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Flow measurement using micro-PIV and related temperature distributions within evaporating sessile drops of self-rewetting mixtures of 1-pentanol and water

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Abstract Recently interest has arisen in the use of so-called self-rewetting mixtures for micro-scale heat transfer systems. Such fluids, in which the surface tension can increase with increasing temperature, are expected to offer superior evaporative cooling performance by extending the region of operation before dry-out of the heated surface sets in. Whilst improved performance has been shown in some practical situations using these fluids, it is not entirely clear as to the mechanism of such improvements.

We have studied the flow within evaporating sessile drops of 1-pentanol-water mixtures using micro-PIV and have observed three stages in the evaporation process. During the first stage there appears to be a single toroidal vortex with flow inwards along the base of the drop. The vortex only occupies the central region of the drop and appears to pulsate, reducing in size during evaporation. This is followed by a second transition stage to a third stage in which the flow is directed radially outward, as observed by us for pure water droplet evaporation and in the latter stages of ethanol/water drop evaporation. Temperature measurements, using IR thermography suggest that the initial stage of evaporation may be controlled by thermal Marangoni effects as opposed to the concentration driven Marangoni flows postulated for ethanol-water mixtures.

Keywords: Marangoni flow, surface tension, evaporation, heat transfer

1. Introduction

Micro scale cooling systems have reached the stage where evaporative cooling technologies are required to match the high heat fluxes generated locally (*e.g.* on silicon chips) without excessive local temperature increases. Such cooling technologies rely on knowledge of the interaction of both local evaporation and replenishment of liquid to the surface. Local dry-out of the surface may inhibit re-wetting and in the case of heat pipes leads to failure of the complete device.

Sessile drop evaporation provides a simple model of the evaporative process that is amenable to observation and has been used by many researchers to provide insight to the process of evaporation on the micro-scale [1]-[5]. Variations in the drop diameter, profile, surface temperature distribution and internal flow patterns with time provide an insight into the nature of the local variation in evaporative flux. However, in order to ascertain the mechanism of local evaporation and how this depends on concentration and temperature variations, a quantitative knowledge of the local convection within the drop is required.

Studies leading to accurate measurement of flow velocities within such drops are rare even for pure liquids [2], [3]. We believe that we are the only ones to have made such measurements so far in drops formed from binary mixtures [3] and we know of no previous studies of velocities in drops formed from so-called 'self-rewetting fluids'. With pure water we measured velocities along the base of the drop that were directed radially outward and whose magnitude was consistent with the assumption that the majority of evaporation occurs at the contact line. For ethanol water mixtures, we observed three distinct phases: 1) an initial stage characterised by multiple vortices rotating around a vertical axis, the total number decreasing and the size

of each increasing as evaporation proceeds; 2) a transition stage in which the vorticity decays rapidly to zero and during which there is a sudden spike in the outwards radial velocity along the base of the drop; and 3) a final stage characteristic of that observed for pure water.

Vochten *et al.* [6] were the first to report on mixtures of higher alcohols and water, whose behaviour is significantly different from ethanol and water in that they appear to have an unusual characteristic: they experience a minimum in surface tension with changes in temperature, with the surface tension being an increasing function of temperature above this minimum value (Fig 2 shows data from our own laboratory on the behaviour of butanol-water and pentanol-water mixtures). All other pure fluids and most mixtures have a surface tension that decreases with increasing temperature. Wilhelmy's method was used to determine the surface tension and observed this unusual behaviour in aqueous mixtures of butanol through to nonanol and for some branched alcohols. Ono *et al.* [7] confirmed Vochten's observations for aqueous mixtures of butanol and pentanol using what they believe to be a more accurate measurement technique of surface tension, that of the minimum bubble pressure method. They also compared the behaviour of these fluids to that of ethanol in water. Several researchers (*e.g.* Savino et al. [8]. Lappa et al [9]) have investigated Marangoni instabilities at the liquid vapour interface of such mixtures.

