# BINARY FLUID MIXTURE AND THERMOCAPILLARY EFFECTS ON THE WETTING CHARACTERISTICS OF A HEATED CURVED MENISCUS

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### **ABSTRACT**

An investigation of the thermocapillary effects on a heated, evaporating meniscus formed by binary fluid mixtures of wetting liquids in a vertical capillary pore system has been conducted. Experiments were conducted to primarily observe the wetting characteristics of the binary fluid mixture and how they are affected by the dynamics associated with the heating of and evaporation from a The results have demonstrated that interfacial thermocapillary stresses arising from liquid-vapor interfacial temperature gradients that degrade the ability of the liquid to wet the pore can be counteracted by introducing naturally occurring concentration gradients associated with distillation in binary fluid mixtures without affecting the heat transport capacity of the system.

# NOMENCLATURE

Volumetric concentration (%)

 $c_{p}$ Constant pressure specific heat (J/kg-K)

Acceleration due to gravity (m/s<sup>2</sup>) g

h Wicking height (m)

 $\overset{h}{\overset{fg}{K}}$ Latent heat of vaporization (J/kg)

Curvature (1/m) L Length (m) Molecular mass M

Real part of the index of refraction n

Pressure (Pa) P

Radius of the capillary tube (m) r Radius of the condenser (m)

R

T Temperature (°C)

Average liquid velocity (m/s) V Cartesian coordinate (m) X

Slope of surface tension (N/m-K) γ

Absolute viscosity (Pa-s) μ θ Contact angle (degrees)

Density (kg/m<sup>3</sup>) ρ

Surface Tension (N/m) σ

# **Subscripts**

Concentration C D Decane flow Flow Liquid 1 o Reference P Pentane Reservoir

Saturation sat Solid-liquid sl Solid-vapor Thermocapillary TC

Vapor

#### INTRODUCTION

Heat transport devices capable of dissipating high intensity heat energy as high as 200 W/cm<sup>2</sup> are required for cooling electronics; hypersonic and re-entry vehicles; satellites; propulsion and thermal energy recovery systems; cryoprobes; permafrost stabilizers; and roadway deicers among others. Of the heat transport devices presently under consideration in this regime, most utilize the latent heat of vaporization via liquid-vapor phase change. Relevant to the present research are passive capillary driven phase change devices (Chang and Hager, 1990). In these devices, the phase change occurs in a liquidsaturated porous or grooved media and capillary forces provide the driving potential for the liquid flow from the condenser to the evaporator. Ultimately for low temperature devices, the rate at which the condenser can resupply liquid to the evaporator limits the heat transport. In practice, however, this capillary heat transport limitation is rarely achieved (Pratt et al., 1998). One possible explanation is that the theory over-predicts the wetting characteristics since they are based on a 'maximum capillary potential' which presumes that the liquid within the porous structure is perfectly wetting and static conditions exist at the evaporating menisci. Dynamic effects, other than those due to viscous flow losses, are not considered.

The speculation here is that the dynamics associated with fluid motion and heat transport in the vicinity of the evaporating meniscus can detrimentally affect the driving capillary potential by degrading the wetting ability of the working fluid (Ma et al. 1998, Pratt and Hallinan 1997). The change in wettability is a result of a nonisothermal liquid-vapor interfacial temperatures near the contact line arising from both non-uniform substrate wall temperatures and nonuniform evaporation. Either or both of these influences yield surface tension gradients on the liquid-vapor interface. These surface tension gradients result in thermocapillary stresses acting near the contact line which can degrade the wettability of the liquid as has been seen by Ehrhard and Davis, 1991; Hocking, 1995; Sen and Davis, 1982, and Anderson and Davis, 1994 among others. Ehrhard and Davis (1991) showed that the spreading of a drop on a surface in the direction of increased wall temperature is retarded relative to the spreading of a similar drop on an isothermal surface. Furthering this work, Hocking (1995) showed that the advancement or spreading of an evaporating drop is retarded due to the evaporation process. Sen and Davis (1982) showed that, for a slot configuration, surface tension gradients create a fluid surface flow field which also affects the liquid wettability. Anderson and Davis (1991) analytically demonstrated that the flow field was coupled to the temperature field through the thermocapillarity as discussed by Sen and Davis (1982).

