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RESEARCH ARTICLE

Marangoni convection in evaporating meniscus with changing contact angle

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Abstract In this work, the Marangoni convection in the liquid phase of an evaporating meniscus interface in open air has been studied for varying contact angles. Ethanol undergoes self-evaporation inside a capillary tube of borosilicate glass with internal diameter of 1 mm. The evaporation is not uniform along the meniscus interface pinned at the capillary tube mouth, and this creates a gradient of temperature between the wedge and the centre of the meniscus. It is this temperature difference and the scale (1 mm) that generate a gradient of surface tension that is acknowledged to drive the vigorous Marangoni convection in the meniscus liquid phase. In previous studies of this configuration, the meniscus has mainly been concave and for this reason, other researchers attributed the differential temperature along the meniscus to the fact that the meniscus wedge is closer to the tube mouth and also further away from the warmer liquid bulk than the meniscus centre. The present study investigates concave, flat and convex meniscus by using a syringe pump that forces the meniscus to the wanted shape. With the present investigation, we want to further demonstrate that it is instead the larger evaporation at the meniscus triple line near the wedge that controls the phenomenon. Flow visualization and infrared temperature measurements have been performed. For concave and convex meniscus, the temperature measurements are in line with the predicted trend; the Marangoni vortices for these two menisci shapes spin in the same direction according to the temperature differences along the meniscus. For a flat meniscus, an intriguing experimental evidence has been found: the temperature difference is inverted with respect to concave and convex menisci, but surprisingly, the Marangoni vortices spin in the same direction than for concave and convex menisci.

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1 Introduction

Evaporation is an important two-phase mechanism by which large amounts of heat and mass are transferred between the liquid and vapour phases. Several important industrial applications rely on evaporation from drying of paints, to combustion, to computer chips cooling, to heat pipes, just to mention a few. Evaporation leads to a temperature drop at the liquid–vapour interface, and it is this reduction of temperature respect to the surrounding ambient that supplies the heat to sustain the phase change. Temperature differences along a thin liquid film have long been reported to generate vigorous convection, which was first attributed to the role of surface tension by Pearson in 1958, while the Marangoni number was first introduced in 1960 by Scriven and Sternling (1960). The temperature difference along or across the thin liquid layer was imposed. More recently, a different experiment has been studied (Buffone and Sefiane 2004a, b; Buffone et al. 2005; Dhavaleswarapu et al. 2007; Chamarthy et al. 2008), where a curved meniscus interface is formed inside a small capillary tube; the liquid (an alcohol) evaporates spontaneously in the open air because of its low partial pressure in air. The evaporation is not uniform (Buffone and Sefiane 2004) being more intense at the meniscus wedge than in the centre, and this causes temperature differences, which were first reported by Buffone and Sefiane (2004). Since surface

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tension varies significantly with temperature and the small tube size involved (here below 1 mm), this generates gradients of surface tension that drive the liquid from warmer regions (meniscus centre) to colder ones (meniscus wedge). This vigorous liquid motion has been referred as to Marangoni convection.

Marangoni convection is very important to many industrial applications; among these are drying of paints, ink-jet printing, welding (Frischat et al. 1980), glass manufacturing (Ramanan and Korpela 1990), crystal growth (Schwabe 1981) and the recently intensively researched evaporation of drops (Sefiane et al. 2008; Savino and Fico 2004; Sobac and Brutin 2012; Hu and Larson 2005; Zhang et al. 2014).

When an alcohol fills a capillary tube, a concave meniscus is formed, and this meniscus is then pinned at the tube mouth. During evaporation, the pinned meniscus remains fixed at the tube mouth, while the second meniscus inside the tube recedes (moves towards the pinned meniscus at the tube mouth) to account for the mass lost due to evaporation at the pinned meniscus. The differential evaporative cooling effect along the curved meniscus interface has been found to be responsible for the differences in temperature, and consequently, surface tension that drives the Marangoni convection reported in many studies (Buffone and Sefiane 2004a, b; Buffone et al. 2005). Wang et al. (2008) postulate a different explanation related to the diffusion of vapour in still air above the meniscus. They argue that the evaporation rate is higher at the meniscus wedge than at the centre; this is because the meniscus wedge is closer to the tube mouth than the centre, and therefore, the vapour transports more efficiently at the wedge. Pan et al. (2011) also argue that the wedge is further away from the warmer liquid bulk than the centre for a concave meniscus.

