

Intermittent cryogen spray cooling for optimal heat extraction during dermatologic laser treatment

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Abstract

Fast heat extraction is critically important to obtain the maximal benefit of cryogen spray cooling (CSC) during laser therapy of shallow skin lesions, such as port wine stain birthmarks. However, a film of liquid cryogen can build up on the skin surface, impairing heat transfer due to the relatively low thermal conductivity and higher temperature of the film as compared to the impinging spray droplets. In an attempt to optimize the cryogen mass flux, while minimally affecting other spray characteristics, we apply a series of 10 ms spurts with variable duty cycles. Heat extraction dynamics during such intermittent cryogen sprays were measured using a custom-made metal-disc detector. The highest cooling rates were observed at moderate duty cycle levels. This confirms the presence, and offers a practical way to eliminate the adverse effect of liquid cryogen build-up on the sprayed surface. On the other hand, lower duty cycles allow a substantial reduction in the average rate of heat extraction, enabling less aggressive and more efficient CSC for treatment of deeper targets, such as hair follicles.

1. Introduction

In laser dermatologic surgery and cosmetic treatments, non-specific absorption by epidermal melanin competes with absorption of radiation in subsurface target chromophores. If not controlled, high epidermal temperatures can lead to blistering, dyspigmentation or scarring, which limits the radiant exposure that can be safely applied. For some clinical indications, such as port wine stain birthmarks (PWS), this limitation adversely affects the success of laser therapy in many patients. Pre-cooling of the epidermis with cryogenic sprays permits the safe

application of laser pulses with higher energy (Welch *et al* 1983, Nelson *et al* 1995), leading to an improved outcome in laser therapy of PWS (Nelson *et al* 1995, Waldorf *et al* 1997, Chang and Nelson 1999) and hemangiomas (Chang *et al* 1998, 2001), rhytides (Kelly *et al* 1999), and hair removal. Despite the wide clinical use of cryogen spray cooling (CSC) and considerable research efforts in recent years, our understanding of the involved heat-transfer mechanisms and dynamics remains incomplete. It is therefore not certain whether existing CSC devices have been optimized for their respective applications.

In particular, laser therapy of PWS requires cooling with high spatial selectivity (Nelson *et al* 2000, Verkruyse *et al* 2000b), which necessitates the use of short cryogen spurts (up to 100 ms) with rates of heat extraction from the skin as high as possible. In theory, the heat extraction rate is maximal when the sprayed mass flux matches the evaporation rate at the cooled surface (Estes and Mudawar 1995). When the cryogen mass flux exceeds the evaporation rate, a thin film of liquid cryogen can build up on the skin surface. Due to the relatively low thermal conductivity of the cryogen compound ($0.082 \text{ W m}^{-1} \text{ K}^{-1}$ versus $0.53 \text{ W m}^{-1} \text{ K}^{-1}$ for human dermis), and higher temperature of the film as compared to the impinging spray droplets, such a build-up may significantly impair heat extraction from the skin.

Using fast-flashlamp photography, the accumulation of liquid cryogen on an epoxy tissue model was observed at spurt durations as short as 20 ms (Verkruyse *et al* 2000a). Build-up of a cryogen film with a commercial vascular treatment device (DCD/ScleroPLUS, Candela, Wayland, MA) and laboratory spray nozzles with similar geometries (straight tube, inner diameter $d_n = 0.7 \text{ mm}$) was subsequently reported for epoxy tissue models (Aguilar *et al* 2001a, 2001c, Torres *et al* 2001) and human skin *in vivo* (Nelson *et al* 2000, Torres *et al* 2001).

Verkruyse *et al* (2000a) observed a higher cooling efficacy with wider nozzles ($d_n = 1.4 \text{ mm}$), despite the higher cryogen mass flux, which contradicts the aforementioned theoretical consideration. The authors tentatively concluded that higher-momentum droplets from the wider nozzles penetrated through and/or partly removed the cryogen film, thereby enhancing heat extraction from the substrate. This hypothesis was later confirmed by measurements of spray droplet size and velocity (Aguilar *et al* 2000, 2001a), measurements of steady-state heat extraction (Aguilar *et al* 2001c) and measurements of the average heat-transfer coefficient in a 100 ms CSC spurt for both nozzles, using a metal-disc detector (Svaasand *et al* 2003).

Unfortunately, the high-momentum spray from the wider nozzle induced considerable patient discomfort in preliminary tests, likely to be prohibitive for routine clinical use (Verkruyse *et al* 2000a). Alternative modifications of nozzle geometry, combined with optimization of spraying distance, would thus be required to achieve similar cooling efficacy. However, since such changes often affect other spray characteristics (e.g., spray droplet temperature, sprayed area) (Aguilar *et al* 2000, 2001a), application-specific optimization of the nozzle design remains a nontrivial task.

We investigate below an alternative approach to maximizing the heat extraction rate, with possibly much less effect on other spray characteristics. Using the 0.7 mm spray nozzle, which—apart from depositing too much cryogen—performs well in clinical practice, we attempt to optimize cryogen spray deposition by intermittent spraying, i.e., by applying a series of millisecond spurts with adjustable delays. Our hypothesis is that, if the cryogen film build-up is impairing heat extraction from the sprayed surface, a suitable reduction of mass flux should improve the cooling rate and efficiency.

