

# Droplets 2019

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# Stability of Evaporating Droplets on Chemically Patterned Surfaces

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Multilayer inkjet printing is an emerging technology used for the fabrication of electric circuits, fuel cells or solar panels, among others. During printing, surface patches with different wettabilities are created onto which additional liquid is deposited in subsequent steps. This calls for a better understanding of droplets on surfaces with patterned wettability.

We have studied the stability of evaporating water droplets on chemically patterned surfaces with alternating hydrophilic and hydrophobic stripes based on experiments, numerical simulations and a scaling model.

Surface Evolver calculations show that when the contact angle contrast between the stripes is small, a droplet stays intact during evaporation, whereas it breaks up when a critical value of the contrast is exceeded. For increasing hydrophobic contact angles this value increases. These calculations are supported by experiments.

In the unstable regime, a H-shaped droplet is formed (see figure 1a below), with a liquid bridge sitting on the hydrophobic stripe separating two liquid fingers on the neighbouring hydrophilic stripes. Owing to evaporation, the width of the bridge decreases until it reaches a critical value (figure 1b). This is the point where the capillary surface becomes unstable, and the droplet breaks up into two disconnected liquid volumes (figure 1c). The breakup dynamics is mainly dominated by the wettability of the hydrophilic stripes. According to experiments, a better wettability leads to a faster breakup process.

The critical width of the capillary bridge increases for an increasing ratio of the hydrophilic and the hydrophobic stripe width. A simple scaling model based on the Young-Laplace equation is used to estimate the critical width: It is reached when the maximum achievable pressure inside the liquid fingers on the hydrophilic stripes is exceeded by the Laplace pressure inside the bridge.

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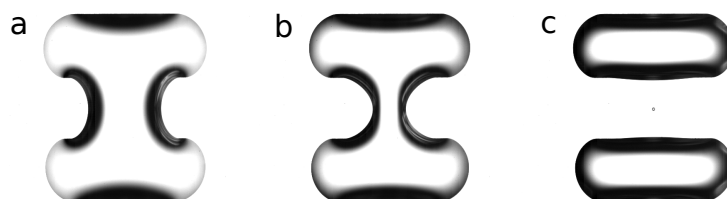


Figure 1: Evaporation behavior of a droplet sitting on a striped patterned surface

## SECONDARY BREAKUP OF LOW VISCOSITY DROPS IN A CONTINUOUS AIR JET

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Secondary breakup/drop breakup affects final spray characteristics of a spray system. Hence, its study has received much attention from researchers. Breakup of the drop depends on the combined action of the aerodynamic force on the drop, surface tension force and viscous force. The higher the surface tension and viscosity of the liquid, the lower is the tendency towards fragmentation [1]. In this study, we investigated the effect of viscosity on the breakup behaviour of the drop by choosing test liquids (water, glycerol (21 wt%) and glycerol (42 wt%)) such that only viscosity differed while the other two properties (density and surface tension) remained nearly the same.

The non-dimensional numbers which govern breakup behaviour of the drop are Weber number and Ohnesorge number: (1)  $We = \rho_g U^2 d_0 / \sigma$ , where  $\rho_g$  is the gas density,  $U$  is the gas velocity,  $d_0$  is the undeformed drop diameter,  $\sigma$  is the surface tension of liquid, (2)  $Oh = \mu_l / \sqrt{\rho_l d_0 \sigma}$ , where  $\mu_l$  is the liquid viscosity, and  $\rho_l$  is the liquid density. In this study, we essentially investigated the effect of a change in Ohnesorge number on the breakup behaviour of the drop.

The experiments were performed at three different Weber numbers for all the test liquids. We observed bag breakup, bag-stamen breakup and sheet-thinning breakup. The Weber numbers corresponding to each breakup mode for all the liquids were kept close to each other so that the effect of only Ohnesorge number on the breakup behaviour can be examined. We measured the following parameters: stream-wise ( $L$ ) and cross-stream ( $2R$ ) length of the drop, the time at which the first instance of breakup is observed, displacement, velocity and acceleration of the drop centroid, and a coefficient of drag ( $C_d$ ). Dissimilarities were observed in the breakup behaviour of different liquids, which will be discussed. Figure 1 below shows the breakup modes observed.

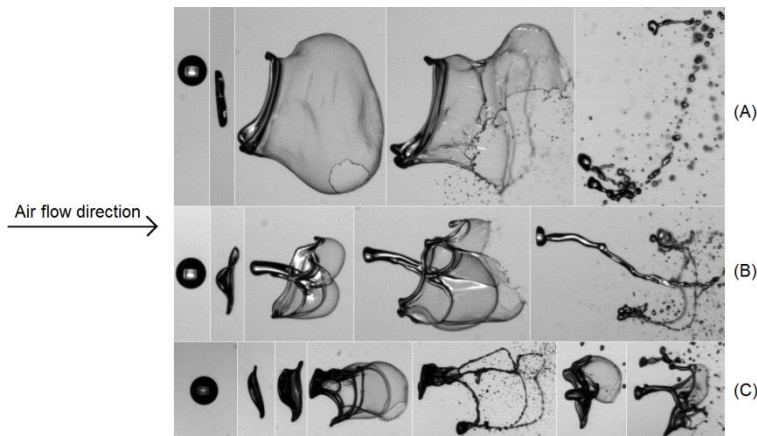


Figure 1: (A) Bag breakup ( $We = 12.7$ , water), (B) bag-stamen breakup ( $We = 22.6$ , glycerol (21 wt%)), (C) sheet-thinning breakup ( $We = 31.7$ , glycerol (42 wt%)).

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# **Numerical Simulations of Coalescence-Induced Droplets Jumping: Effects of Droplet Initial Velocity**

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Coalescence-induced droplet jumping phenomenon on the superhydrophobic surface has a wide range of applications such as hotspot cooling, surface self-cleaning, anti-icing and defrosting. In the process of condensation, it is more common for a moving droplet to impact into another stationary droplet at a certain initial velocity and then they jump from the surface. As for the applications, such as hotspot cooling, if we understand the dynamics of the situation, we can adjust the initial velocity of the moving droplets so that the droplets can jump to the specified hot spot position. Based on these, we used the volume of fluid (VOF) method to simulate the effects of the initial velocity of moving droplets on the jumping process at low Weber numbers ( $We < 1$ ). We analyzed the morphology, velocity and energy of the droplets in each case. Regardless of the initial velocity, the morphological changes of the droplets are similar. An increase in the initial velocity will accelerate this process but will not change the trend. Departure velocity is an important parameter of the whole process. As the initial velocity increases, the droplets will leave the surface at different stages of deformation, which causes the droplet's departure velocity appearing in two stages that the velocity is approximately constant first and then increases. At low Weber numbers, the energy conversion of the droplets is between 4% to 6%. Although the initial velocity is positively correlated with the energy conversion rate when the initial velocity is greater than 0.35m/s. The conversion rate of the whole process is still very low which showed as fig. 1.

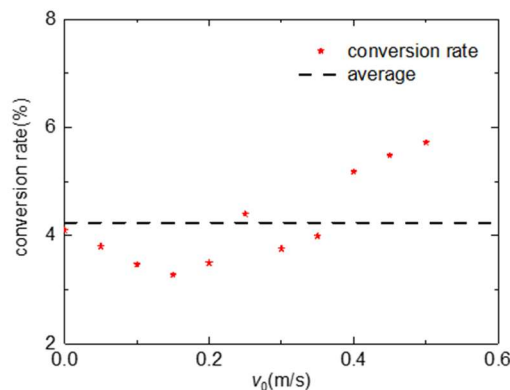


Fig. 1 Energy conversion rate for all cases

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# PHASE-FIELD INVESTIGATION ON THE FORMATION OF DROPLETS AS A RESULT OF SURFACE TENSION AND GRAVITY

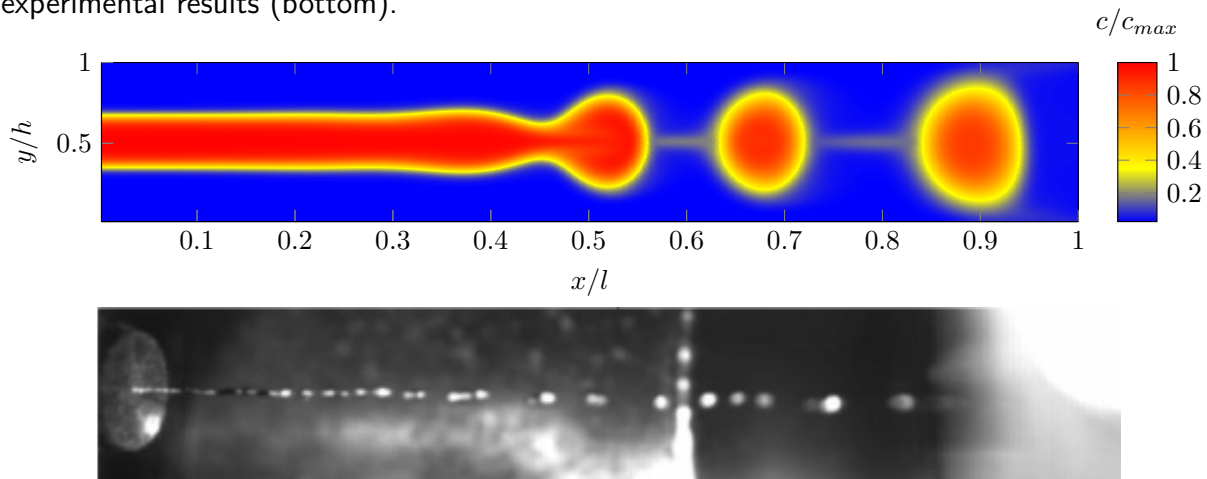
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The breakup of a liquid jet into separated droplets is widely observed in nature as well as in multiple technical applications and is therefore of great interest. Examples are crystal-growth [1], fuel atomization or the manufacturing of metal powders out of a jet of molten material. Plateau and Rayleigh [2, 3] established the fundamental theory of jet breakup considering small disturbances. The breakup occurs due to an instability of perturbations, which are amplified until the droplet finally separates. The droplet size and distance between the resulting droplets are determined by the wavelength of the perturbation with the fastest growth rate. However, Plateau-Rayleigh theory is limited to tiny displacements and does not consider the effect of gravity. Hence, the jet breakup is still subject of several experimental, numerical and theoretical investigations [4, 5]. The present study addresses the liquid jet breakup due to the coaction of surface tension and gravity via experiments and numerical simulations employing a phase field method of Cahn-Hilliard type in conjunction with a Navier-stokes solver. The following figure shows a slice of the computational domain depicting the liquid concentration (top) and experimental results (bottom).



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## WHAT DETERMINES THE DROP SIZE IN SPRAYS?

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### Abstract :

We have worked out the physics of an everyday phenomenon: the break-up of a spray of liquid into droplets. We systematically investigated the different parameters that determine the characteristic drop size in the breakup of sheets emerging from a spray nozzle: the pressure and geometry of the nozzle and the fluid parameters such as viscosity and surface tension. The combined results show that the drop size results from a competition between fluid inertia and surface tension which allows for the prediction of the drop size from the Weber number and the geometry of the nozzle. The drop size distribution is found to be described by a compound Gamma distribution with two parameters that are related to the polydispersity and roughness of the fluid ligaments that result from the breakup of the sheet.

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Kooij, S., Sijs, R., Denn, M. M., Villermaux, E., & Bonn, D. 'What Determines the Drop Size in Sprays?' **2018** *Physical Review X*, 8(3), 031019.



## MIXING WITHIN IMPACTING AND COALESCING DROPLETS OF DIFFERENT SURFACE TENSION

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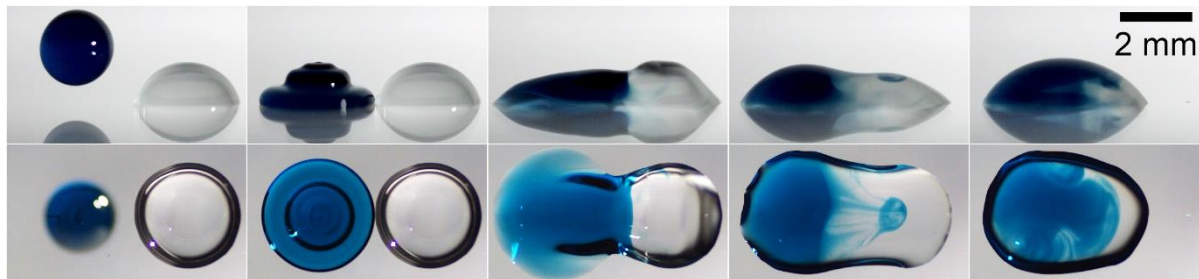
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Effective mixing between impacting and coalescing droplets of different fluids in contact with a solid substrate is a crucial requirement for emerging printing technologies. Whilst recent studies have observed surprisingly little mixing between impacting and coalescing droplets of the same fluid [1], surface tension gradients could play a significant role in enhancing mixing when the droplets consist of different fluids through the contribution of Marangoni flow. An additional factor affecting the internal dynamics is the substrate's wettability.

This work investigates the coalescence of a sessile and an impacting droplet of different fluids (several ethanol/water mixtures), with a range of surface tensions but similar densities and viscosities, in the surface tension dominant regime. Using two colour high-speed cameras, the internal dynamics and associated mixing are captured simultaneously from both the side (at up to 25,000 FPS) and below (at 7,200 FPS, through the substrate). This arrangement exposes both the detail of the flow through the depth of the droplet (side view) and the flow structures without distortion from the droplet's curved surface (bottom view). Either a pH indicator or an inert dye is used to visualise the internal dynamics, with multiple lateral separations studied.



Our results systematically elucidate the role of surface tension gradients and resulting Marangoni flow on the internal dynamics, with significantly faster mixing exhibited compared to droplets with the same fluid properties. Various fascinating physical mechanisms are uncovered, including a jet across the free surface of the originally sessile droplet, which can either be enhanced or suppressed by modifying the relative surface tension between the original droplets (see the image sequence, where the surface tensions of the impacting and sessile droplets are 50 mN/m and 58 mN/m, respectively). Not only do our results expose beautiful physical phenomena, but they also have practical implications regarding lateral separation and order of deposition for enhancing mixing within impacting and coalescing droplets of different surface tension.

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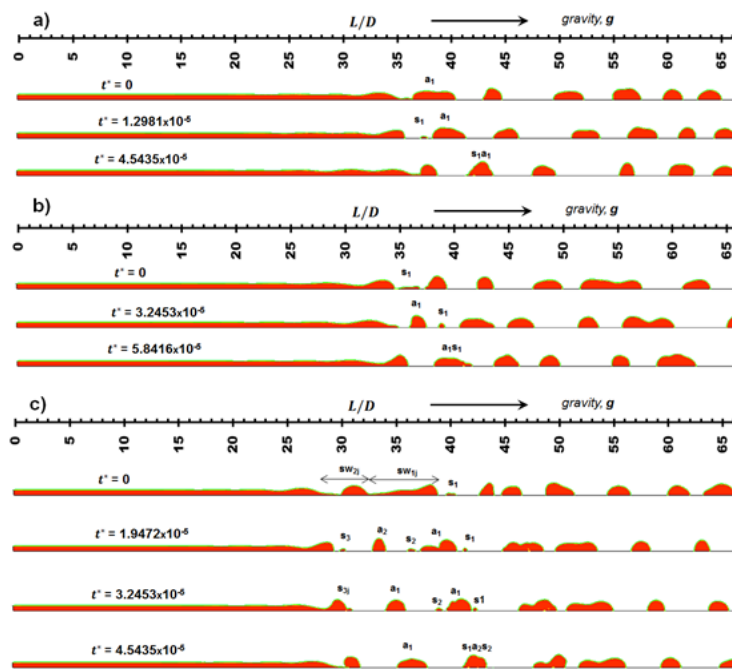
# FORMATION AND MERGING OF SATELLITE DROPLETS DISINTEGRATED FROM LAMINAR LIQUID JETS

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Numerical simulations of laminar liquid jets are presented that reveal the formation of satellite droplets resulting from (i) the non-linear disintegration of jet liquid core and (ii) coalescence of two primary droplets. For the case (i), when satellites disintegrate from the jet liquid core, they tend to show forward, rear and simultaneous merging responses on both the aft side as well as the foreside of the primary droplets that are present downstream that were evidenced experimentally by Vassallo and Ashgriz (1991). However, for the case (ii) the satellites that are formed show similarities with that observed by Zhang et. al (2009) for two stationary droplets at the onset of coalescence. However, our numerical results predict that when two primary drops disintegrated from a liquid jet, undergoing surface oscillations coalesce, satellites can pinch-off with effective diameter ratios of  $\frac{d_{large}}{d_{small}} \sim 1.15$ . Effect of capillary waves during primary droplet coalescence and the subsequent pinch-off dynamics for satellite formation is investigated in this study.



**Fig. 1.** Liquid volume fraction profiles showing a) forward merging, b) rear merging and c) simultaneous merging of satellite droplets emanated from a liquid jet with  $We=16.48$ .

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## Suppression of coffee-stain effect by local shielding of evaporation at drop edge

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A coffee drop drying on a table top leaves a ring-like stain, which is well-known as "coffee-stain effect"[1], i.e., when a drop is evaporating with a pinned contact line, suspended particles are carried by a capillary flow from interior towards the edge, and left concentrated along the original drop edge. The reason lies on that the maintaining of pinned contact line requires replenishing of fluids from the center, which is normally enhanced by the singularity of evaporation rate at the contact line. The ubiquitousness of the effect makes it paradigmatic for scientific research but in practice, difficult to avoid for achieving uniform coating. Within all the studies on the control of the deposit formality by drop evaporation, the approaches mainly focus on internal flow manipulation[2] and particle-solid interactions[3]. In particular, Marangoni effect in evaporative multicomponent systems, either solutal or thermal, is used quite often as an effective way to suppress coffee-stain effect by continuously mixing suspended particles during evaporation. However, recent experimental observations on the evaporation of multicomponent drops show complex physicochemical dynamics, including phase separation[4,5] or crystallization[6], triggered by selective evaporation, which may bring uncertainties into the systems: e.g., the suppression of evaporation[5] or the gravitational effect in flow field[7]. Here, we present a simple but efficient mechanism differing from conventional methods of coffee-stain effect avoidance: hindering the evaporation near the drop edge to suppress the radial capillary flow which supposes to replenish the liquid loss at the pinned contact line. We achieve a local shielding of volatile liquid near an evaporating drop edge by utilizing non-volatile meniscus arising from a primary lubrication film. It is observed that the blocking of local evaporation near the contact line, the radial capillary flow is reduced to only stochastic motions, and as a result, accomplishing a uniform coating without the coffee-stain effect.

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# HYDRODYNAMIC INSTABILITY OF AN EVAPORATING LIQUID LAYER IN A CYLINDRICAL POOL

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Motivated by a recent space experiment which consists of an evaporating liquid layer (FC-72,  $Pr=12.38$ ) contained in a shallow cylindrical pool ( $H/R=0.2$ ) under a bidirectional temperature difference, we numerically investigate the hydrodynamic instabilities to observe under which condition a potential transition from axisymmetric to a fully 3D flow pattern take place. The bidirectional temperature difference is due to a higher temperature imposed at both the bottom and sidewall of the cylindrical container while the temperature far from the liquid surface is assumed to be constant equal to ambient temperature. The evaporation cooling effect at the free surface air/liquid is considered through a varying boundary condition; in dimensionless form this equation is controlled by the Biot number ( $Bi$ ). Consequently, there will be both vertical and horizontal temperature differences inside the liquid layer. Under this bidirectional temperature difference, the effects of different  $Bi$  numbers and dynamic  $Bond$  ( $Bo$ ) numbers on the critical Marangoni number ( $Ma_c$ ) of the flow pattern transferring to fully 3D are studied. Numerical results show that when the Marangoni number is smaller than some critical  $Ma_c$ , it exhibits 2D axisymmetric flow pattern, while when the Marangoni number exceeds the critical condition, the flow field undergoes a transition to a 3D structure. We present results for varying  $Bo$ ,  $Bi$ ,  $Ma$ , and  $\Delta T$  corresponding to our ground and micro-g experiments and give the critical values of these parameters for the transition from 2D axisymmetric flows to full 3D ones with an emphasis on the evaporation effect.

