# Design and Construction of Experimental Device to Study Cryogen Droplet Deposition and Heat Transfer

Matthew D. Keller<sup>1,2</sup>, Guillermo Aguilar<sup>2,3</sup>, and J. Stuart Nelson<sup>2,3</sup>

<sup>1</sup>Harvey Mudd College, Claremont, CA 91711 <sup>2</sup>Beckman Laser Institute, University of California, Irvine, CA 92612 <sup>3</sup>Department of Biomedical Engineering, University of California, Irvine, CA 92612

## ABSTRACT

Cryogen spray cooling (CSC) is used to pre-cool the epidermis during laser dermatological procedures such as treatment of port wine stain (PWS) birthmarks. It is known that PWS patients with medium to high epidermal melanin concentrations are at a high risk of epidermal thermal damage after laser irradiation. To avoid this complication, it is necessary to maximize CSC efficiency and, thus, essential to understand the mechanical and thermal interactions of cryogen droplets with the sprayed surface.

It has been observed that cryogen sprays exhibit droplet rebound as droplets impinge on the skin surface. Studies of water droplet impact on hard surfaces have shown that droplet rebound may be suppressed by dissolving small amounts (a few percent) of diverse polymer or surfactant solutions prior to atomization. To investigate the possibility of suppressing the rebound of cryogen droplets in a similar way, we have constructed a device that allows observation of the impact, spreading, and rebound of individual water and cryogen droplets with and without these solutions, and their influence on cryogen/surface dynamics and heat transfer. Our preliminary studies show that dissolving a 4% non-ionic surfactant in water reduces droplet rebound and thickness of the residual liquid layer. The maximum spread of water droplets after impact can be described within 20% accuracy by a previously developed theoretical model. The same model provides an even more accurate prediction of the maximum spread of cryogen droplets. This study will aid the analysis of future results and design conditions of new studies, which will recreate conditions to determine if added surfactant solutions suppress droplet rebound and lead to improved CSC efficiency.

### 1. INTRODUCTION

During dermatologic laser therapy epidermal melanin represents an "optical barrier" through which light must pass to reach the targeted dermal chromophore. Epidermal melanin absorption of laser light causes localized heating which, if not controlled, may lead to permanent complications such as scarring and dyspigmentation. The risk of non-specific epidermal thermal injury has been effectively minimized by the introduction of skin cooling procedures, such as cryogen spray cooling  $(CSC)^{1,2,3}$ , in which the epidermis is cooled selectively by applying a short cryogen spurt (10-100 ms) onto the skin surface while minimally cooling the deeper targeted chromophores<sup>4</sup>. Shortly after spurt termination a short laser pulse (0.5-40 ms) is aimed at the same area of skin.

Despite the improvements in dermatologic laser therapy, CSC commercial nozzles are inadequate to provide sufficient epidermal protection, particularly for skin types with relatively high epidermal melanin concentrations (e.g., skin types III and higher). Suboptimal CSC may be attributed, in part, to the forceful impact of cryogen spurts on the skin surface, which causes significant droplet rebound and splatter of the liquid cryogen layer that is formed shortly after spurt initiation. Therefore, to enhance CSC efficiency it is necessary to understand the dynamic phenomena of cryogen droplet deposition on skin (impact, spreading, and rebound) and heat transfer thereof. Moreover, it is necessary to seek alternative mechanisms to suppress droplet rebound and splatter of liquid cryogen and, thus, enhance heat extraction through the skin surface during CSC.

Although various studies of droplet spread and rebound have been carried out for water and other Newtonian fluids<sup>5,6,7</sup>, as well as for atomization of liquid films<sup>8,9</sup>, few studies have recognized that reducing surface tension of the cooling liquid enhances spreading and suppresses droplet rebound during spray deposition. Qiao and Chandra<sup>10</sup> observed that the addition of 100 to 1000 ppm of sodium dodecyl sulphate (surfactant) to water droplets reduced the liquid/solid

static contact angle and reduced the lifetime of droplets, resulting in enhanced heat transfer. A more recent study by Bergeron et al<sup>11</sup> showed that 100 ppm of polyethelene-oxide were sufficient to suppress the rebound of a single water droplet from a hydrophobic surface. More recently, Pikkula et al.<sup>12</sup> used cryogen soluble surfactants to reduce cryogen film thickness during CSC and improve heat transfer.