In addition to reducing the CSC rate and efficiency, the build-up of cryogen film on skin surface has other clinically important disadvantages (Majaron *et al* 2001b). First, it extends

the cooling time over the user-specified spurt duration, reducing control over the treatment parameters. Second, the associated 'spillover' may significantly cool the skin surface adjacent to the treated area, which raises concerns with regard to possible cryo-injury and therapeutic effect of subsequent laser irradiation of the adjacent sites. Third, the consequent formation of frost from ambient water vapour may obstruct the physicians' sight, and scatter the incident laser radiation, thus compromising the light dose delivered into the skin.

In comparison with PWS, laser treatment of deeper subsurface targets (e.g., leg veins, hair follicles) involves even higher radiant exposures and longer laser pulses (Ross *et al* 1999, Nelson *et al* 2000, Zenzie *et al* 2000). Consequently, longer cryogen spurts should be employed, raising concerns with regard to possible cryo-injury (Zenzie *et al* 2000, Tunnell *et al* 2002a). Intermittent spraying offers a practical way to reduce the cooling aggressiveness associated with current CSC devices, allowing their adaptation to novel dermatologic laser applications. A similar idea was earlier explored by Anvari *et al* (1996), but using a very different parameter set (typically 1 s intervals between the 50 ms cryogen spurts, 10–120 s long sequences applied in parallel with continuous-wave laser irradiation), leading to very different temperature evolution in the tissue.

Quantitative assessment of the heat-transfer coefficient during CSC has been a somewhat controversial issue. The reported values range from $40\,000\text{ W m}^{-2}\text{ K}^{-1}$ (Anvari *et al* 1995, 1998) down to $2000\text{ W m}^{-2}\text{ K}^{-1}$ (Torres *et al* 1999, Pikkula *et al* 2000). Discrepancies between results from similar spraying setups indicate inaccuracies of some experimental approaches. In the present study, we use a metal-disc detector, similar to that used by Aguilar *et al* (2001a) and Svaasand *et al* (2003). The detector is comparatively simple and reliable, and allows measurements of heat extraction dynamics with high temporal resolution (Majaron *et al* 2001a).

2. Instrumentation and methods

2.1. Intermittent spraying setup and settings

The cryogen used in our measurements (1, 1, 1, 2 tetrafluoroethane) is FDA approved for use in laser dermatological applications. The compound with a boiling point at $-26.2\text{ }^{\circ}\text{C}$ is kept in a pressurized steel canister at room temperature. A hose connects the canister with an automobile fuel injector, controlled electronically by a function generator and a digital delay generator (DS345 and DG535, by Stanford Research Systems). Attached to the body of the valve is a straight-tube nozzle with an inner diameter of $d_n = 0.69\text{ mm}$. The spraying distance was set to either 50 or 60 mm.

The intermittent spraying sequences used in this study consist of individual spurts with durations fixed at $t_s = 10\text{ ms}$. The delays between each successive spurt (t_d) are varied from 1 to 40 ms, thus varying the duty cycle, $r = t_s/(t_s + t_d)$, from 0.91 down to 0.20 (figure 1). In order to allow a comparison with a continuous 100 ms spray, the number of spurts is adjusted to keep the total duration of the spraying sequence as close to 100 ms as possible. The 10 ms spurt duration allows a large selection of duty cycle values within the $\sim 100\text{ ms}$ time interval—the maximal CSC duration currently used in clinical practice. Shorter t_s , however, might compromise other spray characteristics, since it can take up to 20 ms for the cryogen spray to fully develop (Aguilar *et al* 2000).

2.2. Measurement of the heat extraction rate

Heat extraction rate during CSC was measured using a custom device (described earlier in Majaron *et al* 2001a). The device consists of a thin disc made of high-purity copper (Cu-110,

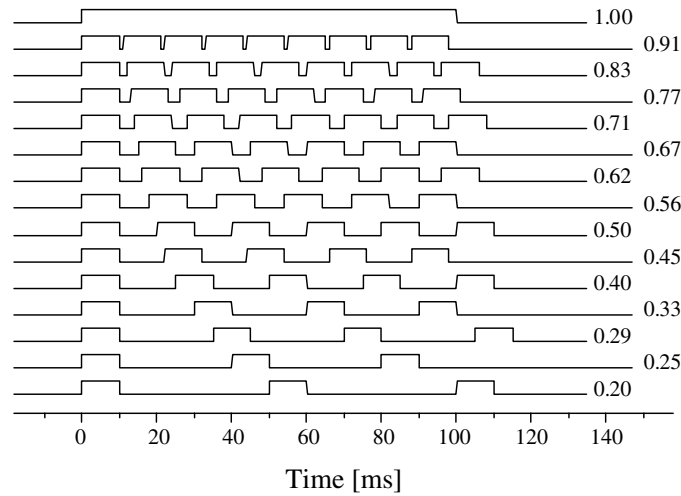


Figure 1. Intermittent spraying sequences consist of 10 ms cryogen spurts separated by 1–40 ms time intervals and are 90–115 ms long. The corresponding duty cycles (r) are marked next to each line. A continuous 100 ms spray is used as a reference.

diameter 7 mm), embedded in an epoxy resin plate of equal thickness. While the front surface of the disc, roughened with sand paper to resemble human skin, is exposed to cryogen spray, its temperature is measured by a thermocouple (type K, 300 μm bead) soldered to the back surface. The temperature data, $T(t)$, are acquired by a dedicated A/D converter and software (InstruNet, Omega Engineering, Stamford, CT) at a sampling rate of 1000 Hz. The disc and plate are supported by a block of expanded polyurethane, which prevents heat exchange across the back surface of the disc.