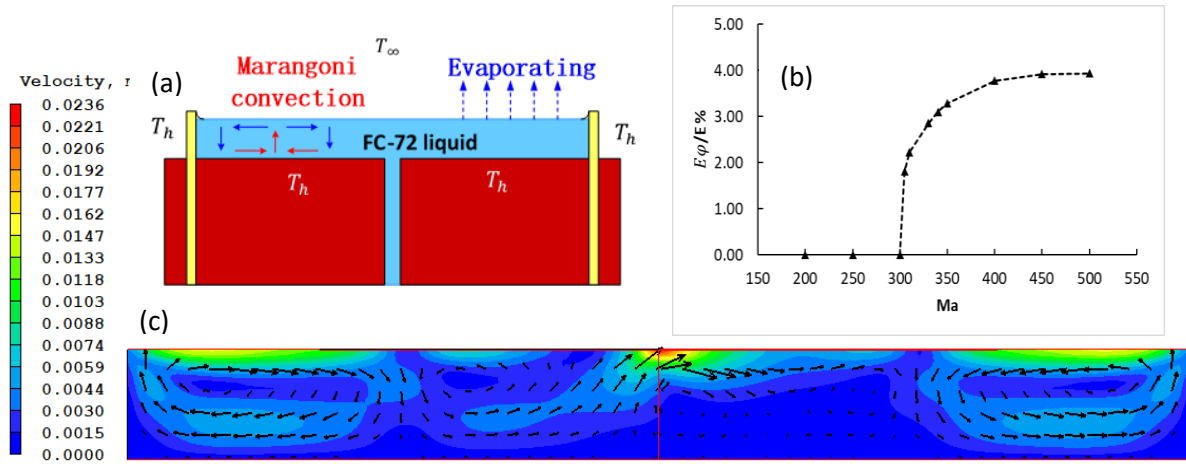


Figure: (a) A schematic of the experimental setup. (b) An example of identification of the transition to a fully 3D fluid flow by measuring the azimuthal kinetic energy compared to the total kinetic energy ( $E_\phi/E$ ) of the flow system; the ratio is plotted vs. the Marangoni number,  $Ma$ . (c) An example of the velocity field of 3D flow pattern at  $Ma=500$  in a  $(r, z)$  section.

ACKNOWLEDGEMENTS : This research was financially supported by the National Natural Science Foundation of China (Grants Nos. 11532015, U1738119), the China's Manned Space Program (TZ-1) and China Scholarship Council (CSC).

# “COFFEE-RING” WIDTH MODIFICATION DUE TO TEMPERATURE GRADIENT ON THE SUBSTRATE

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## Abstract

We experimentally investigate the coffee-ring deposits obtained after evaporation of a  $1.1\ \mu\text{l}$  water droplet containing different polystyrene colloidal particles ( $0.1$ ,  $1.1$ , and  $3.0\ \mu\text{m}$  diameter) with an imposed temperature gradient on the surface of the glass substrate. The temperature gradient was achieved by using two thermoelectric coolers underneath the substrate and was confirmed by the infrared thermal camera measurements. High-speed visualization was employed to record the time-varying droplet evaporation. The three substrate temperature gradients obtained were  $4\ ^\circ\text{C}/\text{mm}$ ,  $2.5\ ^\circ\text{C}/\text{mm}$ , and  $1.75\ ^\circ\text{C}/\text{mm}$ . The deposits are qualitatively visualized under an optical microscope and scanning electron microscope and are compared with the deposits obtained on constant temperature substrate. The measurements showed that there is a substantial difference in the ring width on the hot side ( $T_H$ ) ring perimeter than the cold side ( $T_L$ ) ring perimeter, as shown for the  $1.75\ ^\circ\text{C}/\text{mm}$  case in Figure 1. Due to the imposed temperature gradient, the hot side droplet is at lower surface tension than the cold side and the Marangoni convection loop changes inside the droplet into a single loop unlike the two loops on a constant temperature substrate. The fluid and the particles flow from the hot side to the cold side along the droplet surface.

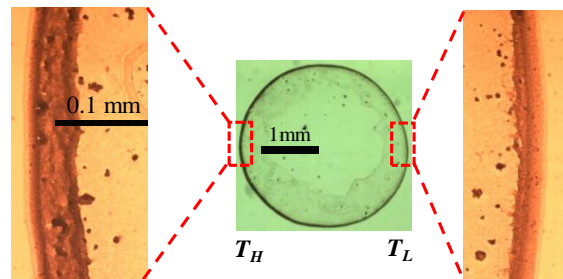


Figure 1. A typical ring-like deposit in the present measurement. The zoomed-in view rings are measured at hot side ( $T_H$ ) and cold side ( $T_L$ ).

## THE DRYING OF BLOOD

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The University of Edinburgh, Leverhulme Research Centre for Forensic Science

In order to pursue criminals as quickly and efficiently as possible, forensic analysts must glean a wealth of information from a limited set of evidence in a short amount of time. This study aims to expand the information acquired from a crime scene by increasing understanding of blood from its drying patterns.

Blood is a biological colloidal system, the drying of which has been examined by the forensic and soft matter communities through the use of anticoagulants. Anticoagulants such as EDTA and Tri-sodium Citrate extend the lifetime and transportability of the blood by preventing coagulation, making it easier to perform laboratory experiments. To expand the usefulness of this research to real-life crime scenes where coagulation naturally occurs, coagulation was reintroduced to the samples via the addition of calcium chloride in order to examine how the process affects a change in particle size, drying rate, and mass change. Image analysis of a drying timelapse, measurements of mass change over time, and drip patterns were used to examine how the gelation of a coagulated blood varies from an anticoagulated blood, and what underlying mechanisms differ between the two.

# Water droplets evaporation rates enhanced by acoustic field in a cylindrical resonator

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Motivated by the potential use of water droplet aerosol as a load or a source in two-phase thermoacoustic devices, [We](#) study the effect of an acoustic field on the evaporation rates of low-density water aerosol in a cylindrical acoustic resonator experimentally. Using a combination of 2D mass flow visualization and PIV techniques, evaporation rates were quantified under the influence of a low frequency (110 [Hz]) acoustic standing wave at various Acoustic Pressure Amplitudes [APA]. Water droplet aerosol evaporation and settling velocities were monitored between the resonator's pressure and velocity anti-nodes. In addition, acoustic intensities were measured during the experiments. The evaporation rate showed a significant increase with the application of the acoustic field in the resonator, largely dependent on the APA. The results revealed two distinct regimes of acoustically enhanced evaporation rate: linear for APA values in the range  $600[\text{Pa}] < \text{APA} < 1000[\text{Pa}]$ , and exponential in the range  $1160[\text{Pa}] < \text{APA} < 2000[\text{Pa}]$ . Such behavior may be explained through combined influence of fluctuations of the boundary layer surrounding each droplet induced by the pressure fluctuations, and the periodic fluctuations in droplets velocity. The latter is argued to be the acoustic streaming of water vapor away from the droplet surrounding, becoming significant at higher APA, enhancing droplets evaporation rates. Significant variations of the acoustic intensity values were detected during the experiments, despite the constant mechanical power provided by the loudspeaker. Such variations in acoustic intensity are shown to be a useful marker for measurements of droplets evaporation rate.

ACKNOWLEDGEMENTS: Supported by a grant from the "Dead Sea Works".

# **VISUALISATION OF THE VAPOUR CLOUD INDUCED BY EVAPORATION OF INTERACTING DROPLETS.**

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Multiple droplets evaporate in the communal environment of their neighbours resulting in a behaviour that is different to a single droplet. Whilst the evaporation of isolated droplets has been studied extensively, little is known about the evaporation dynamics and resulting deposits of multiple interacting droplets due to its inherent complexity. The presence of a close neighbouring droplet is expected to increase the vapour concentration between the droplets, thus reducing the local evaporation rate. This asymmetry in the vapour can give rise to alterations in the fluid flow [1] and asymmetry in the drying patterns [2, 3].

In this study, we use Mach-Zhender (MZ) interferometry to measure the vapour distribution and its dynamical behaviour around a pair of interacting pendant droplets of highly volatile liquids. The MZ interferometer enables the determination of the variations in refractive index induced by the vapour cloud [4]. By varying the droplet radius  $R$  and droplet separation  $L$ , the dependence of the asymmetry in refractive index around the droplets, hence inter-droplet interaction strength, on  $L/R$  is established. The dependence will also be compared to that obtained from evaporation rate measurements.

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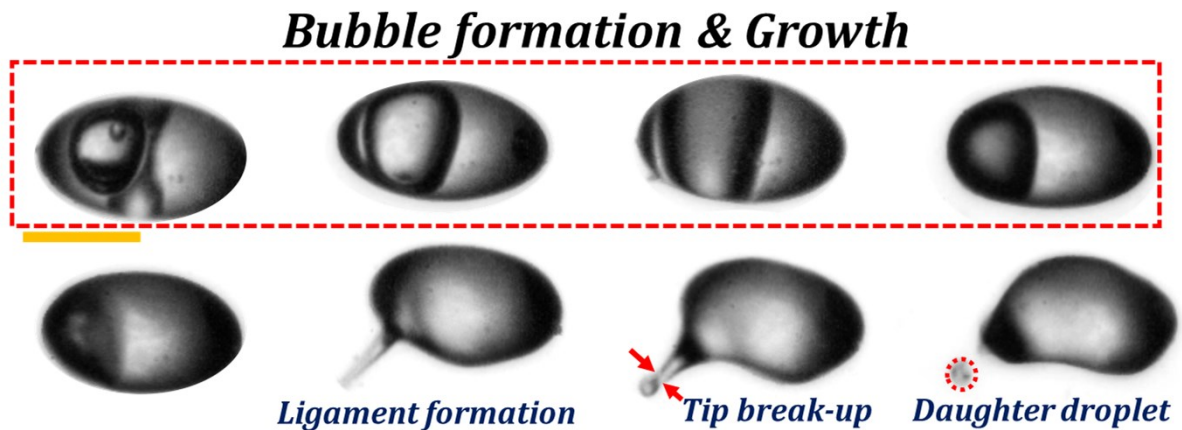
## BOILING IN NANOFUEL DROPLETS

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Inclusion of metal/metalloid particles in conventional hydrocarbon fuels has unearthed the possibilities of next generation fuels. Recent research stride on nanofuel (Base Fuel + Nanoparticles) combustion has shown a unique pathway of droplet secondary atomisation, stemming from heterogeneous boiling. Presence of nanoparticles (NPs) aids internal ebullition in nanofuel droplets. Rupture and expulsion of the bubbles generate high-speed ligaments which further undergo tip break-up. The resulting daughter droplets enhance the mass transfer rate. For instance, during combustion they act as a fuel parcels from droplet surface to the flame envelope.



In the current work, we report detailed analyses of evaporation and atomisation characteristics of nanofuel droplets in a contactless environment (acoustic levitation) under external radiative heating. We explore the critical parameters for bubble incipience by varying base fuel vapour pressure, initial droplet size, and the input laser power. A time scale analysis considering orthokinetic NP aggregation, evaporation lifetime, and bubble growth rate is presented to elucidate the mechanism of internal boiling. A theoretical non-dimensional time scale ( $\tau^*$ ) is coined to estimate the minimum value of the droplet size necessary for exhibiting boiling.

**ACKNOWLEDGEMENTS:** Authors are thankful to the Council of Scientific and Industrial Research (CSIR) and DST Swarnajayanti Fellowship for the financial support.

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## EFFECT OF NEGATIVE MAGNETOPHORESIS INSIDE AN EVAPORATING FERROFLUID DROPLET

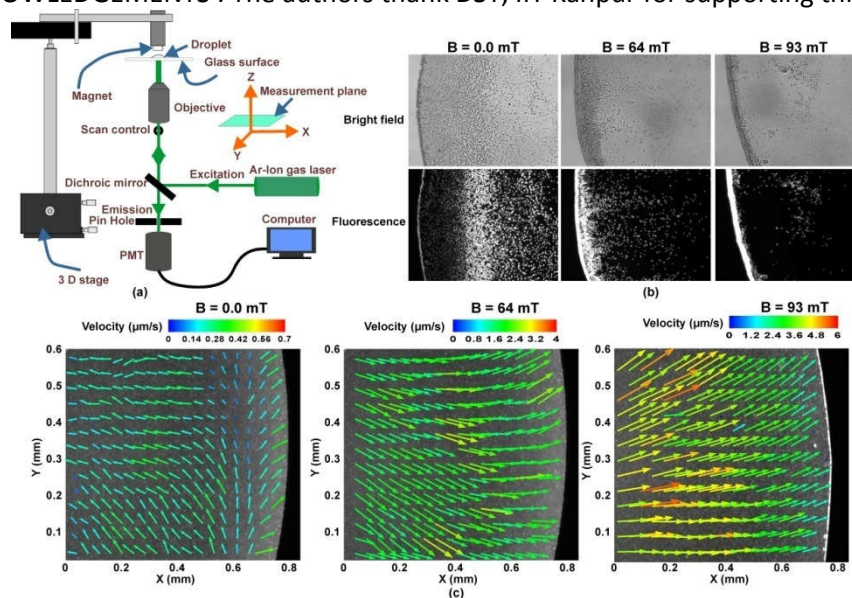
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Diamagnetic particles present inside a paramagnetic fluid experience a negative magnetic force due to mismatch in magnetic susceptibilities. Negative magnetophoresis is the migration of the particles away from the magnetic source. Effect of negative magnetophoresis on the fluorescent particles present inside an evaporating ferrofluid droplet on a glass substrate as a function of magnetic field strength has been studied. The confocal microscopy technique has been used for the visualisation of the particles motion and drying pattern of evaporating ferrofluid droplet. Both bright field and fluorescence technique have been conducted to show the deposition of diamagnetic particles and magnetic particles after complete drying of droplet. Micro-PIV technique has been used to measure the velocity of diamagnetic particles during evaporation under different magnetic field strength. The mixture of diamagnetic and magnetic nanoparticles are deposited at the contact line in the absence magnetic field. When the magnetic field is applied to the droplet during evaporation process, the magnetic particles migrate towards the higher magnetic field region (center of droplet) while diamagnetic particles drift towards the lower magnetic field region (contact line). The combined effect of magnetic force and drag force drive the diamagnetic particles towards the contact line region. The diamagnetic particles are deflected at a higher velocity with increase in the magnetic field strength.

ACKNOWLEDGEMENTS : The authors thank DST, IIT Kanpur for supporting this work.



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# THE EFFECT OF THE THERMAL CONDUCTIVITY OF THE SUBSTRATE ON THE LIFETIMES OF AN EVAPORATING DROPLET

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Determining the lifetime of an evaporating sessile droplet on a solid substrate is an important part of understanding many industrial processes, such as ink-jet printing, coating, and spray cooling, as well as drug delivery systems and chemical spill containment. Consequently, in recent years there has been a rapid growth of experimental and theoretical research into droplet evaporation. Previous authors [1] have shown that the instantaneous evaporation rate of a droplet depends on the thermal conductivity of the substrate. In the present work we use a combination of analytical and numerical methods to study the effect of the thermal conductivity of the substrate on the evolution, and hence the lifetime, of an evaporating droplet. In the limit of a thin droplet on a thin substrate we obtain analytical expressions for the lifetimes of droplets evaporating in various modes of evaporation [3]. In the general case of a non-thin droplet we use numerical methods to calculate the lifetimes of droplets evaporating in various modes of evaporation on substrates of various thermal conductivities. In general, droplets on less conductive substrates are found to have longer lifetimes than those on more conductive ones.

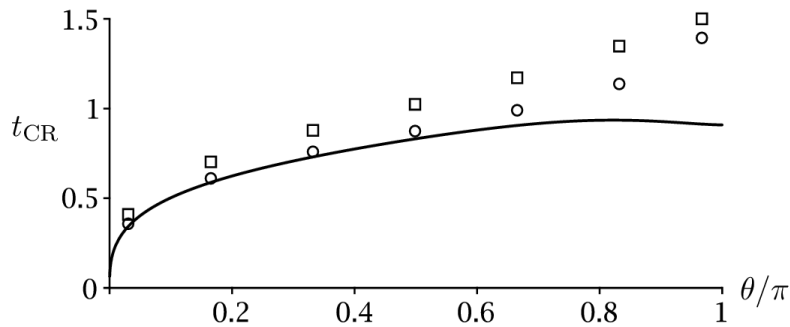


Figure 1: Scaled lifetimes of droplets evaporating in the constant radius (CR) mode  $t_{CR}$  as a function of the scaled initial contact angle  $\theta/\pi$ . The solid curve denotes the lifetime of a perfectly conducting droplet on a perfectly conducting substrate obtained in [2], while the circles and the squares are the present numerical results for a droplet of water on a poorly conducting substrate (PTFE) and on a highly conducting substrate (aluminium), respectively.

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## Digital holographic investigation of micro-liter well

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Current study deals with the evaporation of volatile liquids from the micro litter wells. Extensive studies have been carried out for studying the physics of evaporation of micro litter size droplets however few studies are available on evaporation from micro/nano litter wells. Evaporation of liquid from micro litter wells has applications in several engineering and medical diagnostic systems, solid state sensors, protein crystallization growth and micro DNA array handling etc. Main objective of current work is to study the distribution of vapour mole fraction field above an evaporating micro litter well. The results from this study can help us to understand the local evaporation rate distribution and gradients of vapour mole fraction in radial and normal directions. Gravimetric analysis is performed to measure the global evaporation rate with time. Digital holographic Interferometry is used to calculate the vapour mole fraction above the evaporating well. Phase unwrapping has been carried out using Goldstein algorithm. A well of 5 mm diameter and 2 mm depth is machined on Plexi glass and hexane is used as volatile liquid in the present study. The vapour cloud shows a plume like behaviour. The extent of vapour cloud extends to the three times of well size. This information can be very beneficial for the micro well design in DNA sequencing such that interaction between the neighbouring well can be eliminated leading to improved sensitivity.

