PHASE DOPPLER ANEMOMETRY STUDIES OF SPRAY FREEZING

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ABSTRACT

The freezing of sprays by cold or cryogenic fluids has many potential applications in the food and pharmaceutical industries, mainly as a precursor to freeze drying. Phase Doppler Anemometry (PDA) has been used to characterise spray-freezing by measuring the size and velocity of individual droplets passing different points positioned vertically below the spray nozzle. This was performed in a spray freezing chamber, using a two component high power, high resolution PDA system, with a receiver positioned at an angle of 70 degrees to maximise collection of refractive scatter. Droplet size distributions were normalised with respect to the total number of droplets/particles detected. Some detected droplets were rejected by the PDA software for the measurement of size and velocity. Furthermore, dropping the chamber temperature (from +20 to –60°C) and increasing the distance from the nozzle (from 38 mm to 220 mm) increased the rejection rate. This might be due to the onset of drop freezing which could interfere with refractive scatter. The apparent freezing behaviour of hydraulic nozzle sprays was similar to that predicted by a simple model for drop freezing, and supports the idea that PDA can be used as a non-invasive probe of drop freezing.

INTRODUCTION

Spray-freezing involves the solidification of an atomised liquid by contacting it with an inert cooling medium below the freezing point of the liquid, which for aqueous solutions requires sub-zero temperatures. Various cooling media can be used such as liquid nitrogen, cold air and dry ice. Spray-freezing can be used to generate products in their own right or as a precursor to lyophilisation. Complex microstructures can be generated in the resultant droplets by the pattern of formation of ice crystals, and this yields a porous structure if the ice is allowed to sublime. Relatively little has been published on the technique to date yet the potential for generating products from thermally labile materials with controlled microstructures, renders this an exciting area for development in the food and pharmaceutical industries.

The spray-freezing process in a cold gas is complex and involves a number of mechanisms. Firstly, one must consider the fluid mechanics of the spray - its formation and the movement of individual spray drops with respect to each other and the gas. Second is the heat transfer between the gas phase and the droplets, which depend on the local conditions of gas temperature, droplet temperature and droplet-gas slip velocity. Finally, there is the freezing mechanism itself.

A recent study of single droplet freezing [1] indicates that the drop freezing process is composed of a number of stages: (i) initial cooling to a supercooled temperature, (ii) nucleation, (iii) recalescence, whereby rapid crystal growth occurs at the expense of sensible heat from the supercooled droplet, and (iv) further, slower crystal growth which is limited by heat transfer from the gas.

To date few studies have been made of drop freezing within sprays. The aim of this study is to use Phase Doppler Anemometry (PDA) to perform measurements on a spray freezing system.

PDA is a single point optical diagnostic technique, and is able to measure the instantaneous velocity and diameter of individual drops or particles [2] [3] [4]. This method of characterisation is widely used in research and process optimisation relating to particle streams and liquid sprays. Applications include the analysis of atomised liquids in fuel injection systems in internal combustion engines [3] and spray drying [5].

The experiments described here have been used to measure vertical velocity and droplet/particle diameter along the centreline of the spray at various distances from the nozzle (up to 220 mm) and at various chamber gas temperatures (ambient down to -62°C). Most experiments were performed using a pneumatic (twin-fluid) nozzle, but a few experiments were also performed with a hydraulic (pressure) nozzle. Apart from the different drop size ranges produced by the nozzles, the pneumatic nozzle introduces a significant quantity of ambient temperature gas into the spray which could affect spray freezing.

