Modeling Cryogenic Spray Temperature and Evaporation Rate
Based on Single-Droplet Analysis

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Abstract

Cryogenic sprays are of interest in dermatology for cooling human skin during pulsed laser treatment of port wine stain (PWS) birthmarks. In the present study, a Phase Doppler Interferometer (PDI) was used to obtain preliminary measurements of mean diameters and mean velocities of tetrafluoroethane (HFC-134a) spray droplets, generated by an atomizing nozzle of a commercial dermatological instrument. Our measurements show significantly smaller diameters than those theoretically predicted for water sprays (by at least one order of magnitude), indicating a high evaporation rate due to the low boiling temperature of the cryogen at atmospheric pressure (T_b=-26°C). Using these preliminary measurements, along with a single-droplet evaporation model, we estimate the mean spray temperature, T_{cryo}, as a function of distance from the nozzle (z). In the model, we assume that a droplet decreases in diameter in accordance with the D^2-law. We further consider deceleration of the droplet due to the drag force. To compute $T_{cryo}(z)$, we incorporate the instantaneous droplet velocity and diameter into a phase-change heat transfer balance, which includes heat convection with the surrounding air. The $T_{cryo}(z)$ predicted by our single-droplet model are in reasonable agreement with the mean temperatures obtained experimentally using a straight-tube nozzle, similar to the commercial atomizing nozzle, in the measured range of z=1.8-12.5 cm. Further studies are underway and will be presented in future papers.

1. Introduction

Cryogenic sprays are of interest in various medical applications, *e.g.*, cooling human skin during laser treatment of port wine stain birthmarks (PWS) [1-2]. Patients are treated with laser pulses that induce permanent thermal damage to the targeted PWS blood vessels [3]. However, absorption of laser energy by melanin causes localized heating of the upper skin layer (epidermis), which may result in scarring or dyspigmentation [4]. By applying a cryogen spurt to the skin surface for an appropriately short period of time (typically 10 to 100 ms), the epidermis is cooled selectively prior to the application of the laser pulse [5]. The objective of this precooling is to achieve the largest possible temperature difference between the epidermis and the deeper targeted PWS blood vessels [6]. To achieve optimal cooling selectivity, it is necessary to control precisely the precooling time [7]. For that purpose, cryogen spray cooling (CSC) offers a suitable solution,

and, indeed, it has been used in commercial dermatological instruments for several years. Nevertheless, the characteristics of the sprays produced by these commercial instruments are not well established yet. In order to improve the cooling selectivity, we need to design atomizing nozzles that provide a high heat extraction rate from the skin surface and, ideally, cover a sprayed area of about the same diameter as the laser beam. For that purpose, we need to have a better knowledge and control of the spray characteristics, such as the mean droplet size, velocity, and temperature, and understand the effect of these parameters on the heat extraction rate from the skin surface.

To date, there have been a significant amount of studies on water sprays, from which empirical correlations have been developed to predict various spray characteristics. For instance, Harmon [8], Tanasawa & Toyoda [9], and Elkobt [10] have proposed correlations to estimate the Sauter mean diameter (SMD) of droplets produced by

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plain orifice atomizers (similar to those used on the cryogenic sprays produced by commercial devices). Unfortunately, none of these correlations account for liquid evaporation, which is likely significant during cryogenic sprays. Studies of droplet evaporation for liquid fuel sprays have led to the development of a simple model, which takes into consideration the evaporation rate of droplets under quiescent conditions ($\partial m/\partial t$) [11]. Moreover, in order to account for the convective effects on droplets in motion relative to the surrounding air, a semi-empirical correction was applied to this model, and the predictions using such corrected evaporation rate ($\partial m'/\partial t$) were found to be in a reasonable agreement with experimental data [12].

Compared to water and fuel sprays, there is very limited work on cryogenic sprays. Ingebo [13-15] worked with two-fluid type nozzles, where a high velocity gas flow is used to atomize the liquid cryogen. He studied the effect of the gas temperature, gas properties, and vaporization on the spray droplet size. However, the nozzles he used are different kinds of atomization devices than those used for PWS treatment.

