.18(a) SIZE DISTRIBUTION OF THE RESULTANT SURFACTANT AEROSOL FROM VARIOUS NOZZLES.
FIG. 6.18(b) SIZE DISTRIBUTION OF THE RESULTANT SURFACTANT AEROSOL FROM VARIOUS NOZZLES.
<table>
<thead>
<tr>
<th>NOZZLE</th>
<th>VOLUME-SURFACE MEAN DIAMETER $\mu m$</th>
<th>MASS MEDIAN DIAMETER $\mu m$</th>
<th>PERCENT BY VOLUME BELOW $5 \mu m$</th>
</tr>
</thead>
<tbody>
<tr>
<td>IV (250 x 380)</td>
<td>5.3</td>
<td>6.8</td>
<td>33%</td>
</tr>
<tr>
<td>320 x 350</td>
<td>6.0</td>
<td>7.4</td>
<td>24</td>
</tr>
<tr>
<td>320 x 450</td>
<td>5.2</td>
<td>6.3</td>
<td>35</td>
</tr>
<tr>
<td>450 x 450</td>
<td>5.7</td>
<td>7.0</td>
<td>28</td>
</tr>
<tr>
<td>Run 1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>450 x 450</td>
<td>5.3</td>
<td>6.6</td>
<td>32</td>
</tr>
<tr>
<td>Run 2</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>640 x 450</td>
<td>6.2</td>
<td>9.2</td>
<td>16</td>
</tr>
<tr>
<td>640 x 350</td>
<td>Very large. Not properly atomized.</td>
<td></td>
<td></td>
</tr>
<tr>
<td>II (690 x 700)</td>
<td>9.4</td>
<td>12</td>
<td>8.0</td>
</tr>
<tr>
<td>III (380 x 500)</td>
<td>7.3</td>
<td>8.9</td>
<td>16</td>
</tr>
<tr>
<td>Metered Spray (500 x 250)</td>
<td>3.9</td>
<td>4.8</td>
<td>55</td>
</tr>
</tbody>
</table>

**TABLE 6.5** SIZE ANALYSES OF THE SOLUTE AEROSOL
In Fig. 19 the mass median diameter is plotted against the diameter of the downstream nozzle. It is seen that for nozzles whose orifices are of similar diameter, the relationship between mass median diameter and orifice diameter is tolerably linear. The slope is \(1.6 \times 10^{-2}\).

Therefore:

\[
\text{Mass median diameter} = 1.6 \times 10^{-2} \times \text{(orifice diameter)}.
\]

Similarly:

\[
\text{Volume surface mean diameter} = 1.2 \times 10^{-2} \times \text{(orifice diameter)}
\]

Also the fraction by volume below 5 \(\mu\)m particle diameter is given, very roughly, by:

\[
\text{Fraction below 5 } \mu\text{m} = 0.17 \left(1 - \frac{D}{d}\right)
\]

where \(D\) is orifice diameter in mm.

For nozzles whose diameters are not equal, two cases may be considered. Firstly, when the upstream orifice is smaller than the downstream, there is some evidence to suggest that atomization is marginally improved. This is to be expected because as this first orifice becomes smaller, the temperature fall across it rises and the mass fraction of propellant evaporated increases. Thus a propellant of higher gas-to-vapour ratio is presented to the final, downstream, orifice.

When the upstream orifice is larger than the downstream, atomization efficiency deteriorates rapidly. Thus, compared with a mass median diameter of 7.0 \(\mu\)m for the 450 x 450 nozzle, that of the 640 x 450 nozzle was 9.2 \(\mu\)m, while the 640 x 350 nozzle failed to atomize properly, producing pools of surfactant on the microscope slides. Indeed during one run in the determination of mass flow-rate, this nozzle produced a stream of unatomized propellant. From consideration of the geometrical configuration of the nozzle this is perhaps to be expected because the two orifices are in line and it is possible therefore, as with any of these nozzles, for propellant to pass
FIG. 6.19. VARIATION OF AEROSOL SIZE DISTRIBUTION WITH NOZZLE DIAMETER.
directly in liquid form from one orifice to the other. In the case when the first orifice is larger than the second however, this tendency is considerably enhanced because the required temperature fall across this first orifice, and hence propellant evaporation, are substantially reduced. This explanation is supported by data from the metered spray. This has the same ratio of upstream to downstream orifice diameter but produces a very fine spray. Further the parameters of the size distribution of this spray correlate well with those of the continuous sprays whose orifices are of similar diameter.
6.12 Metastability

It was stated in Section 4 that the propellant in the expansion chamber was in a metastable state, at a pressure which was generally lower than the S.V.P. corresponding to its temperature. An expression for the degree of metastability was determined empirically. Defining the degree of metastability, $\Delta P_{\text{meta}}$, as the difference between the S.V.P. of propellant at expansion chamber temperature minus the expansion chamber pressure, or

$$\Delta P_{\text{meta}} = \text{S.V.P}_{T_e} - P_e$$

then the empirical expression

$$\Delta P_{\text{meta}} = 1.80 \times 10^3 \left( \frac{D_2}{D_1} \right)^{2.5} \left( \text{S.V.P}_{T_e} \right)$$

This is shown in Fig. 6.20, together with experimental values of $\Delta P_{\text{meta}}$. These were obtained from data given in Appendix 1.3.

In using this equation the value of the expansion chamber temperature $T_e$, with which to determine the S.V.P., may be obtained approximately from a simplified relationship for the temperature fall across the first orifice; this is given in sub-section 6.3.
S.V.P OF PROPELLANT - EXPANSION CHAMBER
PRESSURE, KNm²

FIG. 6. MEASUREMENT OF THE PROPELLANT IN THE EXPANSION CHAMBER.

[DOWNSTREAM ORIFICE] 4

DI = 750 µm

DI = 450 µm

DI = 250 µm

20 × 10⁻¹² m⁴

EMPIRICAL EXPRESSION FOR: DI = 320 µm.

KEY:

□ △ ○ •

DI

250 µm

320 µm

450 µm

700 µm
CONCLUSIONS AND RECOMMENDATIONS FOR FURTHER WORK

7.1 Conclusions

A theoretical expression has been produced by means of which it is possible to predict the axial velocity of the gaseous component of a continuous spray generated by passing a liquified-gas propellant through a two-orifice nozzle assembly. From this expression values for off-axis velocities may be determined by assuming a Gaussian transverse distribution of velocity. Also an approximate value of the velocity of the particulate component of the spray may be obtained from a relationship determined semi-empirically, between particle diameter and the relative velocity between particle and gaseous component.

The primary expression, referred to above, requires three independently determined empirical factors. They are the position of the apparent origin of the spray, the angle of divergence of the spray and the degree of metastability of the propellant in the expansion chamber of the nozzle assembly. The first of these, the position of the apparent spray-origin, was determined photographically. It is taken as 30mm upstream of the nozzle for twin-orifice nozzles; its value for other nozzle assemblies is discussed in Section 6. The divergence angle was shown to be approximately equal to that of an air jet. Finally the degree of metastability is given by a simple expression in terms of the orifice dimensions and the S.V.P. of the propellant; generally metastability appears as a small correcting term.

