



## Self-induced Marangoni flow in evaporating alcoholic solutions



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### ABSTRACT

The self-induced Marangoni convection in alcoholic solutions is the subject of the present experimental investigation. Pure ethanol and its mixtures with 5%, 10% and 20% in weight of water are presented and discussed. In particular, Marangoni flow in horizontal pipes from 100 to 1000  $\mu\text{m}$  inner diameter is studied. Vortex spinning frequency, average particle tracers velocity and evaporation rate are measured and discussed. The evaporation rate increases and the evaporation flux decreases at bigger tube sizes in line with previous investigations; pure ethanol has higher evaporation rate and flux than ethanol/water mixtures. The spinning frequency and the average tracer particles velocity decrease for increasing water content in the mixtures. All of these findings are due to evaporative cooling effect which is higher at the meniscus wedge (where the triple-line region is found) than at the meniscus center; this causes a difference in temperature between the wedge and the center that generates a gradient of surface tension driving vigorous Marangoni convection, that has been reported and analyzed. The experimental results are explained on the basis of a numerical model including evaporation, vapor diffusion, heat and mass transfer from the liquid to the surrounding ambient and the Marangoni effects.

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### 1. Introduction

Many industrial applications such as combustion, spreading of drops, evaporation and condensation of liquid films rely on thermocapillary phenomena. Usually, thermocapillary flow is important in applications with temperature gradients on free surfaces such as crystal growth [1], welding and glass manufacture [2]. Recent extensions of these studies find applications to problems encountered in micro-electro-mechanical devices, or for the development of more efficient heat transfer and thermal control systems, micro heat pipes, micro fluidic systems. An overview of different scientific developments on the subject, with particular attention to potential technological applications, is presented in Ref. [3].

Phenomena arising from interfacial tension gradients have been initially discovered in the frame of chemical engineering processes, where fluid flows arising from surface composition differences play a major role. Solutal Marangoni instabilities typically arise in systems involving mass transfer of surface-active compounds. However, there are many other situations where interface tension-driven effects have practical importance, for instance convective flows induced in non-uniformly heated liquids by surface tension variations induced by temperature gradients (sometimes

addressed as thermocapillary convection). In the 1950s Pearson [4] investigated a liquid layer heated from below and attributed the observed convection pattern to surface tension; he also introduced for the first time the Marangoni number.

Unfortunately, in most circumstances under normal gravity conditions on Earth, Marangoni effects are masked by natural convection induced by density differences. For this reason, surface tension effects within liquids have attracted the attention of several scientists in microgravity experiments on space laboratories. On the other hand, the role of surface tension becomes dominant decreasing the Bo number, i.e. decreasing the system size and or in microgravity conditions. This is the case of evaporating drops or menisci commonly used in a number of heat transfer devices like heat pipes and micro-channels using porous wicks and grooved structures [5].

Kavehpour et al. [6] have shown that the evaporation of silicon oil drop can lead to interfacial temperature gradients that generate thermocapillary stresses. Zhang and Chao [7] attributed the convective patterns in an evaporative liquid layer to the cooling of the interface because of the higher evaporation rates. Pratt and Hallinan [8] and Pratt et al. [9] investigated the stability of pentane inside a vertically oriented capillary tube with the meniscus positioned below a heater. They observed that thermocapillary stresses arising from temperature gradients are responsible for meniscus recession down the capillary tube. The authors did not investigate the hydrodynamics of the flow in the meniscus liquid phase.

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## Nomenclature

$x$	liquid mole fraction		
$y$	liquid vapor fraction		
$c$	concentration		
$D$	ethanol/air diffusion coefficient		
$D_L$	ethanol/water diffusion coefficient		
$h$	heat transfer coefficient		
$H_{LV}$	latent heat of vaporization		
ID	inner diameter		
$J$	evaporative mass flux		
$P$	pressure		
$P_s$	saturated vapor pressure		
$\dot{q}$	heat flux		
$S$	surface coordinate		
$T$	temperature		
$V$	velocities		
		<i>Greek symbol</i>	
		$\rho$	density
		$\mu$	viscosity
		$\alpha$	thermal diffusivity
		$\sigma_T$	surface tension derivative
		$\tau$	viscous shear stress
		<i>Subscripts</i>	
		$L$	liquid
		$V$	vapor
		$S$	saturated

Evaporation in capillary tubes has been also the subject of a number of publications by one of the present authors. Evaporation rate and fluxes on a single receding and double menisci (one of which pinned at the capillary tube mouth) has been reported in Buffone and Sefiane [10] for different tube sizes and alcohols; the authors also reported the vortex cross section frequency with some maps of  $\mu$ -Particle Image Velocimetry ( $\mu$ -PIV). The authors conclude that the non-uniform evaporation at the meniscus interface is responsible for a strong convection in the liquid phase driven by surface tension gradient. The mass flux and the vortex frequency are found to be higher for smaller tube sizes.

