Heat Transfer Enhancement and Applications of Femtosecond Laser Processed Metallic Surfaces

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HEAT TRANSFER ENHANCEMENT AND APPLICATIONS OF FEMTOSECOND LASER PROCESSED METALLIC SURFACES

By
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A THESIS

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HEAT TRANSFER ENHANCEMENT AND APPLICATIONS OF
FEMTOSECOND LASER PROCESSED METALLIC SURFACES

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In the present work, functionalized 304 stainless steel metallic surfaces were created with the use of a Femtosecond Laser Surface Processing (FLSP) technique. The laser processing technique produces self-organized micro/nanostructures on the surface. The heat transfer performance of various FLSP functionalized surfaces were characterized through pool boiling and Leidenfrost experiments. Enhancement in both the nucleate and film boiling heat transfer were observed through an increase of the critical heat flux and heat transfer coefficient as well as shifts in the Leidenfrost temperature respectively. For both experiments, a polished reference sample was used as a baseline line to compare against the functionalized metallic surfaces. Using deionized water as the working fluid, a maximum critical heat flux of 142 W/cm$^2$ and a maximum heat transfer coefficient of 67,400 W/m$^2$-K were found for the processed samples compared to 91 W/cm$^2$ and 23,000 Wm$^2$-K for the polished sample. The Leidenfrost temperatures on the FLSP and polished surfaces were experimentally determined using the droplet lifetime technique. Extraordinary shifts in the Leidenfrost temperatures as high as 175 °C were recorded for a laser processed surface relative to a polished reference sample. Enhancement of the film boiling heat transfer was also observed as water droplets were found to evaporate up to 33% faster on processed surfaces compared to polished surfaces. Enhancement of both the film boiling and
nucleate boiling heat transfer is attributed to increased wettability and capillary wicking of the FLSP surfaces. Besides excellent two-phase thermal transport properties, self-propelled liquid droplets on heated angled FLSP surface microstructures were also achieved. Experiments are carried out to characterize the dynamics and mechanisms of self-propelled liquid droplets on angled FLSP surface microstructures. It was found that the droplet motion direction on angled FLSP surfaces is opposite of that for a surface with conventional ratchet microstructures reported in the literature. A new mechanism for a self-propelled droplet on asymmetric three dimensional self-assembled microstructured surfaces is proposed.
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INTRODUCTION

Two phase heat transfer is a very attractive way of transferring energy due to the very high heat transfer coefficients that typically accompany it. The most common type of two phase heat transfer is boiling which refers to evaporation of a liquid at a solid-liquid interface [1]. Boiling is a very dynamic event as first described by Nukiyama [2] who identified that there is a unique curve that describes the boiling process. This “boiling curve” is shown in Figure 1.1 and gives the heat flux with respect to surface temperature.

![Figure 1.1 Standard boiling curve for a liquid-solid interface](image-url)
As illustrated by Figure 1.1, the amount of heat transferred between the liquid and solid interface is highly dependent on the surface temperature. At temperatures just above the saturation temperature of the liquid, nucleation sites begin to activate and vapor bubbles are generated. These bubbles are initially isolated, but as the surface temperature and heat flux are increased, additional nucleation sites begin to activate and the generated vapor begins to coalesce to form vapor columns and slugs. These vapor columns and slugs promote mixing between the liquid and vapor and increase the convection and thus heat transfer coefficient [1]. As the surface temperature increases up to the critical heat flux, the coalescence of the vapor pockets begins to prevent the rewetting of the surface and starts to insulate the surface. This results in a maximum heat flux, referred to as the critical heat flux (CHF).

As the surface temperature is further increased beyond the critical heat flux, the formation of vapor pockets between the liquid and solid becomes very unstable and results in a very quick formation of a stable vapor layer. When this stable vapor layer is formed, the system experiences a minimum heat flux. This minimum heat flux is also referred to as the Leidenfrost point. At temperatures above this Leidenfrost point, the liquid is effectively insulated from the hot surface by a stable vapor layer. This type of boiling is referred to as film boiling. During film boiling the heat fluxes are very low and the surface temperatures are very high. In most industrial applications the nucleate boiling region is desirable due to the high heat transfer rates and relatively small change in surface temperature. Typically film boiling is avoided at all costs because it usually results in damage to equipment and is thus also referred to as burnout.
A shift of this boiling curve can have a significant impact in many industrial applications. With the advancement of technology comes an increased demand for high power heat dissipation as well as very high temperature heat transfer surfaces. In electronic cooling, processors have very high power densities and are beginning to require new methods for thermal management while in the power plant industry components need to withstand extremely high temperatures. To suit these demands, the boiling curve must be modified. These modifications come in the form of increasing critical heat flux and heat transfer coefficient for high power density applications and increasing the Leidenfrost temperature to suit high temperature power plant applications. The most common way of shifting the boiling curve is by modifying the surface with an addition of micro/nanostructures. The addition of micro and nanostructures on the surface can be achieved through many methods which all have different benefits.

In this study, 304 stainless steel surfaces were used as the experimental heat transfer surface. The surface was modified with the use of a new surface enhancement technique referred to as Femtosecond Laser Surface Processing (FLSP). This technique creates a self-organized micro/nano structure on the surface. This microstructure arrangement can take on a variety of forms ranging from spikes and mounds to pyramids. These microstructures are also coated in a porous layer of nanoparticles during the formation process. These surfaces were tested for both nucleate and film boiling heat transfer performance. An angled variation of these microstructures was also tested and found to promote sustained self-propelled droplet motion.
1.1 Related Literature Review

1.1.1 Pool Boiling Critical Heat Flux and Heat Transfer Coefficient Enhancement Techniques

Two-phase heat transfer on micro and nanostructured surfaces has attracted much research interest in recent time [3]. This is due to the observed high heat transfer coefficients and critical heat flux. Most pool boiling experiments for enhanced heat transfer have been conducted on micro and nanostructured surfaces fabricated using complex fabrication techniques such as etching and thin film depositions carried out in highly controlled environments (i.e., cleanroom). These techniques have been very effective at increasing the critical heat flux (CHF) as well as heat transfer coefficients (HTC) through a combination of increased surface roughness, wettability, and porosity. These microfabrication techniques have been used to create very organized arrays of microstructures ranging from pillars to microchannels and have demonstrated CHF values of 100 – 200 W/cm$^2$ [4]–[6] for pillars and 100 W/cm$^2$ [7] for microchannels. When nanoscale features are added to these microstructures, the critical heat flux can be further increased. Values as high as 230 W/cm$^2$ [4] have been reported, showing that hierarchical structures can significantly increase the performance of heat transfer surfaces. In addition to surface microstructuring, it has also been shown that varying surface chemistry, such as provided by a combination or network of hydrophilic and hydrophobic areas on a surface can significantly enhance the heat transfer performance as well. Hydrophobic surfaces lead to easily activated nucleation sites, a decrease in the
onset of nucleate boiling, and a possible increase in heat transfer coefficients, but result in a much lower critical heat flux [8]–[10]. Hydrophilic surfaces meanwhile delay the formation of a stable vapor layer from forming and thus delaying the critical heat flux. A combination of these types of surfaces allows for an optimization of the heat transfer performance [8]–[11].

In addition to microstructures, silicon and copper nanowire coatings have also been used for two-phase heat transfer enhancement. These types of surfaces have been shown to produce CHF values in the range of 120-250 W/cm$^2$ [12]–[14]. Such CHF enhancements have been attributed to high nucleation site density, superhydrophilicity, and enhanced capillary wicking.

All of the previously mentioned experimental results were obtained with a heat transfer surface comprised of a silicon base material which is advantageous in small applications such as electronics cooling. These types of surface enhancement techniques are, however, not practical for applications which require metallic heat transfer surfaces and much larger areas.

The enhancement of heat transfer using metallic surfaces is especially important for large scale operations like in the nuclear power plant industry. Some work has been done to enhance heat transfer with a metallic base surface using techniques such as anodizing processes or material deposition to achieve the desired micro/nanostructures; however these techniques are often applicable to a limited type of base material. For zircaloy-4 (commonly used in the nuclear industry), it has been
shown that the critical heat flux can be increased up to about 200 W/cm$^2$ by using a simple anodizing method [15], [16]. This work resulted in a very good enhancement of the critical heat flux but no real enhancement of the heat transfer coefficients. In other studies, alumina was deposited onto platinum using atomic layer deposition [17] and zinc oxide was deposited onto copper and aluminum surfaces in order to enhance two-phase heat transfer [18]. The alumina deposition on platinum resulted in a CHF of around 120 W/cm$^2$ while the zinc oxide covered surface only reached maximum heat fluxes of about 80 W/cm$^2$. Both techniques resulted in an enhancement of the heat transfer coefficients. Anodizing has also been applied to aluminum surfaces [19] to induce a nanoporous layer. This resulted in a CHF of around 90 W/cm$^2$ but minimal heat transfer coefficient enhancement. Other enhancement techniques have also been applied to metallic surfaces such as copper. Surface sintering and the addition of nanorods have been shown to increase the heat transfer coefficients of both types of surfaces but with no major enhancement of the critical heat flux [20], [21].

1.1.2 Film Boiling and Parametric Effects on Shifting Leidenfrost Temperature Shift

As stated previously, at temperatures well above the nucleate boiling region and critical heat flux, a stable vapor film is present between the liquid and solid interface. This vapor film acts to greatly reduce the amount of energy transferred between the liquid/solid interface. The point at which this vapor film first becomes stable is referred to as the Leidenfrost temperature [22] and results in a minimum heat flux with respect to the boiling curve given in Figure 1.1. A droplet in the Leidenfrost state is also supported in a nearly frictionless state by this vapor layer [22]–[24].
Because the Leidenfrost temperature represents the maximum surface temperature at which efficient heat transfer can occur, understanding the fundamental mechanisms governing the Leidenfrost temperature is of great importance. Knowledge of these mechanisms in addition to the enhancement of the nucleate boiling region will allow for further modification of the boiling curve to suit additional industrial applications. The Leidenfrost temperature plays an important role in specific applications such as thermal management, power generation, and in drag reduction [25]–[27].

The Leidenfrost state, being an interfacial phenomenon, is expected to be governed by the chemical properties such as composition and surface energy and thermo-physical properties such as density and thermal conductivity of the liquid/solid interface in addition to the topographic (nano- and microscopic structures) characteristic of the solid surface [28]. Earlier Leidenfrost models are based on hydrodynamic instability, homogeneous and heterogeneous metastable nucleation, thermomechanical effects, and wettability effects. These methods are summarized by Bernardin and Mudawar [29] and were found to have relatively low accuracy in predicting the Leidenfrost temperature for various scenarios. These Leidenfrost models were developed for smooth and ideal surfaces and thus are not robust enough to accommodate complex engineered surfaces. More recent studies have identified the importance of surface roughness, surface wettability, and nanoscale porosity on predicting and shifting the Leidenfrost temperature. These parameters can be controlled by novel surface nanofabrication and chemical treatment processes. The
following paragraph summarizes the literature on research efforts to control the Leidenfrost temperature.