The development of the predicting equation for axial velocity required the following subsidiary expressions. They were expressions for the mass flow-rate of propellant through the upstream and downstream orifices (short tubes), the mass flow-rate through a two-orifice nozzle assembly, the temperature and hence the quality of propellant in the expansion chamber, and also the exit velocity of the spray. The expressions
for the mass flow-rate through the downstream orifice and exit velocity, also from the downstream orifice, were based on the assumption of mass-limiting flow.

Experimental support for the primary expression was given for three nominally twin orifice nozzles of orifice diameter 0.33mm, 0.45mm and 0.60mm. Experimental support for the subsidiary expressions was given for orifice diameters ranging from 0.25mm to 0.70mm and orifice diameter ratios from about 0.5 to 2.0, although exit velocity data were available for only part of this range.

A feature of the expressions developed was that without undue loss of accuracy, they could generally be reduced to simple functions of orifice diameters $D_1$, $D_2$; thus trends are readily predictable. They are applicable for a propellant, at 20°C, consisting of, by weight, 60% propellant 12, 40% propellant 11, with a solute "Span 85", 1% by weight. These simplified expressions are listed below

1. Mass flow-rate ($\dot{m}$) $= 6.6 \times 10^3 \ D_1 D_2 \ \text{kg/sec}$

2. Temperature fall across the upstream orifice ($\Delta T_1$)
   $$= 3.0 \ \frac{D_2^2}{D_1} \ \text{deg C}.$$  

3. Quality of propellant (mass fraction evaporated) in the expansion chamber ($q$)
   $$= 0.0061 \ \Delta T_1 = 0.018 \ \frac{D_2^2}{D_1}.$$  

4. Mean velocity of gaseous component of the spray ($u_{ga}$)
   $$= \frac{1}{(x + z) \ \tan \alpha} \sqrt{\frac{2}{\pi}} \ \frac{j}{\rho_{air}} \ e^{-y^2/x^2 \tan^2 \alpha}.$$
Here $J$ is the momentum efflux whose determination was discussed in Section 4. Also, $x$ and $y$ are downstream and off-axis distances respectively, and $\alpha$ is the $1/e$-velocity half-angle.

5. Axial particle velocity is given by:

$$u_a (1 + \text{function}(d_p^2))$$

where \(\text{function}(d_p^2)\) was discussed in Section 6.

6. A further, empirical, expression was derived for the mass median diameter (MMD) of the spray of residual solute, "Span 85", generated by nominally twin orifice nozzles. This was:

$$\text{MMD} = 1.6 \times 10^{-2}D$$

where $D$ is the orifice diameter. Distributions from some dissimilar orifice nozzles were given in Section 6.

The above equations, and their more precise forms given in Section 6, generally enable spray properties to be determined in terms of fundamental parameters. They also are of assistance in the design of inhalation devices. In this respect it is clear from the above, that in order to produce a spray suitable for inhalation therapy the orifice diameters should be as small as possible. This has the dual advantage that both spray size and velocity are minimised. It could possibly be advantageous, however, to use a nozzle assembly whose ratio, $D_2:D_1$, was greater than 1; this would improve the quality of the propellant in the expansion chamber. However limited evidence available suggests that, with respect to particle size, the diameter of the downstream orifice is the overriding factor when this orifice is the larger. Thus it is considered that both diameters should be reduced to a
size compatible with free flow of propellant suspension through each orifice. Generally therefore this would suggest a nominally twin orifice nozzle assembly.

The remarks above are considered to apply also to metered dose aerosol generators. This is because they have been shown to follow the same trends as the continuous spray devices. An exception however is given by the comparison between the size distributions from metered and continuous sprays when the diameter of the upstream orifice was about twice that of the downstream. The size distribution of the metered spray was appropriate to that of a twin orifice device of orifice diameter equal to that of the smaller downstream orifice, while the continuous spray device failed to atomize properly. This gives an additional design criterion that in-line orifices should be avoided. A further point regarding nozzle design for metered sprays is that the metering device should be of simple design. This is to avoid two-part spray emission, observed on high speed cine films during preliminary experimentation. In this, a small but significant proportion of the spray was emitted under low pressure, poorly atomized, after the main discharge.

The above has considered nozzle design criteria related to variables investigated experimentally in this work. Other variables may now be considered. An increase in the saturated vapour pressure of the propellant has been shown to produce a considerably finer spray. Polli (51) has shown that roughly doubling the gauge pressure from 210 kN/m² to 530 kN/m² more than halves the mass median diameter which was reduced from 3.2 to 1.3 μm.

A similar trend was observed by Lefebvre and Tregan (38). Increasing the propellant pressure will increase the momentum of the spray. However the momentum and hence the spray velocity are roughly proportional to the square root of propellant gauge pressure. Further, as deposition is proportional to the square of particle diameter and first power only of the velocity, there would appear therefore to be a nett advantage in increasing the pressure.
Spray angle may next be considered. Generally the greater the divergence angle of the spray the greater is the attenuation of spray velocity. Also, the greater is entrainment of air which would promote evaporation of the droplets. Thus it would appear advantageous to increase the spray angle, although operation in the confined region of the mouth would considerably modify the above model.
7.2 FURTHER WORK

This investigation into aerosol generators was necessarily limited, and the need for further investigation of a number of effects has become apparent. The most significant of these are listed below.

1. **Propellant pressure and temperature**

   This work was carried out with one propellant solution at one temperature. It is desirable to determine the effect of both change in propellant pressure and of temperature. Calculations of the thermodynamic properties of the propellant would be simpler if a single component propellant were used.

2. **Coagulation**

   A factor that could significantly affect the size distribution of the residual aerosol is coagulation within the spray. This effect merits investigation.

3. **Propellant evaporation and the concentration of surface active agent**

   The probability of impaction of the spray on the back of the throat is determined by, among other factors, the size distribution of the spray in that region. This is significantly influenced by the evaporation rate of the spray. A high propellant evaporation rate is therefore desirable. In this connection it has been shown by Orr (47), for example, that the presence of a small proportion of surface active material can reduce evaporation rate by many orders of magnitude. Thus the minimum concentration of surface active agent should be used, compatible with efficient drug dispersion and smooth operation of the metering valve. A second obvious disadvantage of excess surfactant is that ultimately the size distribution of the residual aerosol increases with surfactant concentration, as
discussed in Section 5.

4. Critical flow

The theoretical description of the flow through the downstream orifice was based on the assumption of choking flow in this orifice. No direct verification of this assumption was made however. It would be instructive therefore to determine the effect on mass flow-rate of variation of ambient pressure. A more detailed study of the variation of spray velocity in the immediate vicinity of the nozzle would also add to the understanding of choking effects, assuming these to be present.

5. Applicability to metered sprays

The trends found to be applicable to continuous sprays, were found generally to be applicable to metered sprays. However a more detailed description of the metered spray would be desirable. This would necessitate the simultaneous measurement of instantaneous pressure and temperature in the expansion chamber together with that of the spray velocity, at various stages in the metering cycle.

6. Both atomization and metastability of the propellants were treated empirically. A fundamental approach to both would be desirable.