APPARATUS AND EXPERIMENTAL METHOD

Spray-freezing chamber

Spray freezing was carried out in a specially constructed cylindrical chamber (height 1.5 m x diameter 0.8 m) which included five vertical (removable) windows to permit PDA measurements to be made. The sprayed liquid was distilled water which was atomised using either a pneumatic (XA Pr050) or a hydraulic nozzle (WL053), both supplied by Bete Ltd, Lewes, Sussex, UK. The nozzle housings required air heating to prevent freezing of the feed stream within the nozzle and avoid blockage. The feed pressures for the pneumatic nozzle were 3 barg for the atomising gas and 2 barg for the liquid feed. The pressure of the liquid feed for the hydraulic nozzle was 5 barg. In both cases the liquid feed was supplied from a pressurised feed tank to maintain constancy of flow. Before spraying took place the chamber was first purged with dry nitrogen gas from a cylinder to remove humidity from the chamber and then cooled using a liquid nitrogen supply. A number of chamber temperatures were used (20°C, -22°C, -42°C and –62°C), which were maintained by controlling the flows of liquid nitrogen and nitrogen cylinder gas to the chamber.

Phase Doppler Anemometry set-up

A two component high power, high resolution LDA/PDA transmission system was used which has been well documented [3]. The configuration for this application was: laser powers of 120 and 250 milli-watts per beam with coincident measurement volumes of diameters of 56 and 59 microns and fringe spacings of 4.40 and 4.64 microns for the 488 and 514 nm laser beam wavelengths respectively. A Dantec 57X10 receiver optical system was positioned at a scattering angle of 70 degrees while the aperture micrometer setting was set to 0.5 mm. This optical configuration resulted in an effective measurement volume length of 0.1 mm and a maximum drop size measurement capability of 233 microns. A Dantec 58N10 Covariance signal processor was used in this study. The Doppler signal frequency bandwidth was set to 45 MHz match which resulted in an experimental velocity bandwidth of -46 to 160 m/s.

Experimental method

Great care was made to ensure that the chamber was clear of fog from previous experiments before an experiment was performed. Significant fogging of the apparatus was observed during the experiments, but a “window of opportunity” of a few seconds between the establishment of a steady spray and gradual fogging of the chamber was sufficient to allow measurements to take place. Typically 3 seconds were allowed for measurements with the pneumatic nozzle, during which typically 200 000 drops were measured with typical mean diameter of 15 microns. For the hydraulic nozzle only a matter of 5 000 droplets could be measured in this time, due to a smaller number of much larger drops (mean size typically 75 microns).

The measurement volume was located on the centreline of the spray at various distances vertically below the nozzle orifice (60, 100, 160 and 220 mm for the pneumatic nozzle, and 38 and 108 mm for the hydraulic nozzle). Droplet size distributions (DSDs) were normalised with respect to the total number of droplets/particles detected by the PDA equipment (i.e. the total spray population). Repeats were made of selected experiments.