Our long-term objective is to acquire a better understanding of cryogenic sprays in order to optimize their use in biomedical applications. For that purpose, we are utilizing Phase Doppler Interferometry (PDI), Pulsed Photothermal Radiometry (PPTR), Fast-Flashlamp Photography (FFLP), and other experimental techniques, in order to obtain systematic measurements of mean droplet diameter (D) and velocity (V), as well as the mean spray temperature (T_{cryo}). To this extent, however, we have focused on the characterization of different atomization nozzles to improve current commercial designs. With the aid of PDI and miniature thermocouple sensors, we have obtained estimates of the initial values of mean droplet diameter, velocity, and temperature at the nozzle exit, *i.e.*, D_0, V_0, and $T_{cryo,0}$. With these estimates available, in combination with a fuel evaporation model and phase-change heat transfer analysis, we predict T_{cryo} as a function of the distance z from the nozzle. The benefit of this effort is not only to establish a simple model that relates fundamental spray parameters, *i.e.*, V, D, and T_{cryo}, but also to use it as an indirect way to determine the evaporation rate ($\partial m'/\partial t$).

In summary, the objectives pursued in this work are: (*i*) to obtain preliminary measurements of the initial values D_0, V_0, and $T_{cryo,0}$ of cryogenic sprays produced by commercial-type nozzle devices; (*ii*) to obtain experimental measurements of T_{cryo} as a function of z; and (*iii*) to incorporate the measurements performed in *i* into a single-droplet convection-diffusion model, and compare them to experimental measurements of $T_{cryo}(z)$ performed in *ii*. If this model proves to be appropriate, it may serve as an indirect measurement of $\partial m'/\partial t$, which is an important parameter in the design of new nozzles.

2. Experimental Setup and Procedures

2.1 Phase Doppler Interferometry (PDI)

A PDI (Aerometrics, Sunnyvale, CA, USA) was utilized to characterize a commercial cryogen atomizing nozzle used for PWS treatment in dermatology. Figure 1 shows a sketch of the PDI experimental configuration.
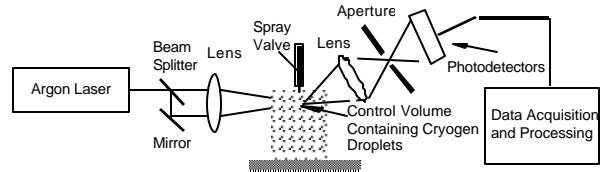

Figure 1. Schematic diagram of a Phase Doppler Interferometer used for droplet size and velocity characterization.

PDI measurements provide information about the size and velocity distributions, statistical mean diameter, size-velocity correlation, and time-of-arrival analysis of droplet size and velocity. However, at this point, we only focused on the mean droplet diameter (D) and mean velocity (V). As a meaningful measurement of particle size, we use the Sauter mean diameter, which represents the size of a droplet with the same ratio of surface area to mass (or volume) as that of the entire spray. This mean diameter is likely to be a more important factor in the characterization of droplet evaporation than the arithmetic mean diameter, because its surface area dependence provides a useful droplet shape dependency and volume dependency and, therefore, may better characterize heat and mass transfer of the droplets. The D and V measurements were taken at three distances from the nozzle (3.3, 5.0 and 8.7 cm), from which D_0 and V_0 were estimated, as will be discussed below.

2.2 Cryogen Temperature Measurements

To measure $T_{cryo}(z)$, we used a type-E thermocouple with a bead diameter of approximately 90 ?m. The estimated response time of this thermocouple is about 5 ms in still water, and its accuracy is ? 1% for the range of temperatures measured. This thermocouple was embedded in a thin epoxy layer (6 by 6 cm^2 and 2 to 3 mm in thickness), not more than 20 ?m below the sprayed surface. The spray is aimed at the epoxy layer surface, which is sequentially moved away from the nozzle. The purpose of this layer is to provide a rigid support and to avoid water condensation and frost formation around the sensor. Figure 2 shows a sketch of the experimental configuration.
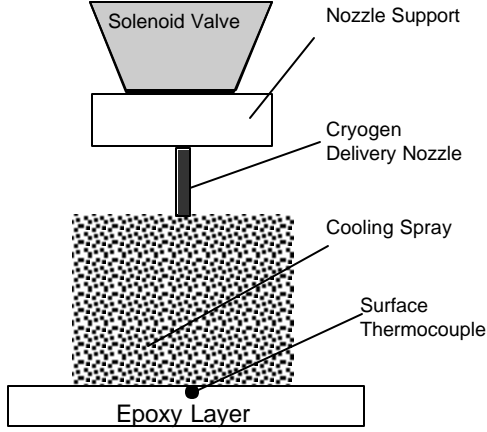
Figure 2. Schematic diagram of T_{cryo} measurements.

