Droplets 2019
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EXPLORING THE DISSOLUTION LAG EXPERIENCED BY DRIED PARTICLES ON INHALATION IN THE AEROSOL PHASE USING SINGLE PARTICLE MEASUREMENTS

N. C. Armstrong Green[1], A. Haddrell[1] , J. Reid[1] , D. Lewis[2] and T. Church[2]

[1]School of Chemistry, University of Bristol, Bristol, BS1 TS, United Kingdom

[2]Chiesi Limited, Bath Road Industrial Estate, Chippenham, Wilts, United Kingdom

natalie.armstronggreen@bristol.ac.uk

Dry powder inhalers (DPIs) have become a popular inhalation delivery system due to good drug stability and a minimal need for patient coordination. The powdered mixture consists of large, course carrier particles, e.g. lactose monohydrate, and an active pharmaceutical ingredient (API). Given that the efficacy of the drug is dependent on where the dose is delivered, it is important to understand the physico-chemical properties of the aerosol to predict recieved dose.

Once inhaled, the dry particles experience a warm, humid environment where, depending on their solubility properties, can take up water. A particle that fully dissolves in the aerosol phase will grow during inhalation, the magnitude of growth is controlled by its hygroscopicity. This dynamic size change influences the deposition mechanism, thus the deposition fraction. A particle that partially dissolves or remains solid, could potentially lead to a different deposition pattern depositing on the lung surface in a different physical state.

To be reported for the first time are the dissolution measurements in the aerosol phase of crystalline and amorphous systems (Fig. 1). An adapted electrodynamic balance (EDB) has been used to replicate the dissolution process of dry powder particles in the aerosol phase experienced in the lung. The EDB traps single particles within its core and monitors size/phase change as a function of time (0.01s resolution) and environmental conditions (relative humidity,RH, & temperature which can be changed <0.1 seconds).

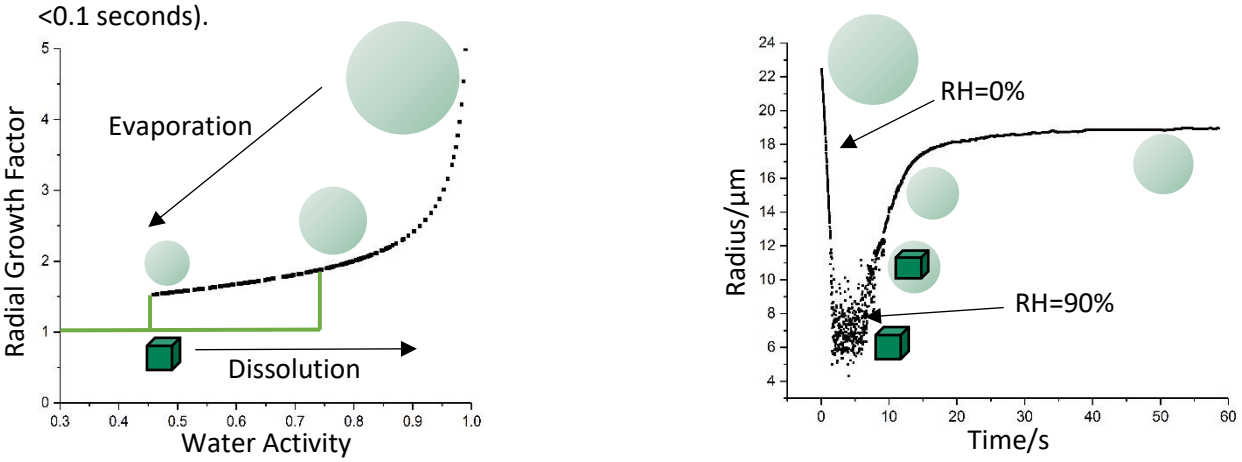

Figure 1. A) The efflorence and deliquesence cycle of sodium chloride, where water activity is equivalent to RH, i.e. a sodium chloride droplet will crystalise at ~45% RH and need an RH of 75% to become an aqueous droplet. B) A dried,crystalline sodium chloride particle is trapped in the EDB, after a set time the RH is switched to 90%. A dissolution lag of ~3seconds is seen before the particle starts to take up water and dissolve to form a homogenous droplet.

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DEVELOPMENT OF VIBRATING PENDANT-DROP RAMAN SPECTROMETER

T. Inoue, J. Kohno

Department of Chemistry, Gakushuin University, Tokyo, Japan

c536d84fd7hyi@gmail.com

Raman spectroscopy is one of important methods to acquire molecular information, which, however, suffers from low sensitivity. We use liquid droplets to improve the sensitivity. The Raman scattered light enhances its intensity within the droplet surface, because the droplets act as an optical cavity with a very high quality factor. In this study, we developed a pendant drop spectrometer to observe the Raman scattered light with high sensitivity. We also applied vibration to the pendant drop to make a deformed droplet, which further enhanced the Raman intensity.

A spherical pendant drop of H_2O was generated at the edge of a capillary (O.D. 360 μm, I.D. 50 μm) by using a syringe pump (5 μl/min). The droplet was irradiated with a second harmonic of a Q-switched Nd:YAG laser (280 μJ/pulse). The Raman scattered light passed through a long pass filter for removal of the Rayleigh scattering light and divided into two by a half mirror. One was fed into a CCD camera to observe the pendant drop, the other was fed into a CCD spectrometer to obtain the spectrum. For the generation of the deformed droplet, the capillary was connected to a piezo-driven speaker applied a voltage of 100 V at 410 Hz.

Figure 1 a and b show the images of spherical and deformed pendant drop of H_2O, respectively. The diameter of the deformed droplet was about 0.21 mm larger in the vertical direction than the spherical droplet with a diameter of 0.66 mm.

Figure 2 a and b show the Raman spectra obtain from the spherical and the deformed pendant drop of H_2O, respectively. In the Raman spectrum, a peak assignable to OH stretching vibrational mode ($\sim$3410 cm^{-1}) is observed with a narrower bandwidth than that of a spontaneous Raman spectrum. The contour of the Raman spectrum is interpreted by considering a stimulated Raman scattering (SRS). The SRS band shape is expressed by the following equation; $\exp\{A(S_I(\omega)-1)\}$, where A is the gain factor, $S_I(\omega)$ is the spontaneous Raman spectrum.[1] The gain factor was 13.0 and 15.7 for spherical and the deformed droplet, respectively. This result indicates that the SRS of the deformed droplet is enhanced more efficiently than that of the spherical droplet.

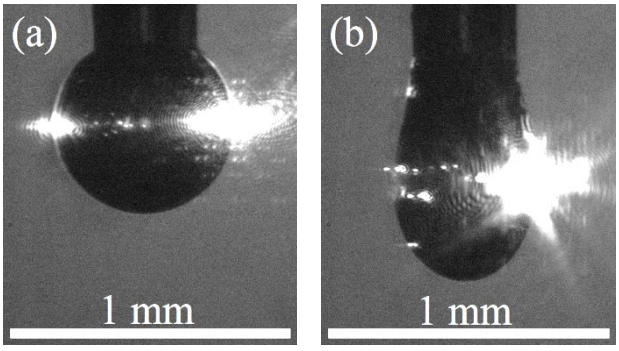

Figure 1 The pendant drop images of (a) spherical and (b) deformed droplet of H_2O.

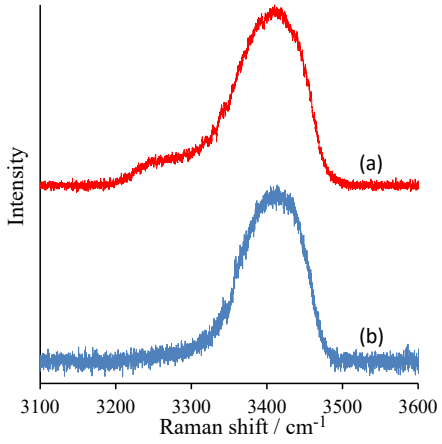

Figure 2 The Raman spectra of (a) spherical and (b) deformed droplet of H_2O.

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Investigations of the viscosity of atmospherically relevant organic particles

L. Lalemi and B. R. Bzdek*

School of Chemistry, University of Bristol

*Presenting author email: ll14323@bris.ac.uk

Atmospheric aerosols influence the absorption and scattering of solar radiation, the formation of clouds, and human health.[1] Organic molecules are a major component of atmospheric aerosols.[2] The viscosity of an aerosol particle affects diffusion rates within the particle, thereby affecting reactivity, as well as partitioning timescales of semivolatile molecules, affecting the responsivity of the particle to changes in ambient conditions (e.g. relative humidity, temperature, gas phase composition). Predictive frameworks to describe particle viscosity are required in order to better capture the dynamic behaviour of aerosols. In this work, we investigate the viscosity of a ternary water/sucrose/citric acid system over a wide range of relative humidities. This chemical system is chosen as a proxy for organic aerosol mixtures.[3] Particle viscosity is investigated using holographic optical tweezers. Two droplets (< 10 μm) are captured in individual optical traps and equilibrated to a desired relative humidity. Viscosity is retrieved through coalescence of the two levitated viscous droplets. The timescale of coalescence allows determination of the droplet viscosity. The benefit of performing these measurements in aerosol rather than in the bulk is that supersaturated solute states inaccessible to bulk approaches can be studied, allowing investigation of the entire range of relative humidity, spanning viscosities from 10^{-3} Pa s to $>10^{9}$ Pa s.[4] These optical tweezers results are compared to results obtained from complementary approaches in order to better resolve the differences and limitations among these approaches. The results also shed light on the applicability of the Stokes-Einstein equation to atmospherically-relevant systems.

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INVESTIGATING BACTERIAL LOSS OF VIABILITY IN AEROSOLS

Henry Oswin[1], Mara Otero[1], Allen Haddrell[1], Jonathan Reid[1]

[1]University of Bristol, Bristol, United Kingdom

ho1451@bristol.ac.uk

Bacterial transport in aerosols is a key step in the transmission of diseases such as Tuberculosis[1] (TB) and pneumonia[2]. These diseases create a large global health burden, causing the deaths of millions of people every year[3]. Studying the way bacteria behave within aerosol droplets, and the factors influencing their survival, will be important in developing countermeasures to limit the spread of these diseases.

It has become apparent that the viability of many bacterial species is negatively impacted by aerosolisation[4]. This loss of viability is likely to be a key factor in determining the risk of airborne transmission for various pathogens, yet its cause is thus far undetermined. We aim to probe the impact of various parameters on this loss of viability, to provide a more detailed understanding of the mechanisms involved. This has required the development of a highly controlled system in which bacterial aerosols can be produced and maintained under defined and highly reproducible conditions[5].

We have developed a system in which piezoelectric droplet dispensers are used to produce identical droplets, which are charged and then trapped in an electric field. By altering the relative humidity around the droplets, and the solute concentration within the droplets, the effect of droplet drying kinetics and droplet size on bacterial viability can be explored. We can alter the atmosphere surrounding the droplets, allowing for the impact of oxygen concentration and the presence of other gases on the viability loss to be assessed. The droplets can be deposited onto a range of substrates, allowing for a various techniques to be used to determine the viability of the bacteria within the droplets. The droplets can be deposited onto solid growth media allowing for assessment of the number of bacteria able to form colonies. Droplets can be deposited onto microscopy slides, so that live/dead staining can be used to assess the membrane integrity of bacteria within the droplets, and so that the morphology of cells post aerosolisation can be examined.

ACKNOWLEDGEMENTS: The authors thank DSTL and EPSRC for supporting this work.

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CONTINUOUS OPERATION OF LEIDENFROST ROTORS ON TURBINE-INSPIRED SUBSTRATES

Prashant Agrawal[1], Gary G. Wells[1], Rodrigo Ledesma-Aguilar[1], Glen McHale[1], Anthony Buchoux[2], Khellil Sefiane[2], Adam Stokes[3], Anthony J. Walton[3], Jonathan G. Terry[3]

[1]Smart Materials & Surfaces Laboratory, Faculty of Engineering & Environment, Northumbria University, Newcastle upon Tyne, UK; [2]School of Engineering, Institute for Multiscale Thermofluids, The University of Edinburgh, Edinburgh, UK; [3]School of Engineering, Institute for Integrated Micro and Nano Systems, The University of Edinburgh, Edinburgh, UK

prashant.agrawal@northumbria.ac.uk

In the Leidenfrost effect a liquid droplet placed on a surface heated at temperatures significantly higher than its boiling point vaporizes instantly and levitates on a cushion of its own vapour[1]. In this thin-film boiling regime, the vapour layer acts as a virtually frictionless surface on which liquid droplets can self-propel by asymmetrically texturing the substrate[2]. Here we investigate the rotation of a liquid pool coupled to a non-volatile solid plate on heated turbine-like substrates[3,4]. By replenishing the liquid working substance we sustain the rotation of the surface tension coupled rotor assembly. We alter the wettability of the plate and the turbine to enhance the rotation stability and expand the operating temperature range**.** Experimental observations of the rotation characteristics are further supported using an analytical model. The dynamic analysis of these Leidenfrost rotors paves the way for developing mm and sub-mm scale heat engines for applications in extreme environments, such as at microscales or in space and planetary exploration.

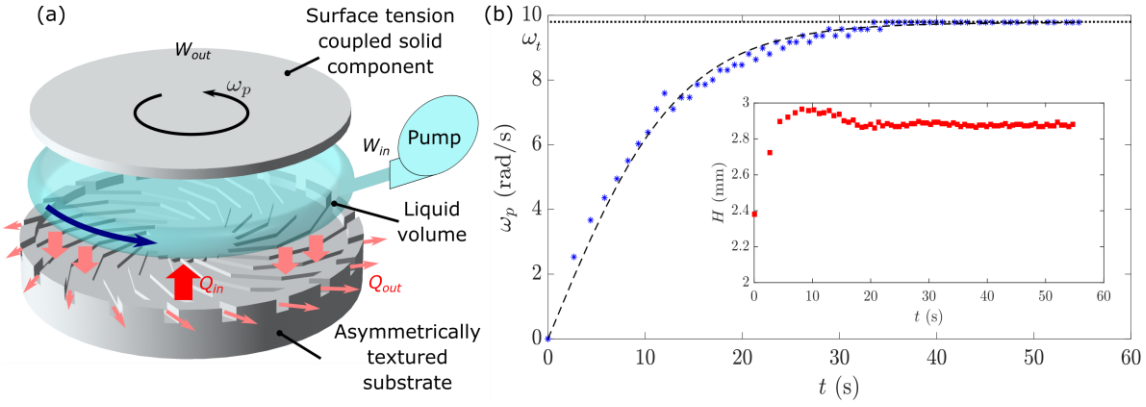

Figure 1: (a) Experimental configuration of a Leidenfrost rotor; (b) Variation of angular speed of the plate and height of liquid volume (inset) with time

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Droplet re-icing characteristics on solid surfaces

Fuqiang Chu[1] and Dongsheng Wen[1,2]

[1] School of Aeronautic Science and Engineering, Beihang University, Beijing, China; [2] School of Chemical and Process Engineering, University of Leeds, Leeds, UK

Email of presenting author: chu_fuqiang@126.com

Droplet icing exist widely in nature and in industry, causing numerous problems such as imperiling the flight safety of an aircraft, deteriorating the heat transfer of a heat exchanger, and reducing the power generation efficiency of a wind turbine.[1] To avoid the hazards of icing, understanding the droplet icing mechanisms is essential. Generally, droplet icing experiences two stages including nucleation/recalescence and freezing, where the former is a rapid kinetically controlled process during which the droplet becomes clouding, and the latter is a heat transfer controlled process.[2] Here, we reported a different icing process of a droplet, which called re-icing. First, we cool the solid surface, freezing a water droplet, and then rise the surface temperature to make the ice droplet melt. Before the ice droplet finish its melting, we reduce the surface temperature again to make the droplet re-ice. According to our observation, the re-icing is quite different from the general icing process. During the nucleation stage, ice nucleation direct nucleates from the unmelted ice cover floating in the upper part of the droplet. The nucleation dendrites gradually stretch downward, like a sword, untill they reach the bottom of the droplet. During this stage, the droplet keeps transparent, without the clouding penomenon (see Fig. 1). The freezing stage of re-icing is similar to that of the general icing process that freezing front begins to grow upwards. Except that bubble formation (reported in our previous work [3]) can be seen clearly due to the transparency of the droplet (see Fig. 1). In addition, the final ice droplet shape after re-icing process is also different from that after the genieral icing process. Compared with the latter, the ice droplet shape after re-icing is more spherical, and the pointy tip formed at the top of the droplet is blunter (see Fig. 1). We expect that our work may deepen the understanding of icing physics and help improving anti-icing techniques.

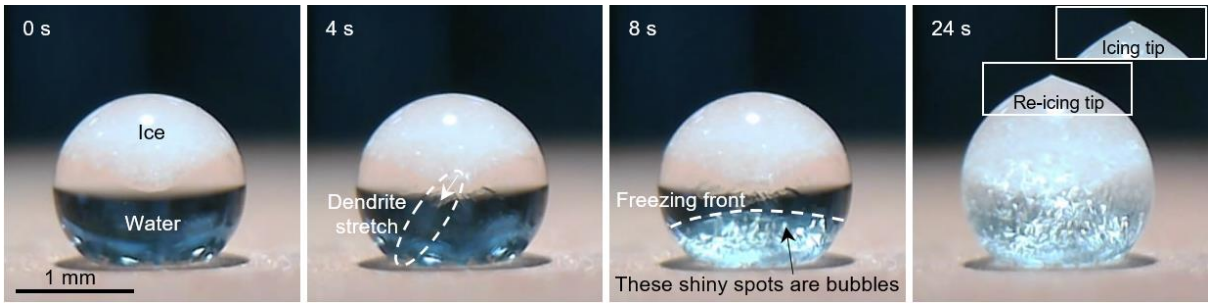

Fig. 1 Re-icing process of a droplet on solid surfaces

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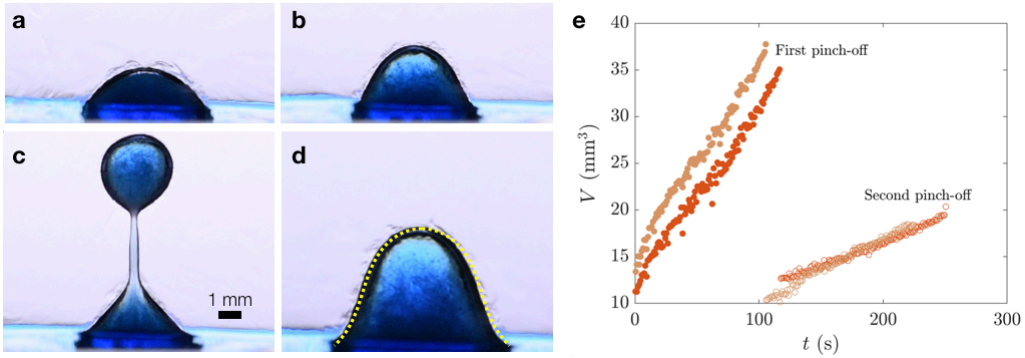
THE SPONTANEOUS MIXING AND BREAKING OF A WATER DROP IN ANISE-OIL (AND SOME ETHANOL)

O. R. Enríquez[1], M. Zürcher[1], P. Peñas-López[2], C. Diddens[2], and J. Rodríguez-Rodríguez[1]

[1]Carlos III University of Madrid, Madrid, Spain; [2]University of Twente, Enschede, The Netherlands

oenrique@ing.uc3m.es

We deposit a water droplet at the bottom of a mixture of anethole (anise oil) and ethanol; where there is at least twice as much of the former than the latter. The ensuing events are dominated by the mass transfer of host fluid into the droplet. Shortly after deposition the droplet (dyed blue for contrast) starts oscillating, presumably due to Marangoni stresses that arise that arise as the composition of both liquids start to change spatially along the interface. The latter also triggers a radial gravity current, moving outwards from the drop (see Fig. 1a-1c), which suggests that (lighter) ethanol is preferentially dissolving into the droplet, making this current rich in (heavier) anethole. This is confirmed by the droplet's growth and its deformation due to buoyancy (panel b), which evidences a reduction in its average density. Observation of Fig. 1d reveals that the reduction is not uniform: the top part of the drop is fainter than the bottom one, indicating that the entrained, undyed fluid is mostly concentrated near the top. Despite this composition gradient and the flows taking place both inside and outside the drop, its shape seems to be well predicted by the equations of hydrostatics assuming a uniform composition [1] (see Fig. 1d). One free parameter is adjusted to fit the hydrostatic solution to the drop shape, namely a Bond number defined as $Bo = \Delta\rho g\, R_0^2/\sigma$, with R_0 the radius at the base of the drop, g the gravity, $\Delta\rho$ the density difference between the drop and the host liquid and σ the interfacial tension. Since these last two parameters are actually unknown, the Bond number obtained through this fitting sets a relation between them.



(a-c) A water droplet (dyed blue for contrast) in a 5:1 (volume) anethole:ethanol solution. Time after deposition: 33 s (a), 63 s (b), and 69 s (c). (d) Best-fitting hydrostatic solution (yellow line) obtained by adjusting the value of the Bond number (Bo = 2.53) for the same droplet (zoomed-in), 2 s before detachment. (e) Droplet volume for two experimental realizations in a 6:1 anethole:ethanol solution. The two detachment events shown for each case reveal that the mixing process is much slower after the pinch-off event.

Eventually the growth in volume and the reduction in the average density make the droplet detach (Fig. 1c) and a new cycle begins. However, as can be seen in Fig. 1e, the mass transfer process becomes much slower after a pinch-off event, as a consequence of the liquid left in the drop having a composition closer to that of the host solution than the initial drop.

