Liquid metal based convective cooling systems

A thesis submitted in fulfilment of the requirements for the degree of
Doctor of Philosophy

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Declaration

I certify that except where due acknowledgement has been made, the work is that of the author alone; the work has not been submitted previously, in whole or in part, to qualify for any other academic award; the content of the thesis/project is the result of work which has been carried out since the official commencement date of the approved research program; any editorial work, paid or unpaid, carried out by a third party is acknowledged; and, ethics procedures and guidelines have been followed. I acknowledge the support I have received for my research through the provision of an Australian Government Research Training Program Scholarship.

Jiuyang Zhu

26th of April 2018
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Abstract

Forced convection is one of the major mechanisms used for cooling of electronic systems. However, the problems associated with the size, fabrication, integration and maintenance of conventional convective cooling systems has limited utility of such technologies for thermal management of miniaturised electronic components. The existence of fluidic interfaces between the pump and the hot spot not only increases the footprint of the cooling system but also may cause additional problems such as the leakage of the coolant medium as well as reducing the response time of the pump. A new class of convective cooling systems, which can address these limitations can facilitate the generation of highly integrated electronic systems.

Gallium based liquid metal alloys have been recently used for making soft electronic and microfluidic devices. These alloys, which inherit the properties of both liquid and metal, have been utilised for making a new class of soft microfluidic elements such as pumps, mixers, valves, heaters and electrodes. These advances have motivated me to develop a novel class of liquid metal based convective cooling systems, which can be used for temperature regulation of both flow-through and flow-free systems in both static and dynamic modes.

As my first contribution, I demonstrated the utility of liquid metal pumps for the localised cooling of hot spots. These pumps, which consist of millimetre size liquid metal droplets, can be easily installed at desired locations of the system. Applying a low voltage square wave signal creates sufficient surface tension gradient across the droplet, providing a continuous flow of coolant
medium through the cooling channel. The flow rate can be readily modulated by varying the frequency of the signal. Furthermore, the high thermal conductivity of liquid metal droplet allows it to serve as a heat sink, enhancing the dissipation of heat into the cooling channel. This hybrid pump-heat sink can reduce the temperature of localised hot spots, and has been characterised in various operating conditions.

As my second contribution, I demonstrated the unique features of liquid metal pumps for the transient cooling of hot spots. The elimination of mechanical moving elements and interconnecting tubes significantly reduces the response time of these pumps, enabling them to reduce the temperature of hot spots in a few seconds.

As my third contribution, I investigated the capability of liquid metal pumps for producing customised temperature profiles inside an isolated flow-free liquid chamber. A pair of liquid metal pumps is located along the opposite sides of the chamber to induce vortices inside the liquid chamber. The configuration and rotational velocity of the vortices can be easily modulated by varying the frequency of the signal. Customised temperature gradients can be produced inside the liquid chamber, breaking the inherent limitations of diffusive heat transfer in isolated chambers.

As my fourth contribution, I studied the ability of liquid metal pumps for creating controllable spatiotemporal temperature gradients inside an isolated liquid chamber. The rapid reconfiguration of vortices and the dominance of convective heat transfer enable the rapid change of temperature profile inside the chamber.
The versatility of liquid metal based convective cooling systems facilitates their incorporation into miniaturised yet highly integrated electronic devices. Accordingly, the simplicity and flexibility of such cooling systems enable their integration into the emerging soft and stretchable devices.
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<th>Description</th>
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<tbody>
<tr>
<td>PDMS</td>
<td>Polydimethylsiloxane</td>
</tr>
<tr>
<td>PMMA</td>
<td>Poly-methyl methacrylate</td>
</tr>
<tr>
<td>DC</td>
<td>Direct current</td>
</tr>
<tr>
<td>NaK</td>
<td>Sodium-potassium</td>
</tr>
<tr>
<td>CNT</td>
<td>Carbon nano tube</td>
</tr>
<tr>
<td>CrO$_2$</td>
<td>Chromium dioxide</td>
</tr>
<tr>
<td>CuO</td>
<td>Copper oxide</td>
</tr>
<tr>
<td>Al$_2$O$_3$</td>
<td>Aluminum Oxide</td>
</tr>
<tr>
<td>SEM</td>
<td>Scanning electron microscope</td>
</tr>
<tr>
<td>NaOH</td>
<td>Sodium hydroxide</td>
</tr>
<tr>
<td>SDS</td>
<td>Sodium dodecyl sulfate</td>
</tr>
<tr>
<td>EDL</td>
<td>Electrical double layer</td>
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\[ \Delta T = \left| \langle T_{\text{one vortex}} \rangle_{t=0\,s} - \langle T_{\text{zero vortex}} \rangle_{t=15\,s} \right|, \]  

(e)

Temperature oscillations of the regions located at the hot spot upstream, hot spot downstream, vortex centre, sidewalls and opposite the hot spot. Green and pink regions correspond to the first and second parts of the one vortex mode whilst the yellow region corresponds to the zero vortex mode. Dashed lines display the phase shift of various regions compared against the hot spot.

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\[ \Delta T = \left| \langle T_{\text{one vortex}} \rangle_{t=0\,s} - \langle T_{\text{zero vortex}} \rangle_{t=15\,s} \right|, \]  

(e)

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Research contributions

First author contributions:


Co-author publications:


CHAPTER 1

Introduction

1.1 An overview of cooling mechanisms for electronic systems

The ongoing advancements in electronics, microfabrication, and packaging technologies along with the tendency for developing highly integrated, embedded and portable electronic systems such as smartphones has led to enhancing the capacity of these systems by adding more components into them while constantly shrinking their size. The combination of two factors has resulted in the continuing increase of the heat dissipation per volume, and consequently the surface heat flux of electronic systems. For example, the surface heat flux of computer chips has increased almost exponentially from 1 W/cm\(^2\) in 1980 to 10 W/cm\(^2\) in 2000, as presented in Figure 1.1 [1]. Similar trends can be found for almost every other electronic devices in the market.

![Figure 1.1: The evolution of surface heat flux in computer chips over 60 years [1]. Reproduced with permission from IEEE.](image-url)
Surface heat flux and the maximum allowable temperature are among the two important specifications of electronic systems, which are determined by the manufacturer [2]. The surface heat flux depends on the size of the electronic system while the maximum allowable temperature of conventional electronic systems is generally 85 °C [1]. The heat released from the external surface of electronic systems is dissipated to the surrounding environment via free convection, forced convection or radiation mechanisms. The type of cooling mechanism is generally selected according to the surface heat flux, and the temperature difference between the hot surface and the surrounding environment, as presented in Figure 1.2 [2, 3]. This figure is based on the thermal properties of air, water and dielectric liquids such as fluorochemicals, with the latter used for immersion cooling [4].

![Figure 1.2: Existing cooling mechanisms for electronic devices categorised according to surface heat flux and the temperature difference between the surface and the surrounding environment [2]. Reproduced with permission from McGraw Hill Education.](image-url)
According to Figure 1.2, air natural convection is only practical for surface heat fluxes of less than 0.05 W/cm² beyond which the temperature difference exceeds the 65 °C level (assuming that the ambient temperature is 20 °C, the maximum allowable temperature of the system will be 85 °C). Air forced convection can raise the maximum surface heat flux to 0.3 W/cm², which implies the addition of a fan and a large metallic heat sink to increase the cooling surface area of the electronic component. Alternatively, the immersion of the electronic device in a bath filled with an inert and dielectric fluorochemical liquid can push the maximum surface heat flux to 2 W/cm².

Water forced convection, astonishingly, increases the maximum heat flux level to 20 W/cm². This can be further increased by incorporating coolant media with higher thermal conductivities such as nanofluids [5] or liquid metals [6]. Forced convection is generally achieved by recirculation of the coolant medium in a closed loop, and the consequent cooling of the coolant medium in a radiator. The heat transfer coefficient, and consequently the amount of heat flux removal can be increased by applying a water jet onto the hot spot. This method, which is known as jet impinging, leads to the formation of a thin layer of the coolant over the hot surface [7, 8]. Further increase of heat transfer coefficient can be achieved by phase change (evaporation) of the coolant medium within the immersion tank (also known as pool boiling) [9] or more sophisticated spray cooling systems [10, 11], as comprehensively reviewed in [12].
1.2 Forced convective cooling systems

The scope of this research is limited to single-phase forced convection of localised hot spots by recirculation of water based solutions through closed loop channels or inducing rotational flows inside isolated liquid chambers, which are referred to as ‘flow-through’ and ‘flow-free’ cooling systems, respectively, throughout this thesis.

A ‘flow-through’ cooling system is composed of six major components, including (i) a heat source, (ii) a heat sink to enlarge the cooling surface area, (iii) a closed-loop conduit to recirculate the coolant medium, (iv) a pump to drive the cooling medium, (v) a heat exchanger for absorbing the heat from the coolant medium, and (vi) a fan to enhance the convective heat transfer along the heat exchanger, as schematically presented in Figure 1.3.

![Figure 1.3: Schematics of a flow-through convective cooling system](image-url)
Alternatively, a ‘flow-free’ cooling system is composed of five major components, including (i) a heat source, (ii) a heat sink to enlarge the cooling surface area, (iii) an isolated liquid chamber containing the coolant medium, (iv) a vortex generator for inducing rotational flow inside the chamber, and (v) a fan to enhance the convective heat transfer along the free surface of the liquid chamber, as schematically presented in Figure 1.4.

**Figure 1.4:** Schematics of a flow-free convective cooling system

This thesis aims to develop novel ‘pumping’ and ‘heat sinking’ mechanisms for miniaturised ‘flow-through’ cooling systems, and ‘vortex generation’ mechanisms for miniaturised ‘flow-free’ cooling systems, as discussed in the following sections.
1.3 Pumping mechanisms

Current microfluidic flow control are mostly depends on mechanical pumps, which driving force is generated by moving parts, including peristaltic pump, gear pump, plunger pump, pneumatic pump and diaphragm pump, as described in Table 1.1. Because of its multiple advantages such as adjustable flow control, fast response time, and relatively small size, which have been widely used in many applications. Despite these advantages, the moving parts of mechanical pump also cause energy loses due to heat generated by friction and their rather complicated fabrication process. Furthermore, these moving parts enable precise control of fluids but also increase the complexity, which limits the usage of microfluidic devices in less equipped environments. To simplify the liquid driving mechanism and fully utilize the advantage of microfluidics, non-mechanical pumps with no moving parts can be used instead.

Non-mechanical pumps with no moving part function with electro-magnetic, electro-osmosis, electro-wetting, electro-chemical and thermosyphon generation. These non-mechanical pumps provide precise control over flow rate without the aid of an external power source and are capable of turning on and off in response to specific analysis in solution. However, these kinds of pump produce relatively low flow rate, and in order to operate they often need very high voltages to generate driving force. Alternatively, non-mechanical pumps only applicable for some specific liquids, such as electromagnetic pump can only drive conductive liquid. Selected pumping mechanisms are briefly discussed in Figures 1.5 to 1.8.
Table 1.1. A summary of pumping mechanisms for miniaturised fluidic systems

<table>
<thead>
<tr>
<th>Pump type</th>
<th>Principle</th>
<th>Reference</th>
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<tbody>
<tr>
<td>Plunger (syringe)</td>
<td>The displacement of a plunger drives the liquid</td>
<td>[13-16]</td>
</tr>
<tr>
<td></td>
<td>A similar principle is used but the plunger is displaced manually</td>
<td>[17]</td>
</tr>
<tr>
<td>Gear</td>
<td>Rotation of gears drives the surrounding liquid</td>
<td>[18, 19]</td>
</tr>
<tr>
<td>Peristaltic</td>
<td>Compression and relaxation of a tube by means of rotating gears drives the liquid in a closed loop</td>
<td>[16, 20, 21]</td>
</tr>
<tr>
<td>Pneumatic</td>
<td>Positive/negative gas pressure is used to push/pull the liquid</td>
<td>[22, 23]</td>
</tr>
<tr>
<td>Diaphragm</td>
<td>The harmonic expansion and compression of piezoelectric diaphragms is utilised for the pumping of liquid</td>
<td>[24-27]</td>
</tr>
<tr>
<td>Centrifugal</td>
<td>The rotational motion of a disc drives the liquid due to centrifugal force</td>
<td>[28, 29]</td>
</tr>
<tr>
<td>Electromagnetic</td>
<td>The combination of electric and magnetic fields drives a conductive liquid under Lorentz force.</td>
<td>[6, 30]</td>
</tr>
<tr>
<td>Method</td>
<td>Description</td>
<td>References</td>
</tr>
<tr>
<td>-----------------</td>
<td>-----------------------------------------------------------------------------</td>
<td>------------</td>
</tr>
<tr>
<td>Electroosmosis</td>
<td>The motion of liquid close to the sidewalls under an external electric field drags the liquid through the entire channel</td>
<td>[31, 32]</td>
</tr>
<tr>
<td>Electrochemical</td>
<td>Gas bubbles are generated and expanded by means of electrolysis to pump the liquid</td>
<td>[33, 34]</td>
</tr>
<tr>
<td>Capillary</td>
<td>The adhesion of liquid molecules to the surface of the channel drives the liquid</td>
<td>[35-37]</td>
</tr>
<tr>
<td>Thermosyphon</td>
<td>The liquid is driven under buoyancy driven forces caused by temperature gradient</td>
<td>[38, 39]</td>
</tr>
<tr>
<td>Electrowetting</td>
<td>The surface tension gradient caused by an external voltage is used for driving droplets</td>
<td>[40, 41]</td>
</tr>
<tr>
<td>Thermocapillary</td>
<td>The surface tension gradient caused by temperature gradient is used for driving droplets</td>
<td>[42, 43]</td>
</tr>
</tbody>
</table>
**Peristaltic pump:** Convective cooling is conventionally used in electronic devices for cooling of hot spots. Typically, the coolant is driven through a closed loop channel by means of a pump, and a heat sink is later implemented in the loop to release the heat absorbed by the coolant. An elegant example is the hybrid liquid metal-water cooling system demonstrated by Deng et al., in which liquid metal is recirculated inside the loop by means of a peristaltic pump (Figure 1.5) [44]. The heated liquid metal then enters a radiator which is cooled by water to release the heat. The flow rate of the coolant can be easily adjusted by varying the rotational speed of the peristaltic pump. Moreover, the pump is not sensitive to the coolant medium. Despite these advantages, the utilisation of liquid interfaces such at recirculating tubes increases the footprint of the cooling system, and importantly can cause leakage. This also reduces the response time of the pump, which limits the capability of the system to regulate the temperature in the event of a heat surge. Additionally, the reliance of rotating elements (gear in the case of peristaltic pump) is prone to fatigue, which requires routine maintenance.