Recent studies of rewetting of liquids along inclined heated plates by Ha and Peterson (1994) and Chan and Zhang (1994) showed that the maximum wicking height measured was beneath that predicted using the typical Laplace-Young equation by as much as thirty percent. Given the observations of Ehrhard and Davis (1991), Hocking (1995), Sen and Davis (1982), and Anderson and Davis (1994), Pratt and Hallinan (1997) established and experimentally verified the relationship between the liquid-vapor interfacial temperature gradient and the wetting characteristics of a liquid within small pores. They showed that thermocapillary stresses acting near the contact line of the advancing liquid front inhibit the wetting of the liquid thereby reducing the wicking height.

The degradation predicted leads to a reduction in the capillary pumping potential in capillary heat transfer devices and thus a reduction in their ability to transport energy (Pratt et al. 1998). Thus the question arises as to how to minimize this reduction. The degradation arises from thermocapillary stresses along the liquid-vapor interface due to the reduction in surface tension with an increase in temperature. These stresses must be minimized. One possible solution is the introduction of a small amount of a relatively high surface-free-energy fluid into the working fluid. This would result in an increase in the concentration of the less volatile fluid with increase in temperature or from the evaporation for the lower surface-free-energy fluid. This would thus result in a concentration gradient of the more volatile working fluid in the opposite direction to the temperature gradients previously detailed (see Figure 1).

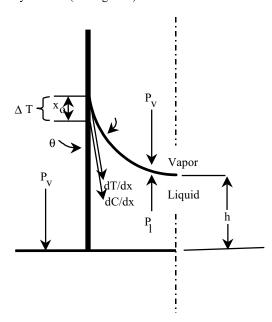


Figure 1 - Interface description

# **EXPERIMENTAL**

The experiment was designed so that macroscopic wetting characteristics could be observed for a heated and evaporating meniscus within a capillary pumped loop system. Specifically, it was

designed to determine the effects of binary fluid mixtures on the thermocapillary stresses arising near the contact line of evaporating menisci within capillary pores. To accomplish this, a single pore capillary pumped heat transfer device was constructed as shown in Fig. 2.

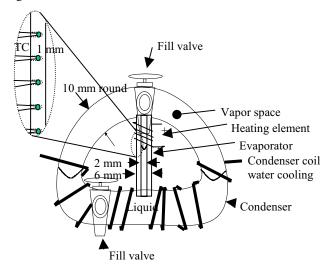


Figure 2 Schematic of a single pore capillary pumped loop

The set-up shown is a closed, single pore evaporator capillary pumped loop. It consists of a single capillary pore evaporator with large pore condensers and vapor channels. Heat is introduced via electrical resistance heating element mounted on the outer diameter of the tube. All lengths were measured using a cathetometer that has an associated bias limit of  $\pm~0.0005$  cm. The inner and outer diameter measurements have an error of  $\pm~0.00005$  mm and  $\pm~0.05$  mm, respectively.

The single tube capillary pumped heat transfer device shown in Fig. 2 was placed within a vacuum chamber that was used to minimize convection heat losses. The chamber was equipped with feedthroughs for thermocouples, pressure transducers, heater power connections, and cooling fluid lines. A roughing vacuum pump was used to evacuate the system. The system was then filled to the appropriate level with the working fluid. The working fluid consisted of a binary fluid mixture of n-pentane and decane. The properties for which are presented in Table 1. Concentrations of 0, 3, 5 and 10 percent by volume of decane were examined. To control the temperature of the condenser region, a refrigerated circulator was used to pump cooling fluid through a heat exchanger placed on the exterior of the condenser region.