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EFFECT OF SURFACTANT ON DROPLET BREAKUP IN T-JUNCTION UNDER ASYMMETRIC PRESSURE DIFFERENCE

Qingqing Gu[1], Haihu Liu[2] and Yonghao Zhang[1] (Underline presenting author)

[1]James Weir Fluids Laboratory, Department of Mechanical and Aerospace Engineering, University of Strathclyde, Glasgow G1 1XJ, UK; [2]School of Energy and Power Engineering, Xi'an Jiaotong University, 28 West Xianning Road, Xi'an 710049, China

haihu.liu@mail.xjtu.edu.cn

Understanding of droplet dynamical behavior in microchannels is of great importance in many biomedical and biotechnological applications. Droplet deformation, breakup and coalescence can be manipulated by adding surfactants [1]. Here the droplet formation in a microfluidic T-junction is studied with presence of an insoluble surfactant. A recent improved lattice Boltzmann color-gradient model, coupled with the finite difference method that solves the convection-diffusion equation, is used to simulate the immiscible two-phase flows considering the impact of surfactant on the interfacial tension. Three flow regimes, i.e. non-breakup, breakup with tunnels, and breakup with permanent obstruction, are observed as reported by the literature [2]. A new definition of asymmetric factor is proposed to capture the effect of surfactant concentration. The influence of surfactant on flow-regime-transition and the corresponding critical Capillary number are also investigated.

ACKNOWLEDGEMENTS: This work is supported by the National Key Research and Development Project of China (No. 2016YFB0200902) and the National Natural Science Foundation of China

(No.51876170). The authors would like to thank the ``UK Consortium on Mesoscale Engineering Sciences (UKCOMES)" (http://www.ukcomes.org/) funded by the UK Engineering and Physical Sciences Research Council (EPSRC) under grant number EP/R029598/1 to provide computing time on ARCHER. And the simulations are also performed on ARCHIE-WeSt funded by EPSRC (EP/K000586/17). Qingqing Gu is grateful for the scholarship provided by China Scholarship Council.

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OPTIMISING LEIDENFROST HERRINGBONE RATCHET PROPULSION WITH NUMERICAL SIMULATIONS

M.T. Parnell[1], D. Wood[1] and L.E. Dodd[1]

[1] Faculty of Engineering and Environment, Northumbria University, Newcastle upon Tyne, UK

M.Parnell@Northumbria.ac.uk

The Leidenfrost effect describes the interaction when a droplet is placed on a surface with temperature significantly higher than the boiling point of the liquid. Here, the underside of the droplet is instantly vaporised, and this constantly replenishing vapour layer cushions the droplet allowing it to float [1]. Due to the symmetry of the system on flat surfaces, a droplet larger than the capillary length of the liquid will not move. However, these droplets can be made to self-propel by adding asymmetries to the system. A ratcheted surface will channel the vapour in a net direction, due to viscous friction, which causes the droplet to propel across the surface [2]. The applications of Leidenfrost propulsion have been limited because the droplet position cannot be easily controlled. Due to the virtually frictionless nature of this effect, any lateral motion cannot be corrected without external input, i.e. walls. In order to achieve droplet directional control without the need for walls, two ratchet surfaces can be angled towards one another in a herringbone pattern, which allows for further droplet control and even self-centring [3]. When the droplet passes from one side of the substrate to the other, the force on the droplet changes such that it always acts towards the central boundary, causing the droplet to oscillate. It has been shown that ratchet height can affect the ability of the droplet to centre, however further control of droplet position had not been investigated. In this work, in order to further investigate the droplet position, Matlab has been used to create a numerical model, which estimates and predicts the effect of the propulsion force on each side of the herringbone ratchet, the effect of the opposite side of the structure as the droplet approaches the centre and the effect of the force when the droplet is above both sides (on or near the centre). By changing the flare angle of the herringbone ratchet, which is defined as the angle between the ratchet and the desired direction of travel, the droplet's position can be optimised. The model was used to predict the ideal parameters to provide a minimised centring distance without compromising the velocity of the droplet transport, which provided a flare angle of 56°. The gives an 8% improvement for centering distance, with a 6% higher velocity and 15% less overshoot, compared a herringbone ratchet with a flare angle of 45°. These trends were confirmed experimentally, with excellent agreement. Optimising frictionless droplet transport could have wide ranged applications, from fluid transport, to multi-phase cooling, and even energy generation [4-5].

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ON-DEMAND PODUCTION OF FEMTOLITER DROPLETS IN MICROCHANNELS USING ELECTRIC FIELD PULSES

M. Shojaeian and S. Hardt

Institute for Nano- and Microfluidics/TU Darmstadt, Darmstadt, Germany

hardt@nmf.tu-darmstadt.de

We present a method allowing to produce monodisperse aqueous droplets with volumes in the femtoliter range in a microchannel on demand [1], with the typical channel dimensions being significantly larger than the droplet diameter. The method utilizes pulsed electric fields deforming the interface between an aqueous and an oil phase and pinching off droplets. Water and xanthan gum solutions are considered as disperse-phase liquids, and it is shown that the method can be applied even to solutions with a zero-shear rate viscosity more than 10^4 times higher than that of water. We explore the different regimes in which no droplets, single droplets and multiple droplets are produced. The dependence of the process on the pulse amplitude can be utilized to tune the droplet size, with the smallest droplets having diameters between 3 and 4 µm. The figure below shows the production of a single droplet of xanthan gum solution, where the numbers indicate the time in milliseconds. To demonstrate the applicability of the electric-field-driven droplet generator, it is shown that the droplets can be used as versatile biological reaction compartments. In that context, gene transcription and protein biosynthesis are demonstrated, and it is verified that biomolecules inside the aqueous droplets such as small RNAs can be diffusionally activated from the outside to induce a ligand-driven biochemical switch.

ACKNOWLEDGEMENTS: The authors acknowledge the support from the LOEWE CompuGene Project, funded by the Hessian Ministry of Science and Art.

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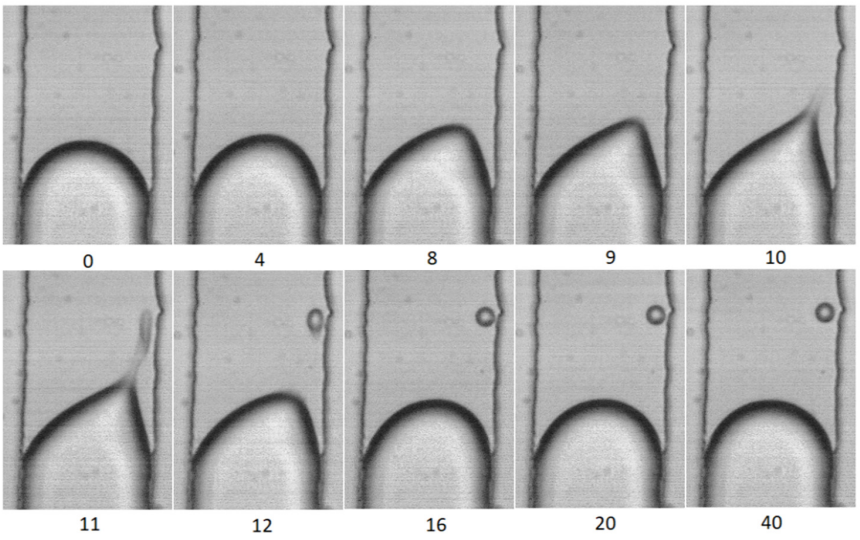

COMBINED CFD-POPULATION BALANCE MODELLING OF EMULSION FORMATION IN TURBULENT FLOWS

A. Vikhansky[1], D. Eskin[2]

[1]Siemens PLM Software, Trident House, Basil Hill Road, Didcot OX11 7HJ, UK; [2]Skolkovo Institute of Science and Technology, Moscow, 121205, Russia
alexander.vichansky@cd-adapco.com

Multiphase flows are characterized by a wide spectrum of diameters of the involved droplets . Since the droplets size distribution (DSD) is in permanent evolution due to breakage and coalescence, one has to integrate a population balance modelling (PBM) method into a CFD code. A combined CFD-PBM method should be robust and computationally non-expensive, it is why we recently developed and implemented the adaptive multiple size-group (A-MuSiG) method. In order account for the multi-disperse nature of the flow, the disperse phase is divided into M size-groups and - following the Eulerian modelling framework - each group has its own momentum, volume fraction, etc.; that is, it is treated as a separate phase in every aspect but name. The method doesn't use any a priori discretisation of the DSD and, as it widens and shifts toward large/smaller characteristic sizes due to coagulation/breakup, diameters of the size-groups adaptively follow the DSD. It is achieved by solution of a tri-diagonal system of linear algebraic equations, so the computational cost of the adaptivity scales linearly with the number of the groups, and the method is stable for dozens of size-groups.

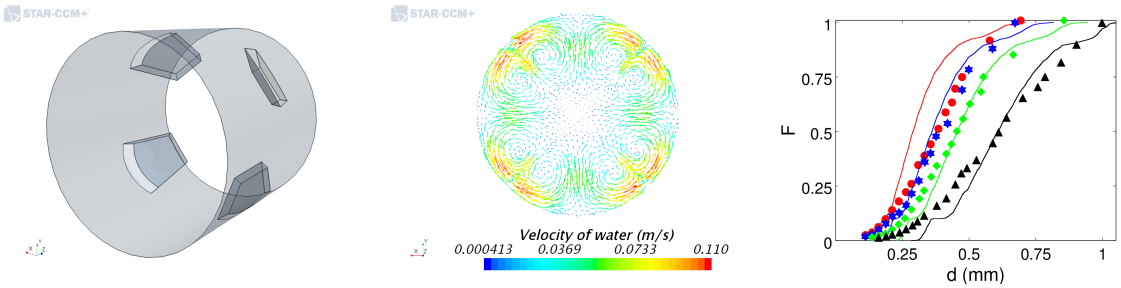

Figure 1: One element of the HEV mixer; velocity in the transverse plane; DSD: experimental (symbols) and predicted for $\mathrm{Re} = 7.5 \times 10^3,\ 10.0 \times 10^3,\ 12.5 \times 10^3,\ 15.0 \times 10^3$.

In the present work a novel model for the droplets breakup rate is proposed; it predicts a wide DSD (in agreement with the experimental data). We simulate emulsion formation in different industrial and laboratory devices: oil pipe-line flow, Couette device, static mixer. Fig. 1 shows results of the calculations for high-efficiency vortex (HEV) mixer. The HEV mixer is a straight pipe, along which tab arrays are fixed; each of the nine arrays consists of four trapezoidal baffles fixed at a 30 degrees angle to the wall. Main role of the baffles is to generate large-scale longitudinal vortices and enhance intensity of the turbulence. Both DSD and level of turbulence are similar to what has been observed in experiment. It is shown that most of droplets are broken near the wall, where turbulence dissipation rate is maximum. Droplets are transported to and out of the wall by the vortices, generated by the baffles.

Special attention has been paid to performance of the A-MuSiG method. Number of the size-groups have been varying from 3 to 10; due to the adaptive nature of the method, even the smallest number of size-groups (3) suffice for reliable prediction of the Sauter mean diameter.

DEVELOPMENT OF A MICROFLUIDIC METHOD FOR PREPARING MONODISPERSED MICRO-PARTICLES WITH CONTROLLABLE SIZES AND MECHANICAL PROPERTIES

C. Zhang, R. Grossier, N. Candoni and S. Veesler

CNRS, Aix-Marseille University, CINaM (Centre Interdisciplinaire de Nanosciences de Marseille), Campus of Luminy, Case 913, F-13288 Marseille Cedex 09, France

zhang@cinam.univ-mrs.fr

In this communication, we introduce and characterize a simple microfluidic setup for the preparation of monodispersed alginate micro-particles displaying controllable sizes and mechanical properties without using surfactant. The final objective is to produce micro-particles mimicking red blood cells. Ca-alginate (alginate cross-linked by calcium) micro-particles are fabricated to achieve this goal. Alginate hydrogel micro-particles are also widely applied in medical and biological fields. [1]

As shown in Figure 1 (a), first, Na-alginate micro-droplets with the desired size and concentration are emulsified in dimethyl carbonate (DMC) by using the droplet-based microfluidic technology. Second, alginate micro-droplets shrink gradually (Figure 1 (b)) because of the water extraction by DMC (mechanism comparable to evaporation) [2]. At last, after collecting in DMC and drying in air, an *ex situ* cross-linking with a $CaCl_2$ solution is performed and Ca-alginate micro-particles are obtained.

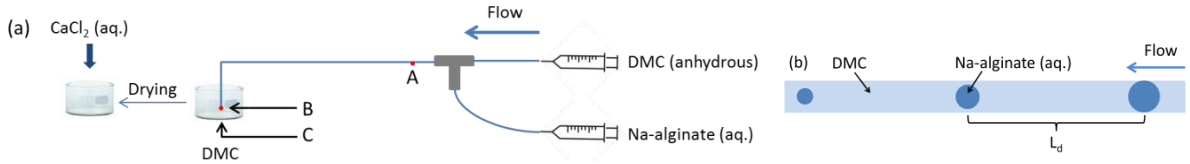

Figure 1. Schema of (a) the microfluidic setup where A, B and C are observation points after micro-droplet generation, at the end of the tube and at the bottom of the collecting vial respectively; (b) generation and shrinkage of micro-droplets in the tube.

Before cross-linking, different parameters, such as the initial Na-alginate concentration $\mathbf{C}_i$ (0.006wt%-1wt%), the channel length, the distance between two successive droplets $\mathbf{L}_d$ and the initial droplet diameter $\mathbf{d}_i$ (110µm-220µm) at the point A, are varied to study their effect on the final alginate micro-particle formation. Observed by scanning electron microscope, these particles are spherical with a smooth surface. We also find that the final micro-particle diameter $\mathbf{d}_f$ at the point C is influenced by both $\mathbf{d}_i$ and $\mathbf{C}_i$. However, the final alginate concentration $\mathbf{C}_f$ in the micro-particle only depends on $\mathbf{d}_i$.

Cross-linking by a $CaCl_2$ solution induces alginate micro-particles swelling. Moreover, both optical and electron microscopes unravel the porous internal structure of micro-particles. This is due to the Na-alginate diffusion in the micro-particle during the water extraction. Finally, a micro-tweezer is used to measure the mechanical properties of alginate micro-particles before and after cross-linking.

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DRYING KINETICS AND PARTICLE FORMATION FROM COLLOIDAL MICRODROPLETS SUSPENSION

J. Archer, F. K. A. Gregson, J.S. Walker, R. E. H. Miles, and J. P. Reid

School of Chemistry, University of Bristol, Bristol, BS8 1TS, UK

Email : justice.archer@bristol.ac.uk.

The morphology and surface properties of dried particles emerging from the drying of colloidal droplet suspensions, e.g in spray drying technologies, pharmaceuticals and food processing are strongly influenced by the general dynamics of the drying process. In particular, the final dry particle morphology is influenced by the competition between the diffusion of the suspended particles in the droplet to the droplet surface and convection, which is normally quantified by the dimensionless Péclet number (Pe)[1]. In this work, a single-particle levitation instrument: the electrodynamic balance[2] (EDB) is used to study the drying kinetics of micron-sized droplets of colloidal silica suspensions for a range of Péclet numbers through variable evaporative parameters (relative humidity, temperature, initial droplet composition and size). To relate the droplet drying kinetics to the final dry particle morphology and properties, a complementary droplet-chain technique[3] is used to reproduce the droplet drying kinetics and to collect the final dry particles for offline analysis with scanning electron microscopy (SEM).

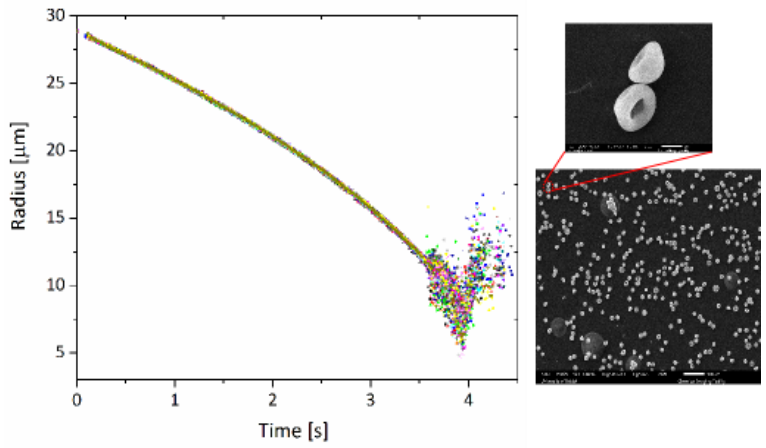

Fig. 1 : Left - The EDB droplet-drying evolutions of 105 hydrophilic silica droplets overlaid (initial volume concentration 0.5 % v/v) at 40 % RH and 294K, demonstrating the reproducibility in the drying kinetics. The right SEM micrographs show the final dry silica products with *Pe ≈ 21,* reproduced with the droplet-chain technique for the same initial droplet parameters and drying conditions.

ACKNOWLEDGEMENTS : The authors acknowledge the funding support from the EPSRC under grant code EP/N025245/1.

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FLUID FLOW STRUCTURES IN AN EVAPORATING DROPLET DEPENDING ON THE DROPLET GEOMETRY AND PROPERTIES OF LIQUID AND SUBSTRATE

E.S. Melnikova[1,2], A.A. Gavrilina[1,2] and L. Yu. Barash[1,2]

[1] National Research University Higher School of Economics, 101000 Moscow, Russia
[2] Landau Institute for Theoretical Physics, 142432 Chernogolovka, Russia
barash@itp.ac.ru

We investigate numerically quasi-steady internal flows in an axially symmetrical evaporating sessile droplet. The fluid dynamics of an evaporating sessile drop, effects of the heat conduction in the droplet and substrate and diffusion of vapor in air have been taken into account jointly. The equations have been solved by finite element method using ANSYS Fluent. Influence of gravity on the droplet shape is taken into account, and the effect of droplet size has been considered. Temperature distributions and the corresponding quasistationary fluid flow structures are obtained for evaporating sessile droplets of 1-hexanol, 1-butanol and ethanol. The phase diagrams containing information on the number and orientation of the vortices depending on the contact angle and the ratio of substrate to fluid thermal conductivities [1], are obtained and analyzed for various values of parameters.

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TURNING DROPS INTO BUBBLES: CAVITATION BY DIFFUSION TROUGH AN ELASTIC NETWORK

M.A.Bruning[1], M.Costalonga[2] , J.H.Snoeijer[1] and A.Marin[1]

[1]Physics of Fluids Group, Faculty of Science and Technology, Mesa+ Institute, University of Twente, Enschede, The Netherlands; [2]Department of Mechanical Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts, USA

m.a.bruning@utwente.nl

Plants have found an elegant way to combine fluid mechanics and elasticity to generate rapid motion. By making use of the elasticity of their tissue structure and the pressure differences in their internal liquid conducts, plants can easily achieve surprisingly fast movements, useful for instance in spore dispersion[1].

In this work, we study this conversion from elastic to kinetic energy using a model experiment: an evaporating water drop trapped in the bulk of a poro-elastic gel (Polydimethylsiloxane). The droplet shrinks due to evaporation, which follows a slow diffusive-limited process and can be modeled accordingly. During the droplet shrinkage, the pressure in the liquid decreases as a consequence of the build-up of elastic stress at its interface. This underpressure eventually leads to the birth of a cavitation bubble. Using high-speed imaging, we access the dynamics of the bubble growth and subsequent oscillations. We show that these observations can be understood through a modified Rayleigh-Plesset equation to account for elasticity. The experiments reveal the cavity dynamics over the extremely disparate timescales of the process, spanning 9 orders of magnitude. Such a model system could serve as a new paradigm for motile synthetic materials. Finally, since natural systems typically present multiple cells/droplets/bubbles, the evaporation and cavitation of multiple droplets are investigated to gain understanding on their interactions.

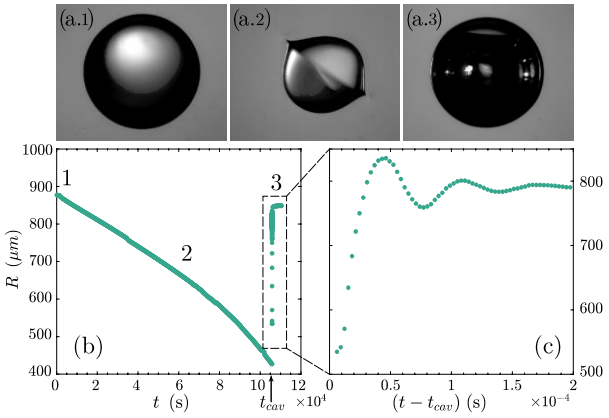

Figure 1: Different phases in the life of a droplet in a dry and elastic medium: (a.1) The initial water drop in a PDMS-based gel shrinks by evaporation. (a.2) Creasing at the gel interface. (a.3) Cavitation bubble is created. (b) The effective droplet radius in, illustrating the disparity of time scales, from a day to a fraction of a millisecond. The corresponding instants for the images in the (a)-panels are also indicated. (c) A zoom of the bubble radius dynamics just after cavitation occurred at t_{cav}

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INFLUENCE OF CAPILLARY ORIGAMI ON THE EVAPORATION OF SESSILE DROPS ON SOFT MEMBRANES

Y. Chen[1], D. Orejon[1], K. Sefiane[1], P. Valluri[1] and V. Koutsos[2]

[1]Institute for Multiscale Thermofluids, The University of Edinburgh, Edinburgh, United Kingdom;

[2] Institute for Material Processes, The University of Edinburgh, Edinburgh, United Kingdom

Yuhong.Chen@ed.ac.uk

ABSTRACT: Drops drying on soft substrates are commonly present in daily life and in nature, such as perspiration on human skin and raindrops on tree leaves. In particular, the surface tension of a drop can fold a thin soft sheet/membrane, which is known as 'capillary origami'. Such folding of a membrane takes place when the capillary energy is larger than the elastic bending energy[1-3]. Recently, capillary origami has attracted increasing attention due to its virtue to provide a simple and cost-effective method for the fabrication of a predetermined 3D structure at the micro/nano scale[4-5]. However, research in the topic scarcely investigates the influence of capillary origami on drop evaporation. Hence, in the present work we carry out experiments of water drops evaporating on PDMS membranes of different thickness. The different membrane thickness enables the absent, partial and complete folding of the membrane of the same area. Drops of 5 μl are generated by a syringe pump and gently deposited on the square PDMS membranes. An x-y translation stage is used to precisely control the deposition of drop at the center of the membrane to ensure symmetrical folding of the membrane. The membrane is supported on a hydrophobic PTFE substrate, sitting on a microbalance with a weighing resolution of ± 0.01 mg. The mass of the drying drop in time is then retrieved. In addition, a digital microscope camera and an optical camera are set to simultaneously record the side-view and top-view profiles of the drop and the membrane, respectively. An LED light is positioned opposite to the side-view camera to provide the right illumination and contrast for the further drop and membrane shape analysis. After the complete drop evaporation, videos are processed with a custom-written Matlab code to extract the profile evolution of the drop and the folding angle of the membrane. Preliminary results show that the degree of folding of the membrane reduces the average evaporation rate of the drying drop lengthening the drop lifetime.