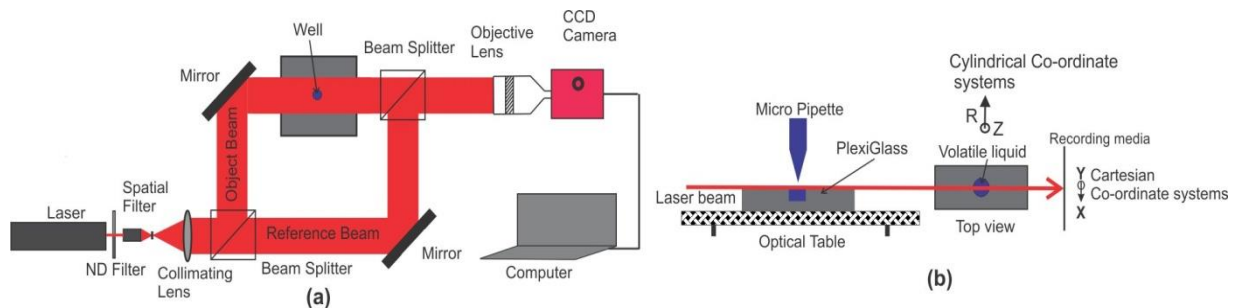


Figure 1(a) Schematic of DHI setup and (b) well arrangement

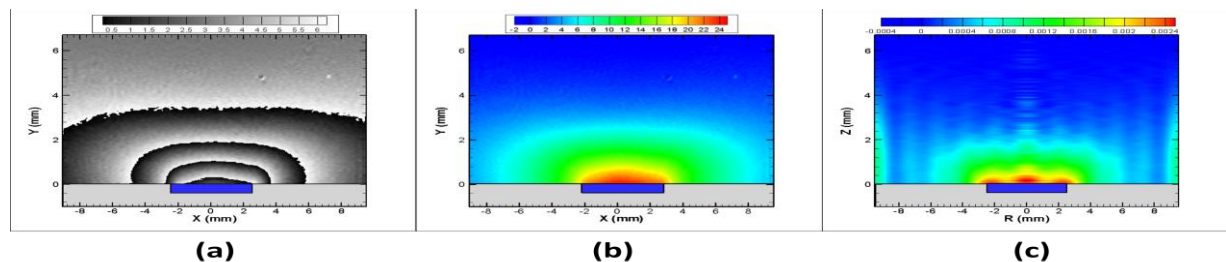


Figure 2 (a) Wrapped phase map for liquid evaporating from 5 mm diameter and 2 mm depth well at time  $t=0+$ , (b) unwrapped phase distribution and (c) vapour cloud above the evaporating well

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## TRANSMISSION OPTICAL IMAGING OF CONTRACTING SESSILE MICRODROPLETS

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Small sessile microdroplets are useful tools to study crystal nucleation, and benefit of microfluidic techniques able to generate large numbers of identical sessile microdroplets<sup>1</sup>. The concomitant observation of the dynamics of such collection of microdroplets is of importance for the study of nucleation, a stochastic process: large number of experiments has to be done in order to be statistically relevant. But the observation of such large populations is challenging regarding resolution per microdroplet: the larger the population, the lesser the resolution. Here, we propose a simple and efficient image processing technique allowing to characterize microdroplets dynamics (evaporation, dissolution or contraction) and to detect nucleation in each microdroplet despite poor image resolution<sup>2</sup>. In practise, water or NaCl solution microdroplets are covered by oil and evaporated under an optical microscope in transmission configuration. An image of just deposited microdroplets is presented in figure 1. With image-processing, each microdroplet is individually followed throughout its complete evaporation process. To do so, each microdroplet and its vicinity is resumed to a scalar: the standard deviation of pixels' grey levels inside a region of interest containing the microdroplet image. Following the time-evolution of this standard deviation has been proven to be a sensitive parameter to detect crystal nucleation. Additionally, this method permits distinguishing different evaporation modes like constant contact angle or constant contact diameter. Experimental results are compared to finite element simulation. Comsol Multiphysics is used in order to model droplet evaporation and transmission optical image. Axisymmetric geometry of a sessile droplet is presented in figure 2. White light source is used for illumination and the electric field intensity is calculated. Both experimental and simulation results are in good agreement.



Figure 1. Image of a just generated microdroplet array.

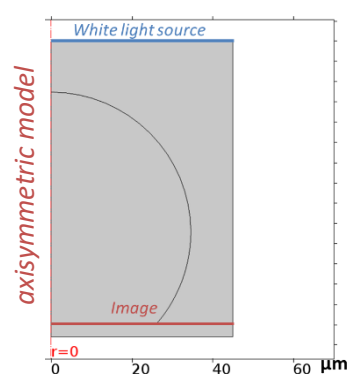


Figure 2. a) Side view on a sessile water drop covered by oil. b) Modelling geometry of a sessile drop. The scale is 10  $\mu\text{m}$

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## EXPLORING THE ENTIRE EVAPORATIVE LIFETIME OF INDIVIDUAL DROPLETS WITH HIGH TIME RESOLUTION IMAGING

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Many industries, including pharmaceutical, food and cosmetic, rely on the rapid evaporation of liquid droplets into dry particles to form their products. The aerodynamic properties of drying and dry particles are also important safety concerns for large industries such as nuclear power and combustion. Understanding the evaporation process is essential, because the specific conditions, as well as the starting composition of the liquid solution, can dramatically affect intermediate droplet morphologies and, ultimately the structure of the dry particle. Few tools exist for making these measurements, and those which do are often constrained to examining individual regions of the evaporation profile (such as freshly dispensed, pre-crystallised or post-crystallised particles), or have a time resolution which prohibits rapid morphological changes from being observed.

In this work we show how the falling droplet chain instrument can be applied to observe the full evaporation lifetime of an evaporating droplet, from dispensing to crystallisation (Fig 1). The instrument uses a droplet-on-demand dispenser to create a uniform chain of droplets which fall within a temperature controlled column. Observing the chain at different positions is equivalent to observing different stages of the evaporative lifetime of an individual droplet. Variably delayed stroboscopic imaging allows observation of the evaporating droplet with a temporal resolution of 1ms and below. Large samples of the dry particles are collected at the bottom of the column allowing off-line SEM analysis of the dry particle morphology and comparison with the evaporation profile. This instrument allows phenomena such the different stages of the crystallisation process and the chaotic rapid evaporation of freshly generated droplets to be explored.

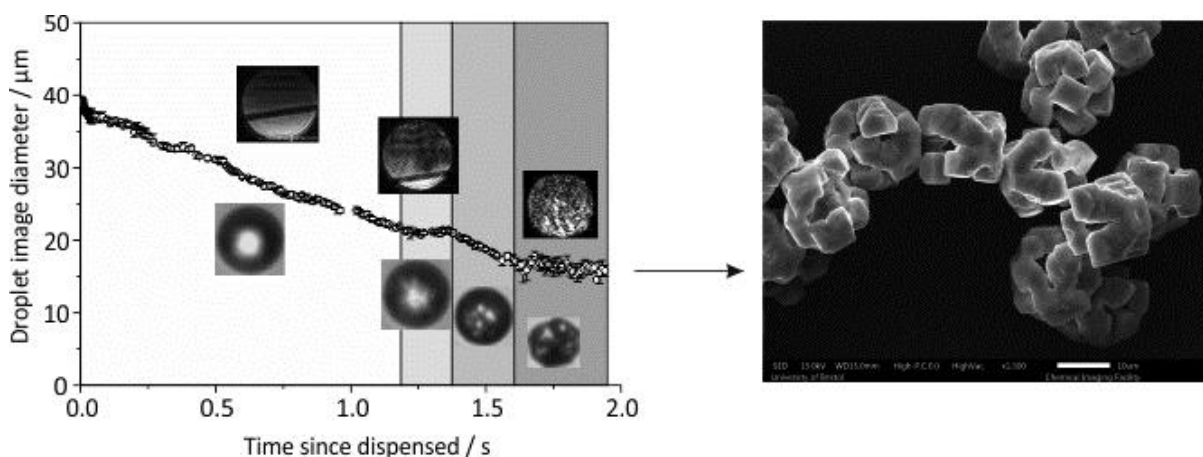


Figure 1: (Left) An example evaporation profile for NaCl aerosol, with a starting solution of 50g/L, in dry conditions at 296K. The shaded regions correspond to different morphological regimes in the drying process (homogenous droplet, inhomogeneous droplet, semi-crystallisation and dry particle). The inset pictures are representative stroboscopic (below) and light scattering (above) images of the different regimes. (Right) Corresponding SEM image of the dry particles collected following the evaporation process.

**ACKNOWLEDGEMENTS:** The authors acknowledge the funding support from the EPSRC under grant code EP/N025245/1 and IRSN under grant code S100100-101.



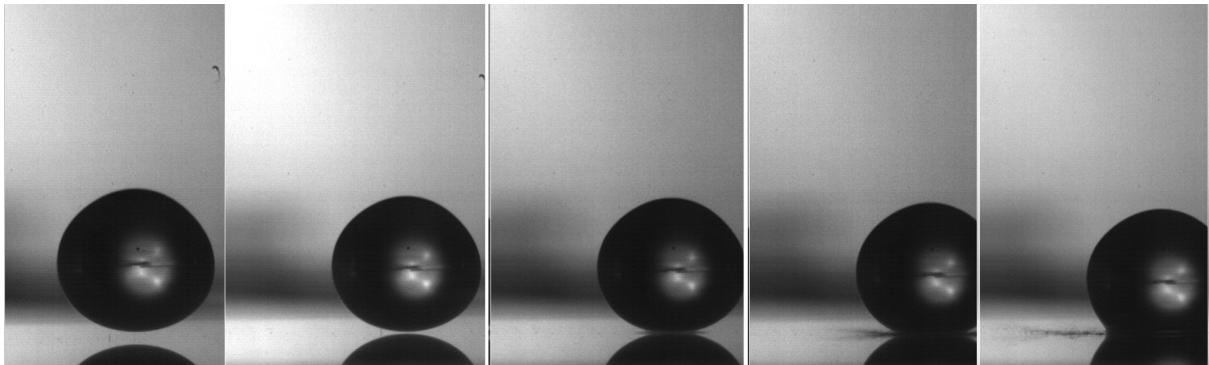
## Splash Or No-Splash!

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Drop impacts are usually studied for impact velocity to  $\sim 5$  m/s, which is limited by typical ceiling heights in laboratories. We have constructed a novel facility consisting of a 25 m-long vacuum tube, where we reach an impact speed up to 23 m/s in a controlled manner. In this way, we can access new corners of the parameter space, which have not been investigated previously. We can generate impacts with  $Re \sim 10^5$  and  $We \sim 10^4$ . By using a water glycerine mixtures our  $Re$  can be systemically varied over three orders of magnitude while keeping the Weber number very large. Our focus is here to investigate the interplay of the three forces of viscosity, surface tension, and inertia on splashing phenomena, which we observe using a high-speed camera at up to 5M frames per second, using a Kirana-5m video camera. As we increased the impact speed the lamellar thickness reduces significantly, while the jetting velocity reaches 1 km/s, or near 50 times the impact speed. We highlight the effect of varying the ambient pressure on the bottom radius of curvature of the drop. We also investigate impacts on liquid surfaces and how ejecta sheet is affected.



**FIGURE 1.** Example of a drop of water impacting a solid surface with impact speed of 21 m/s, where the surrounding pressure was reduced to  $P_{atm} = 50$  kPa. The lamella emerge at about 1 km/s

**ACKNOWLEDGEMENTS:** The authors acknowledge funding support from KAUST.

# HIGH DENSITY RATIO LATTICE BOLTZMANN SIMULATIONS OF IMMISCIBLE DROP COLLISION

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## Abstract:

The physics and dynamics of a ternary fluid system are of special interests for a variety of practical applications, including combustion engines, ink-jet printing, and oil recovery. Recent experiments demonstrated that if fuel and water are colliding in a combustion chamber, the water can be encapsulated by the fuel. This causes micro-explosions, potentially enhancing the burning rate[1].

In this contribution, I will present a numerical investigation of the collision between two immiscible droplets which, depending on the combinations of interfacial tensions, can lead to bouncing or encapsulation after the impact Fig.(1). To account for the Inertial effects, I employ a high density ratio Free energy Lattice Boltzmann model, recently developed in my group. The model describes multiphase and multicomponent fluids and allows to achieve density contrasts of order  $10^3$ [2].

By varying the impact speed, the relative liquid viscosities, the drop size and the impact parameter, I explore a wide range of Weber and Ohnesorge number, and elucidate the collision mechanisms in terms of the interplay between capillarity, inertia and viscous dissipation.

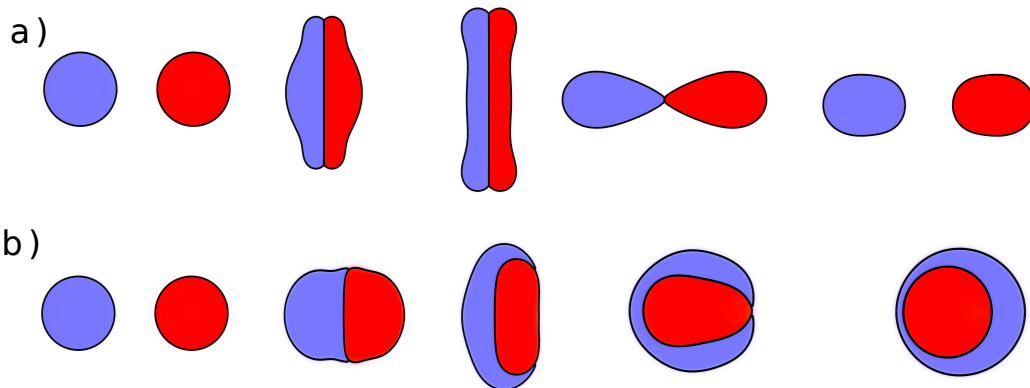


Figure 1:  
Collision between immiscible drops: a) Bouncing of droplets after impact b) Droplet encapsulation after impact.

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## **DROPLET IMPACT ON MONOFILAMENT POLYESTER FABRIC**

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Although droplet spreading on smooth surfaces is well known, spreading on textile materials is still not fully understood. Compared to a solid surface, on textile the liquid can penetrate the holes in the fabric but also spontaneously flow through the porous networks inside the fabric (wicking), making droplet spreading more complex compared to smooth surfaces. Understanding droplet spreading on textile materials is important for applications in the textile industry and forensic research.

We study droplet impact on thin monofilament polyester fabric as a function of the fabric pore size and its wettability. First, the difference between droplet spreading on a smooth surface (stainless steel) and the fabric is investigated where the fabric is either placed on a substrate or suspended in the air. We show that a droplet spreads less on the fabric compared to the smooth surface. Furthermore, a difference in spreading is observed between the spreading on fabric with and without substrate due to the liquid penetrating the fabric. Secondly, we compare experimental results with Lattice Boltzmann simulations and show that the decreased spreading on the fabric with substrate is due to an increased viscous dissipation in the spreading droplet.

## Quasi-normal impacts and the kinematic match for walking droplets

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In 2005, Yves Couder [1] and collaborators discovered that coalescence of a droplet with an underlying bath may be inhibited permanently by vertical oscillation of the bath, resulting in a dynamical system resembles a ball bouncing on an oscillating trampoline. Moreover, bouncing droplets can become unstable to lateral perturbations, showing a tendency to “walk” along straight lines. Each bounce triggers new surface waves and, in turn, the resulting wave field affects the droplet trajectory after each impact. The droplet then become a motile wave source that is guided by the wave field.

We model droplets as spheres impacting on the free surface of the bath [2] and we show that the resultant problem can be decomposed into an axisymmetric vertical impact and the unforced surface waves. The method imposes only the natural kinematic and geometric constraints and yields detailed predictions of the changes in the contact area and pressure field during each impact. We simulate droplets walking on a vertically oscillating bath and compare the results to experimental reports available in the literature [3,4,5] with remarkable agreement. Furthermore, we show that this model is able to capture the recently discovered phenomenon of “superwalking” droplets [6].

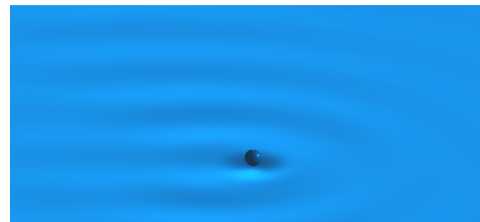


Figure 1: Simulation results for walking droplet.

**ACKNOWLEDGEMENTS:** The authors acknowledge the support of EPSRC project EP/N018176. Authors also thank Adam Damiano and Rahil Valani for sharing details of their experimental measurements.

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## Acoustic study of a water droplet impact on a water surface

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The impact of a liquid droplet on a liquid surface produces a recognizable sound. Although the generation of this sound was related to the underwater entrainment of vibrating air bubbles [1] (see fig.1), most previous studies have dealt with the hydrodynamic aspect but very little with the acoustic one.

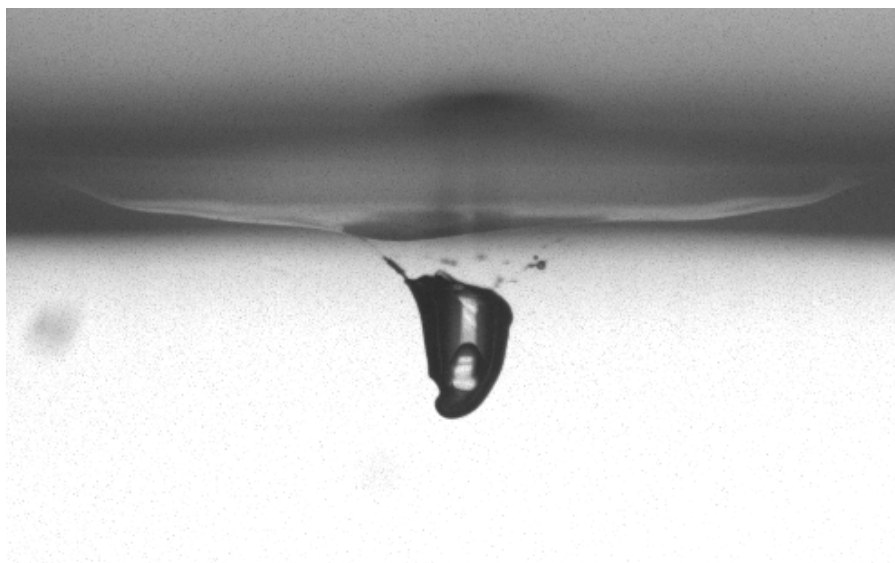
Depending on the droplet diameter and its impact velocity, e.g., several regimes for bubble production were reported. The so-called "irregular entrainment", concerned by this study, occurs for large droplets at high impact velocities (see ref. [2], Fig. 5). The latter received less attention because of its non-repeatability and its randomness.

We study the acoustic signal and the hydrodynamic behaviour of the liquid during an irregular entrainment. Hydrophone and microphone recordings were synchronized with a high-speed camera video. Time and frequency analysis were performed on the acoustic signals whereas the interface features were highlighted thanks to a tracking algorithm. The results were compared to existing models.

ACKNOWLEDGEMENTS: The authors thank Le Mans Acoustique for the funding support.

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*Fig.1 : Example of an air bubble created during an impact of a water droplet on a liquid surface.*

# Freezing morphologies inside a droplet impacting on a cold surface

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(Dated: March 30, 2019)

## Abstract

The solidification behaviour of a droplet after it smashes onto a cold surface is determined by the initial energy of the droplet and the applied undercooling to the substrate. In this work, we use Total-Internal-Reflection (TIR) technique to reveal and characterise various freezing morphologies of a droplet after it impacts onto a cold surface. This technique allows temporally and spatially resolved insights into the nucleation events that occur within the liquid next to the cold surface on an evanescent length scale. The sequences of experimental snapshots in Fig. 1(a-d) highlight different freezing morphologies of a liquid hexadecane droplet after it impinges on flat cold surfaces maintained at various undercooling. We for the first time show that at sufficiently high undercooling a peculiar freezing morphology exists that involves sequential advection of *frozen fronts* from the centre of the droplet to its boundaries. We examine this phenomenology by coupling the elements of classical nucleation theory with large scale hydrodynamics. We also report a self-peeling phenomenon of the frozen splat that is driven by the existence of a unique crystalline state for *n*-alkanes.

