CHARACTERIZATION OF CRYOGEN SPRAY COOLING WITH THIN-FILM THERMOCOUPLE

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ABSTRACT

Cryogen spray cooling (CSC) has been used effectively to protect the epidermis during laser dermatologic surgery. However, the temperature reduction in human skin induced by CSC has not been reliably determined due to the short cooling period and significant temperature gradient in the skin. Therefore, CSC has not been optimized for different laser dermatologic surgery procedures. Although it would be desirable to measure in situ human skin temperature, embedding a sensor within 100 μm beneath the skin surface is not feasible. In addition, infrared skin temperature measurement is also not workable because the skin is covered with a cryogen layer of unknown thickness. In this study, we selected an epoxy which has similar thermal properties to that of human skin as our cooling target. Thin-film thermocouples (TFTC) were deposited directly onto the epoxy substrate using micro-fabrication techniques to minimize the thermal contact resistance between TFTC and the substrate. The negligible mass of TFTC also creates a minimal disturbance to heat flow across the surface. Due to the difference in thermoelectric property between thin film and leading wire, special sensor design and calibration procedures were developed. TFTC were calibrated from -46 to 50°C. The thermoelectric sensitivity is around 50-60% of that of bulk material. Skin phantom temperature reductions produced by a commercial medical laser nozzle at different spray durations were measured using the TFTC. The results not only help to elucidate the mechanisms involved in interaction between cryogen spray and human skin but also provide a thermal boundary condition for numerical modeling of laser dermatologic surgery.

INTRODUCTION

Currently, pulsed laser is the treatment of choice for vascular birthmarks, such as port wine stains and hemangiomas. Yellow light (λ = 585-595 nm wavelength) emitted by the pulsed dye laser is preferentially absorbed by hemoglobin (major chromophore in blood) in the blood vessels where, after being converted to heat, causes thermal damage to the malformed blood vessels. However, non-specific light absorption by epidermal melanin competes for absorption of laser radiation in subsurface target chromophores. If not controlled, high epidermal temperatures can lead to blistering, dyspigmentation or scarring. Pre-cooling of the epidermis with a millisecond cryogen (tetrafluoroethane, R-134a) spurt greatly improves patient comfort, safety and permits the application of laser pulses with higher energy[1, 2]. Despite the wide clinical use of cryogen spray cooling (CSC) and considerable research efforts in recent years, the temperature reduction in human skin or skin phantom induced by CSC has not been reliably determined. It is, therefore, not certain whether existing CSC devices have been optimized for their respective clinical applications.

During CSC, the epidermal temperature may change from 30°C to -26°C within milliseconds, and a significant temperature gradient is expected because of the low thermal diffusivity of human skin. The performance of conventional wire thermocouples may be unsatisfactory for temperature measurements during CSC due to their relatively long response time (3 ms) [3] and large bead diameter (30 – 90 μm) [3-7] as compared to the thickness of human epidermis (50 – 100 μm).
In fact, CSC heat flux derived from temperature measurement with conventional wire thermocouples embedded in different substrates (epoxy, copper, etc.) ranges from 75 – 1,100 kW/m² [3-7]. In this study, thin-film thermocouples (TFTC) were developed for temperature measurements during CSC.

**NOMENCLATURE**

Put nomenclature here.

**THIN-FILM THERMOCOUPLE**

TFTC were deposited directly onto the substrate surface using microelectronic thin-film fabrication techniques, which minimize the thermal contact resistance between the TFTC and the surface. TFTC have two inherent advantages as compared to conventional wire thermocouples: 1) thickness of only a few micrometers which provides an extremely rapid thermal response and high spatial resolution; and 2) negligible mass which creates a minimal disturbance to fluid flow over the surface and heat transfer in the substrate. Because of these unique features, TFTC technology was originally developed for application to superalloys used in jet aircraft engines [8-12]. Thereafter, TFTC were used to measure surface temperatures in sliding contact situations [13] and rapid contact solidification processes [14]. Temperature measurements in rabbit eye were also achieved with a TFTC on a quartz probe [15]. In summary, TFTC technology has been shown to provide a minimally intrusive means of measuring temperature with an extremely short response time (5 μs) and high spatial resolution [14].

