The Leidenfrost Phenomenon on Structured Surfaces

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Abstract The lifetime of a droplet released on a hot plate decreases when the temperature of the plate increases. But above some critical value of the temperature, the lifetime suddenly increases. This is due to the formation of a thin layer of vapour between the droplet and the substrate. This layer plays a double role: first it thermally isolates the droplet from the plate and second it allows the droplet to "levitate." This effect was discovered by Leidenfrost in 1756, but remains an active field of research nowadays, motivated by a wide range of applications. The Leidenfrost point is affected by the roughness or microstructure of the surface. In this work, a silicon surface with different micro-structured regions of square-pillars was prepared such that there is a sharp transition (boundary) between areas of different pillar spacing. The Leidenfrost point was identified in experiments using water drops ranging in average size from 8 μL to 24 μL and the behaviour of the droplets was recorded using a high-speed digital camera. We show that the Leidenfrost point can vary by up to 120 °C for pillar spacings varying from 10 microns to 100 microns. If the drop is placed on the boundary between structured sections, the drop becomes asymmetric. Drop motion may also be observed and some occurrences of drop spinning have been seen. In this paper we present experimental data on Leidenfrost behaviour of drops placed structured surfaces and on the boundary between surfaces with different micro-structures.

Keywords: Leidenfrost, Boiling, Microstructure, Wettability

1. Introduction

Dutch physician, Herman Boerhaave (1668 – 1738), reported in 1732 that, rather counter-intuitively, ethanol droplets deposited on superheated metal surfaces, did not boil rapidly as expected and indeed persisted longer than droplets on cooler surfaces. In 1756, Dr. Johann Gotlob Leidenfrost (1725 – 1794), made similar observations about the lifetime and mobility of water droplets deposited on a hot silver spoon in his paper, 'De Aquae'. Although Boerhaave was the first scientist to have recorded this strange phenomenon, it is from Dr. Leidenfrost that it takes its name. [1-3]

Above the Leidenfrost Point (LFP), droplets begin to float on an insulating layer of their own vapour (see Fig. 1(b)), resulting in a local maximum for droplet lifetime and a substantial increase in mobility, due to the large reduction in friction. This effect is observed by millions of people every day when water is put into a hot frying-pan. In popular culture, the effect was made famous when US prime-time show 'MythBusters' recreated a demonstration by physicist Dr. Jearl Walker, where a finger can be dipped briefly into molten lead and removed unharmed due to the insulation provided by Leidenfrost. [4-5]