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EVAPORATION OF A SESSILE DROPLET IN A SHALLOW WELL

H.-M. D'Ambrosio[1], T. Colosimo[2], S. K. Wilson[1], B. R. Duffy[1], C. D. Bain[2], and D. E. Walker[3]

[1]University of Strathclyde, Glasgow, United Kingdom; [2]Durham University, Durham, United Kingdom; [3]Merck Chemicals Ltd, Southampton, United Kingdom

hannahmay.dambrosio@strath.ac.uk

The evaporation of sessile droplets occurs in a wide variety of physical contexts, with numerous applications in nature, industry and biology. Examples of practical applications include blood splatter in forensic science, DNA mapping and gene analysis in the medical industry, the spreading of pesticides on leaves, and coating technologies. In particular, the evaporation of droplets plays a key role in the manufacture of Organic Light Emitting Diode (OLED) displays. In this talk I will formulate and analyse a mathematical model for the evolution of a thin droplet undergoing diffusion-limited evaporation on a non-uniform substrate, specifically in an axisymmetric well similar to those encountered in OLED manufacturing applications. I will use the model to describe the evolution of the height profile, contact radius, and hence the volume of the droplet, from its initial configuration until total evaporation. In particular, I will show how the evolution of the droplet is qualitatively dependent on the cross-sectional profile of the well. I will then compare the theoretical predictions of the model with detailed experimental results for the evaporation of droplets in cylindrical wells of various sizes, and show that theory and experiment (and, in particular, the critical times at which the droplet first touches down and at which total evaporation occurs) are in excellent agreement.

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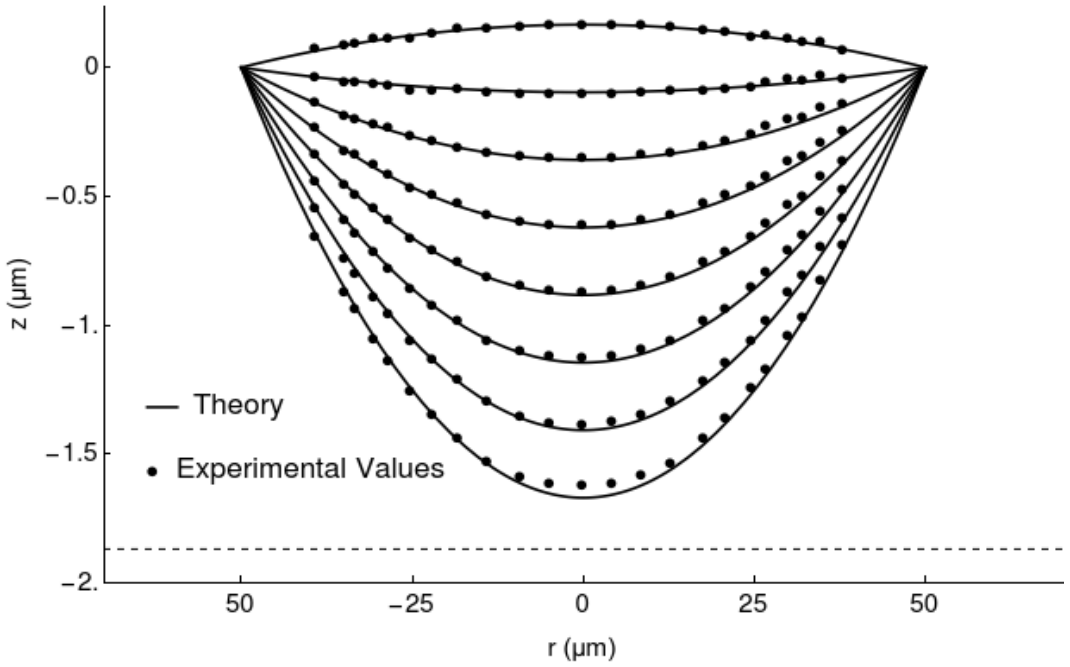

Figure: Comparison of the height profile predicted by the theoretical model with experimental data for a methyl benzoate droplet evaporating in a cylindrical well of depth 1.87 μm and radius 50 μm at eight equally-spaced times.

A MODEL FOR EVAPORATION AND SOLIDIFICATION OF SINGLE DROPLETS OF SEA WATER IN A MIST CURTAIN USED FOR SHIELDING INTENSE THERMAL RADIATION OF A FIRE

L. A. Dombrovsky[1,2], V. Yu. Levashov[3], A. P. Kryukov[4], J. X. Wen[5]and S. Dembele[1]

[1]Department of Mechanical Engineering, Kingston University, London, SW15 3DW, UK
[2]Joint Institute for High Temperatures, 17A Krasnokazarmennaya St, Moscow, 111116, Russia
[3]Institute of Mechanics of Moscow State University, 1 Michurinskiy Prosp., Moscow, 119192, Russia
[4]Moscow Power Engineering Institute, 14 Krasnokazarmennaya St, Moscow, 111250, Russia
[5]University of Warwick, Coventry CV4 9YP, UK

ldombr@yandex.ru

Water mist curtains have received considerable attention from researchers and engineers for protection against thermal radiation from fires. Both the known experimental studies and computational models [1-3] considered pure water. However, some applications related to maritime transport or off-shore petrochemical platforms motivates the need for similar research for sea water. The present paper is focused on modeling complex behavior of saline water droplets during their evaporation and subsequent solidification under the action of intense visible and near-infrared flame radiation. The absorption of radiation by a single droplet or solid particle is calculated using the Mie theory and its modification for the case of two-layered spherical particles [4]. It was shown in [5] that this approach is applicable even for cubic crystals of salt. The evaporative cooling of droplets and convective cooling of salt particles are taken into account in a computational model for the transient heat transfer problem. At the present stage of the work, the simple approaches are considered for modeling the processes involved, and the reference solutions like [6] for evaporation are discussed for possible verification purposes. However, the known hysteresis behavior of the salt solution is taken into account. A series of calculations makes possible the comparison between the radiative heating and evaporation of droplets of pure water and more complex multi-phase processes in droplets of sea water at all stages of the process. The results obtained will be used in a subsequent analysis of protective properties of uniform and multi-layered sea-water curtains.

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STUDYING HYDROTHERMAL WAVES IN AN EVAPORATING SESSILE DROP BY COMPUTER SIMULATION

A. A. Gavrilina[1,2], L. Yu. Barash[1,2]

[1] National Research University Higher School of Economics, 101000 Moscow, Russia; [2] Landau Institute for Theoretical Physics of the Russian Academy of Sciences, 142432 Chernogolovka, Russia

gavrilinasanya@gmail.com

Over the past decade hydrothermal waves (HTWs) in evaporating droplets have been intensively studied experimentally, theoretically and numerically [1, 2]. We study unsteady internal flows in an evaporating sessile droplet on a heated substrate.

We have carried out detailed three-dimensional computer simulation of hydrothermal waves in an evaporating droplet depending on the parameters of the problem. The diffusion of vapor in air, the hydrodynamics in the drop, effects of the thermal conduction in the three phases and thermal radiation have been considered.

The equations have been solved numerically by finite element method (FEM) using ANSYS Fluent. To verify the model and algorithms, the flows in the drop are simulated and compared with the experimental results [3, 4]. The findings of the research illustrate how model parameters, such as the substrate temperature, volatility of the liquid etc., may influence the wave patterns in the droplet. The study of HTWs has potential important technological and biomedical implications.

ACKNOWLEDGEMENTS: This work was supported by grant No. 18-71-10061 from the Russian Science Foundation.

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PARTICLE MIGRATION IN INKJET-PRINTED DROPLETS

J. G. J. Goodall[1], L. Yang[1], C. D. Bain[1]

[1]Durham University, Durham, UK;

jack.g.goodall@durham.ac.uk

The evaporation of sessile droplets can be a useful method to pattern substrates, with inkjet printing technology being particularly good at the selective deposition of functional materials. A predictive understanding of formulations is necessary in order to design systems in which the internal flows generated during drying do not lead to undesirable non-uniform morphologies.[1] Composition or temperature gradients across the liquid-vapour interface have been shown to induce Marangoni flows which can redistribute suspended material,[2] however studies have mainly taken place on microlitre droplets.

Here we report experiments on the internal flows of inkjet-printed picolitre droplets in which high-speed cameras are used to follow the trajectories of light-scattering tracer particles and record the droplet profile. Solutal Marangoni flows are generated in a selection of solvent mixtures and solutions however at these smaller length-scales different morphologies are observed. Instead of obtaining uniform deposits, particles are seen to migrate across flow streamlines[3] to collect in groups in ethanol-water mixtures, ethylene glycol-water mixtures and sucrose, lactose, sodium chloride and sodium nitrate solutions, demonstrating the prevalence of particle migration in a disparate range of chemical systems. A weak particle-size dependence to the migration is noted and a diffusiophoretic mechanism proposed.

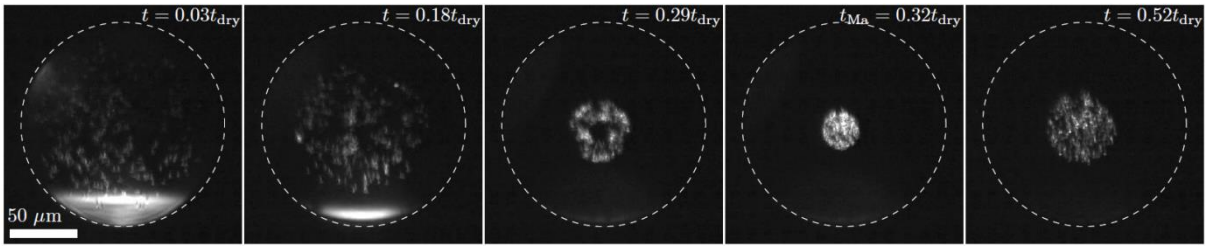

Figure 1. Particle migration towards the centre of an evaporating droplet of a 50:50%v ethanol-water mixture at *RH* = 50% on a cleaned glass coverslip. The dashed line is the position of the contact line. t_{Ma} is the time when the Marangoni flows ended and the collected group had its minimum radius while the overall drying time of the droplet was 2.4 s.

ACKNOWLEDGEMENTS: The authors acknowledge support from the Engineering and Physical Sciences Research Council under grant code EP/N025245/1.

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POWER SPECTRUM AND MACHINE LEARNING ANALYSIS APPLIED TO DRIED BLOOD DROPLETS

L. Hamadeh, S. Imran, M. Bencsik, G. Sharpe, M. Johnson and D. J. Fairhurst

Nottingham Trent University, Department of Physics and Mathematics, School of Science and Technology, Nottingham, Clifton Campus, NG11 8NS, Nottingham, United Kingdom

Lama.hamadeh@ntu.ac.uk

One of the most interesting and everyday natural phenomenon is the formation of different patterns after the evaporation of colloidal droplets deposited on a solid surface. The analysis of dried patterns resulting from biological liquids, such as blood, has recently gained a lot of attention [1,2], experimentally and theoretically, due to its potential application in biomedicine and forensic science. This work presents an entirely novel approach to studying human blood droplet drying patterns that could be extended for use on any circular patterns. We took blood samples from 30 healthy young men before and after exhaustive exercise which is well known to cause large disturbances in blood chemistry. We objectively and quantitatively analysed 1800 dried blood droplet images by developing sophisticated image processing analysis routines and optimising a multivariate statistical machine learning algorithm. We look for statistically relevant correlations between the patterns and exercise-induced changes in blood chemistry. An analysis of various measured physiological parameters is also investigated. We use a machine learning algorithm, which is an optimisation to a statistical model that combines Principal Component Analysis (PCA) [3] and Linear Discriminant Analysis (LDA) [4] method. We apply this technique to the logarithmic power spectrum of the images, and are able to predict with up to 95% accuracy whether a droplet has been taken before or after physical exercise. Interestingly, we find that the predictive power is improved if we average over all the images taken per volunteer per condition.

ACKNOWLEDGEMENTS: The authors thank Nottingham Trent University for supporting this work.

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Evaporation of dilute sodium dodecyl sulfate droplets on a hydrophobic substrate

Wojciech Kwieciński, Tim Segers, Sjoerd van der Werf, Arie van Houselt, Detlef Lohse, Harold Zandvliet, and Stefan Kooij

Email: w.kwiecinski@utwente.nl

Evaporation of surfactant laden sessile droplets is omnipresent in nature and industrial applications such as inkjet printing. Soluble surfactants start to form micelles in an aqueous solution for surfactant concentrations exceeding the critical micelle concentration (CMC). Here, the evaporation of aqueous sodium dodecyl sulfate (SDS) sessile droplets on hydrophobic surfaces was experimentally investigated for SDS concentrations ranging from 0.025 to 1 CMC. In contrast to the constant contact angle of an evaporating sessile water droplet, we observed that, at the same surface the contact angle of an SDS laden droplet with a concentration below 0.5 CMC first decreases, then increases, and finally decreases resulting in a local contact angle minimum (see Figure 1). Surprisingly, the minimum contact angle was found to be substantially lower than the static receding contact angle and it decreased with decreasing initial SDS concentration. Furthermore, the bulk SDS concentration at the local contact angle minimum was found to decrease with a decrease in the initial SDS concentration. The location of the observed contact angle minimum relative to the normalized evaporation time and its minimum value proved to be independent of both the relative humidity and the droplet volume and thus, of the total evaporation time. We discuss the observed contact angle dynamics in terms of the formation of a disordered layer of SDS molecules on the substrate at concentrations below 0.5 CMC. The present work underlines the complexity of the evaporation of sessile liquid-surfactant droplets and the influence of surfactant-substrate interactions on the evaporation process.

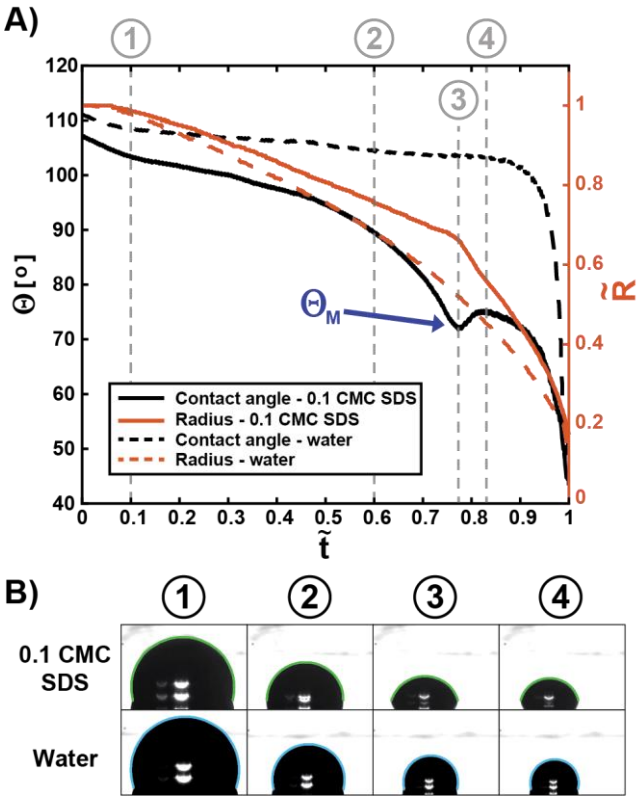


Figure 1. A) Contact angle (black lines) and normalized radius (red lines) of an evaporating 0.5 µL pure water droplet (dashed lines) and of an evaporating 0.5 µL SDS laden droplet (0.1 CMC, solid line) as a function of the normalized time. B) Snapshots of the evaporating SDS laden droplet (top) and of the pure water droplet (bottom) at the different moments in time marked in panel A). The droplet contours, as found by the image processing script, are marked by the green line for the SDS laden droplet and in blue for the water droplet.

EVAPORATION OF SALINE SESSILE DROPLETS: NUMERICAL ANALYSIS WITH A VOF METHOD

C. Lalanne[1,2], F. Lequien[1] and J. M. Fullana[2]

[1]Den-Service de la Corrosion et du Comportement des Matériaux dans leur Environnement (SCCME), CEA, Université Paris-Saclay, F-91191 Gif-sur-Yvette, France; [2]Sorbonne Université, CNRS, UMR 7190, Institut Jean Le Rond ∂'Alembert, Paris, France

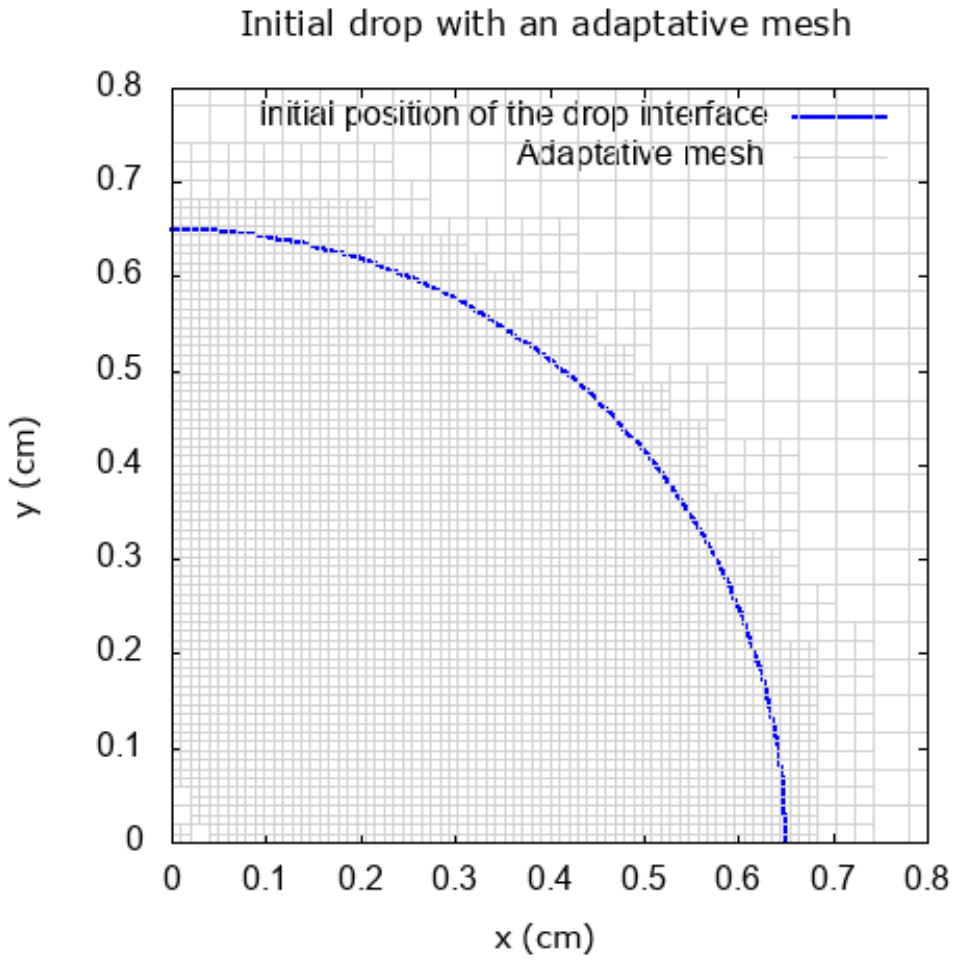
cecile.lalanne@cea.fr

Droplets evaporation is a phenomenon present in everyday life. The residues droplets may leave at the end of evaporation are important to consider, since they testify to the presence of complex mechanisms taking place within such a drop.

The formation of these peripheral residues, also called "coffee stain effect" firstly investigated by Deegan [1], appears in many industrial processes: for instance, in the printing domain [2] or in microfluidics [3]. They are also encountered with any type of liquid mixture with non-volatile particles such as salty water, as in marine atmospheres with sprayed droplets carried by the wind from the sea. The droplet deposits on the surface of exposed structures and becomes saline sessile droplet. In this particular case of the saline sessile droplet deposited on metallic surfaces under atmospheric conditions, the electrolyte solution inside the droplet reacts with the surface, leading to differential aeration corrosion. Evans [4-5] showed that spatially separated cathodic and anodic activities under the droplet are induced by the variation in electrolyte thickness and hence the diffusion of oxygen to the saline solution-metal interface between the droplet edge and its center. The Evans droplet model is specific for static sessile droplets which does not evaporate with time because of particular conditions. However, in nature, droplets commonly evaporate with the variation of the relative humidity and the temperature due to the day/night cycles.