The differential evaporation rate along a drop with large temperature differences has also been extensively studied in recent years (Sefiane et al. 2008; Savino and Fico 2004; Sobac and Brutin 2012; Hu and Larson 2005; Zhang et al. 2014). The mechanism at work in drops is the same that in the curved meniscus of the present case. The meniscus or drop triple line region (where vapour, liquid and solid meet) is of paramount importance because the evaporation and heat transfer peak in this region, generating an evaporative cooling effect of the liquid–vapour interface. It seems that it is this differential evaporation and the consequent temperature differences that drive a vigorous convection inside the liquid phase of the meniscus and the drop.

Instabilities of the toroidal Marangoni vortex have been recently reported in Pan et al. (2011) for a meniscus formed inside a capillary positioned vertically with the meniscus at the bottom in order to minimize buoyancy effects. The authors showed that the Marangoni flow in a concave meniscus is always symmetrical to the tube axis, whereas that on a convex meniscus is reversed with respect to the concave meniscus, which results in an instability that is easily perturbed into a non-axisymmetric rolling motion. Additionally, it was found that the Marangoni flow from the meniscus wedge to its centre is not to be found as stable as the opposite flow. Pan et al. (2011) argue also that for a meniscus convex enough, the wedge should become warmer than the centre and the Marangoni convection should reverse.

The Marangoni convection inside the evaporating meniscus is three dimensional (Buffone et al. 2005; Chamarthy et al. 2008; Minetti and Buffone 2014) due to the competing gravity and surface tension forces. For a horizontally oriented capillary tube, in horizontal optical diametrical sections, two counter-rotating vortices are found; instead, for a vertical optical diametrical section, a single vortex is found. This is due to gravity distorting the symmetry in vertical optical diametrical sections.

In the present investigation, the authors conduct a new experiment. In previous experiments, the meniscus had always a concave shape for horizontally positioned tubes and also for tubes vertically positioned and plunged inside a liquid pool. In the present experiment, the tube is positioned horizontally and only one meniscus is let to form and pinned at the tube mouth. At the other end of the tube, a syringe pump is connected, which supplies the capillary with a fixed flow rate so as to keep the pinned meniscus at a given shape. Three meniscus shapes were investigated, namely concave, flat and convex. The goal of this experiment is to prove or disprove the theory postulated by Wang et al. (Wang et al. 2008) and Pan et al. (2011) that the meniscus wedge is colder than the its centre because it is closer to the tube mouth and further away from the liquid bulk.

2 Experimental facility and procedure

The experimental facility employed for flow characterization and contact angle measurement in this study is reported in Fig. 1; on the left is a photograph of the apparatus, and on the right is a schematic of the same apparatus. It comprises a microscope, a complementary metal-oxide semiconductor (CMOS) camera, a micro-syringe pump, the test cell with the capillary tube and a laptop with specialized software for image acquisition and processing. The working liquid is ethanol, and the capillary tube is made of borosilicate glass with ID of 1 mm and OD of 1.2 mm with a length of 100 mm. The capillary tubes and the liquid are used as received from the manufacturers. The DMK23GV024 CMOS camera has 752 × 480 pixels

