9. Marangoni Flows

Marangoni flows are those driven by surface gradients. In general, surface tension σ depends on both the temperature and chemical composition at the interface; consequently, Marangoni flows may be generated by gradients in either temperature or chemical composition at an interface. We previously derived the tangential stress balance at a free surface:

$$\boldsymbol{n} \cdot \boldsymbol{T} \cdot \boldsymbol{t} = -\boldsymbol{t} \cdot \boldsymbol{\nabla} \boldsymbol{\sigma} \quad , \tag{9.1}$$

where \boldsymbol{n} is the unit outward normal to the surface, and \boldsymbol{t} is any unit tangent vector. The tangential component of the hydrodynamic stress at the surface must balance the tangential stress associated with gradients in σ . Such Marangoni stresses may result from gradients in temperature or chemical composition at the interface. For a static system, since $\boldsymbol{n} \cdot \boldsymbol{T} \cdot \boldsymbol{t} = 0$, the tangential stress balance equation indicates that: $0 = \boldsymbol{\nabla} \sigma$. This leads us to the following important conclusion:

There cannot be a static system in the presence of surface tension gradients.

While pressure jumps can arise in static systems characterized by a normal stress jump across a fluid interface, they do not contribute to the tangential stress jump. Consequently, tangential surface stresses can only be balanced by viscous stresses associated with fluid motion.

Thermocapillary flows: Marangoni flows induced by temperature gradients $\sigma(T)$.

Note that in general $\frac{d\sigma}{dT} < 0$ Why? A warmer gas phase has more liquid molecules, so the creation of surface is less energetically unfavourable; therefore, σ is lower.

Approach Through the interfacial BCs (and $\sigma(T)$'s appearance therein), N-S equations must be coupled



Figure 9.1: Surface tension of a gas-liquid interface decreases with temperature since a warmer gas phase contains more suspended liquid molecules. The energetic penalty of a liquid molecule moving to the interface is thus decreased.

to the heat equation

$$\frac{\partial T}{\partial t} + \boldsymbol{u} \cdot \boldsymbol{\nabla} T = \kappa \nabla^2 T \tag{9.2}$$

Note:

- 1. the heat equation must be solved subject to appropriate BCs at the free surface. Doing so can be complicated, especially if the fluid is evaporating.
- 2. Analysis may be simplified when the **Peclet number** $\mathcal{P}e = \frac{Ua}{\kappa} \ll 1$. Nondimensionalize (9.2): $\boldsymbol{x} = a\boldsymbol{x}', t = \frac{a}{U}t', \boldsymbol{u} = U\boldsymbol{u}'$ to get

$$\mathcal{P}e\left(\frac{\partial T'}{\partial t'} + \boldsymbol{u'} \cdot \boldsymbol{\nabla'}T'\right) = \nabla^2 T'$$
(9.3)

Note:

 $\begin{aligned} \mathcal{P}e &= \mathcal{R}e \cdot \mathcal{P}r = \frac{Ua}{\nu} \cdot \frac{\nu}{\kappa} \ll 1 \text{ if } \mathcal{R}e \ll 1 \text{, so one has } \nabla^2 T = 0. \end{aligned} \\ \text{The Prandtl number } \mathcal{P}r = O(1) \text{ for many common (e.g. aqeous) fluids.} \\ \textbf{E.g.1 Thermocapillary flow in a slot (Fig.9.2a)} \\ \text{Surface Tangential BCs } \tau = \frac{\Delta\sigma}{L} = \frac{d\sigma}{dT} \frac{\Delta T}{L} \approx \mu \frac{U}{H} \text{ viscous stress } U \sim \frac{1}{\mu} \frac{H}{L} \Delta \sigma. \end{aligned}$



Figure 9.2: a) Thermocapillary flow in a slot b) Thermal convection in a plane layer c) Thermocapillary drop motion.

E.g.2 Thermocapillary Drop Motion (Young, Goldstein & Block 1962)

can trap bubbles in gravitational field via thermocapillary forces. (Fig.9.2c).

E.g.3 Thermal Marangoni Convection in a Plane Layer (Fig.9.2b).

Consider a horizontal fluid layer heated from below. Such a layer may be subject to either buoyancy- or Marangoni-induced convection.

Recall: Thermal buoyancy-driven convection (Rayleigh-Bernard) $\rho(T) = \rho_0 (1 + \alpha(T - T_0))$, where α is the thermal expansivity. Consider a buoyant blob of characteristic scale d. Near the onset of convection, one expects it to rise with a Stokes velocity $U \sim \frac{g\Delta\rho}{\rho} \frac{d^2}{\nu} = \frac{g\alpha\Delta T d^2}{\nu}$. The blob will rise, and so convection will occur, provided its rise time $\tau_{rise} = \frac{d}{U} = \frac{d\nu}{g\alpha\Delta T d^2}$ is less than the time required for it to lose its heat and buoyancy by diffusion, $\tau_{diff} = \frac{d^2}{\kappa}$.