In order to investigate the effect of evaporation on corrosion, we study dynamic sessile droplets. The aim is to enrich and develop a numerical and experimental analysis of the corrosion under droplet, so as to accurately predict its impact on the lifetime of the materials.

We propose a numerical model of evaporation dynamics of salty sessile drops, built with Basilisk, a free software which implements finite volume methods for the Navier-Stokes equations [6]. We introduce a two-phase resolution to describe the salt concentration and the Marangoni flow. We focus on pure water droplets and we perform a parametric study with different droplet geometrical characteristics and atmospherical conditions. In addition, we also consider droplets of various salt concentrations to highlight the impact of salt concentrations, relative humidities, drop sizes and contact angle.



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MARANGONI-ENHANCED SPREADING AND DROPLET DRYING

Lisong Yang and Colin D. Bain

Department of Chemistry, University of Durham, Durham DH1 3LE, UK

c.d.bain@durham.ac.uk

The drying of liquids is a complex process that can hide a wealth of dynamics occurring inside the liquid. The fundamental understanding of the process lends us a key to change the distribution of the deposit, which is of importance in spraying agriculture, spraying coating, inkjet printing and fabrication of (opto)electrical and biological devices. The coffee-ring effect (CRE) is well-known phenomenon in nature and functional printing, manifested as ring-like deposit of solutes or suspended particles at the three-phase contact (TPC) line, leading to non-uniform deposits that limit the quality and performance of final product. There are two essential requirements for CRE: first, the contact line is pinned; second, the droplet is a spherical cap, which is the shape that minimises the free energy of the surface. Simple geometric considerations then result in a capillary flow from the droplet apex to the contact line[1]. Faster solvent evaporation at the contact line compared to the apex enhances this outward capillary flow. Various strategies have been developed to suppress the CRE over the last two decades. Broadly speaking, people alter internal flow pattern and/or steer solutes away from the outward capillary flow to minimise CRE. The strategies heavily focus on *the drying phase* of the droplet.

We present a CRE-control strategy based on Marangoni-enhanced spreading[2] of a pL-droplet of isopropanol (IPA) containing a small amount of a second alcohol, such as 2-butanol. Enhanced evaporation of the more volatile solvent with a lower surface tension (IPA) at TPC leads to outward Marangoni flow from apex to contact line, which result in the enhancement of the spreading and delay of the contact line pinning. Surface stress increases radial outward velocity and leads to deviation from spherical cap. Lack of curvature in resulting 'pancake' means that there are no Laplace pressure gradients to drive capillary flows to the contact line – fluid flow slows down. Uniform deposit can be obtained.

We developed another Marangoni-enhanced spreading strategy based on condensation of water from moist air onto alcohol (IPA) droplet. As condensation is fastest at the contact line and water raises the surface tension of alcohol at the TPC, there is again an outward Marangoni flow from apex to the contact line. The spreading and drying curves depend on relative humidity (RH) with Marangoni-enhanced spreading at RH above ~ 45%. A minimum in the drying time of droplet is observed with increasing RH. The droplet shape is significantly affected with a transformation from spherical shape at low RH (<45%) to pancake-shape at medium RH (45-55%) and conical shape at high RH (>60%). At medium and high RH (>55%), we observed peripheral films and a fingering instability.

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Shrinkage of microdroplets in confined and sparingly miscible media

J. M. Zhang[1], Y. Chen[1], D. Lohse[1] and A. Marin[1]

[1]Physics of Fluids Group, University of Twente, The Netherlands.

Jiaming.zhang@utwente.nl

Multiphase systems with well-controlled interfacial properties are found in a large variety of systems like solvent-extraction applications, single-molecule analysis or even molecular gastronomy. The dispersed phase is shaped as nano- and picoliter droplets, which are typically generated in a confined microchannel. Surprisingly, even the two phases are regarded as immiscible, such droplets can vanish in relatively short times. In many applications such as drying of colloid suspensions[1], microcapsules generation[2] and protein crystallization[3], the droplet shrinkage in the microchannel is not only expected but desired. In all these cases, precise control of the dissolution rate of droplets is crucial. Epstein & Plesset[4] studied theoretically the case of a gas bubble dissolving in an infinite liquid environment, which was later extended to liquid droplets by Duncan & Needham[5]. However, the presence of confined boundaries such as microchannels will affect dissolution behavior of droplets. Unfortunately, related studies are still lacking. In this work, we experimentally and numerically study the dissolution of both single and multiple picoliter droplets in a confined and immiscible liquid medium by using a PDMS-based microfluidic droplet generator. We show in our results the limits in which the classical Epstein & Plesset[5] model can still be used to reproduce the experimental results, and in which ones numerical models to account for the channel geometry are necessary.

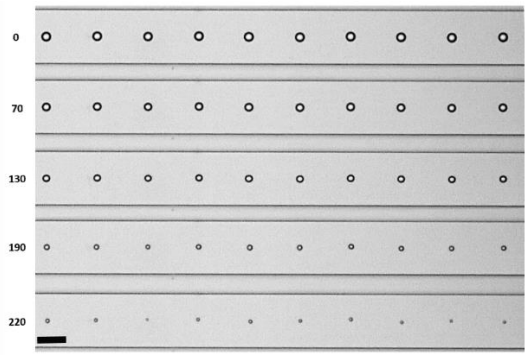

Figure 1. Water droplets of initial size 8.5 µm shrinking in a confined channel filled with silicone oil.

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A DROP DOES NOT FALL ON A STRAIGHT LINE: A RATIONALE FOR THE WIDTH OF STALAGMITES

J. Parmentier[1], S. Lejeune[1], F. Bourges[2], D. Genty[3], J.-C. Maréchal[4], V. Terrapon[1] and T. Gilet[1]

[1]Aerospace & Mechanics Department/University of Liège, Liège, Belgium; [2]Géologie-Environnement-Conseil, Saint-Girons, France; [3]EPOC/University of Bordeaux, Bordeaux, France; [4]BRGM/University of Montpellier, Montpellier, France.

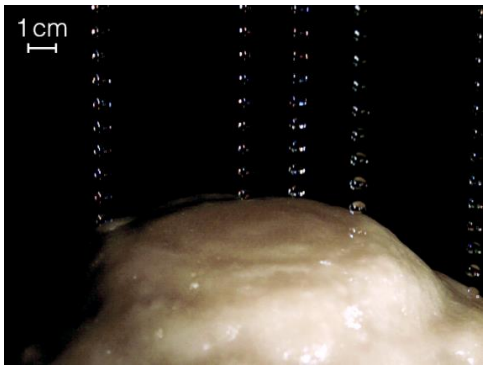
jparmentier@uliege.be

Beyond their outstanding beauty, stalagmites help to understand past climate and hydrology. While the chemical reactions underlying their formation is fairly well understood, little is known about the fluid dynamics at play during their growth. Calcite is brought by drops detaching from the stalactite. The drops fall and impact a very thin film of water that covers the stalagmite. For most stalagmites, the drop velocity is sufficient to generate a splash, with the formation of a corona (fig. - left).

In previous models of stalagmite growth, it was commonly accepted that drops fall on a straight vertical line from the stalactite, thereby feeding the stalagmite film from one central point. Our measurements however revealed that the impact points of drops originating from a single stalactite are scattered over several centimeters (fig. – right). We postulate that drops do not fall straight, but rather follow a wavy trajectory owing to their aerodynamic interaction with the surrounding air.

We have taken high-speed movies of drops splashing on a wide variety of stalagmites in several caves of the south of France. The drop radius, velocity, impact point, radius of the spreading lamella, and stalagmite width were measured by image processing. We show that there is a strong positive correlation between the falling height, the impact point dispersal and the stalagmite width.

We propose a theoretical model of the drop's fall that includes aerodynamic forces. Owing to some lift of random direction that represents the effect of vortex shedding, the drop experiences a random walk, and its impact point on the stalagmite is scattered. All parameters (temperature, composition of drip water and atmospheric partial pressure in CO_2) equal otherwise, for small falling heights, the stalagmite width is mostly set by the maximum spreading of the liquid lamella, while for large falling heights, the stalagmite radius increases proportionally to the impact point dispersal.



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THE ROLE OF THE DYNAMIC CONTACT ANGLE ON SPLASHING

M. A. Quetzeri-Santiago[1], K. Yokoi[2], A. A. Castrejón-Pita[3] and J. R. Castrejón-Pita[1]

[1]Queen Mary University of London, London, UK; [2]Cardiff University, Cardiff, UK; [3]University of Oxford, Oxford; UK

m.a.quetzerisantiago@qmul.ac.uk

A drop impacting onto a solid dry substrate can, among other several results, splash or spread over the solid surface. The result depends not only on the droplet properties and speed, but on a wide range of parameters. Although many studies have aimed at finding scaling arguments to characterise splashing, the exact combination of parameters and their influence have remained elusive.

In this work we perform a systematic study of liquid droplets impacting onto various solid substrates ranging from completely wetting to superhydrophobic. The experimental approach uses high-speed imaging and image analysis to recover the contact angle as a function of the spreading velocity. While our results indicate that wettability plays a small role on the splashing on wettable substrates (validating past works) they also demonstrate that splashing and drop spreading greatly depend on the contact angle for hydrophobic and superhydrophobic substrates. In addition, we show that, under our experimental conditions, liquids spread with a maximum advancing contact angle greater than 87 degrees, regardless of the liquid or substrate properties. Our results also show that existing dimensionless groups, i.e. the splashing parameter (K) and the capillary number (Ca), are not appropriate to characterise the splashing behaviour. Finally, we show that the splashing ratio β [1], appropriately divides the splashing and no-splashing behaviour. These findings have strong repercussions in several industrial environments such as in liquid dispensing, liquid coating, sprays, drug delivery and any other application where splashing can affect coating performance or compromise cleanliness (contamination).

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SPREADING DYNAMICS OF FERROFLUID DROPS POST-IMPACT ON HYDROPHILIC SURFACE

Nilamani Sahoo[1*], Devranjan Samanta[1], and Purbarun Dhar[1]

[1]Department of Mechanical Engineering,

Indian Institute of Technology Ropar

Punjab-140001

[*] *Corresponding author: E-mail- nilamani82@gmail.com*

Abstract

Spreading dynamics of liquid droplets after impact on a solid surface provides important insights to develop and optimize numerous technologies, such as inkjet and 3D printing, spray cooling of hot surfaces, spray coating and sintering[1]. Recently, some studies attempted to explore the spreading dynamics of liquid metal and ferrofluid droplets in presence of a magnetic field[2,3]. These studies investigated the experimental and theoretical maximum spreading after impact for different orientations of the magnetic field. In the present work, the temporal variations of spreading factor (the ratio of the three-phase contact line diameter (D) at any time to the pre-impact droplet diameter (D_0) for stable, water based ferro-colloids, under the influence of vertical magnetic fields. Hydrophilic surfaces (glass) have been considered as the substrate and aqueous Fe_3O_4 nanoparticle (50-60 nm) based colloids have been employed. It is observed that the transient variation of spreading factor ($\psi = \frac{D}{D_0}$) decreases on increasing the magnetic Bond number (Bo_m), as shown in Fig.1.

The magnetic Bond number is expressed as the ratio of magnetic force to interfacial forces ($Bo_m = \frac{B^2 D_0}{\mu_0 \sigma}$) where B, D_0, μ_0, and σ represent the magnetic field induction, initial droplet diameter, magnetic permeability of free space, and surface tension, respectively. The field

decelerates the drop spreading on hydrophilic surface due to vertical orientation of the field. It is seen from Fig.2 that the transient spreading diameter reduces with increasing field strength (manifested though the Bo_m). Fig.3 illustrates the effect of the Weber number on the spreading factor, at a constant magnetic field strength of 0.9T (Bo_m= 22550). While the expected increase in spreading factor with We is noted, the capillary instabilities typical at higher We impact are observed to be absent (inset Fig. 3). It is possible that the magnetic force dampens out the capillary instabilities that arise during the spreading event (for high We impacts).

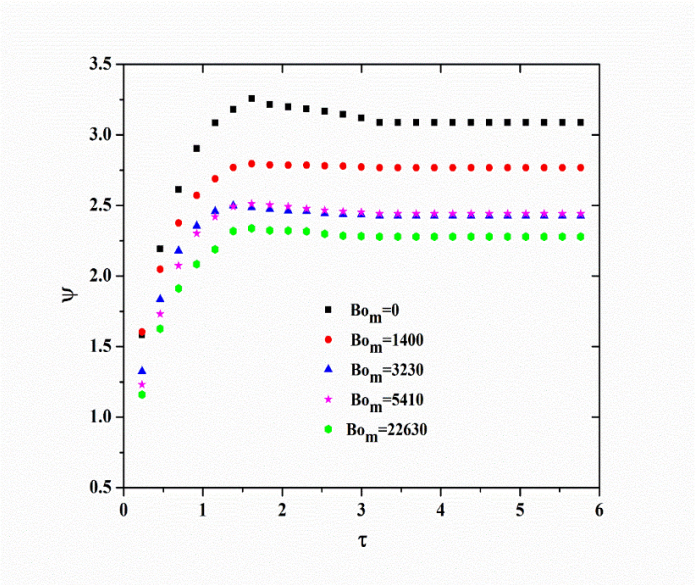

Fig.1 Temporal variation of spreading factor with non-dimensional time ($\tau = \frac{t^*V_0}{D_0}$) where t and V_0 denote time and initial velocity of droplet (before impact).

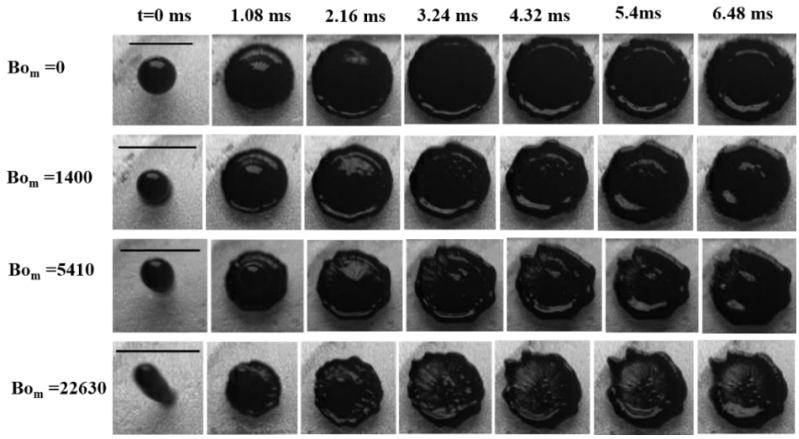

Fig.2 Over-head images of post impact droplet dynamics at We=60 and different Bo_m. The magnitude of scale bar is equal to 7mm for each case.

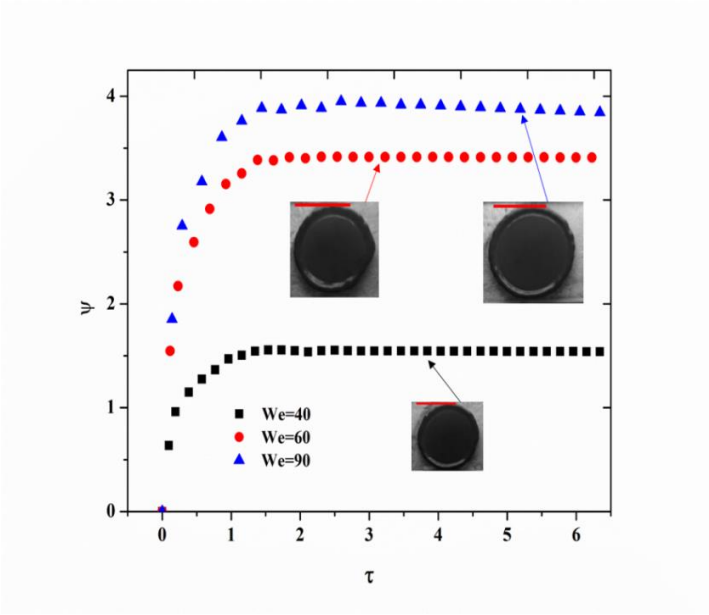

Fig.3 Effect of We on transient spreading factor at Bo_m=22550. The magnitude of scale bar is equal to 7mm. The Weber number is defined as the ratio of inertial force to surface tension force

$$(\mathrm{We}=\frac{\rho V_0^{\,2} D_0}{\sigma}).$$

ACKNOWLEDGEMENT

We would like to thank the Ministry of Human Resource Development, Govt. of India, for the doctoral scholarships.

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SINGULAR JETS FROM THE COLLAPSE OF CRATERS AT A POOL SURFACE

Y. S. Tian[1] and S. T. Thoroddsen[1]

[1]King Abdullah University of Science and Technology (KAUST), Thuwal, Saudi Arabia

yuansi.tian@kaust.edu.sa

The collapse of drop-impact craters can generate a fast singular jet from a dimple which forms at its bottom. A finite-time singularity in the bottom curvature of the crater has in the past been considered as the cause of this singular jet. Self-similar capillary-inertial solutions predict that the radius of the cavity will collapse with time, as $R \sim t^{2/3}$. However, Thoroddsen *et al.*[1] used a high-speed camera to demonstrate that the final collapse has a power-law closer to a purely inertial collapse, with $R \sim t^{1/2}$. They also observed no curvature singularity.

Herein, we use two synchronized high-speed cameras to study the dimple collapse. One is an ultra-fast camera, capable of up to 5 Mfps, which tracks the crater collapse. The second high-speed camera captures the corresponding speed of the singular jet, at up to 400 kfps. The experiment is performed inside a vacuum chamber to control the ambient pressure. The fastest velocity of singular jets is found to be ~ 130 m/s and occurs at reduced pressure without crater pinch-off.

ACKNOWLEDGEMENTS: This research was supported by King Abdullah University of Science and Technology (KAUST) under Grant No. URF/1/2621-01-01.

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Abstract for Droplets 2019

Melting, rounding, bouncing, sticking: The dynamics of magma droplets in volcanic eruptions

Fabian B. Wadsworth & Edward W. Llewellin
Department of Earth Sciences, Durham University.

Explosive volcanic eruptions produce prodigious volumes of ash – micron-to-millimetre sized particles of volcanic glass. Ash is lofted into the atmosphere where it poses a hazard to aviation, and is deposited on surrounding land where it poses numerous hazards to the local population. The ash starts out as droplets of polymeric silicate liquid, produced when magma undergoes fragmentation induced by internal stresses during transport to the Earth's surface. On eruption, the droplets cool rapidly to form ash, but may subsequently be re-melted if ingested into a jet engine. Whilst in the molten state, the droplets are involved in a range of physical processes. They can rapidly round under surface tension, and can impact each other and variably coalesce; this is important because it changes the shape and size distribution of the ash, modifying the hazard that it poses on eruption. In a jet engine, they can impact hot surfaces and either bounce or stick, with obvious consequences for engine operation. We present the results of a programme of experimental and numerical work that characterizes and quantifies these processes. We identify regimes of behaviour based upon dimensionless governing parameters, and compare them with the regimes described in previous work on droplet dynamics. In some cases the state of the art in the physics of droplets is directly applicable to magma droplet dynamics, such as non-isothermal spreading of viscous droplets on surfaces, and capillary rounding. In other cases, magma droplet processes in nature expose gaps in our understanding, such as the process of a viscous jet thinning behind a moving droplet that crosses the Weissenberg number transition, or re-bound of viscoelastic droplets impacting a surface. To illustrate these points, we present the case study of a solid volcanic ash particle being ingested in the engine of a commercial jet aircraft. In this scenario, the ash particle melts in the combustion chamber, rounds under surface tension, and may impact the moving turbine blades where it either sticks and spreads on the surfaces or rebounds and is expelled in the exhaust. For each component of this problem, we present theoretical analysis and, where possible, the results of high-temperature experiments. Finally, we draw analogies between magma droplet dynamics, and processes in ceramic and glass processing industries, motivating future work in these areas beyond the Earth Sciences.

DROP IMPACT ON A FAST-MOVING RIGID SOLID PLATE PROJECTED BY A COILGUN IN A VACUUM CHAMBER

T. Ashida[1], M. Watanabe[1], K. Kobayashi[1], H. Fujii[1] and T. Sanada[2]

[1]Hokkaido University, Sapporo, Japan; [2]Shizuoka University, Hamamatsu, Japan

masao.watanabe@eng.hokudai.ac.jp

In the cleaning processes of manufacturing semiconductor devices, two-fluid spray cleaning technique is widely used[1]: however, the detailed cleaning mechanism has not been clarified yet. In this study, we focus our attention to the elementary process of this cleaning technique, i.e. a single drop impact on a solid surface. An intriguing question we raise is whether we can suppress the occurrence of splash at a higher impact velocity if we only reduce the surrounding gas pressure.

A water drop of radius R=1.1 mm was released from a rest needle in a stainless vacuum chamber with transparent polycarbonate observation windows; then, the free falling drop was brought into collision with a vertically upward flying rigid solid impact plate which was projected by an iron bullet accelerated by a coilgun. The impact plate consisted of a cover glass with a static contact angle of 60˚ and a surface roughness *Ra* of 2.1 nm, adhered to an acrylic plate. The surrounding gas pressure was varied between 1 and 100 kPa, and the relative impact velocity was varied between 4.2 and 33 m/s, where the free falling velocity of the drop was 1.5 m/s at the collision The drop impact on a solid plate with high relative impact velocity and the subsequent splash was recorded by a Shimazu HPV-1 high-speed video camera at a frame rate of 1,000,000 fps with long-distance microscopes at pixel resolution of 16.1 μm/px.