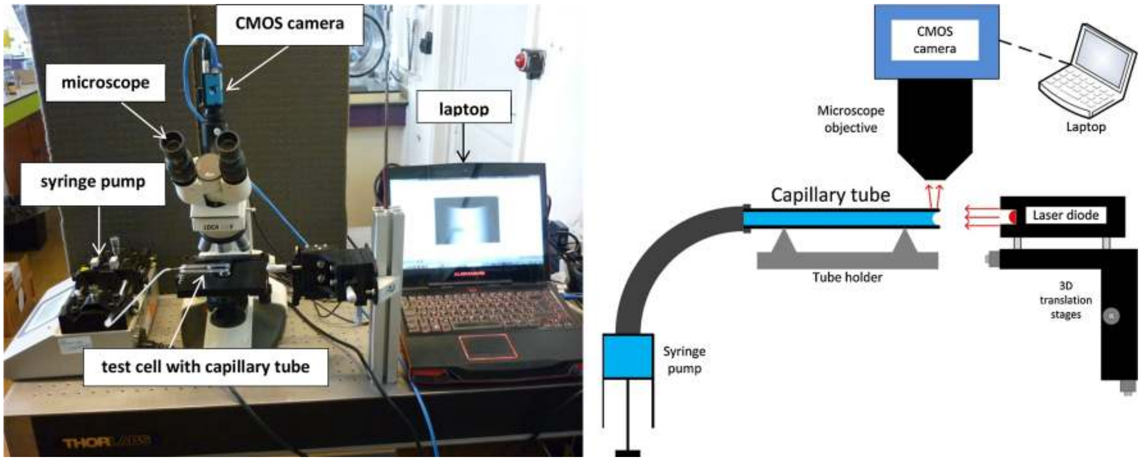
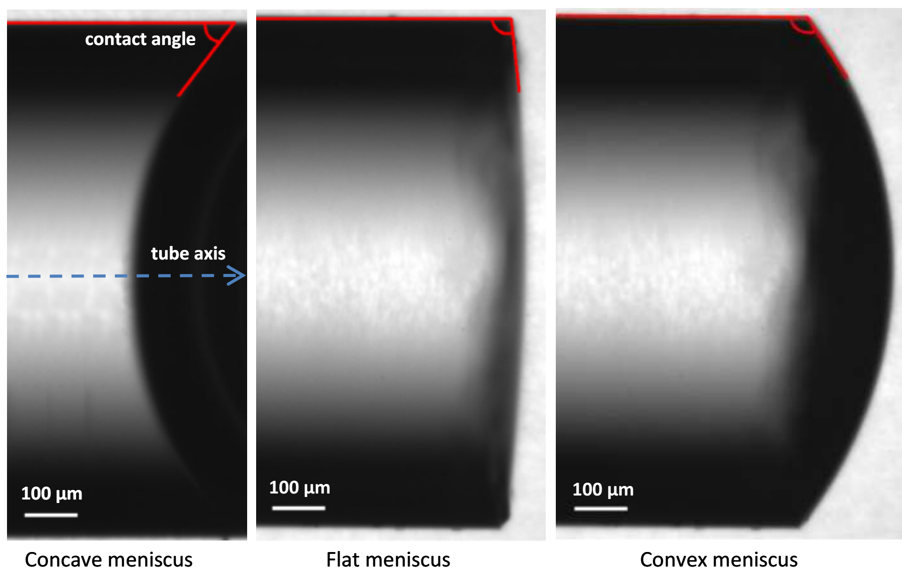


Fig. 1 Experimental set-up for flow visualization and contact angle measurement. On the *left* a photograph of the experimental apparatus and on the *right* the schematic of the same apparatus

Fig. 2 Different meniscus shapes as a function of imposed mass flow rate. In *red,* the measured contact angle



array, and given the microscope magnification, a resolution of around 2 µm is achieved. Both horizontal and vertical optical diametrical sections of the capillary tubes have been analysed in this study. Apart from the results presented on Fig. 2, the samples were illuminated along the tube axis by a red laser diode fixed close to the capillary mouth to better contrast the images.

Different from previous studies of one of these authors, in the present investigation, only one meniscus is present, and it has been pinned at one tube mouth. The syringe pump injects a defined flow rate of ethanol in the capillary such that it compensates the evaporation at the tube mouth and keeps a steady meniscus shape (concave, flat or convex). The imposed ethanol flow rates were as follows: 0.5 µl/min for the concave meniscus, 0.6 µl/min for the flat meniscus and 0.8 µl/min for the convex meniscus.

The meniscus shape was monitored before taking all the measurements presented below, and no appreciable change was observed. After the measurements were taken, the meniscus shape was again inspected and no appreciable change was detected.

To check for repeatability, three different runs have been performed at three different flow rates. The contact angles were then measured under white light according to Fig. 2 and are reported in Table 1. They displayed a relatively good reproducibility.

The flow was also seeded with polystyrene latex beads with an average diameter of 5 µm to unveil the flow structure inside the liquid phase of the meniscus and detect the flow direction for changing contact angle. In particular, the tracer velocity has been measured along the two Marangoni vortex sections along the tube axis in the part

Table 1 Measurement of contact angle for different meniscus shapes

Meniscus shape	I° measurement	II° measurement	III° measurement	Mean value
Concave	66	63	63	64
Flat	95	92	91	92.7
Convex	126	124	128	126