![Figure 1.5: A hybrid liquid metal-water cooling system by using peristaltic pump for recirculation of liquid metal along the cooling channels and water for cooling of the liquid metal inside the heat exchanger [44]. Reproduced with permission from Springer Nature.](image)
**Diaphragm pump:** The limitations associated with conventional pumps have inspired researchers to use miniaturised pumps. These pumps are small, and therefore can be installed close to the hot spot region, reducing the length of tubes and other fluidic interfaces. The small footprint of miniaturised pumps also facilitates the incorporation of multiple pumps within the system. Piezoelectric diaphragm micropumps are very common for this purpose. These pumps are commercially available, have a high stroke volume and a short response time [45]. Piezoelectric diaphragm pumps can be easily integrated into microfluidic systems, as an example demonstrated by Akagi et al. for perfusion of immobilised zebrafish embryos with various chemicals (**Figure 1.6**) [24]. Despite these advantages, these pumps are rather expensive (> $200) and require a relatively large actuation voltage.

**Figure 1.6:** A microfluid system equipped with a piezoelectric diaphragm pump for perfusion of immobilised zebrafish embryos [24]. Reproduced with permission from Elsevier.
**Electromagnetic pump:** The moving elements in conventional pumps cause friction, power loss, and structural damage in such pumps. Several solutions have been proposed to eliminate the moving elements of conventional pumps. This includes the application of electromagnetic pumps, which involves the recirculation of conductive liquids in the presence of magnetic and electric fields due to Lorenz force [46]. This concept has been traditionally used for driving sodium-potassium (NaK) liquid metal through nuclear reactors [47], and recently has been implemented for cooling of smaller devices. One example is the electromagnetic pump reported by Deng et al. [6], which recirculates gallium based liquid metal alloy through a closed loop for cooling an electronic device (Figure 1.7). These alloys have a low viscosity and a high thermal conductivity making it suitable for convective cooling of hot spots [48]. Despite these advantages, gallium based liquid metal alloys are corrosive and can easily damage metallic tubes due to embrittlement [49]. Also these alloys are rather expensive (1 ml is about $8) compared to other coolants such as water or oil.

![Diagram](image.png)

**Figure 1.7:** An electromagnetic pump for recirculation of liquid metal inside the channels: (a) schematics, and (b) cooling system with fins [6]. Reproduced with permission from IEEE.
**Thermosyphon pump**: Liquids can also be recirculated passively through the fluidic system. Capillary driven flow is the most common method for passive pumping [50]. Despite simplicity, capillary flow is limited to a short period before the entire channel is fully wet, and therefore is more suitable for disposable point-of-care devices. In addition, capillary flow is sensitive to ambient conditions such as relative humidity. Thermosyphon effect, the flow of liquid under buoyancy gradient (natural convection) is an alternative method of passive pumping, which is widely used in solar heaters [51]. The existence of hot spots facilitates the recirculation of flow by means of thermosyphon effect, as for example demonstrated by Peipei et al. [38]. In this work, the heat produced by a heating block is utilised for recirculation of the coolant medium through the cooling channel with the thermal performance of the system characterised using water and a gallium based liquid metal alloy (Figure 1.8). Despite simplicity, the system has a limited flow rate, and a long response time due to capacitance of the medium.
Figure 1.8: Driving the coolant through the system under the thermosyphon effect: (a) Schematics, (b) Experimental set-up, (c) Temperature contours using liquid metal as the coolant, and (d) Temperature contours using water as the coolant [38]. Reproduced with permission from ASME.
1.4 Heat sinking mechanisms

Miniaturisation of electronic devices has led to substantial increase of their power density, making the thermal management of such systems very challenging. At the same time, cooling of miniaturised devices using conventional air cooling systems (comprised of a fan and a metallic heat sink) is impractical due to their limited space. These limitations have inspired researchers to seek alternative cooling systems, as summarised in Table 1.2. This includes the integration of cooling units such at thermoelectric devices, also known as peltiers, at the hot spots [52], implementation of microfluidic channels to facilitate the recirculation of coolant medium in a limited space [53], utilisation of microfluidic channels with locally patterned heat sinks to enhance the convective cooling at desired locations [54], and recently droplet based cooling systems which take advantage of discrete volumes of the cooland medium rather than a continuous volume [55].

The compactness, price, ease of fabrication, integration, operation and maintenance as well as the ability to adopt with more complicated (for example 3D electronic circuits) are among the parameters, which determine the type of heat sink for a specific electronic system. Importantly, the ability to utilise multiple heat sinks, and deal with transient heat surges are among the parameters, which are becoming increasingly important in future miniaturised electronic systems, as briefly discussed in Figures 1.9 to 1.12.
Table 1.2. A summary of heat sink types in microfluidics

<table>
<thead>
<tr>
<th>Type</th>
<th>Principle</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermoelectric cooling</td>
<td>Integration of thermoelectric cooler into the microfluidic device to facilitate the cooling of hot spots</td>
<td>[52, 56, 57]</td>
</tr>
<tr>
<td>Microfluidic channels</td>
<td>Arrays of high aspect ratio microfluidic channels are integrated onto the hot spot to facilitate the convective cooling</td>
<td>[53, 58-62]</td>
</tr>
<tr>
<td>Microfluidic channels with assembled micro/nano particles</td>
<td>Thermally conductive micro/nano particles can be assembled along the walls of the channel to enhance the convective cooling</td>
<td>[54, 63, 64]</td>
</tr>
<tr>
<td>Droplet based</td>
<td>Discrete droplets driven by electrowetting facilitate the delivery of heat from the local hot spots</td>
<td>[55, 65, 66]</td>
</tr>
</tbody>
</table>
**Microfluidic channels:** The thermal resistance of a heat sink, which defines the ratio of temperature difference across the heat sink to the amount of dissipated heat ($\theta = \Delta T/\dot{Q}$), is inversely proportional to the thermal conductivity of the coolant medium surrounding the heat sink [67]. Given that water is about 25 times more thermally conductive than air, recirculation of water based coolant through microfabricated heat sinks can significantly reduce the thermal resistance of compact heat sinks. More importantly, the thermal resistance of a compact heat sink is inversely proportional to the width of the fluidic channels, as demonstrated by Tuckerman [53], which makes the high aspect ratio channels desirable for microfluidic heat sinks. The amount of dissipated heat can be easily controlled by changing the flow rate of the medium recirculating through the channels. Microfluidic based heat sinks can be integrated into compact electronic components in customised configurations, as presented by Sarvey et al. (Figure 1.9) [59].

![Figure 1.9: Microfluidic heat sink for localised cooling of hot spots: (a) Schematic, and (b) Assembled microfluidic heat sink integrated into the electronic system [59]. Reproduced with permission from IEEE.](image)
The versatility of microfluidic based heat sinks makes them quite compatible with 3D integrated circuit technologies. An elegant example is an electronic circuit with high aspect ratio ‘through silicon vias’ embedded within a microfluidic based heat sink, as recently presented by Oh et al. [61]. In this work, arrays of copper rods are embedded within silicon pillars located across the microfluidic channel to interconnect different layers of the 3D circuit (Figure 1.10).

Figure 1.10: A microfluidic heat sink composed of silicon micropillars embedded with copper for enhancing the thermal conductivity: (a) Schematic of the microfluidic heat sink, (b) SEM image of micropillars, (c) Cross-sectional view of micropillars [61]. Reproduced with permission from Elsevier.
Microfluidic channels with assembled micro/nano particles: The limitations associated with the fabrication and integration of heat sinks have inspired researchers to develop soft heat sinks, which can be patterned and removed on-demand. An elegant example is the dynamic nanofin heat sinks by Yi et al., in which magnetic CrO$_2$ nanoparticles are assembled along the sidewalls of a microfluidic channel using a permanent magnet (Figure 1.11) [54, 63]. The location, configuration and the size of this soft heat sink can be readily controlled by varying the flow rate of the medium, the concentration of nanoparticles as well as the location and intensity of the magnetic field. More importantly, the patterned fins can be dismantelled by simply removing of the magnetic field. Despite these advantages, the high concentration of nanoparticles leads to the formation of densely packed structures along the sidewalls, which might cause unwanted pressure drop and secondary flows along the channel. Also, it is not possible to control the gap between the assembled nanorods, which means that the assembeled nanofin structure behaves as a continous block of nanoparticles rather than an array of isolated nanorods, significantly reducing the overall surface area of the assembeled nanofin.
**Figure 1.11:** Nanofin heat sinks created by magnetic patterning CrO$_2$ nanoparticle along the sidewalls of a microfluidic channel close to the hot spot: (a) Schematics of the system, (b) Bottom-view of the channel in the absence of nanoparticles, and (c-e) The formation of nanofin along the sidewalls 10 mins after the application of magnetic field and in the presences of nanoparticles at various flow rates of 10, 40 and 120 μl/min [54, 63]. Reproduced with permission from Royal Society of Chemistry.
Droplet based: An alternative solution for convective cooling of hot spots is the use of discrete volumes of coolant media in the form of droplets. Different mechanisms have been proposed for transportation of droplets. Among them is electrowetting-on-dielectric, which has been extensively used for manipulation of droplets (transportation, merging, splitting, dispensing) in microfluidic systems [65, 66]. An elegant example is the droplet-based cooling system demonstrated by Sung-Yong et al. [55], in which droplets are continuously transported along the electrodes to absorb heat from the hot spot of the system (Figure 1.12). The temperature rise of droplets depends on their axial velocity, which can be controlled by varying the applied voltage. This enables the rapid and customised cooling of multiple hot spots using a very low volume of coolant medium. Despite these advantages, the instability and evaporation of droplets, and importantly the need for a reservoir for the continuous injection of coolant medium are among the limitations of this technique.

Figure 1.12: A digital microfluidic system used for cooling of hot spots: (a) Coolant droplet is transported between two parallel plates using electrowetting on dielectric mechanism, (b-c) The droplet is transported to the target hot spot to absorb heat, (d) The heated droplet is replaced by a cool droplet and this process is repeated [55]. Reproduced with permission from Royal Society of Chemistry.
1.5 Vortex generation mechanisms

The laminar characteristics of flow inside the microfluidic structures (and in general miniaturised systems), implies diffusion as the dominant mode of momentum, heat and species exchange within such structures [68]. Diffusion is a slow process, and often leads to formation of linear gradients of velocity, temperature, and concentration of species inside the microfluidic systems, depending on diffusive properties of the liquid, including its viscosity, thermal conductivity, and mass diffusivity. Several strategies have been proposed to augment and accelerate the diffusion process in microfluidic systems. This includes prompting local convection flows, also known as advection or chaotic flows. Under certain conditions governed by the size and geometry of the structure as well as the viscosity and velocity of the liquid, the chaotic flow can form one or multiple vortices. Vortices have a predictable flow pattern with their rotational flow velocity increasing linearly from the centre of the vortex toward its outer edge. Vortices have been extensively used in microfluidic systems for enhancing the mixing of neighbouring flows [69], isolation of suspended particles based on their dimensions [70], trapping of particles using secondary mechanisms such as dielectrophoresis [71] as well as stimulation of mechano-sensitive endothelial cells [72]. This is particularly important in isolated liquid chambers, where the net flow is zero.

Microfluidic vortex generators can be classified into passive and active groups, as summarised in Table 1.3. Passive vortex generators rely on the flow of the medium inside the microfluidic structures to induce secondary flows in the form of vortices. Such secondary flows are generally induced by modifying the geometry of the channel to enhance the likelihood of producing transverse
flows. Common structures include channels with asymmetric ridges along their surfaces [69], curved channels to induce Dean vortices [73], sudden expansion of channel cross section [70]. Passive vortices can also be produced inside the microscale droplets when moving inside a carrier fluid [74]. In contrary, active vortex generators rely on external stimuli such as mechanical [75], acoustic [76], electrokinetic [77], thermal [78], electrothermal [71], and electrowetting [79] to induce vortices. Active vortex generators are generally more complex than passive ones but provide more control over the direction, rotational speed, and timing of vortices, as briefly discussed in Figures 1.13 to 1.16.
Table 1.3. A summary of vortex generation mechanisms in microfluidics

<table>
<thead>
<tr>
<th>Mechanism</th>
<th>Principle</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hydrodynamic</td>
<td>Pattening assymmetric ridges along the surface of the channel produces transverces flows</td>
<td>[69, 80]</td>
</tr>
<tr>
<td></td>
<td>Application of flow through curved channels at high Dean numbers induces Dean vortices</td>
<td>[73, 81, 82]</td>
</tr>
<tr>
<td></td>
<td>Sudden expansion of the channel cross section induces passive vortices</td>
<td>[70, 83-85]</td>
</tr>
<tr>
<td></td>
<td>The recirculation of liquid inside the moving microscale droplets causes vortices</td>
<td>[74]</td>
</tr>
<tr>
<td>Mechanical</td>
<td>Rotation of a rotating elements creates vortices inside the channel</td>
<td>[75, 86]</td>
</tr>
<tr>
<td>Acoustic</td>
<td>Oscilating sharp edges (acoustic streaming)</td>
<td>[87, 88]</td>
</tr>
<tr>
<td></td>
<td>Oscillating trapped bubbles</td>
<td>[76]</td>
</tr>
<tr>
<td>Electrokinetic</td>
<td>Electroosmotic driven flow along the sidewalls is utilised for producing vortices</td>
<td>[77, 89, 90]</td>
</tr>
<tr>
<td>Method</td>
<td>Description</td>
<td>References</td>
</tr>
<tr>
<td>---------------</td>
<td>-----------------------------------------------------------------------------------------------</td>
<td>------------</td>
</tr>
<tr>
<td>Thermal</td>
<td>The creation of local hot spots produces buoyancy driven forces that induce vortices inside the liquid chamber</td>
<td>[78, 91]</td>
</tr>
<tr>
<td>Electrothermal</td>
<td>The creation of non-uniform electric fields creates electrothermal forces that induce vortices inside the liquid chamber</td>
<td>[71, 92]</td>
</tr>
<tr>
<td>Electrowetting</td>
<td>The recirculation of liquid inside the electrowetting driven droplets produces vortices</td>
<td>[79, 93]</td>
</tr>
</tbody>
</table>
**Hydrodynamic vortices:** Vortices can be generated passively by inducing secondary flows through the microfluidic channels. For example, patterning an array of asymmetric ridges along the bottom surface of the channel can produce transverse flows, which consequently leads to the formation of a pair of asymmetric vortices along the channel even at low Reynolds numbers. These structures, which are also known as herringbone structures, have been commonly used as passive mixers in microfluidic devices ([Figure 1.13](#)) [69].

