

**ChE 385M Surface Phenomena
University of Texas at Austin**

**Marangoni-Driven Finger Formation
at a Two Fluid Interface**

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Introduction

Marangoni phenomena are driven by gradients in surface tension on the interface of a fluid. The gradients in surface tension can arise from an uneven distribution of surfactant molecules on the surface of the fluid or from temperature gradients along the interface. Gradients in surface tension induce the flow of fluid on the interface from areas of low surface tension to areas of high surface tension, or equivalently, fluid will flow from areas of high temperature to areas of low temperature on the interface. Marangoni phenomena are believed to be the cause of the formation of finger-like structures during some interfacial polymerizations. The surface tension gradients in the interfacial polymerization system arise from the temperature gradient induced by the heat of reaction. Perturbation of the interface can lead to an instability causing the finger-like formation that is observed. This report presents the results of a linear stability analysis of the formation of “fingers” on a simplified two-fluid system.

Model

The model system chosen for this investigation consists of two immiscible fluids of infinite depth with uniform pressure throughout both fluids. The fluids are assumed to be incompressible fluids with constant physical properties. A linear temperature gradient is assumed across both fluids and it is also assumed that the surface tension between the fluids varies linearly with temperature. The linear stability analysis is performed by assuming a disturbance on the interface and determining the growth or decay of the disturbance. A schematic of the disturbed interface and the expected evolution of an unstable interface is shown in Figure 1. The initial perturbation is represented as a sinusoidal wave. The line extending through both fluids represents the temperature gradient across the fluids. As the wave propagates through the fluids, the crest of the wave will be at a higher temperature than the trough of the wave. The temperature difference will result in a surface tension gradient causing fluid from the crest of the wave to flow to the trough of the wave. The flow of fluid into the trough of the wave can lead to instabilities of the form labeled “fingers” in Figure 1. However, if the surface tension forces arising from the curvature of the interface are dominant, the disturbance will decay.

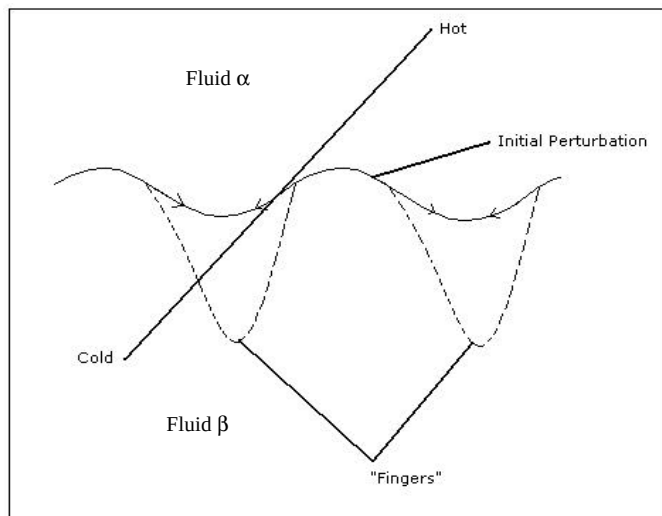


Figure 1. Schematic Representation of Model System

Linear stability analysis

As mentioned earlier, a linear stability analysis consists of assuming a disturbance on the interface and determining how the disturbance evolves. The evolution of the disturbance is determined by applying the equations of fluid mechanics and specifying all boundary conditions. Any terms that are nonlinear in the disturbance are ignored. All the details of the linear stability analysis can be found in the Appendix. In the following analysis the coordinate system is defined with the origin at the interface of the two fluids. The positive y direction is the direction pointing into fluid α .

The disturbance on the interface is assumed to be small and take the following form.

$$h = \hat{h} \exp(St + ikx) \quad (1)$$

where $\hat{h}$ is the amplitude of the disturbance, S is the frequency, and k is the wave number. Equation (1) represents a sinusoidal disturbance on the interface of the fluid.