The increase in surface tension with increasing temperature should make these fluids desirable for evaporation system applications in which the temperature will rise as the onset of dry-out occurs; the increase in surface tension should draw in fluid by Marangoni induced convection to rewet the surface. Thus these fluids have come to be known as self-rewetting fluids. Savino *et al.* [8] have exploited this property of the fluids in improving the performance of heat pipes.

Flow patterns generated within such fluids were observed due to evaporation under reduced gravity in a wickless heat pipe, by seeding the flows with tracers particles. They have observed flow patterns for both butanol-water mixtures and ethanol-water mixtures, showing clear differences that they believe to be beneficial between butanol and ethanol.

We wish to go one step further in terms of measurement of the velocities within such systems rather than just the qualitative flow pattern. For this we use a micro-PIV system as used in our previous studies and the use of sessile drops in order to compare the behaviour to previous studies with other fluids.

2. Experiments

2.1 Preparation of Mixture

In this study the evaporation process of 0.12 µl sessile drops of a solution of distilled water with 2 %vol of 1-pentanol on a heated substrate is investigated. Sample bottles were washed with detergent, rinsed thoroughly with distilled water and dried in an oven. Reagent grade pentanol and distilled water were then carefully pipetted into the sample bottle which was immediately sealed. A similar preparation technique was used for 1-butanol in water. Fresh solutions were made up each day. Cover-slides were prepared by immersing in distilled water within an ultrasound bath and then drying within the oven.

2.2 Measurement of Surface Tension

The surface tension of both aqueous alcohol mixtures was determined using the pendant drop method at a range of temperatures in an environmental chamber, with the contact angle determined using an FTA 200 instrument.

2.3 Apparatus for PIV study

The laser and microscope arrangement, as described in our previous work on ethanol-water mixtures, was used for the velocimetry study [2], [3]. However, a modified cover slide arrangement is used to enable the surface on which the droplet sits to be heated to a known temperature. The substrate heating device (fig 1) consists of 3 heaters mounted on a copper block to distribute the heat and a PTFE mounting ring, both to protect and to reduce heat losses to the microscope. To obtain uniform heat distribution the resistors are

placed in an angle of 120° on the top of the copper block. For better heat transfer a thermal paste (Xigmatek PTI-G3606) is spread between resistors and copper block. Measurements of temperature by means of IR thermography reveal a uniform temperature across the cover slide prior to placing the drop on the slide. Because we measure temperature of the upper surface of the substrate, knowing thermal resistances and heat flux is irrelevant for the purpose of this study. To obtain a close distance between droplet and microscope, a hole is drilled into the middle of the copper block where the cover slides are inserted. The heating device is circular so that it fits exactly under the opaque cap providing a shield to laser light and precise placing of the droplet.
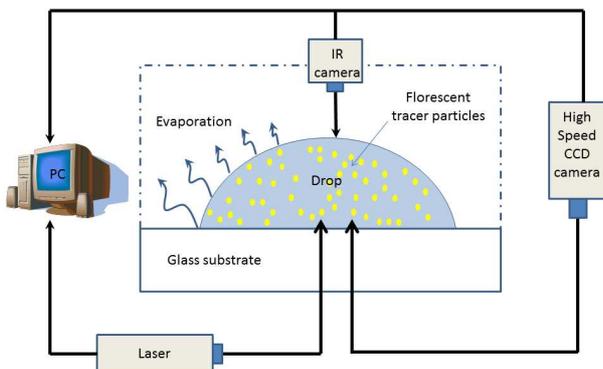

Fig 1: Schematic diagram of the experimental

arrangement.