Heat flux across the sprayed disc surface (j_Q) is determined as

$$j_Q = -\frac{\rho c V}{A} \frac{dT}{dt} = -\rho c d \left(\frac{dT}{dt} \right) \quad (1)$$

where ρ (8920 kg m^{-3}), c (390 $\text{J kg}^{-1} \text{K}^{-1}$), V and A represent the density, specific heat, volume and surface area of the copper disc, respectively, and d is its thickness. The minus sign ensures positive j_Q values when the disc is cooled. Assuming a convective boundary condition at the disc–spray interface (Carslaw and Jaeger 1959), j_Q is expressed by the heat-transfer coefficient (h), indicative of the involved heat-transfer mechanisms, and cryogen spray temperature (T_c)

$$j_Q = h(T - T_c). \quad (2)$$

By equating (1) and (2), h can be calculated as

$$h = -\frac{\rho c d}{(T - T_c)} \left(\frac{dT}{dt} \right). \quad (3)$$

Equation (1) assumes a uniform temperature T over the whole disc volume. All measurements and results, therefore, represent averages over the disc surface area. For the same reason, the validity of (1) and (3) is limited by the thermal diffusion time over the disc thickness, $t_d \sim d^2/\alpha$. By using a thin disc ($d = 0.42$ mm) of high-diffusivity material (Cu-110: $\alpha = 1.14 \times 10^{-4} \text{ m}^2 \text{ s}^{-1}$), t_d is reduced to ~ 1.5 ms. After accounting for the thermocouple bead size, the theoretical temporal resolution of the detector is ~ 3 ms.

From (3), and assuming a constant h , the disc temperature would relax exponentially towards T_c with a relaxation time

$$\tau = \frac{\rho cd}{h}. \quad (4)$$

For the anticipated range of values, $h = 5000\text{--}10000 \text{ W m}^{-2} \text{ K}^{-1}$, for example, $\tau \approx 150\text{--}300 \text{ ms}$. Our detector is thereby suitable for measurements of heat extraction dynamics during $\sim 100 \text{ ms}$ long spraying sequences.

2.3. Spray temperature measurements

Spray temperature was measured using a bare thermocouple (same type as used in the disc detector), positioned on the spray cone axis 50 or 60 mm from the nozzle. While the thermocouple response time in still water is specified at 40 ms (at 100 °C), it responds in a millisecond when positioned near the spray nozzle tip. In order to eliminate the influence of ambient water condensation and freezing (Majaron *et al* 2001b), these measurements were performed in a dry-air chamber, at relative humidity below 5%.

3. Results

Figure 2 presents copper disc temperatures, $T(t)$, during and after CSC with intermittent sprays of varying duty cycle (r , marked next to each curve), and a continuous 100 ms spray ($r = 1.00$). All spraying sequences start at time $t = 0$ and last for 98–115 ms (see figure 1). Each curve represents an average of two measurements under identical experimental conditions. At the 50 mm spraying distance (figure 2(a)), the curves are practically indistinguishable for r ranging from 1.0 to 0.62. At lower r , the temperature curves separate out as cooling becomes less and less aggressive.

A similar pattern is observed at the longer spraying distance, $z = 60 \text{ mm}$ (figure 2(b)). The most important difference is, however, that the cooling efficacy starts to *diminish* with r increasing above 0.71. Cooling with the continuous spray ($r = 1.00$) is thus less effective than that with a similarly long intermittent spray at $r \sim 0.7$.

An overview of the above described trends is shown in figure 3, which presents the minimal temperatures obtained in each scan (T_{\min}) as a function of r . At $z = 60 \text{ mm}$ (circles), increasing the duty cycle beyond $r \sim 0.7$ diminishes systematically CSC efficacy. The lowest value T_{\min} , obtained at $r = 0.71$, is 5 °C lower than that with the continuous spray. At $z = 50 \text{ mm}$ (squares), however, this effect is barely observable, and the T_{\min} are lower than those at $z = 60 \text{ mm}$ for all duty cycle values. The latter effect can be attributed to the plausible trends of spray droplet density, diameter, and/or velocity to decrease with increasing distance from the spraying nozzle (Aguilar *et al* 2000, 2001a, 2001b).

Figure 4 presents the surface density of heat extracted from the copper disc in the same experiments, $q_s = \rho cd(T_i - T_{\min})$, where $T_i = 22 \text{ °C}$ is the initial disc temperature. The maximal values, $q_s = 6.2 \text{ J cm}^{-2}$ at the spraying distance of $z = 50 \text{ mm}$ (squares) and 5.5 J cm^{-2} at $z = 60 \text{ mm}$ (circles), are obtained around $r \sim 0.7$. The open triangles indicate net spraying time, t_{net} , defined as a product of the number of spurts in the spraying sequence (n) and t_s . Data points corresponding to the same t_{net} values are connected with solid lines.