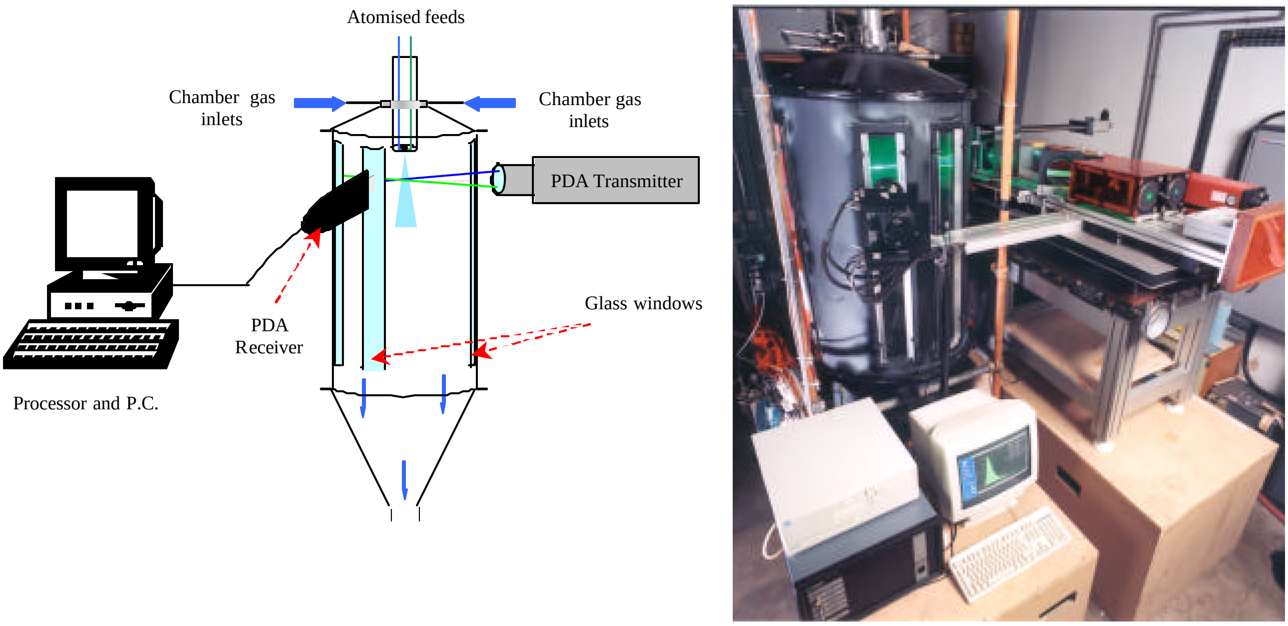

Figure 1 – Schematic of spray-freezing rig and PDA apparatus (left) and photograph during measurements (right).

RESULTS

Figure 1 shows DSDs from initial tests performed at ambient temperature using the pneumatic nozzle. Average drop sizes were observed to be in the region of 10 to 15 µm. There is only a minor variation between the DSDs measured at different distances. When the experiments were repeated at a chamber temperature of –42°C, however, a marked difference could be seen between the DSDs recorded at a distance of 60 mm and those at greater distances (Figure 2). The peaks at greater distances are shifted to lower values and the size of the peaks has also decreased. This is as a result of droplets being detected by the PDA but the PDA software has not returned a diameter (or velocity) value as the internal validation check on the measurements have failed. Thus some droplets have been rejected by the PDA for size measurement. By comparing Figure 1 and Figure 2 one might reasonably infer that if the underlying DSDs of the spray at the two temperatures are similar, then the "missing" or "rejected" droplets or particles can be deduced from the difference between the normalised distributions (for a particular size class) at ambient and sub-zero temperatures.

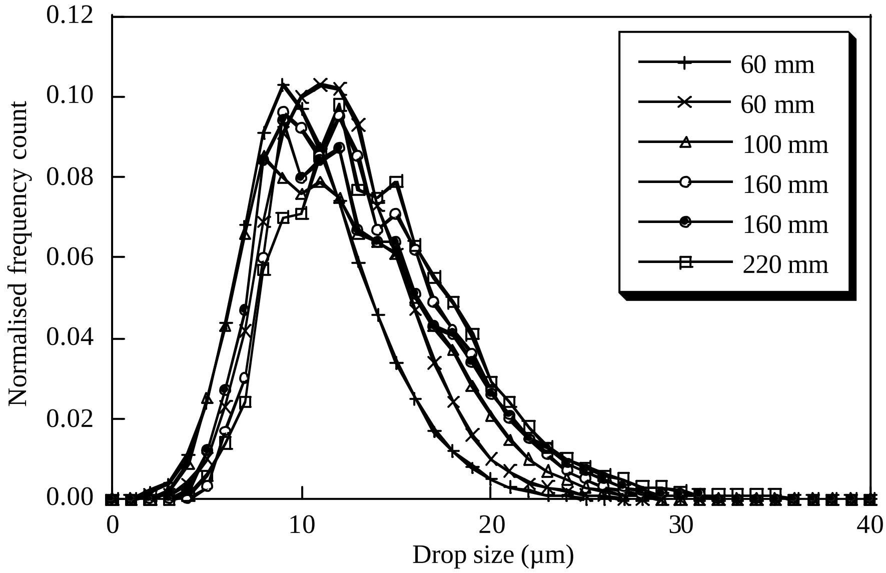

Figure 1. Drop size distribution (normalised against total number of detected objects) for a pneumatic water spray at ambient temperature and at various distances from the nozzle.