7. The analysis of the double-spark photomicrographs and the microscope analysis of the residual aerosol were direct and were considered to be reliable. However these direct methods were time consuming and tedious. Automatic methods would be desirable. In this connection the use of the "Quantimet" automatic particle size analyser (41) may be considered.
8. The work has dealt with one facet of the study of inhalation therapy. It has shown that in all probability, the main factor limiting the efficiency of liquefied gas aerosol generators is the minimum size of the nozzle diameters. This will be limited by their tendency to clog during operation with powder suspensions. Therefore future work should perhaps be concentrated on this aspect of the study. The possible use of propellant vapour as an alternative means of powder dispersion may profitably be investigated. The advantages in terms of rapid velocity decay have been demonstrated.
### GLUED TWO-ORIFICE NOZZLES

<table>
<thead>
<tr>
<th>NOZZLE</th>
<th>UPSTREAM ORIFICE</th>
<th>DOWNSTREAM ORIFICE</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>DIAMETER (µm)</td>
<td>THICKNESS (µm)</td>
</tr>
<tr>
<td>II</td>
<td>690</td>
<td>610</td>
</tr>
<tr>
<td>III</td>
<td>375</td>
<td>485</td>
</tr>
<tr>
<td>IV</td>
<td>250</td>
<td>390</td>
</tr>
<tr>
<td>V</td>
<td>565</td>
<td>415</td>
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### DISMOUNTABLE ORIFICE DISCS

<table>
<thead>
<tr>
<th>NOMINAL DIAMETER (µm)</th>
<th>POSITION IN NOZZLE ASSEMBLY</th>
<th>UPSTREAM DIAMETER (µm)</th>
<th>DOWNSTREAM DIAMETER (µm)</th>
<th>THICKNESS (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>250</td>
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<td>D</td>
<td>435</td>
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<td>370</td>
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<td>640</td>
<td>U/D</td>
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<td>U</td>
<td>675</td>
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<td>700</td>
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APPENDIX 1.1  
NOZZLE DIMENSIONS
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<tr>
<th>NOZZLE</th>
<th>TEMPERATURE OF PROPELLANT (°C)</th>
<th>MEAN DISCHARGE RATE STANDARD DEVIATION</th>
<th>STANDARD ERROR OF THE MEAN (g/sec.)</th>
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<tbody>
<tr>
<td>250 x 350</td>
<td>20.7</td>
<td>.57</td>
<td>.02</td>
</tr>
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<td>20.9</td>
<td>.77</td>
<td>.00</td>
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<td>20.9</td>
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<td>.02</td>
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<td>320 x 700</td>
<td>20.5</td>
<td>1.31</td>
<td>.05</td>
</tr>
<tr>
<td>450 x 350</td>
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<td>1.17</td>
<td>.04</td>
</tr>
<tr>
<td>450 x 450</td>
<td>20.4</td>
<td>1.42</td>
<td>.02</td>
</tr>
<tr>
<td>450 x 700</td>
<td>20.6</td>
<td>1.88</td>
<td>.02</td>
</tr>
<tr>
<td>680 x 350</td>
<td>20.7</td>
<td>1.56</td>
<td>*</td>
</tr>
<tr>
<td>680 x 450</td>
<td>20.7</td>
<td>2.08</td>
<td>.06</td>
</tr>
<tr>
<td>680 x 700</td>
<td>20.7</td>
<td>2.84</td>
<td>.02</td>
</tr>
</tbody>
</table>

* One reading only. During the second run, the propellant streamed undispersed, from the nozzle.
<table>
<thead>
<tr>
<th>NOZZLE</th>
<th>250 x 350</th>
<th>320 x 350</th>
<th>320 x 450</th>
<th>320 x 700</th>
<th>450 x 450</th>
<th>450 x 700</th>
<th>640 x 350</th>
<th>640 x 450</th>
<th>680 x 700</th>
</tr>
</thead>
<tbody>
<tr>
<td>MEAN UPSTREAM PRESSURE PRIOR TO SPRAYING(\text{KPa})</td>
<td>408</td>
<td>413</td>
<td>414</td>
<td>410</td>
<td>411</td>
<td>408</td>
<td>414</td>
<td>414</td>
<td>403</td>
</tr>
<tr>
<td>MEAN UPSTREAM PRESSURE (RUNNING)(\text{KPa})</td>
<td>407</td>
<td>410</td>
<td>413</td>
<td>404</td>
<td>406</td>
<td>400</td>
<td>407</td>
<td>408</td>
<td>400</td>
</tr>
<tr>
<td>MEAN EXPANSION CHAMBER PRESSURE(\text{KPa})</td>
<td>336</td>
<td>350</td>
<td>324</td>
<td>245</td>
<td>364</td>
<td>303</td>
<td>400</td>
<td>387</td>
<td>359</td>
</tr>
<tr>
<td>MEAN UPSTREAM TEMPERATURE(\text{°C})</td>
<td>19.55</td>
<td>19.7</td>
<td>20.1</td>
<td>20.6</td>
<td>20.0</td>
<td>20.3</td>
<td>20.4</td>
<td>20.1</td>
<td>20.15</td>
</tr>
<tr>
<td>MEAN EXPANSION CHAMBER TEMPERATURE(\text{°C})</td>
<td>14.3</td>
<td>16</td>
<td>14.7</td>
<td>9.6</td>
<td>17.1</td>
<td>14.1</td>
<td>19.8</td>
<td>18.7</td>
<td>17.5</td>
</tr>
<tr>
<td>PRESSURE DIFFERENCE ACROSS UPSTREAM ORIFICE ((\Delta P_1))(\text{KPa})</td>
<td>Mean</td>
<td>7.1</td>
<td>60</td>
<td>88</td>
<td>162</td>
<td>41.2</td>
<td>96</td>
<td>7.6</td>
<td>22</td>
</tr>
<tr>
<td>Standard deviation</td>
<td>.7</td>
<td>10</td>
<td>11</td>
<td>3</td>
<td>2</td>
<td>2</td>
<td>2</td>
<td>.7</td>
<td>3</td>
</tr>
<tr>
<td>Standard error of mean</td>
<td>.4</td>
<td>4</td>
<td>7</td>
<td>2</td>
<td>1.5</td>
<td>1</td>
<td>1</td>
<td>.5</td>
<td>1.5</td>
</tr>
<tr>
<td>TEMPERATURE DIFFERENCE ACROSS UPSTREAM ORIFICE ((\Delta T_1))(\text{°C})</td>
<td>Mean</td>
<td>5.2</td>
<td>3.6</td>
<td>5.3</td>
<td>11</td>
<td>2.9</td>
<td>6.2</td>
<td>0.56</td>
<td>1.3</td>
</tr>
<tr>
<td>Standard deviation</td>
<td>0.1</td>
<td>.25</td>
<td>.1</td>
<td>.1</td>
<td>.1</td>
<td>.1</td>
<td>.1</td>
<td>.1</td>
<td>.25</td>
</tr>
<tr>
<td>Standard error of mean</td>
<td>0.05</td>
<td>.1</td>
<td>.07</td>
<td>.07</td>
<td>.07</td>
<td>.07</td>
<td>.05</td>
<td>.07</td>
<td>.1</td>
</tr>
<tr>
<td>CORRECTED TEMPERATURE DIFFERENCE(\Delta T_1^*)(\text{°C})</td>
<td>5.9</td>
<td>4.2</td>
<td>5.8</td>
<td>10.8</td>
<td>3.4</td>
<td>6.3</td>
<td>0.7</td>
<td>1.6</td>
<td>2.9</td>
</tr>
</tbody>
</table>