2.4 Experimental procedure

Micro-PIV was used to obtain velocity information within evaporating sessile drops. 0.12 μl (0.03 μl) droplets of a solution of 98% vol. distilled water and 2% vol. 1-pentanol, seeded with 0.04% solids of fluorescent micro-spheres (1μm diameter, Nile-red, carboxylate modified FluoSphere® beads of density 1.05g/cm3) are injected onto the clean, heated glass cover slide, sitting on an inverted microscope (Leica DM15000 M) to yield sessile drops of about 1.25mm diameter. Due to the presence of particles in the fluid, the contact line remained pinned throughout.
A New Wave Pegasus pulsed diode laser emitting at 527nm, synchronized with a

Dantec Dynamics Nanosense II camera (512x512 pixels) [at 10Hz (for temperatures of 30°C and 40°C), 20Hz (50°C and 60°C), 50Hz (70°C and 80°C) and 100Hz (90°C)] was used to cause the particles to fluoresce at 575nm and the resulting images captured. The resolution of the images was 320 pixels/mm. The height, above the base of the drop, of the plane in which the velocities were determined was set by adjusting the focus on the microscope. For all results reported here, the height was approximately 10μm off the base of the drop and the depth of field of the microscope lens was approximately 20μm. Velocities of the particles in the horizontal plane were determined by cross-correlation of successive images [10] for 16x16 pixel interrogation windows with 50% overlap to yield a 63 by 63 vector array, using Dantec Dynamics Flowmanager 4.71 software. A peak validation algorithm was performed with vectors rejected if the tallest correlation peak was less than 1.2 times the 2nd peak. Typically, up to 15% of the vectors were rejected at this stage.

Velocity maps were created from image pairs for every 0.5 seconds over the lifetime of the drop and for every 0.05 seconds over the region where the flow pattern was changing. From the velocity map, vorticity around a vertical axis could be evaluated. Prior to calculating the vorticity a moving average filter was applied, substituting vectors by an average based on an 11x11 array centered on the vector position, before a mask was used to exclude any spurious vectors outside the edge of the drop. The vorticity around an axis perpendicular to the image plane at position m, n was calculated from   where Δx and Δy are the grid spacing. The vorticity is shown in terms of colour coding on the velocity vector maps in Figure 5.

2.5 Surface temperature measurements

To determine the temperature distribution on the surface of the heated droplet at substrate temperatures in a range of 30°C to 90°C with steps of 10°C a "FLIR SC7000" camera with a "MWIR Macro 1:3.0" objective is used. For

heating a thermo-resistor connected to a variable DC power supply is used. The top of the thermo-resistor is equipped with the same cover slides used for the PIV experiments. To guarantee a good conduction between the thermo-resistor and the cover slide and an even temperature distribution a metallic high-temperature paste "WOLFRA-COAT C" is applied between the resistor and the cover glass.

Droplets of a binary mixture of 98% vol. distilled water and 2% vol. 1-pentanol of approximately 1mm in diameter are placed on the heated cover slide and the surface temperature map is recorded over the lifetime of the drop. The emissivity of the mixture was assumed to be approximately 1.
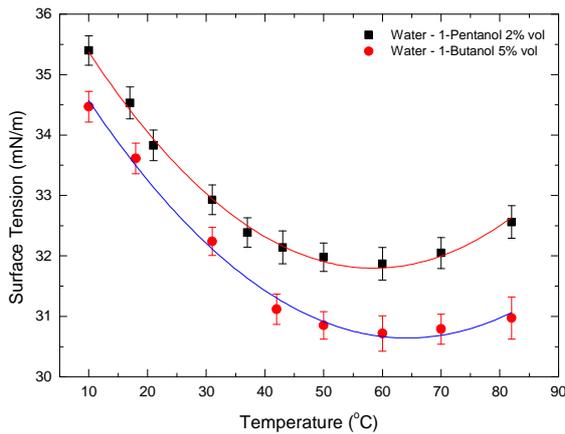
3 Results

Figure 2: Surface Tension versus Temperature for aqueous mixtures of n-butanol and of n-pentanol.