The evolution of the interface (kinematic condition) is determined by taking the total time derivative of the surface defined by the interface. This leads to an equation relating the time evolution of the interface to the velocity of the fluid evaluated on the interface.

$$u_y \Big|_{y=h} = \frac{\partial h}{\partial t} \quad (2)$$

Where u_y is the y -component of the fluid velocity evaluated on the interface. So in order to determine the evolution of the disturbance it is necessary to determine the velocity of the fluid on the interface. This is accomplished by employing the equations of fluid mechanics. The governing fluid mechanics equations are the continuity equation and the equations of motion. The assumptions stated earlier lead to the following linearized forms of the continuity equation and equations of motion for each fluid.

$$\nabla \bullet \underline{u}_a = \nabla \bullet \underline{u}_b = 0 \quad (3)$$

$$r_a \frac{\partial \underline{u}_a}{\partial t} = -\nabla P_a + m_a \nabla^2 \underline{u}_a \quad (4)$$

$$r_b \frac{\partial \underline{u}_b}{\partial t} = -\nabla P_b + m_b \nabla^2 \underline{u}_b \quad (5)$$

where $\underline{u}_j$ is the velocity vector of fluid j , m_j is the viscosity of fluid j , r_j is the density of fluid j , and P_j is the pressure in fluid j . All the variables in equations (3) through (5) represent disturbances away from the initially quiescent state.

Solutions of the following form are assumed for the velocity and pressure ($j=\alpha,\beta$).

$$\underline{u}_j = \underline{\hat{u}}_j(y) \exp(St + ikx) \quad (6)$$

$$P_j = \hat{P}_j(y) \exp(St + ikx) \quad (7)$$

where $\underline{\hat{u}}_j(y)$ and $\hat{P}_j(y)$ are the amplitudes of the velocity and pressure disturbance in fluid j respectively.

Substituting equations (6) and (7) into the continuity equation and the equations of motion lead to the following differential equations.

$$\hat{u}_{x,j} = -\frac{1}{ik} \frac{d\hat{u}_{y,j}}{dy} \quad (8)$$

$$m_j \frac{d^2 \hat{u}_{x,j}}{dy^2} - \hat{u}_{x,j} (r_j S + m_j k^2) = ik \hat{P}_j' \quad (9)$$

$$m_j \frac{d^2 \hat{u}_{y,j}}{dy^2} - \hat{u}_{y,j} (r_j S + m_j k^2) = \frac{d\hat{P}_j}{dy} \quad (10)$$

where $\hat{u}_{x,j}$ is the x-component of the velocity disturbance amplitude in fluid j , and $\hat{u}_{y,j}$ is the y-component of the velocity disturbance amplitude in fluid j .

Equation (8) can be used to eliminate u_x in equations (9) and (10). Then equations (9) and (10) can be combined to eliminate the pressure. The resulting equation is a fourth order linear ordinary differential equation with constant coefficients.

$$m_j \frac{d^4 \hat{u}_{y,j}}{dy^4} - (r_j S + 2m_j k^2) \frac{d^2 \hat{u}_{y,j}}{dy^2} + \hat{u}_{y,j} (r_j S + m_j k^2) = 0 \quad (11)$$

The solutions to equation (11) for both fluids are of the form

$$\hat{u}_{y,b} = C1 \exp(ky) + C2 \exp(-ky) + C3 \exp\left(\left(\frac{r_b S + m_b k^2}{m_b}\right)^{\frac{1}{2}} y\right) + C4 \exp\left(-\left(\frac{r_b S + m_b k^2}{m_b}\right)^{\frac{1}{2}} y\right) \quad (12)$$

$$\hat{u}_{y,a} = A1 \exp(ky) + A2 \exp(-ky) + A3 \exp\left(\left(\frac{r_a S + m_a k^2}{m_a}\right)^{\frac{1}{2}} y\right) + A4 \exp\left(-\left(\frac{r_a S + m_a k^2}{m_a}\right)^{\frac{1}{2}} y\right) \quad (13)$$

Where $C1$, $C2$, $C3$, $C4$, $A1$, $A2$, $A3$, and $A4$ are constants to be determined with boundary conditions. Since the velocities are bound at infinity $C2$, $C4$, $A1$, and $A3$ can be eliminated immediately. The remaining constants are determined from the equality of the velocity of both fluids on the interface, the pressure differential across the interface resulting from the curvature of the interface, and from the tangential stress balance on the interface. All the boundary conditions can be evaluated at $y=0$ for a small disturbance.