In this study, we describe the initial steps taken towards the construction of an experimental device to study the impact, deposition, rebound, and heat transfer of cryogen droplets used commercially for dermatologic laser surgery (tetrafluoroethane: R-134a). The objective is to determine whether or not the addition of small amounts of either polymer or surfactant solutions can suppress rebound and splattering of liquid cryogen and, thus, enhance efficiency of heat transfer through the skin surface.

# 2. MATERIALS AND METHODS

### **Pressure Chamber**

In order to keep the droplets in a liquid state it was necessary to build a vessel that could be pressurized to the cryogen (R-134a) saturation pressure (670 kPa at 25°C). The construction of the device was based on a cross-shaped four-way PVC union. This particular configuration was ideal as it provided room for an impact surface in the base, delivery system above, and windows at the sides to allow a light source and camera (see Figure 1). PVC was also chosen for easy handling and because it is rated for more than twice the saturation pressure of R134-a.

Four modified PVC end plugs were placed and cemented into each branch of the main piece. The bottom plug was a female-threaded reducer which allowed us to insert the desired impact surface and tighten it adequately to seal. The top plug was fitted with several modifications: the cryogen delivery system (described below), valve leading to the pump for pressurization, pressure gauge and safety valve, temperature sensor, and device to wipe off the impact surface without releasing the chamber pressure. The side plugs were fitted with polycarbonate windows to allow for imaging of the droplets. One side provided a light source while a high-speed camera was placed on the other side.

### **Impact Surfaces and Sensors**

Three impact surfaces were constructed using epoxy because it provides a smooth surface and has thermal properties similar to those of human skin. The first is merely a smooth surface to provide good quality images of the droplet impact. The other two surfaces include temperature sensors to measure heat extraction through the surface. The first of these impact surfaces consists of a thin foil on top of an epoxy substrate. A miniature thermocouple (50-70  $\mu$ m bead diameter) was placed between the foil and the substrate to measure rapid changes in temperature due to heat exchange between the cryogen droplet and the surface. The second surface of this kind consists of two small silver discs embedded in the epoxy. Each disc has a thermocouple soldered to the back of it. This sensor allows simultaneous measurements of surface temperatures at two positions across the impact surface.

### **Cryogen Delivery System**

To deliver individual droplets into the pressure chamber, a dropper system was implemented, which consisted of a small stainless steel tank that could be filled with cryogen or water. The tank was connected to a fine control needle valve, which allowed liquid to flow through to a small nozzle made of stainless steel hypodermic tubing with an inner diameter of  $250 \,\mu\text{m}$ . The tank was connected to an external pump to increase pressure within the device (see Fig. 1).

### Imaging

A high speed camera (Photron Fastcam PCI 10K, Itronics, Westlake Village CA) was used to acquire digital images of droplet impact and ensuing behavior. All image sequences were captured at a rate of 500 frames per second.

### Procedure

The above described device was assembled and securely tightened to avoid leaks. The small tank was then filled with either water or cryogen to provide test droplets. For the experiments involving water droplets, the chamber was pressurized with air up to the desired pressure. For the cryogen droplet experiments, the chamber was pressurized with cryogen vapor to saturate the local atmosphere within the device and minimize evaporation. This was done by filling

the chamber with cryogen gas up until the chamber was at equilibrium pressure with the supply tank. Then, pressurized liquid cryogen was sprayed into the chamber until it stopped evaporating.

Once the chamber was pressurized, a hand pump was used to increase the pressure within the small tank to about 138 kPa (20 psi) above the chamber pressure. We then slowly turned the fine control valve to allow a drop to begin to form on the end of the nozzle. As the drop began to grow, the camera was triggered to begin image acquisition.



**Figure 1**: Schematic of experimental setup including: small tank (1), nozzle (2), PVC chamber (3), impact surface (4), chamber pressure control valve (5), safety valve (6), light source (7), and camera (8).