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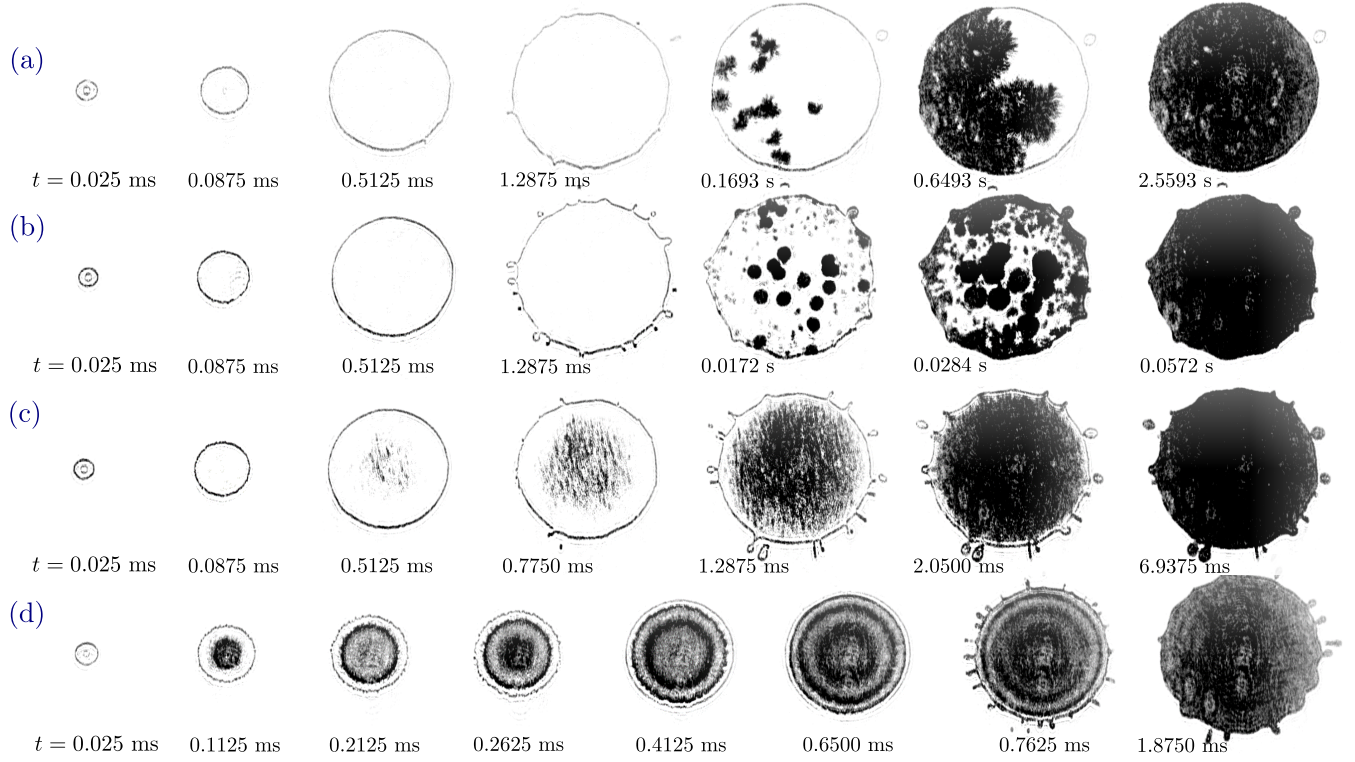


FIG. 1. Sequences of experimental snapshots highlighting the effect of substrate undercooling.  $\Delta T = T_m - T_s =$  (a) 2 K (b) 4.5 K (c) 8 K (d) 12 K on the freezing morphology of an impacting droplet;  $T_m$  is the melting temperature of the liquid and  $T_s$  is the temperature of the substrate. In the shown cases the impact velocity  $U = 3.8$  m/s and the temperature of the droplet at the time of impact  $T_d = 20^\circ\text{C}$ .

# POST-IMPACT SPREADING OF FERROFLUID DROPLET ON SOLID SURFACES UNDER THE INFLUENCE OF HORIZONTAL MAGNETIC FIELD

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Droplet impact dynamics on solid surfaces have been studied extensively by many researchers over the past few decades because of its numerous applications, viz. fire suppression sprinkler systems, inkjet printing<sup>1</sup>, spray painting and spray cooling to name a few. In this study, the focus is on the spreading dynamics of ferrofluid on different substrates under the influence of different magnetic field strength. Droplets of water based 7.5% Fe<sub>3</sub>O<sub>4</sub> solution have been impacted from different heights on glass and Teflon surfaces. The magnetic Bond number is defined as  $B_m = \frac{B^2 D_0}{\mu_0 \sigma}$  where B represents magnetic field induction, D<sub>0</sub> is the initial droplet diameter,  $\mu_0$  is the magnetic permeability of free space, and  $\sigma$  represents the surface tension. The morphology of the impacted droplets under the influence of horizontal magnetic field have been studied by parameters namely, transverse spreading factor ( $\psi_T$ ) and axisymmetric spreading ratio ( $\zeta$ ). Transverse spreading factor ( $\psi_T$ ) is defined as the ratio of post-impact transverse length, perpendicular to the magnetic field to the initial droplet diameter. Similarly, axisymmetric spreading ratio ( $\zeta$ ) is expressed as the ratio of the post-impact transverse spreading length to the longitudinal spreading length. Figure 1 depicts the influence of magnetic bond number on temporal distribution of transverse spreading factor. It has been observed that with the increasing magnetic field strength, the transverse spreading increases for same impact velocity. It has also been noted from figure 2 that the axisymmetric spreading ratio ( $\zeta$ ) increases with increasing magnetic bond number for same weber number. Therefore, more spreading has been observed in horizontal magnetic field as compared to without field effect for ferrofluid on both the substrates.

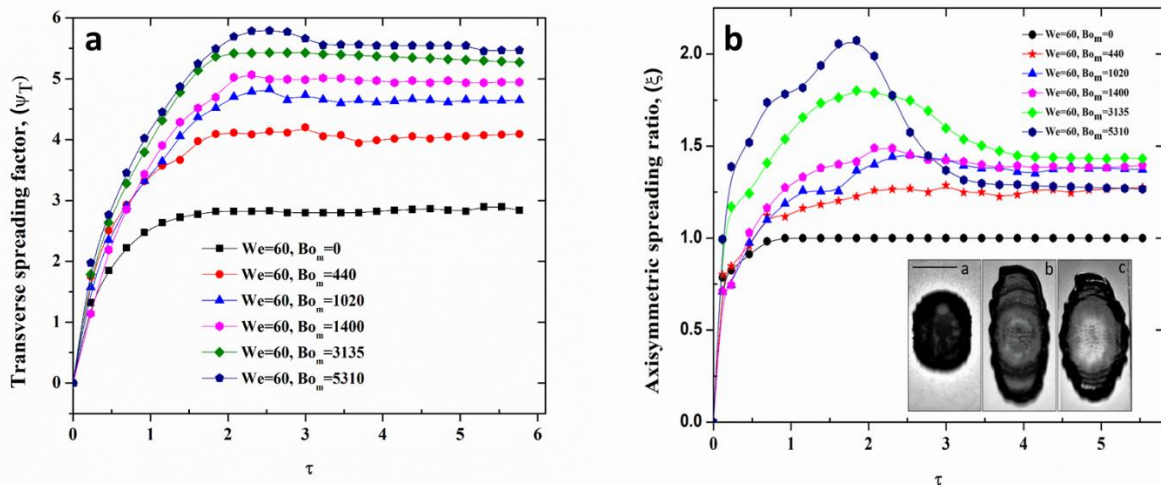


Figure 1: Temporal variation of post-impact transverse spreading factor and axisymmetric spreading ratio of ferrofluid droplet on glass substrate.

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## BUBBLE COLLISIONS ON A FIBER ARRAY

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Bubbles in liquid flow channels can affect momentum, mass, and heat transfer in a liquid-gas two phase flow, and controlling bubbles is thus crucial in various engineering and natural systems. Researchers have suggested various methods to control bubbles using mesh structures. We here present an experimental and theoretical investigation of bubble collisions on parallel arranged fibers. We visualized the impact outcomes using high speed videography, leading to the classification of the outcomes into three modes: capturing, single bubble rising, and splitting (see Figure 1). The experimental results reveal that the impact mode is mainly determined by the rising speed of the bubble and the distance between the fibers. The impact outcomes are shown in a regime map, and we explain the regime boundaries through theoretical analysis. Our study provides insights into bubble filtration or generation for improving heat and mass transfer using bubbles.

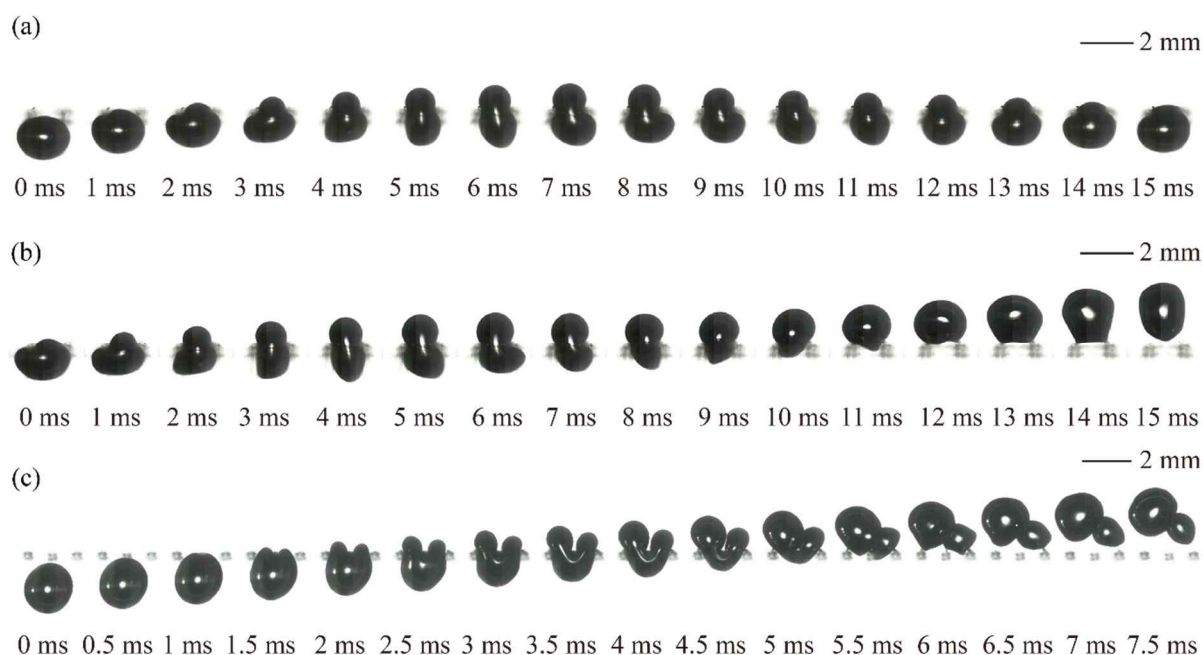


Figure 1. Three outcome modes of bubble collisions on parallel arranged fibers.

**ACKNOWLEDGEMENTS:** This work was supported by a grant of the National Research Foundation of Korea (NRF) funded by the Ministry of Science and ICT (grant no. 2017R1E1A1A01073599) and a grant of the Korea Health Industry Development Institute (KHIDI) funded by the Ministry of Health & Welfare (grant no. HI18C0432), Republic of Korea.

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## RELAXATION OF LIQUID DEFORMATION UNDER IMPACTING DROP

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Slippery surfaces have recently gained a lot of attention owing to its self-cleaning, anti-icing and anti-fouling properties. Of particular importance is the dynamics of impacting drops on such surfaces. Here, we report experiments of complete drop bouncing on a liquid surface. Dynamic optical measurements of the liquid-gas interface, during and after impact, are recorded using reflection digital holographic microscopy. This is complemented by simulations based on lubrication theory, providing a direct comparison to the experimental profiles.

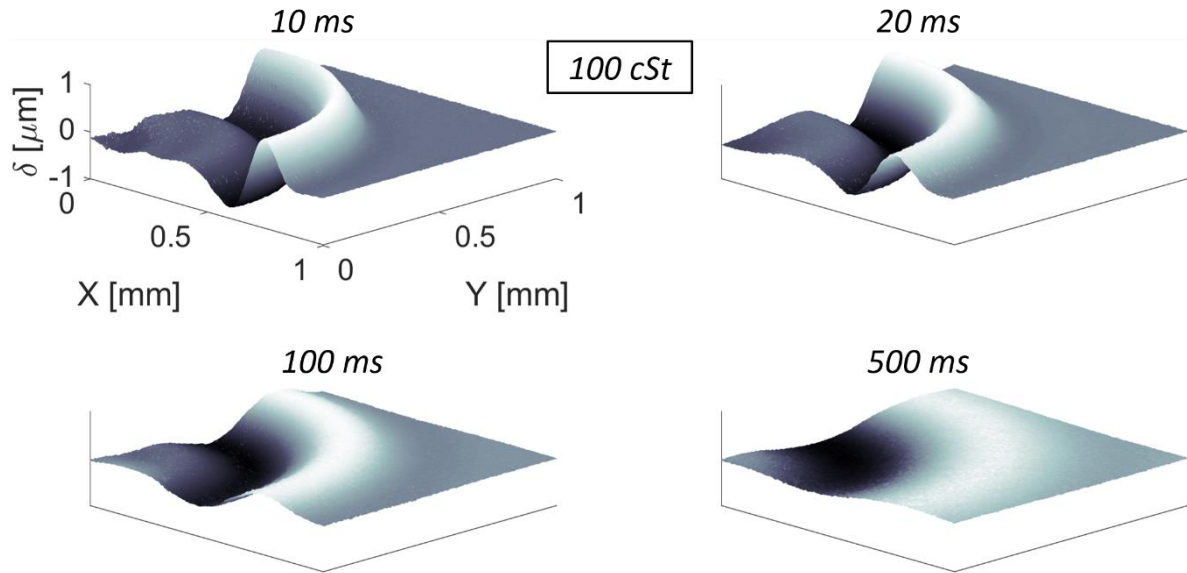


Fig: Relaxation of liquid deformations upon drop impact. Drop diameter  $D = 1.25 \text{ mm}$ , impact speed  $v = 0.13 \text{ m/s}$  and initial film thickness  $h_0 = 15 \mu\text{m}$ .

## Combining emulsion solvent evaporation with inkjet printing: Preparation and deposition of polymeric microcapsules and particles

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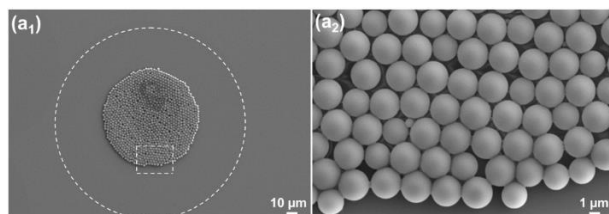
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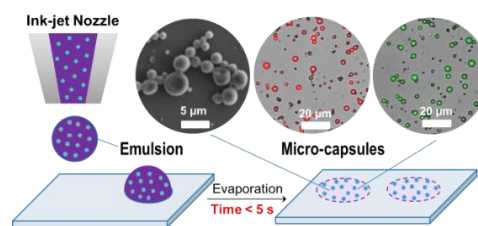
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**ABSTRACT:** The combination of emulsion solvent evaporation with ink-jet printing allows the rapid fabrication of polymeric particles or microcapsules at a target location on a surface.<sup>[1]</sup> Oil/water emulsions with polymer contained inside the oil phase were used as inks. Polymeric particles formed in-situ after being printed onto a substrate because of the preferential evaporation of the oil phase. Circular deposits of spherical polymer particles with uniform thickness are obtained. The effects of the hydrophobicity of substrates and the physical properties of the oil on the morphology of the deposits was explored. The size of particles can be well controlled by varying the initial size or concentration of emulsion droplets. Monodispersed emulsion droplets can be generated in a microfluidic device, and a monolayer deposit of uniform polymer particles with ordered hexagonal packing is obtained (Figure 1).

The same method was developed further to prepare and deposit polymer microcapsules (Figure 2). The oil droplets contain a shell-forming polymer and a core-forming fluid that is a poor solvent for the polymer. After the emulsion is printed onto the substrate, the good solvent evaporates by diffusion through the aqueous phase, which drives the phase separation between the polymer and poor solvent to form microcapsules. This method has been demonstrated for microcapsules with various shell-forming polymers and core-forming poor solvents. Cargoes such as fluorescent dyes or active ingredients can be encapsulated. We discuss the physical parameters that need to be controlled for the successful fabrication of microcapsules in inkjet printing. The method for rapid, in-situ encapsulation could be useful for controlled-release applications including agrochemical sprays, fragrances, functional coatings and topical medicines.



**Figure 1.** SEM images of a monolayer deposit of polymer particles.



**Figure 2.** Preparation and deposition of polymeric microcapsules.

**ACKNOWLEDGEMENTS:** This work was funded by EPSRC under Grant EP/N025245/1.

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## PARAMETRIC OPTIMIZATION FOR HIGHER QUALITY/RESOLUTION ELECTROSTATIC JET PRINTING

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Electrostatic jet printing (ESJET) is a novel technique that can achieve a very small deposition of different materials. In this technique an electric field is applied to a liquid meniscus coming out of a nozzle. The electric stress then deforms the meniscus into a cone shape (Taylor cone) and due to the singularity at the apex, a fine jet, much smaller than the nozzle in size is produced. The jet then breaks up down stream into droplets. Its advantage over conventional ink jet is that the droplets are much smaller than the nozzle size, a droplet volume down to 0.004 pico liters can be deposited compared to 5 pico liters in conventional inkjet printing, which means higher resolution can be achieved. Two important aspects have to be considered in printing: resolution and quality.

To make sure that these aspect are fulfilled, we developed theoretical models that allow us to have a full control over the printing parameters. We developed a model to predict the jet length. Also, another model to predict the printed line width. Finally, we developed a model to predict the stable jetting conditions. As a result of this optimisation a very small lines and droplet pixels with high quality were printed easily using ESJET technology. The improvement that can be made by knowing the jet length for example is shown in Fig. 1.

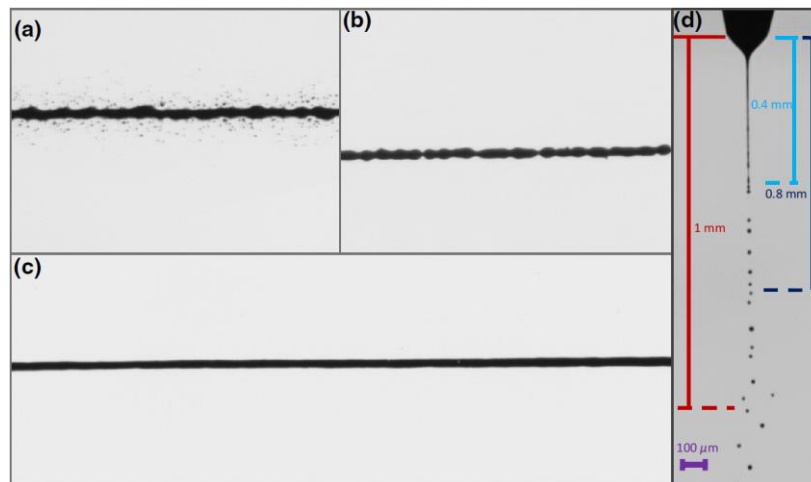


FIG. 1. 5  $\mu\text{m}$  copper lines printed at a printing distance of (a) 1 mm (b) 0.8 mm, and (c) 0.4 mm. The jet breakup during the printing is shown in (d).

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## Bubble entrainment from an acoustically driven meniscus in a piezo-acoustic drop-on-demand inkjet nozzle

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In piezo acoustic Drop-On-Demand (DOD) inkjet printing a single droplet is produced for each piezo driving pulse. A phenomenon that disturbs the droplet formation process is the entrainment of bubbles in the ink channel, as shown in the figure below. Here, bubble pinch-off from an acoustically driven meniscus in a DOD printhead (MicroDrop Technologies GmbH) was studied for various acoustic driving waveforms. The piezo actuation pulse sets into motion a slosh mode of the printhead, resulting in a large amplitude meniscus motion with a frequency on the order of 10 kHz. It also actuates a piezo longitudinal resonance mode, which introduces a low-amplitude 100 kHz component to the meniscus motion. The slosh mode, piezo longitudinal resonance mode, and the falling edge of the rectangular piezo driving pulse destabilize the retracted concave meniscus when propelled outward, by jet formation due to a combination of geometrical focusing of the flow and an inhomogeneous pressure gradient field. Two well-timed outward accelerations of the meniscus result in the formation of a central jet surrounded by a toroidal jet. A phase mismatch of the oscillatory behavior of the two jets leads to the enclosure of an air cavity leading to bubble entrainment through pinch-off. It is shown that, next to pulse timing, the driving pressure is a control parameter of the entrainment process and that the threshold for bubble pinch-off can be increased by suppressing the piezo longitudinal resonance mode by waveform design.