Since T_{cryo} may vary as a function of time, we consider only the average readout of the thermocouple after a steady state condition has been established, *i.e.*, during significantly long spurts (typically a few seconds). In this way, the temperature gradients in the vicinity of the thermocouple bead can be considered negligible, and therefore the measured values are representative of the substrate surface temperature.

The substrate surface temperature is not necessarily equal to T_{cryo}; however, given the vigorous evaporative and convective heat transfer that takes place at the droplet-substrate interface, we expect this difference to be very small (in the order of 1 to 2 $^{\circ}$C). This hypothesis is substantiated on recent studies of similar cryogen sprays, which report h_{cryo} values in the range between 2400 to 40000 W/m^{2}K [16-18].

Since at this point we are mostly interested on the general trend of T_{cryo} as a function of distance from the nozzle, rather than tight-accuracy measurements, this way of estimating T_{cryo} is more than adequate. In order to precisely determine T_{cryo}, other parameters, such as the substrate heat conductivity and h_{cryo}, must be determined by other means. We are presently conducting further experimental work in this regard, which will be presented in future studies.

2.3 Droplet Evaporation

According to Ranz and Marshall [12], the evaporation rate of a single droplet subject to forced convection conditions ($\partial m'/\partial t$) is given by:

$$\frac{\partial m'}{\partial t} \approx 2p\, D \left\{\frac{k_g}{Cp_g}\right\} \ln(1 + B_M)\left[1 + 0.3\, Re_d^{0.5}\, Pr_g^{0.33}\right] \quad (1)$$

where k_g and Cp_g, are, respectively, the thermal conductivity and specific heat of the air-cryogen vapor mixture surrounding the evaporating droplet. Re_d and Pr_g are the droplet Reynolds and Prandtl numbers, and $B_M = Y_{c,s}/(1 - Y_{c,s})$ is the mass transfer number, where Y_{cs} is the cryogen mass fraction at the droplet surface, defined as $Y_{c,s} = 1/[1 + (1 - P_?/P_{c,s}) M_a/M_c]$ [19]. M_a, M_c, $P_?$, and $P_{c,s}$ are the air and cryogen molecular weights, and the ambient and cryogen vapor pressure at the droplet surface, respectively. The term in the square brackets in Equation 1 accounts for the forced convection due to the relative motion of the droplet with respect to its surroundings. Without this term, the evaporation rate corresponds to that of a single droplet under quiescent conditions ($\partial m/\partial t$).

2.3.1. Instantaneous Droplet Size (*D*)

Let us assume that the decrease of diameter for a single droplet as a function of time is given by the diameter square-law (D^2-law):

$$D^2 \approx D_0^2 - \beta\, t \quad (2)$$

where D_0 is the initial drop diameter and β is the evaporation constant under quiescent conditions [20].

In terms of the droplet diameter, the evaporation rate ($\partial m/\partial t$) of a single droplet may be expressed as:

$$\frac{\partial m}{\partial t} \approx \frac{p}{2}\, \rho_c\, D^2\, \frac{\partial D}{\partial t}. \quad (3)$$

where ρ_c is the liquid cryogen density. Equation 2 may be introduced into Equation 3 to yield

$$\frac{\partial m}{\partial t} \approx \frac{p}{4}\, \rho_c\, \beta\, D \quad (4)$$