As seen in figure 4, intermittent sprays can not only extract more heat than continuous sprays of similar duration, but also use less cryogen because t_{net} is roughly proportional to the sprayed cryogen mass. If we introduce cooling efficiency as the ratio between q_s and net

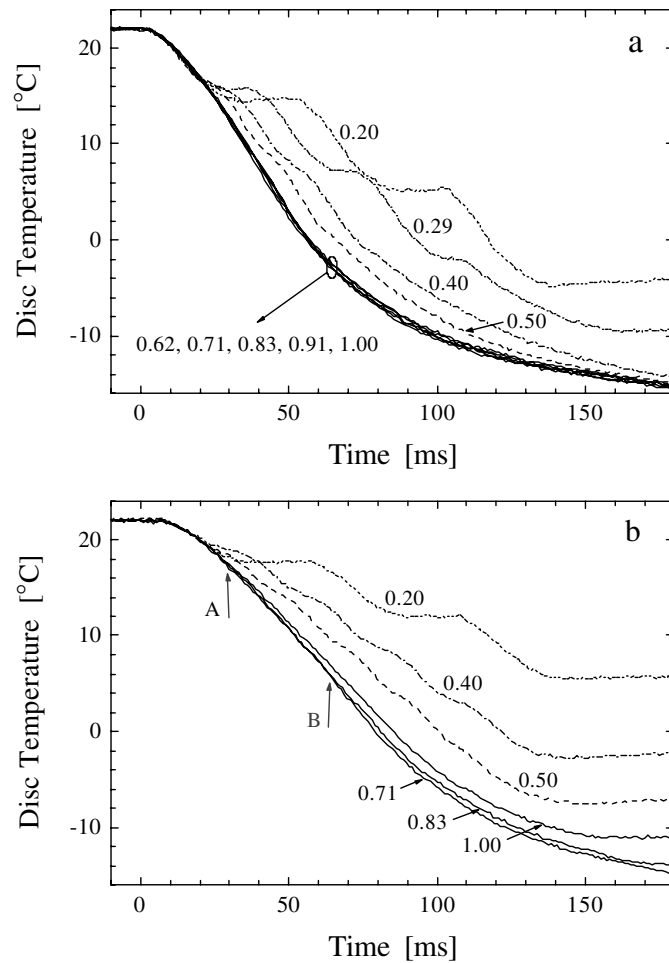


Figure 2. Temperature of the copper disc during intermittent CSC with varying duty cycles, r (marked next to each curve). The spraying distance is (a) $z = 50$ mm and (b) $z = 60$ mm.

spraying time ($\eta_c = q_s/t_{\text{net}}$), its value at $r = 0.71$ exceeds that in a continuous spray by 44% (at $z = 60$ mm). When further decreasing r , η_c stalls at a value 70% above that in continuous spray, around $r = 0.3$.

Figure 5 displays the heat extraction dynamics, $j_Q(t)$, calculated from the temperature measurements shown in figure 2(b) ($z = 60$ mm) using (1). Fast Fourier transform (FFT) filtering with a 10 ms or 5 ms window (for r higher and lower than 0.7, respectively) was applied to suppress the noise induced by derivation over time.

The heat flux reaches the highest value ($j_Q = 56 \text{ W cm}^{-2}$) in the intermittent spray with a moderate duty cycle of $r = 0.71$ (light solid line). At $r = 0.83$, j_Q stalls at a lower value ($\sim 50 \text{ W cm}^{-2}$) approximately 40 ms after the spray begins (dash-dot line, note the arrow 'C'). With a continuous spurt (heavy solid line), $j_Q(t)$ deviates from the former two curves after ~ 20 ms of spraying, and stalls at approximately 48 W cm^{-2} . A prominent common feature is the abrupt decrease in j_Q before the end of the spraying sequence, around $t = 80$ ms with $r = 0.83$ and 1.00 (arrow 'D'), and even sooner with $r = 0.71$.

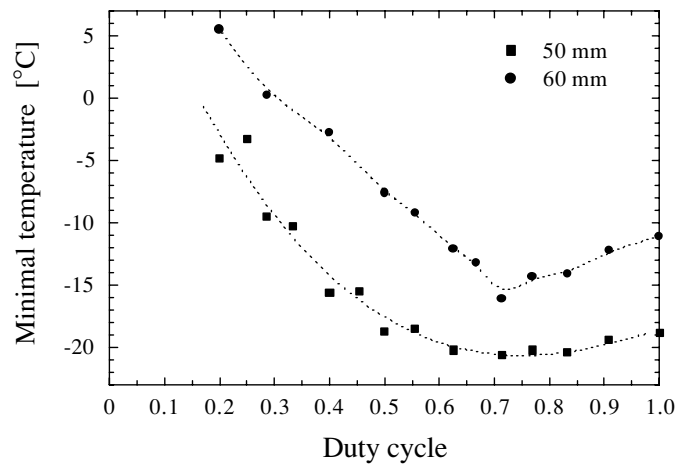


Figure 3. Minimal disc temperatures (T_{\min}) obtained in sequential CSC with varying duty cycles, r (squares: $z = 50$ mm, circles: $z = 60$ mm). The dotted curves are a visual guide, not based on theory.

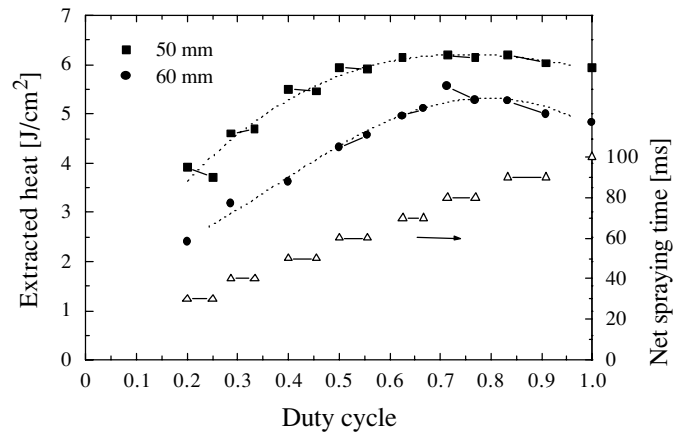


Figure 4. Surface density of heat extracted from the copper disc with sequential CSC (q_s), as a function of r (squares: $z = 50$ mm, circles: $z = 60$ mm). Dotted curves are polynomial fits (a visual guide). Triangles mark the net spraying time ($t_{\text{net}} = nt_s$) of each sequence.