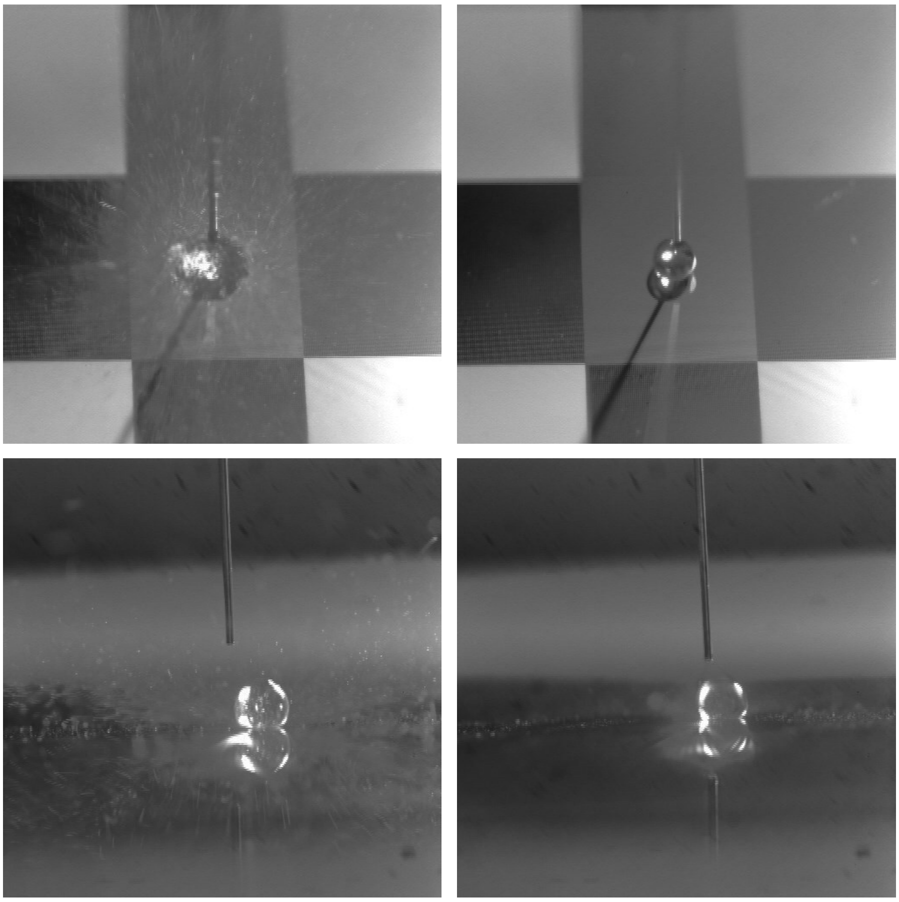

Figure 1 – (a) High-speed camera recording of a droplet below the LFP boiling instantaneously on contact with the heated surface compared with (b) where the surface is above the LFP so the droplet levitates on an insulating vapour layer, preventing rapid boiling. (c) and (d) show side-on views of a droplet boiling and a Leidenfrost droplet.

The droplet levitation resulting from the Leidenfrost effect has been harnessed to direct droplet motion through mechanical macro-scale ratchets and has even been used to produce mazes. [6-7]

In many industrial situations, operating in the Leidenfrost boiling regime is highly undesirable due to the inhibition to heat transfer. In metallurgical processes such as quenching and spray-cooling, the Leidenfrost effect inhibits the control of cooling by means of the insulating vapour layer. Fast cooling is necessary to maintain the mechanical strength of material, while uniform temperature profiles prevent deformation. [8-9]

The Leidenfrost effect has implications in both high heat flux microelectronic cooling systems and in fire-fighting emergencies where it is necessary for rapid heat removal. Indeed, accident reports following the Fukushima nuclear disaster have cited the inverse Leidenfrost effect as a contributor to the inability to cool the fuel system effectively while following emergency procedures. [10-11]

Being able to alter the LFP of systems to either avoid or deliberately enter the Leidenfrost boiling regime is of particular interest across a wide variety of industrial applications. Being able to raise the LFP, by as much as 100 °C, can vastly extend the temperature range where Leidenfrost does not need to be considered for both process design and safety.

Recently, there has been particular interest on the influence of micro- and nano-structures being used to alter the LFP for enhanced boiling. Kwon *et al.* [16] at MIT carried out a systematic study of the effect of microstructure on the LFP of a water-silicon system over a range of micropillar parameters.

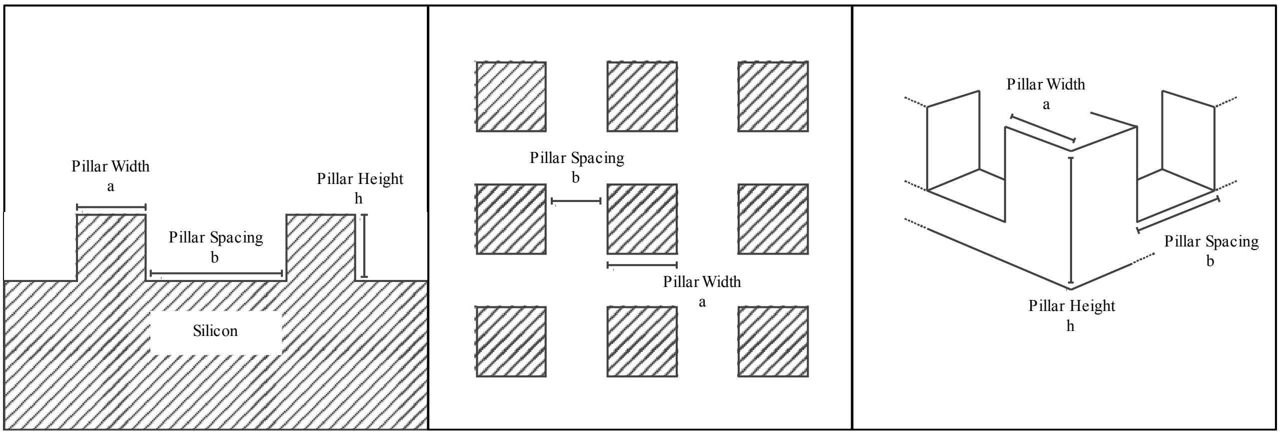

Figure 2 – (a) Cross-sectional view of the micropost array (b) Top-down view of the micropost array (c) 3D Projection of the micropost array.

2. Theory

Although the Leidenfrost Effect has been observed for nearly 300 years, the fundamental principles which govern it are relatively poorly understood and there has been much debate over which factors are most important in defining the LFP.