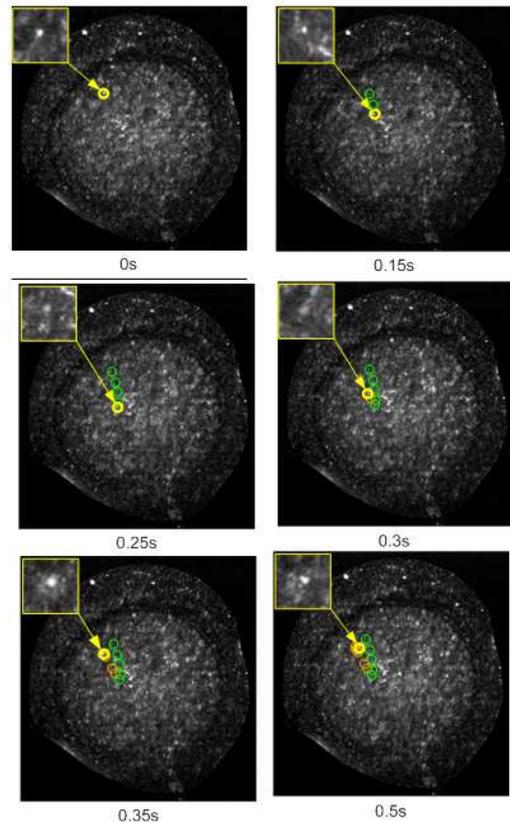

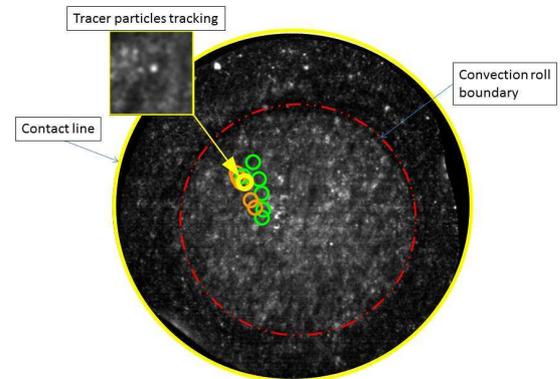Figure 3: The track of one particle for one rotation within the vortex

Figure 4: Pulsating convection roll highlighted.

We observe three distinct phases in the flow patterns during evaporation: an initial stage where we observe what appears to be a toroidal vortex, whose dimensions decrease with time, but with periodic oscillations, a short transition stage characterised by a high radial velocity outwards and then a third stage where the flow is all directed uniformly outwards towards the contact line. Figure 5 shows these three stages in terms of typical

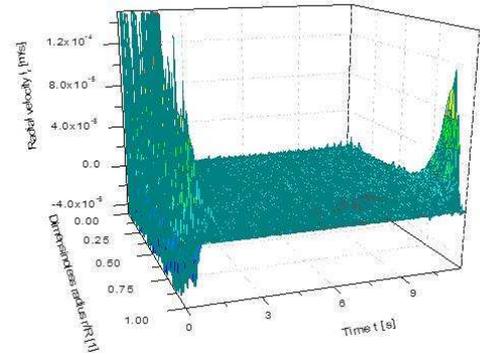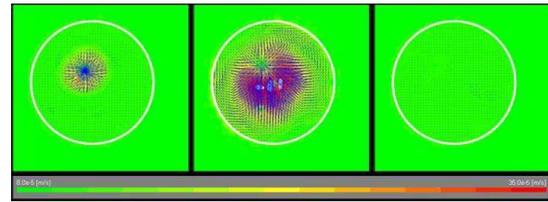velocity maps for each of the stages at three different temperatures (30°C, 70°C and 90°C). The initial stage is significantly affected by temperature, with the size of the vortex reducing and the velocities within it increasing as the temperature increases. The vortex also tends to move to an asymmetric position as the temperature rises.
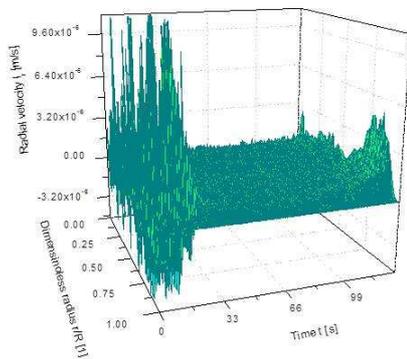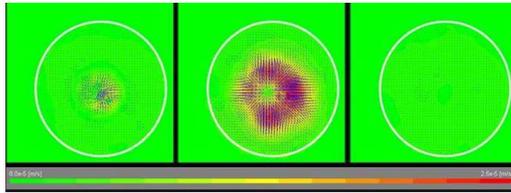
(a)
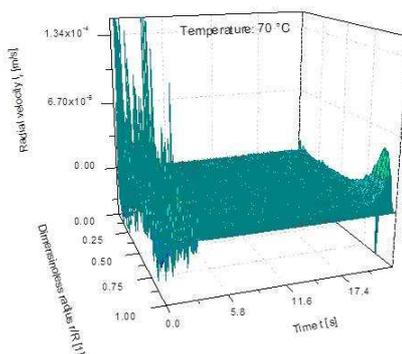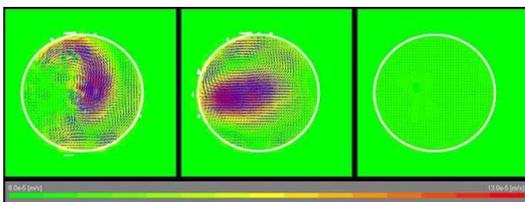
(b)