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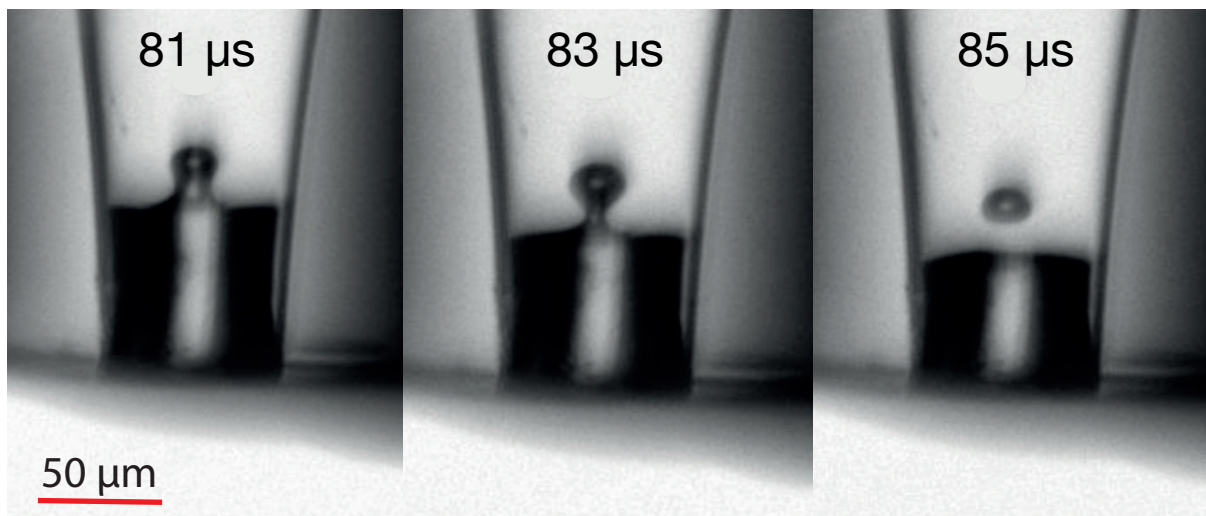


Figure: Bubble entrainment in a 70 µm diameter nozzle of a piezo Drop-on-Demand inkjet printhead.



## LIQUID CRYSTALLINE SELF-ASSEMBLY INSIDE LIQUID MARBLES: MILLIMETRE-SIZED SPHERES WITH TAILORED STRUCTURAL COLOR

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Liquid marbles (LMs) are drops of liquids coated with solvophobic colloid particles. These peculiar objects have attracted vivid interest because they constitute a robust platform for transporting liquids on various substrates. LMs show a plethora of unusual properties [1], for instance they behave as soft solids that can be reversibly deformed and easily manipulated [2]. In addition, although the particle layer at the surface of the LM prevents the enclosed liquid from wetting the supporting solid, interactions between the liquid and the phase surrounding the LM are allowed. The idea of exploiting LMs as ‘mini-reactors’ for creating new materials has been proposed soon after their discovery. However, despite its potential, this interesting concept has been rarely used so far.

On the other hand, over the last decades, there has been increased interest in the liquid crystal (LC) self-assembly of soft matter confined within curved, flexible interfaces. This is due to the fascinating physical phenomena that take place in such geometries, as well as the demonstrated potential for new applications [3]. Especially concerning optical applications, biomimetic approaches utilizing bio-derived materials based on cellulose, have been broadly developed. A prominent example is aqueous solutions of hydroxypropylcellulose (HPC), where short-pitch chiral nematic LC phases are formed above a critical polymer concentration. At sufficiently high concentrations, the chiral nematic pitch is reduced to sub-micrometre values and the material shows structural coloration [4].

In this contribution, we show that LMs can serve as a robust miniature platform for monitoring and controlling the self-assembly of bio-derived polymers into a LC structure that yields bright structural colour. Our strategy was to combine the ability of HPC to form chiral nematic LCs with the extraordinary properties of LMs. We therefore prepared millimetre sized LMs by coating droplets of aqueous HPC solutions with hydrophobic silica nanoparticles. By immersing the LMs into organic liquids with poor (yet finite) water miscibility, we were able to precisely extract a desired amount of water. This allowed the polymers to organize in a chiral nematic structure, the periodicity of which could be chosen by tuning the final concentration in the LM. This approach resulted in LMs with selective Bragg light reflection that can be programmed to be anywhere from the UV to the IR range. Interestingly, the photonic response of the LMs is thermosensitive: increasing (resp. decreasing) the temperature led to a red (resp. blue) shift of the reflected wavelength.

ACKNOWLEDGEMENTS: We thank Luxembourg National Research Fund for supporting this work.

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## A Mathematical Model for the One-Drop-Filling Process

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Liquid crystal devices (LCDs) are ubiquitous in modern day life, and faster and more accurate manufacture of these devices is required to meet increasing global demand. The optimisation of the manufacturing process generally involves attempting to reduce manufacturing time by increasing flow velocities and filling speeds. However, implementing such changes runs the risk of causing deformation to the director structure which is crucial to the display.

A common process of filling the liquid crystal layer in LCDs used by display manufacturers is the One-Drop-Filling (ODF) process. In the first stage of the ODF process, a bottom substrate is coated with an alignment layer in order to correctly orient the liquid crystal layer within the cell. Droplets of liquid crystal are then dispensed onto the bottom substrate and are allowed to equilibrate. In vacuum, a top substrate is then lowered at a constant speed onto the droplets, squeezing them, causing them to coalesce, and eventually creating a continuous thin film of liquid crystal within the cell. Although this method is an efficient way to fill liquid crystal cells used in LCDs, it is known that it can cause deformation to the director structure within the liquid crystal cells known as “ODF mura” (as shown in Figure 1(left)) which, in turn, can affect the optical performance of the final display.

We propose a model for the squeezed coalescence of droplets during the squeezing stage of the ODF process to provide insight into the formation of this ODF mura. This model assumes that the timescale for changes in the shape of droplets due to surface tension is much longer than that of squeezing, and so the effects of surface tension can be neglected. This assumption allows the calculation of the spreading behaviour of multiple squeezed droplets to be determined by geometrical methods using conservation of volume.

We obtain implicit expressions for the droplet boundary speed of multiple squeezed coalescing droplets which allow us to make direct comparisons with experimental photographs of the ODF mura (as shown in Figure 1(left) and (right)). Specifically, the deformation to the director structure at any point is assumed to be proportional to the droplet boundary speed as it passes over that point. The deformation to the director structure calculated shows a remarkable similarity to experimental photographs of the ODF mura. Motivated by the success of this model we further investigate the flow and director within multiple squeezed coalescing droplets and within a single isolated squeezed droplet using Ericksen-Leslie continuum theory to further understanding the formation of ODF mura .

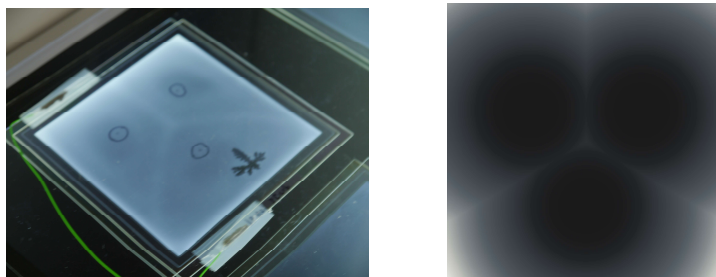


Figure 1 : (left) experimental image of ODF mura (right) theoretical calculation of droplet boundary speed.

ACKNOWLEDGEMENTS: This work is supported by an EPSRC iCASE studentship sponsored by Merck KGaA. We would like to thank Dr. David Wilkes and Dr. Leo Weegels of Merck KGaA for their collaboration.

## VARIATION OF EXPERIMENTAL CONDITIONS TO OPTIMISE MACHINE-LEARNING DISCRIMINATION OF PATTERNS FROM DRIED BLOOD DROPLETS

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In a world where medical industries struggle for funding, we are in need of cheap, simple and easy ways to detect medical conditions. Our work aims to create reliable image processing of dried blood droplets using machine learning algorithms which will be verified within sport science to track exhaustion and subsequently extended to biomedical fields for diagnosing diseases. Physiological conditions can change the composition of blood and have been shown to affect the patterns left from dried blood droplets [1]. The change in these patterns can determine whether the blood of the volunteer is, for example, healthy, anaemic [1] or exhausted [2]. Understanding which experimental parameters influence the features of the droplet will enable the technique to move from the laboratory to the field. Within this study, we have maximised the discrimination between blood taken before, during and after exhaustive exercise. To optimise the machine learning algorithm we varied many experimental parameters including: lighting conditions, camera settings, droplet volume, time of imaging, substrate and humidity. The raw images were treated following a previous methodology [2] which includes averaging over droplets from identical conditions, colour-channel optimisation, and a one-dimensional Fourier transformation to identify angular patterns while preserving radial gradients. The database of treated images are then processed using the machine learning algorithm to aid optimisation of the discrimination between the different conditions. The algorithm combines both Principal Component Analysis (PCA) [3] method and Linear Discriminant Analysis (LDA) [4]. Previously this technique was able to predict with up to 95% accuracy. Our findings will improve this accuracy and enable the method to be developed for use in the field.

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## DESIGNING A LOW-COST SYSTEM FOR THE SCREENING OF BACTERIAL TOXINS

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It is estimated by the Royal Society of Chemistry that diagnostics comprises 3-5% of healthcare spending, yet impacts 70% of healthcare decisions<sup>1</sup>. Therefore, developing low-cost point of care diagnostic tools has the potential to impact the diagnosis and treatment of potentially fatal diseases in developing countries.

Traditionally, the main use of liquid crystals (LCs) has been for LC display (LCD) purposes. Recently however, liquid crystal droplets have received increased exposure due to their potential in a variety of applications, including biosensors<sup>2</sup>. Indeed, their potential for low-cost, sensitive sensors makes them exciting systems to study.

Here, we describe a biosensor that employs phospholipid-coated nematic liquid crystal droplets to detect antimicrobial peptides (AMPs)<sup>3</sup>. Mono-disperse lipid-coated LC droplets with diameters in the range of 10s of microns are produced using microfluidic devices with a flow-focusing configuration. We describe the detection of the model AMP Smp43, from the venom of the North African scorpion *Scorpio maurus palmatus*. In our system, the AMP disrupts the lipid monolayer, causing a clear observable change in the nematic liquid crystal droplets, from a radial to a bipolar configuration. Where the droplets are captured individually in bespoke microfluidic traps, the switch occurs at concentrations <6µM, well within the biologically active range.

We also examine the response of the lipid-coated LC droplets when immobilised in systems such as hydrogels. The designs we consider are selected with the view that this technology can be used as a dipstick test for AMPs, taking us closer to the goal of a low-cost, disposable point of care diagnostic tool.

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## CAPILLARY RETRACTION OF AN AXISYMMETRIC LIQUID LIGAMENT

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We study capillary retraction of an axisymmetric viscous liquid ligament through experimental, numerical and analytical methods.

The experimental setup consists of a continuous liquid-jet system: an oscillating pressure modulation induces surface instabilities to break the stream into a train of equally spaced droplets; ligaments are originated between them. We analyse their contraction dynamics through high-speed imaging.

We perform numerical simulations with the open-source solver Basilisk to integrate full 3D Navier-Stokes equations for a diphasic system in an axisymmetric configuration.

We corroborate numerical results with a theoretical model which uses the well-known Eggers one-dimensional (1D) approximation [3] for a free-surface slender cylindrical flow. We study the 1D-model equations through asymptotic analysis to derive an expansion of the ligament profile for long times, once a steady state is reached after a transient regime. Three distinct regions with different characteristic length and time scales are identified in the retraction domain: a growing spherical rim, a steady filament section and an intermediate matching zone. Our analytical arguments can explain emergence of capillary ripples and instability evolution.

Analytical, numerical and experimental results show good agreement.



### ACKNOWLEDGEMENTS:

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## SODIUM IONS AND ENDOTHELIAL GLYCOLYX INTERACTIONS UNDER FLOW CONDITIONS

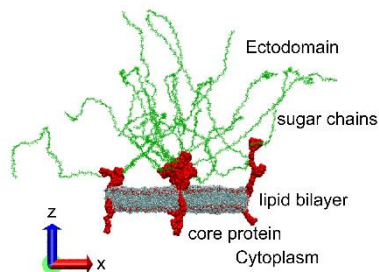
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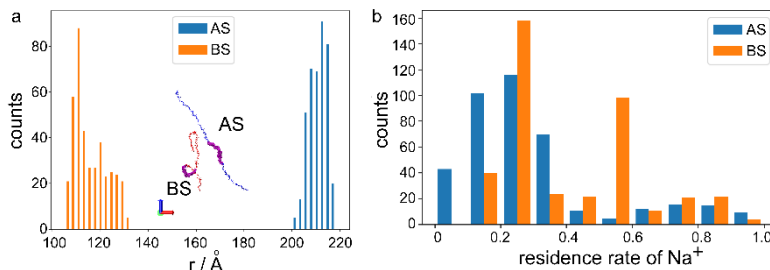
The endothelial glycocalyx layer (EGL) is a thin layer with a thickness of 50 to 500 nm coating endothelial cells. Vascular diseases are intimately associated with ion concentration around the EGL [1, 2]. Study of ions and EGL interactions would help to advance understanding and cure of endothelial glycocalyx-related vascular diseases.

In this research, large-scale molecular dynamics simulations were conducted to investigate the interactions of  $\text{Na}^+$  ions with the EGL under physiological flow conditions. Figure 1 shows a small patch of the EGL under investigation. The biomolecules are solvated in the NaCl solution with a concentration of 0.1 M.



**Figure 1** A small patch of the endothelial glycocalyx. Ions and water molecules are not shown.

Two sugar chains with identical components but different initial conformations were selected. The residence rates of  $\text{Na}^+$  ions around these two sugar chains were compared. Results show that the corner structure has a higher residence rate of  $\text{Na}^+$  ions than its stretching counterpart (Figure 2). Thus, the corner confines  $\text{Na}^+$  ions, whereas the stretching structure facilitates  $\text{Na}^+$  ion movement. The  $\text{Na}^+$  ion transports across the EGL under varying flow velocity conditions were also compared.



**Figure 2** Residence rate of  $\text{Na}^+$  ions around two sugar chains with identical components but different conformations

This research provides new insights into the ion-EGL interactions, which adds to our understanding of microvascular fluid exchange.

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# JOINT EFFECT OF ADVECTION, DIFFUSION AND CAPILLARY ATTRACTION ON A SPATIAL STRUCTURE OF PARTICLE DEPOSITIONS FROM EVAPORATING DROPLETS

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Self-organization of colloidal particles in drying droplets is of fundamental and applied interest. In the experiment [1] deposition in the form of a ring and a spot as a result of evaporation of picolitre drops of water inks and various solvent mixtures were obtained. These sediments are characterized by a well-ordered hexagonal structure of the particle arrangement. In the case of annular deposition, separate chains of particles were formed in the central part. Understanding such processes is important in a number of applications, for example, in the formation of transparent conductive films [2].

In this work, we present a simplified model, which allows us to perform computer simulations of the particles transport in an evaporating droplet with a contact line pinned to a hydrophylic substrate. The model accounts for advection in the droplet, diffusion and particle attraction by capillary forces. On the basis of the simulations, we analyse the physical mechanisms of forming of individual chains of particles inside the annular sediment [3].

The results show that the annular sediment is formed by advection and diffusion transport. The close packing of the particles in the sediment is possible if the evaporation time exceeds the characteristic time of diffusion-based ordering. The chains are formed by the end of the evaporation process due to capillary attraction of particles in the region bounded by a fixing radius, where the local droplet height is comparable to the particle size. At the beginning of the evaporation, the annular deposition is shown to expand faster than the fixing radius moves. However, by the end of the process, the fixing radius rapidly outreaches the expanding inner front of the ring. The snake-like chains are formed at this final stage when the fixing radius moves toward the symmetry axis. The resulting structure and density of particle distribution  $\rho$  are shown in Fig. 1.

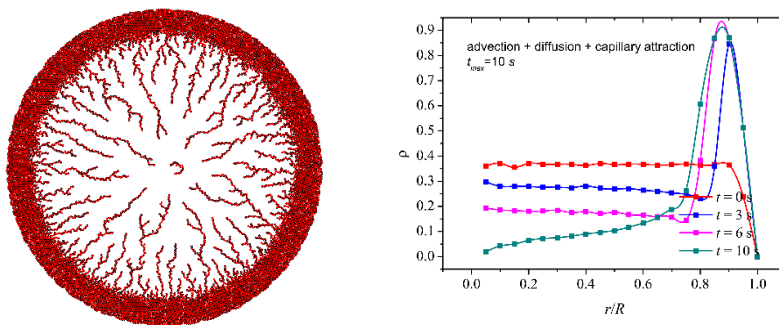


Figure 1. The spatial structure and density of particle distribution  $\rho$ , obtained by means of computer simulations.

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## A LEVEL SET METHOD FOR TWO-PHASE ELECTROHYDRODYNAMICS

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Electrohydrodynamics (EHD) deals with the dynamics of fluid flow in the presence of an electric field. EHD has gained extensive attention in recent years due to its relevance to a wide range of industrial applications, ranging from microfluidics, heat exchangers, to electro-spraying and ink-jet printing. In the present study, we develop a computational model for two-phase EHD based on a level set method and a leaky-dielectric model. We first test the method by studying the steady deformation of a droplet subjected to an electric field. Fig. 1(a) shows the Taylor deformation parameter as a function of electric capillary number  $CaE$ , which represents the relative importance of electrical stress compared with surface tension. Two important ratios defined as  $R$ ,  $Q$  (ratios of conductivity and permittivity, respectively, between the droplet and the matrix fluid) play significant roles in determining the shape of the droplet. From the results, it is seen that when  $R > Q$ , the droplet deforms to a prolate shape; while it deforms into an oblate shape when  $R < Q$ . The present results agree well with the analytical solution of Feng (2012). We also study the transient deformation of a droplet subjected to an electric field and shear flow. We compare the time evolution of the Taylor shape parameter of droplets under various situations with the numerical results of Mahlmann (2009) and obtain good agreement (shown in Fig. 1(b)).

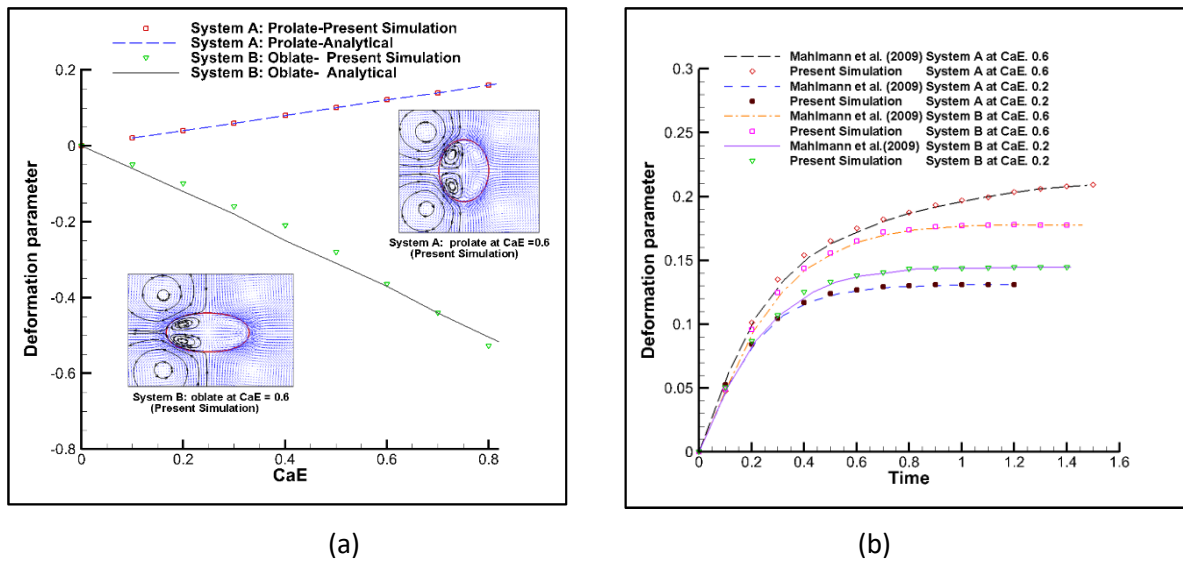


Fig. 1: (a) Taylor deformation parameter as a function of electric capillary number ( $CaE$ ) for a droplet in an electric field; (b) Time evolution of the deformation parameter when droplet subjected to both electric field and shear flow. Here system A has  $(R, Q) = (2, 0.5)$  and system B has  $(R, Q) = (0.5, 2)$ .  $Re = 0.1$ .