We focus ourselves on the occurrence of splash immediately after the impact, which is widely known as prompt splash. It is well recognized that prompt splash occurs when the value of both the Weber and Reynolds numbers are large[2]; prompt splash on a smooth solid surface presented at high Weber number can be understood as an instant creation of filaments at the first contact of the impact drop with the surface, followed by breakup of fingers resulting in secondary droplets flying obliquely upward[3]. However, this is not the only type of splash that occurs immediately after the impact. Our experimental results illustrate the other types of splashes that occurs immediately after the impact, which leads the suggestion that more detailed classification of prompt splash is greatly required to elucidate the dynamics of high-speed drop impact immediately after the impact. We also examine whether the previously proposed analytical models that predict the splash threshold can explain out results.

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HARVESTING ENERGY FROM WATER DROPLET BASED ON CHARGE TRAPPING AT HYDROPHOBIC POLYMER SURFACES

H. Wu[1], Dirk van den Ende[1], Guofu Zhou[2] and Frieder Mugele [1]

[1] Physics of Complex Fluids Group, MESA+ Institute, University of Twente, 7500 AE Enschede, The Netherlands

[2] South China Academy of Advanced Optoelectronics, South China Normal University, 510006, Guangzhou, China.

hao.wu@utwente.nl, f.mugele@utwente.nl

Natural water motion provides a renewable energy, which we could use but haven't successfully harvested. In this work, we present a novel water droplet energy harvesting technology based on the charge trapping in fluoropolymer films. We find that charges can be spontaneously trapped at fluoropolymer/water interfaces and this phenomenon is enhanced by applying an electric fields. We quantify the trapped charge density and its life time by using electrowetting. When a droplet impact the polymer surfaces with trapping charges, to compensate the trapping charges on the polymer surfaces, the counter charges will migrate between the bottom electrode and the liquid/solid interfaces. In such a way, a current is generated due to the motion of water droplet. The first charge trapping based water energy harvester from rain droplet will be shown. The generated current/voltage/power are found affected by the parameters such as the load resistance and the liquid conductivity. This energy harvesting method can be applied toward not only water droplets, but also other types of moving water contact lines, as encountered e.g. in ocean waves.

BUBBLE ENTRAPMENT AND FINE JETTING DURING IMPACT OF IMMISCIBLE DROP ON A POOL

Ziqiang Yang[1] and S. T. Thoroddsen[1]

[1]King Abdullah University of Science and Technology (KAUST), Thuwal 23955-6900, Saudi Arabia

ziqiang.yang@kaust.edu.sa

We investigate the impact of a drop into a deep immiscible liquid pool, with focus on bubble entrapment and the formation of fine jetting during the crater collapse. We use a heavy perfluorohexane drop which impacts on a water pool, where the two liquids are immiscible. The drop first deforms and stretches out at the bottom of the crater, before collapsing during its rebound, entrapping a small bubble and shooting out fine vertical jet. Several air-entrapment scenarios are observed, such as microbubbles left behind by film ruptures and bubble pinch-off from the dimple. We also see bubbles entering the jet. The entrapment regimes are mapped in the Weber - Froude number space. The size of entrapped bubbles at different impact conditions is scaled with Weber number, We. The formation of fine jetting is also studied. The fastest jets are observed with the speeds of 45 m/s and occur at the multi-dimple case without pinch off. At low Weber numbers a variety of other entrapment phenomena appear.

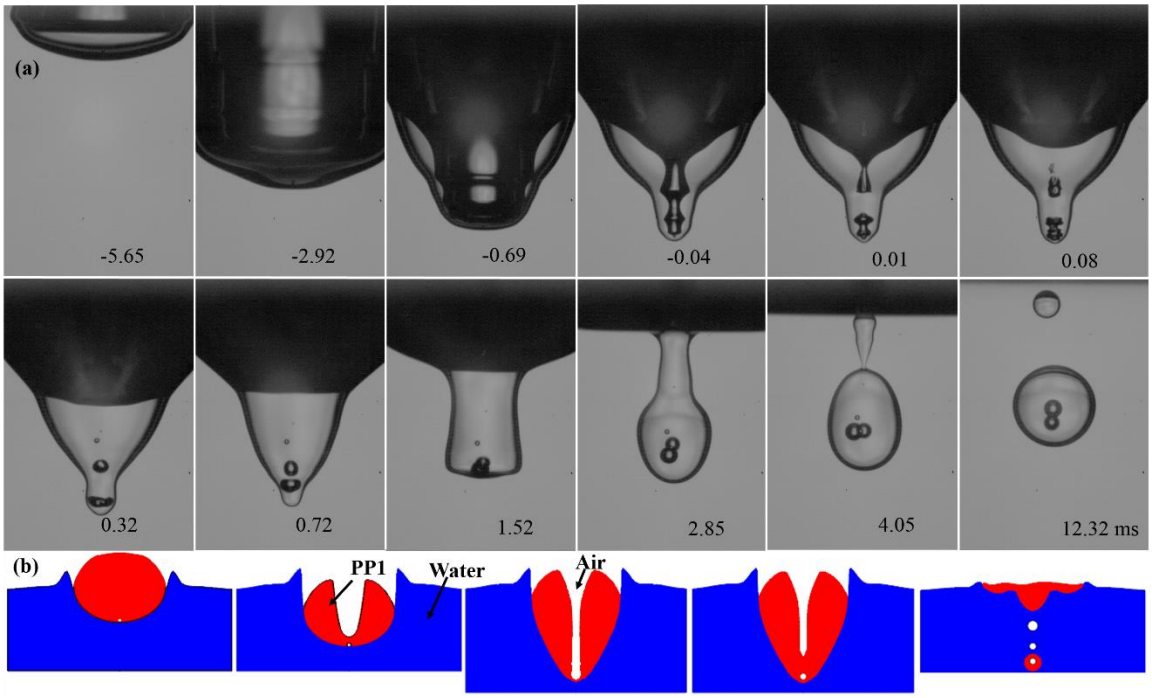

Figure 1. Close-up of typical bubble entrapment process during the crater evolution at U=1.72 m/s, D =1.3 mm, Re = 4215, Fr= 259, We= 493. The video is taken at 75 kfps. Air bubbles are entrapped through the top of the drop.

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

R. Iqbal*, S. Dhiman*, K.S. Jayaprakash* and A.K. Sen
Department of Mechanical Engineering, IIT Madras, Chennai, India, 600036
Email: ashis@itm.ac.in

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[1]. There are inherent limitations associated with the existing various active[2,3] (magnetic- contamination due to the magnetic nanoparticles, electrical and acoustic- peripheral setup for the actuation) and passive[4] (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 (2θ) and groove thickness (δ) submerged in an oil-laden (silicone oil, ρ_{oil}~995 kg/m^3) surface of thickness (h). Upon dispensing an aqueous droplet of diameter (d) on the converging end, the interfacial properties [($\boldsymbol{S_o > 0}$, where $\boldsymbol{S_o = \gamma_w - (\gamma_o + \gamma_{wo})}$), where S_o is the spreading parameter, γ_w, γ_o and γ_{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~2.7 mm) into the diverging groove with an oil-layer thickness of 1.5 mm. **Fig. 1d** shows the coalescence of four droplets (d~3.9 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.

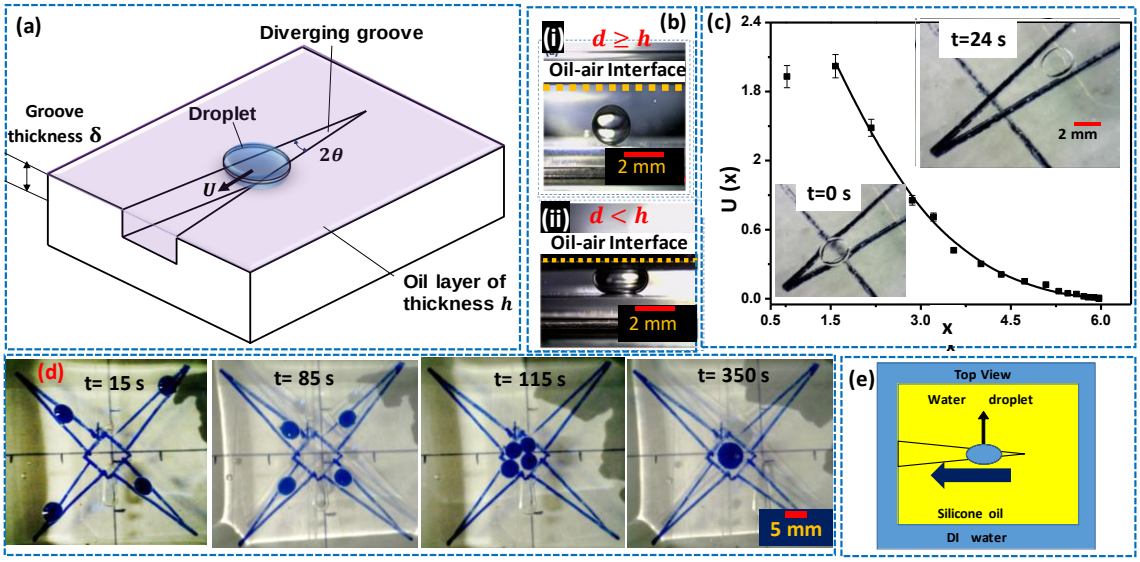

Fig. 1 (a) Schematic of the droplet transport mechanism **(b)** Shapes of water droplet (d~2.7 mm) over an oil layer thickness of (i) 4mm and (ii) 1.5 mm. **(c)** Variation of velocity $U(x)$ with x for $d = 2.7\ mm$, $2\theta = 15^o$, $\delta = 1\ mm$ and $h = 1\ mm$ (**inset:** time-lapsed images of 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

SOME NOVEL ANALYTICAL SOLUTIONS IN MICROFLUIDICS OF SESSILE EVAPORATING DROPLET: MICROFLUIDICS, PARTICLE-FLOW INTERACTION, EVAPORATION

P.V. Lebedev-Stepanov[1,2]

[1]Photochemistry Center of the FSRC «Crystallography and Photonics» RAS, Moscow, Russia;

[2] National Research Nuclear University MEPhI, Moscow, Russia

petrls@yandex.ru

The investigation of evaporating liquid droplets deposited onto flat surfaces is of great importance for physical, engineering and medical applications. The novel analytical solutions of some mathematical problems for droplet with spherical and cylindrical symmetries might have especial fundamental interest. The report proposes several new theoretical results in the field of evaporation and hydrodynamic flows inside a small sessile droplet, as well as near small spherical particles placed into such hydrodynamic flows.

1) Lubrication theory gives the boundary conditions for liquid flow inside the droplet that determined on the surface of the droplet with small contact angle Fig. 1a [1]. These conditions are wrong for arbitrary contact angles (Fig.1 c) [2]. We propose the boundary conditions for general case that includes the arbitrary contact angles . 2) Stokes' flow inside evaporating droplet was described in [3]. New analytical solution for Navier-Stokes' viscous stationary fluid flow near liquid or solid spherical surface is considered. The theory can be applied to the description of viscous flows of a liquid inside a slowly evaporating hemispherical drop fixed on a flat horizontal substrate. The described flows are an exact solution of the stationary linear Navier-Stokes' equation (the assumption of small Reynolds numbers). 3) Yu.O. Popov [4] elaborated the useable model of evaporation of sessile drop of liquid based on J.C. Maxwell diffusion model and electrostatic analogue. Another solution of this problem based on J.C. Maxwell reflection method is proposed.

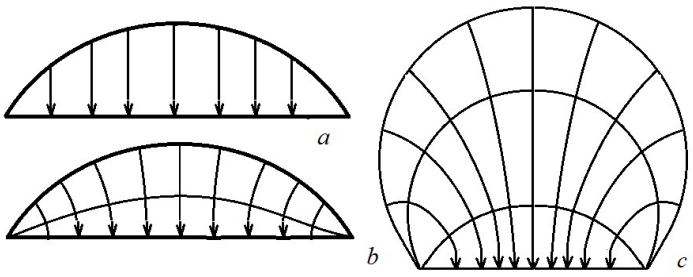

Fig.1

Pictures showing the trajectories of displacement of the points on surface of drying droplets with acute contact angle during evaporation with a pinned contact line: a) lubrication theory (wrong); b), c) our model.

ACKNOWLEDGEMENTS: Author thanks Russian Foundation for Basic Research (grant No 19-02-00937) for supporting this work.

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OSCILLATING SESSILE DROPLET TENSIOMETRY

S. Sharma, B. Hallmark and D.I. Wilson

Department of Chemical Engineering and Biotechnology, University of Cambridge, UK

ss2531@cam.ac.uk

Determining the rheology of complex fluids can be challenging when they are only available in small quantities. This is frequently the case with biological fluids such as the viscoelastic liquid generated by pitcher plants to trap their insect prey: these change properties with age and time so one has to conduct tests on the limited amount available in a single pitcher, in the field. Filament stretching methods [1] offer one route to determining the extensional behaviour of such liquids but quantification of the response requires knowledge of the liquid surface tension. Pendant drop tensiometry [2] requires knowledge of the fluid density, which can be challenging outside the laboratory.

One way to measure a liquid's surface tension is vibrate a small sessile droplet (Bond number < 1) and correlate its resonance frequency with the surface tension. Existing treatments [3, 4] of this phenomenon have employed a simplified one-dimensional capillary wave approach and predict the resonance frequencies of different modes of an oscillating droplet with first order approximations. We present a more general treatment for solving the wave equation inside the domain of a sessile droplet, for contact angles ranging 0 to 180°, with no-slip and free surface boundary conditions. Results are presented for Newtonian liquids. The frequencies predicted lie within 4% of reported experimental values. As a result, the model can be used to predict the surface tension of a sessile droplet. Using different base materials allows the wettability of the liquid to be determined.

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PATH SELECTION OF A SPHERICAL CAPSULE IN A MICROFLUIDIC BRANCHED CHANNEL

Z. Wang[1], A. V. Salsac[2], D. Barthès-Biesel[2], W. Wang[1] and Y. Sui[1]

[1]School of Engineering and Materials Science, Queen Mary University of London, London, E1 4NS, UK ; [2]Université de Technologie de Compiègne, CS 60319, 60203 Compiègne, France

y.sui@qmul.ac.uk

Path selection of a deformable capsule suspended in an external fluid flowing through a branched microchannel is an important problem that is relevant to understand blood-related flows in the microcirculation, aerosol particle deposition in respiratory airways, or to develop microfluidic technologies to separate or enrich suspensions. Here we computationally study the motion of an initially spherical capsule flowing through a straight channel with an orthogonal lateral branch, using a three-dimensional immersed-boundary lattice-Boltzmann method. The capsule is enclosed by a strain-hardening membrane and contains an internal fluid of the same viscosity as the fluid in which it is suspended. Our primary focus is to study the influence of the geometry of the side branch on the capsule path selection. Specifically, we consider the case where the side branch cross-section is half that of the straight channel and study various bifurcation configurations, where the branch is rectangular or square, centred or not on the straight channel axis. The capsule is initially centred on the axis of the straight channel. We impose the flow rate split ratio between the two downstream branches of the bifurcation. We summarise the results obtained for different capsule-to-channel size ratios, flow Reynolds number Re (based on the parent channel size and average flow speed) and capsule mechanical deformability (as measured by the capillary number) in phase diagrams giving the critical flow rate split ratio above which the capsule flows into the side branch. A major finding is that, at equal flow rate split between the two downstream branches, the capsule will enter a branch which is narrow in the spanwise direction, but will not enter a branch which is narrow in the flow direction. For $Re \leq 5$, this novel intriguing phenomenon primarily results from the background flow, which is strongly influenced by the side branch geometry. For higher values of Re, the capsule relative size and deformability also play specific roles in the path selection. The capsule trajectory does not always obey the classical Fung's bifurcation law [1], which stipulates that a particle (in Fung's case, a red blood cell) enters the bifurcation branch with the highest flow rate. We also consider the same branched channels operating under constant pressure drop conditions and show that such systems are difficult to control due to the transient additional pressure drop caused by the capsule. The present results obtained for dilute systems open new perspectives on the design of microfluidic systems, with optimal channel geometries and flow conditions to enrich cell and particle suspensions.

ACKNOWLEDGEMENTS: The authors thank the Royal Society for supporting this work.

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DIGITAL MICROFLUIDICS ON FLEXIBLE SURFACE ACOUSTIC WAVE DEVICES

R. Tao[1], X. Tao[2], Y. Wang[2], H. Torun[1], G. McHale[1] and Y. Q. Fu[1]

[1]Northumbria University, Newcastle upon Tyne, UK; [2]Zhejiang University, Hangzhou, China

r.tao@northumbria.ac.uk

Abstract: One of the most challenging technological barriers for wearable healthcare devices is to deliver the efficient sensing and actuating (fluid/sample manipulation) functions on a flexible substrate. These devices need to be able to transport and adhere liquids and sense their properties when in widely differing physical orientations. Flexible acoustic devices with thin film technologies on flexible substrates have the potential for both sensing and fluid manipulation functions[1–4]. However, acoustic wave propagation is adversely and significantly influenced by commonly used flexible substrate materials such as polymers, resulting in significant attenuation and dissipation of acoustic waves and energies. Moreover, most hydrophobic coatings and textures which provide ease of liquid transport along the device surface also reduce the ability to adhere liquids[5,6]. Here, we report a novel concept of highly efficient, programmable, flexible, bendable and re-deformable 3D acoustofluidic functions using SAW devices fabricated on thin aluminium substrates. This targets one of the most challenging issues in flexible acoustofluidics, which is the compromise for performance between deformability and plasticity, by using thin Al foils/plates and its hierarchical surface created on ZnO/Al plates. In addition, by altering the topography and chemical properties of the ZnO surface, we create a surface structure which is shear hydrophobic, but tensile hydrophilic. This enables easy transport of liquid droplets along the surface, whilst simultaneously enabling droplets to be adhered when the substrate is angled or inverted. We systematically characterised our devices on ZnO/Al substrates and compared their performance against their counterparts on ZnO/Si. We demonstrate significant improvement in key microfluidic parameters including small receding contact angle, small contact angle hysteresis, lower threshold power, higher pumping acceleration and speed. We then demonstrate the capability of 3D droplet pumping on curved surfaces realized by re-deformable ZnO/Al based SAW devices for wearable point-of-care applications.

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Fabrication of Monolayer Polymeric Particles by Inkjet Printing of Uniform Emulsion Produced by Microfluidics

Y. Wang [1], L. Yang[2] and J. Shi[2], R. Deng*, C.D.Bain*

Department of Chemistry, Durham University, Stockton Road, Durham DH1 3LE, U.K.

**E-mail: rhd.deng@gmail.com*

**E-mail: c.d.bain@durham.ac.uk*

ABSTRACT: Emulsion solvent evaporation is a well-established method for generating microparticles from solutions of polymers in volatile organic solvents dispersed in an aqueous medium. Previous work[1] has shown that this approach can also be used to deposit particles by inkjet printing where the particles are formed during the drying of a liquid ink on a substrate. The particle size distribution, however, was very broad. Here we demonstrate that inkjet printing of oil-in-water emulsions produced by microfluidics can generate micron-sized particles with a narrow size distribution (coefficient of variation ~ 10%) and that these particles can self-assemble into ordered arrays with hexagonal packing (Figure 1). The conditions under which drops can be printed with a minimum of break up and coalescence of the oil droplets in the emulsion are explored. Factors affecting the size of the particles and the morphology of the deposit are described. This study uses polystyrene in dichloromethane as a model system, but the approach can be generalized to the production of structured and functional particles. Further research will be focusing on observing the encapsulation process of polymeric microcapsules.

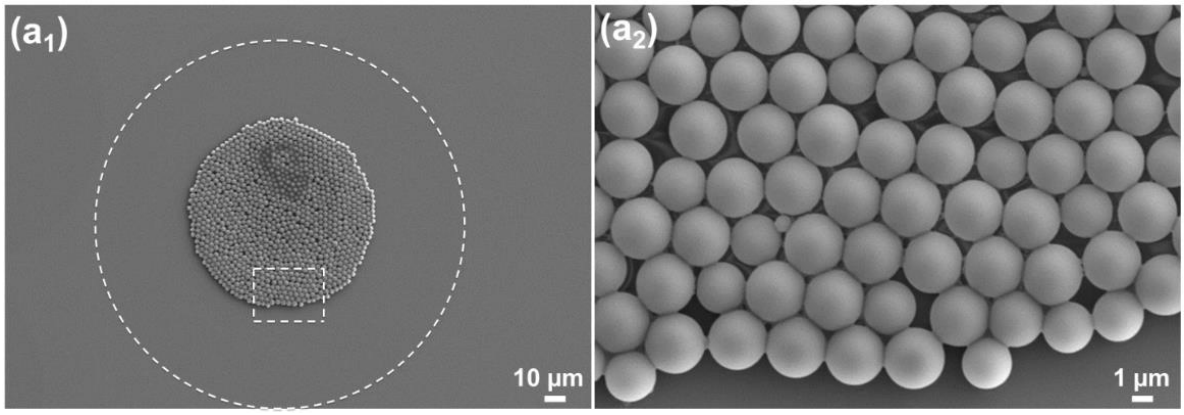

Figure 1. SEM image shows the morphology of the deposit with a monolayer of polystyrene particles.