(c)

Figure 5: Velocity maps showing a) representative velocity patterns for the three stages of evaporation at (a) 30°C, (b) 70°C and (c) 90°C

In order to observe the transition stage more clearly, further sets of runs, of shorter duration, were conducted, but with higher frequencies of illumination and image recording. These revealed that there is a peak in the radial velocity just before the start of the third stage in which the velocity reduces to that representative of a uniform mass flow towards the contact line to match the local evaporation from this region.

Figure 6 shows the temperature distribution at the surface of the drops for the same temperatures as the velocity maps in Figure 5. The infra-red measurements reveal that for heated substrates above 30°C the outer edge of the drop is at the highest temperature, as expected due to the thin layer of liquid in this region. However, what was not expected is that for the highest substrate temperature, 90°C, there is also a hot-spot at the centre of the drop, presumably the result of hot fluid being convected to this surface region by internal fluid motion. For a temperature of 30°C the temperature profile is almost uniform across the drop due to the low evaporation rate

at this temperature.

4 Discussion

There is some similarity between the observations made here and the observations made previously [3] for aqueous ethanol solutions, in that both the aqueous pentanol and aqueous ethanol have three distinct regimes of flow over the duration of droplet evaporation, with the third stage in both cases being identical to that observed by us for pure water [2].
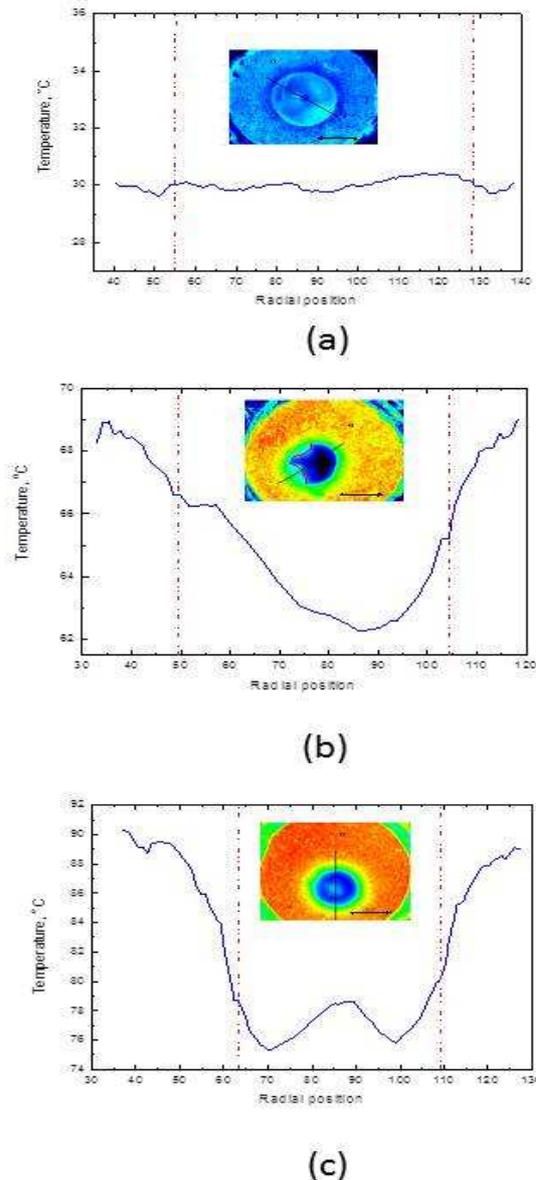

(a)

(b)

(c)

Figure 6: Infra-red maps of the upper surface of the drop at substrate temperatures of (a) 30°C, (b) 70°C and (c) 90°C.