The equality of the velocities of the two fluids on the interface can be expressed as the equality of the components of the velocity on the interface. This boundary condition reduces to the following two equations.

$$\hat{u}_{x,b} = \hat{u}_{x,a}, \quad y=0 \quad (14)$$

$$\hat{u}_{y,b} = \hat{u}_{y,a}, \quad y=0 \quad (15)$$

The assumption of initially uniform pressure in the two fluids results in the pressure differential arising from the curvature of the interface only. This boundary condition is expressed by the Young-Laplace equation, which relates the pressure difference across the interface to the curvature of the interface. The Young-Laplace equation leads to the following linearized form of the pressure differential across the interface.

$$\hat{P}_b - \hat{P}_a = -g \frac{\partial^2 h}{\partial x^2}, \quad y=0 \quad (16)$$

The tangential stress balance relates the tangential stress on the interface to gradients in surface tension. After linearization, the tangential stress balance reduces to

$$(t_{xy}^a - t_{xy}^b) = -\frac{\partial g}{\partial x} = -\frac{\partial g}{\partial T} \frac{\partial T}{\partial x}, \quad y=0 \quad (17)$$

where t_{xy}^j is the xy -component of the stress tensor for fluid j .

$$t_{xy}^j = m_j \left(\frac{\partial u_{x,j}}{\partial y} + \frac{\partial u_{y,j}}{\partial x} \right) \quad (18)$$

Equations (14) through (17) are used to solve for the remaining unknown constants in equations (12) and (13). After evaluation of the constants, equations (2) and (6) are used to find a relationship for S as a function of k . This is the relationship that is needed to determine the wavelength for the fastest growing mode. The fastest growing mode is the growth mode that is observed in an experiment in which an instability occurs.

Results

The stability of the disturbance is determined by the value of S in equation (1). The approach taken to determining S was to use the four boundary conditions to determine the constants in equations (12) and (13). Then the kinematic condition was used to determine how S varied as a function of k . The kinematic condition resulted in a non-linear equation in S , which had to be solved numerically.

Two arbitrary fluids with the physical properties listed in Table 1 were used in the calculation of S . A reference surface tension of 25 dyne/cm at 25°C was used in the calculation.

Table 1 Physical Properties of Fluids Used in Calculations

Fluid	Density(g/cm ³)	Viscosity (cP)
α	1	80
β	1.3	100

Figure 2 is a plot of the calculated values of S versus k for a temperature gradient of 1°C/ft. Positive values of S will result in an unstable disturbance. Figure 2 indicates that instabilities will occur for long wavelength disturbances while short wavelength disturbances will decay. The point at which S crosses from positive to negative is called the cutoff wave number for the disturbance. Stated differently, any values of k less than the cutoff value will result in an unstable disturbance, while values of k greater than the cutoff will result in a decaying disturbance. The cutoff wave number for the system depicted in Figure 2 is 6.6 cm⁻¹. This cutoff wave number corresponds to a cutoff wavelength of 0.95 cm. The most dangerous mode or maximum value of S occurs at a wave number of 3.7cm⁻¹.

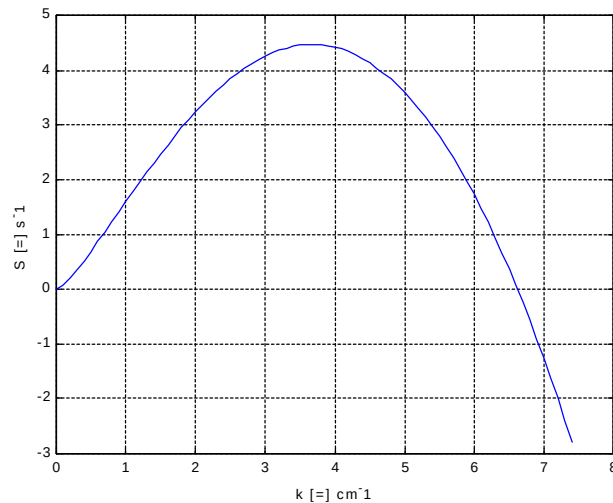


Figure 2 Calculated Plot of S versus k

The most dangerous mode has an inverse time constant of 4.47 s^{-1} . The most dangerous mode is the growth mode that is observed in an experiment in which an instability occurs. Therefore, the disturbance wavelength that would be observed in an experiment for this system would be approximately 1.7 cm. This is a length scale that could easily be achieved in a laboratory setting indicating that this mechanism is a possible explanation for the formation of “fingers”.

