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Experimental Study of Cryogen Spray Properties for Application in Dermatologic Laser Surgery
Guillermo Aguilar*, Boris Majaron, Emil Karapetian, Enrique J. Lavernia, and J. Stuart Nelson

Abstract—Cryogenic sprays are used for cooling human skin during laser dermatologic surgery. In this paper, six straight-tube nozzles are characterized by photographs of cryogenic spray shapes, as well as measurements of average droplet diameter, velocity, and temperature. A single-droplet evaporation model is utilized to predict average spray droplet diameter and temperature. The results show two distinct spray patterns—sprays for 1.4-mm-diameter nozzles (wide nozzles) show significantly larger average droplet diameters and higher temperatures as a function of distance from the nozzle compared with those for 0.5–0.8-mm-diameter nozzles (narrow nozzles). These results complement and support previously reported studies, indicating that wide nozzles induce more efficient heat extraction than the narrow nozzles.

Index Terms—Droplet size, nozzle geometry, skin cooling.

I. INTRODUCTION

Lasers are used for treating hypervascular skin lesions, such as port wine stain birthmarks, and the clinical application of interest in this work, and several papers summarize the progress to date [1]–[3]. To remove these lesions, patients are treated with laser pulses that induce permanent thermal damage to targeted blood vessels, typically located 200–500 μm below the skin surface. However, nonspecific absorption of laser energy by epidermal melanin, localized within the most superficial skin layer, can lead to undesirable damage such as scarring or dyspigmentation [4]. To prevent thermal injury to the epidermis, short sprays of cryogen are sprayed on the skin prior to laser exposure [5]. In order to achieve optimal cooling of the epidermis with minimal cooling of the subsurface target, it is necessary to control precisely the cryogen spurt duration [6] and spray properties. Although technology for cryogen-spray-cooling (CSC)-assisted laser therapy has been commercially available for several years, only a few studies have focused on measuring systematically the properties of cryogen sprays produced by different nozzle geometries [7]–[10]. Further studies are needed to better understand how spray properties could be modified to obtain optimum cooling efficiency and selectivity.

In this paper, the dependence of spray properties on nozzle geometries and their variation as a function of distance from the nozzle are studied by characterizing cryogen sprays from six different nozzles. First, the effect of nozzle diameter ($D_N$) and length ($L_N$) on spray shape is observed using fast-flashlamp photography. Then, the average droplet diameter $D$, velocity $V$, and temperature $T$ are measured at various distances from the nozzle tip $z$. Subsequently, we compare our experimental results of $D$ and $T$ with an earlier developed single-droplet evaporation model [11]. Finally, our experimental results are discussed in the context of those obtained during recent studies aimed at measuring the heat extraction from skin during CSC [12]–[14].

II. EXPERIMENTAL SYSTEMS AND PROCEDURES

A. Spray Forming Systems

The cryogen utilized in the present study is 1,1,1,2-tetrafluoroethane (R-134a) with a boiling temperature $T_B$ of $-26^\circ$C at atmospheric pressure. This is the only FDA approved cryogen for laser dermatologic surgery. The cryogen is kept in a container at a saturation pressure of 660 kPa (95.7 psi) at 25°C, and delivered through a standard high-pressure hose to the nozzles under study via a fuel injector, used to control electronically the spurt duration.

In some previous studies [2], [8], fuel injectors without nozzle attachments have been used. However, we have observed that it has also been reported [8] that fuel injectors without nozzles may produce hollow cone sprays, which would undoubtedly induce inhomogeneous heat extraction that would be undesirable for applications of CSC in dermatologic laser surgery. For this reason, the present study was carried out using only fuel injectors with nozzle attachments. Four straight-tube nozzles with two different lengths ($L_N$) and two different inner diameters ($D_N$), and two commercial nozzles used for clinical laser treatment (ScleroPLUS and GentleLASE, Candela Corporation, Wayland, MA) were studied. Table I shows the dimensions of all six nozzles under study and the terms used to identify them.