At duty cycles below 0.7, $j_Q(t)$ oscillates synchronously with the spraying pattern (figure 5, dashed line). Eventually, the intermittent spray starts functioning as a sequence of individual 10 ms spurts, with heat extraction completely ceasing between each successive spurt ($r = 0.20$; short-dash line).

Finally, the h values and dynamics are assessed using (3). The cryogen spray temperature (T_c) is set to -59 °C or -60 °C, as measured at $z = 50$ and 60 mm, respectively. Figure 6(a) ($z = 50$ mm) shows that h varies markedly during both continuous and intermittent cryogen sprays. Within the accuracy of the analysis, the results are indistinguishable for r between 1.00 and 0.71. Initially, the $h(t)$ curves rise steeply, reaching peak values of $h_{\text{max}} = 11\,000$ – $11\,500$ $\text{W m}^{-2} \text{K}^{-1}$ at $t = 40$ – 50 ms. At this point, heat extraction is abruptly quenched and h drops to 4000 $\text{W m}^{-2} \text{K}^{-1}$ by the end of the spraying sequence ($t \approx 100$ ms). The considerable cooling effect at later times ($h = 1000$ – 2000 $\text{W m}^{-2} \text{K}^{-1}$) is attributed to evaporating cryogen

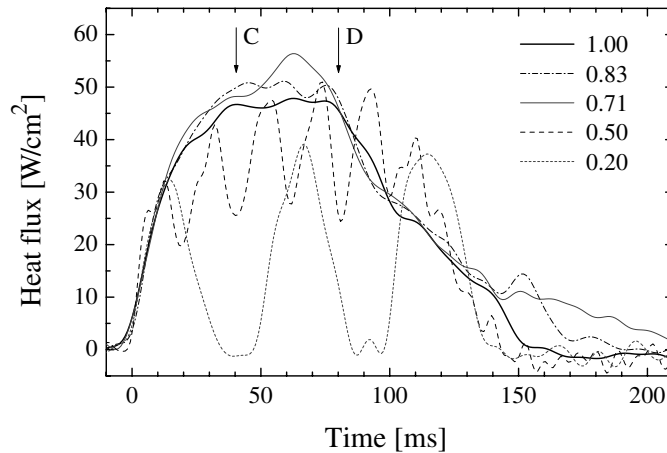


Figure 5. Heat flux dynamics at the sprayed surface, $j_Q(t)$, calculated from disc temperature scans in figure 2(b) ($z = 60$ mm). The duty cycle values (r) are given in the legend.

layer, residing on the disc surface for 150–250 ms after spurt termination (Nelson *et al* 2000, Verkruysse *et al* 2000a, Majaron *et al* 2001b, Pikkula *et al* 2001).

At $r = 0.50$, $h(t)$ displays pronounced oscillations (figure 6(a), dashed line), with the envelope matching the $r > 0.7$ curves. The average h is thus reduced in comparison with the former two examples. Such a trend continues towards $r = 0.20$, where h_{\max} temporarily reaches $12\,000\text{ W m}^{-2}\text{ K}^{-1}$; yet $h(t)$ turns negative between the individual spurts, indicating heating of the disc detector (short-dash line).

At $z = 60$ mm, in comparison, $h(t)$ rises less swiftly in the initial part, and the peak values are consistently lower (figure 6(b)). The largest value, $h_{\max} = 8800\text{ W m}^{-2}\text{ K}^{-1}$ at $r = 0.71$, is 11% higher than that in the continuous spurt. The quenching of heat extraction occurs later than in figure 6(a), at $t = 70\text{--}80$ ms for r between 0.71 and 1.0. With r decreasing below 0.5, the quenching occurs even later, or not at all, h_{\max} values decrease monotonically, and heat extraction stops promptly after the spraying sequence terminates.

4. Discussion

4.1. Experimental approach

CSC efficacy was recently characterized using epoxy blocks with embedded micro-thermocouples. A heat-transfer coefficient of $h = 2400\text{ W m}^{-2}\text{ K}^{-1}$ (and $T_c = -44\text{ °C}$) was reported for 100 ms cryogen sprays from a car fuel injector (Torres *et al* 1999), $h = 2000\text{ W m}^{-2}\text{ K}^{-1}$ for a commercial CSC device (DCD/ScleroPLUS by Candela, $d_n = 0.7$ mm) (Pikkula *et al* 2000), and $1600\text{--}3200\text{ W m}^{-2}\text{ K}^{-1}$ for straight-tube nozzles with inner diameters $d_n = 0.7\text{--}1.5$ mm (Pikkula *et al* 2000). When implementing the same experimental approach in another laboratory, Verkruysse *et al* (2000a) obtained a higher value, $h \approx 10\,000\text{ W m}^{-2}\text{ K}^{-1}$ (and $T_c = -49\text{ °C}$), for a nozzle with $d_n = 1.4$ mm. This result was later confirmed by metal-disc measurements in 100 ms sprays from the same nozzle, yielding $h = 10\,000\text{ W m}^{-2}\text{ K}^{-1}$ (Aguilar *et al* 2001a) and $10\,800\text{ W m}^{-2}\text{ K}^{-1}$ (Svaasand *et al* 2003). For the DCD/ScleroPLUS device, the above two studies determined h to be $6300\text{ W m}^{-2}\text{ K}^{-1}$ and $6600\text{ W m}^{-2}\text{ K}^{-1}$, respectively, in good quantitative agreement with values obtained for a similar spray nozzle in the present study (figure 6).