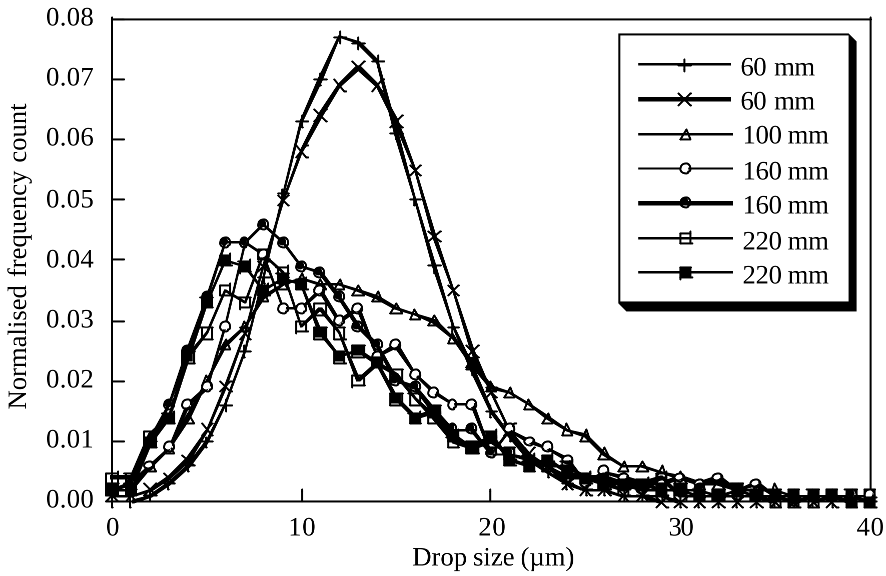

Figure 2. Drop size distribution (normalised against total number of detected objects) for a pneumatic water spray at a chamber temperature of –42°C and at various distances from the nozzle.

DSD data from experiments with PDA measurements taken at the same distance from the nozzle, but at varying chamber temperatures are shown in Figure 3 (60 mm distance) and Figure 4 (160 mm distance). These allow direct comparisons to be made between experiments at ambient and sub-zero temperatures. At a distance of 60 mm a "measurement loss" of droplets is seen to occur as the temperature is dropped from 20°C to –22°C, with a much smaller further reduction (if at all) at lower temperatures (-42°C and –62°C). At 160 mm there is also a reduction in counts when the temperature is lowered, but by a greater extent. It can be further seen in Figures 2 and 4 that dropping the temperature to –22°C and increasing the distance to 100 mm increases the "rejection rate" or "loss" of measurable droplets considerably , but that further decreases in temperature or increases in distance appear to have a marginal or insignificant effect only. The larger droplets appear to suffer the most rejections. The DSDs also seem to approach a "limiting distribution" below which they do not fall.

Experiments with hydraulic nozzles (Figures 5 and 6), also show a loss of measurable droplets as the tempearture is lowered from ambient to –42°C. The results are markedly different to those from the pneumatic sprays as the rejection of droplets appears to instead affect the smallest droplets only. Increasing the distance from 38 mm to 108 mm does not appreciably affect the DSD.