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MAPPING WETTING VARIATIONS ON SURFACES WITH PICONEWTON FORCE AND MICROMETRIC LATERAL RESOLUTIONS

D. Daniel[1], X. Koh[1, 2], Y. Florida[2], A. Sng[1] and N. Tomczak[1]

[1]Institute of Materials Research and Engineering, Singapore; [2]University of Twente, Enschede, Netherlands

daniel@imre.a-star.edu.sg

To characterize the wetting properties of surfaces, the most common approach is to place a *millimetric*-sized droplet on a surface and measure its contact angles. The adhesion and friction forces can then be inferred *indirectly* using Furmidge's relation. While they are easy to implement, contact angle measurements lack the force sensitivity and lateral resolutions to resolve wetting variations on a surface [1-3]. Here, we attach a *micrometric*-sized droplet to an Atomic Force Microscopy (AFM) probe to *directly* measure its adhesion and friction forces to a surface. For water droplets on lotus-effect surfaces and oil droplets on polyzwitterionic brush surfaces (under water), it is possible to map wetting variations with force resolutions of 100 pN and lateral resolutions of 1 µm, better than state-of-the-art techniques. With the proposed AFM setup, it is also possible to study detailed pinning-depinning dynamics as the droplet approaches and retracts from the surface. Our novel AFM imaging mode will greatly extend our understanding of surface science and potentially allow us to probe the wetting properties of surfaces down to the molecular details.

ACKNOWLEDGEMENTS: The authors thank the Agency for Science, Technology and Research (A*STAR) for supporting this work under the PHAROS Advanced Surfaces Programme (grant number 1523700101, project number IMRE/16-2P1203). D. Daniel acknowledges the financial support of the SERC Early Career Award, project number SC25/18-8R1111.

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Experimental investigation of hydrodynamics and heat transport during drop impact on a heated wall covered with electrospun nanofiber mats in a saturated vapor atmosphere

A. Gholijani, T. Gambaryan-Roisman and P. Stephan

Institute of Technical Thermodynamics, Technische Universität Darmstadt, Darmstadt, Germany;

gholijani@ttd.tu-darmstadt.de

The present work is devoted to experimental investigation of hydrodynamics and heat transport during drop impact on a heated wall covered with nanofiber mats. Polyacrylonitrile (PAN) is electrospun on a heater surface, so that the thickness of the nanofiber mats ranges from 25 to 85 µm and the cross-sectional diameter of the fibers is of the order of several hundred of nanometers. The heater consists of a 400 nm chromium (Cr) as a joule heating layer on top of a 400 nm high emissive chromium nitride (CrN) layer. Both layers are sputtered onto an infrared-transparent glass (CaF_2). A syringe pump generates drops of refrigerant FC-72 (perfluorohexane) in a temperature-controlled test cell, kept under pure vapor atmosphere of working fluid.

The temperature field at the solid-fluid interface is directly monitored by an infrared camera. Finite volume method is applied to compute the time-dependent heat flux distribution at the solid-fluid interface from the temperature field by solving transient three-dimensional energy equation. The time-dependent heat flow is assessed by integration of local heat flux over the drop footprints. The temporal evolution of contact radius is derived by image post-processing. The initial drop diameter and the impact velocity have been kept constant at 0.94 mm and 0.46 m/s, respectively. The cell temperature is set to 54 °C corresponding to system pressure of 0.95 bar. The measured wall superheat ranges from 2 to 15 K.

Figure 1 shows the temporal evolution of the contact line radius (left) and cumulative heat flow (right) during drop impact on a bare heater and on a heater covered by 25 µm thick nanofiber mat. As shown, the drop impingement event on the impermeable surface is subdivided into three phases: spreading, receding and sessile drop. However, the presence of nanofiber eliminates the receding phase, since the contact line is pinned at the end of spreading phase. This agrees with the results on water drop impact and evaporation on a nanomat-coated heated steel foil in air [1]. If the heater is covered with nanofiber mat, the cumulative heat flow is almost two times larger at 150 ms after impact. However, during iintial stages of impact, it is slightly lower than case of bare heater.

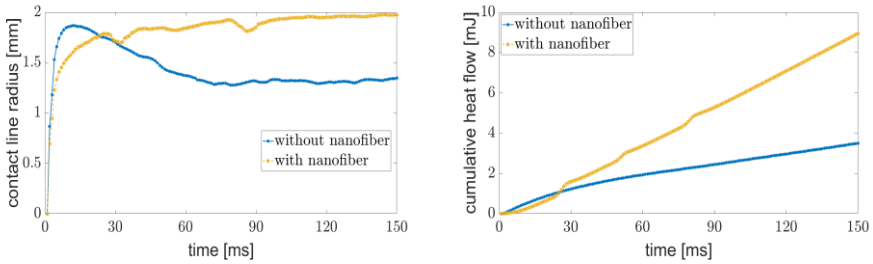

Fig. 1. Contact line radius (left) and cumulative heat flow (right) during the drop impact on a heated wall without and with 25 µm thick nanofiber (ΔT=3 K).

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INVESTIGATION OF PATTERNS FROM DESSICATED DROPLETS OF PHARMACEUTICAL PREPARATIONS TO STUDY THE IMPACT OF SUCCUSSION DURING DILUTION

M. O. Kokornaczyk[1], S. Würtenberger[2] and S. Baumgartner[1,3]

[1] Society for Cancer Research, Hiscia Institute, 4144 Arlesheim, Switzerland
[2] Scientific & Regulatory Affairs, Hevert-Arzneimittel GmbH & Co. KG, 55569 Nussbaum, Germany
[3] Institute of Integrative Medicine, University of Witten/Herdecke, 58313 Herdecke, Germany

m.kokornaczyk@vfk.ch

It is known that strong shaking of pharmaceuticals may cause degradation of some substances (including proteins) and also may cause particle formation, although the underlying mechanisms are not yet fully understood. Investigations into the impact of mechanical energy on pharmaceuticals are therefore important. The droplet evaporation method (DEM) has been previously applied to characterize phenomenological aspects of patterns obtained from desiccated droplets of pharmaceutical preparations (1,2).
In the present experimentation we analyzed five pharmaceutical preparations (*Echinacea* 10^{-2}, *Baptisia* 10^{-3}, *Baptisia* 10^{-4}, *Luffa* 10^{-4}, and *Spongia* 10^{-6}) manufactured following the prescriptions of the European Pharmacopoea (3) for homeopathic preparations, i.e. each dilution step in a ratio 1:9 was followed by shaking; this procedure was repeated until the desired dilution was reached (e.g. for a dilution 10^{-3} three times). The samples were prepared in three variations each: skaken by the application of (i) 100 or (ii) 10 succussion strokes between the consecutive dilution steps, and (iii) prepared as an unsuccussed, only gently mixed control (without foam generation). The stability of the experimental set-up was examined by positive control experiments. DEM experimental protocol (2) consisted in the evaporation of droplets of the analyzed preparation *per se*, placed on microscope slides and under controlled conditions. The resulting patterns were photographed in dark field. The images (4'026 in total) were subjected to a computerized pattern evaluation consisting of the measurement of their grey level distribution, aspects of texture, and fractality.
In all five analyzed preparations, the succussed (100 and 10 times) and unsuccussed variants could be significantly differentiated by means of at least three of the five analyzed image evaluation parameters. Significant differences between all variants were found in two preparations in one parameter per preparation. In most cases, the application of succussion strokes reduced homogeneity and complexity of the DEM patterns and increased the gaps between the structures. The control experiments showed reasonable experimental model stability.
Phenomenological studies focusing on the mechanical impact on pharmaceutical preparations could be important also for manufacturing and handling of liquid drugs, mixing of solutions, and the transportation of pre-filled, ready to use injections.

The authors declare that there is no conflict of interest.

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SLIDING BEHAVIOR OF A DROPLET ON SURFACES HAVING MICRO-GROOVES

Manish Kumar[1], Rajneesh Bhardwaj[1] and Kirti Chandra Sahu[2]

[1]Indian Institute of Technology Bombay, Mumbai, India; [2]Indian Institute of Technology Hyderabad, Sangareddy, India

Email: manishk.iitb@gmail.com

We study the sliding behaviour of droplets migrating along the transverse and longitudinal directions of the micro-grooved surface experimentally. The sliding angle of the droplet is measured by gradually tilting the surface using a rotating stage mechanism until the droplet starts to move. The dynamics is recorded using a high-speed camera. When droplet migrates in the transverse direction, a "Stick-slip" type of motion is observed, i.e. the advancing and receding contact lines attach and detach sequentially. In contrary, In the case of longitudinal migration of the droplet, we did not see any significant change in the relative position of advancing and receding contact lines; the dynamics is found to be similar to the motion of droplet on a smooth surface. Figure 1 shows the effect of direction of migration and pitch of the micro-groove surface on the sliding angle of the droplet. It is found that as we increase the pitch of the microgrooves, the sliding angle of a droplet decreases in both directions of migration. Sliding angle also decreases with the increase in the droplet volume, as the gravitational component increases in the direction of inclination. The sliding angle in the transverse direction of moigration is found to be larger than that observed in the longitudinal migration of the droplet. A theoretical analysis is also conducted to corroborate our experimental findings. The trends observed in our experiments agree with the predictions of the theoretical model. This study is recently published in Langmuir [1].

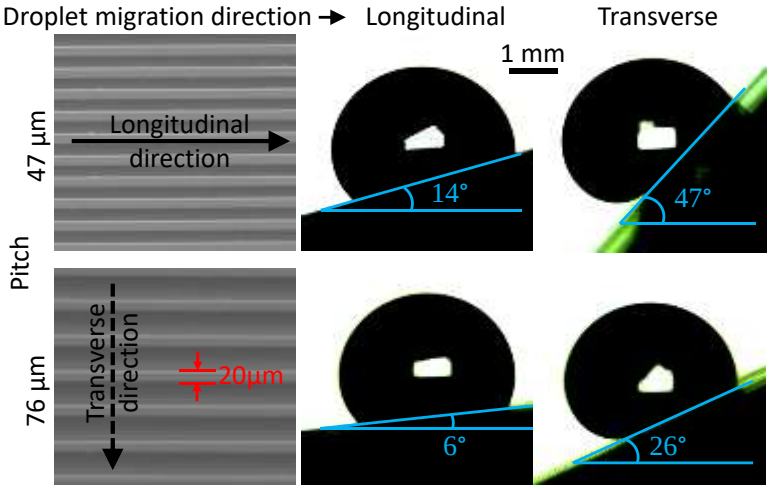

Figure 1: Droplet sliding behavior on micro-grooved surfaces with different pitch and direction of motion. The first column of images shows SEM of micro-grooved surfaces, and the second and third column shows the side visualization of droplets sliding on the micro-grooved surface. Sliding angles of a droplet on these surfaces are indicated.

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SELF-PROPELLED DROPLET TRANSPORT ON LIQUID SURFACES

G. Launay[1], M.S. Sadullah[2], G. McHale[1], R. Ledesma-Aguilar[1], H. Kusumaatmaja[2], G.G. Wells[1]

[1]Northumbria University, Newcastle-upon-Tyne, UK; [2]Durham University, Durham, UK

gaby.launay@northumbria.ac.uk

The ability to direct droplets on flat surfaces has many practical applications in microfluidics, bio-assay and analytical chemistry. We herein show that efficient droplet propulsion can be achieved by using dual length scale roughness to create slippery surfaces with a gradient of wettability. Those surfaces have a unique combination of advantages, including long distance transport, accurate velocity control and the ability to capture droplets prior to motion (Figure 1).

To transport a droplet, a surface needs to provide both a force propelling the droplet in a specific direction and a high droplet mobility. Here, the propelling force is generated by a non-uniform micro-scale roughness, that creates an imbalance in the droplet contact angles [1]. The high mobility is ensured by the addition of a layer of nano-scale roughness imbibed with oil [2]. This layer prevents the contact between the droplet and the solid, hence creating a slippery "liquid surface". The resulting surfaces can propel droplets on several times their diameter, as well as against gravity (Figure 1). Impacting droplets can also be captured prior to motion (Figure 1b) because of the strong vertical adhesion.

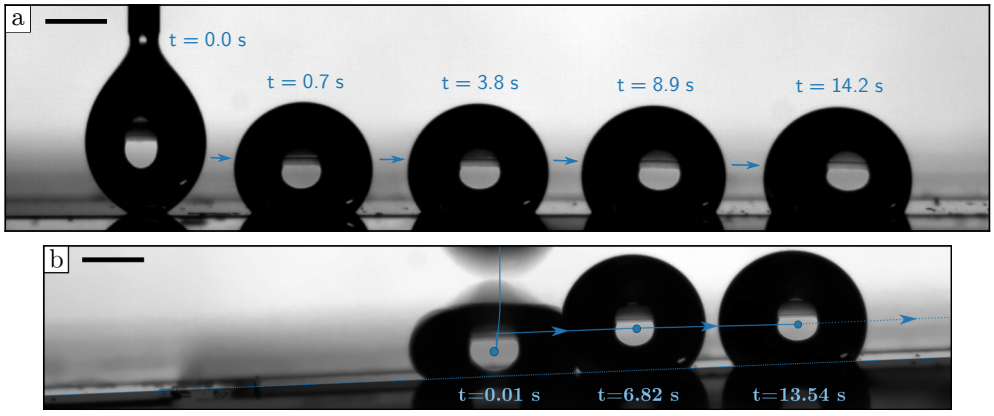

Figure 1: Droplet self-propulsion on shaped liquid surfaces. (a) Droplet moving by several times its size on a flat surface. (b) Droplet self-propulsion on a tilted surface. Because of the strong vertical adhesion, the droplet is successfully captured (no rebound).

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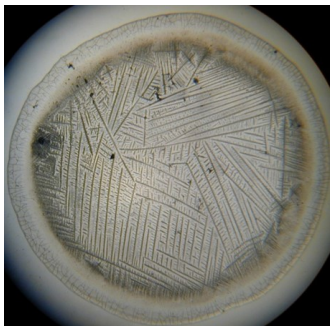
MORPHOLOGICAL ANALYSIS OF IMAGES OF DRIED DROPLETS OF SALIVA FOR DETERMINATION THE DEGREE OF ENDOGENOUS INTOXICATION

P.V. Lebedev-Stepanov[1,2,3], Buzoverya M.E.[3,4], Vlasov K.O.[3,5]and Potekhina Yu.P.[3,5]

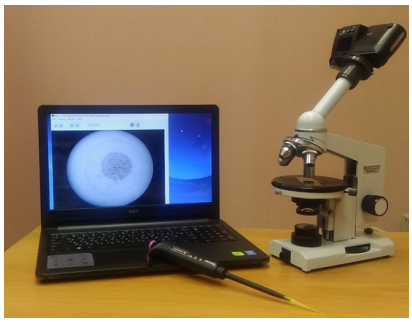
[1]Photochemistry Center of the FSRC «Crystallography and Photonics» RAS, Moscow, Russia; [2]National Research Nuclear University MEPhI, Moscow, Russia ; [3]Morphotest LLC, Satis, Nizhniy Novgorod region, Russia ; [4]Sarov State Physics Technical Institute, Sarov, Russia ; [5]Privolzhsky Research Medical University, Nizhniy Novgorod, Russia

petrls@yandex.ru

Endogenous intoxication (EI) is an integral pathology which gives the general information about state of health of the patient. There is a correlation of biochemical data (costly, require long-term invasive studies) and morphological indicators based on image analysis of the visual image of a droplet of saliva dried in standard conditions (a simple and cheap non-invasive procedure). It is important to develop cheap, fast, non-invasive, computerized morphological methods for a medical diagnostics of EI based on the unique clinical experience obtained in the past decade to apply for mass rapid screening and monitoring of patients during the clinical examination, and also for individual checkup the health of any patient. The method is proposed and laboratory setup is elaborated for determination of the EI by determination of morphological properties of dried pattern of evaporated saliva droplet. The set of microphotographs of the salivary dried patterns is formed taking into account the degree of EI [1]. Criteria for expert evaluation of the EI degree from images of saliva dried pattern image are proposed. For the first time, computer software was created and tested to determine the EI degree. This makes it possible to process a large number of digital images of saliva patterns, which allows evaluating the monitoring of the patient health (Fig.1). Proposed method is a combination of bioinformatics and biochemistry approaches for obtaining diagnostic information from a morphological analysis of standardized dried patterns of saliva.



a



b

Fig.1. a: dried pattern of the saliva which corresponds to the 2-nd degree of EI (real size of the sample is 7 mm); b: laboratory setup.

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Transient snapping gel structure under geometrical confinements

Y. Liu[1,2], A. Sun[2], Z. Li[1,2], Y. Li[2], B. B. Xu[2] and H. Lu[1]

[1]Harbin Institute of Technology, Harbin, China; [2]Northumbria University, Newcastle, UK

Email: yingzhi2.liu@northumbria.ac.uk

The formation of well-defined and functional three-dimensional (3D) structures by buckling of thin sheets subjected to spatially nonuniform stresses is common in biological morphogenesis and has become a subject of great interest in synthetic systems, as such programmable shape-morphing materials hold promise in areas including drug delivery, biomedical devices, soft robotics, and biomimetic systems. Given their ability to undergo large volume changes in swelling in response to a wide variety of stimulus, hydrogels have naturally emerged as a key material in this field.

This work describes the design and fabrication of a soft robotic gripper technology by using the stimuli-responsive hydrogel materials and structural design, which enable instability morphology to be spatially distributed and generated at the surface of gel structure. The investigation considers non-Euclidean shell theory to understand how curvature stimuli induce mechanical instabilities in spherical shells (preliminary results in Fig.1), as well as snap-through shape transitions depending on the amount of stimulus and the deepness of the shell. Autonomous snapping efficiency is assessed by plotting the normalized width change as function of the time. We characterize the reproducibility of the snapping process, and the hysteresis of each transition. We quantitatively understand the snapping by making a scaling analysis of the dominant factors and construct an analytical phase diagram that captures well the transitions found in experiments. A conceptual gripper design will be designed to demonstrate the autonomous gripping under the different stimuli, i.e. salt concentration, PH, temperature, IR, etc.

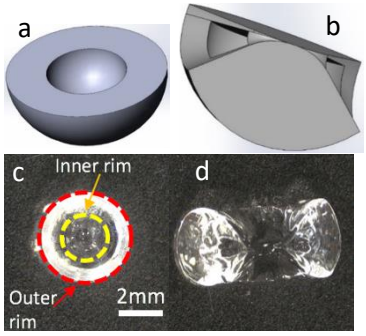

Fig.1 Schematic of hemisphere shell at (a) initial state and (b) snapping state; Optical microscopic images of hemisphere shell at (c) initial state and (d) swelling induced snapping state.

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Simulation of droplet spreading on wettability confined diverging tracks based on a lattice Boltzmann multiphase model for fluid with high-density ratio

Rongye Zheng[1,2], Da Xu[3], Haihu Liu[3], Jinju Sun[3] and Yonghao Zhang[2] (Underline presenting author)

[1]School of Mechanical Engineering and Automation, Fuzhou University, Fuzhou, China ; [2]Department of Mechanical and Aerospace Engineering , University of Strathclyde, Glasgow, UK ; [3]School of Energy and Power Engineering, Xi'an Jiaotong University, Xi'an, China

ryzheng@fzu.edu.cn

Spontaneous droplet transport on wettability confined tracks is important in various applications, such as rapid transport and mixing of droplets in microfluidic devices, enhanced dropwise condensation, etc. We carried out simulations of droplet spreading on wettability confined diverging tracks based on a lattice Boltzmann multiphase model that allows for fuild with high-density ratio [Computers & Fluids 2013, 73, 175-186]. A good agreement between the simulation and the experiment [Langmuir 2018, 34, 1899−1907] is obtained. The overall energy gradient is found to be approximately linear with the spreading speed. The influences of the structure parameters on the spreading characteristics are also investigated, hopefully to gain a better understanding on the spreading behavior of a droplet on wettability confined diverging tracks.

ACKNOWLEDGEMENTS: The authors thank EPSRC (Engineering and Physical Sciences Research Council) under grant number EP/L00030X/1, EP/K000586/17 and China Scholarship Council for supporting this work.

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SPREADING DYNAMICS OF WATER ON SOLUBLE THIN FILMS PATTERENED WITH HYDROPHOBIC DROPLETS

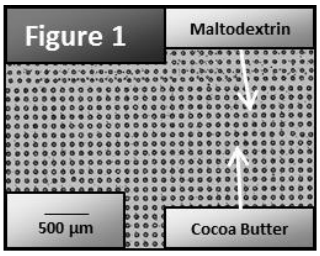
S. S. Melides, J.E. Brown, J.L. Keddie, M. Ramaioli

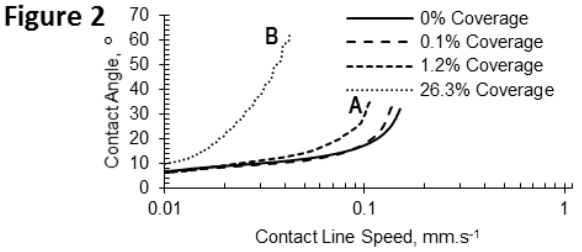
University of Surrey, Guildford, United Kingdom

S.Melides@surrey.ac.uk

Fat located at the surface of food powders is well understood as the cause of poor dissolution and wetting of many food products, such as instant beverages, soups and sauces, and particularly dairy formulas. The wetting of homogeneous soluble films has been studied by previous authors[1,2] who found that evaporation and condensation is the dominant mechanism of hydration ahead of the moving contact line of a sessile drop. Both have an impact on the contact angle (θ) and contact line speed (*U*), depending on the film thickness and the sorbed water volume fraction.

This research introduces surface heterogeneity by inkjet printing hydrophobic droplets (cocoa butter) onto hydrophilic films (maltodextrin DE19) to observe the impacts on the spreading of water. Droplets of cocoa butter (diameter 60 µm) were deposited in controlled area fractions (0-90%) of the film surface, as either regular (Figure 1) or randomised patterns, and the water spreading dynamics were evaluated (2 µL or 10 µL droplets at 55% RH). Increasing the area coverage of cocoa butter increased θ, for a given *U*. A surface coverage of 26.3% was found to halve *U* after 10 s of spreading (Figure 2). Image analysis of the water droplet edge shows that water surrounds fat and alters both the contact line shape and length. High fat coverages were found to pin the water on the surface and strongly slow down its spreading.





