# Three-Dimensional Marangoni cell in self-induced evaporating cooling unveiled by µ-Particle Image Velocimetry and Digital Holographic Microscopy

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# Abstract

 $\mu$ -Particle Image Velocimetry has successfully been used to map the velocity fields on diametrical horizontal and vertical cross sections of a transparent glass circular tube of 900 $\mu$ m diameter filled with ethanol. A Digital Holographic Microscope has been used to trace the trajectory of a tracer particle inside the liquid phase of an evaporating meniscus formed at the mouth of a 1mm square borosilicate tube filled with ethanol. The Marangoni flow cells are due to the self-induced differential evaporating cooling along the meniscus interface that creates gradients of surface tension which drive the convection. The competition between surface tension and gravity forces along the curved meniscus interface disrupts the symmetry due to surface tension alone. This distorts the shape of the toroidal Marangoni vortex. This is clearly seen in the  $\mu$ -Particle Image Velocimetry velocity maps. Thermocapillary instabilities of the evaporating meniscus are reported in the present work by analysing the trajectories of a tracer particle. It is found that the trajectory of the tracer makes different shaped three-dimensional loops and every four loops it returns to the first loop. By analysing several loops it was found that the characteristic frequency of the periodic oscillatory motion is around 0.125 Hz.

Keywords: µ-Particle Image Velocimetry, Digital Holographic Microscopy, Marangoni Convection

## Introduction

Phase change is a key process in numerous industrial applications such as heat pipes [1-2], condensers, combustion, crystal growth [3], glass manufacture [4] just to mention a few. Evaporation produces large heat transfer rates involving small quantities of working liquid at nearly constant temperature. The small temperature differences are important in many applications such as in the DNA replication described in [5] where a flat heat pipe has been used.

When phase change happens at relatively small sizes, typically below 1mm in characteristic dimension and for organic liquids, surface tension forces become important and tend to dominate over gravity forces. Marangoni convection, driven by surface tension gradients, overcomes buoyancy. The evaporation of a volatile liquid and the stability of a meniscus interface created inside a capillary tube have been investigated [6-7] where external heating has been applied with an electric heater; the

authors demonstrated experimentally that the meniscus becomes unstable and recedes at some applied heat load. The authors did not investigate the hydrodynamics in the liquid phase of the meniscus. The characterization of Marangoni convection of volatile liquids inside capillary tubes has been the subject of numerous publications by one of these authors [8-10].  $\mu$ -Particle Image Velocimetry [8], InfraRed Thermography [9] and Thermocromic Liquid Crystals [10] have been used to map the twodimensional velocity field in the liquid phase and the temperature distribution on the external tube and along the meniscus interface. It has been found that the liquid evaporates more at the meniscus wedge than at the centre which creates a difference in temperature which is big enough to generate gradient of surface tension that drives the Marangoni convection. The authors also found that the flow pattern is distorted in diametrical vertical sections of the tube which was attributed to the action of gravity. The same subject has been later investigated more in details [11-12]. The authors found that the evaporation flux is not a linear function of the tube size as reported in [13], rather a power function of the tube diameter; they also showed that below 75 $\mu$ m tube size the flow distortion disappears due to the fact that surface tension becomes much more important than gravity at these sizes.

What has been elusive to date is the three-dimensional structure of the Marangoni toroidal vortex that forms in the liquid phase of the meniscus and is driven by surface tension gradients. This due to the limitation of experimental techniques due to the small tube sizes involved (below 1mm). In the present work  $\mu$ -Particle Image Velocimetry is used to unveil the two-dimensional flow patterns in horizontal and vertical diametrical sections of the capillary tube. Then one single tracer particle has been used to map the three-dimensional trajectory by using a very powerful technique: Digital Holographic Microscopy.

## **Experimental techniques**

In this work the authors characterise the self-induced Marangoni convection arising from differential evaporation of ethanol along the curved meniscus interface formed inside borosilicate capillary tubes. Two techniques are used to unveil the three-dimensional flow pattern in the meniscus liquid phase:  $\mu$ -Particle Image Velocimetry ( $\mu$ -PIV) and Digital Holographic Microscopy (DHM).

 $\mu$ -PIV relays on using tracers particles which are illuminated by coherent light from a laser. A microscope and a CCD camera are used to follow the particles; the  $\mu$ -PIV pairs of images are recorded and stored in a dedicated PC. The cross-correlation is performed by using DANTEC PIV software suite. The experimental apparatus is reported in Figure 1 and is the same as that used in [8], where specific details are given. For  $\mu$ -PIV measurements a round tube of 900 $\mu$ m ID has been used.