B. Spray Shape and Droplet Diameter

A progressive-scan CCD camera with a shutter speed of 60 μs was used to photograph the spray shapes. A flash lamp provided illumination gating by 5-μs pulses that “freeze in” the image...
of moving cryogen droplets. Twelve spray shape images were captured for each nozzle at an acquisition rate of 30 frames per second.

An Ensemble Particle Concentration & Sizing apparatus (EPCS, Insitec/Malvern, Worcestershire, U.K.) was used to measure average droplet diameter \(D\). The operating principle of this apparatus is based on low angle laser light scattering (LALLS). The light scattered by spray droplets is collected by a set of 30 concentric ring detectors capable of measuring over a droplet diameter range of 2–200 \(\mu\)m with 3% accuracy, according to the manufacturer. To determine droplet diameter from the input signals, EPCS uses a computer program (RT Sizer, Insitec/Malvern, Worcestershire, U.K.), which is based on the Mie theory of light-particle interactions. A more detailed description of this apparatus may be found elsewhere [15]. The laser beam diameter of this apparatus is normally 10 mm, but using an inverted beam expander, it was reduced to 3.3 mm to obtain a smaller probe cross section. Measurements were taken along the centerline of the spray cone at various distances from the nozzle with a minimum separation of 5 mm. A realistic estimate of the experimental error of our measurements is 15%–20%.

We use Sauter mean diameter (SMD or \(D_{20}\)) as a meaningful quantity of droplet diameter. SMD represents an average droplet diameter with the same volume to surface area ratio as that of the entire spray. For fuel combustion applications, Lefebvre [16] highly recommends the use of this average diameter, since it is least susceptible to a large spread in the droplet diameter distribution. Since water condensation and freezing can affect temperature measurements, these experiments were conducted in a chamber filled with dry air (relative humidity below 5%) [17].

### TABLE I

| Nozzle             | \(L_N\) [mm] | \(D_N\) [mm] | \(L_N/D_N\) |
|--------------------|--------------|--------------|-------------|
| Short wide (SW)    | 31.8         | 1.4          | 23          |
| Long wide (LW)     | 63.5         | 1.4          | 46          |
| Short narrow (SN)  | 31.8         | 0.7          | 46          |
| Long narrow (LN)   | 63.5         | 0.7          | 93          |
| ScleroPLUS TM (SP) | 25.4         | 0.8          | 33          |
| GentleLASE TM (GL) | 18.0         | 0.5          | 35          |

III. RESULTS

Fig. 2 shows photographs of fully developed (i.e., steady-state) sprays produced by the six nozzles under study. For the SW and LW nozzles, cryogen exits the nozzle in...
Fig. 1. (a) Sketch of time of flight procedure (TFP) setup. (b) Normalized valve input and photodetector signals. In this example, the LN nozzle tip was positioned at $z = 60$ mm from the laser beam during a 100-ms cryogen spurt. The solid line represents the photodetector signal, and the dashed line represents the valve input signal. The time interval between the first signs of intensity attenuation of the photodetector signal and that when the signal stalls around an average lower value is referred to as spray development time ($t_d$), which is 20 ms for this example.

Fig. 2. Photographs of cryogenic spray shapes produced by the nozzles. Length of field of view is 17 mm.

a jet-like fashion [Fig. 2(a) and (b)], compared with the SN and LN nozzles, for which a more pronounced cone angle and more finely atomized spray is noted [Fig. 2(c) and (d)]. Although the influence of $L_N$ on spray shape is less apparent than the effect of $D_N$, the somewhat higher intensity of the reflected light seems to indicate a denser spray core with longer nozzles. Photographs of the SP and GL sprays show large differences in their patterns [Fig. 2(e) and (f)]. However, the SP handpiece housing partially obstructs the spray causing some droplet scatter [Fig. 2(e)]. When the housing is removed, the spray shape is similar to that of SN, LN, and GL nozzles. Based on these photographs, it appears reasonable to distinguish between “jet-like” and coarsely atomized sprays, produced by wide nozzles with $D_N = 1.4$, and the cone-like, finely atomized sprays produced by narrow nozzles with $D_N = 0.5–0.8$ mm.