In Buffone and Sefiane [11] the authors measure the temperature profile along the meniscus interface at the tube mouth and on the side of the tube. The measurements have been performed for different alcohols and tube sizes and clearly show the sink effect at the meniscus triple line due to the peak of evaporation in that region. Buffone et al. [12] reported the important distortion of the toroidal Marangoni convection vortex inside capillary tubes horizontally positioned for methanol and ethanol; they found by the use of  $\mu$ -PIV that the horizontal cross sections of the vortex cells are symmetrical with respect to the tube axis, whereas the vertical cross section is asymmetrical. Dhavaleswarapu et al. [13] reproduced the results of Buffone and Sefiane [10] and Buffone et al. [12] by analyzing five different tube sizes ranging from 75 to 1575  $\mu\text{m}$  using  $\mu$ -PIV. They found that the evaporation rate and flux scale parabolically and hyperbolically respectively instead of linearly as in Buffone and Sefiane [10]; the vorticity scales hyperbolically with the tube diameter in contrast with the linear relationship of Buffone et al. [12]. Dhavaleswarapu et al. [13] argue that Buffone and Sefiane [11] and Buffone et al. [12] used only three tube sizes for their experiments.

Chamarthy et al. [14] reported  $\mu$ -PIV measurements of horizontally oriented capillary tubes with ethanol as working fluid; in particular they looked at different horizontal cross sections of the tube and also at the vertical cross section. They found distortion of the flow field which they attributed to the action gravity; interestingly they reported that there is no distortion of the flow pattern for capillary sizes of 75  $\mu\text{m}$ . This is a very interesting finding with important repercussions in industrial and laboratory applications such as crystal growth. Instabilities of the toroidal Marangoni vortex have been reported in Pan et al. [15]; the authors positioned an evaporating meniscus of ethanol in a 625  $\mu\text{m}$  cylindrical channel. They show that the Marangoni flow in a concave meniscus is always symmetrical to the tube axis whereas that on a convex meniscus loses symmetry with only one vortex occupying the whole

channel. Additionally, it was shown that the inwards (from the meniscus wedge to its center) Marangoni flow is found not as stable as the outwards flow. They also found that for the inward flow to lose symmetry the bulk fluid must be warmer than the meniscus and the Marangoni number must be above a specified value.

Instabilities on the Marangoni flow, the meniscus position and its temperature were reported in Buffone et al. [16] for ethanol in a 900  $\mu\text{m}$  cylindrical horizontal channel. The authors developed a linear stability analysis and attributed the instabilities to the competition of gravity and surface tension forces. Marangoni oscillatory periodic instabilities in a 1 mm square glass tube filled with ethanol have also been recently reported in Minetti and Buffone [17], where digital holographic microscopy has been used to trace a seeding particle in its three-dimensional trajectory.

In the present work Marangoni flows induced by the self-evaporation in horizontal capillary tubes have been studied considering binary mixtures. Pure ethanol and ethanol/water mixtures of different concentrations have been investigated using capillary tubes of 100, 200, 400, 600, 800 and 1000  $\mu\text{m}$  internal diameters. Works on Marangoni convection in binary mixtures are rare as also reported by Zhang et al. [18]. The authors work with liquid films of NaCl and water in open air; they find that evaporation is important to the flow pattern at the beginning whereas the Soret effect becomes significant at later stages. For the present experimental investigation, to the authors' knowledge it is the first time that such study inside capillaries tubes has been carried out. The Marangoni convection is characterized by measuring the evaporation rate, the Marangoni vortex spinning frequency and the velocities in the liquid. The reported Marangoni convection is self-induced because of the differential evaporation rate along the curved meniscus interface. Analyses of the experimental results are given under the assumption of diffusion limited evaporation process in the vapor phase. Marangoni convection and particle trajectories have been reconstructed numerically using a computational fluid dynamic model.

## 2. Experimental setup

Capillary tubes of borosilicate glass with ID of 100, 200, 400, 600, 800, and 1000  $\mu\text{m}$  with a total length of 100 mm have been used. The tubes are used as received from the manufacturer (Vitro-Com). For all flow characterization the experimental apparatus consists of a microscope, a CCD camera with  $752 \times 480$  pixels, nylon tracer particles with average diameter of 15  $\mu\text{m}$ , and a computer with "home-made" software (Fig. 1).

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