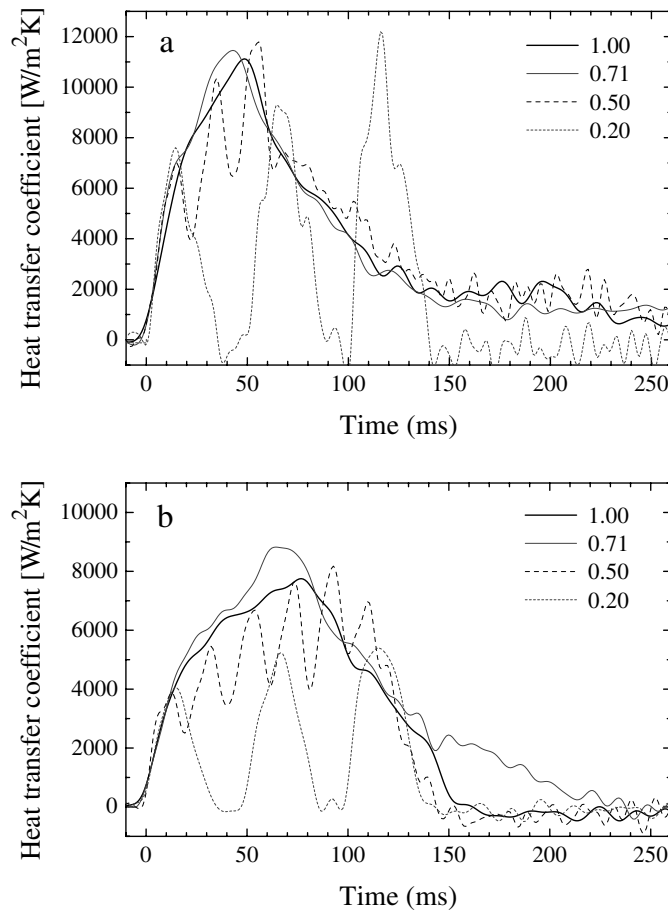


Figure 6. Heat-transfer coefficient dynamics, $h(t)$, at different duty cycles, r (see the legend): (a) $z = 50$ mm and (b) $z = 60$ mm.

While the epoxy block represents a practical model for studying the temperature evolution inside human skin, we believe that its use for determination of j_Q and h may be problematic. The ill-posedness of the involved inverse analysis introduces large uncertainties into the best-fit results in the presence of experimental noise (Verkruysse *et al* 2000a). For the same reason, this approach is very sensitive to experimental parameters, such as the thermocouple depths (Tunnell *et al* 2002b). Systematic errors may result from the thermocouple bead size (relative to high thermal gradients in the substrate), heat conduction through the wires (Valvano and Pearce 1995, Tunnell *et al* 2002b) and inaccuracies in the thermal properties of the epoxy, which may also vary with temperature (Verkruysse *et al* 2000a). Moreover, the measurements become increasingly insensitive to h at high heat extraction rates, since the thermal resistance of the epoxy starts limiting j_Q (Verkruysse *et al* 2000a, 2000b).

Extracting dynamic information ($j_Q(t)$, $h(t)$) is especially difficult (Tunnell *et al* 2002b). This is an important limitation because cryogen spurts shorter than 100 ms are currently used in clinical practice, while up to 20 ms may be required for a cryogen spray to fully develop (Aguilar *et al* 2000).

The metal-disc technique avoids or alleviates most of the aforementioned problems. In particular, the temperature gradients in metals are orders of magnitude smaller, and relax much faster than in epoxy. The thermal properties are precisely known and vary only marginally in the relevant temperature range. These two advantages were exploited in analysis of stationary CSC based on measurements of temperature gradient in a thin copper rod with a variable surface temperature (Verkruysse *et al* 2000a, Aguilar *et al* 2001c). This experimental approach, as well as the metal-disc technique, enables studies of lateral inhomogeneities in CSC by scanning a small detector across the spray or by implementing a detector array.

The metal-disc detector has a simple structure, and determination of heat extraction dynamics from acquired data is straightforward. This enables reliable and reproducible measurements of j_Q and h , allowing analysis of minor modifications of CSC conditions, such as r and z in the present study.

As mentioned earlier, the theoretical temporal resolution of our detector is ~ 3 ms. In practice, however, the thermocouple (TC) data may lag behind the disc temperature evolution unless the thermal contact between the TC bead and metal disc is superior to that between the disc and cryogen spray. In a direct comparison performed in our laboratory (Majaron *et al* 2001a), a TC soldered to a silver disc ($d = 0.7$ mm) indicated a peak value of $h_{\max} = 6000 \text{ W m}^{-2} \text{ K}^{-1}$. Simultaneous measurements with a separate TC, dipped in thermal paste, pressed into a tiny bore on the back of the same disc and fixed with a drop of conductive epoxy, produced severely distorted cooling dynamics and $h_{\max} = 3500 \text{ W m}^{-2} \text{ K}^{-1}$. Note that it would be essentially impossible to recognize the systematic error in the latter result without having available the soldered TC data. (Given the high viscosity of most epoxy resins and their tendency to shrink while curing, it is conceivable that non-optimal thermal contact between the TC and epoxy substrate may contribute to underestimation of j_Q and h in epoxy-block measurements.)