It has been shown that the Leidenfrost temperature for a water droplet of 1.88 mm diameter on stainless steel is about 290 °C [30]. Similar Leidenfrost temperatures have also been reported in previous works [29]–[35]. More recent experiments have been focused on understanding which of the above-mentioned mechanisms has the largest effect on the Leidenfrost temperature. Contact angle and surface roughness have been shown to critically affect the Leidenfrost temperature [36]–[38]. As a general trend, rendering a material more hydrophilic increases the Leidenfrost temperature and rendering a material more hydrophobic decreases the Leidenfrost temperature. Various coatings and cleaning methods have been used to modify contact angles in order to understand how these changes can shift the Leidenfrost temperature [25], [37]–[40]; typical Leidenfrost temperature increases were on the order of about 30 °C for superhydrophilic samples while the hydrophobic processing resulted in reductions of about 100 °C. The effects of porosity and micropost structures on the Leidenfrost temperature have also been reported. A 50% increase in porosity of an aluminum oxide surface resulted in a 45 °C increase in Leidenfrost temperature [41]. Adding nanopores to a SiO₂ surface resulted in a shift of 85 °C. The addition of 15 µm tall microposts to this nanoporous SiO₂ surface generated an additional increase of about 94 °C in the Leidenfrost temperature [42]. Hydrophobic surfaces have also been created on stainless steel using a picosecond laser machining process. This method resulted in a contact angle of 115° and a reduction of about 120 °C in the Leidenfrost temperature [43].
1.1.3 Self-Propelled Liquid Droplet Transport

Controlling and moving liquid droplets is very important in many applications such as microfluidics, ink-jet printing, lab-on-a-chip, and fuel injection for combustion applications. Liquid transport can also increase heat transfer efficiency due to an increase in convection. Fluids are conventionally moved through the application of asymmetric potentials such as a pressure gradient (pumps, compressors, etc.) or an electric field (electroosmotic pumps). In microfluidics applications, liquid droplets can be moved and controlled with an asymmetric potential created by varying surface tensions from chemical and thermal gradients[44]–[49] as well as with the use of magnetic fields [50]. These methods have the disadvantage of producing very slow droplet velocities (60 \(\mu\)m/s to 6 cm/s) as well as typically being limited to a small working distance. An alternative to these methods, which has been recently garnering interest in the scientific community, is self-propelled Leidenfrost droplets on asymmetric surfaces.

As stated previously, a droplet in the Leidenfrost state is in a nearly frictionless state due to the vapor layer supporting the droplet [26], [51], [52]. As a result it takes very little force to initiate and sustain droplet motion. This characteristic of droplets in the Leidenfrost state has been recently exploited in self-propelled droplets on ratchet surfaces [26], [51]–[56]. Ratchet surfaces have been shown to be very effective at moving liquid droplets over relatively long distances with considerably high speeds, 5-40 cm/s [26], [51]–[56]. Recently, tilted micropillars have been shown to also result in Leidenfrost droplet motion [57]. Regardless of microstructure arrangement, there has
been a general consensus in the literature that the motion of self-propelled Leidenfrost droplets is in the direction opposite to the direction that the microstructures are tilted. It was found recently that the droplet motion directionality can be dependent on the microstructure size and surface temperature [57]. It is important to understand what can cause a change in directionality of a self-propelled Leidenfrost droplet in order to design functionalized surfaces that provide optimum liquid transport control.

1.2 Experimental Objectives and Goals

The goal of this experimental work is to determine if functionalized metallic surfaces fabricated with the FLSP technique are a viable means for enhancing interfacial two-phase heat transfer. This is determined by experimentally evaluating unique laser processed surfaces in nucleate pool boiling as well as in droplet film boiling. Surface characteristics were changed between each sample in order to understand the fundamental mechanisms that govern both nucleate pool boiling and droplet film boiling. These surface characteristics were used to determine a trend with the corresponding results in order to suggest future surface characteristics for further heat transfer enhancement. Angled microstructured surfaces were also created in order to determine if liquid transport can be induced by the hot surface itself. The mechanisms that govern this liquid motion were also investigated. This work is to serve as a preliminary study into manufacturing an optimal metallic heat transfer surface that provides excellent performance throughout the entire boiling curve due to a combination of enhanced nucleate boiling, film boiling, and liquid transport.
CHAPTER 2  
FEMTOSECOND LASER SURFACE PROCESSING

2.1 FLSP Multiscale Surface Formation and Mechanisms  
Multiscale surfaces (surfaces with roughness on both the micrometer and nanometer scales) are commonly applied for the fabrication of advanced wettability surfaces that range from superhydrophobic to superhydrophilic [58]–[63]. These surfaces are considered to be biologically inspired as they often mimic the surfaces of plant leaves or other biological surfaces; one iconic example is the superhydrophobic lotus leaf, which exhibits self-cleaning properties due in part to multiscale surface features [64], [65]. For such structured surfaces, the relative sizes of both micrometer and nanometer scale structures are critical for the control of not only the contact angle, but also the adhesion and wetting state (e.g. the fully-wetting Wenzel state or the hybrid Cassie-Baxter state) [65].

Femtosecond laser surface processing (FLSP) is rapidly emerging as a powerful and dynamic method for the fabrication of biologically inspired multiscale surface structures. Using this process, surfaces generally consist of self-organized, quasi-periodic micron-scale conical or mound structures that are covered in a layer of nanoparticles [66]–[73]. These surface structures are formed through a complex combination of multiple growth mechanisms including laser ablation, capillary flow of laser-induced melt layers, and redeposition of ablated material.

The size and shape of self-organized surface structures fabricated via FLSP are controlled through various fabrication parameters including the laser fluence, the
number of laser shots per area incident on the sample, and the composition and pressure of the atmosphere during processing. In the present study, a range of multiscale surface morphologies were fabricated on 304 stainless steel and then utilized to demonstrate heat transfer enhancement. The variations in the surface morphology are generated through modification of both the laser fluence and the number of pulses incident on the sample. The fluence and shot number were chosen as control parameters as they represent two contrasting methods of controlling the total dose of laser energy transferred to a substrate. To illustrate this, consider that a given amount of laser energy can be transferred to a target substrate through either a small number of laser pulses with a large fluence or through a large number of laser pulses with a small fluence. However, the laser fluence critically impacts the laser-matter interaction mechanisms attributed to the development of multiscale structures. A shot-by-shot study of the ability of the laser fluence to influence the physical formation mechanisms of the self-organized surface structures was published by Zuhlke [73]. This study showed the ability to utilize this control to fabricate multiscale metallic surface structures that rise above the original surface. Thus, control of the laser dose via a calculated selection of both the laser fluence and the number of pulses on the sample is a convenient method to produce a range of unique surface morphologies. A subset of this range is demonstrated in Figure 2.1, which documents three distinct classes of surface structures fabricated using FLSP: nanostructure-covered pyramids (NC-Pyramids), below-surface-growth mounds (BSG-mounds), and above-surface-growth mounds (ASG-mounds). The
required laser fluence necessary to generate the structures increases from left to right in Figure 2.1.

**Figure 2.1** Scanning electron microscope images of three classes of surface morphologies fabricated by FLSP. The top image of each panel was taken at 45° to show the structure height relative to the original surface; the bottom image of each panel was taken at normal incidence to show the size and separation of the structures.

NC-Pyramids are formed with a laser fluence near the ablation threshold with several thousand pulses, whereas both BSG-mounds and ASG-mounds are formed with laser fluence values several times the ablation threshold. Each of these surfaces is comprised of micron-scale conical structures that are covered with a layer of nanoparticles. However, the structures differ greatly in terms of the height, width and separation of the microscale structures as well as in the thickness of the nanoparticle layer. Specifically, there are three primary mechanisms by which self-organized multiscale surface features grow in response to incident laser irradiation as depicted in Figure 2.2; the balance of these mechanisms is determined by the laser fluence.
The first mechanism, preferential valley ablation (PVA), is a geometry-driven process in which laser light is scattered off of defects on the sample surface that induces a higher laser fluence and thus increased ablation in the valleys between scattering sites. Upon irradiation with multiple pulses, this PVA process is the initial driving force for the formation of the microscale structures. As the structures grow, the increased subtended area of the sidewalls reduces the fluence and amplifies this effect. PVA plays an important role in the formation of all three of the surface morphologies described in Figure 2.1. The primary difference between NC-Pyramids and BSG-mounds is that NC-Pyramids require a defect in the material to serve as a scattering site, whereas the fluence is sufficiently high in BSG-mound creation to form surface defects via hydrodynamic ablation [72], [74]. ASG-mounds are formed with an even higher fluence, which can cause two other formation mechanisms to occur: fluid flow of the surface melt induced by the femtosecond laser by capillary effects [75] and the redeposition of ablated material via vapor-liquid-solid growth [62], [63], [76]. These two mechanisms...
are responsible for the upward growth of ASG-mounds. A detailed description of the development of these structures is beyond the scope of this paper; a detailed description of the formation of BSG-mounds, ASG-mounds, and NC-Pyramids is provided by Zuhlke, et al [73], [77].

2.2 FLSP Component Specifics

A general schematic of the FLSP is given in Figure 2.3. The fabrication laser was a Ti:Sapphire (Spitfire, Spectra Physics) that produced pulses of approximately 50 femtoseconds duration with a central wavelength of 800 nm at a 1 kHz repetition rate. The laser power was controlled through a combination of a half-wave plate and a polarizer. The pulses were focused using a 125 mm focal length plano-convex lens (PLCX-25.4-64.4-UV-670-1064) with a broadband antireflection coating covering the laser spectrum. The sample was placed on a computer-controlled 3D translation stage and translated through the beam path of the laser in order to process an area larger than the laser spot size. The number of pulses incident on the sample was controlled by adjusting the translation speed of the sample. For the fabrication of the angled microstructures used in the self-propelled Leidenfrost droplet experiment, the incident angle of the laser was changed with respect to the normal to create these angled microstructures.
A slight modification of the FLSP setup was made during the transition between experiments. The initial setup also consisted of a refractive Gauss-to-top hat beam shaper (Eksma Optics, GTH-4-2.2FA). This was used to generate a top hat beam with a square profile. The intention of this component was to ensure a uniform laser fluence throughout the cross section of the laser. This additional component was used in the fabrication of processed surfaces used in the Leidenfrost shift experiment. It was later found that more consistent surfaces could be produced with a Gaussian shaped laser beam. Thus, the Gauss-to-top hat beam shaper was removed. The processed surfaces used in the pool boiling and self-propelled Leidenfrost droplet experiments were fabricated with this Gaussian beam profile.
CHAPTER 3
POOL BOILING CRITICAL HEAT FLUX AND HEAT TRANSFER COEFFICIENT ENHANCEMENT

3.1 Boiling Surfaces and Characterization

The impact of multiscale surface structures on the heat transfer coefficient and critical heat flux of stainless steel was investigated through the characterization of four FLSP-generated samples with a polished sample as a control. These surfaces were fabricated with the process outlined in Chapter 2. The defining physical characteristics for the samples analyzed in the present study as well as the laser parameters used in fabrication are summarized in Table 1; scanning electron microscope (SEM) images and 3D profiles of the samples are shown in Figure. 2. The 3D profiles of the sample surfaces were generated using a 3D confocal laser scanning microscope (Keyence VK-X200). The peak-to-valley height, RMS surface roughness, and surface area ratio (total area of the microstructures divided by the projected area) are measured with the Keyence instrument. The separation between the microstructures is determined by a 2D Fast Fourier Transform analysis of the SEM image.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Fluence (J/cm²)</th>
<th>Number of Shots</th>
<th>Peak to Valley Height (µm)</th>
<th>Surface Roughness (µm)</th>
<th>Surface Area Ratio</th>
<th>Separation (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Polished</td>
<td>0.7</td>
<td>840</td>
<td>1.1</td>
<td>1.4</td>
<td>1.09</td>
<td>1.0</td>
</tr>
<tr>
<td>S1</td>
<td>1.1</td>
<td>840</td>
<td>7.2</td>
<td>4.5</td>
<td>3.79</td>
<td>5.9</td>
</tr>
<tr>
<td>S2</td>
<td>2.1</td>
<td>840</td>
<td>31.3</td>
<td>1.8</td>
<td>8.99</td>
<td>26.1</td>
</tr>
<tr>
<td>S4</td>
<td>4.1</td>
<td>250</td>
<td>35.8</td>
<td>7.4</td>
<td>4.7</td>
<td>26.1</td>
</tr>
</tbody>
</table>