APPENDIX 1.3

TEMPERATURE AND PRESSURE MEASUREMENTS OF THE PROPELLANT DURING NOZZLE FLOW
### Nozzle II

<table>
<thead>
<tr>
<th>RANGE - NO. OF FRAMES</th>
<th>MEAN VELOCITY STANDARD DEVIATION IN THE GIVEN PARTICLE SIZE RANGE (μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>&lt;5μm</td>
<td>3</td>
</tr>
<tr>
<td>5mm</td>
<td>3</td>
</tr>
<tr>
<td>10mm</td>
<td>3</td>
</tr>
<tr>
<td>20mm</td>
<td>3</td>
</tr>
<tr>
<td>40mm</td>
<td>3</td>
</tr>
<tr>
<td>80mm</td>
<td>6</td>
</tr>
<tr>
<td>120mm</td>
<td>6</td>
</tr>
<tr>
<td>140mm</td>
<td>3</td>
</tr>
</tbody>
</table>

**Appendix 1.4**

Variation of particle velocity within a spray with the distance from the nozzle.
## APPENDIX 1.4

### VARIATION OF PARTICLE VELOCITY WITHIN A SPRAY

WITH THE DISTANCE FROM THE NOZZLE.
<table>
<thead>
<tr>
<th>RANGE</th>
<th>MEAN VELOCITY STANDARD DEVIATION</th>
<th>STANDARD ERROR OF MEAN (m/sec.)</th>
<th>IN THE GIVEN PARTICLE SIZE RANGE (μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>NO. OF FRAMES</td>
<td>&lt; 5 μm</td>
<td>5 - 10</td>
<td>10 - 15</td>
</tr>
<tr>
<td>0mm</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>3</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>5mm</td>
<td>-</td>
<td>28</td>
<td>27.5</td>
</tr>
<tr>
<td>-</td>
<td>-</td>
<td>4</td>
<td>4</td>
</tr>
<tr>
<td>2</td>
<td>-</td>
<td>3</td>
<td>3</td>
</tr>
<tr>
<td>10mm</td>
<td>-</td>
<td>30</td>
<td>29</td>
</tr>
<tr>
<td>-</td>
<td>-</td>
<td>2</td>
<td>4</td>
</tr>
<tr>
<td>2</td>
<td>-</td>
<td>1.5</td>
<td>3</td>
</tr>
<tr>
<td>20mm</td>
<td>-</td>
<td>23</td>
<td>25</td>
</tr>
<tr>
<td>1</td>
<td>one frame only</td>
<td></td>
<td></td>
</tr>
<tr>
<td>40mm</td>
<td>13.5</td>
<td>14</td>
<td>15</td>
</tr>
<tr>
<td>2</td>
<td>3</td>
<td>2</td>
<td>2</td>
</tr>
<tr>
<td>1.5</td>
<td>2</td>
<td>1.5</td>
<td>1.5</td>
</tr>
<tr>
<td>80mm</td>
<td>14</td>
<td>15</td>
<td>16</td>
</tr>
<tr>
<td>5</td>
<td>5</td>
<td>5</td>
<td>3</td>
</tr>
<tr>
<td>2</td>
<td>3.5</td>
<td>3.5</td>
<td>3.5</td>
</tr>
<tr>
<td>160mm</td>
<td>4.1</td>
<td>4.3</td>
<td>4.2</td>
</tr>
<tr>
<td>1</td>
<td>1</td>
<td>.5</td>
<td>.6</td>
</tr>
<tr>
<td>2</td>
<td>.7</td>
<td>35</td>
<td>.4</td>
</tr>
</tbody>
</table>

APPENDIX 1.4 VARIATION OF PARTICLE VELOCITY WITHIN A SPRAY WITH THE DISTANCE FROM THE NOZZLE.
NOZZLE 320 x 700  (Not plotted in Fig. 6.15)

<table>
<thead>
<tr>
<th>RANGE - NO. OF FRAMES</th>
<th>&lt;5μm</th>
<th>5 - 10</th>
<th>10 - 15</th>
<th>15 - 20</th>
<th>20 - 30</th>
<th>30 - 40</th>
<th>40 - 50</th>
<th>50 - 60</th>
</tr>
</thead>
<tbody>
<tr>
<td>0mm</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>26</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>2</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>2</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>5mm</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>32</td>
<td>-</td>
<td>34.5</td>
<td>-</td>
</tr>
<tr>
<td>2</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>1</td>
<td>-</td>
</tr>
<tr>
<td>10mm</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>29</td>
<td>-</td>
<td>-</td>
<td>30</td>
</tr>
<tr>
<td>2</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>1</td>
</tr>
<tr>
<td>20mm</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>29</td>
<td>-</td>
<td>32</td>
<td>26</td>
</tr>
<tr>
<td>2</td>
<td>-</td>
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<td>-</td>
<td>2</td>
<td>2</td>
<td>-</td>
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<tr>
<td>40mm</td>
<td>-</td>
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<td>27</td>
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<td>30</td>
<td>30</td>
</tr>
<tr>
<td>4</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>4</td>
<td>2</td>
<td>2</td>
<td>-</td>
<td>2.5</td>
</tr>
</tbody>
</table>

APPENDIX 1.4. VARIATION OF PARTICLE VELOCITY WITHIN A SPRAY WITH THE DISTANCE FROM THE NOZZLE.
### APPENDIX 1.4

**VARIATION OF PARTICLE VELOCITY WITHIN A SPRAY**

**WITH THE DISTANCE FROM THE NOZZLE.**

<table>
<thead>
<tr>
<th>RANGE</th>
<th>MEAN VELOCITY</th>
<th>STANDARD DEVIATION</th>
<th>IN THE GIVEN PARTICLE SIZE RANGE (μm)</th>
<th>STANDARD ERROR OF MEAN (m/sec.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>NO. OF FRAMES</td>
<td>&lt; 5μm</td>
<td>5 - 10</td>
<td>10 - 15</td>
<td>15 - 20</td>
</tr>
<tr>
<td>0mm</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>particles identifiable on only one frame</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>30</td>
<td>29.5</td>
<td>29.5</td>
<td>29</td>
</tr>
<tr>
<td></td>
<td>5mm</td>
<td>5</td>
<td>5</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>7</td>
<td>7</td>
<td>1</td>
</tr>
<tr>
<td>10mm</td>
<td>-</td>
<td>20</td>
<td>25</td>
<td>25</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>2</td>
<td>2</td>
<td>1</td>
</tr>
<tr>
<td>20mm</td>
<td>-</td>
<td>20</td>
<td>25</td>
<td>25</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>time interval too large on second frame</td>
<td></td>
<td></td>
</tr>
<tr>
<td>40mm</td>
<td>16</td>
<td>17</td>
<td>18</td>
<td>22</td>
</tr>
<tr>
<td></td>
<td>5</td>
<td>4</td>
<td>5</td>
<td>4</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>3</td>
<td>3</td>
<td>3</td>
</tr>
<tr>
<td>80mm</td>
<td>9</td>
<td>9</td>
<td>10</td>
<td>11</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>7</td>
<td>7</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td>160mm</td>
<td>6.5</td>
<td>7</td>
<td>8</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>7</td>
<td>7</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