Fig. 3 shows SMD measurements using the EPCS instrument. Solid and hollow symbols illustrate results for the wide and narrow nozzles, respectively. Error bars representing standard deviations of a total of three to five measurements at the same $z$ are included on each data point. The first characteristic noted is that there is practically no variation in $D$ between the wide nozzles, and between the narrow nozzles, within the range of $z$ measured. A two-sample independent $t$-test between the measurements of the wide and narrow nozzle sprays yields a P-value <0.01, demonstrating that the difference in $D$ of these two spray groups is statistically significant at a 95% level. These measurements confirm the existence of two atomization patterns, as suggested above: larger droplet diameters (10–15 μm) produced by the wide nozzles, and smaller droplet diameters (2–6 μm), produced by the narrow nozzles. Also shown in Fig. 3 (dashed lines) are the droplet diameters predicted using a single-droplet evaporation model [11]. As may be seen, the model predictions of $D$ describe the experimental data reasonably well for the wide nozzles at $z > 100$ mm. The rather large discrepancy at shorter $z$ and, in particular, with the narrow nozzles is discussed in the following section.

Fig. 4 shows results of the TFP experiments performed with the four straight-tube nozzles, at $z = 3, 33, 63, 123,$ and 153 mm. At each location, eight spurts were averaged to
In contrast, there was a large mismatch between the measured and calculated values. A two-sample independent t-test between the measurements of the wide and narrow nozzle sprays yields a P-value of ~0.01, demonstrating that the difference in T between these two spray groups is also statistically significant at a 95% level. Also shown in Fig. 5 (dashed lines) are the spray temperatures predicted by the single-droplet evaporation model [11], which will be discussed below.

IV. DISCUSSION

Based on photographs of spray shapes (Fig. 2), it is apparent that nozzle length (L_N) does not have a significant impact on overall spray shape, and that nozzle diameter (D_N) ultimately dictates the spray properties, despite the partial spray atomization that the valve itself may induce. These qualitative observations are confirmed by average droplet diameter (Fig. 3) and temperature (Fig. 5) measurements, which show strong similarities between the sprays produced by wide nozzles with D_N = 1.4 mm and, similarly, between those produced by narrow nozzles with D_N = 0.5–0.8 mm.

Using EPCS, it was not possible to obtain reliable diameter measurements closer than 90 mm from the tip for the wide nozzles (Fig. 3), most likely because the spray density was too high [Fig. 2(a) and (b)]. In contrast, for narrow nozzles, it was possible to obtain diameter measurements as close as z = 15 mm, because such sprays were better atomized [see Fig. 2(c) and (d)]. The D(z) of narrow nozzle sprays clearly show a maximum at 60–70 mm, and a twofold increase in D within the first 50 mm from the nozzle tip. This behavior suggests the presence of droplet coalescence, as observed earlier in liquid and metal sprays [18], [19]. Similarly, the experiments with wide nozzles indicate a slight increase in D, or at least a tendency for that value to remain constant in the range of z from 90 to 150 mm. Interestingly, the ratio between the nozzle diameters of the SN (or LN), and GL is 1.4, yet there is no significant difference in their spray shapes, droplet diameter, and temperature. Alternatively, the slightly larger ratio between the narrow and wide nozzle diameters (2.0) introduces substantial differences in spray shapes, droplet diameter, and temperature, indicating that there is a critical nozzle diameter between 0.8 and 1.4 mm where a large change in the atomization pattern occurs.

The dashed curves shown in Fig. 3 are predictions of the single-droplet evaporation model [11], which requires the input of the initial average droplet diameter (D_0), initial velocity (V_0), and initial temperature (T_0). These parameters must be selected to match the experimental data or deduced from theoretical assumptions. For these experiments, a value of D_0 = 25 μm for the wide nozzle sprays produced the best fit within z = 90–200 mm. In contrast, there was a large mismatch for the narrow nozzle sprays, regardless of the value of D_0 chosen (Fig. 3). This is a clear indication that the single-droplet evaporation model is not sufficiently accurate to describe droplet diameter for all sprays under the conditions used for our studies.