The control sample in the present study was 304 stainless steel polished to a mirror finish through the use of a series of buffing compounds. As can be seen from Fig. 3.1, the FLSP samples have mound like microstructures surrounded by deep valleys.
Although it cannot be seen from the SEM images in Figure 3.1, the mound like microstructures are covered in a layer of nanoparticles [77]. This nanoparticle layer will be further discussed in a later section. Samples S1 – S3 are a series created with varying laser fluence but constant shot number (N = 840). The surface structures are Below Surface Growth (BSG) Mounds as described earlier. A visual inspection of Figure 3.1 demonstrates that the microscale structures in this series are similar in shape, but increase in size and separation. Quantitative analysis shows that these samples all have roughly equivalent surface area ratios, but have concurrently increasing peak-to-valley height, RMS surface roughness, and microstructure separation. Sample S4 was fabricated with a higher laser fluence in order to generate Above Surface Growth (ASG) Mounds; this sample is characterized by taller, narrower microstructures surrounded by circular pits.
Figure 3.1 SEM and laser confocal microscope images of the femtosecond laser processed surfaces. Left – Confocal microscope 3D profile, Right – SEM images

It is well known that the wetting and the wicking ability of a surface greatly affects its heat transfer performance. The FLSP process has a significant impact on the
wetting and wicking ability of the surface. A Ramé-Hart Model 590 F4 Series Goniometer and Tensiometer was used to measure the contact angle of both the polished and processed samples using deionized water. The contact angle of the polished surface was found to be approximately 80° while FLSP surfaces all had contact angles of nearly zero. When a small droplet is placed on each of the surfaces, the droplet is absorbed in its entirety into the surface in a very fast manner. The surfaces are considered superwicking because the wetting front easily spreads across the entire surface in a matter of a few seconds as confirmed by flow visualization using a high speed camera. The superwicking behavior is attributed to the presence of the porous layer of nanoparticles that induces high capillary wicking actions

3.2 Pool Boiling Experimental Setup and Procedure

A closed system pool boiling experimental setup was used for the heat transfer experiments. This setup is highlighted in Figure 3.2. The experimental setup was designed to accommodate a variety of working fluids. For the presented results, the test rig was filled with eight liters of deionized water. The system was brought to the saturation temperature of the water at atmospheric pressure by the use of an immersion heater (Omega - MT-112E2/120V) controlled by an analog variac. The water temperature and internal pressure were monitored with the use of two K-type thermocouples (Omega - M12KIN-18-U-6-D) and a pressure transducer (Omega - MMA100V5P4C0T4A5CE) connected to a National Instrument Data Acquisition Board. The water was degassed for a half hour before measurements were taken; the evaporated water was directed through a coil condenser (Quark Glass - qc-6-4sp) and
noncondensable gases were vented to the atmosphere. The condenser was supplied with cold water with the use of a chiller. The experimental setup also includes a bladder accumulator for regulating the overall system pressure. This feature of the boiling rig was not used for the present experiment as all measurements were taken at atmospheric pressure. Two Lexan view windows were incorporated into the test rig to allow for flow visualization.

Once the system was allowed to reach the saturation temperature of the water, power to the heater was gradually increased using a copper heating block containing five cartridge heaters (Omega - CSH-203450/120V) controlled with an analog variac. This copper heating block was attached to the upper copper heating block (see Fig. 3.2) with the use of a high temperature solder (McMaster - 7685A12). The upper copper heating block has five thermocouple holes drilled to the center of the block. The thermocouples (located 3.18 mm apart) were used to measure the temperature gradient within the block and to calculate the heat flux. Heat flux values were recorded after the system had reached steady state monitored through an in-house LabVIEW program. The test section consists of a 25.4 mm diameter and .254 mm thick 304 stainless steel disk. The size of the test section was chosen to be sufficiently large in order to eliminate any heater size effects on the critical heat flux. This was done by determining the Bond number for the given heater size. This number gives a ratio of heater size to bubble departure size. A test sample is considered to be sufficiently large if the Bond number is greater than 3 [78]. The Bond number is calculated using Equation 1.
\[ Bo = \frac{g(\rho_l - \rho_v)}{\sigma} D^2 \]  

Where \( g \) is the acceleration due to gravity, \( \rho_l \) and \( \rho_v \) are the liquid and vapor densities respectively, \( \sigma \) is the surface tension of the liquid in air, and \( D \) is the diameter of the heater surface. Using this equation, the Bond number was found to be around 100; therefore it can be assumed that the heater size can be neglected. The stainless steel test section thickness was chosen to minimize the operating temperatures of the heating block at high heat fluxes. The test section was brazed onto the copper heating block using a silver solder paste (Muggyweld - SSQ-6) to ensure an efficient contact between the two. The surface temperature of the test section was obtained with the use of the measured temperature gradient along the heating block. The contact resistance between the copper and stainless steel was neglected due to the very thin and the highly conductive silver solder braze used. A high temperature PEEK plastic insulating bushing was used to insulate the upper copper heating block. Fiberglass insulation (not shown in Fig. 3.2) was used to insulate the lower copper heating block. High temperature silicon o-rings were used to seal between the concentric heating and insulating pieces. To ensure that nucleation would not prematurely occur on the outer edges of the boiling surface, a special epoxy (Mcmaster - 7513A1) was used for bonding dissimilar materials.
3.3 Heat Flux and Uncertainty Calculation

The five equally spaced thermocouples located in the upper heating block were used to measure the temperature gradient along the axis of the heating block and calculate the heat flux. The heat flux was calculated as:
where $k_c$ is the thermal conductivity of the copper, $T_i$ and $T_j$ are the thermocouple temperatures and $x$ is the thermocouple separation distance. The heat flux is calculated between alternating thermocouple locations. To clarify, if the thermocouples are labeled in order from 1 to 5, the heat flux is calculated between temperatures 1 and 3, 3 and 5, 2 and 4, and then 1 and 5. This increase in separation distance reduces the uncertainty of the calculation. The minimum value of $x$ is 6.35 mm. The thermal conductivity of copper was taken to be constant at a value of 401 W/m-K. The temperature gradient and heat flux were measured at the four locations and then averaged. The critical heat flux is determined when the monitored thermocouple temperatures spike on the order of 100 °C and burnout occurs.

Radial heat losses were minimized by insulating the copper heating blocks. The upper heating block was insulated with a PEEK plastic bushing ($k_p = .25$ W/mK) with a thickness of .635 cm. The lower heating block was insulated with fiberglass insulation. The thermocouple measurements were used to show that the upper copper heating block was well insulated and at uniform temperature in the radial direction. The radial heat loss can be estimated across the PEEK bushing if the aluminum housing (see Figure 3.2) is assumed to be at 100 °C since it is in contact with the saturated water. At heat fluxes near the critical heat flux, the maximum temperature in the upper copper heating block was measured to be 216 °C. It is assumed that the copper/PEEK interface is at approximately 216 °C due to the highly conductive nature of the upper copper heating block; hence the radial heat loss can be estimated by Equation 3.
\[ q''_r = k_p \frac{(T_r - T_w)}{r_p} \]  

(3)

Where \( k_p \) is the PEEK thermal conductivity, \( r_p \) is the PEEK thickness, \( T_w \) is the aluminum temperature at the wall, and \( T_i \) is the interface temperature. It was found that this heat flux is approximately 0.5 W/cm\(^2\) which is less than 1% of the total heat flux measured at the critical heat flux. This approximation was also validated using a full 3D finite element analysis of the heating block, test surface, and insulation assembly. The boundary conditions for the simulation are as follows: convective heat transfer coefficient of 23,000 W/m\(^2\)-K at the boiling surface, heat flux of 92 W/cm\(^2\) at the junction of the upper and lower copper heating blocks, a constant temperature of 100 °C at the PEEK/water and aluminum housing/water interface, and a natural convection coefficient of 10 W/m\(^2\)-K at the lower part of the aluminum housing (see Figure 3.2). The simulation results were in excellent agreement with the theoretical approximation obtained by Equation 3.

Uncertainties on the heat flux values as well as the surface temperatures were calculated using the standard error propagation equation. The standard deviation of the heat flux based on the accuracies of the measurement equipment is given as follows.

\[ \Delta q'' = \sqrt{\left(\frac{\partial q''_r}{\partial x} \Delta x\right)^2 + \left(\frac{\partial q''_r}{\partial T_1} \Delta T\right)^2 + \left(\frac{\partial q''_r}{\partial T_2} \Delta T\right)^2} \]  

(4)
The variation of the thermocouple separation ($\Delta x$) is .08 mm and the thermocouple variation ($\Delta T$) is 1 °C. This leads to an estimated error of approximately 7.5% at an average critical heat flux.

The surface temperature was calculated from the measured heat flux. This calculation considered both the copper material as well as the stainless steel wafer. The contact between the two materials was assumed to be ideal due to the highly conductive and very thin layer of silver braze used to join the two. The surface temperature was therefore calculated as follows:

$$T_s = T_1 - \frac{q'' x_1}{k_c} - \frac{q'' x_{ss}}{k_{ss}}$$  \hspace{1cm} (5)

Where $T_1$ is the thermocouple temperature located closest to the surface, $x_1$ is the distance between $T_1$ and the bottom of the stainless steel wafer, $x_{ss}$ is the stainless steel wafer thickness, and $k_c$ and $k_{ss}$ are the copper and stainless steel thermal conductivities respectively, allowing for variation with temperature. The estimated error for the surface temperature can be determined using the following equation.

$$\Delta T_s = \sqrt{\left(\frac{\partial T_s}{\partial T_1} \Delta T\right)^2 + \left(\frac{\partial T_s}{\partial q''} \Delta q''\right)^2 + \left(\frac{\partial T_s}{\partial x_1} \Delta x_1\right)^2 + \left(\frac{\partial T_s}{\partial x_{ss}} \Delta x_{ss}\right)^2}$$  \hspace{1cm} (6)
\( \Delta x_1 \), the variation of \( x_1 \), is 0.07 mm and \( \Delta x_{ss} \) the variation of the wafer thickness is 0.03 mm. With these variations the error in the surface superheat temperature calculation at the critical heat flux is around 6 – 13% depending on the surface.

In a similar fashion, the uncertainty of the heat transfer coefficients can also be determined using the extreme cases of the heat flux and surface temperature uncertainties. Using the standard error propagation method, uncertainties of the heat transfer coefficient range from 11 – 16% depending on the test surfaces.

3.4 Results and Discussion

For each of the sample surfaces tested, measurements were first taken at low heat fluxes and then the heat flux was gradually increased until critical heat flux was reached. The results are shown in Figure 3.3. It can be seen from this figure that there was a drastic difference between the processed and unprocessed surfaces in their heat transfer performance. The processed surfaces consistently outperformed the polished sample at every surface temperature. The polished sample was found to have a critical heat flux of 91 W/cm\(^2\) at a surface superheat of 40 °C. This result closely matches the results found in the literature for a similar smooth metallic surface [15], [79], [80], as well as critical heat flux correlations [81] such as Zuber’s and Kandlikar’s. Zuber’s model is given in Equation 7 and Kandlikar’s model is given in Equation 8.

\[
q_c = K h_f g \rho_f \left[ \frac{\sigma g (\rho_L - \rho_v)}{\rho_v^2} \right]^{1/4}
\]  

(7)

\[
q_c = h_f g \rho_v^5 \left( \frac{1 + \cos \beta}{16} \right) \left[ \frac{2}{\pi} + \frac{\pi}{4} (1 + \cos \beta) \right]^{1/2} \times \left[ \sigma g (\rho_L - \rho_v) \right]^{1/4}
\]  

(8)
The value of the constant K is .131 as determined by Zuber. The terms $h_{fg}$, $\rho_v$, $\rho_L$, $\sigma$, and $g$ are the latent heat of vaporization, the vapor density, the liquid density, the surface tension, and gravity. These properties are evaluated for water at 100 °C. The $\beta$ term in Equation 8 is the receding contact angle for the polished stainless steel surface which was measured to be 70°. Zuber’s equation predicts a critical heat flux of about 110 W/cm$^2$; this prediction overestimates the measured result for the polished sample in the present experiments. This discrepancy can be attributed to the fact that Zuber’s model does not take into account the surface wettability which we know has significant effects on the critical heat flux. Using Kandlikar’s model, the critical heat flux was predicted to be 91.7 W/cm$^2$ and agreed extremely well with our measured value.