*NOZZLE 450 x 450*
Metered Spray

Discharge of 50 mm$^3$ (50 microlitre) of propellant through a metering device (described in Section 2) whose "plunger" orifice diameter = .5mm(20/1000") and "adapter" nozzle = .25mm(10/1000")

<table>
<thead>
<tr>
<th>RANGE</th>
<th>MEAN VELOCITY</th>
<th>STANDARD DEVIATION</th>
<th>STANDARD ERROR OF MEAN (m/sec.)</th>
<th>IN THE GIVEN PARTICLE SIZE RANGE (μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>NO. OF FRAMES</td>
<td>&lt; 5μm</td>
<td>5 - 10</td>
<td>10 - 15</td>
<td>15 - 20</td>
</tr>
<tr>
<td>10mm</td>
<td>-</td>
<td>14</td>
<td>15</td>
<td>17</td>
</tr>
<tr>
<td>3</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>20mm</td>
<td>-</td>
<td>11</td>
<td>12</td>
<td>13</td>
</tr>
<tr>
<td>3</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>40mm</td>
<td>6</td>
<td>6</td>
<td>8</td>
<td>9.5</td>
</tr>
<tr>
<td>3</td>
<td>-</td>
<td>2</td>
<td>2.5</td>
<td>-</td>
</tr>
<tr>
<td>80mm</td>
<td>-</td>
<td>5</td>
<td>7</td>
<td>9</td>
</tr>
<tr>
<td>3</td>
<td>-</td>
<td>-</td>
<td>2</td>
<td>-</td>
</tr>
<tr>
<td>140mm</td>
<td>-</td>
<td>3</td>
<td>9</td>
<td>-</td>
</tr>
<tr>
<td>1</td>
<td>one frame only</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

Appendix 1.4

Variation of Particle Velocity Within a Metered Spray With Distance From the Nozzle
**APPENDIX 1.5**  
**SIZE DISTRIBUTION OF SOLUTE AEROSOL**

**NOZZLE** 320 x 350

<table>
<thead>
<tr>
<th>FLATTENED PARTICLE DIA. (D) (GRATICULE DIV.)</th>
<th>AREA CORRECTION (C)</th>
<th>NO. OF PARTICLES COUNTED</th>
<th>NC($D$)$^3$ (x1000)</th>
<th>% BY VOLUME</th>
<th>% BY VOLUME UNDERSIZE</th>
<th>NC($D$)$^2$ (x1000)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(MAG=100)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0-1</td>
<td>480</td>
<td>24</td>
<td>2.9</td>
<td>1.0</td>
<td>1.0</td>
<td>3.8</td>
</tr>
<tr>
<td>1-2</td>
<td>240</td>
<td>19</td>
<td>17.1</td>
<td>5.9</td>
<td>6.9</td>
<td>10.6</td>
</tr>
<tr>
<td>2-4</td>
<td>96</td>
<td>17</td>
<td>4.8</td>
<td>16.5</td>
<td>23.4</td>
<td>14.9</td>
</tr>
<tr>
<td>4-6</td>
<td>24</td>
<td>21</td>
<td>66.5</td>
<td>22.9</td>
<td>46.3</td>
<td>13</td>
</tr>
<tr>
<td>6-8</td>
<td>16</td>
<td>15</td>
<td>84</td>
<td>29</td>
<td>75.3</td>
<td>11.8</td>
</tr>
<tr>
<td>(MAG=100)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>8-12</td>
<td>1</td>
<td>55</td>
<td>57.3</td>
<td>19.8</td>
<td>95.1</td>
<td>5.5</td>
</tr>
<tr>
<td>12-16</td>
<td>$1/3$</td>
<td>15</td>
<td>14</td>
<td>4.8</td>
<td>99.9</td>
<td>1</td>
</tr>
</tbody>
</table>

+Calibration  
1 Div. = 2.5 μm  
\[ \text{Sum } NC(D)^3 = 290 \times 1.25 \mu m = 6 \mu m \]  
(Spread factor = 2.17)
### NOZZLE 320 x 450

<table>
<thead>
<tr>
<th>FLATTENED PARTICLE DIA. (D) (GRATICULE DIV.)</th>
<th>AREA CORRECTION (C)</th>
<th>NO. OF PARTICLES COUNTED</th>
<th>NC(D) \times 1000</th>
<th>% BY VOLUME</th>
<th>% BY VOLUME UNDERSIZE</th>
<th>NC(D)²</th>
</tr>
</thead>
<tbody>
<tr>
<td>(MAG=400) 0-1</td>
<td>320</td>
<td>59</td>
<td>4.8</td>
<td>1.6</td>
<td>1.6</td>
<td>6.3</td>
</tr>
<tr>
<td>1-2</td>
<td>160</td>
<td>41</td>
<td>25</td>
<td>8.3</td>
<td>9.9</td>
<td>15.2</td>
</tr>
<tr>
<td>2-4</td>
<td>32</td>
<td>75</td>
<td>72</td>
<td>23.8</td>
<td>33.7</td>
<td>22.3</td>
</tr>
<tr>
<td>4-6</td>
<td>16</td>
<td>42</td>
<td>87.1</td>
<td>28.8</td>
<td>62.5</td>
<td>17</td>
</tr>
<tr>
<td>6-8</td>
<td>16/3</td>
<td>26</td>
<td>49</td>
<td>16.2</td>
<td>78.7</td>
<td>6.9</td>
</tr>
<tr>
<td>(MAG=100) 8-12</td>
<td>1</td>
<td>46</td>
<td>47.8</td>
<td>15.8</td>
<td>94.5</td>
<td>4.6</td>
</tr>
<tr>
<td>12-16</td>
<td>1/5</td>
<td>3</td>
<td>16.8</td>
<td>5.6</td>
<td>100</td>
<td>.1</td>
</tr>
</tbody>
</table>

Calibration

1 Div. = 2.5 μm

\[ \frac{\text{Sum NC}(D)^3}{\text{Sum NC}(D)^2} = 302.5 \]

\[ \frac{\text{Sum NC}(D)^2}{\text{Sum NC}(D)^2} = 72.4 \]

\[ \text{Surface mean diameter} = \frac{\text{Sum NC}(D)^3}{\text{Spread factor = 1.96}} \times 1.25 \mu m = 5.2 \mu m \]