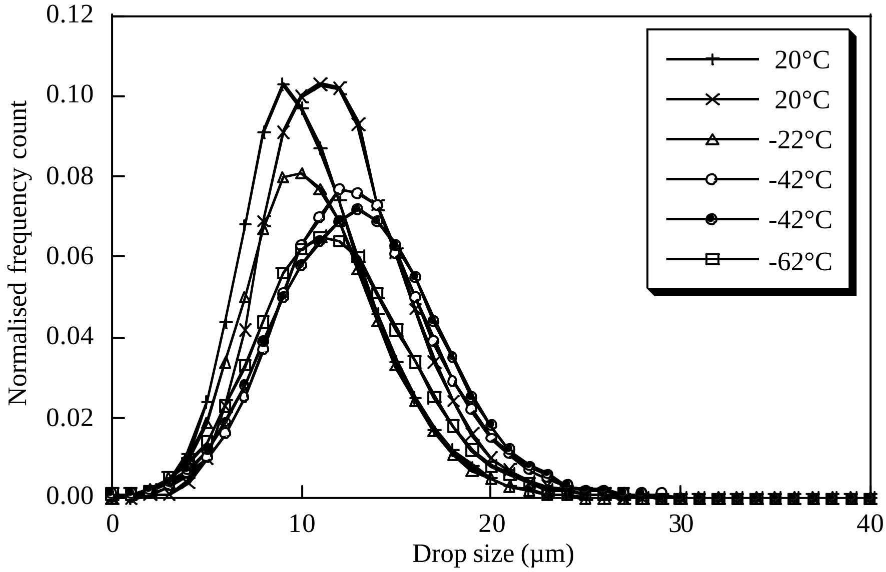

Figure 3. Drop size distribution (normalised against total number of detected objects) for a pneumatic water spray 60 mm from the nozzle and at various chamber temperatures.

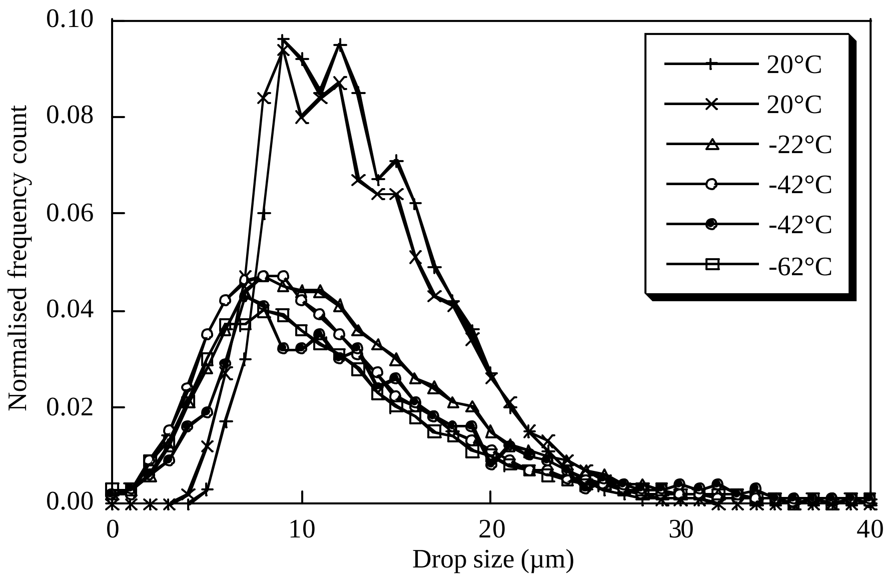

Figure 4. Drop size distribution (normalised against total number of detected objects) for a pneumatic water spray 160 mm from the nozzle and at various chamber temperatures.

DISCUSSION

Th results clearly show that frequency counts normalised against the total number of detected (not measured) droplets decrease when PDA measurements are made at sub-zero temperatures. The most likely explanation for this is droplet freezing. The PDA experiments described here were set up to measure refractive scatter, which is the predominant mode of scatter for liquid droplets. Reflective scatter is more typical for solid particles, and produces the maximum intensity for PDA observations at a different receiver angle. In a situation where objects change from liquid droplets to solid particles (as occurs in spray-freezing) there is thus bound to be some change to the PDA response to the spray population, from mainly refractive to mainly reflective scatter. Furthermore, the refractive signal can be disrupted by even partial solidification. This may well occur just after the nucleation and recalescance stages, as a significant fraction of water can transform into ice during the usually very rapid recalescence stage. This may therefore explain the apparent rejection of droplets by the PDA at low temperatures.