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## **SLOW GAS MICROFLOW PAST A SPHEROID: SOLUTION BASED ON GRAD'S MOMENT EQUATIONS USING THE BOUNDARY ELEMENT METHOD**

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Grad's 13-moment transport equations are solved numerically for slow, steady flow of a gas past a prolate or oblate ellipsoid of revolution at low Mach numbers. It is assumed that the conditions are such that the molecules mean free path can be of the same order as the characteristic size of the spheroid. This scenario is more likely to occur when the particle size is in the nano- or micrometer range. In this case, rarefaction or non-equilibrium effects such as velocity slip and temperature jump at the gas-solid interface may be conspicuous and the validity of the Navier-Stokes equations to model the flow becomes dubious. Instead, Grad's 13-moment equations (Grad, 1949; Struchtrup, 2005) represent a set of partial differential equations for the macroscopic variables velocity, temperature, density, heat-flux, and stress deviator in the gas capable of modelling non-equilibrium effects. These equations for the bulk are supplemented with a set of conditions at the gas-solid boundary derived in a consistent manner that relate the velocity slip and temperature jump with components of the stress deviator and heat flux in the gas at the boundary.

Under the assumptions made, we show that Grad's equations reduce to the Stokes flow equations for the pressure and a pseudo-velocity coupled with the Laplace equation for the heat-flux potential. We cast this set of differential equations as a system of boundary integral equations that combines integral equations from Stokes flow with those from potential theory and solve it using the boundary element method. We use this formulation to compute the net drag produced by the streaming gas flow on both prolate and oblate spheroids of revolution with various aspect ratios as a function of the Knudsen number (ratio of molecular mean free path to particle characteristic length). This method opens a new path for investigating other gas non-equilibrium phenomena that can be modelled by the same set of equations, such as thermophoresis, on non-spherical geometries.

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## A LATTICE-BOLTZMANN MODEL OF ELECTROCAPILLARITY

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Dielectrophoresis and electrowetting have become widely used techniques for controlling and manipulating small amounts of liquids. Applications of dielectrophoresis include the transport, and separation of liquids and other particles of different electric permittivity. Examples of electrowetting applications are electronic paper displays, adjustable lenses, and lab-on-a-chip devices.<sup>1,2</sup> In both cases, the underlying phenomena can be encompassed under electrocapillarity, the interaction of electric fields with multiphase systems where the effects of surface tension are comparable with electrostatic forces. Usually, one of the phases is a conducting liquid surrounded by an ambient dielectric or two liquids of different electric permittivities. Fundamental aspects in electrocapillarity are still open for investigation; this mainly concerns the dynamical aspects, for example, the motion of contact lines and the shape that the liquid interface acquire in the presence of electric stresses.<sup>2,3</sup> This is important since most of the applications rely a precise control of the liquids and by placing electrodes in clever arrays. Simulations on electrocapillarity are timely as they may provide insights to address these aspects. In this contribution, we propose a simple lattice-Boltzmann method that is capable of simulating electrocapillarity. We use a binary fluid model that includes capillary phenomena and extend the algorithm to include the forces produced by electric fields. The electric field and the charges are derived from a potential function, which we obtain by a relaxation method.<sup>4</sup> We first validate our method by comparison against the experimental observations. Then, we examine the morphology of droplets under dielectrophoretic stresses.

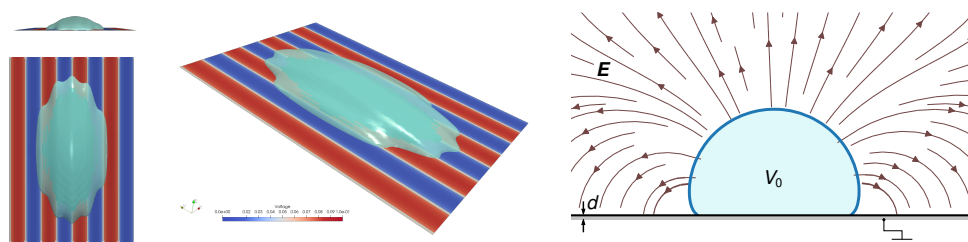


Figure 1: Example of simulations of droplet spreading by dielectrophoretic forces (left) and electrowetting (right).

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## SELF-ASSEMBLY OF COLLOIDAL PARTICLES INTO EVAPORATING SESSILE DROP OF WATER-GLYCEROL AND WATER-ETHYLENE GLYCOL BINARY SOLUTIONS

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The evaporating sessile drop of binary solution is an interesting object of theoretical and experimental investigations due to its important fundamental and practice applications. Recently, the concentration Marangoni driven migration of particles in such a system was experimentally investigated [1] There was experimentally shown that the polystyrene nanoparticles were moved by toroidal vortex flow acted inside a drop, and the direction of this flow depends on solvent components types (ethanol/water, isopropanol/water, metoxypropanol/water), ratio of mixture, particle size and environmental conditions. In such a system, the thermocapillary Marangoni number is much smaller than concentration Marangoni number, so that the concentration gradient is a main origin of Marangoni instability. In our study, colloidal nanoparticles self-assembly into evaporating sessile drop of binary solvent mixture water-glycerol ( $C_3H_5(OH)_3$ ) deposited on a flat substrate is considered. The components of such a solvent have highly contrasting properties. Under normal conditions, pure liquid water has approximately a 10 000 times higher evaporation rate and viscosity a 1000 times less than pure glycerol has. Also, we investigated the water-ethylene glycol solution.

Recently, we developed the evaporation model of sessile drop of binary solvent mixture (with infinitely soluble in each other components) based on Hu and Larson solution for single solvent sessile drop and Raoult law for saturated vapor density of components of binary mixture in wide range of undimensional molar binary concentration of the components. (The Raoult law declares that saturated vapor density of first component has a linear reducing dependence on the percent of second component presence in binary solution if this addition of second one is small enough) [2].

In this report we suggest a new physical model of particles ensemble self-assembly in drop of binary solvent mixture as a further development of recently elaborated model for evaporating droplet of pure solvent [3] with account of phenomenological results obtained in our experiments. The computer modeling results are compared with the previously mentioned experiments.

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## DESICCATION OF PARTICLE-LADEN SESSILE DROPS: SIMULATION AND MODELING

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For droplets containing dispersed particles or a solute, a complete mathematical description of processes, leading up to the dried patterns, is extremely difficult since different interconnected process should be considered simultaneously.

Models of evaporation, heat and mass transfer, and crust formation are discussed and compared.

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## **SIMULATION OF TRANSPORT OF CHARGED DROPLETS IN AN ELECTROSPRAY IONIZATION SOURCE OF A MASS SPECTROMETER**

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Electrospray ionization mass spectrometry (MS) is a powerful analytical technique that provides both qualitative and quantitative information in chemical and biological applications. The detection limit of a mass spectrometer is highly dependent on the ion transmission efficiency from the ionization source to the MS inlet. During this process, transport of droplets generated in an electrospray ionization source chamber plays an important role. The droplet transport efficiency is determined by several factors, including the geometric design of the ion source chamber, the gas flow field, liquid flow rate, voltage applied to the ionization capillary, charge status of droplets, volatility of droplets, etc. In this study, numerical simulations of the transport of charged droplets were carried out to predict and thereby further understand droplet transport behaviour in an ion source chamber with a high-velocity plume.

ANSYS Fluent [1] was customized to simulate flow and charged droplet transport. User-defined functions were developed to solve a time-dependent electric field with the transient local charge density updated with a particle-in-cell method. Droplets were tracked using a discrete phase method. Simulations considered the dominant forces on charged droplets and included drag force with a slip correction, Brownian force, thermophoretic force, Saffman lift force and electric force with a real-time space-charge effect. Details about this method can be found in [2].

In this study, two different geometric designs of an ion source chamber, a cylinder and a cylinder with a hemispherical bottom, have been considered to study the effect of nebulizer gas flow field and geometry on droplet recirculation. Both charged and neutral droplets were simulated to study the influences of space charge and electric field on droplet dispersion and transport. Droplet destination and size distribution were then statistically calculated and compared to available experimental measurements.

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## DROPLET RETENTION AND SHEDDING ON SLIPPERY SUBSTRATES

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When liquids come into contact with most solid surfaces, they experience contact line pinning, caused by physical or chemical heterogeneities. This contact line pinning limits the ease at which a droplet can be moved. By imbuing a porous or structured surface and with a lubricant that is immiscible with the working fluid, one can create a slippery liquid infused porous surface (SLIPS). Water droplets on such surfaces have little to no contact with the underlying solid surface and therefore have little contact line pinning or hysteresis. However, the resultant high mobility of the droplets means that accurately controlling and determining the position is challenging<sup>1</sup>. Here we present an experimental study into droplet control and positioning on SLIPS using a very simple microstructure and the capillary force generated by the “Cheerios Effect”<sup>1-3</sup>. We show that the step height, lubricant thickness and initial droplet position determine the angle at which a droplet detaches from a step on a tilted substrate<sup>4</sup>. We characterise both the attractive and repulsive capillary forces and show that the detachment angle for a droplet below the step is dependent on how the droplet arrived at its initial position, which indicates that the system has a memory. Such surfaces could be tailored to specific microfluidic applications.

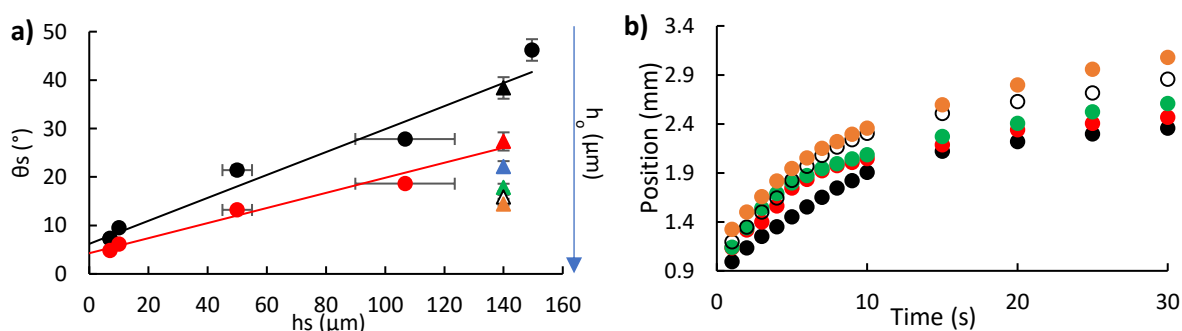


Figure 1: a) Detachment angles given changes in step height and lubricant thickness. b) Repulsion of the droplet from top of step.

ACKNOWLEDGEMENTS: B.V.O would like to thank Northumbria University for funding *via* postgraduate research studentship. The authors would like to thank Dr. Andrew Edwards for assistance during sample production.

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## DROPLET MOTION AND BEHAVIOUR ON THE FLEXIBLE SLIPS (F-SLIPS)

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Controlling the mobility and positioning of liquid droplets interaction with surfaces is crucial for many applications, such as microfluidics, self-cleaning surfaces, inject printing, fog collection etc<sup>1</sup>. Slippery Liquid Infused Porous Surfaces (SLIPS)<sup>2,3</sup> based on Nepenthes pitcher plants, are attractive because they have lubricated surface on which it is easy to move droplets. This ease of movement of droplets on surfaces can come at the cost of droplet control. Since, on a SLIP surface the interaction of the porous structure and the interfacial tensions of the contact line play an important role in the mobility of a droplet, it should be possible to control the velocity of a droplet by changing the parameters of the structured surface<sup>4,5</sup>. In this work we control this by applying a strain to the substrate containing the SLIPS. By increasing strain, at some point, the SLIPS property will break down and the underlying morphology of the surface will halt the moving droplet by pinning<sup>4</sup>. In this work we present a simple method to create a Flexible SLIP Surface (F-SLIPS) with the aim of controlling droplets. Glaco nanoparticles are spray coated on to a flexible material to make them superhydrophobic. Lubricating oil is applied to create slippery surface. The effects of stress and strain on flexible SLIPS is investigated and we investigate how the droplet's motion is effected by the mechanical strain placed on the underlying substrate.

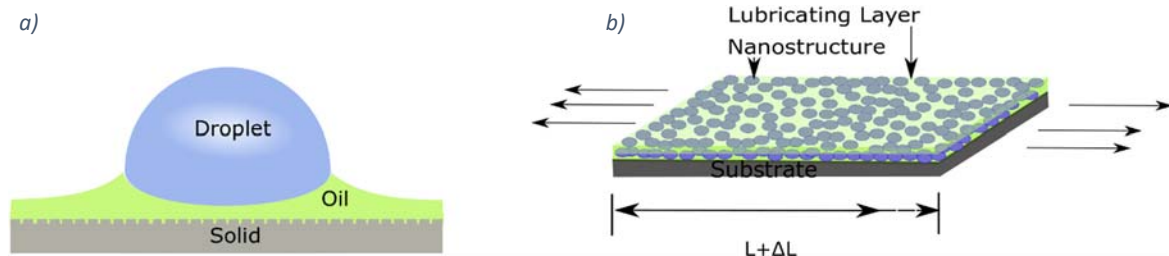


Figure1) a) ) Schematic of droplet on a SLIPS, b) Illustration of a SLIPS with nanostructure and lubricating layer, where upon stretch, the substrate along with nanoparticles and laminating layer goes from the initial length  $L$  to final length  $(L + \Delta L)$

ACKNOWLEDGEMENTS: The authors thank Northumbria University for financial support

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# SELF-PROPELLING LEIDENFROST DROPLETS ON A VARIABLE TOPOGRAPHY SURFACE

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Leidenfrost water droplets can self-propel on heated surfaces with ratcheted topography<sup>1</sup>. For any application, reliability and flow rate are significant concerns for such systems. Here, the effect of the surface profile on reliability and droplet velocity are investigated on a single device with a continuously adjustable millimetre-scale ratcheted surface, see Fig. 1(a). Conditions are described under which the droplet velocity can exceed 350mm/s. The reliability of droplet propulsion is shown to be improved for ratchet geometries with an overhang<sup>2</sup>.

In addition to the experiments, Computational Fluid Dynamics (CFD) simulations are performed to investigate the role of ratchet geometry in droplet and CO<sub>2</sub> puck propulsion, see Fig. 1(b). Vapour flow and resulting forces are computed for a CO<sub>2</sub> puck with a flat base held at fixed distances above the ratchet sawteeth. Overhang is found to reduce the upward vertical force on the droplet by providing an escape route for the vapour with lower impedance, while also reducing the area under the droplet with vapour flow in the reverse direction; both factors leading to enhanced droplet propulsion. The full paper will provide experimental and CFD results that enable description of the phenomenological changes in fluid flow resulting from geometry changes in mm-scale triangular ratchets.

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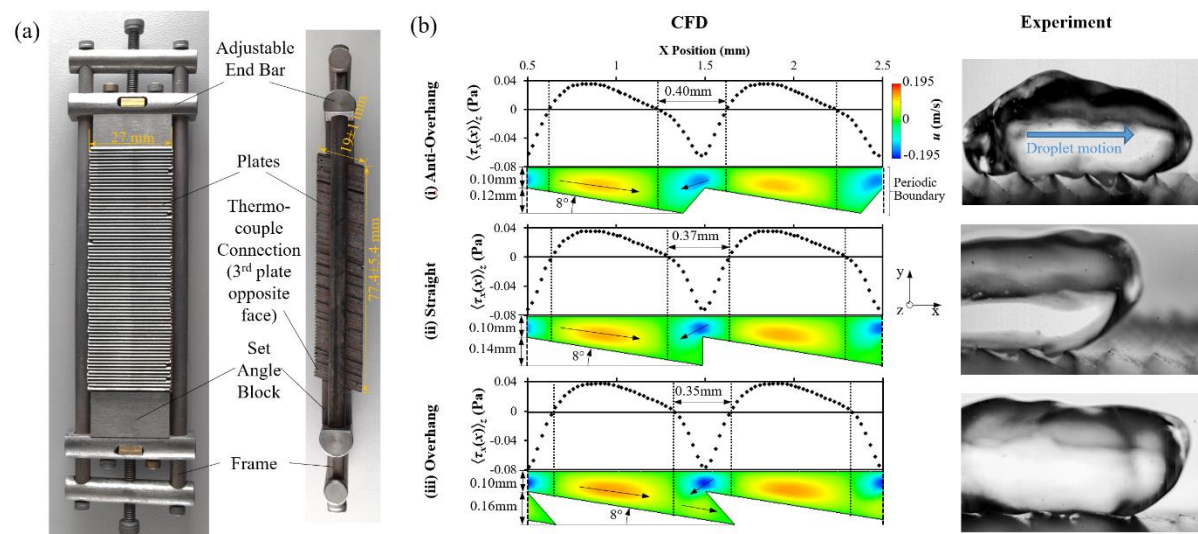


Figure 1: The device is formed of 1mm thick plates each with an initial height of ~20 mm. (a) Plan and side views of the device. (b) CFD and high speed video experimental results for representative ratchet geometries, adapted from [2].

# UNIFIED THEORY FOR ANISOTROPIC DROP GROWTH ON LINEAR PATTERNS

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## Abstract:

The development of anisotropic surface features appears to be an evolutionary advantage achieved by living organisms, allowing a more efficient collection, control, or disposal of liquids. Inspired by the rich spectrum of natural microstructures, artificial patterns of linear topographic and chemical surface features have been proposed to control anisotropic spreading of liquids. Despite the number of experimental and theoretical studies, to date, our understanding of the interfacial instabilities governing anisotropic wetting is still scattered. In this talk I will present a unified theory[1] to describe the shape evolution of liquid drops on anisotropic surfaces, under to volume variations, i.e during condensation and evaporation.

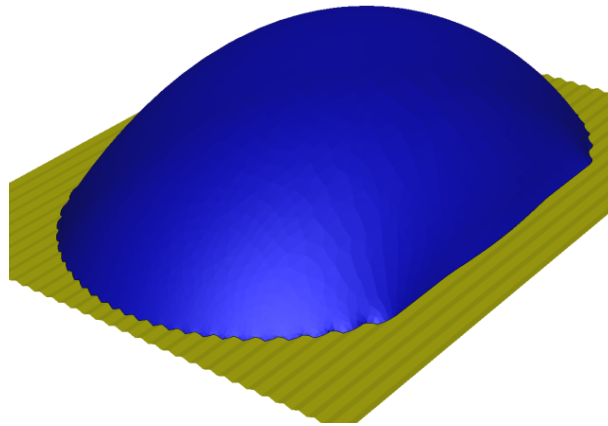


Figure 1:

Numerical calculation of a metastable state of a drop on a linear V-groove pattern.

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# NON NEWTONIAL SLIPPERY LIQUID INFUSED POROUS SUBSTRATES

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## **Abstract:**

Inspired by the Nepenthes pitcher plant, SLIPS (Slippery Liquid Infused Porous Substrates) have been recently introduced. They enhance the liquid mobility due to the presence of a lubricating film between liquid and solid, reducing the contact to solid surface and therefore the driving force required for liquid manipulation. The development of SLIPS leads to applications in areas such as food packaging and biomedical devices, which involve fluids with complex rheological properties. In this contribution I will discuss potential implications of complex fluids in liquid flow on SLIPS.