**APPENDIX 1.5** SIZE DISTRIBUTION OF SOLUTE AEROSOL.
<table>
<thead>
<tr>
<th>FLATTENED PARTICLE DIA. (D)  (GRATICULE DIV. *)</th>
<th>AREA CORRECTION (C)</th>
<th>NO. OF PARTICLES COUNTED</th>
<th>NC(D)^3* (x1000)</th>
<th>% BY VOLUME</th>
<th>% BY VOLUME UNDERSIZE</th>
<th>NC(D)^2*</th>
</tr>
</thead>
<tbody>
<tr>
<td>(MAG=4.00) 0-1</td>
<td>320</td>
<td>70</td>
<td>5.6</td>
<td>1.2</td>
<td>1.2</td>
<td>7.4</td>
</tr>
<tr>
<td>1-2</td>
<td>160</td>
<td>43</td>
<td>26.1</td>
<td>5.5</td>
<td>6.7</td>
<td>15.8</td>
</tr>
<tr>
<td>2-4</td>
<td>32</td>
<td>104</td>
<td>100</td>
<td>20.9</td>
<td>27.6</td>
<td>31</td>
</tr>
<tr>
<td>4-6</td>
<td>16</td>
<td>66</td>
<td>137.8</td>
<td>28.8</td>
<td>56.4</td>
<td>26.8</td>
</tr>
<tr>
<td>6-8</td>
<td>8</td>
<td>34</td>
<td>94.5</td>
<td>19.8</td>
<td>76.2</td>
<td>13.3</td>
</tr>
<tr>
<td>(MAG=100) 8-12</td>
<td>2</td>
<td>51</td>
<td>106</td>
<td>22.2</td>
<td>98.4</td>
<td>10.3</td>
</tr>
<tr>
<td>12-16</td>
<td>1/2</td>
<td>6</td>
<td>8.4</td>
<td>1.7</td>
<td>100</td>
<td>.6</td>
</tr>
</tbody>
</table>

*Calibration
1 Div. = 2.5 μm
Sum NC(D)^3 = 478.4
Sum NC(D)^2 = 105.2

* Volume and surface mean diameter respectively.

Surface mean diameter = \( \frac{\text{Sum NC(D)^3}}{\text{Sum NC(D)^2}} \times 1.25 \mu m = 5.7 \mu m \)

(Spread factor = 2.01)

APPENDIX 1.5 SIZE DISTRIBUTION OF SOLUTE AEROSOL
<table>
<thead>
<tr>
<th>FLATTENED PARTICLE DIA. (D)</th>
<th>AREA CORRECTION (C)</th>
<th>NO. OF PARTICLES COUNTED</th>
<th>NC(D)³ (x1000)</th>
<th>% BY VOLUME</th>
<th>% BY VOLUME UNDERSIZE</th>
<th>NC(D)² (x1000)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(MAG=400)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0-1</td>
<td>320</td>
<td>72</td>
<td>5.7</td>
<td>1.5</td>
<td>1.5</td>
<td>7.6</td>
</tr>
<tr>
<td>1-2</td>
<td>160</td>
<td>37</td>
<td>22.4</td>
<td>5.8</td>
<td>7.3</td>
<td>13.6</td>
</tr>
<tr>
<td>2-4</td>
<td>32</td>
<td>96</td>
<td>92.1</td>
<td>23.9</td>
<td>31.2</td>
<td>28.6</td>
</tr>
<tr>
<td>4-6</td>
<td>16</td>
<td>53</td>
<td>110.2</td>
<td>28.6</td>
<td>59.8</td>
<td>21.5</td>
</tr>
<tr>
<td>6-8</td>
<td>8</td>
<td>34</td>
<td>94.5</td>
<td>24.5</td>
<td>84.2</td>
<td>13.3</td>
</tr>
<tr>
<td>(MAG=100)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>8-12</td>
<td>1</td>
<td>54</td>
<td>56.2</td>
<td>14.6</td>
<td>98.8</td>
<td>5.5</td>
</tr>
<tr>
<td>12-14</td>
<td>1/4</td>
<td>6</td>
<td>3.3</td>
<td>.8</td>
<td>99.6</td>
<td>.3</td>
</tr>
<tr>
<td>14-16</td>
<td>1/4</td>
<td>1</td>
<td>.8</td>
<td>.2</td>
<td>99.8</td>
<td>.3</td>
</tr>
<tr>
<td>16-18</td>
<td>1/4</td>
<td>1</td>
<td>1.2</td>
<td>.3</td>
<td>100</td>
<td>.07</td>
</tr>
</tbody>
</table>

Calibration
1 Div. = 2.5 µm

Sum NC(D)³ = 366
Sum NC(D)² = 90.5

*Volume and surface mean diameter respectively.

Surface mean diameter = \( \frac{\text{Sum NC(D)}^3}{\text{Sum NC(D)}^2} \times 1.25\,\mu m = 5.3\,\mu m \)

(Spread factor = 1.90)
### Size Distribution of Solute Aerosol

<table>
<thead>
<tr>
<th>FLATTENED PARTICLE DIA. (D) (GRATTICLE DIV.)</th>
<th>AREA CONNECTION (C)</th>
<th>NO. OF PARTICLES COUNTED</th>
<th>NC(D) -3×1000</th>
<th>% BY VOLUME</th>
<th>% BY VOLUME UNDERSIZE</th>
<th>NC(D) -2×1000</th>
</tr>
</thead>
<tbody>
<tr>
<td>0-1</td>
<td>160</td>
<td>42</td>
<td>1.7</td>
<td>0.3</td>
<td>0.3</td>
<td>2.2</td>
</tr>
<tr>
<td>1-2</td>
<td>160</td>
<td>18</td>
<td>11</td>
<td>2.2</td>
<td>2.5</td>
<td>6.7</td>
</tr>
<tr>
<td>2-4</td>
<td>32</td>
<td>68</td>
<td>66</td>
<td>13.4</td>
<td>15.9</td>
<td>20.5</td>
</tr>
<tr>
<td>4-6</td>
<td>6</td>
<td>55</td>
<td>114</td>
<td>23.1</td>
<td>39</td>
<td>22.3</td>
</tr>
<tr>
<td>6-8</td>
<td>8</td>
<td>27</td>
<td>77</td>
<td>15.6</td>
<td>54.6</td>
<td>10.8</td>
</tr>
<tr>
<td>(MAG=100) 8-12</td>
<td>2</td>
<td>53</td>
<td>110</td>
<td>22.3</td>
<td>76.9</td>
<td>10.5</td>
</tr>
<tr>
<td>12-16</td>
<td>½</td>
<td>43</td>
<td>61.6</td>
<td>12.5</td>
<td>89.4</td>
<td>4.3</td>
</tr>
<tr>
<td>16-18</td>
<td>1</td>
<td>3</td>
<td>14.7</td>
<td>3</td>
<td>92.4</td>
<td>.9</td>
</tr>
<tr>
<td>(MAG=30) 18-24</td>
<td>1⁄9</td>
<td>25</td>
<td>26.5</td>
<td>5.4</td>
<td>97.8</td>
<td>1.2</td>
</tr>
<tr>
<td>24-30</td>
<td>1⁄9</td>
<td>5</td>
<td>11.1</td>
<td>2.2</td>
<td>100</td>
<td>.4</td>
</tr>
</tbody>
</table>

*Calibration

\[
1 \text{ Div. } = \frac{\text{Sum } NC(D)^3}{2.5 \mu m} = 494
\]

\[
\text{Sum } NC(D)^2 = 79.8
\]

*Volume and surface mean diameter respectively.