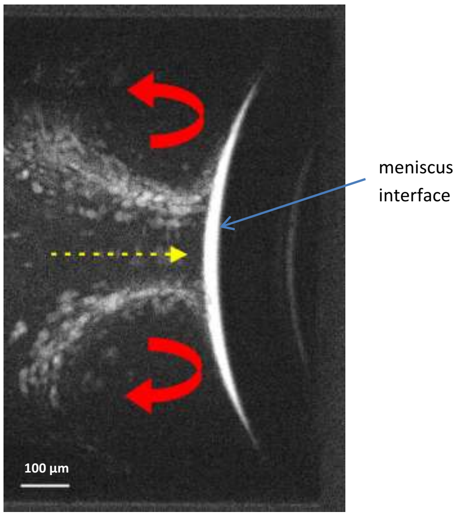


Fig. 3 Two symmetrical optical sections of Marangoni roll for a horizontal diametrical optical section of the capillary tube unveiled by tracer particles with opposite spinning direction shown by *red curved arrows*. The *dashed yellow arrow* is the tube axis, and it is the direction along which the tracers velocity reported on Table 2 has been measured

towards the meniscus interface as shown schematically by the yellow arrow on Fig. 3. The concentration of polystyrene latex beads is at best fractions of a percentage point, and therefore, it is not expected that the surface tension of ethanol is affected by the particles.

The gravitational forces arising from the mismatch between tracers and fluid density are evaluated with the following formula [coming from Stokes's drag law (Markus et al. 1998)]:

$$U_{zP} - U_z = d_P^2 \frac{\rho_P - \rho}{18\,\mu} g$$

where U_{zP} and U_z are the particle and fluid vertical velocity components, respectively, d_P is the particle diameter, ρ_P (1,050 kg/m^3) and ρ (789 kg/m^3) are the particles and fluid density, respectively, µ (0.0012 Pa s) is the fluid dynamic viscosity, and g is the gravitational acceleration. The aforementioned formula for the present case leads to:

$$U_{zP} - U_z \cong 3 \times 10^{-6}\,\mathrm{m\,s^{-1}}$$

where the average particles velocity evaluated along the meniscus ($U_{zP} \cong 2 \times 10^{-3}\,\mathrm{m\,s^{-1}}$) is much higher. Therefore, we can assume that the particles behave as neutrally buoyant. A second, separate experiment has also been conducted to measure the interfacial temperature with an infrared (IR) camera. The infrared camera has been pointed at the curved meniscus interface at the tube mouth in the axial direction. The disposition of the IR camera is the same as used during a previous investigation (Buffone and Sefiane 2004) and is reproduced in Fig. 4. To check the meniscus shape, before IR measurements were taken a high-magnification teleobjective lens connected to a CMOS camera has also been used. The IR camera of the present study is a Flir SC5000 with 320 × 256 pixel resolution, a 20 mK sensitivity and a 1 % accuracy, which translate in 1.5 °C accuracy at full scale for temperature up to 150 °C. The camera is equipped with an indium antimonide (InSb) focal plane array detector and operates in the 2.5- to 5.1-µm wavebands. The IR camera has a microscopic lens with field of view 19.2 × 14.4 mm; the depth of focus is 1 mm. This would allow the measurement of the temperature profile along the entire meniscus, which was not possible with the IR camera used in a previous study (Buffone and Sefiane 2004), where the depth of focus was only 100 µm. However, the better microscopic lens used by Buffone and Sefiane (2004) allowed for better spatial resolution 31.25 µm compared with the 60.19 µm of the present IR camera. For IR measurements, horizontal diametrical temperature profiles of the tubes have been analysed.

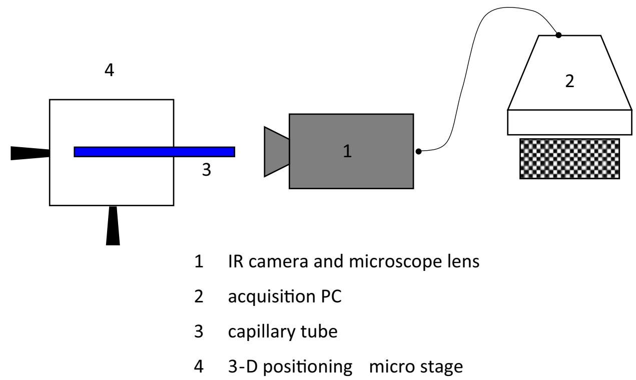


Fig. 4 Sketch of the experimental set-up for IR investigation with the use of FLIR SC5000 camera

It is important to point out that ethanol is a semitransparent fluid to IR at the camera wavelengths of 2.5–5.1 µm. The emissivity of ethanol depends on the liquid thickness as clearly shown also by Brutin et al. (2011) for drops. Therefore, the IR measurements of the present investigation give an indication of the temperature distribution of the liquid close to the meniscus interface, but not of the interface itself.