The configuration of vortices can be controlled according to the pattern of the ridges as well as the flow rate of the medium through the channel. Despite these advantages, the patterning of the ridges requires 3D fabrication procedures. Importantly, a flow of medium should be provided through the channel to induce the vortices, which limits the utility of this method in enclosed liquid chambers where the net flow rate of the liquid is zero.
Figure 1.13: Vortices induced by a staggered herringbone mixer: (a) Schematic of the mixer with assymmetic ridges fabricated along the bottom surface of the channel, and (b) Confocal micrographs of vertical cross sections of the mixing channel showing the induced vortices at different locations of the ridges [69]. Reproduced with permission from The American Association for the Advancement of Science.
Vortices can also be induced by means of curved microfluidic channels. The centrifugal force exerted on the flow leads to radial pressure gradients, dragging the flow from the centre of the channel toward the outer sidewalls. These centrifugal instabilities, which are known as Dean instabilities, can lead to formation of Dean vortices inside the curved channel. The intensity of Dean instabilities can be characterised by Dean number, which is calculated by multiplying the Reynolds number of flow by the square root of channel curvature ratio ($De = Re \sqrt{W/R}$, in which $W$ is the channel width and $R$ the radius of the curvature). Dean instabilities become dominant at Dean numbers of higher than 10. Sudarsan et al. used this principle to develop a passive microfluidic mixer using a curved channel (Figure 1.14) [73]. Dean vortices have also been extensively used for focusing and isolation of microparticles and mammalian cells [81, 94]. Despite simplicity, the formation of Dean vortices relies on high flow rates inside the curved channels, limiting its application in confined chambers with zero net flow.
Figure 1.14: Dean vortices induced inside a curved channel: (a-b) Schematics of flow pattern inside the mixing channel at low and high Dean numbers, and (c) Experimental flow patterns induced at various Dean numbers [73]. Reproduced with permission from National Academy of Sciences.
**Electrokinetic vortices:** Vortices can also be generated actively. The most straightforward way for producing vortices is to implement a microstirrer inside the microfluidic chamber or channel. However, the limitations associated with the integration and maintenance of mechanical actuators have inspired researchers to develop vortex generators, which do not rely on conventional moving elements. One strategy is to induce electroosmotic flow along the sidewalls of the channel, as presented by Suk et al. [77]. In this work, two diodes are embedded within the opposite sidewalls of a microfluidic channel (Figure 1.15). The activation of diodes with a DC signal leads to inducing local electroosmotic flows along the sidewalls. This leads to formation of a vortex inside the channel, which has been used for the rapid mixing of neighbouring flows. The intensity and configuration of the vortices can be easily controlled by varying the magnitude and polarity of the DC signal. Despite simplicity, the structure of the vortices is dictated by the arrangement of the diodes, and cannot be changed once it has been fabricated.
Figure 1.15: Electroosmotic mixer: (a) Schematics of the mixer, composed of two diodes along the two sidewalls of the channel, and (b) Snapshot images of neighbouring flows following the activation of diodes [77]. Reproduced with permission from Royal Society of Chemistry.
**Thermal vortices:** Active vortices can also be generated by applying a temperature gradient inside the liquid. The local heating of liquid induces free convective flows, also known as buoyancy driven flow, which leads to formation of vortices [67]. Such local hot spots can be produced by implementing microelectrodes at the desired locations of the chamber. However, the limitations associated with the fabrication and electrolysis of microelectrodes have inspired researchers to seek alternative solutions. An elegant example of this application is the formation of vortices by applying a laser beam onto the surface of liquid [78]. The adoption of a 1.55 μm laser beam facilitates the absorption of laser energy by the water. The local temperature rise at the surface of the liquid leads to formation of vortices, which leads to focusing of 713 nm polystyrene microparticles (**Figure 1.16**). The rotational velocity and direction of the vortices can be controlled by the location and input power of the laser beam. Despite simplicity and versatility, the vortices are limited to the region very close to the top surface of the liquid.

**Figure 1.16:** Thermally induced vortices produced by applying laser beam into a water chamber at various input powers of: (a) 10 mW, and (b) 60 mW [78]. Reproduced with permission from American Institute of Physics.
1.6 Liquid metal based microfluidic systems

Liquid metals refer to a category of materials, which remain liquid at room temperature and have metallic properties, including high thermal and electrical properties. Mercury is the most common liquid metal, which has been traditionally used for various sensing and actuating applications [95]. Despite these remarkable properties, the toxicity and high vapour pressure of mercury has led to forbidding its application over the last decade.

Gallium based liquid metal alloys, such as eutectic GaIn (75% gallium and 25% indium) and Galinstan (68.5% gallium, 21.5% indium, and 10% tin) have been introduced to fill the mercury’s gap. These alloys have similar properties to mercury but due to their non-toxicity and very low vapour pressure can be safely used. The main difference between these alloys and mercury is the development of a thin gallium oxide layer over their free surface when exposed to oxygen, providing several opportunities and limitations [96].

Gallium based liquid metal alloys can be patterned in customised structures using various lithography, microfluidic enabled (injection into channels), 3D printing and substractive techniques [97]. These patterned structures can be actuated using various chemical, magnetic, optical and electrical stimuli [98]. These alloys have enabled the development of reconfigurable and stretchable electronic devices [99], soft robotic and biotechnological applications [100] as well as microfluidic systems [101]. With respect to microfluidics, gallium based liquid metal alloys have been utilised for the fabrication of channels, pumps, mixers, valves, electrodes, heaters, reconfigurable structures and microscale droplets, as summarised in Table 1.4.
<table>
<thead>
<tr>
<th>Component</th>
<th>Principle</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Channel</td>
<td>3D printed liquid metal is employed as sacrificial ink for fabricating semi-circular polymeric channels</td>
<td>[102]</td>
</tr>
<tr>
<td>Pump</td>
<td>Liquid is driven by inducing surface tension gradient (using electrowetting) across a liquid metal droplet</td>
<td>[103]</td>
</tr>
<tr>
<td>Pump</td>
<td>Liquid is driven by inducing electroosmotic flow using liquid metal electrodes patterned along the sidewalls of the channel</td>
<td>[104]</td>
</tr>
<tr>
<td>Mixer</td>
<td>Liquid is oscillated transversely by inducing an oscillating surface tension gradient (using electrowetting) across a liquid metal droplet</td>
<td>[105]</td>
</tr>
<tr>
<td>Mixer</td>
<td>Laminar streams are mixed due to application of electrohydrodynamic instabilities using liquid metal electrodes patterned along the sidewalls</td>
<td>[106]</td>
</tr>
<tr>
<td>Valve</td>
<td>A liquid metal droplet is moved electrostatically over a thin membrane to open/close the secondary channel</td>
<td>[107]</td>
</tr>
</tbody>
</table>
A liquid metal droplet is moved by inducing surface tension gradient (using ionic imbalance) to block the main channel

<table>
<thead>
<tr>
<th>Electrode</th>
<th>Liquid metal is employed as electrode due to its high electrical conductivity</th>
<th>[104, 106, 109]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Heater</td>
<td>Liquid metal is employed as heater due to its high thermal conductivity</td>
<td>[110, 111]</td>
</tr>
<tr>
<td>Reconfigurable structures</td>
<td>Liquid metal columns are reconfigured along the channel to enable reconfigurable structures</td>
<td>[109, 112]</td>
</tr>
<tr>
<td>Microscale droplets</td>
<td>Liquid metal microscale droplets are continuously generated using flow focusing microfluidic systems</td>
<td>[113-117]</td>
</tr>
<tr>
<td></td>
<td>Liquid metal microscale droplets are generated on-chip using acoustic waves</td>
<td>[118]</td>
</tr>
</tbody>
</table>
1.7 Objectives and Motivation

The primary objective of this research is to develop liquid based forced convective cooling systems, made of gallium-based liquid metal alloys, for the cooling of hot spots through a flow-through system as well as creating customised temperature gradients inside an flow-free liquid chamber.

Given the limitations of existing pumps, heat sinks and vortex generators discussed in the previous section, this liquid metal based system should have the following specifications:

- Simple fabrication process
- Simple integration into miniaturised systems
- Simple operational procedure
- Inexpensive
- Small footprint
- Can be replaced or reconfigured easily
- Can be operated in active mode
- Requires low voltage and consumes low power
- Without conventional moving elements to avoid friction and maintenance
- Highly controllable and predictable
- Versatile (capable of serving as pump, heat sink and vortex generator)
- Short response time capable of operating in dynamic mode
- Can be installed close to the hot spot (avoiding interconnecting tubes)
- Multiple units can be operated simultaneously

These specifications have led to a series of research questions, which will be briefly discussed in the next section.
1.8 Research Questions

The limitations of existing pumping, heat sinking and vortex generation elements, summarised in Sections 1.3 to 1.5 and the need for alternative liquid metal based elements, as specified in Section 1.7, has led to the following four research questions:

**Research Question 1**: Is it possible to develop a liquid based convective cooling system using liquid metal pumps?

This can be broken into the following questions:

1. Is it possible to reduce the temperature of the hot spot by recirculation of basic solutions inside a closed-loop using a liquid metal pump?
2. Can the hot spot temperature be controlled by varying the flow rate of the solution?
3. Is the pumping performance of liquid metal adversely affected by high temperatures?
4. Does the high thermal conductivity of liquid metal droplets allow them to serve as a heat sink?
5. Is it possible to enhance the convective heat transfer of recirculating basic solution by adding thermally conductive nanoparticles into the solution?

**Research Question 2**: Is this liquid metal based convective cooling system capable of rapidly reducing the hot spot temperature?

This can be broken into the following questions:

1. How rapidly the flow rate of recirculating liquid can be changed?
2. Does this lead to rapid change of hot spot temperature?
3. Is this dynamic process repeatable and controllable?
**Research Question 3**: Is it possible to create customised spatial temperature gradients inside an isolated flow-free liquid chamber using liquid metal pumps?

This can be broken into the following questions:

1. Is it possible to induce vortices inside the liquid chamber using a pair of liquid metal pumps?
2. Is it possible to change the configuration and rotational velocity of these vortices on demand?
3. Are these induced vortices capable of changing the spatial temperature gradient of the liquid chamber?
4. How the geometrical properties of the liquid chamber affect the induction of vortices?

**Research Question 4**: Is it possible to create customised spatiotemporal temperature gradients inside the liquid chamber using the aforementioned liquid metal pumps?

This can be broken into the following questions:

1. How rapidly the configuration of vortices can be changed?
2. Is the process of switching from one vortex configuration to another repeatable and controllable?
3. Is it possible to dynamically change the temperature gradient of the liquid chamber by changing the configuration of vortices?
4. Is the above transition process repeatable and controllable?
1.9 Thesis Layout

This research aims to develop novel liquid metal based convective cooling systems for localised cooling of hot spots in flow-through systems as well as creating customised spatiotemporal temperature gradients in flow-free liquid chambers. In particular, the research aims to address the four research questions discussed in Section 1.8. To achieve this goal, the rest of this thesis is structured as outlined below:

Chapter 2 presents a liquid metal based convective cooling system for localised cooling of hot spots. The pumping and cooling performance of the system is examined for various operating conditions. It also demonstrates the utility of the liquid metal pump to serve as a ‘heat sink’. The cooling performance of this system is characterised under both static and dynamic conditions. In dynamic mode, the capability of the system for the rapid cooling of hot spots is explored.

Chapter 3 presents a liquid metal based vortex generator for creating customised spatiotemporal temperature gradients inside an isolated flow-free liquid chamber. It demonstrates the capability of liquid metal pumps to produce highly controllable vortices inside the chamber, and explores how these vortices change the temperature distribution of the liquid. The system is studied under dynamic conditions to investigate whether modifying the configuration of vortices from one to another can lead to rapid change of temperature profiles and eventually temperature gradients inside the liquid chamber.

Chapter 4 summarises the outcomes of this research, and provides some suggestions for future work.
1.10 References


[35] C. Szydzik, K. Khoshmanesh, A. Mitchell, and C. Karnutsch, "Microfluidic platform for separation and extraction of plasma from


CHAPTER 2

An integrated liquid cooling system based on Galinstan liquid metal droplets

2.1 Abstract

The continued miniaturisation of electronic components demands integrated liquid cooling systems with minimised external connections and fabrication costs that can be implanted very close to localised hot spots. This might be challenging for existing liquid cooling systems, as most of them rely on external pumps, connecting tubes, and micro-fabricated heat sinks. This chapter demonstrates an integrated liquid cooling system by utilising a small droplet of liquid metal Galinstan, which is placed over the hot spot. Energising the liquid metal droplet with a square wave signal creates a surface tension gradient across the droplet, which induces Marangoni flow over the surface of droplet and produces a high flow rate of coolant medium through the cooling channel, enabling a ‘soft pump’. At the same time, the high thermal conductivity of liquid metal facilitates the dissipation of heat and extends the heat transfer surface, enabling a ‘soft heat sink’. This facilitates the rapid cooling of localised hot spots. This technology facilitates customised liquid cooling systems with simple fabrication and assembling processes, with no moving parts that can achieve high flow rates with low power consumption.
2.2 Introduction

Thermal management is crucial for further development of high power density, compact electronic devices [1, 2]. Closed loop liquid cooling systems offer higher convective heat transfer coefficient and lower thermal resistance compared to their air cooling counterparts, and have been proven as capable technologies for heat dissipation within compact electronic packages [3]. The evolution of micro/nano technologies has significantly enhanced the capability of liquid cooling systems. The advances in micro fabrication technologies have enabled production of microchannel heat sinks with superior convective heat transfer coefficients, and large surface area to volume ratio [4, 5]. On the other hand, the advances in nanotechnology have led to the development of ‘nanofluids’, a category of fluids with superior thermal conductivity, which are made by adding high thermal conductivity nano sized particles such as carbon nanotubes (CNTs), copper oxide (CuO) and aluminium oxide (Al2O3) into the conventional coolant media [6, 7]. The existing closed loop cooling systems rely on external pumps and connecting tubes to circulate the coolant medium through the system, and therefore occupy a relatively large space, are prone to leakage, and might be rather expensive. The continued miniaturisation and increased power dissipation of electronic components demands new approaches for highly integrated and efficient, yet simple liquid cooling systems.

Gallium-based liquid metal alloys such as eutectic GaIn (75% gallium and 25% indium) [8], and Galinstan (68.5% gallium, 21.5% indium and 10% tin) [9] have recently attracted a significant research attention. Such liquid metal alloys offer remarkable fluidic and metallic properties. This includes high deformability, low viscosity, high surface tension, as well as high electrical and thermal
conductivities [9]. These alloys have a low melting point (lower than 20 °C) and a high boiling point [9], and can be readily used in room temperature. These liquid metal alloys have a low toxicity and very low vapour pressure, and can be considered as a safe replacement for mercury [8, 9]. Various mechanisms have been demonstrated for actuation of EGaIn and Galinstan. This includes electro-chemical [10-12], electro-magnetic [13], photo-chemical [14], nanoparticle-fuelled [15], interfacial tension gradient [16], and thermal gradient [17] mechanisms, enabling such liquid metals to be used in a variety of exciting applications such as soft electronics [18-25], soft motors [10, 15, 26], microfluidics [8, 16, 27, 28], chemical sensing [29, 30], energy harvesting [31], and heat transfer [32, 33].

The intrinsic liquidity and high thermal conductivity of gallium-based liquid metal alloys makes them the favourite coolant medium for liquid cooling of micro devices [34]. The thermal conductivity of these alloys can be further increased by adding high thermal conductivity nanoparticles such as CNTs into the liquid metal [33]. The liquid metal is typically driven through a closed loop by means of a peristaltic pump [34]. However, the problems regarding the cost, maintenance and integration of peristaltic pumps have led to development of alternative pumping mechanisms.