Surface mean diameter = \[
\frac{\text{Sum } NC(D)^3}{\text{Sum } NC(D)^2} \times 1.25\mu m = 8.7\mu m
\]

(Spread factor = 2.0)

APPENDIX 1.5  SIZE DISTRIBUTION OF SOLUTE AEROSOL
METERED SPRAY; 0.5 mm x 0.25 mm NOZZLE

<table>
<thead>
<tr>
<th>FLATTENED PARTICLE DIA. (D)</th>
<th>AREA CORRECTION (C)</th>
<th>NO. OF PARTICLES COUNTED</th>
<th>NC(D)³* (x1000)</th>
<th>% BY VOLUME</th>
<th>% BY VOLUME UNDERSIZE</th>
<th>NC(D)²* (x1000)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(MAG=400)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0-1</td>
<td>160</td>
<td>61</td>
<td>2.4</td>
<td>3</td>
<td>3</td>
<td>3.2</td>
</tr>
<tr>
<td>1-2</td>
<td>160</td>
<td>22</td>
<td>13.4</td>
<td>16.5</td>
<td>19.5</td>
<td>8.1</td>
</tr>
<tr>
<td>2-3</td>
<td>16</td>
<td>50</td>
<td>13</td>
<td>16</td>
<td>35.5</td>
<td>5</td>
</tr>
<tr>
<td>3-4</td>
<td>16</td>
<td>23</td>
<td>16.1</td>
<td>19.9</td>
<td>55.5</td>
<td>4.5</td>
</tr>
<tr>
<td>4-6</td>
<td>16</td>
<td>8</td>
<td>16.6</td>
<td>20.5</td>
<td>76</td>
<td>3.2</td>
</tr>
<tr>
<td>(MAG=100)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>6-8</td>
<td>1</td>
<td>36</td>
<td>12.6</td>
<td>15.5</td>
<td>91</td>
<td>1.8</td>
</tr>
<tr>
<td>8-10</td>
<td>1</td>
<td>10</td>
<td>7.4</td>
<td>9.1</td>
<td>100</td>
<td>.8</td>
</tr>
</tbody>
</table>

Calibration

Div. = \( \frac{\text{Sum NC(D)}^3}{2.5 \mu m} = 81.5 \)

\( \text{Sum NC(D)}^2 = 26.6 \)

* Volume and surface mean diameter respectively.

Surface mean diameter = \( \frac{\text{Sum NC(D)}^3}{\text{Sum NC(D)}^2} \times 1.25 \mu m = 3.9 \mu m \)

(Spread factor = 2.05)

For nozzles II, III and IV the reduced data only is available.

APPENDIX 1.5 SIZE DISTRIBUTION OF SOLUTE AEROSOL
Appendix 2  
**Interferometric photomicrography of flattened droplets on a glass slide.**

Size analyses of the residual liquid solute aerosol were determined by sedimentation on to glass slides, followed by microscope counting of the flattened droplets. The original spherical diameter was determined from the flattened diameter by calculating the "flattening factor" which is the ratio of flattened diameter to spherical diameter. This was determined as described in Section 5, by measuring the focal length of the resultant plane convex lens formed by the flattened droplet; the flattening factor was then obtained geometrically. This method assumes that the angle of contact between droplet and slide is acute and not obtuse. Although there appears to be ample evidence in the literature (57 for example) to justify this assumption, it was considered worthwhile to check, directly, the profile of the droplets. This was done as shown in Fig. A 2.1 using, for convenience at that time, droplets of dioctylphthalate (D.O.P.). This was dissolved in an aerosol propellant and sprayed on to a glass microscope slide, which had been vacuum coated with a semi-transparent film of aluminium. The droplets were viewed with a metallurgical microscope, using a sodium lamp as the source of illumination. Interference fringes were obtained as shown in Fig. A 2.2. The distance between each fringe represented a change in height of:

\[
\text{wavelength of the sodium lamp} \times \frac{0.589}{2 \times 1.485} = 0.198 \, \mu m \text{ for DOP.}
\]

Thus the height of the droplet could be determined by counting the
Fig. A2.1. Interferometric photomicrography of droplets.
PHOTOMICROGRAPH OF DROPLETS, SHOWING THE PRESENCE OF INTERFERENCE FRINGES.
total number of fringes.

For a few droplets, their focal lengths were determined as described previously, by light transmitted through the slide. From these values the height of the droplet could be determined geometrically assuming an acute angle of contact. The two values of droplet height could then be compared.

Results for two droplets are shown below.

<table>
<thead>
<tr>
<th>FLATTENED DROPLET LENGTH µm</th>
<th>FOCAL DROPLET LENGTH µm</th>
<th>DROPLET HEIGHT µm (From focal length)</th>
<th>NO. OF FRinges</th>
<th>DROPLET HEIGHT µm (From fringes)</th>
</tr>
</thead>
<tbody>
<tr>
<td>73 ± 1</td>
<td>358 ± 5</td>
<td>3.9 ± 0.1</td>
<td>18 ± 1</td>
<td>3.6 ± 0.2</td>
</tr>
<tr>
<td>16 ± 0.5</td>
<td>83 ± 10</td>
<td>0.82 ± 0.1</td>
<td>5 ± 1</td>
<td>1.0 ± 0.2</td>
</tr>
</tbody>
</table>

It is seen that agreement between the two methods is acceptable, confirming that as expected the droplets have an acute angle of contact.
Appendix 3 Critical flow of air through some of the orifices used to form two-orifice nozzle assemblies.

It was considered desirable to obtain an independent assessment of the discharge coefficients of the orifices used to form the two-orifice nozzle assemblies. These independently determined values could then be compared with those determined for the flow of saturated liquid propellant and two-phase propellant liquid and vapour.

A critical flow arrangement was used in which ambient air was drawn, by a vacuum pump, through first a rotameter flow meter and then the orifice under test. The pressure on the low pressure side of the orifice was measured with a vacuum gauge. Ambient temperature and pressure were also recorded. The rotameter was calibrated by drawing air from an inverted measuring cylinder whose open end was below water. Atmospheric pressure within the cylinder was maintained by lowering the cylinder as the air was drawn out such that water levels inside and outside the cylinder remained approximately equal. The vacuum gauge was calibrated with a mercury manometer.

For each nozzle the flow rate was increased in steps; at each step the vacuum gauge and rotameter readings were recorded. The results are shown in Fig. A 3.1. It is apparent from the results that the orifices are displaying choking effects with no increase in mass flow-rate beyond a critical pressure. Thus the "orifices" which have a length to diameter ratio of approximately 1 : 1, are behaving as short tubes as would have been expected.

The discharge coefficient for each orifice was determined from the isentropic critical flow equation:

\[
\text{Mass flow-rate} = C_D A \sqrt{\frac{\gamma + 1}{\gamma - 1}} \left( \frac{2}{\gamma + 1} \right)
\]
FIG. A3.1. CRITICAL FLOW OF AIR THROUGH VARIOUS SINGLE ORIFICES.
where $C_Q$ is the discharge coefficient,

$A$ is the orifice area

$\gamma$ is the specific heat ratio, equal to 1.4

and $\rho$ and $P$ are respectively the density and pressure of the ambient air.

From this equation the discharge coefficient of each orifice may be determined. These are given in the chart below.