However, the first stage is distinctly different for the aqueous pentanol, with the presence of a single toroidal vortex, whereas for aqueous ethanol a number of vortices rotating around the vertical axis were observed, this number decreasing to two as evaporation proceeds.

As discussed previously for aqueous ethanol, the initial stage is that in which the majority of the alcohol evaporates, with the second transition stage being explained on the basis of depletion of the last of the alcohol and the third stage representing evaporation of pure water. In the present study for aqueous pentanol, the differences observed in the initial stage of evaporation must represent a significant difference in the behaviour of pentanol relative to that of ethanol. The presence of a single toroidal vortex with inward flow along the base of the drop and, presumably outward flow along the top is inconsistent with Marangoni convection driven by the temperature profile.

Indeed, in the case of the experiment at 30°C, there is no measurable temperature profile along the droplet surface, suggesting that there will be no Marangoni convection due to temperature variations. Given the size of the drop, gravity driven convection is also unlikely, especially as the droplet surface temperature measured is the same as that of the substrate. Hence, the most plausible explanation for this internal convection is as a result of concentration variations. If propanol is preferentially evaporated at or near the contact line, then this region will end up depleted in propanol with a consequent increase in the surface tension in this region. This will cause an outward flow along the top surface that may in turn generate an inward flow along the base of the drop. In this case the inward flow is found over about 75% of the radius of the drop from the centre outwards. This explanation, if correct, is consistent with our expectation of the driving force for ethanol-water droplets, i.e. that the initial vertical flow is driven by concentration-related Marangoni convection. We believe that the reason for the difference in type of vortex

pattern observed, is due to differences in the rate of evaporation. For ethanol, evaporation is very rapid, generating an unstable flow regime, with randomly orientated vortices. However, propanol, being less volatile generates a more stable toroidal vortex.

At higher substrate temperatures, the surface temperature is lower at the apex of the drop and higher near the contact line. For both 70°C and 90°C, the recorded temperatures are above the temperature of the minimum in surface tension versus temperature, with the result that the temperature-based surface tension should be higher at the contact line than further in towards the apex, with temperature-driven Marangoni convection being expected to be outwards towards the contact line. This ought to enhance the effect of the concentration-driven Marangoni convection. For the experiment at substrate temperature of 90°C, the apex also has a higher temperature, suggesting that the temperature driven Marangoni convection is outwards close to the contact line and inwards close to the apex with zero flow at the point of minimum surface temperature. This temperature-driven convection inwards at the surface of the drop is inconsistent with our observation of a n intense inwards flow at the base of the drop that occupies only a small fraction of the radius of the drop. We thus conclude that the concentration-driven convection outweighs the temperature-driven convection and that this convection is strong enough to draw hot fluid from the base of the drop to the apex, thus altering the surface temperature profile.

5 Conclusions

For Propanol-water sessile drop evaporation on heated substrates, we observe three distinct flow regimes, similar but not identical to those we observed for ethanol-water droplets. The second, transition, stage and third stage appear to be very similar for both propanol-water and ethanol-water, being, we believe, caused by the evaporation of the last drops of alcohol and then the evaporation of the remaining water. However, the first

stage is significantly different for the two different alcohols. We believe that for both alcohol-water mixtures, this initial stage is driven by Marangoni convection based on surface concentration profiles. For propanol-water, the single toroidal vortex rather than the multiple vortices observed for ethanol water, may be due to differences in evaporation rate. The temperature profile that is much more organised for propanol-water being radial in nature appears to have no direct bearing on the flows observed.

6 References

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