The liquid metals are electrically conductive, and can be driven using electromagnetic force. This concept has been used for circulating sodium-potassium (NaK) liquid metal through nuclear reactors [35], and recently for driving gallium-based liquid metals through compact electronic devices [32]. The electromagnetic pump has no moving parts, and therefore does not cause any noise and vibration in the system. The flow rate of electromagnetic pumps is
proportional to current density \((J)\), magnetic flux density \((B)\), inverse of liquid viscosity \((\mu)\), and the fourth power of cooling channel hydraulic diameter \((D)\) : 

\[
Q \propto J \cdot B \cdot \mu^{-1} \cdot D^4
\]  

[36, 37]. The latter makes the miniaturisation of electromagnetic pumps challenging, as by reducing the hydraulic diameter, a large current density or magnetic flux density is needed to induce the flow. Furthermore, in the case of using a permanent magnet, the strength of the magnet reduces at high temperatures, and eventually the magnet loses its ferromagnetism at temperatures above Curie temperature [38]. Alternatively, the liquid metal can be circulated through the cooling system by means of thermosiphon effect [39]. The flow rate of thermosiphon pumps is proportional to inverse of liquid viscosity \((\mu)\), temperature difference \((\Delta T)\) and the fourth power of cooling channel hydraulic diameter \((D)\) : 

\[
Q \propto \mu^{-1} \cdot \Delta T \cdot D^4
\]  

[40]. Similarly, the miniaturisation of thermosiphon pumps is challenging, as reducing the channel dimensions implies the presence of a large temperature difference to circulate the flow.

Apart from circulating mechanism, gallium-based liquid metals are rather expensive compared to conventional coolants, and amalgamate to most metallic materials (e.g. copper) [41], which might limit their usage as a coolant medium. Therefore, a miniaturised liquid metal based cooling system with no moving parts, low energy consumption, high flow rates, and capable of circulating inexpensive, non-reactive water-based coolants can benefit the development of future compact electronic systems.

This chapter demonstrates a liquid metal based cooling system by placing a liquid metal droplet over the hot spot. Applying a square wave signal with appropriate amplitude and frequency induces Marangoni flow over the surface of
droplet, which circulates sodium hydroxide (NaOH) solution through the cooling channel. The high thermal conductivity of liquid metal droplet facilitates the effective delivery of heat into the stream of liquid, further enhancing the convective cooling. Proof-of-concept experiments, utilising infrared camera and embedded thermistor, demonstrate the capability of such integrated liquid metal based ‘pump’ and ‘heat sink’ for the rapid cooling of hot spots. The performance of the system is studied under a range of operating conditions. The presented liquid metal based cooling system has no moving parts, and is placed right onto the hot spot without the need for connecting tubes. Thus, it has a short response time and can serve as an auxiliary cooling system in the case of a sudden heat surge within the electronic device.
2.3 Experiment setup

The liquid metal based cooling system is schematically shown in Figure 2.1. The system consists of a closed-loop, open-top polydimethylsiloxane (PDMS) channel with a length of 134 mm and a cross-section of 5 mm × 3 mm, that is referred to as ‘cooling channel’. The cooling channel is permanently bonded onto a thin glass slide (60 mm × 24 mm × 100 μm). This glass slide is equipped with a micro-sized, serpentine-shaped resistive heater with an overall surface area of $2.24 \times 10^{-3}$ cm$^2$, which is patterned on its dry side, as shown in Figure 2.1b, and serves as the hot spot upon energising with DC signal. A thermistor with a diameter of 0.5 mm is placed in contact with the microheater to monitor the temperature of the hot spot. The cooling system is integrated onto a thick PDMSs lab to be thermally insulated, as shown in Figure 2.1c.

A 2 mm diameter copper tape with a thickness of 100 μm is placed over the wet side of the glass slide and aligned with respect to the microheater. A droplet of Galinstan (RG Medical Diagnostics, USA) with a diameter of 3 mm is injected onto the copper tape using a pipette to be immobilised, as shown in Figure 2.1d. The cooling channel is entirely filled with 0.3 M NaOH solution. This solution suppresses the oxidation of liquid metal, is electrically conductive and has a low viscosity, which are essential features for inducing Marangoni flows under continuous electrowetting effect [42].

The immersed liquid metal droplet is actuated by applying a square wave DC signal between the two stainless steel electrodes placed at the two sides of liquid metal droplet, and separated by 10 mm. A thin glass side is placed over the top of the cooling channel to avoid evaporation of NaOH solution. The temperature of the cooling channel is monitored through the top glass slide using an infrared
A camera (FLIR systems, Thermo Vision A320, Sweden) interfaced with ThermaCAM researcher software installed in a work station. The actual experimental setup is presented in Figure 2.2.

Figure 2.1: Schematics of liquid metal based cooling system: (a) Exploded schematic. (b) Close-up image of the microheater, (c) Assembled schematics. (d) Close-up image of Galinstan droplet placed above the hot spot. Reproduced with permission from ACS Applied Materials & Interfaces, 2016, 8, 2173-2180. Copyright (2018) American Chemical Society.
Figure 2.2: (a) Experimental setup. (b) Close-up image of the liquid metal based cooling system.
2.4 Results and Discussion

2.4.1 Operation of liquid metal based cooling system

Proof-of-concept experiments are conducted to demonstrate the capability of the liquid metal based cooling system for the rapid and efficient cooling of a hot spot in a small-scale system. In doing so, a 3 mm diameter Galinstan droplet is placed over the hot spot, and the cooling channel is filled with 0.3 M NaOH solution, as illustrated in Figure 2.3. The microheater is energised with a DC signal (3.5 V, 0.04 A) to provide a hot spot. The immersed Galinstan droplet is then actuated by applying a square wave signal (200 Hz, 5 V<sub>p-p</sub>, 2.5 V DC offset and 50% duty cycle) between the two stainless steel electrodes. The application of such a square wave signal imbalances the interfacial tension between the liquid metal and the surrounding NaOH solution along the surface of liquid metal droplet. Such induced interfacial tension gradient produces Marangoni flow along the surface of liquid metal droplet, and consequently pumps the surrounding liquid through the cooling channel [43].

The flow rate of NaOH is measured to be as ~ 28.1 ml/min. In order to measure the flow rate of NaOH solution through the cooling channel, a droplet of blue food dye is added into the channel. Then, the droplet is actuated with a square wave DC signal with desired magnitude and frequency. The induced flow rate of NaOH solution is estimated by measuring the amount of time required for the flow to circulate one cycle, as shown in Figure 2.3. The average velocity of flow is calculated by dividing the length of the cooling channel by the measured time. The flow rate is then calculated by multiplying the average velocity by the cross sectional area of the cooling channel.
The Reynolds number of NaOH flow through the channel, defined as \( Re_{\text{channel}} = \frac{\rho_{\text{NaOH}} \cdot U_{\text{NaOH}} \cdot D_{\text{channel}}}{\mu_{\text{NaOH}}} \), is obtained as \( Re_{\text{channel}} \approx 110 \), indicating the laminar characteristics of flow even at such a high flow rate (where \( \rho \) and \( \mu \) are the density and dynamic viscosity of the coolant medium, \( U \) is the mean velocity of the flow, and \( D \) is the hydraulic diameter of the channel).

**Figure 2.3:** Actuation of liquid metal based pump with a droplet of blue dye used to visualise the induced flow of NaOH solution. (a-f) Sequential snapshots of flow using a 3 mm Galinstan droplet immersed in 0.3 M NaOH, when operated with a square wave signal (200 Hz, 5 V\(_{p-p}\), 2.5 V DC offset and 50% duty cycle).
The rapid cooling of hot spot is monitored by infrared camera. Sequential snapshots presented in Figure 2.4a-f show the variations of temperature of the cooling channel with respect to time. Before actuating the liquid metal droplet ($t=0$ s), natural convection is the dominant mode of heat transfer within the cooling channel. This leads to accumulation of heat over the hot spot and formation of a hot region with a peak temperature of $36.5 \, ^\circ C$ above the hot spot. The surface of Galinstan droplet is shiny and reflects the infrared radiation [44], and therefore seems as a cavity. In contrast, after actuating the liquid metal droplet, forced convection becomes the dominant mode of heat transfer within the cooling system. The inducing of a counter-clockwise flow through the channel enhances the propagation of heat, as evidenced by the quick migration and dissipation of the yellow contour through the channel. Interestingly, after only one flow cycle, the temperature becomes almost uniform through the channel, and reduces to $\sim 29.5 \, ^\circ C$.

To further investigate the heat distribution, the variations of temperature along three representative lines, which pass through the middle of the two long arms of the cooling channel and the hot spot, are investigated, as shown in Figure 2.4a. The graphs clearly show the non-uniform distribution of temperature along the cooling channel, before actuating the liquid metal droplet ($t=0$ s). The temperature reaches a peak value of $36.5 \, ^\circ C$ over the hot spot and sharply reduces to $25.5 \, ^\circ C$ at the left end of the channel (Figure 2.4g). Conversely, Figure 2.4h shows the uniform distribution of temperature after actuating the liquid metal droplet for 5 seconds. The obtained temperature is $\sim 29.5 \, ^\circ C$, which is $\sim 7 \, ^\circ C$ less than the peak temperature observed in the absence of flow.
Figure 2.4: Heat dissipation through the liquid metal based cooling system: (a-f) Sequential snapshots obtained by infrared camera showing the variations of temperature with respect to time. Temperature distribution along three representative lines, as shown in Figure 2.4a obtained at (g) before actuating the liquid metal, and (h) 5 seconds after actuating the liquid metal). Results are obtained by using a 3 mm Galinstan droplet immersed in 0.3 M NaOH solution, and actuated with a square wave signal (200 Hz, 5 V_{\text{p-p}}, 2.5 V DC offset and 50 % duty cycle). The microheater is energised with a 3.5 V, 0.04 A DC signal. Reproduced with permission from *ACS Applied Materials & Interfaces*, 2016, 8, 2173-2180. Copyright (2018) *American Chemical Society*.
2.4.2 Estimating the flow rate of coolant medium

The physics underlying the operation of the liquid metal based cooling system can be described as follows. In general, the surface tension between the liquid metal droplet and the surrounding NaOH remains constant across the entire droplet. However, creating a surface tension gradient across the liquid metal droplet can lead to the flow of NaOH from the low surface tension region of droplet towards its high surface tension region, which is known as ‘Marangoni flow’, and can be described as follows [27]:

\[
\frac{\partial \gamma}{\partial t}_{\text{Droplet}} = \tau_{\text{Droplet}} = \mu \left| \frac{\partial U_{\text{Surface}}}{\partial \mathbf{n}} \right|_{\text{Droplet}}
\]  \hspace{1cm} (2.1)

where \(\gamma\) is the surface tension, \(\tau_{\text{Droplet}}\) is the shear stress induced over the surface of droplet, \(\mu\) is the viscosity of the solution surrounding the liquid metal droplet, \(U_{\text{Surface}}\) is the induced ‘Marangoni flow’ velocity at the surface of droplet, while \(\mathbf{\tilde{t}}\) and \(\mathbf{\tilde{n}}\) are the tangential and normal vectors along the surface of droplet, respectively.

Immersing of a Galinstan droplet in strong alkalis such as NaOH solution leads to a chemical reaction between the gallium and the alkali solution, which produces gallates such as \([\text{Ga}^{2+}]^{-}\) anions at the interface of droplet-NaOH solution. These anions make the droplet surface negatively charged. This in turn leads to the gathering of positively charged ions at the interface due to ionic adsorption, and creates an electrical double layer (EDL) [43]. In the absence of external potential, the potential difference across the EDL, which is referred to as \(V_{\text{EDL}}\), is uniform across the surface of droplet, and has a magnitude of \(V_0 = q_0/c\), in which \(q_0\) is the initial change of EDL and \(c\) is the capacitance of EDL per unit area. However, applying a voltage across the droplet can imbalance the
distribution of $V_{\text{EDL}}$, and change the surface tension between the droplet and the surrounding NaOH solution (as shown in Figure 2.5a), as described by Lippmann’s equation [42]:

$$\gamma = \gamma_o - \frac{1}{2} c V_{\text{EDL}}^2,$$

where, and $\gamma_o$ is the surface tension when $V=0$.

Applying a voltage across the droplet leads to a voltage drop of $\Delta \varphi$ across the two sides of the droplet, which according to Ohm’s law can be expressed as follows: $\Delta \varphi \propto V_{\text{Electrode}} \cdot D_{\text{Droplet}} / A_{\text{Gap}}$, where $V_{\text{Electrode}}$ is the magnitude of the voltage applied between the two stainless steel electrodes to actuate the liquid metal, $D_{\text{Droplet}}$ is droplet diameter, and $A_{\text{Gap}}$ is the area of the gap between the droplet and the channel walls through which the NaOH solution flows. In this case, the $V_{\text{EDL}}$ at the high and low voltage sides of the droplet is obtained as $V_o + \Delta \varphi / 2$ and $V_o - \Delta \varphi / 2$, respectively. Substituting these values in the Lippmann’s equation, the surface tension difference across the two ends of the liquid metal droplet, is estimated as $\Delta \gamma = c V_o \Delta \varphi$ [29].