**Structure**

The structure of the TFTC is illustrated in Figure 1. It has three components: 1) epoxy substrate; 2) copper (Cu)/nickel (Ni) leading wires; and 3) Cu/Ni thin-film strip which forms the measurement junction. The cured epoxy (EP30-3, Master Bond Inc., Hackensack, NJ) has a thermal diffusivity of 0.84×10⁻⁷ m²/s, which is within 10% of that of human epidermis (0.95×10⁻⁷ m²/s) [16]. To avoid trapping air bubble in the substrate, we mixed the epoxy resin with its hardener carefully, degassed thoroughly and then poured into a mold attached with leading wires and standard thermocouples. As shown in Figure 1A, the leading wires have a U-shape inside the epoxy, which prevents the wire from rotating. Straight wires were used in our unsuccessful initial sensor design because slight wire rotation broke the connection between thin-film strips and leading wires. The Ni strip/wire junction is located far from the sprayed area because it was found that electromotive force (EMF) could be generated if the junction is cooled by spray (Figure 1B). A K-type thermocouple (5TC-TTK, Omega Engineering, Stamford, CT) was installed there to monitor any temperature change. After the substrate cured in 24 hours, the top surface was polished with 600-grit sandpaper, degreased with acetone, cleaned with DI water and blow-dried with nitrogen. Thereafter, stainless steel masks were attached on top of the substrate and thin-film strips of Ni/Cu with a thickness of ~1.5 μm was deposited sequentially using an E-beam evaporator (model CV-14, Airco/Temescale).

**Calibration**

Since the Seebeck coefficient of thin-film strip may not be the same as that of leading wire due to different crystal structure etc, three junctions (Cu/Ni strips, Cu strip/wire and Ni strip/wire) may contribute to the TFTC’s total output. To find out if there is EMF generation, each of these three junctions was cooled separately with a drop of dry ice/alcohol mixture which has a temperature of -76°C. It was found that there was EMF generation when either the Cu/Ni strip (expected) or Ni strip/wire (unexpected) was cooled. No EMF generation was detected when the Cu strip/wire was cooled. We have not found a deposition procedure to eliminate the EMF generation at the Ni strip/wire junction. However, special sensor structure and procedures were developed to minimize the temperature change at the Ni strip/wire junction during calibration and measurement.

To calibrate the TFTC, we submerged only the square part of the substrate into baths of liquid with different temperatures (see Figure 1B). Our hypothesis is that when the junction of Cu/Ni strip reaches the bath temperature, the temperature of the Ni strip/wire junction barely changes because the diffusivity of epoxy is low and the distance between liquid bath and Ni strip/wire junction is great. To verify this hypothesis, we measured temperatures adjacent to the Cu/Ni strip and Ni strip/wire junctions with embedded standard K-type thermocouples (TC1 and TC2 in Figure 1A). The liquid bath temperature was measured with another K-type thermocouple.
Standard thermocouples’ output and the voltage output of the TFTC were recorded with a data acquisition system (InstruNet, GW Instruments, Inc., MA).

Figure 2: Voltage change of TFTC and temperatures changes of TC1 and TC2 over time when the square part of the substrate was submerged into ice/water mixture. Pool temperature is also shown in the figure.

Figure 2 shows the voltage change of TFTC and temperatures changes of TC1 and TC2 over time when the square part of the substrate was submerged into ice/water mixture. It took 15 minutes for TC1's temperature (pink curve) to reach its asymptotic value which is very close to the pool temperature (green curve). Because the TFTC is located closer to the liquid pool than TC1, TFTC’s temperature should also reach pool temperature after 15 minutes. TC2’s temperature reduced from 20°C to 19.5°C. The temperature decrease of TC2 (Ni strip/wire junction) is only 2.5% of that of TC1 (Cu/Ni strip junction). For other liquid pools, this percentage remained below 3%. Therefore, we believe our hypothesis is valid and the following calibration curve can be used to convert the TFTC’s voltage output to temperature when Ni strip/wire junction is kept at room temperature of 20°C.

Another issue associated with TFTC is the nonuniformity of the thin film strips. We found that when the film thickness is less than 1 μm, EMF may be generated when the Ni strip is cooled with cryogen spray. Significant measurement error will be resulted and therefore a film thickness of 1.5 μm is recommended and each sensor much be checked to ensure that no EMF generation when the Ni film’s temperature changes.

Figure 3 shows the calibration curve of the TFTC. The liquid pools were heated water (35 and 50°C), ice/water mixture (0°C), refrigerant R134a (-26°C) and R404a (-46°C) at atmosphere pressure. Each point is an average of TFTC’s voltage and pool temperature in one minute after the former’s temperature reaches its asymptotic value. As seen, a parabolic fit can be used for the temperature range presented herein (-78°C to 44°C). The TFTC’s sensitivity is about 10 μV/°C, which is 50% of that of wire Cu/Ni thermocouple.