The reported critical heat flux of the polished sample agrees well with theory and other values found in the literature, but there is a variation in the surface temperature reported. The polished sample in our work was found to have a wall superheat temperature of 40 °C at the critical heat flux. In various published data [15], [79], [80] the wall superheat temperature has been shown to range from 10 °C to 55 °C. Even though all the above referenced experiments had Bond numbers greater than 3, they significantly differ in heater thickness. It has been reported that heater thickness and thermal conductivity can have an effect on the heat transfer coefficient [82], [83]. The differences in wall superheat between the present experiments and published literature could therefore be likely due to variances in heater thickness.
The maximum CHF observed was with sample S1 which had a CHF of 142 W/cm². All of the processed surfaces had consistently higher CHF values than the polished sample. An overview of the boiling performance for each test sample is given in Table 3.2.

Table 3.2 Summary of boiling performance for each test surface

<table>
<thead>
<tr>
<th>Sample</th>
<th>CHF (W/cm²)</th>
<th>CHF Surface Superheat (°C)</th>
<th>Onset of Nucleate Boiling Superheat (°C)</th>
<th>Maximum Heat Transfer Coefficient (W/m²-K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Polished</td>
<td>91</td>
<td>39.8</td>
<td>10.8</td>
<td>22,900</td>
</tr>
<tr>
<td>S1</td>
<td>142</td>
<td>29.2</td>
<td>8.6</td>
<td>48,600</td>
</tr>
<tr>
<td>S2</td>
<td>118</td>
<td>22.6</td>
<td>8.7</td>
<td>52,200</td>
</tr>
<tr>
<td>S3</td>
<td>109</td>
<td>22.8</td>
<td>8.2</td>
<td>47,800</td>
</tr>
<tr>
<td>S4</td>
<td>122</td>
<td>18.1</td>
<td>7</td>
<td>67,400</td>
</tr>
</tbody>
</table>

Although the four processed samples had the same contact angle, there is a significant difference between the samples which can no longer be related to a change in the contact angle. It is well known that reducing the contact angle to zero does not result in reaching a limit on critical heat flux enhancement and that additional mechanisms become dominant. As can be seen from Table 3.2 and Figure 3.3, there is a definite trend in CHF enhancement for samples S1-S3. For these surfaces the surface area ratio is nearly constant but the critical heat flux increases with decreasing structure height and spacing. The reason for this increase in CHF for these samples can be related to the microstructure geometry. As can be seen from Figure 3.1, the area between the microstructures increases from S1 to S3. For S1 the mound structures are densely packed and narrow channels or cracks are formed between microstructures. This network of channels, in addition to the nanoparticle layer on the microstructures, allows
for high capillary wicking effects which help to quickly replenish the heated surface with cold liquid after local evaporation occurs and consequently delay the critical heat flux. As the sample number increases, the separation and size of the microstructures also increase; this results in the formation of deep pits in between the microstructures instead of the channels. These deep pits and holes reduce the wicking potential of the surfaces and the wicking effect is mainly dominated by the presence of the layer of nanoparticles covering the mound structures.

Figure 3.3 Heat fluxes with respect to surface superheat for both the laser processed and polished stainless steel surfaces

As mentioned earlier, sample S4 is slightly different than the other samples. However, its critical heat flux enhancement can still be explained by the same approach. S4 resulted in the second highest CHF of 122 W/cm² observed, however had a larger peak-to-valley height as well as structure spacing. The most notable geometric difference between S4 and the rest is the surface area ratio as its surface area ratio is significantly higher than the other samples. This increase in the surface area ratio is the
reason for the increase in critical heat flux over sample S2 which had a much denser microstructure arrangement. The larger surface area ratio results in more surface area in contact with the liquid and thus can compensate for a lack of wicking ability due to the larger microstructure spacing. The increase in surface area ratio can also result in a better wetting surface as described by the classic Wenzel model for a droplet on a rough surface. The contact angle measurements taken were not able to prove a difference in wettability between the processed surfaces due to the highly wicking nature of the processed surfaces and the limits of the measuring device. Hence, S4 has higher CHF than S2 and S3 because of its relatively higher wettability. It can be therefore concluded from the observed results, that the enhancement in CHF is due to a combination of both surface wettability and capillary wicking.

The local maximum superheat temperature that arises in S2-S4 can be related to the thermal conductivity of the surface and the active nucleation site density. Since the thermal conductivity of stainless steel is relatively low compared to other metals such as copper, local sites with different heat fluxes can occur [82]. As the number of active nucleation sites increase the average surface temperature would then decrease. Because the processed surfaces have roughness on both the nano and microscale, there is a wide range of potential nucleation sites that could be activated at high heat fluxes. Near the critical heat flux, the maximum amount of nucleation sites is active and thus reduces the surface temperature. The polished sample does not have this nano and microscale roughness so there are no additional nucleation sites to activate at high heat fluxes and thus the curve remains nearly linear.
The shift of the boiling curves to the left with the processed surfaces is very advantageous to heat transfer applications as it corresponds to an enhancement of the heat transfer coefficients and allows for a large amount of heat to be transferred at relatively low surface temperatures. The steep slope of the curve is also advantageous because of the relatively small surface temperature change (around 10 °C for S4). For all processed surfaces investigated, significant enhancement of the heat transfer coefficients were observed in comparison to the polished surface.

Heat transfer coefficients based on the projected area were determined for each of the test samples and plotted in Fig. 3.4. A summary of the maximum heat transfer coefficients is also given in Table 3.2. The maximum heat transfer coefficient values varied from 22,900 to 67,400 W/m²-K. As expected for nucleate boiling, the heat transfer coefficients increase with increasing heat flux. The shape of each curve looks

![Figure 3.4 Heat transfer coefficient with respect to wall heat flux for each sample surface](image)
nearly the same for heat flux values up to 90 W/cm$^2$. Beyond this point the slope of the curve for the processed samples changes significantly. This change in slope is a result of the additional nucleation sites which become active at higher heat fluxes (e.g., smaller radius cavities); the probability of active nucleation sites in this region is expected to be different for each test sample.

Samples S1-S3 have very similar heat transfer coefficient curves. This is expected since these surfaces have nearly identical surface area ratios. The curves of S1-S3 diverge at around 90 W/cm$^2$. Beyond this point the heat transfer coefficients follow the trend of increasing microstructure peak-to-valley height. The microstructures act like cooling fins for heat to be conducted through. As the height of the microstructures increase, the more efficiently the surface is cooled and thus increases the heat transfer coefficient. Sample S4 consistently has higher heat transfer coefficients than all the other samples. The overall enhancement of the heat transfer coefficients can be attributed to efficient nucleate boiling and an increase in the surface area ratio. The increase in the slope beyond the divergent point (at 90 W/cm$^2$) can also be related to the microstructure peak-to-valley height. As the height is increased the change in slope is also increased. The enhancement of the heat transfer coefficients at high heat fluxes is due to a combination of the tall nature of the microstructures [82] and the higher surface area ratio, as well as a higher probability of finding a nucleation site which can be activated at higher heat fluxes.
As for the Onset of Nucleate Boiling (ONB), it can be seen from Figure 3.3 and Table 3.2 that the ONB occurred at much lower wall superheat values for the processed surfaces when compared to the polished surface. This is due to the nano and microscale features present on the processed surfaces which allow for nucleation sites that activate with less energy. Figure 3.5 shows the difference in nucleation between S4 and the polished surface at low heat fluxes further supporting the above mentioned statement. As can be seen from Figure 3.5, sample S4 has a much higher nucleation site density and produces much smaller bubbles that quickly detach from the surface compared to the polished sample. This difference in bubble size and departure rate and diameter also occurred at higher heat fluxes and surface temperatures. Sample S4 was found to have the smallest ONB at around 7 °C compared to about 10 °C for the polished sample.
Figure 3.5  Near Onset of Nucleate Boiling: Top – polished sample, 13 °C superheat and 3 W/cm², Bottom – S4, 7.7 °C and 2 W/cm²
CHAPTER 4
LEIDENFROST TEMPERATURE SHIFT AND FILM BOILING ENHANCEMENT

4.1 Film Boiling Surfaces and Characterization

A polished 304 stainless steel surface sample was used as a reference and baseline for comparison to the laser-structured surfaces. This polished sample was first wet-sanded with 600 grit sandpaper and then polished to a mirror finish with the use of a series of buffing compounds. The impact of the surface morphology as fabricated via FLSP on the Leidenfrost point was then studied with five distinct laser-processed surfaces. Each processed surface was fabricated with a different combination of laser fluence and number of laser shots in order to produce unique geometric microstructures. The fluence and shots were chosen to vary the shape and spacing of the microstructures while keeping the average height approximately constant. Table 4.1 highlights the various parameter values used to process each sample as well as measured surface specific characteristics of the actual sample used in testing. The separation between microstructures increases with each sample number.

<table>
<thead>
<tr>
<th>Design Parameters</th>
<th>Measured Parameters</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample Name</td>
<td>Fluence (J/cm²)</td>
</tr>
<tr>
<td></td>
<td>Shots</td>
</tr>
<tr>
<td></td>
<td>Average Height (µm)</td>
</tr>
<tr>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
</tr>
<tr>
<td>S1: BSG – Mounds</td>
<td>1.1</td>
</tr>
<tr>
<td>S2: BSG - Mounds</td>
<td>1.1</td>
</tr>
<tr>
<td>S3: ASG - Mounds</td>
<td>1.4</td>
</tr>
<tr>
<td>S4: ASG - Mounds</td>
<td>1.4</td>
</tr>
<tr>
<td>S5: NC - Pyramids</td>
<td>0.1</td>
</tr>
</tbody>
</table>
A full 3D surface profile was obtained for each of the fabricated samples using a Keyence VK-X100 laser confocal microscope. From this data, the average height, surface area ratio, and surface roughness were measured. The average height represents the average peak-to-valley height of the microstructures. The surface area ratio or roughness factor [84] is the total surface area of all the microstructures divided by the cross sectional area. The structure separation was determined through a Fast Fourier Transform (FFT) analysis of scanning electron microscope images. The contact angle was measured with a 1 µL drop of deionized water at room temperature using a Ramé-Hart Model 590 F4 Series Goniometer and Tensiometer. Contact angles were measured on 5 drops in 5 different places and each drop was measured 10 times. This gave an average angle deviation of about 0.7°.

Figure 4.1 shows the SEM images and 3D profilometry scans of the unique geometric surface structures (S1 through S5) used in the Leidenfrost experiments. Samples S1 and S2 contain BSG-mounds and are characterized by smooth round tops. Samples S3 and S4 contain ASG-mounds that were fabricated with increased laser fluence and are characterized by deep holes separating pointed structures. Sample S5 contains NC-pyramids, which are densely-packed pyramidal structures fabricated with many laser pulses at a low laser fluence. The gradual increase in structure size and separation can also be easily seen from the 3D scan images in Figure 4.1.
Figure 4.1 SEM (left panel) and 3D profile images (right panel) of the tested surfaces
The creation of these surface micro/nanostructures also had an effect on the contact angles. The polished reference surface was found to have a contact angle of 80°. The addition of micro/nano structures reduced the contact angle for all surfaces. Figure 4.2 shows the resulting contact angles for each of the surfaces. Surfaces S3 and S4 are considered “superwicking” surfaces and have a contact angle of 0°. These two surfaces are similar to previously published accounts of superwicking surfaces fabricated using FLSP [85], [86]. The contact angles measured do not match angles predicted by the Wenzel model [84] as the Wenzel model does not take into account surface chemistry and the presence of nanoparticles which promote capillary wicking which reduces the contact angle.