Using fast photography, Pikkula et al. [8] reported SMD measurements for various nozzles. Exact comparisons of average droplet sizes are difficult since nozzles are not the same, except for the SP nozzle. For this nozzle, their reported SMD value...
is 38.3 µm at z = 40 mm, while our SMD measurement is 4.46 ± 0.6 µm (≈ 9 times smaller). We believe that the difference could be due to the fact that the photographic images used by Pikkula et al. were only resolving droplet diameters larger than 9.5 µm. According to our data, this size limit leaves out the lower half of the total range of droplet diameters measured by our EPCS system. Also, since their measurements focused on the centerline of the spray, the large fraction of smaller droplets that exist near the periphery could not be captured, leading to an overestimated droplet diameter.

Although not shown here, droplet size distributions measured by EPCS showed a small fraction of large droplet diameters (> 25 µm) for the wide nozzle sprays, while they were almost nonexistent for narrow nozzle ones. This is in accordance with the spray photographs, which show a few large droplets near the narrow nozzle sprays’ periphery [Fig. 2 (a) and (b)] and, also, with the larger SMD values measured for the wide nozzles as compared to those of the narrow nozzles.

It should also be noted that there is a small effect of \( L_N \) on droplet diameter for both the wide and narrow nozzles (Fig. 3), where \( D \) is smaller for shorter nozzles. This is also supported by the TFP velocity measurements (Fig. 4), where \( V_{1,2} \) is always greater for shorter nozzles. Although the cryogen vessel pressure is constant, \( V_0 \) can differ between nozzles of the same \( D_N \) since the shorter nozzles induce less drag on the cryogen flows, yielding a higher \( V_0 \) (Fig. 4). Higher velocity droplets enhance the evaporation rate due to increased convection, thus yielding smaller droplet diameters.

Overall spray measurements such as \( D \) and \( V_{1,2} \) provide insight into the mechanisms of cryogenic spray atomization and evaporation. However, in order to understand in more detail phenomena such as droplet coalescence, it is necessary to observe the temporal and local variations in the spray properties. For this purpose, we have conducted preliminary measurements of local droplet velocity using a phase Doppler particle analyzer (PDPA, TSI Inc., St. Paul, MN). This apparatus is based on the principles of light scattering interferometry, and permits simultaneous droplet diameter and velocity measurements over a small (~ 1 mm³) probe volume formed by the intersection of two off-phase laser beams of the same wavelength. A more detailed description of this apparatus can be found elsewhere [20].

In Fig. 6, PDPA velocity measurements taken at various locations across the spray cone at \( z = 60 \) mm are shown for four nozzles (SW, SN, SP, and GL). Each measurement represents a local average of the axial velocity component \( (V_z) \) of \( \sim 10^3 \) droplets in a fully developed spray. The large velocity gradient across the spray cone is accompanied by a noticeable shift in the velocity distribution, as shown by the measurements carried out at the spray cone centerline \( r = 0 \) [Fig. 7(a)] and near the periphery \( r = 8 \) mm [Fig. 7(b)], both at \( z = 60 \) mm and for the SN nozzle. The velocity distribution at \( r = 0 \) is practically symmetric, centered at \( \sim 42 \) m/s with a spread of \( \pm 20 \) m/s, while that at \( r = 8 \) mm is markedly skewed, with a maximum at \( \sim 2 \) m/s and a spread of \( -7 \) and \( +18 \) m/s. Most distinctly, a relatively large fraction of droplets show negative \( V_z \) near the cone’s edge, which demonstrates the presence of recirculation zones. Although not conclusive, note that these measurements
were taken at $z = 60$ mm, which is the same distance at which $D$ is maximal for this nozzle (Fig. 3). It is likely that the recirculation zones aids to collisions and coalescence of droplets, as described earlier. We are presently conducting systematic studies to address these issues.