Equation 4 represents the evaporation rate of a single droplet under quiescent conditions, expressed in terms of β. If the forced convection condition at the droplet interface is considered, a convective evaporation constant (β') should replace β, and thus, the corresponding convective evaporation rate, $\partial m'/\partial t$, would be given by Equation 1. Therefore, β' may be obtained by equating the modified Equation 4 with Equation 1:

$$\beta' \approx \frac{8k_g\, \ln(1 + B_M)}{Cp_g\, \rho_c}\left[1 + 0.3\, Re_d^{0.5}\, Pr_g^{0.33}\right] \quad (5)$$

2.3.2 Instantaneous Droplet Velocity (*V*)

To estimate the time evolution of the droplet velocity, we have assumed a deceleration caused by the drag force (F_D) acting on a round sphere of surface area A, *i.e.*, $F_D = ? \rho_c\, C_D\, V^2\, A$, where C_D is the drag coefficient and A the droplet surface area. By incorporating this ex-

pression into the Newton's 2nd law, the instantaneous velocity of the sphere can be computed as:

$$V ? V_0 ? \left\{ \frac{F_D t}{m_0 ? \frac{? m'}{? t} t} \right\} \tag{6}$$

2.4 Mean Droplet Temperature

To compute the mean temperature of a droplet, we first establish an energy balance between times t_0 and t_1, where t_0 represents the initial time where a pure liquid droplet is assumed, and t_1 represents any subsequent time at which part of the droplet has evaporated. Therefore:

$$Q_0 = Q_{1,l} + (Q_{1,g} + Q_{evap}) - Q_{conv,} \tag{7}$$

where:

$Q_o = m_o Cp_{,0,l} T_o$, is the total thermal energy of a pure liquid cryogen droplet.

$Q_{1,l} = m_{1,l} Cp_{1,l} T_1$, is the total thermal energy of the remaining liquid droplet after time t_1.

$Q_{1,g} = m_{1,g} Cp_{1,g} T_1$, is the total thermal energy of the cryogen vapor after time t_1.

$Q_{evap} = m_{1,g} L$; is the latent heat driven out by the evaporation process, where L is the latent heat of vaporization.

$Q_{conv} ? ?h_d A ?T_? ? T_1 ? dt$ is the heat transfer between the droplet and the atmosphere, where h_d is the heat transfer coefficient at the droplet-surface interface.

Expressing the energy balance in terms of temperatures T_0 and T_1 and solving for T_1 yields,

$$T_1 ? \frac{m_0 Cp_{0,l} T_0 ? ?m_0 ? m_{1,l} ?L ? ?h_d A ?T_? ? T_1 ? dt}{m_{1,l} Cp_{1,l} ? ?m_0 ? m_{1,l} ? Cp_{1,g}} \tag{8}$$

The coefficient h_d may then be expressed in terms of the droplet Nusselt number (Nu), which for forced convection acting on a single droplet is defined as:

$$Nu ? \frac{h_d D}{k_g}. \tag{9}$$

Substituting h_d in Equation 8 using Equation 9 yields

$$T_1 ? \frac{m_0 Cp_{0,l} T_o ? ?m_0 ? m_{1,l} ?L ? ?Nu k_g ? D ?T_? ? T_1 ? dt}{m_{1,l} Cp_{1,l} ? ?m_0 ? m_{1,l} ? Cp_{1,g}} \tag{10}$$

According to Ranz and Marshall [12], Nu may be ex-

pressed by the following correlation:

$$Nu ? 2 ? 0.6 Re_d^{0.5} Pr_g^{0.33} \tag{11}$$

Equation 10 allows us to estimate the mean droplet temperature at time t_1, as a function of D_0, V_0, and T_0. Finally, in order to represent this temperature as a function of the distance from the nozzle, we compute the distance that a single droplet has traveled after time t_1 by integrating Equation 6 over time, $i.e.$, $z = ?_0^{t_1} V \, dt$. Evidently, the mean droplet temperature, T_1, then represents the mean spray temperature, T_{cryo}, and the initial droplet temperature, T_0, represents the initial spray temperature, $T_{cryo,0}$, $i.e.$, the temperature at the exit of the nozzle.