#### **Droplet Spread Model**

A theoretical model<sup>13</sup> that predicts the maximum spread of impacting non-evaporating droplets was tested against our experimental data. The model is based on various energy balances between the different stages a droplet goes through before, during and after impacting on a hard surface. The following equation describes the maximum diameter  $(d_m)$  that a given droplet of diameter D can reach after impact as a function of three parameters: static contact angle  $(\theta)$ , Reynolds number (Re) and Weber number (We):

$$\left[\frac{1}{4}(1-\cos\theta)+0.2\frac{We^{0.83}}{Re^{0.33}}\right]\left(\frac{d_m}{D}\right)^3 - \left(\frac{We}{12}+1\right)\left(\frac{d_m}{D}\right) + \frac{2}{3} = 0.$$
 (1)

Re is a commonly used dimensionless number in fluid mechanics which represents the ratio of inertial to viscous forces acting on a moving fluid We is another dimensionless number which represents the ratio of inertial to surface tension forces. These numbers are defined as:

$$Re = \frac{\rho VD}{\mu}$$
(2)

$$We = \frac{\rho V^2 D}{\sigma}$$
(3)

where  $\rho$ , V, D,  $\mu$ , and  $\sigma$  represent droplet density, impact velocity, diameter, dynamic viscosity, and surface tension, respectively. V was computed using the following analytic equation:

$$V = \left\{ \frac{1}{k} mg \left( 1 - e^{\frac{2k}{m}(y - y_0)} \right) \right\}^{1/2}$$
(4)

where  $k = 2 \times 10^{-4}$  kg/m is the air resistivity,  $y \cdot y_0 = 65$  mm was the distance between the nozzle tip and the impact surface, *m* the droplet mass and g = 9.81 m/s<sup>2</sup> the gravity acceleration constant.

## 3. RESULTS

#### Water

Images shown in Fig. 2 illustrate the impact of a water droplet on a rigid epoxy surface. The classical spread, ring formation, re-attachment, and an incipient but appreciable rebound are observed in frames corresponding to 14 - 18 ms.



Figure 2: Impact sequence of a water droplet

#### Water with surfactant

A 4% in weight of proprietary non-ionic surfactant (by Akzo Nobel Inc., Chicago, IL) was added to water to decrease droplet surface tension. Figure 3 shows a sequence of images from an instant before impact (0 ms) up until a time when no appreciable motion was observed (86 ms). Apparently, the decreased surface tension and/or viscoelastic properties of this solution cause an increased spread of the droplet, a thinner cryogen layer on the surface, and a significant suppression of the droplet rebound.



Figure 3: Impact sequence of a water droplet with surfactant

# Cryogen

Fig. 4 shows the sequence of an impacting cryogen droplet. For both water and cryogen droplets, the same nozzle and nozzle-to-surface distance were used. These images were taken with the chamber pressure at 593 kPa (86 psi) and dropper pressure at 731 kPa (106 psi). The cryogen droplet behavior is very similar to that of water with surfactant. It is important to note that because the surface tension of cryogen is less than water, the initial cryogen droplet diameter (*D*) is smaller that that of water (as noted in the image at 0 ms). Therefore, the impact velocity of the cryogen droplet is smaller, as seen from Eq. 4.



Table 1 lists the density  $\rho$ , viscosity  $\mu$ , surface tension  $\sigma$ , measured static contact angle  $\theta$ , impact velocity V, Re, We, droplet diameter D, maximum spread  $d_m$ , and  $d_m/D$  of water, water + surfactant, and cryogen for the three experiments described above.

Table 1	1
---------	---

	$\rho$ [kg/m <sup>3</sup> ]	μ[Pa s]	$\sigma$ [kg/s <sup>2</sup> ]	θ	<i>V</i> [m/s]	Re	We	<i>D</i> [mm]	$d_{\rm m}$ [mm]	$d_m/D$
Water	999.8	$10 \times 10^{-4}$	0.073	83	0.83	2734	31.2	3.3	7.0	2.12
Water +	999.8	$10 \times 10^{-4}$	0.049	34	0.74	2135	32.4	2.9	8.5	2.93
Surf										
R134a	1206.0	2×10 <sup>-4</sup>	0.0085	31	0.43	4576	47.2	1.8	9.8	5.44

#### **Droplet Spread Model**

Fig. 5 shows model predictions (solid lines) of the normalized maximum spread  $(d_m/D)$  as a function of Re. Both curves were computed using Eq. 1 for a We = 30. This We number is an approximate value close to that computed for both water droplet experiments shown in Table 1. The corresponding  $\theta$  measured for each droplet was also used as input to the model. As one would expect, the spread is larger for smaller static contact angles and increases with larger Re. Symbols represent the measured  $d_m/D$ , which correspond to the values listed in the last column of Table 1. As seen in both water experiments, there is an overprediction of about 20% of the maximum spread that an impacting droplet may reach.