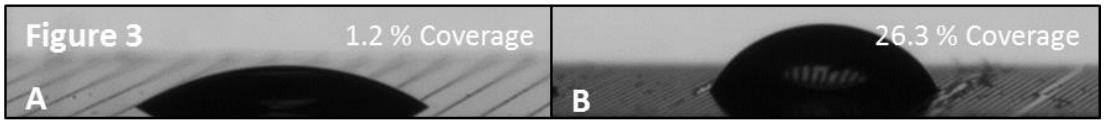


Figure 1. Micrograph of a typical heterogeneous thin film of maltodextrin DE19 (26.3% coverage by regular printing of cocoa butter). **Figure 2.** θ vs U for 0, 0.1, 1.2 and 26.3% area coverage of 60 µm droplets of deposited cocoa butter on 275 nm maltodextrin films at 55% RH. **Figure 3.** Images of point A and B in Figure 2 of water droplets spreading on surfaces with (A) 1.2% and (B) 26.3.% coverage after 0.5 s after depositing the water droplet.

ACKNOWLEDGEMENTS: The authors thank the EPSRC for partial funding of S.S.M's PhD Study.

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HIERARCHICAL LUBRICANT INFUSED SURFACES WITH ENHANCED DROPLET MOBILITY

D. Orejon[1,2], Y. Maeda[3,4], F. Y. Lv[5], P. Zhang[5], Y. Takata[2,3]

[1]Institute for Multiscale Thermofluids, School of Engineering, University of Edinburgh, EH9 3BF, UK
[2]International Institute for Carbon-Neutral Energy Research (WPI-I2CNER), Kyushu University, 744 Motooka, Nishi-ku, Fukuoka 819-0395, Japan
[3]Department of Mechanical Engineering, Kyushu University, 744 Motooka, Nishi-ku, Fukuoka 819-0395, Japan
[4]Kubota Corporation Research & Development, Sakai Plant, Osaka 590-0823, Japan
[5]Institute of Refrigeration and Cryogenics, Shanghai Jiao Tong University, Shanghai 200240, China

d.orejon@ed.ac.uk

Dropwise condensation DWC has received increasing exponential attention in the past decades steaming from novel fabrication and coating techniques, which enables the production of a wide range of surface structures and functionalizations with unique properties. Amongst these surfaces, lubricant infused surfaces LIS have been proposed due to their excellent repellency and low-adhesion, which make them ideal in anti-fouling, microfluidcs, thermal management, etc. [1].

In this work, we investigate the condensation perfomance of different LISs with surface structure ranging from hierarchical micro-/nano- to solely nano-structured ones. Superhydrophobic surfaces before impregnation were prepared as in the work of Zhang *et al*. [2]. Then, LISs were subjected to condensation experiments in an environmentally (temperature and relative humidity) controlled chamber. The dynamics of condensation were imaged for ca. 4 hours and special attention was paid to the dynamics of droplet growth and droplet shedding. On one hand, no major differences were found during droplet growth and/or on the breath figures during both growth and coalescence with neighbouring drops, which is consistent with literature [3]. On the other hand, when looking into the droplet shedding performance, important differences are found depending on the surface structure present underneath the oil and the condensate. The size and the velocity of the shedding droplets was found to decrease and increase, respectively, in the presence of hierarchical micro-/nano-structures, when compared to solely nano-structured LISs. On hiearchical LISs, the small nature of the shedding droplets with sizes in the sub-millimetre range, enhances the frequency of refreshed area for re-nucleation and growth, which is characteristic of hight heat transfer.

To conclude, we propose a revisited surface force analysis to account for the different shedding performance depending on the surface structure underneath the condensate. Moreover, we compute the different droplet growths (solely due to condensation, due to coalescence and due to coalescence during shedding) coupled with the varius shedding performances (size and velocity of shedding droplets) to demonstrate the greater heat transfer performance of hierarchical LISs.

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MATERIALS BASED ON POLYMER-PARTICLE-OIL EMULSIONS: DROPLET STABILISATION AND EVOLUTION

Dominika Zabiegaj, Farzaneh Hajirasouliha

Smart Materials and Surfaces Laboratory, Faculty of Engineering and Environnent, Northumbria University, Newcastle upon Tyne, NE1 8ST, United Kingdom
dominika.zabiegaj@northumbria.ac.uk

Emulsions are known as a class of disperse systems consisting of two immiscible liquids: the liquid droplets (the disperse phase) are dispersed in a liquid medium (the continuous phase). Where, several classes may be distinguished: oil-in-water (O/W), water-in-oil (W/O) and oil-in-oil (O/O).

Common emulsions are inherently unstable and, thus, do not tend to form spontaneously. They can be obtained through shaking, stirring, exposure to power ultrasound or homogenization. Over time, emulsions tend to revert to the stable state of the phases comprising the emulsion. That is why to improve their stability so that the size of the droplets does not change significantly with time and phase separation will not occur, an appropriate surface active agent can be used [1].

The study presented here faces the problem of emulsion stabilisation by exposure of the rapeseed oil to the mixtures of Poly(vinyl alcohol), PVA, and organic carbon post combustion particles[2]. Emulsions of oil-in-polymer, and polymer-in-oil, as well as oil/polymer-particle mixtures, in various ratios, have been obtained in homogenisation process. Then, the systematic study on their stability, droplet size evolution (as shown on Figure 1ab) and, suitability to undergo a polymerisation process, to from a stable solid material [3] as shown on Figure 1c, was performed. The process revealed a formation of an emulsion in submicron scale in the oil-in-polymer and polymer-in-oil emulsion.

After solidification, obtained materials have been characterized in terms of structural evolution and morphology, involving porosity shape and size.

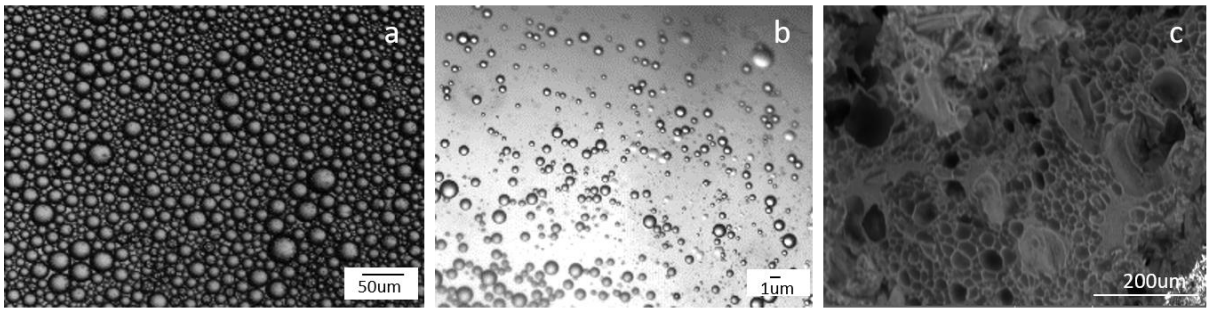

Figure 1 Macro (a) and submicron (b) PVA-in-Oil emulsion in liquid state and after polymerisation (c).

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DROPLET MANIPULATION ON MICROGROOVED SURFACES WITH STRUCTURAL CONTRAST

H. Zhao[1], C. Mackenzie-Dover[1], D. Orejon[1], M.E.R. Shanahan[2] and K. Sefiane[1]

[1]The University of Edinburgh, Edinburgh, UK; [2]University of Bordeaux, Talence, France

S1770360@sms.ed.ac.uk

On a horizontal surface, a droplet can spontaneously move under a chemical[1] or a structural[2] wettability gradient. In the past two decades, drop manipulation has attracted increasing interest due to its potential applications in many industrial and bio-medical fields. To this extent microfluidics[3] is one of the fields where the precise droplet control and positioning is required. This study systematically investigates the droplet motion and its behaviour due to the presence of a wettability contrast and provides further insights for the estimation of the moving distance and speed of the droplet. Microgrooves were utilized to create different wettability contrasts on the surface, i.e., sharp contrast across two different surfaces, as shown in Figure 1(left). Water droplets of ca. 9 µl were deposited at the interface and the horizontal motion of the droplet towards the less hydrophobic region was monitored and analysed. Upon deposition, droplets experience a vertical oscillation (Figure 1(right)), which is believed to help to overcome the hysteresis. A criterion based on the structural wetting contrast for predicting the moving distance and the velocity of the droplet is here proposed. This study provides further insights for the surface design aiming to control droplet distance and speed for a specific purpose. Future work will involve the study of multiple interfaces in sequence aiming to achieve a continuous gradient to make the droplet move further, as well as the study of different type of liquids for practical applications.

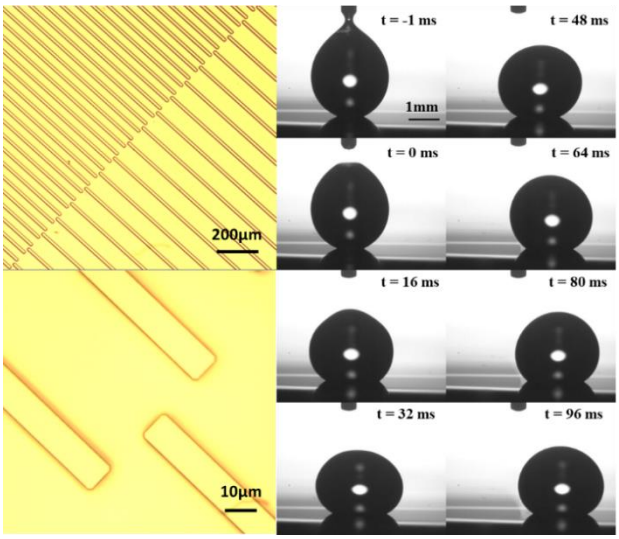

Fig.1 Left: Microscope pictures of the microgrooved surface. Right: An example of the droplet motion.

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How drops move on a wettability contrast

A. Askounis[1], Y. Kita[2], C. Mackenzie Dover[3], Y. Takata[2] and K. Sefiane[3]

[1]University of East Anglia, Norwich, UK; [2]University of Edinburgh, Edinburgh, UK, [3]Kyushy University, Fukuoka, Japan

Email: a.askounis@uea.ac.uk

uphill[1]. A plethora of works has been reported since where motion was induced via external thermal[2], electrical[3]

Manipulation of drop motion is paramount in a variety of areas from biomedical microfluidics to self-cleaning surfaces. In their seminal work, Chaudhury and Whitesides chemically treated a surface to create a wettability gradient which allowed drops climbing uphill[1]. A plethora of works has been reported since where motion was induced via external thermal[2], electrical[3] and/or other inputs. Nonetheless, the underlying principles dictating the motion of drops placed at the interface between two surfaces with different wettabilities, i.e. wettability contrast, remain elusive. We conducted a series of drop motion experiments on micro-pillared, superhydrophobic surfaces with a variety of wettability gradients (Figure 1 (a)) due to differences in geometrical features such as pillar dimensions, pitch and shape. We found a linear relationship between drop velocity and pillar density (Figure 1 (b)), due to surface energy minimisation. In particular, the drop is deformed during deposition (detachment from needle) resulting in excess energy which in turn induces drop motion to the more stable and lower energy surface. Lastly, we proposed a theory to describe the direction of drop motion when placed on a wettability contrast shown in Figure 1 (c).

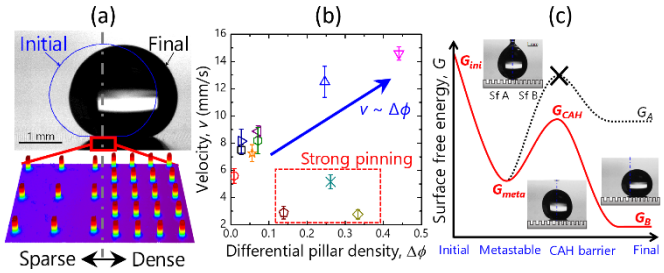

Figure 1: (a) Optical image of drop motion on wettability contrast, (b) Drop velocity as a function of pillar density, (c) Schematic illustration of proposed theory. Adapted with permission from [4].

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CONTACT LINE DYNAMICS AND HYSTERESIS MEASUREMENTS ON SOCAL SURFACES

H. Barrio-Zhang[1], É. Ruiz-Gutiérrez[1], G. G. Wells[1] and R. Ledesma-Aguilar[1]

[1]Smart Materials and Surfaces Laboratory, Northumbria University, Newcastle Upon Tyne NE2 1TN, United Kingdom

hernan.barrio-zhang@northumbria.ac.uk

Solids are not uniform and exhibit roughness at micrometric scales. The chemical and physical imperfections that cause roughness affect the interaction between a liquid-gas interface and the solid, namely on its apparent contact angle and pinning behaviour. This is the reason why surface topography has become a promising variable for controlling wettability, adhesion, mobility and liquid transport[1]. Contact Angle Hysteresis (CAH) is a measure of the static friction exerted by a solid surface on a liquid droplet[2]. Slippery Omniphobic Covalently Attached Liquid (SOCAL) surfaces are a novel way to produce surfaces with ultra-low CAH through acid-catalyzed graft polycondensation of dimethyldimethoxysilane[3]. This study focuses on the interaction of pure water droplets on ultra-smooth SOCAL surfaces. Our results show that when CAH approaches zero, its measurement becomes challenging. Hence, we develop a method to measure CAH by analysing the behaviour of the contact line during the slow relaxation of the liquid-gas interface on a SOCAL surface. By analysing the asymptotic behaviour of the contact line, we are able to quantitatively determine hysteresis.

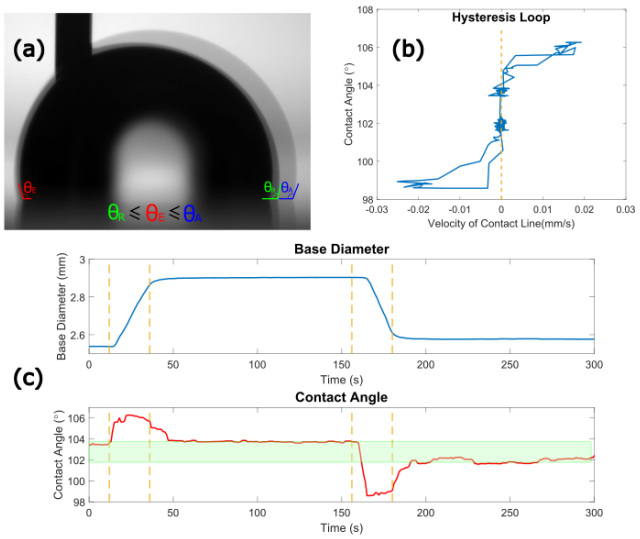

Figure: **(a)** Experimental method: The droplet is at equilibrium angle (θ_E), then inflated/deflated to obtain the advancing angle (θ_A) and receding angle (θ_R). **(b)** Hysteresis loop showing contact line velocity behaviour of the experiment with respect to the contact angle. **(c)** Graphs showing the evolution of the base diameter of the droplet and the contact angle over time. The yellow markers indicate when pumping in or out is performed during the experiment and the green band is the approximate value of CAH.

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SHAPE LOCKING PHENOMENA DURING THE DROPLET TRANSPORT ON DIFFERENT ENERGY GRADIENT SURFACES

I. U. Chowdhury[1], P. S. Mahapatra[1], A. K. Sen[1] and M. K. Tiwari[2]

[1]Indian Institute of Technology Madras, Chennai, India; [2]University College of London, London, United Kingdom. Presenting author email - *me17d023@smail.iitm.ac.in*

Self-driven droplet transport on an open surface is becoming more popular for various microfluidics applications. In an energy gradient surface, droplet moves from low energy (low wettability) region to high energy (high wettability) region. Numerical simulation of droplet transport on different linear energy gradient surface is performed on COMSOL. Level set method is used to track the interface. It is observed that the droplet attains an identical shape on different energy gradient surface at a particular time which is called as shape locking of the droplet. At shape locking, wetted area, height, front contact angle and the rear contact angle of the droplet is identical on different energy gradient surface. It should be noted that the droplet is at different positions for different energy gradient surfaces at the shape locking time. This shape locking is because of the identical surface energy of the droplet at the shape locking time for different energy gradient surface.

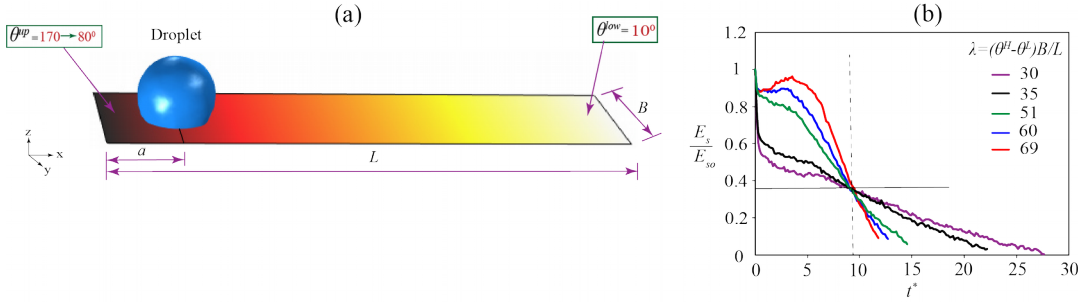

Figure 1 :a) A glycerin droplet of initial diameter d_0=0.5 mm is placed on an energy gradient surface of length L=3.5 mm and width B= 1.5 mm. The downstream contact angle θ^L is fixed as 10^0 whereas the upstream contact angle θ^H varies from 170^0 to 80^0. The droplet is placed at a distance of a=0.5 mm from the left. b) Non-dimensional surface energy E_s/E_{s0} variation with non-dimensional time $t^*(=t\sigma/L\mu)$ for different energy gradient λ. Here, E_{s0} is the initial surface energy of the droplet.

After the droplet is deposited on the solid surface, the droplet tries to minimize its surface energy due to which droplet starts to move. During the droplet transport, one part of the energy is absorbed by the viscous dissipation whereas the other part of the energy is converted to the kinetic energy of the droplet. It is observed that the kinetic energy E_k is 2-3 order less than the surface energy E_s; hence, in the present case, the surface effect is much more dominant than the inertial effect. The surface energy E_s is continuously decreasing because of the continuous spreading of the droplet. The surface energy E_s is calculated as $E_s = \sigma_{lv}(A_{lv} - A_{ls} \cos\theta_c)$. Where θ_c is the average contact angle of the droplet footprint, A_{lv} is the surface area of the droplet exposed to liquid vapor, A_{ls} is the wetted area of the droplet and σ_{lv} is the surface tension of the droplet. Figure 1(b) shows that E_s is identical at t^*=9.5 and hence the shape locking of the droplet. The primary objective of the present work is to demonstrate the shape locking phenomena of the droplet on different energy gradient surfaces.

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CONTROLLED NANOSCALE DEFECTS TO STUDY WETTING HYSTERESIS AND DYNAMICS

R. Lhermerout[1] and K. Davitt[1]

[1]Laboratoire de Physique de l'Ecole normale supérieure, ENS, Université PSL, CNRS, Sorbonne Université, Université Paris-Diderot, Sorbonne Paris Cité, Paris, France

Kristina.Davitt@ens.fr

Numerous models have attempted to describe how contact angle hysteresis and dynamics are affected by defects or inhomogeneities on the underlying solid surface [1], but comparison to systematic experiments is lacking. Here we report an experimental study of the wetting dynamics where we have unprecedented control of the size, shape and density of defects at the nanoscale [2].

Defects on silicon are obtained by colloidal deposition followed by high-temperature sintering of silica nanobeads. Higher temperatures yield flatter defects and therefore lower pinning energies. We then measure the dynamic contact angle over 7 decades in velocity for a variety of liquids.

We find that the hysteresis scales linearly with the defect density n and quadratically with their height h_0 (Figure 1), consistent with the often-cited scaling originally developed for strong, independent defects [3]. However, even for defects whose individual pinning energies are estimated at $100\ k_{\mathrm{B}}T$ we find no evidence of thermally-activated dynamics. We discuss how this illustrates the considerable room for improvement in our understanding of whether a real actualization of disorder is weak or strong and exhibits individual or collective pinning.

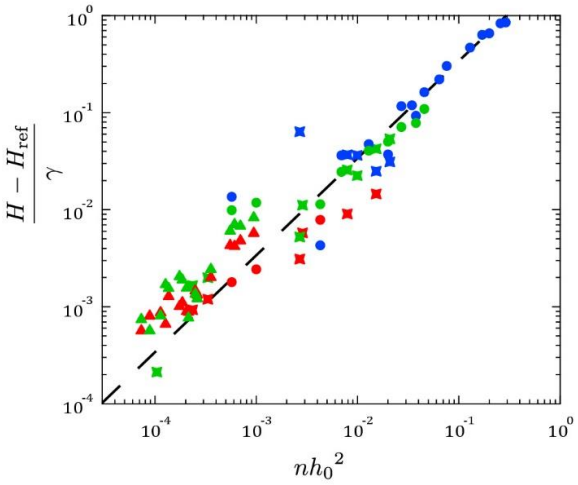

Figure 1 : Hysteresis as a function of defect concentration n and height h_0 for a variety of liquids (surface tensions γ). The hysteresis H_{ref} on a reference devoid of defects has been subtracted.

ACKNOWLEDGEMENTS: The authors acknowledge the ANR grant REALWET for supporting this work.