<table>
<thead>
<tr>
<th>NOZZLE</th>
<th>DISCHARGE COEFFICIENT</th>
<th>REYNOLDS NUMBER (approx)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Based on minimum area</td>
<td>Based on mean area</td>
</tr>
<tr>
<td>700</td>
<td>0.82</td>
<td>0.82</td>
</tr>
<tr>
<td>640</td>
<td>0.87</td>
<td>0.84</td>
</tr>
<tr>
<td>Y</td>
<td>0.83</td>
<td>0.79</td>
</tr>
<tr>
<td>450 (D)</td>
<td>0.89</td>
<td>0.865</td>
</tr>
<tr>
<td>320</td>
<td>0.83</td>
<td>0.805</td>
</tr>
<tr>
<td>250</td>
<td>0.81</td>
<td>0.81</td>
</tr>
<tr>
<td>Mean</td>
<td>$0.84 \pm 0.03$</td>
<td>$0.82 \pm 0.025$</td>
</tr>
</tbody>
</table>

The mean discharge coefficient for the six orifices tested is 0.84 based on minimum area and 0.82 based on mean area. These values are in good agreement with a critical discharge coefficient of 0.83 given by Grace and Lapple (24) for a 1.6 mm (1/16 inch) dia. thick plate orifice of thickness 1.6 mm; the Reynolds number was approximately $2 \times 10^4$. This is also the value to be expected from a sharp edged short tube with a head loss coefficient of 0.5.
Appendix 4 Stereoscan electronmicrography of residual aerosol droplets.

Towards the end of this project it became possible to use a stereoscan electronmicroscope. A number of micrographs were consequently taken of the residual aerosol from a metered spray sedimented on to cleaned glass discs, which were then glued on to the microscope sample holders. In order to render the specimen electrically conductive "silver dag" was painted round the edge of the glass disc and the specimen was vacuum coated with platinum.

Two types of sprays were used. The first contained a powdered drug dispersed in the surfactant "span 85". The second contained a powdered drug dispersed in soya lecithin. The first is shown in Fig. A 4.1(a) while the second is shown in Fig. A 4.1(b). It is seen that in both cases, all or part of the surfactant has remained on the glass discs, although the soya lecithin (in (b)) has assumed a shrivelled appearance, suggesting that a volatile component has evaporated leaving behind a non-volatile component.

It would appear from these results that electronmicrography, with its attendant high resolution, is applicable to the size determination of aerosols without the need for tedious replication techniques.
FIG. A.4.1(a)

Mag. = 2700

FIG. A.4.1(b)  STEREOSCAN PHOTOMICROGRAPHS.

Mag. = 11000
**NOMENCLATURE**

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>Orifice area</td>
</tr>
<tr>
<td>B</td>
<td>Constant defined in Section 6</td>
</tr>
<tr>
<td>C&lt;sub&gt;1&lt;/sub&gt;, C&lt;sub&gt;2&lt;/sub&gt;, C&lt;sub&gt;3&lt;/sub&gt;</td>
<td>Constants defined in Section 4</td>
</tr>
<tr>
<td>C&lt;sub&gt;P&lt;/sub&gt;, C&lt;sub&gt;V&lt;/sub&gt;</td>
<td>Specific heats at constant pressure and volume</td>
</tr>
<tr>
<td>C&lt;sub&gt;1&lt;/sub&gt;, C&lt;sub&gt;g&lt;/sub&gt;</td>
<td>Concentration of liquid, gas</td>
</tr>
<tr>
<td>D</td>
<td>Orifice diameter</td>
</tr>
<tr>
<td>D&lt;sub&gt;p&lt;/sub&gt;</td>
<td>Flattened droplet diameter</td>
</tr>
<tr>
<td>d</td>
<td>Particle diameter or differential operator as appropriate</td>
</tr>
<tr>
<td>e</td>
<td>2.718</td>
</tr>
<tr>
<td>F</td>
<td>Focal length</td>
</tr>
<tr>
<td>f</td>
<td>Divisor defined in Section 4, or &quot;function of&quot;</td>
</tr>
<tr>
<td>G</td>
<td>Mass flow-rate per unit area</td>
</tr>
<tr>
<td>H</td>
<td>Enthalpy (total)</td>
</tr>
<tr>
<td>h</td>
<td>Enthalpy (specific)</td>
</tr>
<tr>
<td>J</td>
<td>Momentum flux</td>
</tr>
<tr>
<td>k</td>
<td>( \omega/\sigma )</td>
</tr>
<tr>
<td>l</td>
<td>( \phi/\sigma )</td>
</tr>
<tr>
<td>( \dot{m} )</td>
<td>Mass flow-rate</td>
</tr>
<tr>
<td>m</td>
<td>Mass</td>
</tr>
<tr>
<td>N</td>
<td>Number</td>
</tr>
<tr>
<td>N&lt;sub&gt;\text{Web}e&lt;/sub&gt;</td>
<td>Weber number ( \frac{1}{2} \rho_g \frac{u^2 d}{\sigma} )</td>
</tr>
<tr>
<td>N&lt;sub&gt;Re&lt;/sub&gt;</td>
<td>Reynolds number ( \frac{u d \rho}{\eta} )</td>
</tr>
<tr>
<td>P</td>
<td>Absolute pressure</td>
</tr>
<tr>
<td>Q</td>
<td>Volume flow-rate</td>
</tr>
<tr>
<td>q</td>
<td>Mass fraction of propellant evaporated (quality)</td>
</tr>
</tbody>
</table>
Critical pressure ratio
Standard deviation
Temperature °C or K as appropriate
Time
Velocity
Volume
Specific volume
Molecular weight
Ratio of volume of vapour to volume of liquid. $X = \frac{\rho_1}{\rho_5}^{1-q}$
Dimension - usually longitudinal
Dimension - usually transverse
Distance, from the nozzle, of the apparent origin of the spray

**GREEK SYMBOLS**

Half-angle of jet divergence or void fraction as appropriate
Ratio of specific heats $C_p / C_v$
Finite difference
Small difference
Viscosity
Latent heat of vaporisation
Density
Distance from the axis of a jet where the velocity falls to $(\frac{1}{6})$ of the axial value
Distance from the axis of a jet where the vapour concentration falls to $(\frac{1}{6})$ of the axial value
Impaction parameter
Distance from the axis of a jet where the liquid droplet concentration falls to $\left(\frac{1}{8}\right)$ of the axial value

**SUBSCRIPTS**

1,2 First and second orifices respectively of two-orifice nozzle assembly

a Axial

amb Ambient

c Critical or mass-limiting

e Expansion chamber

g Gas or vapour

i Initial

l Liquid

n Relates to a region just beyond the nozzle housing exit where the static pressure within the jet has returned to ambient but before significant atmospheric entrainment has taken place.

o Nozzle exit

P Constant pressure

p Particle

Q Relates to discharge coefficient

rel Relative

s Sonic

V Constant volume

vel Velocity
SUPERSCRIPTS

* Indicates that the value is reduced to standard conditions

- Mean value
  \( \bar{u} \): Time average value of velocity

\:
Denotes a dimensionless quantity

\cdot
Time derivative
1. Benatt, F.G.S. & Eisenklam, P.
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