Surface roughness has always been understood as a key parameter: measurements of the LFP on two surfaces of the same material have been shown to vary greatly depending on whether the material was polished or rough. [12-21]

Kwon *et al.* [16] proposed a mechanism to describe the variation of the LFP due to the presence of the microstructure array. They surmised that as the surface was highly wetting, there would be contact between the tops of the micropost array and the underside of the droplet, and that this contact would result in a force balance between the surface tension ($\Delta P_{cap} \sim \gamma/b$) acting downwards and a vapour pressure (which can be modelled as radial Poiseuille flow) acting upwards. The Leidenfrost state exists if the ratio of these competing vertical forces balances is approximately unity; $\Delta P^*_{vap} / \Delta P_{cap} \sim 1$.

2.1 Structured Surface

A silicon micro-structured surface was produced at the Scottish Microelectronics Centre (SMC). The microstructure design pattern imprinted onto the wafer is illustrated in Fig. 2. All pillars are square-topped; arranged in a grid-like array and have a uniform height of 10 μm.

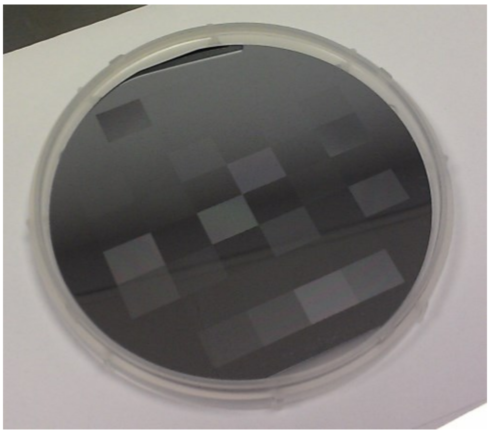

Figure 3 – Photograph of the silicon micro-structured surface produced at the SMC. The regions of different hue indicate different microstructure properties.

	5, 10	10, 10	10, 20	5, 20	
	10, 80	10, 60	10, 50	10, 60	
5, 10	10, 70	10, 20	10, 90	10, 30	5, 10
5, 40	10, 40	10, 100	10, 10	10, 60	5, 5
5, 5	10, 60	10, 10	10, 100	10, 40	5, 40
5, 20	10, 30	10, 90	10, 20	10, 70	5, 20
	10, 70	10, 50	10, 60	10, 50	
	5, 40	10, 40	10, 30	5, 5	

Table 1 – Key: pillar thickness, pillar spacing. Table detailing the microstructure design of the silicon wafer produced, all values are in μm.

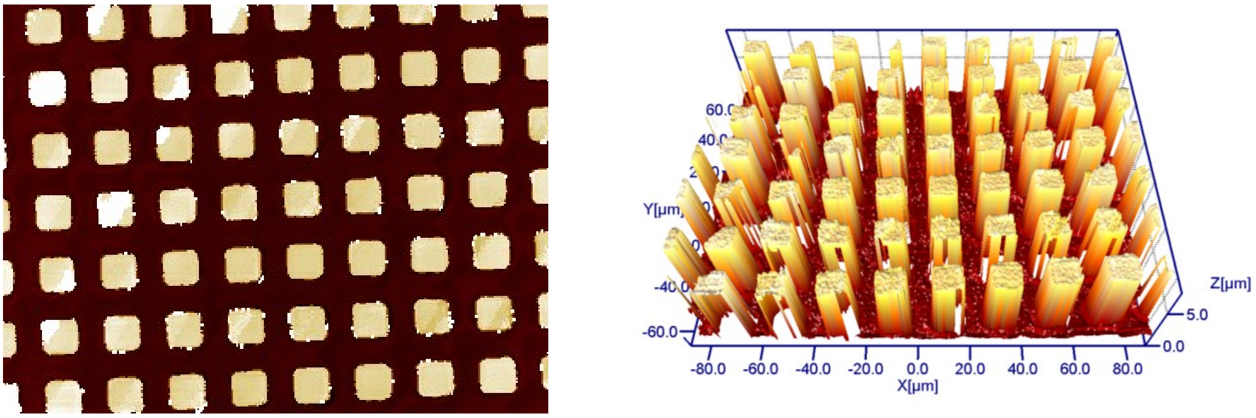

Figure 4 – (a) Top-down interferometer view of a region with 10 µm pillars & 10 µm spacing (b) 3D interferometer projection of a region with 10 µm pillars & 10 µm spacing. Both images correspond to the designs illustrated in Fig. 2.

The wafer (as shown in Fig. 3) was divided into sections, each of which was created with a unique pillar thickness and spacing (detailed in Table 1) to give a wide range of microstructures to examine.