The effect of the magnitude of the temperature gradient was also investigated. The curve in Figure 2 is for a temperature gradient of $1^\circ\text{C}/\text{ft}$. Figure 3 is a plot of the S versus k for temperature gradients of $1^\circ\text{C}/\text{ft}$ and $10^\circ\text{C}/\text{ft}$. Although it is not apparent in Figure 3, there is a slight difference in the two curves. However, the difference for this temperature range is very small. This can be understood by recalling that the model was developed for a small disturbance and that the surface tension varied linearly with temperature. Temperature gradients of $1^\circ\text{C}/\text{ft}$ and $10^\circ\text{C}/\text{ft}$ are significantly small on the scale of the disturbance and therefore the change in the behavior of the disturbance shouldn't be expected to change drastically in this temperature range.

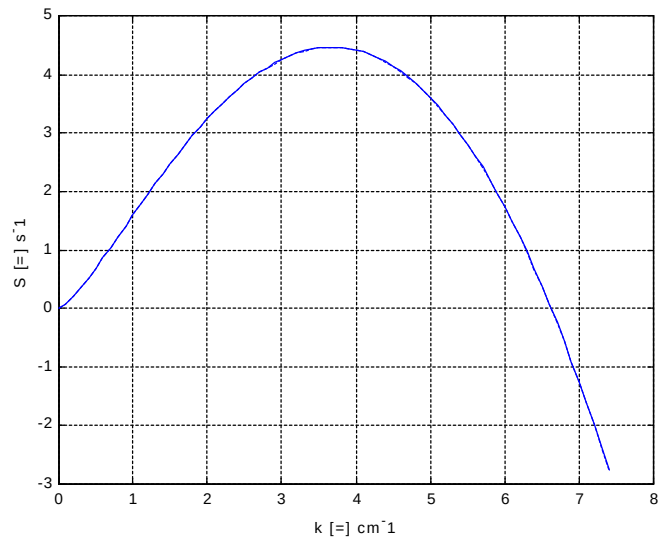


Figure 3 Calculated Plot of S versus k for temperature gradients of $1^\circ\text{C}/\text{ft}$ and $10^\circ\text{C}/\text{ft}$

Conclusion

The linear stability analysis outlined above indicates that marangoni phenomena could be playing a role in the formation of the fingers observed during some interfacial polymerizations. Although the system analyzed in this study is highly simplified, it does provide some insight into the physical phenomena responsible for the formation of fingers during polymerization. The results of the linear stability analysis indicate that long wavelength disturbances are responsible for the formation of the fingers by a marangoni mechanism.

References:

Bird, R.B., W.E. Stewart, and E.N. Lightfoot. *Transport Phenomena*. New York: John Wiley & Sons, 1960.

APPENDIX

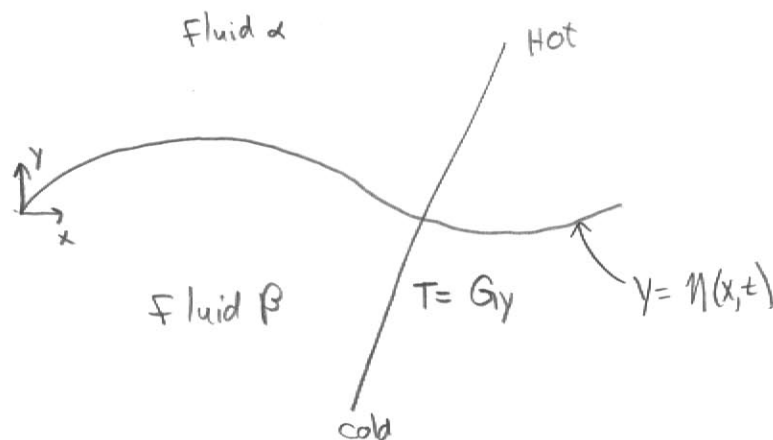
assumptions

constant physical properties
linear temperature gradient

γ linear dependence on T

$$\gamma = \gamma_0 \left(1 - \frac{T}{T_c} \right)$$

neglecting hydrostatics
(uniform pressure throughout)
(both fluids initially)



governing equations:

$$\nabla \cdot \underline{u}_\alpha = \nabla \cdot \underline{u}_\beta = 0 \quad \text{Continuity equation}$$