DHM is used for the first time to track a single tracer particle and unveil the three-dimensional trajectory characteristic of the problem at hand. Over the last 10 years, digital holography has shown a growing interest in the scientific community and particularly in the field of microscopy. Classical microscopy is suffering from a very small depth of focus due to the high numerical apertures (NA) of the microscope lenses and the high magnification. Confocal microscopes strongly enlarges the depth of field of classical microscopes but are not adequate for non-static phenomenon's as it requires a scanning point by point in the three directions. DHM overcomes this classical limitation by increasing the depth of field by a factor of one hundred in a single frame acquisition. Digital holograms are recorded by a CCD camera and processed to compute the complex amplitude of the light field involving the intensity and the phase of the signal. The Kirchhoff-Fresnel propagation is then used to propagate the complex amplitude and to perform a digital holographic refocusing of the sample slide by slide. DHM is an elegant solution for three-dimensional tracking of small particles [14] as it can easily provide their 3D position inside an experimental volume.



Figure 1: Schematic diagram of the experimental setup (unheated tubes horizontally oriented);

g is the gravitational acceleration.

The set-up of the DHM is reported schematically in Figure 2 where all components are mentioned. Holograms of 1280 x 1024 pixels are captured at a frequency of 24 frames/sec covering a field of view of 1057 x 846  $\mu$ m (camera JAI CV-M4). The depth investigation is performed slide by slide in the range [0-1000  $\mu$ m] (the focus plane of the microscope is placed on the bottom wall of the capillary, the propagation direction of light is considered as positive and the reconstruction direction negative). With this magnification, the precision on the position of an object in the X and Y direction is about 1  $\mu$ m and about 20  $\mu$ m in the Z direction. The sample is a 1mm square borosilicate tube; a square section is chosen for DHM measurements to avoid large optical distortions and lens effect.

# **Results and discussion**

The evaporation of a volatile liquid (without external heating) inside a small capillary tube is characterised to be much higher at the meniscus wedge (triple line) than in the middle. This creates differences of temperatures (which have also been measured by InfraRed thermography [9]) that generate surface tension gradients because of the small size (below 1mm). It is the gradient of surface tension that drives the convection reported in this work.



**Figure 2:** Optical setup of the Digital Holographic Microscope. L1, focusing lens; RGG, rotating ground glass for spatial coherence reduction; L2, collimating lens; L3, L4, identical microscope lenses (x10); L5, refocusing lens; CCD, charge-coupled device camera; M1–M3, mirrors; BS1, BS2, beam splitters; S sample.



Figure 3: Superimposed of 20 images of tracers particles.

Figure 3 reports the superimposition of 20 frames to show the Marangoni convection cells in a horizontal diametrical section of a round tube. As can be seen, the Marangoni cells are symmetrical with respect to the tube axis. This can also been seen from Figure 4 which is a  $\mu$ -PIV image reporting the velocity vectors, the streamlines and the vorticity (colour map), this latter being a derivation of the velocity map. In horizontal diametrical sections of the tube gravity does not act and therefore the flow pattern is symmetrical with respect to the tube axis because of the symmetry of the surface tension driving force.



Figure 4: PIV analysis of horizontal diametrical section of the capillary tube.



Figure 5: PIV analysis of vertical diametrical section of the capillary tube (g is the gravitational acceleration).



Figure 6: Tracer particle trajectory (2-D projection).



Figure 7: X, Y and Z time evolution of the tracer particle position.

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Instead in vertical diametrical sections of the tube gravity acts in the same direction as surface tension in the lower part of the tube and against it in the upper part of the tube. Therefore, the flow patter in vertical diametrical sections is strongly deformed as reported in Figure 5. There is a single vortex which occupies all the cross section.

The limitation of  $\mu$ -PIV used here is that it does not allow visualising the three-dimensional nature of the Marangoni vortex. This problem is solved here for the first time with the use of DHM by tracking a single particle.

In Figure 6 the two-dimensional projection of the tracer trajectory is reported. What this figure shows is that the particle makes for each turn a different loop and after four loops it returns to the original loop. In fact the particle trajectory starts at C and finishes at D. It makes five loops from I to V, where this latter is identical to loop I. The dashed parts of the trajectory in Figure 6 are not real because it is difficult to follow the tracer when it is very close to the meniscus interface because of the meniscus curvature.

Figure 7 reports the time evolution of the X, Y and Z components of the tracer as it goes around in the Marangoni three-dimensional cell. From this figure it is clear that the trajectory is periodic (see for instance the loops between L and M). By analysing several loops it was found that the characteristic frequency of the periodic oscillatory motion is around 0.125 Hz.

## Conclusions

This paper reports an experimental investigation of Marangoni convection of evaporating liquid inside borosilicate capillary tubes. The convection is due to differential evaporation along the curved meniscus interface which leads to surface tension gradients that are the driving force. It was reported in previous works (also by one of these authors) that the Marangoni toroidal cell is distorted in vertical diametrical sections of the horizontally oriented tube because of the action of gravity that acts in opposite direction to surface tension. However, the truly three-dimensional nature of the Marangoni vortex had not been reported before the present work. In this work  $\mu$ -Particle Image Velocimetry and the powerful Digital Holographic Microscopy have been used to unveil the three-dimensional shape of the Marangoni roll.

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