Figure 5: Normalized maximum spread of water and water with surfactant droplets as a function of Re for a constant We = 30. Solid lines are model predictions. Symbols are measured values.

Fig. 6 shows model predictions (solid line) of the normalized maximum spread  $(d_m/D)$  as a function of Re for a We = 50, which is close to that computed for the cryogen droplet experiment, as shown in Table 1. The symbol represents the measured  $d_m/D$ , which is also listed in the last column of Table 1. As noted, the model prediction is almost identical. This result is a good indication that the impact and spreading of cryogen droplets can be accounted for by this model. Moreover, the energy balances which led to the development of Eq. 1, could be used in future studies to predict if cryogen droplet rebound may occur under actual CSC conditions.



**Figure 6:** Normalized maximum spread of cryogen droplets as a function of Re for a constant We = 50. Solid line is the model prediction and the symbol is the measured value.

#### 4. **DISCUSSION**

During CSC it is important to note that subcooled cryogen droplets evaporate by two distinct mechanisms: (1) evaporation in flight, as droplets try to reach thermodynamic equilibrium in an open atmosphere; and (2) evaporation by heat transferred to the droplets by the sprayed object. Whereas the latter mechanism makes CSC no different from other more studied spray cooling scenarios (e.g., water spray cooling at atmospheric conditions), the former is a distinct characteristic of refrigerants and cryogens, which makes the phenomena even more complex and difficult to analyze.

One concern in our experiments stemmed from the different surface tensions of water and cryogen. The size of the droplet when it detaches from the nozzle is determined by the surface tension of the liquid and outer diameter of the nozzle, according to the following equation:

$$D = \left(\frac{6d_0\sigma}{\rho g}\right) \tag{5}$$

where  $d_0$  is the outer diameter of the nozzle. The droplet detaches once its weight becomes greater than the force due to surface tension holding the drop to the tip of the nozzle. The droplet size also affects the impact velocity (*V*), as it is evident from Eq. 4. Both of these properties affect the droplet kinetic energy at the moment of impact and therefore, the dynamic behavior of the spread and rebound is not necessarily similar for water and cryogen. An appropriate comparison should be made by ensuring that the kinetic energies of water and cryogen droplets are the same at the time of impact which can be achieved by placing the nozzle at different distances from the impacting surface. A new chamber is being built which will allow adjustable nozzle height. This will produce higher *V* and likely will cause the cryogen droplet to rebound.

The addition of small concentrations of surfactant into a purely viscous liquid (e.g., water and liquid cryogen) diminishes its surface tension and, at the same time, introduces a viscoelastic property. Since the model developed by Mao et al does not include any viscoelastic effect, the relatively good match for both water and water/surfactant droplets with the model predictions (Fig. 5) suggests that the surface tension effect is the most dominant property on the droplet spreading dynamics. The slight mismatch could possibly be attributed to the relatively low V and the uncertainty in  $\theta$ . Also, the good match of the measured cryogen droplet spreading and the theoretical model shown in Fig. 6 suggests that droplet evaporation was properly controlled within the chamber. In the future, it will be interesting to study the effect of cryogen evaporation as the chamber pressure is systematically reduced.

Another improvement to the new chamber will be an automatic triggering device to trigger the camera to begin capturing images when the droplet is falling. This will allow for easier data analysis and more consistent images between trials. Also, new studies will use cryogen-soluble surfactants to study their effect on droplet rebound.

### 5. CONCLUSIONS

The experimental setup constructed for this study allowed successful imaging of the impact of individual water and cryogen droplets. It was shown that the addition of a dilute surfactant solution (4%) to water significantly suppresses the droplet rebound. This result is expected to improve the droplet-surface contact and liquid layer spreading which should be accompanied by an increase in heat transfer through the impact surface. Future studies will focus on the measurement of the instantaneous surface heat flux.