$$\left. \begin{aligned} \rho_\alpha \left(\frac{\partial \underline{u}_\alpha}{\partial t} + \underline{u}_\alpha \cdot \nabla \underline{u}_\alpha \right) &= -\nabla P_\alpha + \mu_\alpha \nabla^2 \underline{u}_\alpha \\ \rho_\beta \left(\frac{\partial \underline{u}_\beta}{\partial t} + \underline{u}_\beta \cdot \nabla \underline{u}_\beta \right) &= -\nabla P_\beta + \mu_\beta \nabla^2 \underline{u}_\beta \end{aligned} \right\} \text{Equations of motion}$$

linearizing equations of motion:

$$\rho_\alpha \frac{\partial \underline{u}_\alpha}{\partial t} = -\nabla P_\alpha + \mu_\alpha \nabla^2 \underline{u}_\alpha$$

$$\rho_\beta \frac{\partial \underline{u}_\beta}{\partial t} = -\nabla P_\beta + \mu_\beta \nabla^2 \underline{u}_\beta$$

assume disturbances of form:

$$\eta = \hat{\eta} \exp(st + i k x)$$

$$P = \hat{P}(y) \exp(st + i k x)$$

$$\underline{u} = \hat{\underline{u}}(y) \exp(st + i k x)$$

Continuity equation:

$$\frac{\partial u_x}{\partial x} + \frac{\partial u_y}{\partial y} = 0$$

substituting disturbance

$$i k \hat{u}_x \exp(st + i k x) + \frac{\partial \hat{u}_y}{\partial y} \exp(st + i k x) = 0 \Rightarrow \frac{\partial \hat{u}_y}{\partial y} + i k \hat{u}_x = 0$$

since $\hat{u}_y$ is only a function of y

$$\boxed{\frac{d\hat{u}_y}{dy} + i k \hat{u}_x = 0}$$

for both fluids
 $\alpha + \beta$

equations of motion:

$$\rho \frac{\partial u_x}{\partial t} = -\frac{\partial p}{\partial x} + \mu \left(\frac{\partial^2 u_x}{\partial x^2} + \frac{\partial^2 u_x}{\partial y^2} \right)$$

$$\rho \frac{\partial u_y}{\partial t} = -\frac{\partial p}{\partial y} + \mu \left(\frac{\partial^2 u_y}{\partial x^2} + \frac{\partial^2 u_y}{\partial y^2} \right)$$

substituting disturbance into the equations of motion + rearranging leads to the following two equations

$$\boxed{\begin{aligned} \mu \frac{d^2 \hat{u}_x}{dy^2} - \hat{u}_x(\rho s + \mu k^2) &= ik \hat{p} \quad \text{① for both fluids } \alpha + \beta \\ \mu \frac{d^2 \hat{u}_y}{dy^2} - \hat{u}_y(\rho s + \mu k^2) &= \frac{d\hat{p}}{dy} \quad \text{②} \end{aligned}}$$

Equations ① + ②, along with the continuity equation can be used to solve for $\hat{u}_y$.

step 1. $\frac{d}{dy}$ ① $\Rightarrow \mu \frac{d^3 \hat{u}_x}{dy^3} - \frac{d\hat{u}_x}{dy}(\rho s + \mu k^2) = ik \frac{d\hat{p}}{dy}$ ①a

step 2. $ik \cdot$ ② $\Rightarrow \mu ik \frac{d^2 \hat{u}_y}{dy^2} - \hat{u}_y ik(\rho s + \mu k^2) = ik \frac{d\hat{p}}{dy}$ ②a

step 3. ①a - ②a

$$\Rightarrow \mu \frac{d^3 \hat{u}_x}{dy^3} - \frac{d\hat{u}_x}{dy}(\rho s + \mu k^2) - \mu ik \frac{d^2 \hat{u}_y}{dy^2} + \hat{u}_y ik(\rho s + \mu k^2) = 0$$

step 4. Substitute continuity equation: $\hat{u}_x = -\frac{1}{ik} \frac{d\hat{u}_y}{dy}$

$$\boxed{\mu \frac{d^4 \hat{u}_y}{dy^4} - (\rho s + 2\mu k^2) \frac{d^2 \hat{u}_y}{dy^2} + \hat{u}_y k^2(\rho s + \mu k^2) = 0}$$

applies to both fluids

4th order linear ODE with constant coefficients.