![Contact angles of tested surfaces. The sample magnification is different to properly show the contact angle of each sample](image)

**Figure 4.2** Contact angles of tested surfaces. The sample magnification is different to properly show the contact angle of each sample

### 4.2 Leidenfrost Experiment

The method used to determine the Leidenfrost temperature was the droplet lifetime evaporation method [29]. This method consists of placing a liquid droplet on a
heated surface and measuring the evaporation time. This method essentially results in an inverse of the boiling curve given in Figure 1.1. The Leidenfrost point corresponds to the surface temperature at which the largest evaporation time occurs. A 4.2 µL (1 mm radius) deionized water droplet was chosen for this experiment in order to ensure that the radius was smaller than the capillary length \( a = \sqrt{\gamma/\rho g} \), where \( \gamma \) is the surface tension, \( \rho \) is the liquid density, and \( g \) is gravitational constant. When the radius is smaller than this length, the droplet is nearly spherical when in the Leidenfrost state [23]. For water, the capillary length is around 2.5 mm. Droplets were also released as close to the surface as possible in order to minimize the impact velocity and corresponding Weber number \( (We = \rho V^2 R/\gamma) \), where \( V \) is the droplet velocity and \( R \) is the radius of the droplet [87]. Ten droplet evaporation times were recorded at each temperature and the average value was plotted. Only droplets that landed softly on the surface and remained completely intact during evaporation were considered for measurement. A Ramé-Hart precision dropper was used to control the size and placement of the droplets. Figure 4.3 shows a CAD illustration of the Leidenfrost experimental setup.
The sample surfaces were fabricated from a 304 stainless steel block with a diameter of 64 mm and a thickness of 15 mm. Because a droplet in the Leidenfrost state tends to move around on the surface in a nearly frictionless manner, a conical depression was machined with a 1° slope and a depth of 0.4 mm at the center of the test surface in order to keep the droplet from rolling off the test area. Note that this machining step was carried out before the FLSP process. The entire conical depression was processed in order to ensure that the droplet was always on the processed surface. The sample surface temperature was controlled through the use of five cartridge heaters (Omega) implanted inside a heating block and connected to a programmable PID temperature controller (Ramé-Hart). A K-type thermocouple was used to monitor the temperature as part of the feedback loop and was embedded 0.8 mm below the center of the conical depression. An additional thermocouple was placed on the outer edge of the conical depression to measure the uniformity of the surface temperature. The temperature on the outer edge was consistently 3 °C less than the center temperature, which was less than 1% of the average operating temperatures in this experiment.

Figure 4.3 Leidenfrost experimental setup
4.3 Results and Discussion

Figure 4.4 shows the data obtained from the Leidenfrost experiments. The Leidenfrost temperature was found to be 280 °C for the polished surface. This number agrees well with Tamura and Tanasawa [30] who reported a Leidenfrost temperature of 290 °C for a droplet of 1.88 mm diameter. The Leidenfrost temperature for surfaces S1, S2, S3, S4, and S5 were 316 °C, 340 °C, 360 °C, 405 °C and 455 °C, respectively. The error bars shown indicate the standard deviation of the ten droplet evaporation times recorded at each temperature. As can be seen from the data, extraordinary shifts in the Leidenfrost temperature, as high as 175°, have been observed for the multiscale micro/nanostructures.

Figure 4.4  Droplet lifetimes with respect to surface temperature for a polished 304 stainless steel sample and five laser-processed samples with varying micro and nanostructures. The temperatures indicated with arrows are the corresponding Leidenfrost temperatures for the given surface.
An increase in the Leidenfrost temperature is typically attributed to a reduction of the contact angle, an increase in the surface roughness, or an increase in the nanoporosity [36]–[38], [88]. Although these properties are often interrelated, controlled experiments have indicated that each of these properties can independently affect the Leidenfrost temperature [37]. Indeed, the controlled increase of the Leidenfrost temperature demonstrated by the series of surface morphologies described in Table 4.1 is attributed to a dynamic balance of each of these factors. For the surfaces studied here, neither the RMS surface roughness nor the surface area ratio provides a direct correlation to a controlled increase in the Leidenfrost temperature. Shifts corresponding to samples S1, S2, and S3 relative to the polished sample can be partially attributed to the gradual reduction in contact angle across this series. Further increases in the Leidenfrost temperatures for samples S4 and S5 cannot be explained by the reduction of the contact angles since the contact angle is 0° for both S3 and S4 and the 15° contact angle of S5 is the highest of any of the laser-processed surfaces. Rather, the increase in the Leidenfrost point from S3 to S4 as well as the extraordinary increase for S5 is primarily attributed to increased nanoporosity. Kim et al [37] explained that nanopores act as sites for heterogeneous bubble nucleation and therefore reduce the temperature difference required for heterogeneous nucleation, consequently resulting in an increase of the Leidenfrost temperature. The degree of nanoporosity is related to the thickness of the nanoparticle layer on the microstructure surface, which is predominantly a function of the number of pulses incident on the sample under the FLSP conditions described here since each laser pulse generates nanoparticles that can
redeposit on the surface. The data in Table 4.1 indicates that sample S4 was fabricated with nearly 1000 more pulses per spot, equivalent to 3 times as many pulses per spot, as sample S3. Sample S5, which exhibited the largest increase in the Leidenfrost temperature, was fabricated with over 33 times as many pulses per spot as sample S4. Figure 4.5 shows a cross-section of the layer of nanoparticles on a NC-pyramid structure taken with a transmission electron microscope. The nanoparticle layer is greater than 4.6 μm thick on the upper portion of the NC-pyramid, which corresponds to nearly one third of the total structure height.
Figure 4.5 Transmission electron microscopy (TEM) images of a 4.6 μm-thick layer of nanoparticles that have redeposited on the NC-Pyramid structures (sample S5) during fabrication. The scale bars in the lower left corners of (a) and (b) are 0.5 μm and 100 nm, respectively. Note that the layer consists of a densely packed array of spherical nanoparticles.

The presence of these self-assembled nanoparticle layers on top of the microstructures plays a major role during intermittent solid-liquid contacts that can
occur when a moving droplet interacts with the surface structures. The nanoparticles promote further wetting of the surface and heterogeneous nucleation during intermittent contact. Kim et al [37] reported that during these intermittent contacts, the velocity of the vapor generated during the heterogeneous nucleation can be greater than that of the critical velocity of the Kelvin-Helmholtz instability. When this occurs, the liquid-vapor interface can be disrupted and the stable vapor film can be destroyed, thus increasing the Leidenfrost temperature.

Intermittent contacts can result from two phenomena. First, surface microstructures may protrude into the liquid droplet when the peak-to-valley height is roughly equal to the vapor layer thickness. It has been reported in the literature that the vapor layer thickness can be in the range of 10-100 µm thick [22]–[24], [89], which is around the same range as the microstructure heights of our surfaces. This hypothesis has also been reported by Kim et al [37], who fabricated 15 µm tall rods on the heating surface. The second way in which droplets can intermittently contact a surface occurs when the momentum of a moving droplet overcomes the resistance of the vapor film. In this case, intermittent contact between the liquid droplet and surface is more likely to occur with increased microstructure spacing as illustrated in the schematic of Figure 4.6 for mound structures (left) and NC-pyramid structures (right). Indeed, there is a correlation between increased microstructure separation (see Table 4.1) and an increase in the Leidenfrost temperature evident in Figure 4.4.
Figure 4.6  Schematic of the impact of nanostructure spacing on the liquid-solid interface in the film-boiling regime. As the spacing between surface structures increases, distortions in the interfacial layer can form that may lead to intermittent contact by moving droplets. The left panel represents the mound structures (S1-S4) and the right panel represents NC-pyramid structures (S5).

Indications of substantial intermittent contact between the liquid droplet and the surface were observed for sample S5; intermittent contact was manifested as brief periods in which the direction of motion of the droplet abruptly changed. This intermittent contact combined with nanoparticle-induced wicking generates violent heterogeneous nucleate boiling, which tends to propel droplets in different directions while increasing the Leidenfrost temperature and decreasing evaporation times above the Leidenfrost temperature.

Finally, an increase in emissivity of the laser-processed surfaces relative to the polished surfaces likely contributes to decreased droplet evaporation times by increasing radiative heat transfer. The processed surfaces have a large emissivity and appear black after processing. The emissivity of sample S2 was measured to be around .75 whereas that of the polished surface was .14. At 500 °C, the evaporation time of a droplet on S1-S4 was reduced between 10-15% compared to the polished sample, while
S5 displayed a 33% reduction in evaporation time. Figure 4.7 shows the difference between the processed and unprocessed surfaces.

![Leidenfrost experiment test sample; black area is processed and outer area is unprocessed](image)

**Figure 4.7** Leidenfrost experiment test sample; black area is processed and outer area is unprocessed

### 4.4 Effects of Fouling on the Multiscale Nano/Microstructures

During the Leidenfrost experiment, a simple surface fouling experiment was conducted. The contact angle of each surface was measured directly before Leidenfrost measurements were taken and was continually monitored throughout the measurement process, which lasted about three days for each sample. No significant changes were observed during the measurement process. After testing was completed, the samples were kept in the open environment (exposed to dust and other particulates) and the contact angles were periodically checked. The contact angles of the samples increased with environmental exposure, eventually rendering the surface hydrophobic as was also reported in [90]. Figure 4.8 illustrates the changes in contact angle observed in S1 and S2.
The contact angle of S1 was measured 14 days after testing was completed and was found to be hydrophobic. The S2 contact angle was measured 28 days after testing was completed and was found to be slightly more hydrophobic than S1. These samples were then placed in an ultrasonic bath with isopropyl alcohol for twenty minutes, rinsed with deionized water, dried, and then the contact angle was measured again. The contact angles returned to nearly the original value as a result of cleaning. The extreme temperatures during the Leidenfrost experiments did not affect the structures on the processed sample. It was found that the contact angle change could easily be restored with simple cleaning. In applications where significant fouling can be suppressed, these structures are expected to retain their wettability.
CHAPTER 5
SELF-PROPELLED LEIDENFROST DROPLETS ON HEATED SURFACES WITH ANGLED MICROSTRUCTURES

5.1 Angled Microstructures for Self-Propelled Droplet Motion

As stated earlier, the Leidenfrost phenomenon in combination with an asymmetric microstructure can result in a self-propelled droplet motion. The advantage of these types of surfaces is that they provide liquid transport without the use of any moving parts or additional components. In theory, these types of characteristics can be integrated into other functionalized heat transfer surfaces for further enhancement. The idea is that surfaces that induce liquid motion will have higher heat transfer coefficients and critical heat fluxes due to the increased convection and increased cool liquid supply. This type of hybrid surface that promotes liquid transport could have a significant effect in pool boiling and especially flow boiling in microchannels, where pressure drop could be significantly reduced due to the self-pumping nature of the surface.

In this experiment, our FLSP surfaces were tested to determine the self-propelling capabilities. As discussed in section 1.1.3, self-propelled Leidenfrost droplets are conventionally achieved with a ratchet-like microstructure. It was also recently found that microposts and pillars tilted at an angle can also provide a self-propelled motion of a liquid droplet and that the droplet directionality was dependent on the microstructure size. In the case of our experiment, the droplet motion directionality was found to be consistently opposite of a conventional ratchet structure regardless of the microstructure size. Figure 5.1 illustrates the Leidenfrost droplet motion directionality corresponding to a conventional ratchet microstructure and the angled self-assembled
microstructures used in the present study. As can be seen on the figure, the distinct surfaces result in opposite droplet motion directionality. This change in directionality and the liquid transport performance are discussed in a later section.