## Gel optical design with Autonomous Focal Shifting for future Ocular applications

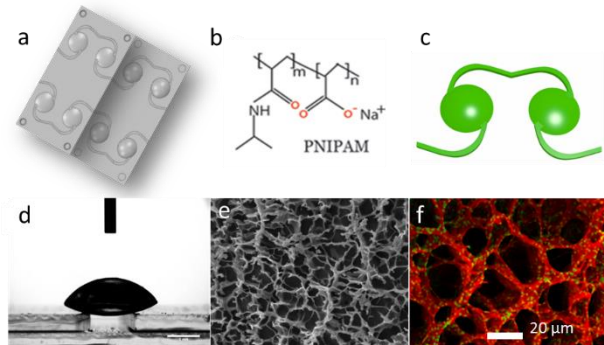
A. Sun<sup>1</sup>, S. Sridhar<sup>1</sup>, D. Wang<sup>1</sup>, Y. Liu<sup>2</sup>, H. Lv<sup>2</sup>, B. Xu<sup>1</sup>

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Stretchable and autonomously self-adaptable transparent materials with elastohydrodynamic character are of high fidelity in tuneable optics and soft robotics research areas [1]. To date, it continues to be a huge challenge to synthesize a highly tuneable liquid based optical system integrating autonomous, dimensionally stable, vari-focal length, and minimal mineral deposition. Moreover, the opto-system's shape should optimally fit the individual test subjects with reduced geometrical aberration and improving image resolution [2]. This work describes the fabrication of intraocular lens using smart hydrogel system combining a soft matrix made of Poly (Acrylamide-co-sodium acrylate) with controlled swelling kinetics and mineral deposition through a preserving solution made of Phosphate Buffer Saline (PBS). The soft matrix has the advantage of being multifaceted to tunability, inexpensive, simple to actuate. We combine experiments, finite element analyses; to design a type of soft mechanical metamaterials that switches its flexibility and design ability to build diverse architectures for achieving various responsive deformations with desired isotropic/anisotropic features. The Young's modulus ( $50 \pm 3$  KPa) and swelling ratio of not more than 5% have also been characterized experimentally as the PBS concentration is increased from 0.01 M to 0.2 M. By introducing chemically tunable stimuli-responsive hydrogel system, a spatially homogenous designed system exhibited a controlled anisotropic swelling and bidirectional shrinking for more than 20 days. We showed that the fabricated hydrogel is also highly transparent, with refractive indices ranging from 1.42 to 1.45 and porosity of  $\sim 14 \mu\text{m}$ . The storage solutions leverages a switchable configuration to influence the swelling deformations of hydrogel to attract less deposition of calcium ions inside the system, thereby resulting in system stability and improved optical transparency for 30 days. Our method discussed above, could significantly promote ionic swelling induced morphological transformation with autonomous response to changing ionic environment intraocular lens by opening new perspectives on practical tuneable optic systems.



**Figure 1:** (a) IOL design mould; (b) The chemical composition for PNIPAM gel; (c) illustration of IOL design; (d) curve data of the design 4 IOL measured by droplet shape analyser (DSA), diameter: 5.669 mm; (e-f) SEM and EDS figures of IOL samples swelling in PBS (0.1M) and store in calcium solution (0.5mM) for 3 days.

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## Smart surface enabled thin layer heterogeneous responsive soft material patterning

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Droplet microfluidic technologies utilizing smart surface with wettability control (e.g. switchable hydrophobic/philic patterns) have shown great promise in thin, uniform bio-content layer patterning previously reported by [1]. Recently, it has also been applied in controlling the formation of encoded multifunctional as heterogeneous hydrogel building blocks have been exploited to form complex hydrogel architectures [2]. Such heterogeneous soft material structures will greatly expand the areas such as the actuators, robots and shape morphing functional patterned soft devices [3, 4, 5, 6].

This work has demonstrated the design, fabrication and characterization of well-defined controllable morphing technology by patterning and controlling the variform of hydrogel droplets onto hydrophobically patterned surface, which to achieve the advanced and complex 3D morphing structures. The investigation considers deposited different composites of functional hydrogel droplets (with swelling ratio mismatch) onto patterned hydrophobic surface (a “two-parallel plate” configuration, shape-controlled by hydrophobic boundaries) to generate the heterogeneous layer (fig. 1). For reconfigurable morphing structures of dynamically shape change, we have generated 3D morphing deformation (fig. 2) responding to discrete levels of stimulation inputs. We believed that the achieved complex multiple morphing devices should hold promise for future applications of biomedical structures, microfluidics, soft engineering and biomimetic systems.

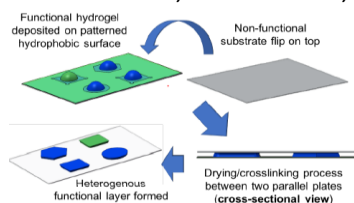


Figure.1 Schematic view of the heterogeneous hydrogel structure patterned by hydrophobic/philic surface

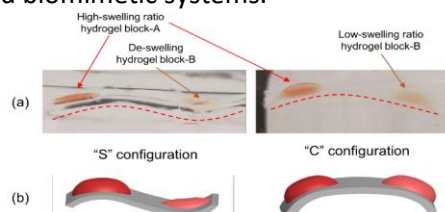


Figure.2 (a) microscopic photos; and (b) schematic illustration showing hydrogel structure shape reconfiguration from "S" shape to "C" shape

**ACKNOWLEDGEMENTS:** The authors would like to thank the support from EPSRC UK Fluid Network (EP/N032861/1).

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# Investigation of equilibrium droplet-shapes on chemically striped patterned surfaces using phase-field method

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## Abstract

Droplet wetting on chemically patterned surfaces is ubiquitous in many applications including inkjet printing, coating, and microfluidics. Due to multiple local free energy minima caused by surface inhomogeneities, the equilibrium droplet-shapes on chemically patterned surfaces are challenging to be predicted. Here, we systematically investigate the equilibrium shapes of droplets on chemically striped patterned surfaces by using a free energy minimization method. The chemically striped patterned substrates in the present consideration are functionalized with stripes of the same width but two different wettabilities. The influences of droplet volume  $V$ , deposition position, and stripe width  $d$  are explored. It is found that the droplet aspect ratio  $\xi$  (ratio of base radii in the direction parallel and perpendicular to the stripes) as a function of the scaled stripe width  $d/V^{1/3}$  shows a periodic oscillation behavior. This interesting observation is the same as the “stick-slip-jump” moving fashion of a slowly condensing droplet on a chemically striped patterned surface. According to the number of the wetted hydrophilic stripes, the diagram of  $\xi$  versus  $d/V^{1/3}$  is divided into six areas. Within each area, the droplets have two different equilibrium shapes, whereas on the boundaries, the droplets have a unique equilibrium shape. Additionally, by comparing the movements of slowly evaporating and condensing droplets, we have observed a hysteresis phenomenon (different  $\xi$  though the same value of  $d/V^{1/3}$  and the same deposition position), which is attributed to the difference of the receding and advancing contact line movements.

ACKNOWLEDGEMENTS: The authors gratefully acknowledge funding of the research through the Gottfried-Wilhelm Leibniz prize NE 822/31-1 of the German research foundation (DFG). Applications of the developed modeling methods for wetting behavior on structured surface are considered within the VirtMat project of the Helmholtz association.



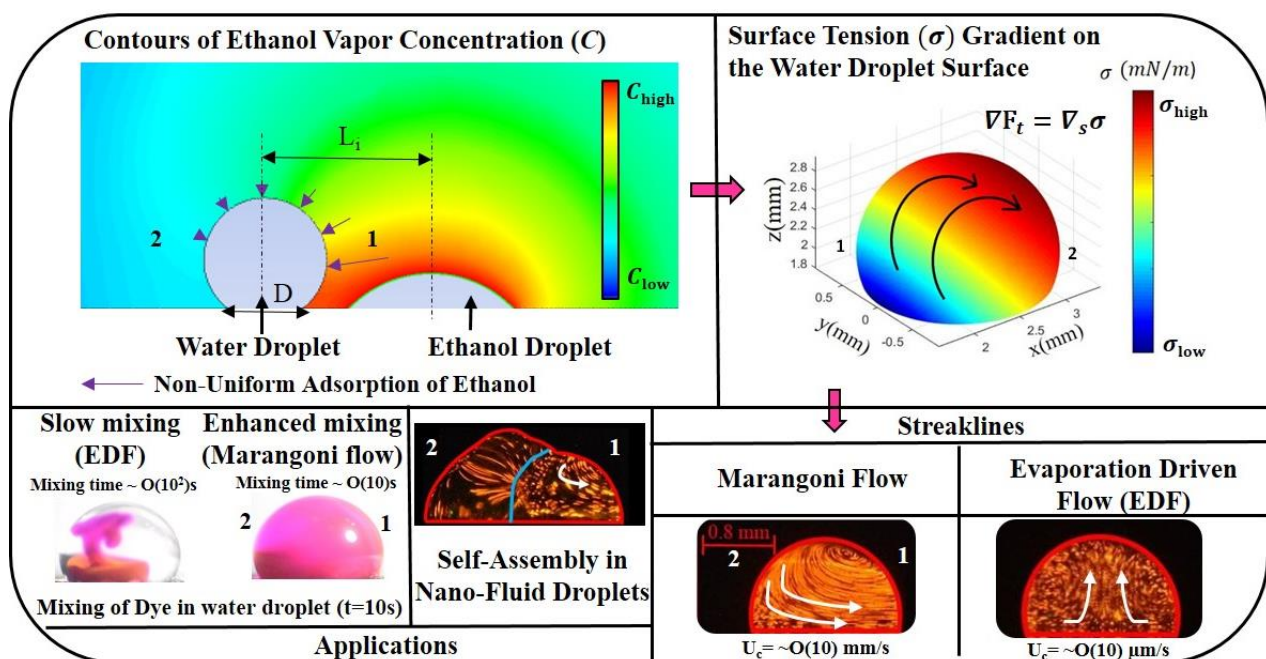
# ALTERATION OF FLOW INSIDE MICROLITRE SESSILE DROPLETS USING VAPOR MEDIATED INTERACTIONS.

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The Study of flow inside sessile droplets has ramifications in diverse range of applications such as lab-on-a chip medical diagnostics, self-assembly and surface patterning. The evaporation induced flow inside microscale droplets is very low ( $U_c \sim O(10) \mu\text{m/s}$ ) owing to its low Reynolds number. The low velocity poses a major bottleneck in mixing reagents while implementing droplets as platform for bioassays. We present a methodology to enhance mixing inside droplets without affecting its global evaporation pattern. We strategically place an ethanol droplet adjacent to water droplet. Evaporation of ethanol creates an asymmetric vapour field around the water droplet. Asymmetric adsorption of ethanol vapour on water droplet creates a surface tension gradient, which induces Marangoni advection inside the water droplet. The flow induced due to Marangoni convection is  $\sim O(10^3)$  times higher than the naturally evaporating droplet, resulting in enhanced mixing. Using vapour mediated flow control mechanism, morphological control of buckling instabilities is achieved in Nano-Fluid Droplets.



**ACKNOWLEDGEMENTS:** The authors thank the Ministry of Human Resource Development (MHRD), Government of India and Department of Science and Technology (DST), Government of India for their generous funding and support.

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## Flow structure of marangoni-contracted sessile droplets of water-diol mixtures

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A droplet consisting of two miscible liquids, placed on a smooth high-energy surface, should spread until it completely wets the surface. However, if one of the components has a higher volatility and surface tension than the other, a quasi-stationary wetting behavior with non-vanishing apparent contact angle can be observed [1]. This behavior is known as "Marangoni-contraction" since it is caused by Marangoni flows toward the center of the droplet, driven by the enrichment of the residual component near the contact line and the associated surface tension gradient. A hydrodynamic-evaporative model predicts that the quasi-stationary state is caused by a balance between Marangoni and capillary flows [2]. It also predicts a power law between the apparent contact angle and the ambient humidity that closely matches to experiments. Nonetheless, this explanation differs from the model in [1], where the precursor film around the droplet contributes to the contraction. Recent work also suggests an impact of buoyancy-driven flows even for small droplets [3]. Resolving the internal flows is therefore essential for discriminating between the contributing mechanisms. Using high-resolution  $\mu$ PIV and simultaneous shadowgraphy of the droplet contour, we study the relation between the flow inside the droplet and its apparent shape. We use aqueous solutions of various short chain carbon diols as a model system in which the surface activity varies while most other properties remain similar. Depending on the surface activity of the diol, its concentration, and the ambient humidity, we observe different flow patterns, indicating that multiple mechanisms are related to the apparent contraction, which remained hidden in a sole observation of the apparent contact angle.

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## DOCTOR BLADE TECHNIQUE AND WETTING DYNAMICS

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The controlled deposition of nanoparticles to form ordered 2D patterns free of defects is a challenging topic with myriad of applications as antireflection coatings [1], plasmonic surfaces [2], electronic [3], etc. However, the process throughput and the scalability are limited for real industrial applications.

Recent studies [4] have shown the viability to deposit nanoparticles with great spacial precision on patterned surfaces by using the doctor blade method where a meniscus of a liquid containing the nanoparticles is forced to move accros a patterned substrate. The combination of the resulting flow, the wetting dynamics at the receding contact line, the contact line pinning plus the evaporation which confines the particles to a small subvolume leads to the nanoparticles deposition.

However, in order to optimize this method and to increase the velocity of the particle deposition it seems necessary a better understanding of the different mechanisms acting at the receding contact line.

In this work we use large-scale molecular dynamic simulations to model the doctor blade technique at the nanoscale. We focus in the study of a pure liquid confined between two plates. The moving of the top plate at constant velocity induce a flow and the displacement of the liquid along the bottom plate where we introduce different chemical and physical heterogeneities. This simple system allows us to analyze the dynamics of the different contact angle of the system as well as to study the contact line pinning on the different heterogeneties. We develop a theoretical model to predict the dynamic contact angles as a function of the affinity between the liquid and the different plates and the velocity of the top plate. Finally, we have studied the mechanism behind the pinning of the receding contact line at the nanoscale as well as the posibility of controlling the pinning time by tunnig the wettability of the substrate. The results obtained open the door to optimize the doctor blade technique for a faster nanoparticle deposition process.

**ACKNOWLEDGEMENTS:** This research was partially funded by UMONS. Computational resources have been provided by the Consortium des Equipements de Calcul Intensif (CECI), funded by the Fonds de la Recherche Scientifique de Belgique (F.R.S.-FNRS) under Grant No. 2.5020.11

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## WETTING ADAPTATION AND CHARGE SEPARATION AT THE INTERFACE BETWEEN POLYMER SURFACES AND ROLLING DROPS

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Separation of electrical charge in water is a long-observed phenomenon exemplified by the dramatic sparks of the famous Kelvin Water Dropper. This effect is witnessed, often with some grief, in the charging of aerosols produced by medical nebulizers and in the breakdown of semiconductor chips rinsed with ultra pure water [1]. A number of groups have begun to decipher the reasons behind the charging of water upon contact with substances such as plastic or glass, accrediting hydroxide ions and their affinity for electronegative surfaces [2]. However, the literature is fairly sparse and the topic is highly charged. Much of the literature does not distinguish between charging from drop impact on a surface, and that from continued rolling. Additionally, very little has been written about the timescales involved in electrical surface adaptation to a series of water drops. We have developed a method for studying the individual charge from many thousands of drops rolled successively across a substrate. On thin insulating layers of OTS (Octadecyltrichlorosilane), as well as on thick insulating layers of Teflon and PDMS, we observe temporal surface adaptation that is dependent on such variables as drop rate, drop velocity, and roll distance. With the obvious prevalence and importance of water droplets in science and in society, a more thorough understanding of this topic could lead to advances in fields as wide and varied as adaptive wetting, tribocharging, electrochemistry, and renewable energy.

ACKNOWLEDGEMENTS: The authors thank ERC Grant no. 340391 Supro for supporting this work.

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doi:10.1002/anie.200701812

## CAPILLARY TRANSPORT OF DROPLETS ON 3D PRINTED CONICAL STRUCTURES

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In most arid regions like deserts, plants and animals have developed specific strategies (such as spines) in order to survive long periods without water [1]. Recent works [2] have proposed that conical shapes could trigger the motion of droplets towards the base. We show that it is possible to achieve such capillary transport with macroscopic 3d printed conical structures. Figure 1 shows a superposition of pictures emphasizing this motion of large droplets on these cones. We present experimental results as well as a model for describing the dynamics. The results obtained herein could be used to develop smart ways for manipulating droplets.



FIGURE 1 – Superposition of successive pictures emphasizing the motion of a large droplet on a 3d printed conical structure. The motion is shown at time  $t = 0s, 2s, 7s$  and  $14s$ .

ACKNOWLEDGEMENTS: The authors thank the WISD-FNRS program (Brussels, Belgium) for supporting this work.

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# RESPONSE OF SESSILE DROPLET TO A SINGLE ELECTRICAL WAVE PERTURBATION

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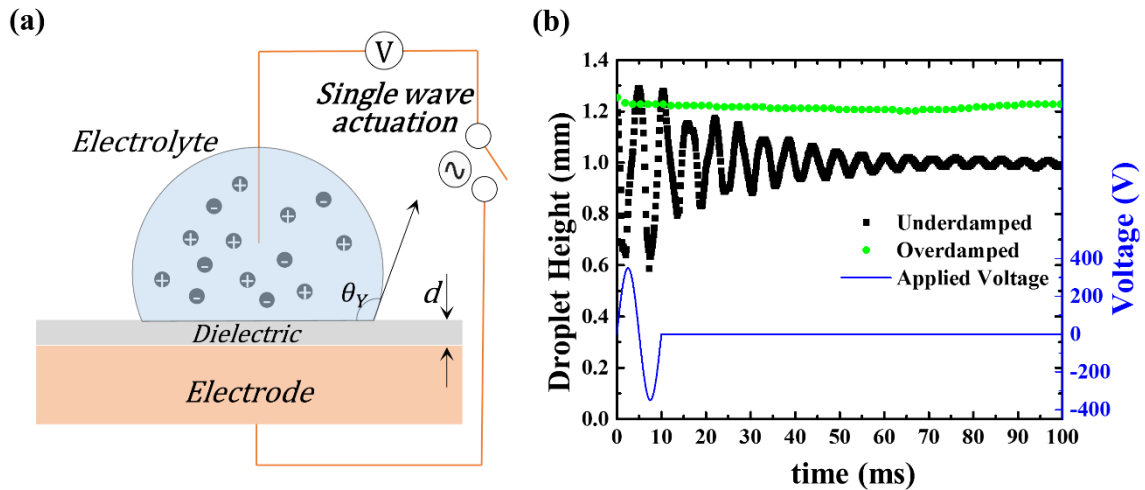


Figure 1. (a) Schematics of the electrowetting setup (b) Droplet height vs. time plot for drops with viscosities of 0.91mPas (underdamped) and 121.71mPas (overdamped), respectively, along with the single sine wave (350V and 100Hz) perturbation in second vertical axis.

A sessile droplet attains an equilibrium on a solid surface based on the magnitude of the solid surface energy and interfacial tension between the liquid and surrounding medium. In a realistic scenario, the wettability of the droplet can be disturbed by means of external perturbing force such as electric field [1]. However, until now it has not been reported the responsive behavior of the drop to a single or series of perturbation waves. In this work, an experimental mechanism was designed to create a controlled perturbation or disturbances in a sessile droplet by applying an external electric field which induces the corresponding changes in the drop shape. Single sine wave of an AC voltage with different frequencies and voltages were applied and the drop dynamics was investigated to scrutinize the drop response while regaining its equilibrium configuration. High speed imaging of a sessile drop disclosed that the drop response varies significantly according to the single wave characteristic (amplitude and frequency) and the properties of the drop (density, surface tension, and viscosity). The drop shape response, after the actuation of the external field, resembles the inertial capillarity phenomenon [2], therefore the role of drop and medium viscosity is studied in detail. Experimental observations permitted us to propose a model, for the variations in the height of the sessile drop, based on a mass-spring-damper system. The damping ratio of the system was observed to be a function of the Ohnesorge number, i.e., viscous, surface tension, and inertial forces in the system.