Fluid B

$$\hat{u}_y = C_1 \exp(ky) + C_2 \exp(-ky) + C_3 \exp\left(\left(\frac{\rho s + \mu k^2}{\mu \beta}\right)^{1/2} y\right) + C_4 \exp\left(-\left(\frac{\rho s + \mu k^2}{\mu \beta}\right)^{1/2} y\right)$$

C_2 + C_3 must be zero to keep $\hat{u}_x$ bound at $-\infty$

$$\therefore \hat{u}_y = C_1 \exp(ky) + C_3 \exp\left(\left(\frac{\rho s + \mu k^2}{\mu \beta}\right)^{1/2} y\right)$$

Fluid α :

$$\hat{u}_y = A_1 \exp(ky) + A_2 \exp(-ky) + A_3 \left(\left(\frac{\rho_\alpha S + \mu_\alpha k^2}{\mu_\alpha} \right)^{1/2} y \right) + A_4 \exp \left(- \left(\frac{\rho_\alpha S + \mu_\alpha k^2}{\mu_\alpha} \right)^{1/2} y \right)$$

$A_1 + A_3$ must be zero

$$\hat{u}_y = A_2 \exp(-ky) + A_4 \exp \left(- \left(\frac{\rho_\alpha S + \mu_\alpha k^2}{\mu_\alpha} \right)^{1/2} y \right)$$

Boundary Conditions

- Equality of Fluid velocities on interface (evaluated at $y=0$ for small disturbance)

$$\hat{u}_y^\alpha = \hat{u}_y^\beta, y=0$$

$$C_1 + C_3 = A_2 + A_4$$

$$\hat{u}_x^\alpha = \hat{u}_x^\beta, y=0$$

$$C_1 k + C_3 \left(\frac{\rho_\beta S + \mu_\beta k^2}{\mu_\beta} \right)^{1/2} = -A_2 k - A_4 \left(\frac{\rho_\alpha S + \mu_\alpha k^2}{\mu_\alpha} \right)^{1/2}$$

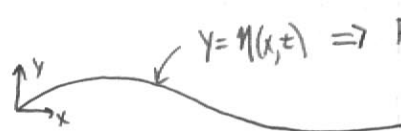
- The Young-Laplace Equation can be used as a boundary condition at the interface

$$P_\beta - P_\alpha = \gamma \nabla \cdot \underline{n} \quad @ y=0$$

The pressures in each fluid are determined using equation (I)

$$P_\beta = \frac{-C_1 \rho_\beta S \exp(ky)}{k} \exp(st+ikx) ; \quad P_\alpha = \frac{A_2 \rho_\alpha S \exp(-ky)}{k} \exp(st+ikx)$$

$\nabla \cdot \underline{n}$


$$y = \eta(x, t) \Rightarrow F(x, y, t) = y - \eta(x, t) = 0$$

$$\underline{n} = \frac{\nabla F}{|\nabla F|}$$

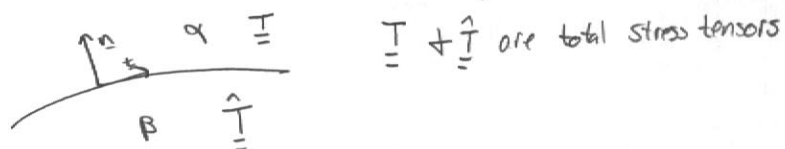
$$\underline{n} = \frac{\delta_y - \frac{\partial \eta}{\partial x} \delta_x}{\left(1 + \left(\frac{\partial \eta}{\partial x}\right)^2\right)^{1/2}} \stackrel{\text{linearizing}}{\approx} \delta_y - \frac{\partial \eta}{\partial x} \delta_x$$

$$\nabla \cdot \underline{n} = -\frac{\partial^2 \eta}{\partial x^2} = +\hat{\eta} k^2 \exp(kt + i k x)$$

$$\therefore -\frac{C_1 \rho_s \exp(0) \exp(kt + i k x)}{k} - \frac{A_2 \rho_s \exp(0) \exp(kt + i k x)}{k} = \delta_0 k^2 \hat{\eta} \exp(kt + i k x)$$