A 4 inch silicon wafer was treated with a photoresist polymer before standard lithography and etching techniques were used to impose the design, through a glass mask, onto the surface. The surface was post-processed with plasma oxidation to remove any residue from the etching process and to ensure that droplets deposited onto its surface are completely wetting (superhydrophilic).

The surface was inspected visually using an optical microscope and then analysed in detail using a white light interferometer to ensure the integrity of the micropillars (see Fig. 4).

3 Experimental Methods

3.1 Goniometer

A digital goniometer was used to measure very accurately the equilibrium contact angles of water droplets on the structured surface. Droplets were deposited using an electronically controlled syringe to produce consistently droplets of 5 µL.

3.2 High-Speed Camera

The surface was heated using a hot plate and the Leidenfrost point was measured for each individual microstructured section. A high-speed Phantom camera was used to record Leidenfrost behaviour in detail, for droplets deposited both on single regions and on the interface between two regions of different structure (Figure 5).

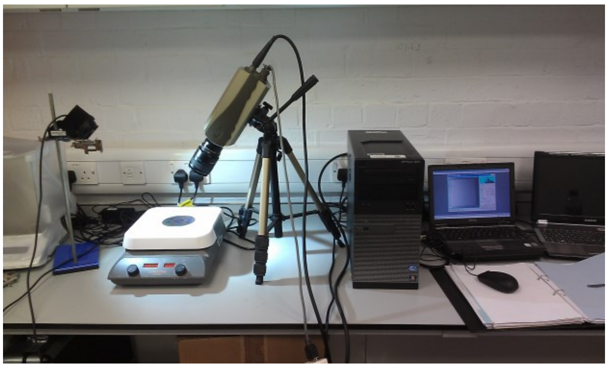

Figure 5 – Photograph of the high-speed Phantom camera apparatus used.

Droplets were carefully placed on the surface (We ~ 1, as high inertia drops can impinge on the pillars and artificially raise the LFP) and the LFP was measured for three separate droplet sizes: 15 µL, 8 µL and 24 µL.

4 Results

Equilibrium contact angles for droplets measured using the goniometer are shown in

Table 2 and were all indeed superhydrophilic and of the range 6.18° – 23.90°.

	16.95°	17.54°	17.65°	20.16°	
	18.15°	17.47°	17.58°	18.22°	
6.18°	19.41°	12.12°	16.80°	17.56°	5.74°
20.09°	17.15°	21.32°	13.88°	21.04°	14.54°
7.82°	18.38°	10.93°	21.71°	23.90°	22.42°
8.11°	16.08°	18.53°	19.28°	20.85°	16.38°
	17.70°	19.42°	20.54°	19.84°	
	8.20°	11.15°	15.25°	6.57°	

Table 2 – Equilibrium contact angles of droplets deposited on the microstructured silicon surface at STP.

For all droplets, the LFP was found to rise almost linearly with the pillar spacing (see Figure 6).

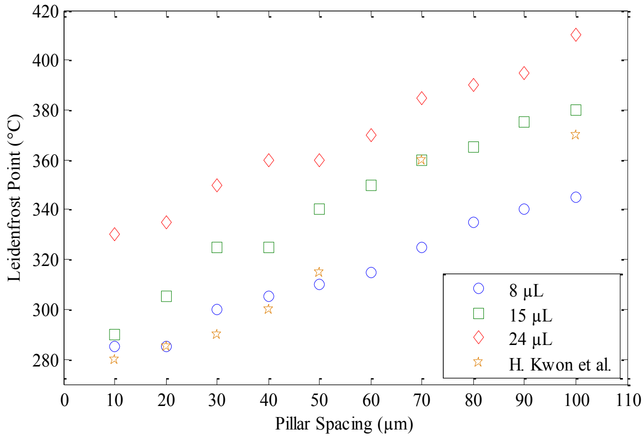

Figure 6 – Measured Leidenfrost points as a function of the pillar spacing.

5 Discussion & Conclusions

Both microstructure and droplet size have a significant influence on the LFP of the silicon surface. The range of the LFP varies from 240°C for the smallest drop size to 410°C. This is a far greater LFP than has previously been recorded in literature for silicon surfaces and an extension of the work by H. Kwon *et al*. Work by Biance *et al*. in 2003, previously explored the effect of drop size on the LFP. [22-23]

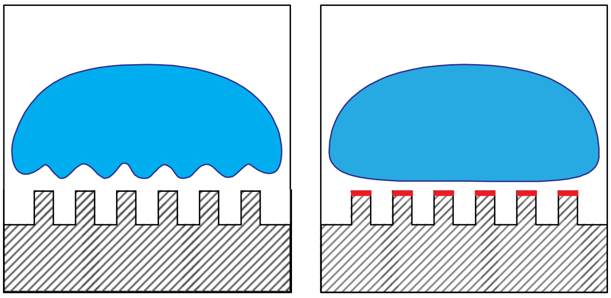

Figure 7 – (a) Underside droplet deformation by the presence of micropillars (b) Effective area for heat transfer is reduced to the tops of the posts.