Combining the above mentioned equations, the flow rate of NaOH through the cooling channel with respect to the influential parameters of the cooling system, including the magnitude of applied voltage, droplet diameter, channel dimension, and the viscosity of the coolant medium, is obtained as follows:

$$Q_{\text{NaOH}} \sim \frac{V_{\text{Electrode}}}{A_{\text{Gap}}} \cdot D_{\text{Droplet}}^2 \cdot \frac{W_{\text{Channel}} H_{\text{Channel}}}{L_{\text{Channel}}} \cdot \frac{1}{\mu_{\text{NaOH}}}.$$
2.4.3 Characterisation of the flow rate of coolant medium with respect to the parameters of the cooling system

The shear stress over the surface of liquid metal droplet can be approximated as below:

\[
\frac{\partial y}{\partial t}_{\text{Droplet}} = \tau_{\text{Droplet}} \rightarrow \tau_{\text{Droplet}} \sim \frac{\Delta y_{\text{Droplet}}}{\pi 0.5 D_{\text{Droplet}}} \quad (2.2)
\]

For internal flows, the relation between the shear stress over the surfaces of the liquid metal droplet and the cooling channel walls can be described as below:

\[
\tau_{\text{Droplet}} \cdot A_{\text{Droplet}} = \tau_{\text{Channel}} \cdot A_{\text{Channel}}
\]

\[
\rightarrow \tau_{\text{Channel}} = \tau_{\text{Droplet}} \cdot \frac{\pi D_{\text{Droplet}}^2}{L_{\text{Channel}} H_{\text{Channel}}} \quad (2.3)
\]

The shear stress over the surface of cooling channel walls can be approximated as below:

\[
\tau_{\text{Channel}} = \mu_{\text{NaOH}} \frac{\partial U}{\partial n}_{\text{Channel}} \sim \mu_{\text{NaOH}} \frac{U_{\text{NaOH}}}{H_{\text{Channel}}}
\]

\[
\sim \mu_{\text{NaOH}} \cdot \frac{Q_{\text{NaOH}}}{W_{\text{Channel}} H_{\text{Channel}}^2} \quad (2.4)
\]

Therefore, the flow rate of NaOH through the cooling channel is obtained as below:

\[
Q_{\text{NaOH}} \sim \tau_{\text{Channel}} \cdot \frac{W_{\text{Channel}} H_{\text{Channel}}^2}{\mu_{\text{NaOH}}} \quad (2.5)
\]

Combining equations 2.2, 2.3 and 2.5, the flow rate of NaOH is obtained as below:
The surface tension gradient over the surface of liquid metal droplet is due to continuous electrowetting, created by applying a voltage difference over the two sides of liquid metal, which can be calculated as: \( \Delta \gamma_{Droplet} = c V_o \Delta \varphi \). As a result, the flow rate of NaOH with respect to the parameters of the system can be described as below:

\[
Q_{NaOH} \sim \tau_{Droplet} \cdot \pi D_{Droplet}^2 \cdot \frac{W_{Channel}H_{Channel}}{L_{Channel}} \cdot \frac{1}{\mu_{NaOH}}
\]

(2.6)

\[
\rightarrow Q_{NaOH} \sim \Delta \gamma_{Droplet} \cdot D_{Droplet} \cdot \frac{W_{Channel}H_{Channel}}{L_{Channel}} \cdot \frac{1}{\mu_{NaOH}}
\]

(2.7)
The equivalent thermal circuit describing the heat dissipation through the liquid metal droplet is shown in Figure 2.5b, using which the temperature difference between the hot spot and the surrounding NaOH flow can be described as follows:

\[ T_{\text{Hot spot}} - T_{\text{NaOH}} = \]

\[ q \left( \frac{L_g}{k_g A_g} l_{\text{glass}} + \frac{L_c}{k_c A_c} l_{\text{copper tape}} + \frac{L_d}{k_d A_d} l_{\text{droplet}} + \frac{1}{h_{\text{NaOH}} A_d} \right) \quad (2.8) \]

where \( q \) is the heat generated at the hot spot, which can be estimated by multiplying the voltage energising the microheater by its current. \( L, A \) and \( k \) are the thickness, surface area and thermal conductivity of the glass, copper tape and liquid metal droplet, \( h \) is the convection heat transfer coefficient of the coolant medium, and \( T \) is temperature. It should be noted that this equation neglects the internal eddies inside the Galinstan droplet induced either by Marangoni flow or the natural convection [45].
Figure 2.5: Working mechanism of actuated liquid metal over the hot spot. (a) Schematic of liquid metal droplet surface charge distribution when an electric potential is applied between the electrodes. The generation of harmonic Marangoni flow is enabled by continuous electrowetting effect at the surface of the liquid metal droplet, upon the application of a square wave DC signal. (b) Schematic and equivalent thermal circuit of heat dissipation through liquid metal droplet, where T is temperature and R is thermal resistance. Reproduced with permission from ACS Applied Materials & Interfaces, 2016, 8, 2173-2180. Copyright (2018) American Chemical Society.
The geometrical and thermo-physical properties of glass slide, copper tape and liquid metal droplet are constant, and therefore the above equation can be simplified as $T_{\text{hot spot}} - T_{\text{NaOH}} \propto q/h_{\text{NaOH}}$. Experiments indicate that $T_{\text{NaOH}}$ does not depend on the flow rate of NaOH solution through the cooling channel, and in fact depends on the ambient temperature, as shown in Figure 2.6.
Figure 2.6: NaOH solution temperature within the left arm of cooling channel measured by infrared camera, when operated the droplet with a square wave signal (5 V_{p-p}, 2.5 V DC offset and 50% duty cycle) of different frequencies: (a) at 1 Hz, (b) at 200 Hz, (c) at 1000 Hz. (d) NaOH solution temperature measured by infrared camera vs droplet operating frequency. (e) NaOH solution temperature measured by infrared camera vs flow rate.
Therefore when energising microheater with a constant voltage, the hot spot temperature will be inversely proportional to convective heat transfer coefficient:

\[ T_{\text{hot \, spot}} \propto \frac{1}{h_{\text{NaOH}}} \]

The \( h_{\text{NaOH}} \) can be estimated as the average heat transfer coefficient of flow over a spherical structure, as described below [46]:

\[
Nu_{\text{NaOH}} = \frac{h_{\text{NaOH}} D_{\text{Droplet}}}{k_{\text{NaOH}}}
\]

\[
= 2 + \left[ 0.4 \, Re_{\text{Droplet}}^{0.5} + 0.06 \, Re_{\text{Droplet}}^{0.66} \right] Pr_{\text{NaOH}}^{0.4} \left( \frac{\mu_{\text{NaOH}}}{\mu_{\text{surface}}} \right)^{0.25}
\] (2.9)

Where \( Nu \), \( k \) and \( \mu_{\text{surface}} \) are the Nusselt number, thermal conductivity, and the viscosity of the coolant medium in contact with droplet surface. Based on the conditions described in Figure 2.4, the Reynolds number of flow across the liquid metal droplet, defined as \( Re_{\text{Droplet}} = \rho_{\text{NaOH}} \cdot U_{\text{NaOH}} \cdot D_{\text{Droplet}} / \mu_{\text{NaOH}} \) is obtained as \( Re_{\text{Droplet}} \approx 80 \). The Prandtl number of NaOH solution, defined as \( Pr_{\text{NaOH}} = C_p_{\text{NaOH}} \cdot \mu_{\text{NaOH}} / k_{\text{NaOH}} \) is obtained as \( Pr_{\text{NaOH}} \approx 4.62 \), where \( C_p \) is the specific heat capacity of the coolant medium.

Using this equation, and based on the conditions described in Figure 2.4, the average \( Nu_{\text{NaOH}} \) and \( h_{\text{NaOH}} \) over the droplet are obtained as 10.62, and 2123 W/m²K, respectively. Moreover, the relation between the heat transfer coefficient and the flow rate is obtained as \( h_{\text{NaOH}} \propto Re_{\text{Droplet}}^{0.5} \) and therefore, the relation between the flow rate and the hot spot temperature can be expressed as \( T_{\text{hot \, spot}} \propto Re_{\text{Droplet}}^{0.5} \).
2.4.4 Characterising the performance of the liquid metal based cooling system under various conditions

A series of experiments are conducted to identify the role of different components of the liquid metal based cooling system, as presented in Figure 2.7. The same conditions described in Figure 2.4 are applied, and the thermal images for different operating conditions are captured while heating the system under steady state conditions, as explained below:

*Case 1:* By inserting an empty cooling channel onto the hot spot (in the absence of Galinstan droplet and NaOH solution) a localised hot region with a peak temperature of 55.2 °C is observed at the left arm of the cooling channel (Figure 2.7a). Natural convection is the dominant mode of heat transfer between the hot spot and the surrounding air, with the low thermal conductivity of the air limiting the dissipation of heat through the cooling channel.

*Case 2:* By inserting a 3 mm Galinstan droplet within the cooling channel such that it locates over the hot spot (in the absence of NaOH solution), a hot region with a peak temperature of 42.7 °C is observed around the liquid metal droplet (Figure 2.7b). Compared to *Case 1*, the peak temperature reduces by 12.5 °C and the hot region expands by 80 %. The thermal conductivity of Galinstan is 16.5 W/mK, which is ~ 650 times higher than that of air. As a result, the droplet serves as a heat sink and facilitates the passage of heat into the cooling channel. Increasing the size of droplet leads to reducing the temperature of hot spot. Enlarging the droplet facilitates the thermal conduction through the droplet, and enhances the natural convection over the surface of droplet.
Case 3: By filling the cooling channel with 0.3 M NaOH solution (in the absence of Galinstan droplet), the peak temperature reduces to 38.2 °C and the hot region is further expanded within the right arm of the cooling channel (Figure 2.7c). The enhanced heat transfer compared to Case 1 is attributed to the thermal conductivity of NaOH solution, which is 23.5 times higher than that of air.

Case 4: By inserting 3 mm Galinstan droplet into the filled cooling channel (combinations of second and third cases), the peak temperature further reduces to 36.8 °C and the hot region occupies almost the entire left arm of the cooling channel (Figure 2.7d). This further confirms the role of liquid metal droplet serving as a heat sink.

Case 5: By actuating the Galinstan droplet (as described for Figure 2.4), the temperature becomes almost uniform throughout the cooling channel and reduces to 29.5 °C (Figure 2.7e). In contrast to Cases 1 to 4, where natural convection is the dominant mode of heat transfer between the hot surface/droplet and the surrounding medium, in this case, the forced convection becomes the dominant mode of heat transfer as the liquid metal droplet induces a flow of NaOH solution through the cooling channel. This means that the droplet serves as a pump and at the same time as a heat sink. This is significant as this configuration enables the hot spot-targeted liquid cooling of electronic devices [2]. Similar trends are observed by increasing the amplitude of the DC signal energising the microheater to 4.5 V, 0.06 A, as shown in Figure 2.7a´-e´.

The temperature contours presented in Figure 2.7a-e are captured using an infrared camera through the top glass slide, and therefore do not reflect the actual temperature of the hot spot. To address this, the temperature of the hot spot is measured using a thermistor (as detailed in Figure 2.1). Experiments are
conducted in different cases, as described above, and also under various DC signals energising the microheater, as presented in Figure 2.7f. For Case 1 (empty cooling channel) the temperature of hot spot is obtained as 65.9 °C. For Case 2 (liquid metal droplet located over the hot spot), the temperature of hot spot reduces to 59.7 °C, which is 6.2 °C less than case 1. For Case 3 (cooling channel filled with NaOH), the temperature of hot spot reduces to 46.6 °C, indicating a 19.3 °C reduction compared to case 1. For Case 4 (combination of cases 2 and 3), the temperature of hot spot reduces to 44.6 °C, which is slightly less than that of case 3. Finally, for Case 5 (actuating the liquid metal droplet to induce flow), the temperature of hot spot reduces to 37.1 °C, which is 28.8 °C and 7.5 °C cooler than cases 1 and 4, respectively. The results confirm the potential of the liquid metal based cooling system for the efficient cooling of hot spots by means of forced convective. A similar trend can be observed by energising the microheater at different voltages. The power density of the microheater is obtained as 62.5 W/cm² when energised with a 3.5 V, 0.04 ADC signal, and increases to 120.5 W/cm² when energised with a 4.5 V, 0.06 ADC signal. Higher voltages are avoided as the thin glass accommodating the microheater can break under excessive temperatures of the hot spot.
Figure 2.7: Analysing the temperature at different operating conditions for the liquid metal based cooling system. Temperature contours measured by infrared camera when energising microheater at 3.5 and 4.5 V: (a-a’) Empty channel. (b-b’) A 3 mm liquid metal droplet placed above the microheater. (c-c’) Channel filled with 0.3 M NaOH solution. (d-d’) 3 mm liquid metal droplet immersed in 0.3 M NaOH solution and placed above the microheater. (e-e’) In the presence of flow, by actuating the liquid metal droplet with a square wave signal (200 Hz, 5 V<sub>p-p</sub>, 2.5 V DC offset and 50% duty cycle). (f) Hot spot temperature measured by thermistor when energising the microheater with different voltages ranging from 1 to 4.5 V. Reproduced with permission from *ACS Applied Materials & Interfaces*, 2016, 8, 2173-2180. Copyright (2018) American Chemical Society.
*Case 6:* Relocating the liquid metal before or after the hot spot leads to slight increase of hot spot temperature (~ 0.3 °C), as presented in Figure 2.8. Unlike *Case 5,* in which the droplet serves as both a pump and a heat sink, in this case, the droplet only serves as a pump. However, the slight difference between the hot spot temperatures demonstrates the potential of the proposed system for cooling of multiple hot spots.
Figure 2.8: Hot spot temperature measured by thermistor for different operating conditions of the liquid metal based cooling system. (a-c) Schematics of liquid metal droplet at different locations with respect to hot spot. (d) Hot spot temperature measured by thermistor vs droplet operating frequency when actuating droplet with a square wave signal (5 V$_{p-p}$, 2.5 V DC offset and 50% duty cycle) at different operating conditions.
Case 7: The convective heat transfer can be further increased by circulating nanofluids through the cooling channel. For example, applying a suspension of carbon black nanoparticle-NaOH solution with a particle concentration of 0.1 wt% (provided by adding 5 mg of carbon black powder with an average diameter of 50 nm and a thermal conductivity of 6-174 W/mK [47] into 5 ml of 0.3 M NaOH solution) reduces the hot spot temperature by 1.2 °C, as presented in Figure 2.9. Similarly, applying a suspension of Al$_2$O$_3$ nanoparticle-NaOH solution with a particle concentration of 0.1 wt% (provided by adding 5 mg of 5 mg of Al$_2$O$_3$ nanoparticles with a diameter of 10-50 nm powder and a thermal conductivity of 20-29 W/mK into 5 ml of 0.3 M NaOH solution) decreases the hot spot temperature by 1 °C. The application of these nanoparticles increases the overall thermal conductivity of the suspension, and in turn enhances the convective heat transfer coefficient of the flow. It is worth mentioning that the p-type semiconducting nanoparticles should be avoided as they could disturb the continuous electrowetting over the surface of liquid metal [48], as previously shown by our group.

Carbon black nanofluid is made by adding 5 mg of carbon black powder (50 nm, Sigma-Aldrich) with a thermal conductivity of $6 \sim 174$ W/mK into 5 ml of 0.3 M NaOH solution, providing the particle suspension with a concentration of ~0.1 wt%.

Similarly, Al$_2$O$_3$ nanofluid is made by adding 5 mg of 5 mg of Al$_2$O$_3$ nanoparticle powder (10–50 nm, Sigma-Aldrich) with a thermal conductivity of $20 \sim 29$ W/mK into 5 ml of 0.3 M NaOH solution, providing the particle suspension with a concentration of ~0.1 wt%. Next, 10 μl SDS (Sodium dodecyl sulfate) surfactant is added into the particle suspension and sonicated for at least
10 min using a high power ultrasonicator (Qsonica Sonicators) to avoid particle aggregation.

**Figure 2.9:** Hot spot temperature measured by thermistor for different operating liquid of the liquid metal based cooling system. (a) Carbon black and Al₂O₃ nanofluids. (b) Schematics of liquid metal based cooling system by circulating nanofluids. (c) Hot spot temperature measured by thermistor vs different operating liquid when actuating droplet with a square wave signal (5 V_{p-p}, 2.5 V DC offset and 50 % duty cycle) at different frequencies.
To further characterise the performance of our liquid metal based cooling system, the Galinstan droplet is actuated with different frequencies and voltages while energising the microheater with a constant DC signal of 3.5 V, 0.04 A, as shown in Figure 2.10.