Previous experimental evidence shows that the impact velocity of actual cryogen spray droplets can be as large as 30 m/s. However, the impact velocity of the cryogen droplets in these experiments was less than 1 m/s, which is not large enough to induce cryogen droplet rebound. Ongoing work is being done to adapt the experimental conditions so that they more closely resemble those existent in CSC. Once this is achieved, and with the choice of suitable solutions for cryogen sprays, we expect to observe a suppression of the droplet rebound and layer spreading similar to that seen in water droplets in this study.

It is expected that droplet rebound suppression through the use of suitable surfactant solutions will have a significant effect on the cryogen-surface contact and liquid layer spreading. Consequently, a more efficient heat extraction should result and, thus, higher radiant exposures could be used during various dermatologic laser therapies without the risk of inducing excessive epidermal damage.

### ACKNOWLEDGEMENTS

This work was supported by a research grant from the Institute of Arthritis and Musculoskeletal and Skin Diseases at the National Institutes of Health (GM62177 to JSN and HD42057 to GA). Institutional support from the Office of Naval Research, Department of Energy, and the Beckman Laser Institute and Medical Clinic Endowment is also acknowledged. Useful discussion with Drs. John A. Viator, Bernard Choi, and B. Samuel Tanenbaum are greatly appreciated.

### REFERENCES

<sup>1</sup> JS Nelson, TE Milner, B Anvari, BS Tanenbaum, S Kimel, LO Svaasand. Dynamic cooling of the epidermis during laser port wine stain therapy. Lasers Surg. Med. 6S: 48, 1994.

<sup>2</sup> JS Nelson, TE Milner, B Anvari, BS Tanenbaum, S Kimel, LO Svaasand. Dynamic epidermal cooling during pulsed laser treatment of port-wine stain. A new methodology with preliminary clinical evaluation. Arch Dermatol 131:695-700, 1995.

<sup>3</sup> JS Nelson, TE Milner, B Anvari, BS Tanenbaum, LO Svaasand, S Kimel. Dynamic epidermal cooling in conjunction with laser-induced photothermolysis of port wine stain blood vessels. Lasers Surg. Med. 19: 224-229, 1996.

<sup>4</sup> W. Verkruysse, B. Majaron, B.S. Tanenbaum, J.S. Nelson. Optimal cryogen spray cooling parameters for pulsed laser treatment of port wine stains. Lasers Surg. Med. 27: 165-170, 2000.

<sup>5</sup> F.H. Harlow, J.P. Shannon. The splash of a liquid droplet. J. Appl. Phys., 38: 3855, 1967.

<sup>6</sup> G. Trapaga, J. Szekely. Mathematical modeling of the isothermal impingement of liquid droplets in spraying processes. Met. Trans. B, 22B: 901, 1991.

<sup>7</sup> J. Fukai, Z. Zhao, D. Poulikakos, C.M. Megaridis, O. Miyatake. Modeling of the deformation of a liquid droplet impinging upon a flat surface. Phys. Fluids A: 2588-2599, 1993.

<sup>8</sup> A.L. Yarin, D.A. Weiss. Impact of drops on solid surfaces: self-similar capillary waves, and splashing as a new type of kinematics discontinuity. J. Fluid Mech. 283: 141-173, 1995.
<sup>9</sup> M. Al-Roub, P.V. Farrell. Atomization of thin liquid films by droplet impact. Atomization and Sprays 7: 531-

<sup>9</sup> M. Al-Roub, P.V. Farrell. Atomization of thin liquid films by droplet impact. Atomization and Sprays 7: 531-547, 1997.

<sup>10</sup> Y.M. Qiao, S. Chandra. Experiments on adding a surfactant to water drops boiling on a hot surface. Proc. R. Soc. Lond. A V: 453, 673-689, 1997.

<sup>11</sup> V. Bergeron, D. Bonn, J.Y. Martin, L. Vovelle. Controlling droplet deposition with polymer additives. Nature 405: 772-775, 2000.

<sup>12</sup> B.M. Pikkula, Y. Domankevitz, J.W. Tunnell, B. Anvari. Cryogen spray cooling: Effects of cryogen film on heat removal and light transmission. Proc. SPIE 4609: 50-56, 2002.

<sup>13</sup> T. Mao, D.C.S. Kuhn, H. Tran. Spread and Rebound of Liquid Droplets upon Impact on Flat Surfaces. AIChE Journal 43: 2169-2179, 1997.