3. Results

To indicate the magnitude of droplet evaporation in cryogenic sprays, we used PDI to obtain measurements of the SMD of sprays produced by a commercial atomizing nozzle (Candela DCD[TM], Wayland MA), and compared the results to the expected SMD predicted for water sprays produced by similar straight-tube nozzles. The results are shown in Figure 3.
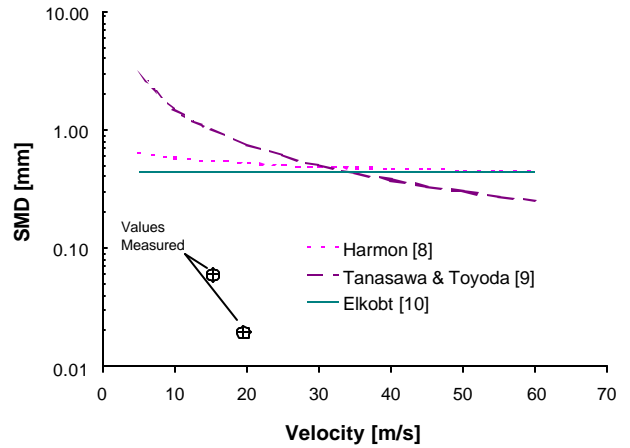

Figure 3. Sauter mean diameter as a function of jet velocity according to the values predicted by correlations applicable to water spays [8-10]. Preliminary measurements of cryogen sprays using PDI are also shown for comparison.

The two data points represent our preliminary results, and the lines represent the predicted values according to various correlations [8-10]. As noted, the Sauter mean diameter measured for a given V is smaller than that expected for water by at least one order of magnitude, indicating a significant reduction in droplet diameter due to the high evaporation rate of cryogenic sprays at atmos-

pheric pressure.

Figure 4 shows the count number distribution for the droplet diameter (a) and the axial velocity (b) measured for the Candela DCD^{TM} device.
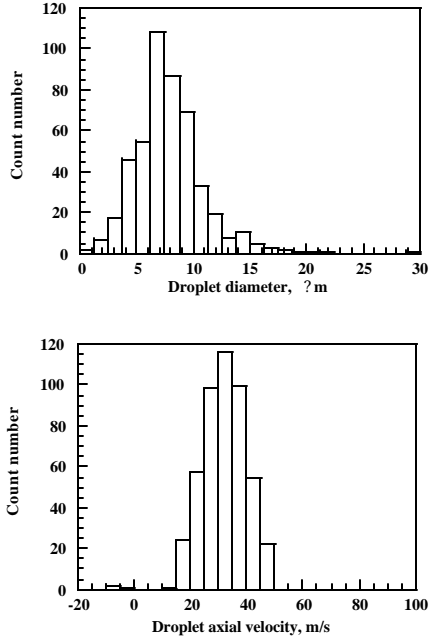
Figure 4. Count number distribution of droplet diameter (a) and velocity (b) using PDI at a distance of $z = 3.3$ cm from the nozzle exit.
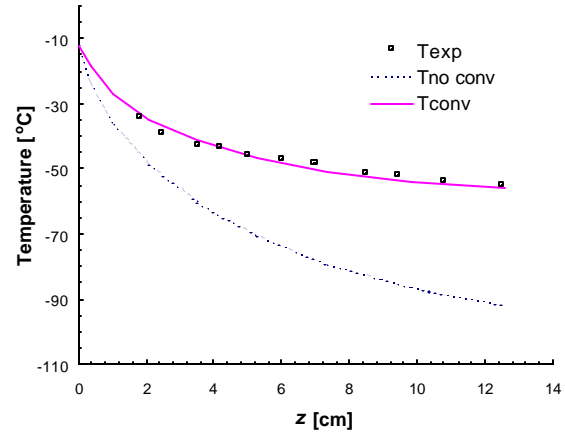
Figure 5. Measurements and model prediction of the spray temperature (T_{cryo}) as a function of distance from the nozzle (z). The initial values used for the computations were: V_0=35 m/s, D_0=25 ?m, and T_0= -12 $^{\circ}$C.