Figure 2.10a shows the variations of NaOH solution flow rate and hot spot temperature (measured by thermistor) with respect to droplet operating frequency when activating the liquid metal droplet with a constant voltage of 5 V<sub>p-p</sub>, 2.5 V DC offset. For visual comparison, the cooling performance of the system is monitored under different frequencies using infrared camera. A flow rate of ~11.5 ml/min is obtained at 1 Hz. The flow rate of NaOH solution keeps increasing until reaching a peak value of ~28.1 ml/min at 200 Hz. Further increase of frequency reduces the flow rate, leading to a flow rate of ~6.9 ml/min at 10 kHz. The decrease of flow rate at high frequency could be attributed to the fact that at high frequencies, the ions cannot be fully redistributed within the EDL before de-electrowetting occurs to generate the maximum pressure difference [29]. Increasing the flow rate of NaOH intensifies the forced convection over the surface of liquid metal droplet ($h_{NaOH} \propto Re_{NaOH}^{0.5}$), leading to reducing the hot spot temperature. This results in a minimum hot spot temperature of 35.5 °C at 200 Hz.

Figure 2.10b shows the variations of NaOH flow rate and hot spot temperature (measured by thermistor) with respect to droplet operating voltages when activating the liquid metal droplet with a constant frequency of 200 Hz. The cooling performance of the system under different voltages is captured by infrared camera. No noticeable flow rate is observed at voltages below 2 V<sub>p-p</sub>, after which the flow rate increases linearly with respect to voltage. However, by
increasing the voltage above 5 V p-p, hydrogen bubbles are formed over the surface of stainless steel electrodes due to electrolysis. The bubbles induce additional pressure drop through the cooling channel and reduce the flow rate of NaOH. The hot spot temperature reduces from 43.5 °C at 2 V p-p to 35.5 °C at 5 V p-p but does not reduce significantly at higher voltages.
Figure 2.10: Characterisation of liquid metal based cooling system: (a) Flow rate and hot spot temperature measured by thermistor vs droplet operating frequency when actuating droplet with a square wave signal (5 V p-p, 2.5 V DC offset and 50% duty cycle). (b) Flow rate and hot spot temperature vs droplet operating voltage when actuating droplet with a square wave signal of 200 Hz. Reproduced with permission from *ACS Applied Materials & Interfaces*, 2016, 8, 2173-2180. Copyright (2018) *American Chemical Society*. 
2.4.5 Investigating the cooling performance of the liquid metal based cooling system under transient conditions

Figures 2.10a-b corresponds to steady operating conditions, in which the NaOH solution is constantly flowing through the cooling channel. Unlike conventional pumps (e.g. syringe or peristaltic pumps), the liquid metal based pump has no moving parts, and can produce high flow rates immediately. More importantly, the pump is located right onto the hot spot without the need for connecting tubes. These two conditions enable the liquid metal droplet cooling system to have a short response time, enabling it to be used in the case of a sudden temperature rise of the hot spot.

The performance of the liquid metal based cooling system in such dynamic (unsteady) cases is assessed, as shown in Figure 2.11a. To mimic a sudden temperature rise, the microheater is energised with a 3.5 V, 0.04 ADC signal, and let the hot spot reach a peak temperature of 44.5 °C, and maintain it for at least 300 s to reach a steady state condition. After which, the liquid metal droplet is actuated with a square wave signal (5 V_{pp}, 2.5 V DC offset) with various operating frequencies ranging from 1 to 10 kHz. The droplet is actuated for at least 30 s to reduce the hot spot temperature. The temperature of hot spot is constantly monitored using the thermistor to capture the cooling dynamics of the system. The results indicate the rapid temperature reduction of the hot spot following the actuation of the liquid metal droplet, after which the hot spot reaches a steady temperature. The amount of time taken to reach a steady temperature is regarded as ‘cooling time’. Figure 2.11b shows the variations of ‘cooling time’ versus droplet operating frequency. The ‘cooling time’ varies from ~5.6 s to 12.9 s with the shortest ‘cooling time’ obtained at 200 Hz.
corresponding to the highest flow rate of NaOH solution, as presented in Figure 2.10a.

Figure 2.11: The performance of liquid metal based cooling system: (a) Hot spot temperature measured by thermistor vs time when actuating droplet at different frequencies. (b) Cooling time vs droplet operating frequency. Reproduced with permission from ACS Applied Materials & Interfaces, 2016, 8, 2173-2180. Copyright (2018) American Chemical Society.
2.5 Conclusion

In summary, this chapter demonstrated an integrated liquid metal based system for the localised liquid cooling of hot spots. Upon application of a square wave DC signal, the liquid metal droplet serves as a ‘pump’, driving the coolant medium through the cooling channel. Owing to its high thermal conductivity, the liquid metal droplet also serves as a ‘heat sink’, conducting heat into the stream of the coolant medium. The flow rate of the medium can be readily tuned by varying the voltage and frequency of the applied DC signal. Suspensions of nanofluids can also be circulated through the cooling channel to enhance the convective heat transfer. The elimination of external connections and moving parts reduces the response time of this cooling system, enabling the rapid cooling of hot spots in the case of a heat surge within the electronic devices. The transient cooling process can be shortened by operating the liquid metal pump at the frequency at which the highest flow rate of the coolant medium is induced. Simple fabrication and assembling processes facilitates multiple/customised liquid cooling systems for thermal management of compact electronic devices with high power density and limited space.
2.6 References


CHAPTER 3

Customised spatiotemporal temperature gradients created by a liquid metal enabled vortex generator

3.1 Abstract

Generating customised temperature gradients in miniaturised flow-free liquid chambers is challenging due to the dominance of diffusion. Inducing internal flows in the form of vortices is an effective strategy for overcoming the limitations of diffusion in such environments. Vortices can be produced by applying pressure, temperature and electric potential gradients via miniaturised actuators. However, the difficulties associated with the fabrication, integration, maintenance and operation of such actuators hinders their utility. This chapter investigates the utilisation of liquid metal enabled pumps to induce vortices inside a miniaturised liquid chamber. The configuration and rotational velocity of these vortices can be controlled by tuning the polarity and frequency of energising electrical signal. This allows for creating customised spatial temperature gradients inside the chamber. The absence of conventional moving elements in the pumps facilitates the rapid reconfiguration of vortices. This enables quick transition from one temperature profile to another, and creating customised spatiotemporal temperature gradients. This allows for oscillating temperature from 35 to 62 °C at the hot spot, and from 25 to 27 °C at the centre of the vortex within 15 seconds. The liquid metal enabled vortex generator can be fabricated, integrated and operated easily, and offers unprecedented opportunities for studying thermo-responsive materials and biological samples.
3.2 Introduction

Generation of temperature gradients is essential for conducting various biological, chemical and physical studies such as magnification of nucleic acids (known as polymerase chain reaction) [1, 2], thermal stimulation of cells for inducing various diseases [3] or expression of temperature-sensitive proteins [4], actuation of thermo-responsive polymers and hydrogels [5, 6], as well as the movement of droplets under thermocapillary effect [7]. Such experiments are generally performed in enclosed chambers such as Petri dishes or multi-well plates. In the absence of liquid flow in such enclosed environments, diffusion becomes the dominant mode of heat and mass transfer within the stored liquid, limiting the ability for producing customised spatial and temporal gradients.

A logical way to overcome the inherent limitations of diffusion is to induce internal flows in the form of vortices within those chambers, as widely used in diffusion-governed microfluidic systems. Vortices can be produced passively, through sudden expansion of channel dimensions [8, 9], the patterning of asymmetric grooves on the bottom or top surfaces of the channel [10, 11], and using curved channels for inducing centrifugal forces [12, 13]. However, the generation of vortices in such devices relies on the existence of liquid flow in the microfluidic channel, which typically does not exist in enclosed chambers.

Vortices can also be generated actively using mechanical, acoustic, thermal and electrical mechanisms. Vortices can be generated mechanically by rotating elements such as stirrers [14] and impellers [15] to recirculate the flow inside the chamber. The resultant vortices have a long range and their rotational speed and direction can be easily controlled. Despite these advantages, the problems associated with fabrication, integration and maintenance of these mechanical
elements inhibits their utility. Vortices can also be induced acoustically by the propagation of acoustic waves through the fluidic system [16, 17]. Alternatively, vortices can be induced indirectly by causing oscillating motions along sharp edges [18] or microscale bubbles trapped inside cavities [19-21] enabling localised and controllable vortices. Despite versatility, and insensitivity to liquid properties, the problems associated with the stability and positioning of the bubbles can limit the utility of such devices [18].

In comparison, thermal vortices are produced by patterning heaters along the chamber walls to cause natural convection [22, 23], creating strong localised electric fields in conjunction with a conductive liquid to cause Joule heating effect [24-26], or applying focused laser beams [27-29]. However, the local heating of stored liquid leads to changing its natural temperature distribution which might be undesirable for some applications, and can accelerate the rate of evaporation. In addition, vortices can be generated by applying strong electric fields inside the chamber. Application of electrically charged solutions induces electrohydrodynamic (also known as electrokinetic) driven flows at the surface of electrodes [30-33], which leads to the formation of vortices inside the chamber. Electroosmosis is among the most common electrokinetic driven flows, which is used for driving and mixing charged solutions in microfluidic structures [34-37]. However, the issues associated with the implementation of electrodes along the surface of the chamber, as well as the presence of unwanted chemical reactions such as electrolysis at the surface of electrodes (which often occur with low frequency signals) can deem such systems impractical.

A vortex generator system, which can be easily fabricated, operated, and maintained, consumes low power, and is capable of producing long range,
controllable and fast responding vortices facilitates the creation of customised spatial and temporal gradients in enclosed chambers.

Gallium-based liquid metal alloys, and in particular EGaIn (gallium-indium alloy) and Galinstan (gallium-indium-tin alloy) are non-toxic liquid metals, which have steadily replaced mercury [38, 39]. These alloys are liquid at room temperature, and can be embedded in soft elastomers such as polydimethylsiloxane (PDMS) to make customised and highly stretchable electronic devices [38, 40-47]. The electrical properties, and hence the specifications of such devices, can be altered by reconfiguring the liquid metal inside the microfluidic structure, enabling highly tunable devices [40, 48-50].

Liquid metal structures can be used in continuous (e.g. long columns flowing inside channels) or discontinuous (e.g. droplets patterned onto predetermined locations of the system [51]) arrangements. Such structures can be activated using various electrical [51-55], magnetic [56, 57], optical [58] and more recently self-powered [59-62] mechanisms. Among these mechanisms, electrical actuation of droplets (covering electrocapillary, electrowetting and electrochemical mechanisms) [63] has attracted a lot of attention due to its simplicity, versatility and controllability. Electrical actuation has also allowed for inducing surface tension driven flows at the surface of stationary droplets [52], enabling pumps [51], mixers [64] and convective coolers [65], which are the building blocks of future liquid metal enabled microfluidic systems [66].

In this chapter, a pair of liquid metal pumps made of Galinstan droplets are utilised to generate highly controllable vortices inside a liquid chamber. Actuation of droplets with a square wave signal induces surface tension driven flows at the surface of droplets, leading to the formation of vortices inside the
liquid chamber. These vortices allow the heat to be dissipated inside the chamber by both convection and diffusion mechanisms. Customised spatial temperature gradients can be created according to the configuration and rotational velocity of vortices, which can be both modulated by the polarity and frequency of the energising signal. Experiments indicate that the vortices can be regenerated or reconfigured almost instantly. This enables customised spatiotemporal temperature gradients to be created in repeatable cycles.
3.3 Experimental setup

The experimental setup presented in Figure 3.1 consists of an open-top cylindrical chamber ($D_{chamber} = 12$ mm, $H_{chamber} = 5$ mm), fabricated from polymethylmethacrylate (PMMA), to accommodate the working liquid. Two narrow channels, fitted with a droplet seat in the middle are patterned at the two sides of the liquid chamber to accommodate two Galinstan droplets ($D_{droplet} = 3$ mm) and the associated stainless steel electrodes required for energising the droplets. A 50 $\Omega$ resistor twined with a copper wire is used as the hot spot, which is positioned inside the liquid chamber using a mechanical holder. The temperature distribution inside the liquid chamber is monitored using an infrared camera (FLIR systems, Thermo Vision A320, Sweden) interfaced with ThermaCAM researcher software (Figure 3.1a). The liquid chamber is filled with 0.3 M NaOH solution up to a height of 4 mm. The Galinstan droplets are independently actuated by applying a square wave signal (200 Hz, 5 V peak-to-peak with 2.5 V offset, 50 % duty cycle) to the stainless steel electrodes (Figure 3.1b).

The application of voltage reduces the surface energy at the interface of liquid metal droplet and surrounding electrolyte, as described by Lippmann equation [52]:

$$\gamma(V) = \gamma_o - \frac{C}{2} (V - V_o)^2$$  \hspace{1cm} (3.1)

Where $\gamma$ is surface tension, $V$ is the potential difference across the electric double layer (EDL) formed at the interface of droplet and electrolyte, $V_o$ is the potential of zero charge, $\gamma_o$ is the maximum surface tension at $V_o$, and $C$ is the capacitance of EDL per unit area. However, the unequal voltage drops along the electrolyte
and droplet (due to their massively different electrical conductivities) leads to unequal distribution of $\gamma(V)$ across the EDL, causing a surface tension gradient along the two hemispheres of droplet. This mechanism, which is known as ‘continuous electrowetting’ drives the surrounding electrolyte from the low surface tension hemisphere of the droplet toward its high surface tension hemisphere [51], and leads to the formation of vortices inside the enclosed liquid chamber. It should be noted that ‘electrowetting’ and ‘electrowetting on dielectric’ mechanisms also take advantage of electrical energy for movement of liquids in microfluidic systems. However, these two mechanisms both rely on the surface tension gradient at the interface of liquid-solid [67], whereas the ‘continuous electrowetting’ mechanism used here relies on the surface tension gradient at the interface of liquid-liquid (liquid metal droplet-electrolyte), eliminating the need for patterning metallic electrodes to apply electrical energy. More importantly, these two mechanisms are only effective for movement of discrete liquid volumes (droplets) [67], whereas ‘continuous electrowetting’ mechanism enables the continuous movement of liquid, as implied by its name, and hence is more useful for creating a flow inside a miniaturised structure.