In order to check the IR camera readings, we took a heated plate from below and covered the upper surface with matt black paint (to have high emissivity, above 0.95).

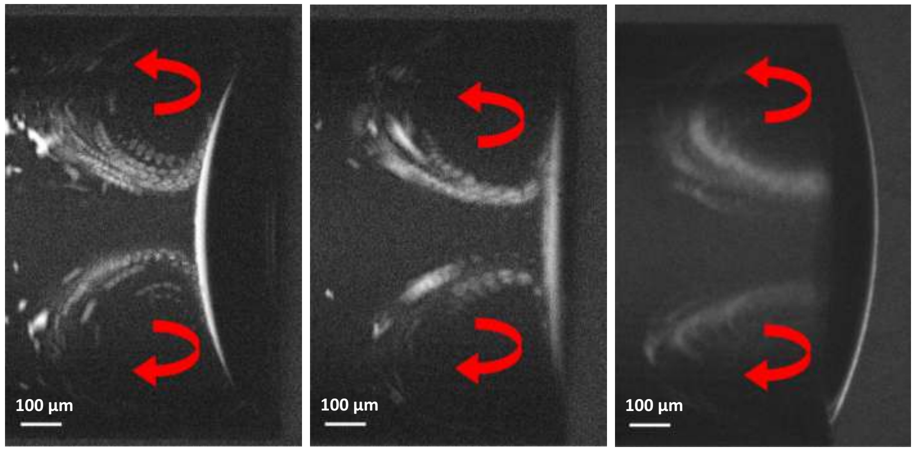


Fig. 5 Marangoni flow structure in horizontal diametrical section of the capillary tube for the three meniscus shapes investigated. *Red curved arrows* indicate the spinning direction of the vortices

We attached a calibrated platinum resistance temperature detector PT100 to the upper surface and compared its temperature with the IR reading for three heater power settings. The difference between IR camera and PT100 sensor readings is found to be less than 6 %.

3 Results and discussion

Figure 5 reports the superimposition of several consecutive frames of the tracers' position. In Fig. 5, arrows are added to show the direction of the Marangoni vortex sections. As can be seen, two counter-rotating symmetrical vortex sections are present in the horizontal diametrical section of the capillary tube; these are optical sections of the toroidal Marangoni vortex also presented and discussed in Buffone et al. (2005) for a concave meniscus.

The tracer particles are accelerated along the meniscus interface because of the surface tension gradient generated by the differences in temperature due to differential evaporation along the meniscus. From the corners of the tube, they move back towards the liquid bulk for continuity and then again for continuity they are accelerated in the middle of the tube towards the meniscus interface. Even without a syringe pump (when both ends of the capillary tube are open and there are two menisci), there is a noticeable supply of liquid from the bulk to account for mass lost at the pinned meniscus. This natural resupply of liquid flow goes to stabilize the Marangoni convection taking place in close proximity of the meniscus interface. The length of the tube inside which there is a vigorous Marangoni convection pattern is roughly one diameter from the meniscus interface.

Figure 5 reports the Marangoni roll structure for the three meniscus shapes: concave (on the left), flat (on the middle) and convex (on the right). As shown, the Marangoni rolls structure appears to be weakly affected by the shape of the meniscus interface. Surprisingly, the direction of the Marangoni rolls is always the same independently of the meniscus contact angle. According to Wang et al. (2008) and Pan et al. (2011), for a concave meniscus, the wedge is closer to the tube mouth and further away from the liquid bulk with respect to the meniscus centre, and therefore, the wedge is colder than the centre of the meniscus. Therefore, the Marangoni rolls spin from the centre to the wedge bringing hotter fluid to the triple line region at the meniscus wedge. For a flat meniscus, the wedge is at the same level as the centre, and therefore, following the arguments of Wang et al. (2008) and Pan et al. (2011), the meniscus should have a uniform temperature. Additionally, for a convex meniscus following the arguments of Wang et al. (2008) and Pan et al. (2011), the wedge is closer to the liquid bulk and therefore should be warmer than the meniscus centre. If the argument of Wang et al. (2008) and Pan et al. (2011) was true, then for flat and convex meniscus there should be a substantial change in the Marangoni flow with the inversion of the spinning direction for convex meniscus. What we see in Fig. 4 instead is that the Marangoni rolls spin always in the same direction regardless of the shape of the meniscus.