This hypothesis would explain the different behaviour observed for the pneumatic sprays (where rejections occured most often with the medium to large droplets) and the hydraulic sprays where the smallest droplets were most affected. With hydraulic sprays, one would expect that the smallest droplets to freeze first as they have the largest surface area to volume ratio. With pneumatic sprays, however, a significant quantity of ambient temperature atomisation gas is introduced with the spray and is likely to shield the droplets from the cold chamber gas. The smallest droplets are most likely to be swept along with pockets of atomisation gas and not come into direct contact with the chamber gas, but the momentum of larger droplets could well take the larger droplets out of a pocket of decelerating atomisation gas.

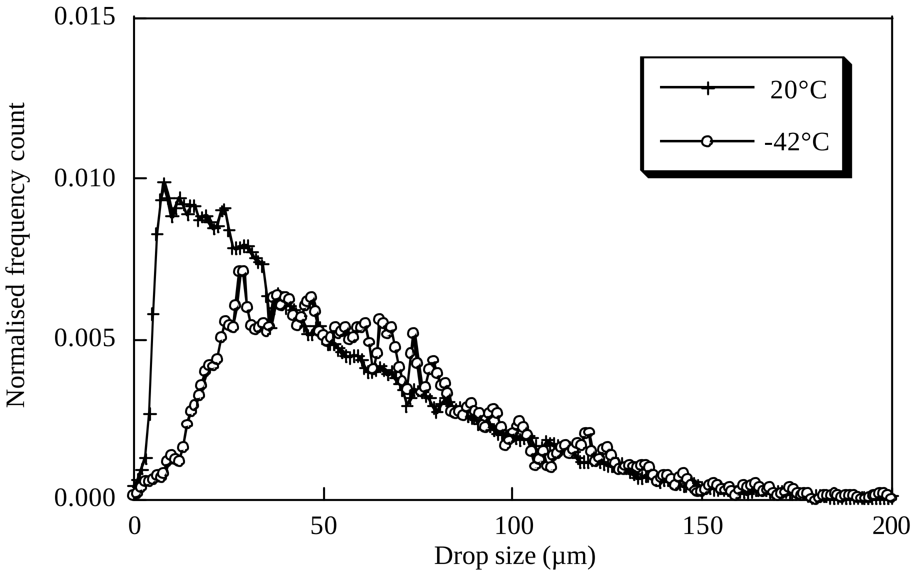

Figure 5. Drop size distribution (normalised against total number of detected objects) for a hydraulic water spray 38 mm mm from the nozzle and at two different chamb er temperatures.