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# Wetting of non-equilibrium liquids

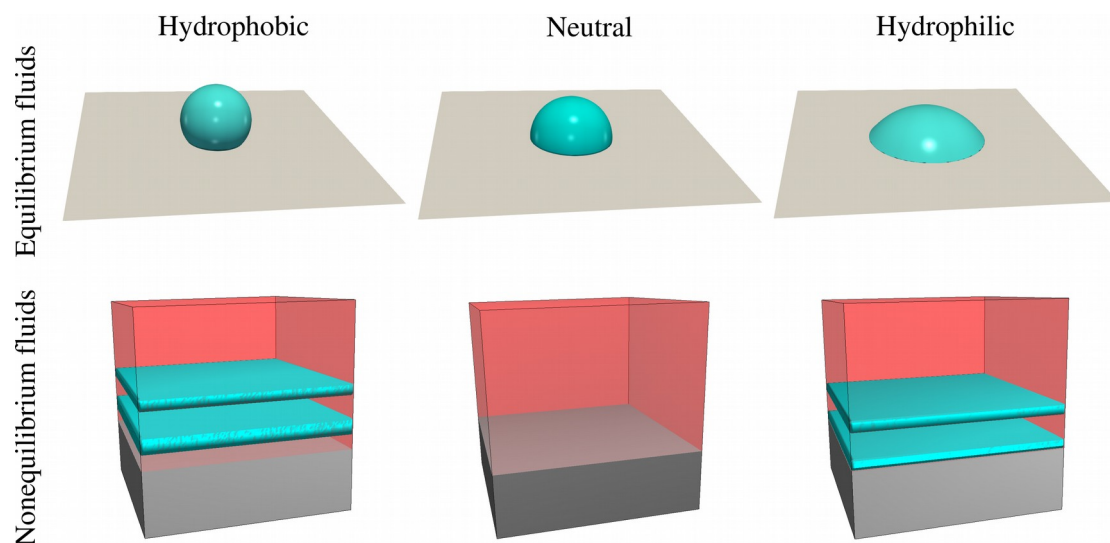
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Wetting is an omnipresent phenomenon in nature which has a great impact on materials design. The classic Young's law proposed in 1805 addressed the static contact angle of homogeneous droplets sitting on a substrate, such as beads of dew drops clinging on leaves. However, in many realms of advanced technological applications, this groundbreaking tenet loses its validity since droplets are mostly non-static and include compositional inhomogeneities engendering diffusion. Here, we demonstrate that when diffusion is taken into account, the static contact angle deviates considerably from Young's law. Most importantly, we find that a hydrophilic liquid spontaneously transforms into a hydrophobic one, being ascribed to a vying effect between wetting and diffusion. We anticipate that the present finding can be applied to fabricate ultra hydrophobic materials.



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## DROPLETS ON BENT FIBERS

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Droplets trapped on fibers are part of our everyday lives : dew droplets appear on spiderwebs, rain drops are hanging under cypress leaves, water droplets are collected by cactus spines. Looking at these phenomena, scientists may wonder what is the maximum amount of fluid that a fiber can hold. Lorenceau et al.<sup>1</sup> determined the critical volume of a water droplet hanging on a horizontal fiber. Here, we address a similar question : we try to find out the maximum volume that a bent fiber can hold<sup>2</sup>. We focus on this geometry because, in nature, some specific plants are able to hold large rain droplets thanks to their Y-shaped leaves. We try to mimic these structures using nylon fibers. It turns out that a bent fiber can hold significantly more water than a horizontal fiber. We determine the critical water volume for different fiber diameters and different angles. Finally, we propose models that are able to predict the maximum droplet size and that could be implemented in future fiber-based microfluidic devices.

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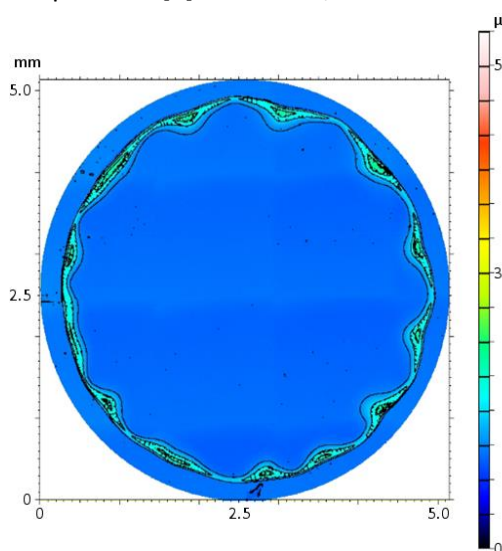
# EXPERIMENTAL INVESTIGATION OF WETTING OF SUBSTRATES WITH THIN SOLUBLE POLYMER COATINGS AND EVAPORATION-DRIVEN SURFACE RESTRUCTURING

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Spreading and evaporation of solvent drops on soluble polymer surfaces is governed by surface energies of substrate and solvent, as well as by the convective and diffusive transport of substrate molecules in the solvent and of solvent molecules in the substrate [1, 2]. During the processes of wetting and subsequent evaporation the substrate surfaces are restructured by diffusive and convective transport of polymer molecules towards or away from the contact line. Depending on the relation between transport mechanisms different structures can be observed after complete evaporation [3]. Until now, this relation has been studied by varying molecular masses of substrate



1: Polystyrene surface topography after spreading and evaporation of a toluene droplet.

molecules as well as choosing different liquids (good and poor solvents). The experiments have been performed with thick substrates. However, the influence of the gas environment on the spreading process of apolar solvents and the resulting surface shape has not been studied systematically.

In this work the spontaneous spreading and evaporation of solvent droplets on thin spin-coated polystyrene films with different molar masses are studied in a closed chamber under a dry or solvent loaded nitrogen atmosphere. The experiments are conducted with different solvents. The spontaneous spreading of droplets is recorded using shadowgraphy. The surface topography is characterized after complete solvent evaporation using a confocal microscope.

It has been found that the molar mass of the polymer substrate has no effect on the initial wetting behaviour. However, our findings suggest that there exists a

relation between the later stages of wetting and the solubility of the substrate which has not been reported in the literature. The formation of structures during solvent evaporation depends strongly on polymer molar mass and shows distinct characteristics like the instabilities shown in figure 1.

**ACKNOWLEDGEMENTS:** The authors thank the Deutsche Forschungsgemeinschaft (DFG) for supporting this work.

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## CHARACTERIZING THE WETTING PROPERTIES OF SOFT POROUS MATERIAL USING A WASHBURN-LIKE APPROACH

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Lubricating grease is a two-phase soft material consisting of a thickener matrix that provides mechanical strength and mobile base oil. The matrix acts as a reservoir that slowly releases the oil during years of bearing operation. Despite the fact that the reliability of most mechanical machinery crucially depends on the lifetime of bearings and their lubrication, the process of oil retention and release by the grease matrix is very poorly understood<sup>[1]</sup>.

In this project, we facilitate the oil-release by extracting the oil from the grease using filter paper. Taken into account the heterogeneity of the pores sizes in the paper<sup>[2]</sup>, the mass of oil imbibed by the paper is quantified by measuring the transmitted light intensity<sup>[3]</sup> using a modified Lambert-Beer relation. The imbibition timescale from the grease is found to be of 2 orders of magnitude longer than that of a drop of the same base oil. The slow oil release results from the high affinity between the grease matrix and the oil, leading to a pressure gradient ( $\sim 5\text{kPa}$ ) opposite to the direction of the extraction flow. To quantify this wetting affinity, we developed a Washburn-like model in which both grease and paper are described as porous media. The creeping oil flow is driven by the difference in wetting properties of the grease matrix and the paper. Furthermore, we found that the imbibition dynamics depends on the oil content of the grease. Even a slight increase ( $\sim 0.2\%$ ) of the wetting energy volume can lead to an increase of a factor 2 in the timescale of imbibition.

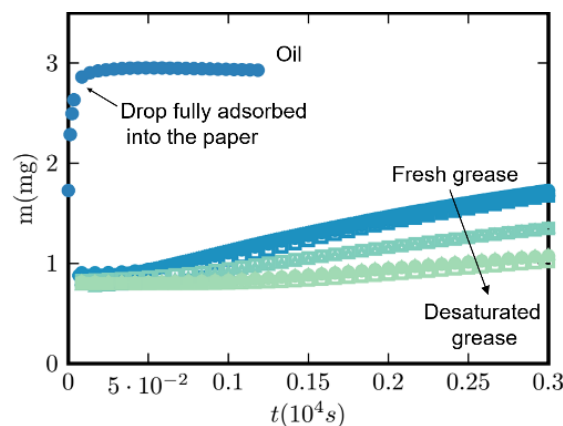


Figure 1 The amount of oil adsorbed in the paper versus time from an oil drop and from de-saturated grease patches (from top to bottom the saturation level decreases from 100% to 65%)

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## Molecular dynamics simulation of water droplet wettability on graphite substrate

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Surface wetting by water droplets has been studied in many disciplines, for example, designing water-resistant fabrics in the textile industries (Xue, 2014); contact lens, worn by approximately 125 million people worldwide, from medical prospect (Menzie, 2010); in the oil industries, wettability plays a significantly role in the field of enhanced oil recovery (EOR), as it controls the oil and water phase flow properties in the complicate pore networks, i.e., and one of approaches to optimize oil recovery is to change the wettability of fluid rocks (Sohal, 2017).

Wettability of water droplet on carbon composed substrate relates to hydrocarbon extraction from the unconventional formation, such as shale or coal bed methane. Normally, wettability is characterized by macroscopic contact angle, however it is controlled by molecular scale interaction force. In this work, Through the molecular dynamics, the contact angle of water droplets over flat and rough substrates is studied. For the roughness, the grooves sizes, fractions and heights dependences were considered. The effect of thermal agitation of substrate particles and water droplet sizes on contact angle are also investigated in this article with a relationship between microscopic and macroscopic contact angle shown and our inferred macroscopic water droplet contact angle ( $79.54 \pm 1.15^\circ$ ) over flat substrate based on simulation results (figure 1) are highly consistent with the value of  $79.3^\circ$  from experimental and theoretical analysis work on the water contact angle on chemically pure graphene (Li, et al., 2013). In addition to, we developed a series of MD simulations for Water-Methane-Graphite system, which allow us to investigate the effect of the intrusive component (methane) on the water droplet contact angle, through which it is seen that the water droplet contact gradually decreases with increasing the amount of methane until the droplet reaches the state where droplet lift-off the substrate (figure 2) from the state of solid-liquids adhesion.

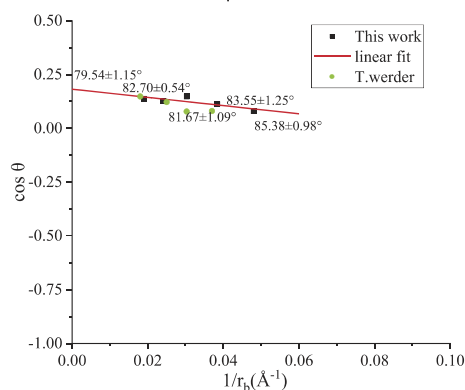


Figure 1 Cosine of the contact angle as a function of the droplet base curvature.

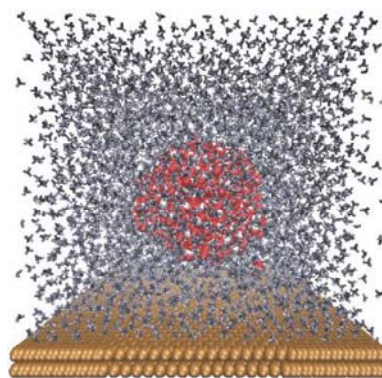


Figure 2 Equilibrium state of a water-methane-graphite system at the droplet lift-off state.

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# EVAPORATION FREE SELF-TRANSPORTATION AND MANIPULATION OF AQUEOUS DROPLETS OVER AN OIL-LADEN DIVERGING GROOVE

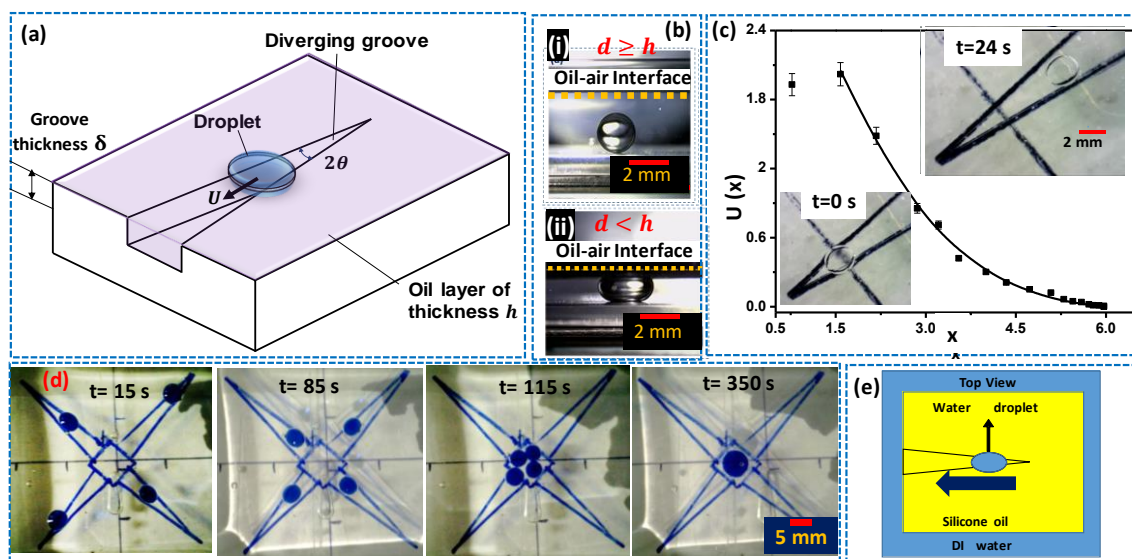
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Self-transportation of the droplets has received tremendous attention in the recent past owing to the rapid advancement in various lab-on-a-chip and point of care microfluidic devices<sup>1</sup>. There are inherent limitations associated with the existing various active<sup>2,3</sup> (magnetic- contamination due to the magnetic nanoparticles, electrical and acoustic- peripheral setup for the actuation) and passive<sup>4</sup> (requirement of temperature and concentration gradient) droplet transportation techniques. Thus there is a need to develop a more efficient droplet manipulation technique. Herein, we report a mechanism that is free of evaporation, pinning and contamination for the transportation and the manipulation of aqueous droplets over an oil-laden diverging groove.

**Fig. 1a** depicts the schematic of the mechanism comprising the diverging groove of groove angle ( $2\theta$ ) and groove thickness ( $\delta$ ) submerged in an oil-laden (silicone oil,  $\rho_{oil} \sim 995 \text{ kg/m}^3$ ) surface of thickness ( $h$ ). Upon dispensing an aqueous droplet of diameter ( $d$ ) on the converging end, the interfacial properties [ $S_o > 0$ , where  $S_o = \gamma_w - (\gamma_o + \gamma_{wo})$ ], where  $S_o$  is the spreading parameter,  $\gamma_w$ ,  $\gamma_o$  and  $\gamma_{og}$  are the surface and interfacial tensions of water, oil and water/oil respectively] facilitates the droplet to enter into the groove to minimize the energy even though the density differences between the water and oil are negligible. Depending on the thickness of the oil layer, the droplet can either get deformed ( $d > h$ ) (by getting compressed and expanded in the vertical and lateral directions respectively) or remain undeformed ( $d < h$ ) as shown in the **Fig 1b**. Upon entering into the oil phase, the droplet also tends to enter the slot (for  $d > h$ ), and get transported through the groove owing to the differential Laplace pressure as the radii of the curvature at the leading and the trailing edge of the droplet becomes unequal due to the diverging nature of the slot. **Fig. 1c** shows the migration of a droplet ( $d \sim 2.7 \text{ mm}$ ) into the diverging groove with an oil-layer thickness of  $1.5 \text{ mm}$ . **Fig. 1d** shows the coalescence of four droplets ( $d \sim 3.9 \text{ mm}$ ) dispensed simultaneously at the converging end of the four-branch star groove network. The proposed droplet transport mechanism can also be used for the extraction of the aqueous droplets from oil as shown in the **Fig.1e**. In conclusion, our simple, facile and passive technique could find applications in droplet manipulation on open surfaces microfluidics, oil-water separation and other biological applications.



**Fig. 1** (a) Schematic of the droplet transport mechanism (b) Shapes of water droplet ( $d \sim 2.7 \text{ mm}$ ) over an oil layer thickness of (i)  $4 \text{ mm}$  and (ii)  $1.5 \text{ mm}$ . (c) Variation of velocity  $U(x)$  with  $x$  for  $d = 2.7 \text{ mm}$ ,  $2\theta = 15^\circ$ ,  $\delta = 1 \text{ mm}$  and  $h = 1 \text{ mm}$  (inset: time-lapsed images of the droplet transportation) (d) Coalescence of droplets dispensed simultaneously at the converging ends of a four-branch star-shaped network (e) Schematic of the mechanism for the extraction of aqueous droplets from oil.

**Acknowledgements:** We thank NCCRD, IIT Madras for extending their facilities for surface tension and interfacial measurements.

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\*equal contribution



## Effect of Surfactants on Drop Coalescence at Liquid/liquid Interfaces

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In this work, the effect of surfactants on the coalescence of a drop with an initially flat aqueous-organic interface was experimentally investigated. As shown in Figure 1, a high-speed Particle Image Velocimetry (PIV) system was implemented to obtain the flow fields inside the coalescing droplets. The aqueous phase was a 78% glycerol solution and the organic phase was Exxsol D80 oil. Different amounts of Span 80 dissolved in the oil phase with surfactant-oil mass ratios below the critical micelle concentration, up to  $\phi = 5 \times 10^{-4}$  were investigated. When the drop impacted on the interface, an oil film was trapped between them. It was found that an increase in the surfactant concentration promoted the deformation of the interface before the film ruptured. A high surfactant concentration also increased the time needed for film rupture. When rupture occurred, two counter-rotating vortices formed inside the droplet on either side of the rupture point, which moved upwards with time. The propagation of the vortices inside the droplet was faster for low surfactant concentrations, while the intensities of the two counter-rotating vortices significantly decreased for increasing surfactant concentration. At the early stages of coalescence after film rupture, the kinetic energy per unit mass was mainly distributed near the bottom part of the droplet, while at later stages it was distributed near the upper part of the droplet.

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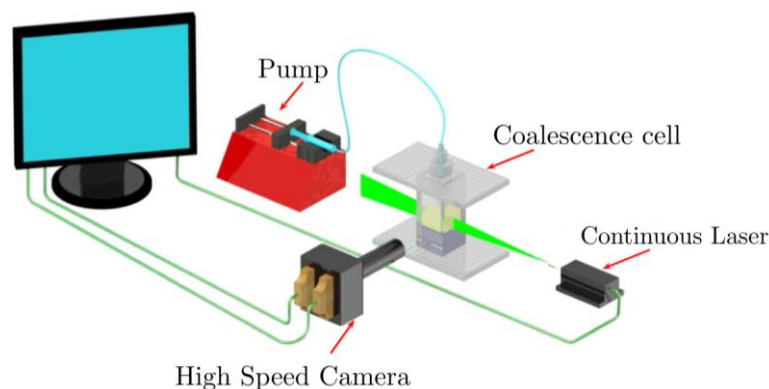


Figure 1 Sketch of the experimental set-up and the PIV system used to investigate the coalescence process

## High Resolution Inkjet Printing of 2D Materials

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Graphene was the first reported 2D material and created huge interest in the field due to its unique material properties.[1] Since then a wider range of 2D materials have been developed including insulators (hexagonal boron nitride h-BN) and semiconductors (MoS<sub>2</sub>).[2] Combining sheets of different materials into stacks, held together by van der Waals forces, forming van der Waals heterostructures allows combinations of the properties of the 2D materials, producing defined properties on-demand determined by the ordering, number and type of sheets in the stack.[3] A number of methods to produce van der Waals heterostructures are under development including: direct mechanical assembly, CVD growth, epitaxy, liquid phase formation and printing[2]–[5].

Inkjet printing offers the potential to mass produce structures at relatively low cost since 2D material flakes can be readily dispersed in inks without loss of their unique properties. In particular, it is possible to develop fine control of the amount of material deposited per unit or droplet area on the substrate. At very low flake number densities single flake per drop printing conditions are possible which, if the difference between the flake area and the droplet area is small enough, will give a high spatial resolution for flake deposition. Subsequent overprinting of drops would then allow flake on flake printing and production of heterostructures. At higher number densities, percolating lateral networks or hole free (< 1%) coverage of a surface could be achieved using a minimum of print passes and material, giving greater control of print output.

Here, the definition of a printing parameter space allowing single flake per drop printing is described. This predicts the concentration of 2D materials required for complete coverage of a substrate, treating flake deposition as independent random events. These are tested experimentally, using Dimatix and Super Inkjet Printers to print inks of 2D materials in the drop volume range from 10 pL down to 1 fL.

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