Criterion for Instability: $\frac{\tau_{diff}}{\tau_{rise}} \sim \frac{g \alpha \Delta T d^3}{\kappa \nu} \equiv \mathcal{R}a > \mathcal{R}a_c \sim 10^3$, where $\mathcal{R}a$ is the Rayleigh number.

Note: for $\mathcal{R}a < \mathcal{R}a_c$, heat is transported solely through diffusion, so the layer remains static. For $\mathcal{R}a > \mathcal{R}a_c$, convection arises.

The subsequent behaviour depends on $\mathcal{R}a$ and $\mathcal{P}r$. Generally, as $\mathcal{R}a$ increases, steady convection rolls \Rightarrow time-dependency \Rightarrow chaos \Rightarrow turbulence.

Thermal Marangoni Convection

Arises because of the dependence of σ on temperature: $\sigma(T) = \sigma_0 - \Gamma(T - T_0)$ Mechanism:

- Imagine a warm spot on surface \Rightarrow prompts surface divergence \Rightarrow upwelling.
- Upwelling blob is warm, which reinforces the perturbation provided it rises before losing its heat via diffusion.
- Balance Marangoni and viscous stress: $\frac{\Delta\sigma}{d}\sim\frac{\mu U}{d}$
- Rise time: $\frac{d}{U} \sim \frac{\mu d}{\Delta \sigma}$
- Diffusion time $\tau_{diff} = \frac{d^2}{\kappa}$

Criterion for instability: $\frac{\tau_{diff}}{\tau_{rise}} \sim \frac{\Gamma \Delta T d}{\mu \kappa} \equiv \mathcal{M}a > \mathcal{M}a_c$, where $\mathcal{M}a$ is the Marangoni number. Note:

- 1. Since $\mathcal{M}a \sim d$ and $\mathcal{R}a \sim d^3$, thin layers are most unstable to Marangoni convection.
- 2. Bénard's original experiments performed in millimetric layers of spermaceti were visualizing Marangoni convection, but were misinterpreted by Rayleigh as being due to buoyancy \Rightarrow not recognized until Block (Nature 1956).
- 3. Pearson (1958) performed stability analysis with flat surface \Rightarrow deduced $\mathcal{M}a_c = 80$.
- 4. Scriven & Sternling (1964) considered a deformable interface, which renders the system unstable at all $\mathcal{M}a$. Downwelling beneath peaks in Marangoni convection, upwelling between peaks in Rayleigh-Bénard convection (Fig. 9.3a).
- 5. Smith (1966) showed that the destabilizing influence of the surface may be mitigated by gravity. Stability Criterion: $\frac{d\sigma}{dT}\frac{dT}{dz} < \frac{2}{3}\rho gd \Rightarrow$ thin layers prone to instability.

E.g.4 Marangoni Shear Layer (Fig. 9.3)

Lateral $\nabla \theta$ leads to Marangoni stress \Rightarrow shear flow. The resulting T(x, y) may destabilize the layer to



Figure 9.3: **a)** Marangoni convection in a shear layer may lead to transverse surface waves or streamwise rolls (**c**). Surface deflection may accompany both instabilities (**b**,**d**).

 ${\it Marangoni\ convection.}$

Smith & Davis (1983ab) considered the case of flat free surface. System behaviour depends on $\mathcal{P}r = \nu/\kappa$. Low $\mathcal{P}r$: Hydrothermal waves propagate in direction of τ .

High $\mathcal{P}r$: Streamwise vortices (Fig. 9.3c).

Hosoi & Bush (2001) considered a deformable free surface (Fig. 9.3d)

E.g.5 Evaporatively-driven convection

e.g. for an alcohol- H_2O solution, evaporation affects both the alcohol concentration c and temperature θ . The density $\rho(c, \theta)$ and surface tension $\sigma(c, \theta)$ are such that $\frac{\partial \rho}{\partial \theta} < 0$, $\frac{\partial \rho}{\partial c} < 0$, $\frac{d\sigma}{d\theta} < 0$, $\frac{d\sigma}{dc} < 0$. Evaporation results in surface cooling and so may generate either Rayleigh-Bénard or Marangoni thermal convection. Since it also induces a change in surface chemistry, it may likewise generate either Ra - B or Marangoni chemical convection.

E.g.6 Coffee Drop

Marangoni flows are responsible for the ring-like stain left by a coffee drop.

- coffee grounds stick to the surface
- evaporation leads to surface cooling, which is most pronounced near the edge, where surface area per volume ratio is highest
- resulting thermal Marangoni stresses drive radial outflow on surface \Rightarrow radial ring



Figure 9.4: Evaporation of water from a coffee drop drives a Marangoni flow.

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