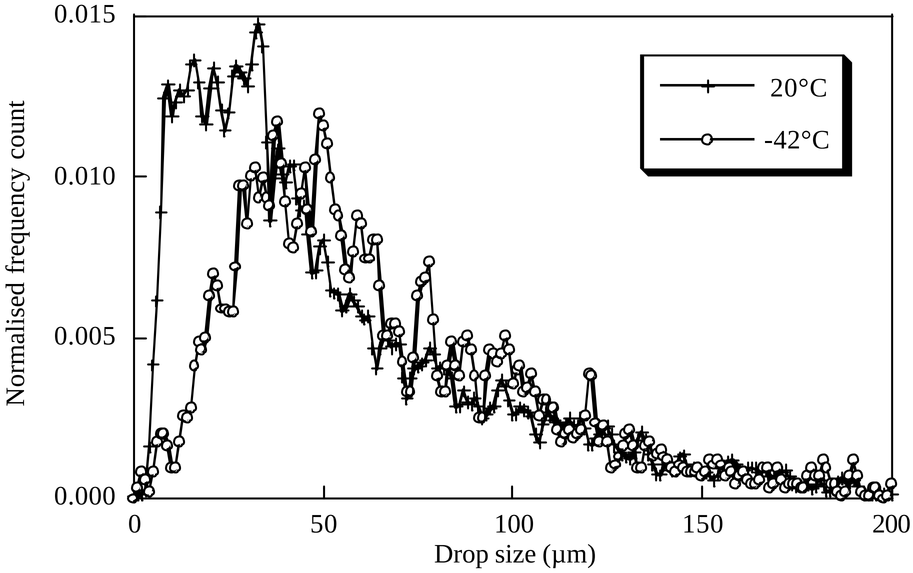

Figure 5. Drop size distribution (normalised against total number of detected objects) for a hydraulic water spray 108 mm mm from the nozzle and at two different chamber temperatures.

Table 1. Estimates of nucleation time of droplets in hydraulic sprays.

Drop size (μm)	2	5	10	20	50	100
Nucleation time (ms)	0.03	0.26	1.3	4.1	22.6	70.0

An order of magnitude estimate for freezing time can be made using a simple single droplet model which can be applied to hydraulic sprays (where droplets encounter only chamber gas) using the Ranz Marshall correlations for heat transfer coefficient [6][7]. Table 1 shows estimations of the time taken for droplets to reach a critical nucleation temperature of –10°C (an estimated value), assuming a terminal slip velocity (a pessimistic predictor of slip velocity), and a chamber temperture of –42°C.

These can be compared to rough estimates of times taken to reach the various nozzle positions using velocity data taken from the PDA experiments. For the hydraulic sprays the mean times taken to reach 38 mm and 108 mm respectively were approximately 1-2 ms and 5-9 ms (for drops over a size range of 20 mm to 150 mm), which suggests a cut-off size (below which drops have experienced nucleation) of around 10 μm for the 38 mm position and around 25 μm for te 108 mm position. Although a precise comparison should not be made on the basis of rough estimates, the order of magnitude agreement is encouraging.

An explanation for the limiting distributions, however, is not obvious. This may be due to the latent heat liberated from those droplets that do solidify heating up the gas and slowing down heat transfer away from the other drops. Alternatively, some partially frozen droplets may still be able to register a size measurement with the PDA if the shape and position of the newly formed ice phase is not too disruptive. This will require further research.

CONCLUSIONS

The results of the study show that at a fixed chamber temperature the rejection rate for PDA size measurements on droplets increased significantly with increasing distance from the nozzle. Similarly it was found that dropping the chamber temperature also increased the rejection rate. An analysis of the normalised drop size distributions showed that in the case of the hydraulic nozzle the rejected drops (those which disappear compared to the distribution at ambient conditions) were almost exclusively at the smaller end of the distribution, whereas for the pneumatic nozzle the “loss” of droplets was more uniform. A difference in behaviour between the two systems might be expected as the pneumatic nozzle injects a significant quantity of atomisation gas at ambient temperature into the spray which is able to shield the spray droplets from the low temperature of the chamber gas. Drop freezing may thus be more chaotic and depend on random contact with small pockets of low temperature chamber gas. Conversely, with the hydraulic nozzle the gas temperature can be assumed to be much closer to the chamber temperature.

A simple model of the freezing process was used to gain an order of magnitude estimate for the freezing time and distance travelled for droplets using diameter and vertical velocity data derived from the PDA experiments. The model confirmed that the distances and temperatures involved with drop freezing taking place were consistent with those observed.

This work therefore strongly suggests that the PDA technique can be used as a non-invasive probe of drop freezing in spray freezing systems, in addition to highly valuable correlated data for drop size and velocity.

ACKOWLEDGEMENTS

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NOMENCLATURE

DSD – drop size distribution
PDA – Phase Doppler Anemometry

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