Table 2 Tracer particle velocity along the capillary tube axis towards the meniscus interface

	Concave	Flat	Convex
Vx (µm/s)	4,077	6,735	2,304

In Table 2, the tracer particle velocity (the component in the capillary tube axial direction) in the path towards the meniscus interface as indicated in Fig. 3 has been measured for concave, flat and convex meniscus by measuring the distance travelled by particles between two consecutive frames as recorded by the camera (10-ms steps between frames). The flat meniscus exhibits the largest particle tracers' velocity. The convex meniscus has the smallest

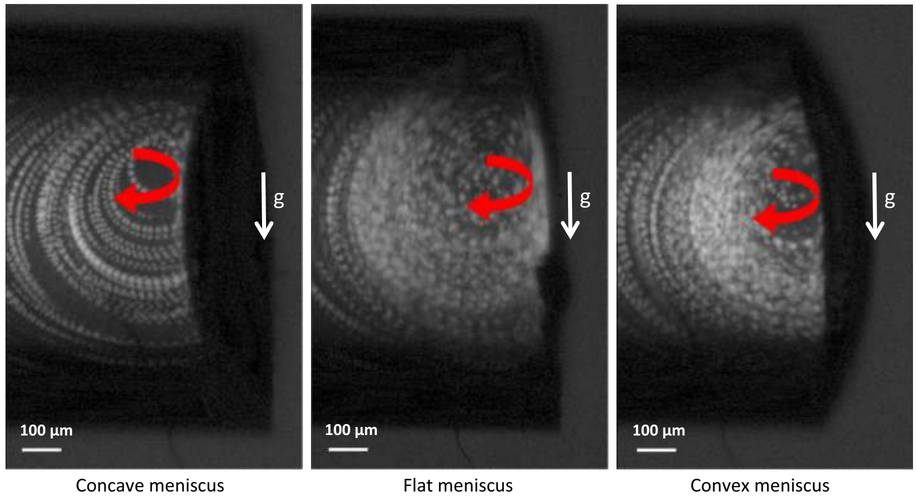


Fig. 6 Marangoni flow structure in vertical optical diametrical section of the capillary tube for the three meniscus shapes investigated. *Red curved arrows* indicate the spinning direction of the vortices. *g* is the gravitational acceleration

tracers' velocity. This experimental evidence is counter-intuitive and needs a more detailed study. It might be due to the shape and dimensions of the Marangoni vortex, which unfortunately cannot be assessed properly in the present study. As the flow rate changes, the inner most part of the Marangoni roll might change in width, and this can have a very important influence on the particle velocity towards the meniscus interface.

Figure 6 shows the flow patterns unveiled by tracer particles in vertical optical diametrical sections of the capillary tubes for concave, flat and convex menisci. It can be seen that there is only one vortex instead of the two counter-rotating vortices of Fig. 5 for horizontal diametrical sections. As indicated by the red arrow in Fig. 6, the spinning direction of the tracer particles is the same for concave, flat and convex menisci, further supporting the evidence shown in Fig. 5 that the direction of the Marangoni roll is the same independently of the meniscus shape.

The IR measurements of temperature profiles along the meniscus interface are reported in Fig. 7 for the three meniscus shapes. The temperature profile for the case of concave meniscus is similar to previous measurements reported in Buffone and Sefiane (2004), which shows a trough at the meniscus wedge where the triple line (where vapour, liquid and solid meet) is found. In the meniscus centre, the temperature is higher than at the wedge. This fact has been attributed to the large evaporation taking place at the meniscus triple line region as clearly shown by Pratt and Hallinan (1997), Pratt et al. (1998), Sartre et al. (2000), Hohmann and Stephan (2002) and Buffone and Sefiane (2005). The large evaporation at the meniscus triple line region compared to its apex is also found in evaporating drops (Steinchen and Sefiane 2005).