Once again the FLSP technique was used to generate stainless steel surfaces with a quasiperiodic pattern of angled surface microstructures. The angled nature of the microstructures was defined by that incident angle of the laser as shown in Figure 2.3. In the present study, two stainless steel samples were fabricated with microstructure angles of 45° and 10° with respect to the surface normal and then utilized to demonstrate the ability to self-propel Leidenfrost droplets. These samples are characterized by mound-shaped microstructures that are covered in a layer of nanoparticles and are angled versions of Above Surface Growth (ASG) Mound structures [73], [77]. The fabrication parameters as well as relevant surface characteristics are described in Table 5.1. The two samples were fabricated with the same pulse energy. Because the laser was incident on the sample at an angle, the spot on the sample was elliptical and not the same size for each sample. The elliptical beam profile on the target sample
(see Figure 5.2 (A)) is due to the non-normal incident angle of the laser. The parallel and perpendicular dimensions given in Table 5.1 refer to spot size dimensions relative to the laser direction.

Table 5.1 Laser Parameters and relevant surface characteristics. The angle of the microstructures is defined relative to the surface normal. The spot diameters and structure spacing values are defined as parallel or perpendicular to the direction of the laser.

<table>
<thead>
<tr>
<th>Structure Angle</th>
<th>Pulse Energy (µJ)</th>
<th>Number of Laser Shots</th>
<th>Spot Dia. (µm) (Parallel)</th>
<th>Spot Dia. (µm) (Perpendicular)</th>
<th>Peak-to-Valley Height (µm)</th>
<th>Structure Spacing (Parallel) (µm)</th>
<th>Structure Spacing (Perpendicular) (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>45</td>
<td>700</td>
<td>500</td>
<td>328</td>
<td>232</td>
<td>17</td>
<td>27</td>
<td>17</td>
</tr>
<tr>
<td>10</td>
<td>700</td>
<td>500</td>
<td>188</td>
<td>224</td>
<td>57</td>
<td>29</td>
<td>30</td>
</tr>
</tbody>
</table>

Figure 5.2 SEM images of the 45° (top) and 10° (bottom): A) Laser damage site on the target sample after exposure to 500 laser pulses with a pulse energy of 700 µJ (400X and 100 µm scale bar), B) Looking at sides of structures (600X and 100 µm scale bar), C) Looking along the microstructures (1200X and 50 µm scale bar), D) Looking normal to the surface (1200X and 50 µm scale bar). The arrows represent the projected direction of the incident laser pulses.

Scanning Electron Microscope (SEM) images of the samples taken from several angles are shown in Figure 5.2. The structure spacing values in Table 5.1 are obtained by a 2D Fast Fourier Transform (FFT) analysis of the images in Figure 5.2(C) and represent the peak values in the directions parallel and perpendicular to the laser. The peak-to-valley structure heights were measured using a 3D Confocal Laser Scanning Profilometer (Keyence, VK-X200); these images are shown in Figure 5.3. The markedly smaller peak-to-valley structure heights of the 45° sample relative to the 10° sample are
due to the larger spot size (see Table 5.1 and Figure 5.2(A)) and thus decreased laser fluence on the sample. This relatively lower laser fluence results in decreased surface fluid flow during processing and thus reduced structure development [73].

![3D laser confocal images of both the 45° sample and 10° sample](image)

**Figure 5.3** 3D laser confocal images of both the 45° sample and 10° sample

The two samples were superhydrophilic; this was determined by measuring 0° contact angles with a Ramé-Hart Goniometer. Due to the superwicking nature of the surface, the droplet would perfectly wet the surface and was not able to be directly imaged. The superhydrophilic nature of the surface is a result of the fabrication process as stated earlier and shown by the previous experiments.

### 5.2 Self-Propelled Droplet Motion Experiments

Each of the experimental samples was fabricated on a 2.5” x 1” piece of polished 316 stainless steel plate. The laser-structured area was 0.5” wide and 2” long and was located in the center of the plate. Each processed sample was then placed onto a leveled copper heating block heated by five cartridge heaters. Four K-type thermocouples (Omega 5TC-GG-K-36-72) were epoxied (Omega OB-200-2) to the surface of the test sample in order to accurately determine the surface temperature.
The surface temperature was monitored with the use of LabVIEW. The surface temperature was controlled through the use of a Ramé-Hart precision temperature controller (Ramé-Hart 100-50) and a thermocouple feedback loop. Droplet size and dispensing was controlled by a Ramé-Hart computer controlled precision dropper (Ramé-Hart 100-22). Deionized water was used as the working fluid with droplet sizes of 10.5 µL (diameter of 2.8mm). This size was chosen because it corresponds to the droplet size that easily detaches from the needle by gravity alone. Droplets were released close to the surface to limit the effects of the impact velocity. From high speed video analysis, using two successive frames immediately before impact, it was determined that the droplets impacted the surface with a velocity of approximately 20 cm/s. This corresponds to a weber number \( \text{We} = \left( \frac{\rho D_0 V_0^2}{\gamma} \right) \) of around 1.5 (\( \rho = 998 \text{ kg/m}^3 \) and \( \gamma = 73 \text{ mN/m} \) at room temperature). Where \( \rho \) is the liquid density, \( D_0 \) is the droplet diameter, \( V_0 \) is the impact velocity, and \( \gamma \) is the surface tension. All videos were recorded with the use of a high speed camera (Photron Fastcam SA1.1), set at 250 frames per second. Figure 5.4 shows a schematic of the experimental setup.
From the high speed video images, droplet velocities across the samples were calculated using an in-house Matlab droplet tracking program which tracks the centroid of the droplet. This program calculates the instantaneous horizontal droplet velocity between successive frames and then gives an average velocity profile for the entire droplet motion. The program was validated against droplet velocities manually calculated from still images using a movie editing software; the two methods were in excellent agreement.

5.3 Results and Discussion

Figure 5.5 shows the data obtained from the droplet motion experiments for the two distinct angled microstructures investigated; velocities presented correspond to the maximum droplet velocities at the edge of the processed surface. Each velocity data point corresponds to an average velocity of ten individual droplets and the error bars correspond to the standard deviation of these ten droplets. As can be seen from the
graph, the two curves have similar features yet significant differences. Both curves exhibit a local maximum towards lower surface temperatures. The 45 degree sample has a maximum velocity of 19.2 cm/s at a surface temperature of 310 °C while the 10 degree sample has a maximum velocity of 13.5 cm/s at a surface temperature of 256 °C. For both samples, droplet velocities gradually decrease as the surface temperature is decreased from the maximum observed velocities. At the lowest temperature recorded, both samples displayed a spike in the droplet velocity. Velocities could not be recorded below 225 °C as violent nucleate boiling resulted in the destruction of the liquid droplets. As the surface temperature is increased beyond the value at the maximum droplet velocity, droplet velocities again decrease but at a much faster rate, especially for the 45 degree sample.

![Graph showing droplet velocities with respect to surface temperature for both processed samples](image)

**Figure 5.5** Droplet velocities with respect to surface temperature for both processed samples

From Figure 5.5, it can be seen that there are two regions of interest. These regions of interest correspond to temperatures above and below the Leidenfrost
temperature of the surface. The Leidenfrost temperatures for the 10° and 45° sample were estimated to be 330 °C and 360 °C, respectively. The Leidenfrost temperature of each surface was estimated by the change in the slope of the curve and the standard deviations of the velocities (Figure 5.5) and the visual differences in the droplet behavior, captured with the high speed video images (Figure 5.6). Looking at Figure 5.5, the slope of the curves changes at 330 °C and 360 °C for the 10° and 45° samples, respectively. To the left of these temperatures, the standard deviations are significantly larger. This indicates that intermittent contact, as indicated in section 4.3 and [37], [51], [91], is occurring and the droplet is not in a stable film boiling state. Because this intermittent contact promotes an explosive type of energy transfer, it results in a wide range of droplet velocities and thus larger standard deviations. Figure 5.6 shows droplets at different locations for temperatures near the Leidenfrost transition temperature for both samples. It can be seen from these images that there is a distinct visual difference in the images of the droplets between the two temperatures. For both samples, the droplets appear to be white in color and not very spherical at temperatures below the Leidenfrost temperature. This indicates that the droplets are being disturbed by intermittent contact. At these temperatures, it can also be seen from the high speed video that the droplets tend to jump and bounce much more frequently and eject smaller satellite drops. This is characteristic of not having a fully developed vapor film between the droplets and the heated surface and thus below the Leidenfrost region. Flow/thermal instabilities lead to the non-spherical shapes and ejection of satellite droplets. At temperatures at or above the Leidenfrost temperature, the
droplets appear to be very spherical and clear in color. This is due to the stable vapor film below the droplet. The Leidenfrost temperatures estimated by this technique are within the expected range for surfaces created by a femtosecond laser process as indicated earlier from the Leidenfrost shift experiment. The variation in the Leidenfrost temperature is due to the differences in the surface microstructures.

![Figure 5.6](image)

**Figure 5.6** Droplets at various positions along the leveled sample at temperatures above and at the respective Leidenfrost temperatures with times from initial contact. A) 10° sample at 320 °C, B) 10° sample at 330 °C, C) 45° sample at 340 °C, D) 45° sample at 360 °C

It can be seen from the graph (Figure 5.5) and the high speed images (Figure 5.6) that there are two distinctly different mechanisms that aid to the motion of the droplet. The dynamic balance between these two mechanisms results in the characteristics of the velocity curves shown in Figure 5.5. At temperatures below the Leidenfrost temperature, droplet motion results from the directional ejection of vapor due to intermittent contact between the liquid droplet and microstructures [37], [51], [91]. When this intermittent contact happens, heterogeneous boiling occurs and vapor
is violently released from the droplet resulting in higher droplet velocities. At temperatures above the Leidenfrost temperature, a stable vapor film is created and thus intermittent contact between the droplet and microstructures is less likely to happen. At these temperatures, the droplet motion mechanism is dominated by viscous stresses that drag the droplet in the direction of the vapor flow. Because this mechanism is not abrupt like in the case of intermittent contact, it produces a smaller but more stable force on the droplet and consequently slower velocities. The local maximums for both samples are most likely due to an optimal combination of these two mechanisms.

The overall larger velocities of the 45 degree sample relative to the 10 degree sample can be attributed to the difference in microstructure angle between the two samples. The 45 degree angle results in a more favorable horizontal force on the droplet during intermittent contact at lower temperatures. The differences at higher temperatures can be explained by a combination of the microstructure size and the viscous drag mechanism. For the 10 degree sample, the droplet velocity decreases very rapidly with increasing temperatures to reach what seems to be a local velocity plateau (e.g., 370 °C). At temperatures higher than 370 °C in the case of the 10 degree sample, droplet velocities increase with increasing temperatures due to the increased heat flux to the droplet and a corresponding higher vapor flow velocity. A similar trend was also reported in the literature [53] with ratchet structures. No velocities were recorded for the 45 degree sample above 380 °C because the droplet no longer displayed a preferential directionality. In these temperature ranges there is little to no intermittent
contact and the dominant mechanism is the viscous drag mechanism. The 45 degree sample has microstructure heights significantly smaller than the 10 degree sample (see Table 5.1). This difference in height is the main reason for the different trends at higher temperatures and the lack of directionality for the 45 degree sample. The viscous drag mechanism is an interaction between the vapor flow, the microstructure geometry, and the droplet base. At high temperatures, the vapor layer is fully developed and relatively thick. In the case of the 45 degree sample, it is likely that the vapor layer is thick enough to effectively isolate the droplet from the surface microstructures and therefore inhibiting interaction between droplet and surface microstructures, hence no self-propelled motion. Since the 10 degree sample has significantly taller microstructures (see Table 5.1), this interaction remains intact at high temperatures and thus the propulsion still occurs.