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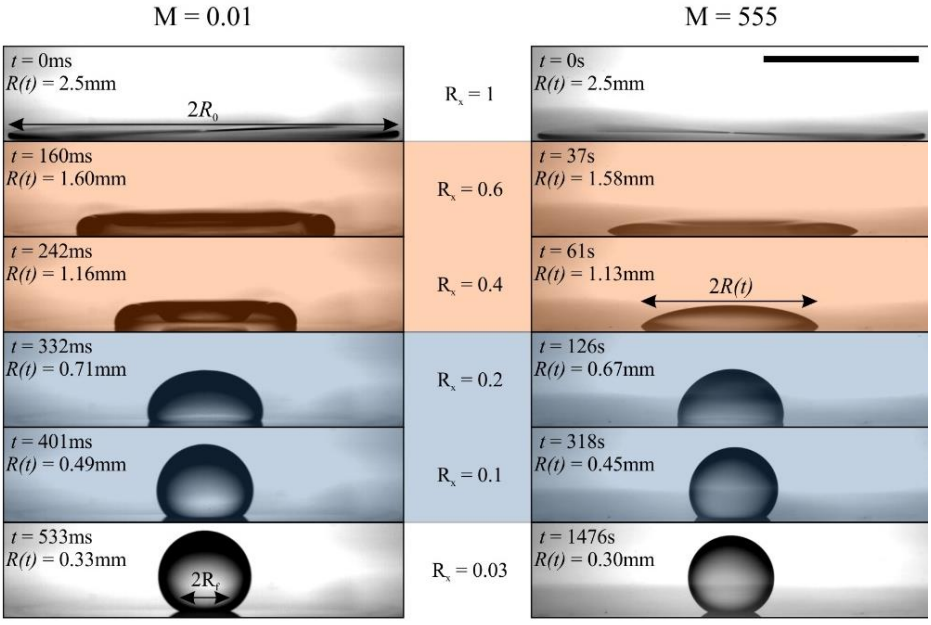
Liquid-in-Liquid Dewetting Dynamics

A. M. J. Edwards[1], R. Ledesma-Aguilar[2], M. I. Newton[1], C. V. Brown[1], and G. McHale[2]

[1]School of Science and Technology, Nottingham Trent University, Nottingham, UK; [2] Northumbria University, Newcastle upon Tyne, UK.

andrew.edwards@ntu.ac.uk

The dynamics of liquid contact lines both wetting and dewetting are critical in many applications including coating, cleaning and microscale patterning. In a recent paper, we demonstrated how interface-localised liquid-dielectrophoresis [1] can be used to create thin liquid films on non-wetting substrates which retract into a single droplet by quenching the electric field [2]. For the first time enabling repeatable and controlled studies of the dewetting phenomenon with a variety of liquids. In this work we apply this technique to create thin liquid-in-liquid films which retract to form a single droplet. We present data covering four orders of magnitude of inner-to-outer liquid viscosity ratio from 10^{-2} to 10^{2} on both wetting and non-wetting substrates. Demonstrating at low viscosity ratios, non-wetting relaxation occurs an order of magnitude quicker than a relaxation on wetting surfaces. While for high viscosity ratios, an unexpected crossover in relaxation timescales is observed between wetting and non-wetting surfaces. We analyse this data in terms of the Cox-Voinov hydrodynamic model and show excellent fits between experiment and data, both for the early time linear speed dewetting regime and the late time exponential approach to equilibrium.



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SELF-PROPELLED WATER DROPS ON BARE GLASS SUBSTRATES

P. E. Galy[1], M. Morel[1], S. Rudiuk[1] and D. Baigl[1]

[1]UMR 8640 PASTEUR, Département de Chimie, École Normale Supérieure, Paris, France

pauline.galy@ens.fr

Miniaturisation of chemical reactions has become a great challenge for the development of micro-analysis devices and micro-chemical reactors. Instead of squeezing fluids in micro-channel networks, having liquids in the form of drops that could move by their own along user-defined pathways appears as an interesting goal for low-cost and simplified microfluidics.

Since the first reports in the early 90's, the so-called self-propelled drops have constituted an example of such systems. Several approaches have been proposed but most of them involved organic drops[1,2,3]. With water, drop motion could be achieved but only using specific surfaces presenting a wetting gradient[4] or addition of reactive compounds such as etching acids[5]. In this presentation, I will describe an exquisitely simple system of water drops that can propel themselves on bare glass substrate and contain no more additive than conventional surfactants[6]. This was achieved by exploiting the adsorption behavior of these surfactants leading to dynamical anisotropy of wetting properties. We precisely analyzed the drop motion (Figure 1-a) and established how it could be tuned by both the nature and the concentration of the surfactant. I will emphasize the performance of this system by demonstrating drop motion as fast as 10 cm/s, complex drop manipulation along user-defined path simply produced using a marker pen, as well as drops capable of moving vertically against gravity (Figure 1-b).

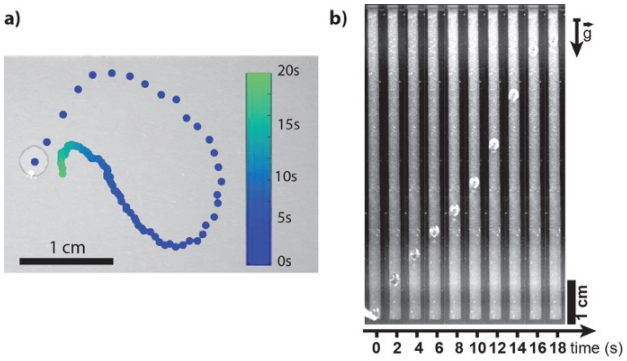

Figure 1 : a) Tracking of a self-moving water drop on a glass substrate.
b) Drop climbing a vertical glass slide along straight pathways.

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INVESTIGATING THE AGEING OF MODEL LIQUID INFUSED POROUS SURFACES

S. Goodband[1], H. Kusumaatmaja[1] and K. Voitchovsky (Underline presenting author)

[1]Department of Physics, Durham University, UK

s.j.goodband@durham.ac.uk

S. Goodband

Slippery liquid infused porous (SLIP) surfaces are a new breed of bio-inspired materials that are being explored as anti-fouling coatings for use in the medical and marine industry. Despite their success as self cleaning surfaces, they are still poorly understood and more work is needed to understand the complex interactions between the porous matrix, the infused liquid and any fouling particles or droplets. In particular understanding how the surfaces age when they are subjected to external perturbations would be useful when considering the environments that SLIP surfaces are required to function in. An example may be that of a SLIPs coating for an ocean liner which would be required to survive the mechanical stresses of waves as well as the corrosive behaviour of the saline environment.

Here, a model liquid infused surface is used, fabricated with commercially available hydrophobic nano-particles (Glaco Mirror Coat™ [1]) and infused with silicone oil. This allows low cost surfaces to be easily made as test-pieces to study SLIP surface behaviour. Using a combination of atomic force microscopy (AFM) and contact angle measurements, the surfaces are studied in detail to investigate the impact of ageing on the surface performance and nano-structure.

It is found that when the surfaces are exposed to prolonged pressure perturbations or to saline solutions the surfaces become less reliable and exhibit high variability in contact angle measurements. The surface struggles to retain its self healing properties, leading to a patchy oil layer, which affects the contact angle of a droplet on the surface by increasing the surface roughness and inducing pinning. Comparing this model system to other SLIP surfaces may be useful in predicting the effect of certain environmental pressures on SLIP surfaces and explain some of the behaviours observed.

ACKNOWLEDGEMENTS: The authors thank EPRSC for supporting this work.

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Graphical Abstract

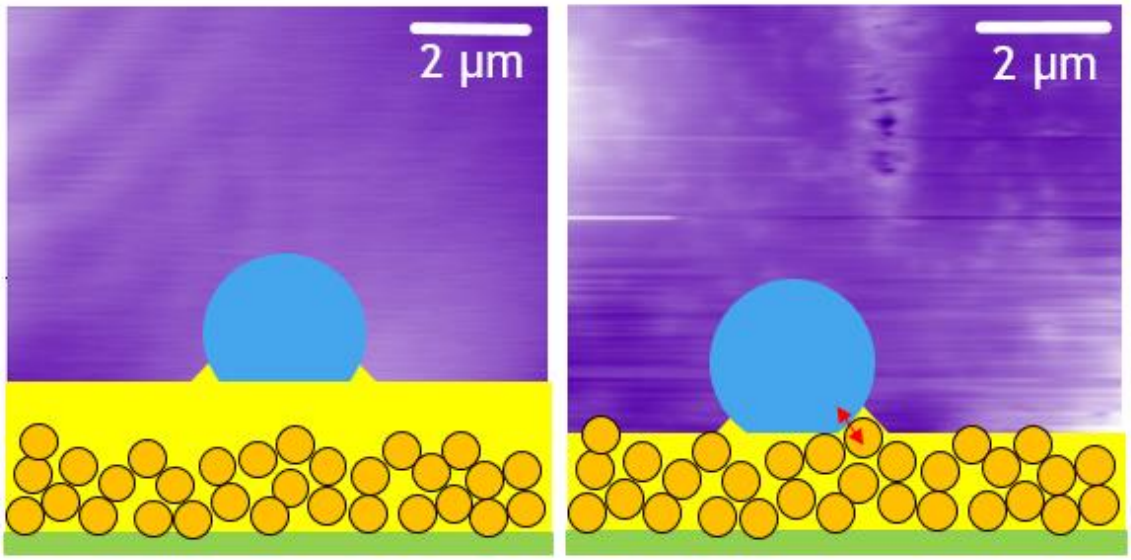

Figure 1: A slippery liquid infused porous (SLIP) surface is shown before and after an ageing process. (left) shows a porous structure with a thick oil layer on the surface which becomes depleted (right) once aged. This leads to surface features interacting with liquid drop and creating surface effects.

DROPLET DYNAMICS ON ROUGH SURFACES

N. Savva and D. Groves

Cardiff University, Cardiff, United Kingdom

savvan@cardiff.ac.uk

We undertake a combined numerical and analytical approach to study the motion of partially wetting droplets on surfaces decorated with smoothly varying topographical features. This work is based on the associated thin-film equation for the evolution of the droplet thickness $h(\boldsymbol{x}, t)$, which accounts for the effects of viscous dissipation, capillarity, slip, and surface heterogeneities, cast in the form

$$h_t + \frac{\sigma}{3\mu} \boldsymbol{\nabla} \cdot \left[h \left(h^2 + 3\lambda^2 \right) \boldsymbol{\nabla} \nabla^2 (h + s) \right] = 0,$$

where μ and σ are the fluid's viscosity and surface tension, respectively, $s(\boldsymbol{x})$ describes the topographical features of the substrate and λ is the slip length, which allows us to alleviate the non-integrable stress singularity that would occur at a moving contact line.[1]

A matched asymptotic analysis is used to extend an earlier study on two-dimensional droplets moving on rough surfaces[2] to the arguably more realistic three-dimensional setting. Our methodology is based on a recent work on the dynamics of droplets on ideally smooth and chemically heterogeneous surfaces and pertains in the limit $\lambda \to 0$.[3] For sufficiently long timescales, the quasi-static approximation can be invoked to deduce a lower-dimensional surrogate model to describe the evolution of nearly circular contact lines.

This model constitutes a system of differential equations for the harmonics of the contact line and is reminiscent of the Cox–Voinov law[1] supplemented with higher-order corrections. Noteworthy is also that this analytical approach may be straightforwardly adapted for other contact line models, by suitably choosing the microscopic scale and formally linking it with with the dynamics in the vicinity of the contact line.[4]

A number of representative cases are discussed, demonstrating that the model typically exhibits very good agreement with accurate solutions to the full problem. A hybrid numerical scheme combining the boundary integral method and the relative merits of the surrogate model is also proposed, which offers improved agreement with the predictions of the full model for strongly deformed contact lines, whilst requiring considerably fewer computing resources.

ACKNOWLEDGEMENTS: We acknowledge financial support from the Engineering and Physical Sciences Research Council (EPSRC) of the UK through the grant EP/P505453.

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Wetting of mineral surfaces by fatty-acid laden oil and brine: carbonate effect at elevated temperature

M.E.J. Haagh, N. Schilderink, M. H.G. Duits, F. Mugele

[1] Physics of Complex Fluids, MESA+ Institute, University of Twente, 7500 AE Enschede, The Netherlands

f.mugele@utwente.nl

The recovery of crude oil from geological reservoirs depends strongly on the competitive wetting of oil and water on the rock surface. Oil companies strive to maximize the recovery by reducing the salinity of the water injected into the rock to displace the oil. This process of Low Salinity Water Flooding (LSWF) was found to be efficient in sandstone reservoirs consisting containing silica and clay mineral surfaces. However, the complex composition of the materials involved, i.e. rock, brine, and oil has so far hampered the identification of origin of the LSWF effect. While binding mechanisms such as divalent cation bridging of acidic oil components to silica and clay surfaces have been proven to be effective in idealized model experiments [1, 2], their relevance under reservoir conditions remains elusive. Here, we present a combination of contact angle measurements and microscopic AFM investigations of idealized mica substrates at slightly elevated temperatures (60°C) and brine compositions. The experiments show that relatively minor variations of the brine composition, specifically, the addition of 2mM of bicarbonate can have a dramatic effect on the wettability [3] by inducing precipitation of very hydrophobic salts of fatty acids from the oil onto the solid surface. The effect is rather universal and depends only weakly on the chain length of the fatty acid. Next to bicarbonate and elevated temperatures, the presence of divalent cations is also essential for the contact angle variation, in agreement with the multiple ion exchange model of LSWF. Reducing the divalent cation concentration in the brine leads to a dissolution of the organic layers and thus to improved water wettability, as required for enhanced oil recovery.

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Cooling Rate Model for Single Metal Droplets Generated via Drop-on-Demand Droplet Generator

Saeedeh Imani Moqadam[1,2], Nils Ellendt[1,2], Lutz Mädler[1,2]

[1]University of Bremen, Faculty of Production Engineering, Bremen, Germany; [2]Leibniz Institute for Materials Engineering IWT, Bremen, Germany

s.imani@iwt.uni-bremen.de

The thermal energy of a free falling, single metal droplet moving in a colder gas is released via convection and radiation [1]. In order to model the cooling rate and understand the solidification behavior during falling, the coupled droplet motion and heat transfer need to be examined. In this study, the cooling rate of the falling droplets is modelled and validated experimentally using a pneumatic drop-on-demand droplet generator. Experiments with three different alloys namely AlCu4.5, CuSn6 and FeNi25, having different melting temperature levels, were carried out. These alloys allow estimating their cooling rate during solidification by quantitative microstructure analysis.

The droplet cooling model includes a model for the droplet trajectory in which drag and gravitation are key parameters. In previous investigations, we found that for pronounced temperature differences between the droplet and the ambient gas, correlations for the drag coefficient need to be extended to include temperature effects [2, 3]. Different correlations for the Nusselt number are examined in the equation for the thermal history of a single droplet together with a formulation for radiation. The equations for the trajectory and thermal history are solved simultaneously. As boundary conditions, the initial velocity of the droplet and its initial temperature are required. While the initial droplet temperature is simply assumed to be equal to the melt temperature in the crucible, its initial velocity is determined from high-speed recordings. Metallographic analyses of the particles were performed to determine the secondary dendrite arm spacing (SDAS) which is related to the cooling rate during solidification [4, 5].

As a result, we obtain reliable sets of well-defined process conditions of the generated particles and their cooling rate which allow us to compare the results with the predicted solidification rates of our cooling model. We found that the results of the model and experiments are in strong agreement.

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DRYING OF AQUEOUS DROPLETS WITH NON-IONIC SURFACTANTS CnEm AND ANIONIC SURFACTANT SDS

J. Shi[1], L. Yang[1], C. D. Bain[1]

[1]Durham University, Durham, UK;

Jing.shi@durham.ac.uk

Understanding the influence of surfactants on drying droplets is of importance for various industries, e.g., printing, spray coating and agricultural spraying. A small amount of particular surfactants can lead to more uniform deposits from drying colloidal drops or polymer solution drops and is often explained by surfactant-driven Marangoni flows[1,2]. However, in addition to surface tension, the addition of surfactants affects various aspects of the dynamic drying process, e.g., the contact angles, the spreading and depinning process, and particle−particle, particle−free interface, and particle−substrate interactions. Further, the surfactant solutions might undergo phase separation and/or phase transition with evaporation of water. The understanding on the drying behaviors of droplets containing surfactants is far from complete.

In this study, we studied systematically the drying behaviours of aqueous droplets with non-ionic surfactants C_nE_m and anionic surfactant SDS. The spreading and drying of the surfactant droplets were studied by contact line tracking and interferometry. Surprisingly, C_nE_m solutions inhibit spreading on hydrophilic surfaces; the higher concentrations stop spreading sooner. SDS solutions at various concentrations show similar spreading behaviour. During drying, the non-ionic surfactant solutions appear to undergo phase separation, with a surfactant-rich film adjacent to the substrate pinned at the contact line with a water-rich droplet sitting atop the surfactant-rich film. The water-rich droplet shrinks in diameter as it evaporates. The surfactant-rich film is thicker for higher concentrations. SDS droplets do not pin, but retract immediately after spreading stops.

The internal flows during drying were observed by addition of tracer particles. Marangoni flows are observed (0-0.6 t/t_{dry} where t_{dry} is the drying time of the droplet) for drying droplets with C_nE_8 (n = 10, 12 and 14) well above cmc; the Marangoni flows observed are mild without obvious circulating eddies. There are inward flows close to the air−liquid surface and outward flows close to the substrate; no obvious Marangoni flows are observed for droplets with $C_{14}E_6$. Compared to the Marangoni flows in C_nE_8 droplets, Marangoni flows are somewhat stronger in drying droplets with SDS, but cease earlier (flows observed for 0-0.3 t/t_{dry}).

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Evaporation of Cu-Al LDH nanofluid droplet for electronic cooling

S. Chakraborty [a], S.K. Saroj [a], and P.K. Panigrahi [a]

[a] *Department of Mechanical Engineering, Indian Institute of Technology Kanpur, Kanpur, India*
email id: samarshi@iitk.ac.in

The current study deals with the synthesis, thermo-physical characterization of Cu-Al layered double hydroxide (Cu-Al LDH) nanofluid (water based), and its potential application in electronic cooling. The main objective of this investigation is to explore the potential of water-based mixed metal nanofluid for low-temperature surface cooling. LDH nanofluid was prepared via co-precipitation technique using sodium hydroxide and nitrate salts of Cu and Al. One-step nanofluid synthesis method has been used to eliminate the need of intermediate particle drying step, and problems related to particle re-dispersion in the base fluid. In order to improve nanofluid stability, an anionic surfactant named Sodium Dodecyl Sulphate (SDS) has been used as a stabilizer. Figure 1(a) shows the detailed schematic diagram of the experimental setup. A CMOS camera is used for the recording of droplet images during evaporation and a thermoelectric cooler has been used to maintain the surface at a constant temperature. Evaporation experiments have been performed by depositing sessile droplets of constant volume (5 µl) on the aluminium surface at different surface temperatures. Four different coolant combinations i.e. water, water-LDH, water-SDS, and SDS stabilized water-LDH have been compared. The water-SDS droplet shows the highest average evaporation rate followed by SDS stabilized water-LDH nanofluid (Figure 1b). Water–SDS and Water-LDH-SDS show similar contact angle and contact line behavior (Figure 1c and 1d) and the overall effect is clearly reflected in Figure 1b. The detailed heat transfer measurements provide effectiveness of the nanofluid for electronic cooling.

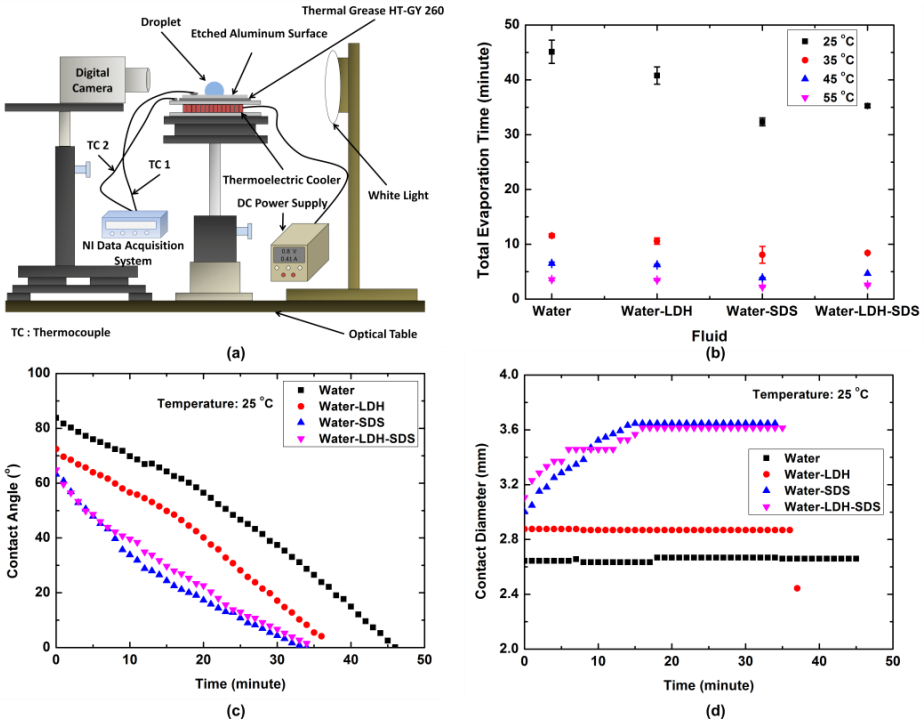

Figure 1 a) Schematic diagram of experimental setup, b) comparison of total evaporation time for different fluids as a function of surface temperature, (c) contact angle, and (d) contact diameter vs. time at 25°C surface temperature.

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