4.2. Heat extraction by intermittent CSC

The presented results demonstrate that intermittent CSC can produce higher cooling rates than continuous sprays of the same duration. For our nozzle and spraying distance of $z = 60$ mm, such a trend persists at $r = 0.7$ – 1.0 , but is reversed at lower duty cycles (figure 3, circles). This is in perfect agreement with spray cooling theory in general (Estes and Mudawar 1995), and the hypothesis of the ‘insulating’ cryogen film (Verkruysse *et al* 2000a, Aguilar *et al* 2001c), in particular.

In figure 5, the $j_Q(t)$ curves obtained with higher duty cycles ($r > 0.7$) start to differ 20 ms into the spraying sequence. If $r = 0.71$ —presumably corresponding to no cryogen layer build-up—is used as a reference, the $r = 1.00$ curve is the first to indicate a reduced cooling effect (see also figure 2, arrow ‘A’). With a somewhat lower average mass flux than in the continuous spray ($r = 0.83$), the deviation—tentatively indicating the cryogen layer build-up—occurs considerably later (figure 5, arrow ‘B’ in figure 2).

At the shorter spraying distance ($z = 50$ mm), the adverse effect of excessive mass flux at high duty cycles is markedly reduced (figures 2(a) and 3). This can be understood by recalling that the average spray droplet diameter and velocity are larger closer to the nozzle (Aguilar *et al* 2000, 2001a). As demonstrated earlier, the impact of high-momentum spray droplets can augment heat extraction by reducing the thickness of cryogen layer at the impact site and/or by enhancing convective heat transfer within it (Verkruysse *et al* 2000a, Aguilar *et al* 2001c).

Heat flux across the cooled surface (j_Q) varies substantially during both continuous and intermittent CSC (figure 5). A prominent common feature is the abrupt decline of j_Q before

the spray terminates, around $t \sim 80$ ms with $r = 0.83$ and 1.00 (arrow 'D'). Since this effect correlates in time with the disc temperature crossing 0°C (figure 2(b)), we attribute it to the freezing of ambient water on the disc surface. We demonstrated earlier that condensation of ambient water and frost formation impair CSC efficacy through the associated release of latent heat (Majaron *et al* 2001b). Accordingly, we attribute the stall in $j_Q(t)$ at $t \sim 40$ ms (arrow 'C') to the onset of ambient water condensation. Combined with the temperature data in figure 2(b), this indicates a dew point at $\sim 14^\circ\text{C}$.

4.3. Heat-transfer coefficient (h)

Our results show that higher h values can result from intermittent as opposed to continuous CSC, due to the influence of the cryogen layer deposition. In addition, h varies markedly during the spraying event in both modes (figure 6). The observed dynamics closely resembles that seen in figure 5, which is understandable since $j_Q(t)$ (1) and $h(t)$ (3) differ only by the slowly varying factor $(T(t) - T_c)$.

However, the rather slow increase of h over 70–80 ms of spraying in figure 6(b) may not accurately reflect the dynamics of the involved heat extraction processes. Optical scattering measurements namely indicated that the cryogen spray from this nozzle stabilizes after ~ 20 ms (Aguilar *et al* 2000). We therefore believe that the discussed effect may result in part from the varying effective spray temperature. Such an effect is conceivable since the surrounding air, which is entrapped by the spray, is becoming progressively colder during the spurt. In this sense, the indicated gradual increase of $h(t)$ could be perceived as an artefact due to the assumption of constant T_c in (3).

In an attempt to improve on this analysis, we replaced T_c in (3) with dynamic spray temperature data, $T_{\text{TC}}(t)$, as measured using a bare thermocouple in a dry-air environment (figure 7(a)). However, the results were difficult to reconcile with prior knowledge of the involved effects. The excessively high initial value ($h \sim 100\,000\text{ W m}^{-2}\text{ K}^{-1}$ after a few ms) dropped very rapidly (to $h \sim 50\,000\text{ W m}^{-2}\text{ K}^{-1}$ at $t = 10$ ms), and converged into the $h(t)$ dynamics obtained from the former analysis at $t \sim 50$ ms.

This observation suggests that $T_{\text{TC}}(t)$ does not represent the actual spray temperature evolution. Instead, the thermocouple (TC) reacts to CSC in a similar way as our disc detector, with heat extraction from the TC bead limited by the value of h (2). In response, $T_{\text{TC}}(t)$ follows a relation similar to (1), with the product ρcd replaced by an unknown quantity (ζ). Consequently, the heat-transfer coefficient, referring to spraying of the bare thermocouple, can be obtained from

$$h'(t) = -\frac{\zeta}{(T_{\text{TC}}(t) - T_c)} \left(\frac{dT_{\text{TC}}}{dt} \right). \quad (5)$$

If $h'(t)$ was exclusively a spray property, this result should match that obtained from the corresponding disc measurement, $h(t)$ (3). Such a comparison is presented in figure 7(b) (for $r = 1.00$ and $z = 60$ mm). With $\zeta = \frac{1}{7}\rho cd$, $h'(t)$ (solid line) matches $h(t)$ (dashed line) within the experimental error, through the first 20 ms of the spurt. At that time, cryogen starts building up on the disc surface (see figures 2(b) and 5), and ambient water condensation starts affecting the disc heat balance shortly thereafter. Since both these effects are suppressed in the bare-TC measurement, the related $h'(t)$ values exceed $h(t)$ for $t > 20$ ms. (FFT filtering with a 5 ms window was applied to suppress the noise in $h'(t)$. Nevertheless, $h'(t)$ values become unreliable and oscillate wildly at $t > 60$ ms, due to the small denominator in (5).)