Table 1 Properties of pentane and decane at 20°C

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Pentane (C <sub>5</sub> H <sub>12</sub> )	Decane (C <sub>10</sub> H <sub>22</sub> )	
Working fluid	Additive	
M = 72.15	M = 142.28	

$\sigma$ = 0.0155 N/m	$\sigma$ = 0.0234 N/m	
n = 1.3545	n = 1.410	
$T_{BOIL} = 36^{\circ}C$	T <sub>BOIL</sub> = 74°C	
ρ= 621.4 kg/m <sup>3</sup>	$\rho = 726.4 \text{ kg/m}^3$	

The capillary tube was heated using a fine nichrome heater wire (spirally wrapped around the tube at the top) having a resistance of 1  $\Omega$ /cm. It was connected to a dc power supply capable of producing up to three amps at thirty volts. A shunt resistor connected in series with the heating element allowed for the current flowing through the heater to be determined by measuring the voltage drop across it.

Calibrated thermocouples were used to measure the wall temperature of the capillary tube and to measure the temperature of the vapor space and liquid reservoir. To determine the temperature distribution of the evaporator region, thermocouples were positioned at 0.5 mm intervals along the capillary pore. This was accomplished by creating two rows of thermocouples 180 degrees apart. Each row had thermocouples placed longitudinally every millimeter and the two rows has a longitudinally offset of 0.5 mm. All automated data were recorded using an A/D board interfaced to a Pentuim III - 500 PC. The associated error in the temperature readings were  $\pm$  0.3°C with a 95% confidence interval.

The refrigerated circulator temperature was used to set the liquid reservoir temperature at 5, 15, and 25°C. Tests were run for each of these system states for variable heat input. The test conditions and power inputs for the tests are summarized in Table 2. The data presented in this table includes the bulk liquid reservoir temperature and the range of the power input considered.

Table 2 Experiment conditions

Concentration	Condenser	Heat input (W)
(% Decane)	temperature (°C)	
0	5	0-1.4
0	15	0-1.4
0	25	0-1.4
3	5	0-2.7
3	15	0-2.6
3	25	0-3
5	5	0-2.1
5	15	0-2.4
5	25	0-2.4
10	5	0-1.8
10	15	0-3
10	25	0-3

Additional tests were conducted to obtain estimates of the radiation and convection losses from the heated tubes and the variations of the wetting characteristics due to bulk liquid reservoir temperature changes.

# **ANALYSIS**

Analysis is presented to qualitatively explain the experimental results. The most easily measurable macroscopic wetting characteristic of a liquid in a pore is the wicking height. For static

interfacial conditions, it can be predicted with the Laplace-Young equation.

$$P_{v} - P_{l} = \sigma K \tag{1}$$

The reduced liquid pressure at the meniscus causes the liquid to wick to a height h, which for an axisymetric pore of radius r and a condenser of radius R (as shown in Fig. 2) gives rise to the following expression

$$\rho gh = \frac{2\sigma(R-r)}{rR}\cos\theta \tag{2}$$

where  $\theta$  is the apparent contact angle and  $\sigma$  is the surface tension of the mixture. When the meniscus is heated via wall heating, dynamic effects can alter this wicking height. One effect of heating is to produce a liquid-vapor interfacial temperature gradient near the contact line. This temperature gradient gives rise to a thermocapillary stress  $(\sigma_{TC})$  emanating from the contact line directed towards the intrinsic meniscus if the contact line region is hotter than the remainder of the meniscus. Additionally, the evaporative transport from the meniscus due to the heat transfer induces liquid flow from the reservoir. Associated with this flow are viscous losses along the wall of the pore. The use of a binary fluid mixture gives rise to an additional stress ( $\sigma_C$ ) along the interface resulting from the naturally occurring concentration which emanates from the contact line region in the opposite direction from the thermocapillary stress. While, both the thermocapillary and the flow loss effects can reduce the wicking height, the concentration effect acts to negate these effects.