5.4 Self-Propulsion Mechanism
Unlike previously published studies in the literature, the direction of liquid droplets in the present study was found to be opposite to that of conventional ratchet microstructures regardless of surface temperature and structure size (refer to Figure 5.1). The mechanism that is widely used to describe the motion of a Leidenfrost droplet on a ratchet surface is known as the viscous drag mechanism [51]. This mechanism is based on the preferential direction of vapor flow underneath the droplet. This vapor flow drags the droplet in a direction opposite to the tilt of the ratchet as a result of viscous stresses. Our experimental results could not be, however, explained by this mechanism; hence a new mechanism for a self-propelled droplet on asymmetric three
dimensional self-organized microstructured surfaces is proposed. A schematic drawing of the proposed mechanism is shown in Figure 5.7. It has been shown experimentally [51] that the vapor from an evaporating liquid droplet flows in the direction of descending slope on the teeth of a ratchet (x-direction on Figure 5.7-Top). When the flow encounters the next ratchet, it is redirected 90° (y-direction on Figure 5.7-Top) and flows down the ratchet channels [51]. Flow in the y-direction is unobstructed; therefore there exists only a net force in the x-direction, which results in the motion of the droplet with the same direction as the vapor flow (Figure 5.7-Top). This also means that each of the ratchet segments is cellular in the x-direction and develops a similar, yet independent, flow and force.
In principle, the physics of the viscous mechanism must also apply to the angled FLSP microstructures. However, as shown by the present experimental results, this theory does not fully describe why droplet motion on the angled FLSP samples is in the opposite direction of that on ratchet structures. If the angled FLSP microstructures were reduced to their simplest form, they would be similar to the ratchet microstructures, however with one critical difference. Because the angled FLSP microstructures are three-dimensional and self-organized, they result in no channel in the $y$-direction unlike with the ratchet structures. This difference is the key to understanding why the direction of droplet motion is different between the two structures. When vapor is released from a droplet on angled FLSP microstructures, the released vapor initially follows a very similar
profile as in the case of the ratchet structures. However because with the angled FLSP microstructures, there is no continuous path in the y-direction, the vapor flowing into the spacing surrounded by neighboring microstructures is forced to be redirected nearly 180° (see Fig. 5.7-Bottom). The redirected vapor drags the droplet with it through the viscous forces and causes the droplet to move in the opposite direction than that reported with the ratchet microstructures. Unlike the ratchet structures, the angled FLSP microstructures provide x and y direction cellular spacing, each independently generating a net force on the liquid droplet. Given the three dimensional and self-organized nature of the angled FLSP microstructures, it is possible to have local vapor flows opposite to the droplet motion, however they do not derail it from its main trajectory.
CHAPTER 6

CONCLUSIONS AND RECOMMENDATIONS

6.1 Pool Boiling Enhancement Conclusions

Through the use of femtosecond laser surface processing (FLSP), functionalized stainless steel surfaces were fabricated with hierarchical micro/nanostructures. The structures consist of mound like microstructures covered by a dense layer of nanoparticles resulting in highly wetting and superwicking surfaces with augmented surface area and increased nucleation site density. Pool boiling heat transfer experimental results revealed that using the laser processed stainless steel surfaces, the critical heat flux can be increased from 91 to 142 W/cm$^2$ while also increasing the maximum heat transfer coefficients from 23,000 to 67,400 W/m$^2$-K. Increases in the critical heat flux has been attributed to increase in surface wettability and wicking capabilities. Processed surfaces with tightly packed mound structures resulted in better wettability and wicking and thus resulted in higher critical heat fluxes. The enhancement in heat transfer coefficients are related to the surface area ratio, structure height, and the active nucleation site density. It was found that the higher the surface area ratio the higher the heat transfer coefficients. It was also found that microstructure peak-to-valley height can result in a cooling fin effect and further increase the heat transfer coefficients at high heat fluxes. The heat transfer enhancement at low heat fluxes can be attributed to the increase in nucleation site density and more efficient bubble departure dynamics.
6.2 Leidenfrost Shift and Film Boiling Enhancement Conclusions

Extraordinary high shifts in the Leidenfrost temperature, up to 175°, have been achieved through the use of multiscale micro/nano structures formed via femtosecond laser surface processing (FLSP). A series of laser-processed surfaces fabricated by varying laser fluence and number of shots incident on the sample demonstrated a controlled increase in the Leidenfrost temperature. Shifts in the Leidenfrost temperature were attributed to reductions in contact angle and substantial capillary wicking due to nanoporosity during intermittent contacts of the droplet with the heated surface. The greatest shift was seen on NC-pyramid structures, which are characterized by 14 μm tall surface features separated by 25 μm that were blanketed with a thick layer of self-assembled nanoparticles. This combination of feature spacing and nanoporosity resulted in significant intermittent contact of the droplet with the surface near film boiling regime, which promoted capillary wicking and nucleate boiling. Further research is needed to determine the limits of the self-assembled nanoparticles on shifting the Leidenfrost temperature and their durability in harsh environments.

6.3 Self-Propelled Leidenfrost Droplet Conclusions

It has been shown in the present work that angled microstructures created through the use of femtosecond laser surface processing can be used to effectively propel liquid droplets in the Leidenfrost state across a heated surface. Angled FLSP microstructures consist of mound-like structures with a rounded top that lean at a specific angle. These structures can be created at nearly any inclination angle. For this study, two surfaces were created with angles of 45° and 10° with respect to the surface normal. Self-propelled droplet motion experiments resulted in maximum velocities of
13.5 cm/s and 19.2 cm/s for the 10° and 45° samples, respectively. These maximum velocities occurred at temperatures well below the corresponding Leidenfrost temperatures of the surfaces. The high velocities at temperatures below the Leidenfrost temperatures of the surfaces are due to intermittent contacts of the liquid droplet with the surface microstructures. When this occurs more energy is transferred to the droplet and vapor is violently ejected from the droplet. This vapor is preferentially directed by the microstructures into one general direction. In comparison to conventional ratchet structures, the angled FLSP microstructures result in droplet motion in the opposite direction. This change in the direction of the droplet motion is due to the three dimensional self-organized nature of the angled FLSP microstructures which leads to a redirection of the vapor flow. The viscous stress forces of the redirected vapor flow move the droplet in a direction opposite to that of the conventional ratchet structures that have been previously reported in the literature.

6.4 General Recommendations

It has been shown in this work that functionalized surfaces created with a femtosecond laser processing technique can have a very significant impact on the heat transfer performance of a metallic surface. The purpose of this work was to serve as a preliminary study of optimizing a stainless steel heat transfer surface with the use of a femtosecond laser surface processing technique. For most heat transfer applications, a high heat transfer coefficient and high critical heat flux are desirable. This is only achieved in the nucleate boiling region. In order to achieve even higher heat transfer coefficients and critical heat fluxes, it is desirable to have surface features with high
surface area ratios and high wetting and wicking abilities. It is believed that if these surface microstructures are also angled it could help promote liquid transport above the heating surface and improve natural convection as well as draw in cool liquid to the hot surface. An improvement of natural convection would result in a higher heat transfer coefficient while drawing in cool liquid to the heating surface would result in a higher critical heat flux. It is also believed that a similar type of surface morphology could have a very large impact in flow boiling in microchannels. In this scenario, there is a combination of liquid and vapor moving through a small containment area. This resembles the motion seen in the self-propelled Leidenfrost droplet experiments. It is believed that the integration of angled microstructures in a microchannel flow boiling setup could result in very high heat transfer coefficients and very low pressured drops due to the self-pumping nature of the surface.

The FLSP technique results in an almost infinite combination of surface parameters. The presented results are very promising for high heat transfer enhancement with metallic surfaces and it is believed that this technique truly has the ability to have a significant impact on the efficiency of interfacial heat transfer in future applications. Thus, further experimental studies should be conducted to determine the absolute potential of this unique surface processing technique.
REFERENCES


APPENDIX

ADDITIONAL EXPERIMENTAL SETUP INFORMATION

A.1 Pool Boiling Experimental Setup Modifications

Throughout the development and design process of each experiment, modifications were made to the experimental setup as well as the measurement procedure. The experimental setups described in the previous sections are the final version after modifications. The pool boiling experimental setup was the most complicated and thus resulted in the most changes during the development period. The heating section of the pool boiling setup required the most modifications and development. These modifications were aimed at improving the sealing of the system as well as increasing the heat conduction efficiency.

The first issue that arose during the development period was effectively sealing the heating system and preventing leaks. Initially the insulating bushing surrounding the heating block was made from PTFE. This material provided good thermal insulation and high temperature stability as well as being easy to machine. This insulating bushing was not originally designed without the O-ring grooves near the boiling surface. The system was sealed by a single O-ring located between the bottom part of the aluminum housing and heating block. This O-ring is depicted in Figure 3.2. The thermocouple holes were drilled through the aluminum housing, the insulating bushing, and the heating block. To seal these holes, epoxy was coated on the outside of the aluminum housing where the thermocouples entered. This technique effectively sealed the heating system and
prevented leaks. Although this technique sealed the system it resulted in other problems. Since the heater system was sealed at the bottom part of the heater assembly, water was able to seep between the insulating bushing and heating block all the way down to the O-ring. This resulted in nucleation occurring within this gap instead of at the face of the boiling surface. This also resulted in water coming into contact with the thermocouples. This resulted in very misleading temperature readings. From this it was determined that the system must be sealed closer to the boiling surface.

In order to move the sealing location closer to the boiling surface, O-ring seals were integrated into the top of the insulating bushing as illustrated in Figure 3.2. This effectively sealed the water from coming in contact with the thermocouples and thus realistic temperature readings were acquired. This method worked well with single material heating blocks but did not accommodate multi material heaters as described by the final design. When dissimilar material heating blocks with very thin stainless steel portions like the final design were used, the O-ring seal was typically below the transition between the two metals. This results in the ability for water to come into contact with the copper. Since the copper is much hotter than the stainless steel and has a higher thermal conductivity this results in premature nucleation at this interaction location.

In order to prevent this premature nucleation, an epoxy seal was used to seal between the insulating bushing and the top of the heating surface. Initially a simple two-part 5 minute epoxy was used for this seal. It was found out that this epoxy would
typically fail during operation and delaminate from the insulating surface and then leak. This delamination was a result of the PTFE insulating bushing. Due to the nature of PTFE, it is very difficult to bond to. This can be only achieved by using an etching process to treat the surface before bonding and a special epoxy must be used. From this knowledge, it was determined that a different material was needed. The insulating bushing was then changed from PTFE to PEEK. PEEK offers a nearly identical thermal conductivity and temperature resistance and can easily be bonded to with a normal epoxy. PEEK is also more ridged than PTFE and thus provides more mechanical strength. Because of the soft nature of the PTFE, insulating bushings were easily damaged during the sample removal process. Switching to PEEK resulted in an increased longevity of the insulating bushing.

The epoxy described in the previous section was specially designed to bond to dissimilar materials, like PEEK to stainless steel. This epoxy had a cure time of around 12 hours. Throughout the experimental measurement process, a simple 5 minute epoxy was also tried with this material combination and was found to have nearly identical success as the special epoxy with the benefit of a much faster cure time. When this 5 minute epoxy was used, an hour of cure time in a dry environment was given to ensure that the epoxy had completely cured.

Originally, it was intended for the main heating block to be a continuous material. The goal of this was to prevent any contact resistances between dissimilar metals. The first heating block was made entirely out of stainless steel. This block
contained the thermocouple locations shown in Figure 3.2. This design resulted in a very high heating block temperature at high heat fluxes due to the relatively low thermal conductivity of the stainless steel. From this it was realized that the amount of stainless steel must be minimized.