PRELIMINARY RESULTS

Effect of Spurt Duration

We have carried out CSC experiments with a nozzle from a commercial medical laser (GentleLase, Candela, MA). The nozzle inner diameter is 0.5 mm, distance from nozzle exit to the TFTC sensor is 30 mm, the angle between the nozzle and the TFTC surface is 60°. Three CSC durations (Δt_{CSC}) of 20, 50 and 100 ms were selected. The measured surface temperature variations with time derived from the above calibration curve are shown in Figure 4. Each curve is the average of three trials and difference between different trials is marginal.

Figure 4: Measured surface temperature variation as a function of time for cryogen spurt duration t_{CSC} = 20, 50, and 100°C; nozzle-surface-distance = 30 mm.

Three distinct heat transfer regimes can be identified from these curves. The first regime is transient heat conduction in which cold cryogen droplets are put into contact with the skin phantom. The surface temperature drops sharply from 20.6°C to -30.5°C within 2 ms. The second regime is forced convection by cryogen spray. The surface temperatures...
decrease gradually during the spurt and reach the minimum surface temperatures of -48.6, -50.2 and -51.5°C for spurt duration of 20, 50 and 100 ms, respectively. The third regime is the residual cryogen evaporation after spurt termination. A plateau of the temperature curve for each spurt duration represents the exhausting of residual cryogen, which was confirmed with the high-speed video recorded simultaneously with temperature acquisition. It is also noted that the actual spurt duration is ~8 ms longer than that of the trigger pulse due to the delay of the valve and the contribution of the remnant cryogen in the valve and tube.

**Non-Uniformity**

High-speed videos of spray-surface interaction were taken at 1000 fps. Selected frames for a spurt duration of 50 ms are shown in Figure 5. Figure 5A shows that the major axis of the wetted area has a length of 15 mm at 30 ms, although the major axis of the impingement area is only 5 mm in length. In Figure 5B, the wetted area reached its maximum at 60 ms, which was 2 ms after spurt termination. The major axis increased by 3 mm only as compared to Figure 5A. It can be clearly seen from Figure 5C that the wetted area can be divided into three regions. Residual cryogen in Region 1 and 3 dried up within 100 ms after spurt termination. However, residual cryogen in Region 2 remained on the surface for more than 600 ms and bubbles could be seen in this region. Due to the non-uniform cryogen deposition, significant non-uniformity in the temperature reduction was resulted.

**Figure 5:** Spread and evaporation of cryogen on the surface of an epoxy substrate.

Temperature reductions at five locations in the wetted area were measured. As shown in Figure 5B, the temperature reduction at location C, which is the intersection point of the nozzle axis and the substrate surface, was measured first. The TFTC was then moved sequentially to the left, right edges of the wetted area (-X₂, X₂) and the midpoints (-X₁, X₁) between location C and -X₂ or X₂.

**Figure 6:** Temperature reductions at five locations in the wetted area (Figure 5B).

Figure 6A shows the surface temperature variations over time during the cryogen spurt and shortly after spurt termination. Within the impingement area (C and X₁), the temperature reduction curves are similar to each other. The minimum temperature at location C was slightly lower than that at location X₁. In the forward direction (-X₁ and -X₂), the surface temperatures decreased slowly before the TFTC at those locations got wetted and thus the temperature decreases should be induced by convection of the cryogen vapor and lateral heat conduction. Thereafter, the trend of temperature variations was similar to that at location C, which consists of a sharp temperature decrease caused by the initial cryogen contact with the TFTC and a gradual decrease due to cryogen convection. However, the minimum temperatures at these locations were much higher than that at location C. It can also be seen that the minimum temperature increased with the distance between these locations and location C. In the
backward direction (X2), the surface temperature variation was quite different to that at all other locations. The surface temperature reached its minimum during the spurt and then increased to a temperature that is 6°C higher than the cryogen’s saturation temperature (Figure 6B). We hypothesize that the cryogen spray did not have sufficient momentum to reach the surface in the backward direction once the cryogen layer reached certain thickness and the isolation effect of a thick cryogen layer made the surface temperature increase.

Figure 6B shows the surface temperature variations over time long after spurt termination. Within Region 2 (locations X1 and X2), the temperature remained around the cryogen’s saturation temperature of -26.1°C for more than 600 ms, which is not desired because frost bite may be resulted if human skin is exposed to this temperature for such a long period of time. However, subsequent laser heating of the epidermis during clinical practice may substantially reduce the risk of frost bite.

CONCLUDING REMARKS

We have developed thin-film thermocouples (TFTC) to characterize cryogen spray cooling (CSC). The sensor has a special structure and calibration procedure due to EMF generation at the Ni strip/wire junction. Using the TFTC, we have obtained some preliminary results of surface temperature variations during CSC. Three distinct heat transfer regimes can be identified, which may be helpful to develop optimal cooling devices for medical lasers.

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