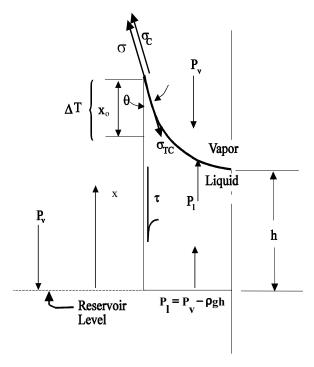


Figure 3 Meniscus within a capillary tube with an induced temperature gradient and a concentration gradient of a binary fluid

A vertical force balance is applied to the control volume defined by the liquid column in the pore shown in Fig. 3 to determine the thermocapillary and flow loss effects on the capillary potential. This results in the following equilibrium condition.

$$(P_l - P_v)\pi r^2 + (\sigma - \sigma_{TC} + \sigma_C)2\pi r \cos\theta - \Delta P_{flow}\pi r^2 - \sigma 2\pi R \cos\theta = 0$$
(3)

with  $P_l - P_v = -\rho gh$ . Dividing by  $\pi r^2$  and substituting yields

$$\rho g h = \frac{2(\sigma - \sigma_{TC} + \sigma_C)}{r} \cos \theta - \frac{2\sigma R}{r^2} \cos \theta - \Delta P_{flow}$$
 (4)

Where 
$$\frac{\Delta P_{flow}}{L_{flow}} = \frac{16}{r \text{ Re}} \rho \langle V \rangle^2$$
 and  $\text{Re} = 2r \langle V \rangle \rho / \mu$  and  $L_{flow}$  is

the flow length from the bottom of the capillary tube to the meniscus

or 
$$\Delta P_{flow} = \frac{8\mu L_{flow}}{\rho \pi r^4} \frac{Q_M}{h_{fg} + c_p (T_{sat} - T_r)}$$
.

This modified version of the capillary pumping potential incorporates yet undefined thermocapillary and concentration forces that can be determined by examining the effects of liquid-vapor interfacial temperature gradients on the surface tension.

$$\sigma = C\sigma_{oD} - C\gamma_D T + (1 - C)\sigma_{oP} - (1 - C)\gamma_P T$$
(5)

So that

$$\sigma_C - \sigma_{TC} = \frac{\partial \sigma}{\partial x} x_o \tag{6}$$

and

$$\frac{\partial \sigma}{\partial x} = \frac{\partial C}{\partial x} \left( \sigma_{oD} - \gamma_D T - \sigma_{oP} + \gamma_P T \right) - \frac{\partial T}{\partial x} \left[ C \gamma_D + \left( 1 - C \right) \gamma_P \right] \tag{7}$$

Finally expression for the thermocapillary and concentration forces are obtainable from Eq. 7 or

$$\sigma_C = \frac{\partial C}{\partial x} \left( \sigma_{oD} - \gamma_D T - \sigma_{oP} + \gamma_P T \right) x_o \tag{8}$$

and

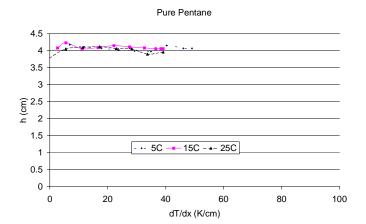
$$\sigma_{TC} = \frac{\partial T}{\partial x} \left[ C \gamma_D + (1 - C) \gamma_P \right] x_o \tag{9}$$

The thermocapillary stress is of the same form as that which was shown to exist by Pratt and Hallinan (1997). To determine  $\sigma_C$  the concentration gradient along the meniscus  $\frac{\partial C}{\partial x}$  must be measured or estimated. The measurement of the concentration that should be achieved thermally and non-intrusively is left for future work.

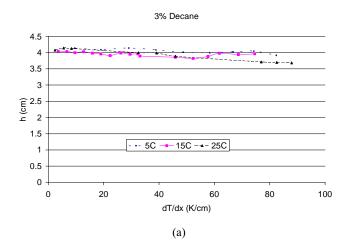
# **RESULTS**

For full examination of the experimentally determined wetting characteristics, data is presented for the conditions detailed in Table 2.

This presented data includes the steady-state wicking height versus wall temperature gradient for concentrations of 0, 3, 5, and 10% decane in pentane. Tests were conducted until the temperature nearest



to the heating element exceeded 110°C or for the pure pentane case, the system became unstable. The x-axis on all plots is set to the same scale to assist in comparisons. Total errors associated with these plots are 2.7% with a 95% confidence interval for the wicking height and 8.5% at a 95% confidence interval for the temperature gradient. Plots depicting the heat transfer are also presented.