$$\boxed{C_1 \rho_s + A_2 \rho_s = -k^2 \delta_0 \hat{\eta}}$$

- A surface stress balance is necessary to determine how gradients in surface tension affect the flow



$$\underline{n} \cdot (\underline{T}_\alpha - \underline{T}_\beta) + \gamma \underline{n} (\nabla \cdot \underline{n}) + \nabla \gamma = 0$$

dotting all terms w/ $\underline{t}$

$$\underline{n} \cdot (\underline{T}_\alpha - \underline{T}_\beta) \cdot \underline{t} + \gamma \cancel{\underline{n} \cdot \underline{t}} (\nabla \cdot \underline{n}) + \nabla \gamma \cdot \underline{t} = 0$$

orthogonal vectors

$$\underline{n} \cdot (\underline{T}_\alpha - \underline{T}_\beta) \cdot \underline{t} + \nabla \gamma \cdot \underline{t} = 0$$

$$\underline{T}_\alpha = -p \underline{I} + \underline{\tau}_\alpha ; \quad \underline{T}_\beta = -\hat{p} \underline{I} + \underline{\tau}_\beta \quad \text{substituting } \underline{T}_\alpha + \underline{T}_\beta$$

$$\cancel{\underline{n} \cdot \underline{t}} (\hat{p} - p) + \underline{n} \cdot (\underline{\tau}_\alpha - \underline{\tau}_\beta) \cdot \underline{t} = -\nabla \gamma \cdot \underline{t}$$

$$\therefore \underline{n} \cdot (\underline{\tau}_\alpha - \underline{\tau}_\beta) \cdot \underline{t} = -\nabla \gamma \cdot \underline{t}$$

$$\underline{\tau}_\alpha = \begin{pmatrix} \tau_{xx}^\alpha & \tau_{xy}^\alpha \\ \tau_{yx}^\alpha & \tau_{yy}^\alpha \end{pmatrix} ; \quad \underline{\tau}_\beta = \begin{pmatrix} \tau_{xx}^\beta & \tau_{xy}^\beta \\ \tau_{yx}^\beta & \tau_{yy}^\beta \end{pmatrix}$$

$$\underline{\tau}_\alpha - \underline{\tau}_\beta = \begin{pmatrix} \tau_{xx}^\alpha - \tau_{xx}^\beta & \tau_{xy}^\alpha - \tau_{xy}^\beta \\ \tau_{yx}^\alpha - \tau_{yx}^\beta & \tau_{yy}^\alpha - \tau_{yy}^\beta \end{pmatrix} ; \quad \underline{n} = n_1 \delta_x + n_2 \delta_y ; \quad \underline{t} = t_1 \delta_x + t_2 \delta_y$$

$$\nabla \cdot (\underline{\underline{\gamma}}_\alpha - \underline{\underline{\gamma}}_\beta) = \delta_x [(\gamma_{xx}^\alpha - \gamma_{xx}^\beta)n_1 + (\gamma_{xy}^\alpha - \gamma_{xy}^\beta)n_2] + \delta_y [(\gamma_{yx}^\alpha - \gamma_{yx}^\beta)n_1 + (\gamma_{yy}^\alpha - \gamma_{yy}^\beta)n_2]$$

$$\nabla \cdot (\underline{\underline{\gamma}}_\alpha - \underline{\underline{\gamma}}_\beta) \cdot \underline{\underline{t}} = [(\gamma_{xx}^\alpha - \gamma_{xx}^\beta)n_1 + (\gamma_{xy}^\alpha - \gamma_{xy}^\beta)n_2]t_1 + [(\gamma_{yx}^\alpha - \gamma_{yx}^\beta)n_1 + (\gamma_{yy}^\alpha - \gamma_{yy}^\beta)n_2]t_2$$

$$t_1 n_1 (\gamma_{xx}^\alpha - \gamma_{xx}^\beta) + t_1 n_2 (\gamma_{xy}^\alpha - \gamma_{xy}^\beta) + t_2 n_1 (\gamma_{yx}^\alpha - \gamma_{yx}^\beta) + t_2 n_2 (\gamma_{yy}^\alpha - \gamma_{yy}^\beta) = -\nabla \gamma \cdot \underline{\underline{t}}$$

$$\nabla \gamma = \frac{\partial \gamma}{\partial x} \delta_x + \frac{\partial \gamma}{\partial y} \delta_y \Rightarrow \nabla \gamma \cdot \underline{\underline{t}} = t_1 \frac{\partial \gamma}{\partial x} + t_2 \frac{\partial \gamma}{\partial y}$$