The IR measurements of Fig. 7 show that the meniscus centre is warmer than the wedge for the concave and convex shapes. This fact is corroborated by the results on

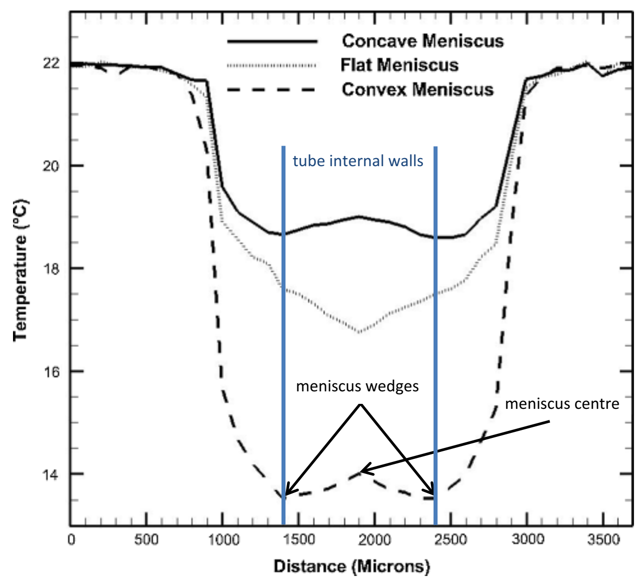


Fig. 7 Temperature profiles of meniscus interface for different meniscus shapes. The *vertical lines* represent the tube internal surfaces

the Marangoni rolls spin direction of Fig. 4, where the tracers moves from the centre to the wedge of the meniscus. What is really intriguing is the temperature profile of Fig. 7 for the flat meniscus. In fact, this temperature profile shows a colder centre than the wedge, which presumably means larger evaporation at the centre than at the wedge. This experimental finding is not corroborated by the Marangoni roll spin direction of Fig. 5 for the flat meniscus; in fact, for the flat meniscus, the spinning direction of the tracers is also from the centre to the wedge of the meniscus interface. Figure 7 also shows that the average temperature of the meniscus is relatively colder going from concave to flat and finally convex meniscus because of an increasing average evaporation rate.

There are two completely different views on the temperature difference along a sessile drop as explained in Zhang et al. (2014). Deegan et al. (2000) studied the deposits of solute in evaporating drops, which migrates to the outer edges of the drop; in their work, they postulate that the apex of the drop is colder than its edge because it is further away from the substrate. Steinchen and Sefiane (2005) studying the evaporation of drops of pure liquid arrive to a completely different conclusion; they state that most of the evaporation takes place at the edge of the drop (where the triple line is found) and that this region becomes therefore colder than the apex of the drop. Zhang et al. (2014) clearly show that for evaporating drops the surface temperature gradient reverses its direction at the critical contact angle. In addition, the profile of surface temperature is monotonic away from the critical contact angle and becomes non-monotonic in the transition around the critical contact angle.

The evaporation from a meniscus formed in a capillary tube such as in the present experimental investigation has attracted some attention in the last decade but has not been extensively studied as the evaporation of drops. In addition, there are only few cases where different meniscus shapes (contact angle) have been studied. It is therefore important that more research is carried out for an evaporating meniscus inside a capillary tube, where many more meniscus contact angles are investigated to better understand the temperature profiles and the consequent Marangoni flow.

4 Conclusions

This paper is an experimental investigation of Marangoni convection in the liquid phase of an evaporating meniscus of ethanol in still air inside a borosilicate capillary tube with an internal diameter of 1 mm. Only one meniscus is pinned at tube mouth, and the mass lost by evaporation is supplied through the opposite end of the capillary tube by a syringe pump. Three different meniscus shapes (contact angles) have been investigated, namely: concave, flat and convex meniscus. The liquid close to the meniscus has been seeded by tracer particles in order to unveil the vigorous Marangoni convection. Infrared temperature measurements of the meniscus interface have also been performed. The goal of the study was to further understand the origin of the Marangoni convection. Other researchers have postulated that the differential evaporation along the meniscus, and its consequent temperature differences are due to the fact that the meniscus wedge is closer to the tube mouth and further away from the warmer liquid bulk than the meniscus centre. The present authors sustain a different explanation; it is the important evaporation rate at the meniscus triple line region close to the meniscus wedge with respect to meniscus centre that creates the temperature differences, which produce surface tension gradients that ultimately drive the Marangoni convection. The present study found that for a concave and convex meniscus, the Marangoni vortices spin in the same direction, and this is corroborated by the temperature measurements, which have a trough at the meniscus wedge and drive warm liquid from the centre to the wedge. An intriguing result has been found for the flat meniscus: the Marangoni vortices spin in the same direction as for concave and convex meniscus, but the temperature measurements shows that the meniscus centre is colder than the wedge. This fact clearly requires some deeper investigation.

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