(b)

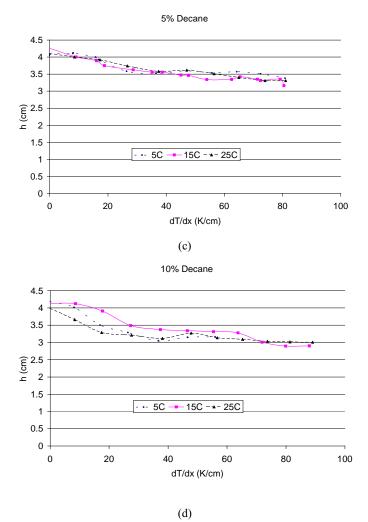
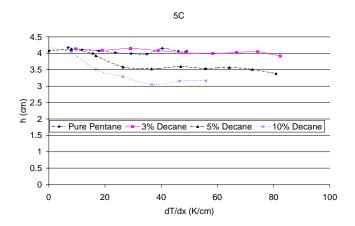


Figure 4 Wicking height versus wall temperature gradient (a) Pure pentane, (b) 3% decane in pentane, (c) 5% decane in pentane and (d) 10% decane in pentane

Figure 4 is a presentation of wicking height versus wall temperature gradient for pure pentane, 3, 5 and 10% decane in pentane. It shows that there is little variation in wicking height with different subcooling conditions of the condenser for all the cases examined.



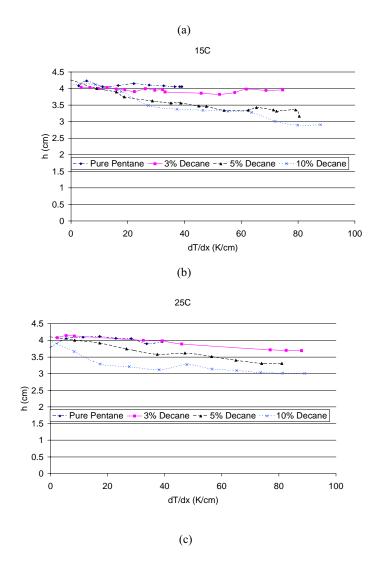
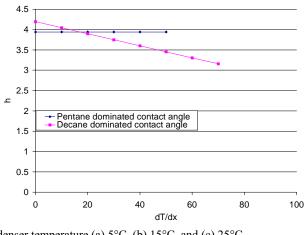


Figure 5 Wicking height versus wall temperature gradient for variable



condenser temperature (a) 5°C, (b) 15°C, and (c) 25°C

In Fig. 5 the data for wicking height versus wall temperature gradient for variable condenser temperature is presented. This figure shows that for low concentrations of decane (3%) the wicking height is not deleteriously affected. However for high concentrations of decane (5 and 10%) the wicking height is drastically reduced. This is a result of the distillation process in that near the contact line, decane is the primary component and thus dominates the wetting characteristic or contact angle. To understand the significance of this assumption, examination of the contact angle is required. Figure 6 is a plot of contact angle versus liquid temperature for pure pentane and decane.

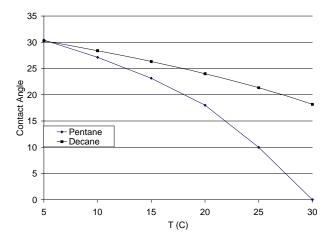


Figure 6 Contact angle versus liquid temperature for pure pentane and pure decane

Figure 6 is semi-empirical in nature. The values presented were obtained by measuring the wicking height of a single pore placed within a large liquid reservoir held at a known temperature and calculating the contact angle using

$$\theta = a\cos\frac{\rho ghr}{2\sigma} \tag{10}$$

Then the Young-Dupre' equation (Eq. (11)) was applied to determine the variation in contact angle with temperature by assuming that the numerator was approximately constant with temperature and allowing the denominator to vary.

$$\cos\theta = \frac{\sigma_{sv} - \sigma_{sl}}{\sigma_{lv}(T)} \quad \text{and} \quad \sigma_{lv} = \sigma_o - \gamma T \tag{11}$$

The contact angle data produced was then used in Eq. 2 to examine the variation in wicking height due solely to bulk liquid temperature variations for pure pentane and a decane in pentane mixture. For pure pentane, the contact angle calculated for pentane was used and for the mixture, the contact angle measured for decane was used. The results of which are presented in Fig. 7.