The actuation of the pumps in opposite directions leads to the formation of one vortex inside the chamber, which is referred to as ‘one vortex’ case. In contrast, the actuation of the pumps in similar directions leads to the formation of two vortices inside the chamber, which is referred to as ‘two vortex’ case. Vortices are visualised by adding 250 $\mu$m glitter flakes into the liquid chamber (Figure 3.1c). The actual experimental setup is presented in Figure 3.2.
Figure 3.1: Liquid metal enabled vortex generator: (a) Schematic of experimental setup comprising of a liquid chamber, two liquid metal pumps with associated electrodes, which are energised by a signal generator, a resistor serving as the hot spot, an infrared camera and a laptop to monitor the temperature of the liquid chamber, (b) Close-up view of liquid chamber, (c) Actuation of Galinstan droplets with a square wave signal induces surface tension driven flow at the surface of droplets, leading to the formation of customised vortices inside the liquid chamber, visualised by the addition of glitter flakes in the liquid. Reproduced from Lab on a Chip, 2017, 17, 3862-3873 with permission from The Royal Society of Chemistry.
Figure 3.2: Mechanical clamp fabricated for holding the heat source inside the liquid chamber.
3.4 Results and Discussions

3.4.1 Generation of vortices

In contrast to previous works using liquid metal pumps [51, 64, 65], here the direction of induced flow is not directly in-line with the direction of voltage drop between the electrodes. Observations reveal that this interesting phenomenon relies on the deformation of spherical liquid metal droplet, resulting in the intrusion of droplet into the liquid chamber upon injection to the droplet seat (Figure 3.3). The deformed shape of droplet is maintained after energising the electrodes. This in turn increases the effective surface area of the droplet, which is in contact with the liquid inside the chamber, enabling strong surface tension driven flows, and consequently vortices inside the enclosed chamber.

![Figure 3.3: Intrusion of liquid metal droplet into the liquid chamber shown (a-b) before after applying voltage, and (c) in isometric view.](image)
In order to visualise the flow patterns at the vicinity of droplets, 100 μm colourant pigments are added into the liquid chamber, and tracked them using an upright USB microscope. The results clearly show the existence of two vortices, with a large vortex recirculating inside the liquid chamber, and a small vortex recirculating above the droplet (Figure 3.4). Interestingly, the two vortices interfaced along the bulged edge of droplet intruding into the liquid chamber.

Figure 3.4: Flow visualisation using 100 μm colourant pigments added into the liquid chamber, showing the existence of a large vortex inside the liquid chamber and a small vortex above the liquid metal droplet.
3.4.2 Theoretical and numerical modelling of vortices

A simplified theoretical model is developed to describe the pumping performance of droplets, as given below and schematically presented in Figure 3.5a:

\[
\frac{\partial y}{\partial t} \cdot A_{\text{droplet}} = \tau_{\text{NaOH}} \cdot A_{\text{NaOH}} = \mu_{\text{NaOH}} \frac{\partial U_{\text{NaOH}}}{\partial r} \cdot 2\pi R'_{\text{chamber}} H_{\text{NaOH}} \quad (3.2)
\]

where \( \gamma \) is surface tension, \( \vec{t} \) is the tangential vector along the surface of droplet, \( A_{\text{droplet}} \) is the effective surface area of droplet, \( \tau_{\text{NaOH}} \) is the time averaged shear stress induced at the surface of droplet, \( \mu \) is the viscosity of the liquid, \( U_{\text{NaOH}} \) is the rotational velocity of NaOH solution, \( r \) is location along the radial axis of the chamber, \( R'_{\text{chamber}} \) is the reduced radius of the chamber due to intrusion of the liquid metal droplet, and \( H_{\text{NaOH}} \) is the height of NaOH solution inside the chamber. Assuming that the system is operating in one vortex mode, \( U_{\text{NaOH}} = r\omega_{\text{NaOH}} \) (\( \omega_{\text{NaOH}} \) is the rotational velocity of liquid), and hence \( \frac{\partial U_{\text{NaOH}}}{\partial r} = \omega_{\text{NaOH}} \), using which the pumping performance of droplets is simplified as follows.

\[
\frac{\partial y}{\partial t} \cdot A_{\text{droplet}} = \mu_{\text{NaOH}} \omega_{\text{NaOH}} \cdot 2\pi R'_{\text{chamber}} H_{\text{NaOH}} \quad (3.3)
\]

**Figure 3.5:** Flow characterisation: Numerical simulations predicting the formation of two vortices which interface along the bulged edge of droplet.
Numerical simulations also predict the formation of large and small vortices at the vicinity of each droplet (Figure 3.6a). The results are obtained by solving Navier-Stokes equations using ANSYS-Fluent software. The boundary conditions include a surface tension gradient at the surface of liquid metal droplet, zero shear stress at the top free surface of the chamber, and no-slip at the other surfaces. The simulations are conducted by applying a surface tension gradient across the surface of liquid metal droplets, as detailed in Figure 3.6b-c.

**Figure 3.6:** Analysis of flow patterns using computational fluid dynamics (CFD) method: (a) Schematics of liquid metal enabled vortex generator used for developing a simplified theoretical model, (b) The simulations clearly predict the formation of a large vortex inside the liquid chamber and two small vortices above the liquid metal droplets, (c) Velocity profiles along the radial axis of liquid chamber at the heights of 3 mm. Reproduced from *Lab on a Chip*, 2017, **17**, 3862-3873 with permission from *The Royal Society of Chemistry*. 
3.4.3 Characterisation of vortices against various operating parameters

Further experiments are conducted to understand the role of droplet intrusion on the pumping performance of droplets. In doing so, the size of the orifice, interconnecting the electrode channel to the liquid chamber, is varied from 1.5 to 3.3 mm (Figure 3.7a-c), and operated the pumps under the reference conditions introduced in Figure 3.1. Experiments reveal three different vortex patterns according to the width of the orifice. The small orifice leads to a local vortex close to the droplets (Figure 3.7a'), the reference orifice leads to a large vortex encompassing the entire liquid chamber (Figure 3.7b'), and finally the large orifice leads to complete intrusion of one of the droplets into the liquid chamber and formation of an assymetric vortex by the other liquid metal droplet (Figure 3.7c').

Applying voltage to stainless steel electrodes also leads to generation of hydrogen bubbles at their surface due to electrolysis of NaOH solution. The majority of the bubbles rise to the surface of liquid and burst next to electrodes but small bubbles can recirculate in the electrode channel before merging and bursting. Electrolysis also leads to gradual dissolving of electrodes. Experiments indicate that stainless steel electrodes can be used for up to 5 hours (corresponding to 20 sets of experiments with an average duration of 15 minutes) without any significant change observed in the pumping performance of droplets.

Reducing the length of the electrode channel increases the current flowing through the channel, and therefore increases the rotational velocity of vortices. However, the electrode channel should be long enough to ensure the majority of hydrogen bubbles generated at the surface of electrodes can burst without entering the liquid chamber.
Figure 3.7: Flow characterization for different size of liquid chamber orifice widths, (a-c) Liquid chambers with small, reference and large orifice widths. (a’-c’) Characterisation of vortices in response to small, reference and large orifices interconnecting electrode channel to liquid chamber. Reproduced from *Lab on a Chip*, 2017, 17, 3862-3873 with permission from *The Royal Society of Chemistry*. 
To investigate the role of liquid chamber diameter on the pumping performance of droplets, the size of the chamber diameter is varied from 6 to 24 mm (Figure 3.8a-c), and measure the rotational velocity of the vortex under reference conditions. Experiments indicate that the vortex rotational velocity varies with respect to chamber diameter, which can be expressed as $\omega_{NaOH} = 399 D_{\text{chamber}}^{-0.8}$ (Figure 3.8d), which is close enough to the simplified model introduced in equation (3.2), which predicts $\omega_{NaOH} \propto D_{\text{chamber}}^{-1}$.

Figure 3.8: Flow characterisation against the size of liquid chamber, (a-c) Liquid chambers with small, reference and large diameters. (d) Liquid metal chamber diameter with the red dashed line corresponding to the curve fitted into the experimental graph. Reproduced from Lab on a Chip, 2017, 17, 3862-3873 with permission from The Royal Society of Chemistry.
On the other hand, generation of surface tension gradient across the liquid metal droplet is facilitated by continuous electrowetting mechanism, which in turn is governed by the pH and molarity of electrolyte as well as the frequency and magnitude of energising square wave signal [51]. Experiments indicate that the pH of electrolyte should be higher than 6.5 to induce sufficient continuous electrowetting at the surface of liquid metal droplet, as previously reported [51]. The molarity of electrolyte determines the concentration of ions, and hence the amount of charges across the EDL formed at surface of droplet, which in turn affects the pumping performance of liquid metal droplets. Experiments show that the molarity of NaOH should be kept higher than 0.15 to produce consistent vortices. Increasing the molarity from 0.15 to 0.3 increases the rotational velocity of the liquid almost linearly. Further increase of molarity does not increase the rotational velocity significantly. This can be due to saturation of charges across the EDL [51].
Furthermore, experiments indicate that the highest rotational velocity of liquid is obtained at a frequency of 200 Hz (Figure 3.9a). This is because the generation of surface tension gradient across the droplet relies on the continuous redistribution of ions across the droplet surface. However, at low frequencies the slow redistribution of ions leads to oxidation of droplet, whereas at high frequencies the ions do not have enough time to be redistributed. The frequency of 200 Hz is optimum, as it balances both conditions. It should be noted that the optimum frequency of droplets is inversely proportional to their diameter [51].

Accordingly, Prandtl number is defined as $Pr = \frac{k}{\rho C_p}$, in which $k$ and $C_p$ are the conduction coefficient and heat specific capacity of the liquid, respectively. Under these conditions, the temperature profile of the liquid is dominated by a hot ring formed along the sidewalls and a cold region inside the middle of the chamber. The rotational velocity of the vortex can be modulated by the frequency of the square wave signal, as detailed in Figure 3.9a, which in turn enables to change the hot spot temperature (Figure 3.9b).
Figure 3.9: Characterisation of vortices against various operating parameters. (a) Vortex rotational velocity against pump actuation frequency, (b) Maximum temperature against pump actuation frequency. Reproduced from *Lab on a Chip*, 2017, **17**, 3862-3873 with permission from *The Royal Society of Chemistry*.
3.4.4 Creating customised spatial temperature gradients

Proof-of-concept experiments are conducted to demonstrate the capability of the liquid metal enabled vortex generator in creating customised spatial temperature gradients inside the liquid chamber (Figure 3.10). Experiments are conducted in three operating conditions, designated as zero, one and two vortex cases. The temperature contours are captured 5 minutes after the activation of the hot spot. The temperature across the free surface of the chamber varies from 22 to 65 °C. However, the temperature contours shown in Figure 3.10 are limited to 44 °C for better visualisation of temperature profiles.

For the case of zero vortex (liquid metal pumps are not actuated), diffusion is the dominant mode of heat transfer inside the liquid chamber, leading to a temperature gradient of 40.1 °C along the radial axis of the chamber (Figure 3.10a). The liquid behaves as a heat sink, with the heat being dissipated through the liquid via diffusion and being lost through the top free surface of the liquid chamber via free convection (Figure 3.10a’).

For the case of one vortex (formed by actuating liquid metal droplets in opposite directions) heat is dissipated by both convection and diffusion mechanisms (Figure 3.10b). Convection becomes the dominant mode of heat transfer along the side walls of the chamber (vortex edge), at which the rotational velocity induced by the vortex is maximum. In contrast, diffusion becomes the dominant mode of heat transfer inside the middle regions of the chamber (vortex eye), at which the rotational velocity is minimum (Figure 3.10b’). The ratio between the convective and diffusive heat transfer mechanisms can be described by Péclet number, defined as Pe = Re Pr, in which Re is the Reynolds number of the flow, and Pr is the Prandtl number of the liquid. The Reynolds number is defined as
Re = \rho UD_{\text{chamber}} / \mu, in which \rho and \mu are the density and viscosity of liquid, accordingly, \( D_{\text{chamber}} \) is the diameter of the chamber, and \( U \) is the rotational velocity of the liquid, as presented in Figure 3.5a.

Figure 3.10: Versatility of liquid metal enabled vortex generator for creating customised spatial temperature gradients: (a-c) Temperature distribution across the top free surface of the liquid chamber for the cases of zero, one and two vortex obtained by infrared camera, and (a’-c’) Equivalent heat transfer diagram for the cases of zero, one and two vortex, showing the formation of hot rings at the edge of vortices where convection becomes dominant. Reproduced from Lab on a Chip, 2017, 17, 3862-3873 with permission from The Royal Society of Chemistry.
Similarly, for the case of two vortex (formed by actuating liquid metal droplets in the same direction), heat is dissipated by both convection and diffusion mechanisms (**Figure 3.10c**). Convection becomes dominant along the edges of vortices whereas diffusion becomes dominant in the eyes of the two vorticities. This creates two hot rings within the chamber with cold regions at each centre, forming an anchor-shaped temperature profile (**Figure 3.10c'**). Actuation of the two pumps with unequal frequencies leads to formation of asymmetric vortices inside the chamber, as presented in **Figure 3.11**.
Figure 3.11: Temperature distribution across the top surface of liquid chamber when operating in two vortex mode while applying different frequencies to the liquid metal pumps.
3.4.5 Analysing the transient response of the vortex generator

Next, the transient response of the vortex generator is investigated by monitoring the temperature contours of the liquid over a period of 60 minutes. The amount of liquid that evaporates via the top surface of the chamber was less than 5 %, and does not affect the overall thermal response of the system. Experiments are conducted under three operating conditions designated as zero, one and two vortex cases. Figure 3.12a-c present temperature contours captured by the infrared camera, 1, 5 and 20 minutes after the activation of the hot spot. Using these contours, the variations of average temperature ($T_{\text{average}}$), temperature standard deviation ($T_{\text{stdev}}$), maximum temperature gradient ($\Delta T_{\text{max}} = T_{\text{hot}} - T_{\text{cold}}$), as presented in Figure 3.10, and hot temperature ($T_{\text{hot}}$) are calculated.

For the case of zero vortex, measurements reveal the gradual increase of $T_{\text{average}}$ over the first 30 minutes, after which it remains almost constant as the system reached its steady state conditions (Figure 3.12d). Similar trends are observed for the cases of one and two vortex, indicating that the vortices do not impact the average temperature of the liquid.

In contrast, measurements show that the existense of vortices can significantly reduce the $T_{\text{stdev}}$ value (Figure 3.12e). For the case of zero vortex, $T_{\text{stdev}}$ varies between 12-14.4 °C over a period of 60 minutes, whereas for the case of one vortex $T_{\text{stdev}}$ varies between 4.3-5.6 °C over the same period, almost one third of the values obtained for the case of zero vortex. Similar trends are observed for the case of two vortex. In contrast to diffusive heat transfer, which relies on a temperature gradient between the hot and cold regions, the convective heat transfer relies on flow velocity, and therefore can occur with a lower temperature gradient. This in turn reduces the value of $T_{\text{stdev}}$ value.
This pattern can be clearly observed below (Figure 3.12f) which presents the variations of $\Delta T_{\text{max}}$ over time. For the case of zero vortex, $\Delta T_{\text{max}}$ is about 39.8 °C whilst for the cases of one and two vortex, $\Delta T_{\text{max}}$ is obtained as 30.2 and 30.4 °C, respectively, which are at least 9.4 °C less than the case of zero vortex. Interestingly, $\Delta T_{\text{max}}$ remains almost constant for all three cases, as the magnitude of diffusive heat transfer remains almost constant for each case. Despite this, for the cases of one and two vortex, the temperature of the cold region ($T_{\text{cold}}$) gradually increases over the first 30 minutes (Figure 3.12b-c), and therefore $T_{\text{hot}}$ should gradually increase over this period to maintain the magnitude of $\Delta T_{\text{max}}$, as shown (Figure 3.12g). The difference between the $T_{\text{max}}$ values across the cases of zero and one vortex is reduced, from 12.7 °C after 5 seconds to 2.5 °C after 30 minutes. This creates opportunities for transient cooling of hot spots, as further discussed in Figure 3.13.
Figure 3.12: Transient response of liquid metal enabled vortex generator: (a-c) Temperature distribution across the top free surface of the liquid chamber at 1, 5 and 20 minutes for the cases of zero, one and two vortex, (d-g) Temporal variations of average temperature ($T_{average}$), temperature standard deviation ($T_{stdev}$), maximum temperature gradient ($\Delta T_{max}$), and maximum temperature ($T_{hot}$). Reproduced from *Lab on a Chip*, 2017, **17**, 3862-3873 with permission from *The Royal Society of Chemistry*. 
3.4.6 Creating customised spatio temporal temperature gradients

Next, the capability of the liquid metal enabled vortex generator for creating customised spatiotemporal temperature gradients is investigated (Figure 3.13). In doing so, the vortex generator is switched between the one and zero vortex modes every 15 s (Figure 3.13a), and monitor the temperature contours of the liquid chamber for a period of 30 minutes (Figure 3.13b-c). Experiments indicate that the vortices and the temperature profiles can be both regenerated in consecutive cycles.