$\underline{\underline{t}}$:



$$\underline{\underline{t}} = \frac{dx \delta_x + dy \delta_y}{(dx^2 + dy^2)^{1/2}} = \frac{\delta_x + \frac{dy}{dx} \delta_y}{(1 + (\frac{dy}{dx})^2)^{1/2}} \stackrel{\text{linearizing}}{\simeq} \delta_x + \frac{dy}{dx} \delta_y$$

$$\text{Now } t_1 = 1, t_2 = \frac{dy}{dx}, n_1 = -\frac{dy}{dx}, n_2 = 1$$

$$(\gamma_{xy}^\alpha - \gamma_{xy}^\beta) \left(1 - \left(\frac{dy}{dx}\right)^2\right) + \left(-\frac{dy}{dx}\right) (\gamma_{xx}^\alpha - \gamma_{xx}^\beta) + \frac{dy}{dx} (\gamma_{yy}^\alpha - \gamma_{yy}^\beta) = -\frac{\partial \gamma}{\partial x} - \frac{dy}{dx} \frac{\partial \gamma}{\partial y}$$

linearizing
assuming small disturbance
assuming small disturbance
assuming small disturbance

$$\therefore \boxed{(\gamma_{xy}^\alpha - \gamma_{xy}^\beta) \simeq -\frac{\partial \gamma}{\partial x} = -\frac{\partial \gamma}{\partial T} \frac{\partial T}{\partial x} \quad y=0}$$

γ_{xy}

$$\gamma_{xy} = \mu \left(\frac{\partial u_x}{\partial y} + \frac{\partial u_y}{\partial x} \right)$$

Surface tension gradient:

$$\frac{\partial \gamma}{\partial x} = \frac{\partial \gamma}{\partial T} \frac{\partial T}{\partial x} \Big|_{y=\eta}$$

$$T = G \eta(y) \Rightarrow \frac{\partial T}{\partial x} = G \frac{\partial \eta}{\partial x} = G i k \hat{\eta} \exp(st + i k x)$$

$$\frac{\partial \gamma}{\partial T} = -\frac{\gamma_0}{T_c}$$

$$\therefore \boxed{\frac{\partial \gamma}{\partial x} = -\frac{\gamma_0 G i k \hat{\eta}}{T_c} \exp(st + i k x)}$$

Substituting in disturbance equations in xy leads to surface stress balance Boundary condition

$$2\mu_p k^2 C_1 + (\rho_p S + 2\mu_p k^2) C_3 - 2\mu_\alpha k^2 A_2 - (\rho_\alpha S + 2\mu_\alpha k^2) A_4 = -\frac{\gamma_0 G k^2 \hat{\eta}}{\Gamma_c}$$

Kinematic condition



$$\frac{dF}{dt} = \frac{\partial F}{\partial x} \frac{\partial k}{\partial t} + \frac{\partial F}{\partial y} \frac{\partial y}{\partial t} + \frac{\partial F}{\partial t} = 0$$

$\downarrow$ $\downarrow$
 u_x u_y

$$-\frac{\partial \eta}{\partial x} u_x + u_y - \frac{\partial \eta}{\partial t} = 0 \quad \therefore u_y|_{y=\eta} = \frac{\partial \eta}{\partial t} = \hat{\eta} S \exp(st + ikx)$$

$\swarrow$
 $O(\eta^2)$

Fluid B

$$u_y|_{y=\eta} = C_1 \exp(k\eta) \exp(st + ikx) + C_3 \left(\left(\frac{\rho_p S + \mu_p k^2}{\mu_p} \right)^{1/2} \eta \right) \exp(st + ikx) = \hat{\eta} S \exp(st + ikx)$$

For small η $\exp(k\eta) \simeq 1 + k\eta + \dots$

$$\therefore \boxed{C_1 + C_3 \simeq \hat{\eta} S}$$

The four boundary conditions were used to find $C_1, C_3, A_2 + A_4$. This was done using Mathematica. Then the kinematic condition was used to determine how S varied w/ k . Because of the non-linear relationship, MATLAB's `fsolve` function was used to generate plots shown in paper.