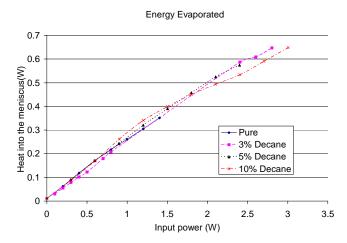


Figure 7 Wicking height versus wall temperature gradient

The wicking height data presented in Fig. 7 was obtained by examining variations in bulk temperature that result in variations in contact angle and surface tension. For the decane dominated case, it was assumed that decane controlled the contact angle, that is, the contact angle used was for pure decane. Figure 7 shows similar variations in wicking height as those seen during testing. Thus the assumption that for high decane concentration mixtures, the decane controls the contact angle seems to be substantiated. This also supports the model developed and presented in Eqs. 7 to 9 because if the wicking height variation is due to changes in contact angle, no net interfacial stress exists. This is apparent if the model is examined. It shows that the thermocapillary stress is balanced by the stress arising from the concentration gradient along the liquid-vapor interface. This balance would yield no net interfacial stress.

Finally, consideration must be made as to how if at all the addition of decane to the working fluid affects heat transfer. To do this two variable must be examined. The first is the amount of energy being evaporated at the meniscus and the second is the temperature of the mensicus. Figure 8 depicts the first of these two parameters for a  $25^{\circ}\mathrm{C}$  condenser temperature.

Figure 8 Heat transferred into the meniscus versus total heat input

Figure 8 is a plot of the total power input into the system via the electric heater versus that which is transferred into the meniscus for the pure pentane case and the 3, 5 and 10% decane in pentane cases. The heat transferred into the meniscus is calculated by applying an energy balance at the meniscus location as shown in Fig. 9. Figure 8 shows that the addition of decane into the pentane has no effect on the energy transferred into the meniscus.

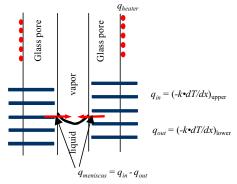


Figure 9 Energy balance at the meniscus

To further substantiate this conclusion examination of the meniscus temperature is necessary. Figure 10 is a presentation of the temperature of the thermocouple closes to the intrinsic meniscus for the two limiting cases that of pure pentane and 10% decane in pentane.

Figure 10 Temperature at the meniscus versus input power - condenser

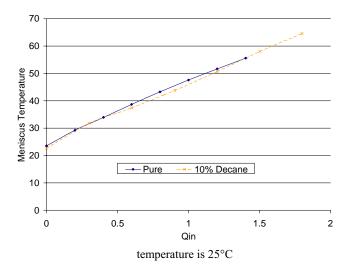


Figure 10 shows that there is no significant variation in the temperature at the meniscus for the two limiting cases. This in conjunction with Fig. 8 demonstrates that the heat transfer characteristics of the system are not noticeably depreciated due to the addition of decane.

# **CONCLUSIONS**

Analysis describing a novel method of negating the deleterious effects of thermocapillary stress on the capillary driven phase change devices has been presented and preliminary experiments have been completed showing its validity. The data also revealed that an "optimum" concentration of decane in pentane existed for which no degradation in wicking height or heat transfer existed. This is a result of non-complete distillation of the pentane in the near contact line region. However, for high concentrations of decane in pentane, substantial reductions in wicking height were observed due to the higher surface-free-energy decane dominating the contact angle characteristics. Also, the addition of decane prolongs instability onset compared to the pure pentane case.

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