From the above determined value of ζ , the TC relaxation time can be estimated from (4) as $\tau' = \zeta/h \approx 20\text{--}40$ ms (for $h = 5000\text{--}10\,000\text{ W m}^{-2}\text{ K}^{-1}$). Despite the small thermal capacity of our TC bead, the CSC heat extraction is too low to ensure that $T_{\text{TC}}(t)$ would follow

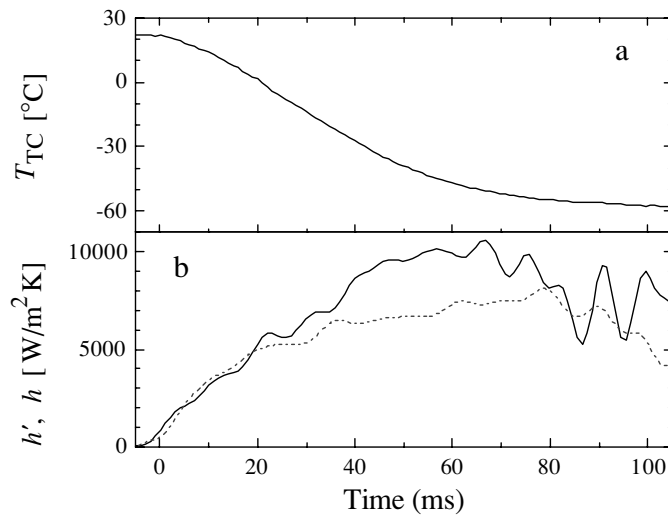


Figure 7. (a) Temperature of a bare thermocouple, $T_{TC}(t)$, during a 100 ms continuous spray ($z = 60$ mm). (b) Comparison of $h'(t)$, calculated from the above data using (5) (solid line) and $h(t)$ from (3) (dashed line; see text for details).

the spray temperature evolution on a millisecond timescale. (Since ζ is proportional to the bead diameter, $\tau' \approx 2\text{--}4$ ms for the smallest commercially available TC with $d_{TC} \approx 30$ μm .) In the absence of reliable spray temperature data, however, it is wiser to use a constant T_c , and combine the dynamics of effective spray temperature, influence of the liquid cryogen layer, and ambient water condensation and freezing, in $h(t)$. To our knowledge, such an approach was used in all recent studies of CSC.

Finally, the texture, chemical composition and thermal properties of the metal-disc detector differ from those of human skin. As has been established in a separate study (Svaasand *et al* 2003), the h values obtained using this method can under certain design considerations be representative of CSC of human skin.

4.4. Implications for CSC-assisted laser medical procedures

For the purpose of laser therapy of superficial dermatological lesions such as PWS, requiring the highest spatial selectivity of cooling, intermittent CSC can be used to maximize the rate of heat extraction by matching the sprayed mass flux to cryogen evaporation off the skin surface. In the present study, intermittent sprays with $r = 0.71$ resulted in 17% higher j_Q (and 15% higher q_s) than a continuous spurt of equal duration, while using 20% less cryogen (at $z = 60$ mm).

It should be noted that, while j_Q and q_s were measured to be higher at $z = 50$ mm, modifications of spraying distance are often undesirable because they affect other spray characteristics (e.g., sprayed area). Also, the higher momentum sprays may induce a prohibitive amount of patient discomfort (Verkruysse *et al* 2000a), and the higher spray density increases the amount of excess liquid cryogen deposited on a patient's skin. In contrast, the absence of a liquid cryogen layer with application of intermittent sprays improves control over the cooling duration and minimizes the formation of frost on adjacent skin sites, thus alleviating the related adverse effects (e.g. obstruction of sight, undesired cooling).

Alternatively, maximizing the rate of heat extraction may not be the ultimate goal in laser treatment of deeper and larger subsurface targets (e.g., hair follicles). For such targets, a prolonged, but less aggressive cooling would be advantageous, providing sufficient epidermal protection while eliminating the risk of cryo-injury (Nelson *et al* 2000, Zenzie *et al* 2000, Majaron *et al* 2001b, Tunnell *et al* 2002a). Intermittent spraying with low duty cycles offers a practical approach to adapting CSC technology for such dermatologic laser applications.

5. Conclusions

The heat extraction rate (j_Q) varies substantially during a 100 ms continuous CSC, in part due to condensation and freezing of ambient water. In our experiments, j_Q temporarily exceeded 80 W cm^{-2} , which corresponds to a heat-transfer coefficient value of $h \approx 11\,000 \text{ W m}^{-2} \text{ K}^{-1}$ (at $T_c = -59^\circ \text{C}$). Intermittent spraying with moderate duty cycles can be used to prevent the accumulation of a liquid cryogen layer on the skin surface, thus increasing the cooling rate and efficiency in comparison with continuous CSC. Preventing the cryogen build-up also helps localize the cooling effect to the desired area and improves control over the actual cooling duration. Alternatively, low duty cycles offer substantially reduced average heat extraction rates, enabling safe and efficient CSC for use in laser treatments of deeper skin structures.

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