Experiments also indicate that the amplitude of temperature oscillations is different across the liquid chamber, and is determined by the difference between the initial temperature profiles of sequential vortex modes. For instance, in the case of the temperature cycles corresponding to zero and one vortex modes, the amplitude of temperature oscillations across the liquid chamber is calculated as:

$$\Delta T = |(T_{\text{one vortex}})_{t=0 s} - (T_{\text{zero vortex}})_{t=15 s}|$$

as presented in Figure 3.13d. This contour reveals that the region close to the hot spot experiences the highest temperature oscillations, whereas the regions close to the middle of the chamber experience the lowest temperature oscillations.

This trend is quantified in Figure 3.13e, which presents the temperature oscillations of five points in consecutive cycles, located at the hot spot upstream, hot spot downstream, vortex centre, sidewalls and opposite the hot spot, designated as $T_{\text{hot-}}$, $T_{\text{hot+}}$, $T_{\text{centre}}$, $T_{\text{sidewall}}$, and $T_{\text{opposite}}$, respectively, as described below.
For the first part of the cycle, operated at one vortex mode, $T_{hot-}$ decreased from 62.9 to 36.5 °C within 5 s (shaded in green) and then increased to 38.8 °C within the next 10 s (shaded in pink), as shown in Figure 3.13e. This correlates to the responses observed in Figure 3.12g, showing the gradual increase of temperature even in the presence of vortices. For the second part of the cycle, operated at zero vortex mode, $T_{hot-}$ increased to 62.9 °C within 15 s (shaded in yellow) before going through the next cycle. Repeating this cycle leads to similar transient patterns with a maximum difference of ±1.1 °C observed between various cycles. $T_{hot+}$, $T_{centre}$, $T_{sidewall}$, and $T_{opposite}$ went through similar transient patterns (Figure 3.13e) but experienced less temperature oscillations compared to $T_{hot-}$, as predicted in Figure 3.13d.

Interestingly, the temperature oscillations of these points are not necessarily in phase with the temperature oscillations of the hot spot, with phase shifts of 180° obtained for $T_{centre}$, $T_{sidewall}$, and $T_{opposite}$, as displayed by dashed lines in Figure 3.13e.
Figure 3.13: Versatility of liquid metal enabled vortex generator for creating customised spatiotemporal temperature gradients: (a) Switching between one and zero vortex modes by setting the duration of each mode to 15 s, (b-c) Temperature distribution at the top free surface of the liquid chamber immediately after switching from one to zero vortex modes, (d) Amplitude of temperature oscillations across the liquid chamber when switching between one-zero vortex modes, calculated as:

$$\Delta T = |\langle T_{\text{one vortex}} \rangle_{t=0\ s} - \langle T_{\text{zero vortex}} \rangle_{t=15\ s}|. $$

(e) Temperature oscillations of the regions located at the hot spot upstream, hot spot downstream, vortex centre, sidewalls and opposite the hot spot. Green and pink regions correspond to the first and second parts of the one vortex mode whilst the yellow region corresponds to the zero vortex mode. Dashed lines display the phase shift of various regions compared against the hot spot. Reproduced from *Lab on a Chip*, 2017, 17, 3862-3873 with permission from *The Royal Society of Chemistry*.
Similar trends are obtained by creating temperature cycles, through the switching between two and zero vortex modes, as presented in Figure 3.14.

**Figure 3.14:** Creation of customised spatiotemporal temperature gradients: (a) Consequential switching between two and zero vortex modes by setting the duration of each mode to 15 s, (b-c) Temperature distribution at the top free surface of the liquid chamber immediately after switching to one and zero vortex modes, (d) Amplitude of temperature oscillations across the liquid chamber when switching between one-zero vortex modes, calculated as: $\Delta T = |\langle T_{\text{one vortex}} \rangle_{t=0\,s} - \langle T_{\text{zero vortex}} \rangle_{t=15\,s}|$, (e) Temperature oscillations of the regions located at the hot spot upstream, hot spot downstream, vortex centre, sidewalls and opposite the hot spot.
Further experiments are conducted to demonstrate the versatility of the liquid metal vortex generator for producing customised spatiotemporal temperature gradients. In doing so, the vortex generator is operated in sequential one, two and zero vortex modes (Figure 3.15a). Three sets of experiments are conducted, in which the duration of each mode ($\Delta t$) was set to 5, 10 and 15 s.

Figures 3.15b-d present sequential temperature contours obtained at one, two and zero vortex modes at $\Delta t=5$ s. Accordingly, Figures 3.15b'-d' illustrate the amplitude of temperature oscillations across the liquid chamber when switching between sequential modes. For example, the amplitude of temperature oscillations corresponding to switching between the one and two vortex modes is calculated as $\Delta T = |\langle T_{one\ vortex}\rangle_{t=0\ s} - \langle T_{two\ vortex}\rangle_{t=5\ s}|$.

Figures 3.15e-g present the oscillations of hot spot temperature at $\Delta t=5$, 10 and 15 s, in which the presence of one, two and zero vortex modes is differentiated using green, pink and yellow shades, respectively. For the case of $\Delta t=5$ s, operating at one vortex mode decreases the hot spot temperature from 66.3 to 54.5 °C (shaded in green). This is followed by successive operation at two vortex mode, which increases the hot spot temperature to 58.2 °C (shaded in pink). Finally, operating at zero vortex mode further increases the hot spot temperature to 66.3 °C (shaded in yellow), as presented in Figure 3.15e.
Figure 3.15: Utility of liquid metal enabled vortex generator for creating more complex spatiotemporal temperature gradients: (a) Sequential switching of one, two and zero vortex modes, (b-d) Temperature distribution at the top free surface of the liquid chamber immediately after switching to one, two and zero vortex modes, (b'-d') Amplitude of temperature oscillations across the liquid chamber for transient conditions obtained by switching from one→two, two→zero and zero→one vortex modes when setting the duration of sequential vortex modes to Δt=5 s, (e-g) Temperature oscillations obtained for the hot spot region at Δt=5, 10 and 15 s, and (h) Variations of the amplitude of temperature oscillations for the hot spot (ΔT\textsubscript{hot}) against Δt. Reproduced from Lab on a Chip, 2017, 17, 3862-3873 with permission from The Royal Society of Chemistry.
The transient response of the system at $\Delta t=10$ and 15 s are given in Figures 3.16 and 3.17. Similar trends are obtained at $\Delta t=10$ and 15 s (Figures 3.16f-g and Figures 3.17f-g) with the following differences: (i) the duration of temperature cycles increased to 30 and 45 s, as expected, (ii) extension of $\Delta t$ changes the pattern of transient temperature variations and leads to formation of additional local peaks and valleys along the curve, and (iii) the amplitude of temperature oscillations ($\Delta T_{hot}$) varies with $\Delta t$ (Figure 3.16h and Figure 3.17h).

These experiments clearly demonstrate the ability of the liquid metal enabled vortex generator for producing customised spatiotemporal temperature oscillations across the liquid chamber. Modifying the order or duration of sequential vortex modes has the potential to produce a diverse range of spatiotemporal temperature oscillations.
Figure 3.16: Creating more complex spatiotemporal temperature gradients for the case of $\Delta t=10$ s: (a) Consequential switching between one, two and zero vortex modes, (b-d) Temperature distribution at the top free surface of the liquid chamber immediately after switching to one, two and zero vortex modes, (b'-d') Amplitude of temperature oscillations across the liquid chamber for transient conditions obtained by switching from one $\rightarrow$ two, two $\rightarrow$ zero and zero $\rightarrow$ one vortex modes when setting the duration of consequential vortex modes to $\Delta t=10$ s, (e-h) Temperature oscillations obtained for the hot spot located at hot spot, vortex centre, sidewalls and opposite to hot spot.
**Figure 3.17:** Creating more complex spatiotemporal temperature gradients for the case of $\Delta t=15$ s: (a) Consequental switching between one, two and zero vortex modes, (b-d) Temperature distribution at the top free surface of the liquid chamber immediately after switching to one, two and zero vortex modes, (b’-d’) Amplitude of temperature oscillations across the liquid chamber for transient conditions obtained by switching from one→two, two→zero and zero→one vortex modes when setting the duration of consequental vortex modes to $\Delta t=15$ s, (e-h) Temperature oscillations obtained for the hot spot located at hot spot, vortex centre, sidewalls and opposite to the hot spot.
3.5 Conclusion

In summary, this chapter demonstrated the utility of miniaturised liquid metal pumps for creating customised spatiotemporal temperature profiles within a small liquid chamber. Surface tension driven flow induced at the surface of liquid metal droplets led to formation of vortices inside the liquid chamber. The regions close to the edge of the vortex experience the highest rotational velocity at which forced convection becomes dominant. Alternatively, the regions close to the eye of the vortex experience the lowest rotational velocity at which conduction becomes dominant. This enabled us to create customised spatial temperature gradients by changing the configuration and rotational velocity of the vortices, which could be easily achieved by altering the frequency and polarity of the signal applied to the pumps.

In the absence of conventional moving elements in the liquid metal pumps, the configuration of vortices could be instantaneously changed. This enabled us to create customised spatiotemporal temperature gradients across the chamber by sequentially switching between zero and one vortex modes. More complex temperature oscillations were produced by switching between one, two and zero vortex modes.

The liquid metal enabled vortex generator can be easily integrated into enclosed structures to enable rapid and controllable temperature gradients within those devices. Covering the top surface of the liquid chamber with a thin glass slide minimises thermal diffusion through the top surface, and facilitates the projection of produced temperature gradients over its free surface. Target thermo-responsive materials can be immobilised onto the free surface of the
glass slide to investigate their response to customised spatiotemporal temperature gradients without the need for implementing multiple hot spots.

Customised temperature profiles can be produced based on the location of the hot spot, configuration and rotational velocity of vortices. More diverse vortex patterns can be produced by applying multiple liquid metal pumps or changing the configuration of the liquid chamber. More complex liquid metal structures can be implemented using various patterning techniques [68-71]. The process of energising liquid metal pumps can be automated to produce more complex temperature profiles and gradients.
3.6 References


CHAPTER 4

Conclusions and Future Work

4.1 Concluding Remarks

This PhD research demonstrated integrated liquid metal based liquid actuators for (i) localised convective cooling of hot spots, and (ii) creating customised spatiotemporal temperature profiles within a small liquid chamber. The outcomes of this research have been presented in Chapters 2 and 3, which are summarised in this chapter.
**Research Contribution 1:** As the first contribution, the author developed a liquid metal based convective cooling system for the localised cooling of hot spots. A small liquid metal droplet was located onto the hot spot. Applying a square wave signal induced sufficient surface tension gradient across the droplet, dragging the surrounding liquid due to continuous electrowetting mechanism. In this condition, the liquid metal droplet served as a ‘pump’. The flow rate of the coolant liquid could be easily tuned by varying the voltage and frequency of the applied square wave DC signal (Figure 2.3 and Figure 2.10). Due to the high thermal conductivity of the liquid metal droplet, it also served as a ‘heat sink’, facilitating the dissipation of heat into the surrounding liquid (Figure 2.7 and 2.8). The operation of this novel cooling system was studied under various operating conditions (Figure 2.10). This addresses my *Research Question 1*.

**Research Contribution 2:** As the second contribution, the author demonstrated the capability of the developed liquid metal based convective cooling system for the rapid cooling of hot spots. The elimination of external tubes and moving parts reduced the response time of the cooling system. Proof-of-concept indicated the capability of the system for reducing the temperature of the hot spot from 44.5 to 35.5 °C within 5.6 s (Figure 2.11). The response time of the system was inversely proportional to the flow rate of the coolant liquid, which could be easily adjusted by changing the frequency of the square wave signal. This addresses my *Research Question 2*. 

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**Research Contribution 3:** As the third contribution, the author developed a liquid metal enabled vortex generator for producing customised spatial temperature profiles within a small liquid chamber. The vortex generator consisted of a pair of liquid metal pumps. Energising the two pumps with square wave DC signals induced vortices inside the chamber (Figure 3.1 and 3.2). The configuration and rotational velocity of the vortices could be easily tuned by changing the polarity and frequency of the signal. In the presence of vortices, convective cooling became the dominant mode of heat transfer within the chamber, facilitating the generation of customised temperature profiles by operating the system in zero, one and two vortex modes (Figure 3.10). This addresses my Research Question 3.

**Research Contribution 4:** As the fourth contribution, the author demonstrated the capability of the liquid metal enabled vortex generator for producing temporal temperature gradients. The short response time of liquid metal pumps, as discussed in Contribution 2, facilitated the rapid switching between zero, one and two vortex modes, and eventually enabled switching from one temperature profile to another within a short amount of time. Temperature fluctuations of 11.8 °C and 14.5 °C were obtained by setting the switching time to 5 and 15 s, respectively (Figure 3.13 to 3.17). This addresses my Research Question 4.
4.2 Future work

The simple fabrication, integration and operation of liquid metal based convective cooling systems along with its short response time and versatility, as presented in the research, offers huge opportunities for further investigation in this field, with a few of them suggested here:

(i) Multiple liquid metal pumps can be integrated into high power density electronic circuits to facilitate the localised cooling of hot spots within such devices. Implementation of temperature sensors facilitates a fully automated cooling system.

(ii) Liquid metal based vortex generator can be integrated into secondary microfluidic devices to facilitate studying the response of temperature-sensitive materials or biological organisms to customised temperature gradients both spatially and temporally.

(iii) The sensitivity of the presented gallium-based liquid metal pumps to the pH value and electrical conductivity of the surrounding liquid limits its widespread application for cooling purposes. This suggests a comprehensive study on surface functionalisation of the current pumps with nano materials or alloying them with other metals to overcome such limitations.