Due to space limitations, the closest measurement was taken at 3.3 cm from the nozzle tip. The D and V at that location are 9.8 ?m and 32.2 m/s, respectively. Based on these and other measurements carried out at 5.1 and 8.9 cm (not shown), it was estimated that a good approximation to the value of V_0 at the nozzle exit (z=0), was 35 m/s. The D_0 value suggested by our preliminary measurements was about 7 to 9 ?m. However, if this value was incorporated into the model, the predictions indicated complete droplet evaporation at only 0.35 cm away from the nozzle exit. Using a value of $D_0 = 25$?m instead, we obtained a reasonable agreement with the model. In recent studies carried out with an optical particle-sizer measuring device (EPCS, Malvern, Worcestershire, UK) we have found out that for this kind of straight-tube nozzles, there are at least two peaks in the size distribution of the droplets. The first one around 10 ?m, and the second around 200 ?m. We were not able to measure the second peak with the PDI measurements because the range selected did not count particles sizes beyond 100 ?m. However, if the whole range is considered, the Sauter mean diameter is around 25 ?m, which is the value of D_0 we used in our model.

Figure 5 shows the temperature measurements of the thermocouple embedded in the epoxy layer as a function of z (symbols), as well as the computations of T_1 (equal to T_{cryo}) resulting from Equation 10 (solid line). T_0 (equal to $T_{cryo,0}$) is another free parameter, which needs to be either measured or estimated in the model. In this case, T_0 was adjusted until it matched the experimental data, which is

As can be seen, the solid curve reproduces reasonably well the experimental data (symbols). Finally, the computations of T_1 without consideration of the convective term in Equation 7 are also plotted for comparison (dotted line). This curve predicts a much faster decrease of the spray temperature, compared to experimental results.

The reasonably good fit of Equation 10 with the experimental data gives us a good certainty that the convective evaporation rate, $\partial m'/\partial t$, has been appropriately considered. Figure 6 shows the variation of $\partial m'/\partial t$ as a function of z, over approximately the range of distances covered in the experiment.
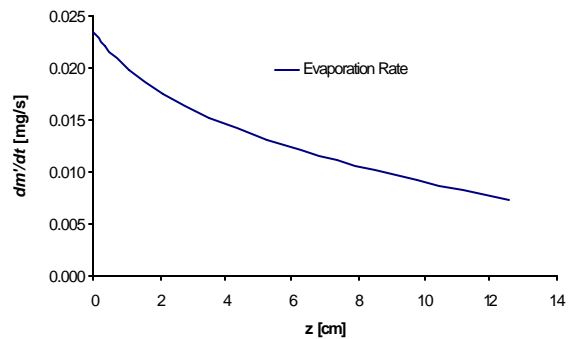

Figure 6. Variation of $\partial m'/\partial t$ as a function of z (computed from Equation 4).

5. Conclusions

1. The D^2-law evaporation model, widely used in fuel spray analyses, can be adequately applied to the analysis of mean droplet size of cryogenic sprays pro-

produced by straight-tube type nozzles.

2. The incorporation of the D^2-law, and a droplet deceleration term, into a phase-change heat transfer balance for a single droplet reproduces quite accurately the temperature decrease of the cryogenic spray produced by a straight-tube type nozzle.

3. The success of Equation 10 in reproducing the experimental data represents an additional tool to determine the evaporation rate $\partial m'/\partial t$ of cryogenic sprays provided estimates of the initial values D_0, V_0 and T_0.

Nomenclature

Cp	specific heat [J/kg K]
D	droplet diameter [$?$m]
k	thermal conductivity [W/m K]
L	latent heat of vaporization [J/kg]
m	mass [mg]
M	molecular weight [g/mol]
$P_{c,s}$	cryogen vapor pressure at the surface [Pa]
V	mean velocity [m/s]
$?$	density [Kg/m^3]
$??$	evaporation constant [m^2/s]

Subscripts

a	air
c	cryogen
g	relative to the air-cryogen vapor mixture
l	liquid
s	relative to the surface
$??$	relative to ambient
0	initial
1	relative to time t_1

Superscripts

'	subject to convective conditions

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