A stainless steel disk was machined to have a thickness of .125”. This disk was then placed on top of a copper heating block. This stainless steel disk was originally sealed with only O-ring seals as described earlier. Thermal grease was applied between the two materials to reduce the contact resistance. It was found that during testing, the heater block temperatures were not significantly reduced and at higher heat fluxes pressure would build up between the two surfaces and the stainless steel surface would be pushed upward and the seal would be broken. It was also determined that the thermal grease still resulted in a significant contact resistance. In the surface temperature calculation, as described earlier, it was found that the surface temperatures were extremely high when neglecting the contact resistance. The calculated surface temperatures were considered to be unrealistic due to the large contact resistance between the two surfaces. A fastening cap was machined to keep pressure on the heating surface in order to maintain contact between the two materials during operation. Even when contact was maintained, it was still determined that the contact resistance was excessive due to the extremely high surface temperatures calculated. Surface temperatures on the order of 30-40 °C superheat were calculated with no nucleation present at the surface leading to the conclusion that the contact resistance was large and the surface temperatures were unrealistic.
In order to reduce the contact resistance of the interface it was determined that a highly conductive bonding agent was needed. The first thought was to use a solder of some sort. The problem with this was that stainless steel is not meant to be soldered and conventional solders would not work. It was then found that a silver braze could be used to bond copper and stainless steel. The silver braze chosen was in the form of a paste with the flux mixed in with it. The stainless steel surface selected was also chosen to be as thin as possible but thick enough to prevent the laser from ablating through it. In extreme cases the laser can ablate down around 100 microns. Because of this, the stainless steel disk thickness was chosen to be .010” or around 250 microns.

The heating system also went through multiple revisions. Initially a copper heater system was designed to connect with the boiling section by direct contact only. A fixture was made to hold heating system in contact with the boiling section. As mentioned earlier, a direct contact results in a high contact resistance. This large contact resistance between the heater system and the boiling section resulted in a system unable to reach critical heat flux. The cartridge heaters had to be completely maxed out to achieve moderate heat fluxes due to this inefficient connection. Thermal grease was added to this interface but like before it had only a slight effect on improving the efficiency.

It was found that this interface can be efficiently connected by solder due to both materials being copper. A high melting temperature solder was selected to connect the two copper blocks. Initially the weight of the copper heating system was supported
by the solder connection alone. Depending on the critical heat flux of the surface, the solder connection didn’t melt before CHF was reached. In the case of higher heat flux surfaces, the solder would melt before CHF was reached and the heating system would be detached. To prevent this premature detachment, a fixture was made to support the weight of the heater. At high heat fluxes when the solder melted the fixture retained the position of the heating system. The surface tension of the liquid solder would keep the two copper blocks efficiently in contact and heat was conducted through a liquid metal film and the critical heat flux could be attained.

Initially, the cartridge heater system was powered with a PID digital controller with a feedback temperature sensor. The idea behind this decision was that the surface temperature could be directly controlled. This method resulted in a very unstable power supply method. The proportional controller was unable to efficiently control the surface temperature and resulted in a continuously spiking surface temperature which never reached a steady state. The power supply was then changed to an analog variac which supplies a constant power. This resulted in a much more controllable power delivery method.

The thermocouples used for measuring the heat flux were also changed throughout the development period. Initially flexible thin gage exposed thermocouples were used. The motivation for this was due to the tight space that the thermocouple must be maneuvered into. The flexible thermocouples actually turned out to be very difficult to install. The insulation tended to snag on the sides of the thermocouple hole
and then wedge in place. Due to the flexible nature of the thermocouples it was also nearly impossible to determine if the thermocouples were installed to the correct depth. The thermocouples were then changed to an exposed 1/32” diameter thermocouple with a stainless steel sheath. These were much easier to install due to the more rigid nature of the thermocouple. The exposed end was chosen because it was believed that it would give a more accurate reading but in reality these were actually very inconsistent. The exposed end of the thermocouple would typically bend around the end of the stainless steel sheath during installation. This resulted in a temperature measuring point not in the center of the thermocouple hole. Also, these thermocouples were prone to destruction during the installation and removal process due to the fragile nature of the thermocouple tip. Finally, the thermocouples were changed to fully sheathed ungrounded thermocouples. These thermocouples provide a consistent measuring point as well as easy installation and robustness.

The final modification made to the pool boiling setup came with the condenser unit. The condenser was connected to the housing through the use of a 1/8” NPT fitting. At very high heat fluxes the pressure would build up in the system and water began to fill up the condenser and resist draining. This resulted in an increased pressure of the system. The connection fitting was changed from 1/8” to a 1/4” NPT fitting. This resulted in a larger opening to the condenser and prevented the condenser from filling up and maintained the pressure at high heat fluxes.
A.2 Pool Boiling Sample Preparation and Installation

When attaching the stainless steel disk to the copper heating block, a thin layer of braze was evenly spread across the surface of the copper heating block. The stainless steel disk was then placed on top of the copper block and secured with a much thicker stainless steel disk and a C-clamp. The thicker stainless steel disk is used to prevent the edges of the thin disk from curling up during the heating process. The heating block was then heated up with an oxy-acetylene torch. It was heated to the point that the copper just begins to glow red. During this process, some of the braze also bonds the thicker stainless steel disk to the heater block. To remove the thicker disk, the heater block is secured in a lathe and a hammer and chisel is used to knock off the thick securing stainless steel disk. Then any silver braze left on the thin stainless steel disk is cleaned off with sand paper. For future sample manufacturing, it is recommended that a larger diameter (1.5” instead of 1.0”) thin disk be used instead of the smaller one. When a securing disk of 1” diameter is clamped to this much larger thin disk, the securing disk will not be bonded to the heater block due to flowing silver braze. The securing disk can then be easily removed and the excess stainless steel material can easily be turned down to the required diameter.

After the stainless steel disk is bonded to the copper block, the stainless steel is polished with the use of a series of sand papers and polishing compounds. After polishing, the sample is processed with the laser. A layer of high temperature solder is applied to the bottom side of the sample where it connects to the heating system. Adding solder at this time greatly increases the ease of attaching the heating system.
Next O-rings are installed in the insulating bushing and the test sample is pressed into the insulating bushing with the thermocouple holes aligned. These two components are then inserted into the pool boiling aluminum housing. The thermocouples are then inserted through the aluminum housing and into the sample. A 5 minute epoxy is used to secure the thermocouples in place. This epoxy is placed on the outside of the aluminum housing. Once this epoxy has dried the heating system is attached to the bottom of the test sample. Like the test sample, a layer of solder is added to the heater in order to promote adhesions of the two surfaces. The heater is turned on and placed in contact with the test sample. Once the solder at both connections has melted the heater is turned off and rapidly cooled with compressed air. After the system has completely cooled down, the top of the boiling surface is sealed with epoxy and let to cure. When removing the test sample these steps are reversed. It was found that removing the thermocouples is easiest when the securing epoxy is softened by heating with a heat gun.

A.3 Leidenfrost Experiment Additional Information

Unlike the pool boiling experimental setup, the experimental setup used for the Leidenfrost experiment was relatively simpler. Because of this simplicity there were very few experimental modifications. Initially, the heater block used was made from oxygen free copper. It was found that at very high temperatures this copper oxidizes and flakes. In order to keep a clean work environment, the heater block material was changed to stainless steel. At high temperatures the stainless steel block does not produces any flakes or debris.
Due to the conical depression machined into the test surface, it was difficult to measure the contact angle of the sample. In order to accurately measure the contact angle of the sample, an additional area outside the conical depression was also processed. This small area allowed for a flat uniform area to test the contact angle. Due to the highly wetting nature of the processed surfaces, a 1 µL droplet was used to measure the contact angle. This droplet size was ideal because the entire droplet could be imaged with the camera setup. With larger sized droplets and a superhydrophilic surface, the droplet spreads across the surface and the droplet edges used for measuring the contact angle cannot be imaged.

When measuring droplet lifetimes the most difficult parts is releasing the droplet from the needle and onto the surface. Depending on the droplet size, the droplet can be released purely by gravity. This size of the droplet depends on the needle size. Surface tension of the water holds the droplet on the needle. The droplet is released when the gravitational forces balance with the surface tension forces. A smaller diameter needle results in a smaller surface area and a smaller surface tension force which in turn results in a smaller droplet that detaches by gravity. The inverse is also true; a larger needle results in a larger droplet that detaches by gravity. In the case of this experiment, the droplet size selected did not easily detach by gravity alone. In order to detach the droplet from the needle a sliding needle stage was used. This sliding stage also incorporated a mechanical stop. The needle was raised and then slide into the mechanical stop resulting in an inertial force that removed the droplet from the needle. The stopping needle height was carefully selected. If the needle is too close to the
surface the droplet will actually bounce back up and reattach to the needle. If the needle is too far away from the surface the impact velocity can be large enough to cause the droplet to breakup on impact. The ideal height is slightly higher than the location that the droplet reattaches to the needle.

In order to determine the Leidenfrost point the maximum droplet lifetime must be recorded. At temperatures below this point, droplets on the surface violently explode and shatter when they come in contact with the surface. It was found that the most efficient way to find the Leidenfrost temperature is to start at much higher temperatures and then work downward. Typically the curve was started at around 400 °C and then the surface temperature was decreased until there was some instability in the droplet evaporation. At this point the surface temperature was slowly decreased until no more droplet evaporation times could be recorded due to transition boiling. The temperature was then increased to finish the entire curve. Only droplets that remained completely intact during the evaporation process had lifetimes recorded. The droplet lifetimes were calculated with the use of video editing software. The video was cut down to the frames at which the droplet first contacts the surface and when the droplet can no longer be seen. The total evaporation time can then be determined.

A.4 Self-Propelled Droplet Experiment Additional Information

During the fabrication of the test samples a 20 degree sample was also created. This sample was also tested for droplet motion but was found to not produce any preferential motion. Microstructures created at this 20 degree angle do not take on the same form as the microstructures discussed earlier. These microstructures appear more
like plateaus and not mounds as depicted before. This configuration also does not display any sort of directional tilt like the previous structures and thus does not result in any preferential droplet motion.

The droplet velocities were calculated with an in-house Matlab droplet tracking program which tracks the centroid of the droplet. An output of this program is shown in Figure A.1. This figure shows a typical motion profile of a droplet moving across the processed surface. This program was used to track the motion of the droplet and calculate the instantaneous horizontal and vertical velocity as well as displacement. The instantaneous velocities were calculated from the instant that the droplet contacted the surface till the droplet transitioned from the processed area to the unprocessed area. This program gives the ability to see the acceleration of the droplet from the initial zero velocity to the maximum velocity as the droplet leaves the processed area. The frame-by-frame velocities were averaged to give a smooth velocity trend with respect to location. The velocity data presented is the maximum droplet velocity as the droplet transitioned from the processed and unprocessed area.

![Typical droplet motion path across the processed surface](image)

**Figure A.1** Typical droplet motion path across the processed surface

An example of the instantaneous x and y velocities of the droplet as well as the x and y position are given in Figure A.2 and A.3.
Figure A.2 Horizontal and vertical velocities of a droplet moving across the surface
The black dots represent the instantaneous points while the red lines are the averaged smoothed curve.

The velocities recorded and used for the data in Figure 5.5 are the velocities recorded at the end of the processed strip. Typically the velocity used is the maximum velocity at the end of the recording period but in a few instances an error would occur at the end of recording. The horizontal velocity curve shown in Figure A.2 actually displays an error. As can be seen at the end of the curve the velocity profile begins to oscillate. This is an error that occurs as the droplet transitions from the processed to
unprocessed area. In this case the velocity recorded would be the velocity located slightly before where the oscillation occurs.

**Figure A.4** Typical error at the end of the droplet motion. The drastic change in velocity at the end is related to the droplet leaving the viewing and measuring window.

Figure A.4 shows another typical error associated with the droplet program. In this case the calculated velocity spikes at the end of the run. This spike is a result of the droplet leaving the viewing and measuring window. The droplet velocity recorded in this case is the maximum velocity located before the spike.