Biance *et al.* [22] showed that droplet evaporation occurs almost entirely through the underside and C. Kruse *et al.* [17] explored the idea of possible distortions in the droplet underside due to the presence of microstructure, shown schematically in Fig 7(a). This increases the available area for heat transfer. As the Leidenfrost state can be said to occur when the lift force created by the evaporating vapour overcomes the downward forces of surface tension and gravity, an increase in the available area for heat transfer to occur should reduce the thermal flux required to achieve it.

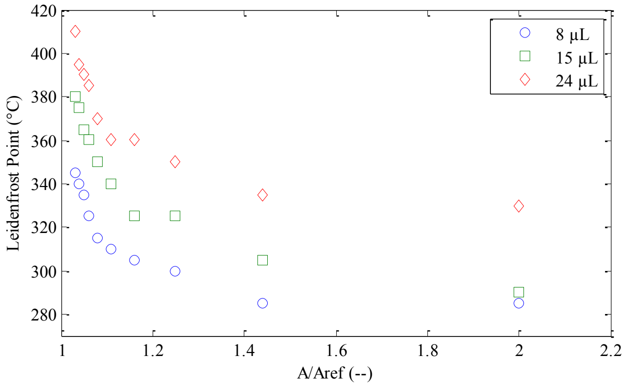

Figure 8 – Measured Leidenfrost points as a function of the Area of the surface divided by the Reference Area of an equivalent smooth surface.

The increased area of the underside of the droplets is too complex to measure, so instead the increase in surface area due to the microposts is used instead. This was nondimensionalised using the base area of a flat surface. Plotting the measured LFP against dimensionless area (Figure 8) shows that in cases where the LFP is highest, the surface area approaches that of a flat surface (LFP

~320°C for the case of the smallest drop), presumably as the distorted area decreases and hence a higher thermal flux is required to achieve the Leidenfrost state.

An additional mechanism is that the micropillars act as conductors, such that the effective area available for heat transfer to occur is equivalent only to the area of the tops of the pillars themselves (Fig. 7(b)) where the vapour film is thinnest, but that conduction distance is reduced.

Plotting, in Fig 13, the LFP measured against this Effective Area for heat transfer (a ratio of the surface area of the tops of the pillars to the area of a flat surface) shows a similar trend to Figure 8. Where there is more surface area available for heat transfer to occur, the LFP is lower.

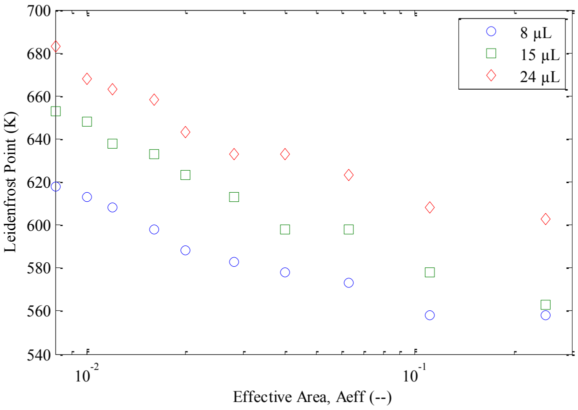

Figure 13 – Measured Leidenfrost points as a function of the Effective Area of the micropost array.

Additionally, it was observed that droplets which initially start in an area where they are in the Leidenfrost boiling regime but are then allowed to move into regions where the LFP is higher, remain in Leidenfrost.

If a droplet is suspended on the interface between two regions of differing microstructure, a strong motive force is observed as it appears to tug violently and rotate erratically. There appears to be some kind of underlying oscillation in the supporting vapour phase, brought about as a result of the varying microstructure, which causes a strong impetus for the droplet to move.

Future Work

As part of an ongoing experimental programme, further recording, by means of the high-speed camera, of the behavior of Leidenfrost drops – in particular, the interactions that occur when a droplet is placed on a boundary between two regions – will be obtained and analysed quantitatively through computational techniques.

As a further point of interest, the lifetime of droplets, below the LFP, will be measured as both a function of surface microstructure and temperature with an aim to provide comprehensive experimental data on the influence of structure, and by extension wettability, on droplet lifetimes.

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