The variation of $T(z)$ for all nozzles shown in Fig. 5 is very consistent with the model predictions (dashed lines). The much poorer match for $D(z)$ suggests that the temperature variation is rather insensitive to variations in $D(z)$. For more finely atomized sprays, which have a larger surface area, removal of latent heat of vaporization from the droplets is more effective, and thus lower temperatures are achieved. In the single-droplet evaporation model, $T_0$ is adjusted until $T(z)$ best fits the experimental data in the range of $10 < z < 100$ mm. At $z > 100$ mm, the temperature data are somewhat higher than those predicted by the model, likely indicating release of latent heat from water vapor deposited on the thermocouple bead [21]. Indeed, signs of condensation and frost formation on the bead were noted at $z > 100$ mm, despite performing the measurements in a dry atmosphere with relative humidity below 5%. For wide and narrow nozzles, the best fits are found at $T_0 = -26 \, ^\circ C$ and $-29 \, ^\circ C$, respectively, yielding best predictions for distances up to 90 or 120 mm, where a sudden rise in temperature indicates complete droplet evaporation, in perfect agreement with experimental results (Fig. 5).

Note that the initial temperature estimate ($T_0$) for wide nozzles coincides with the cryogen boiling temperature ($T_b = -26.2 \, ^\circ C$), which is the expected temperature after sudden expansion through the valve, provided that evaporation within the hose and nozzle are negligible. Alternatively, the somewhat lower $T_0$ estimated for narrow nozzles ($-29 \, ^\circ C$) may indicate some evaporation within the nozzle and consequently better atomization at the nozzle exit. Such an interpretation is consistent with the spray photographs (Fig. 2), which show finer atomization with the narrow nozzles.

The present results relate to recent studies, which demonstrated a significant difference between the interface heat transfer coefficient ($h$) values obtained with the two distinctive spray patterns discussed in this work, namely, values around 10 800 and 7200 W/m$^2$·K for the SW and SN nozzles, respectively [14]. Verkruysse et al. [12] attributed similar differences to a buildup of a liquid cryogen layer on the sprayed surface, which—being thicker for narrow nozzle sprays—impairs heat extraction efficiency more than with the wide nozzle sprays. Based on fast-flashlamp photography of sprays and preliminary surface heat flux ($q$) measurements, the authors hypothesized [12] and later confirmed [13], that the greater impact of larger and faster droplets, such as those produced by wide nozzles, may partly or completely remove the liquid cryogen layer and, consequently, extract heat from the substrate more efficiently.

On the other hand, the more forceful impact of the wide nozzle sprays occurs at the expense of somewhat higher spray temperature, which may partially counteract the enhancement of $q$ achieved with wide nozzles.

In view of these results, we may infer that the surface heat extraction could be enhanced by increasing the current nozzle diameter and/or shortening its length. One should be aware, however, that changes in nozzle design affects other spray properties, such as temperature and area covered, which could pose some practical limitations. Aiming to optimize CSC, we have recently investigated two alternative solutions: fine adjustment of the distance between the nozzle and skin [7], and sequential cryospraying for precise control of the cryogen mass flux [22], [23].

V. CONCLUSION

1) Two distinctive cryogenic spray patterns have been identified for the nozzles under study: coarser (jet-like) sprays, produced by nozzles with inner diameters ($D_N$) of 1.4 mm (wide nozzles); and finely atomized sprays (larger cone), produced by nozzles with $D_N = 0.5$–0.8 mm (narrow nozzles). Nozzle length ($L_N$) has but a small impact on spray shape.

2) A distinctive average droplet diameter $D$, velocity $V_{1,2}$, and temperature $T$ with distance from the nozzle $z$ is observed in each of these two patterns. The wide nozzle sprays show $D$ ranging from 10 to 15 μm, $V_{1,2}$ dropping from 60 to 80 m/s to <5 m/s, and $T$ varying from $-26 \, ^\circ C$ to $-60 \, ^\circ C$ for the range of distances covered (0 < $z$ < 225 mm). The narrow nozzle sprays exhibit $D$ between 2 and 6 μm, $V_{1,2}$ reducing from 15 to 30 m/s to <5 m/s, and $T$ between $-29 \, ^\circ C$ and $-57 \, ^\circ C$, for the range of distances covered (0 < $z$ < 250 mm).

3) A single-droplet evaporation model represents $T(z)$ reasonably well for all nozzles. However, the model does not adequately represent $D(z)$ in the presence of recirculation zones and droplet coalescence, as substantiated by preliminary PDPA velocity measurements.

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