

# A Mathematical Model for Core-Annular Flows with Surfactants

*Un Modelo Matemático para Flujos Centro-Anulares  
con Surfactantes*

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## Abstract

The stability of core-annular flows is of fundamental scientific and practical importance. The interface between two immiscible fluids can become unstable by several physical mechanisms. Surface tension is one of those mechanisms of practical importance. We include in our model the effects of insoluble surfactants. A full problem is derived considering the surfactant transport equation. We carried out an asymptotic solution of the problem when the annulus is thin compared to the core-fluid radius and for waves which are of the order of the pipe radius. We obtain from matched asymptotic analysis a system of two coupled nonlinear partial differential equations for the interfacial amplitude and the surfactant concentration on the interface.

**Key words and phrases:** core-annular flow, surfactants, interfacial tension.

## Resumen

La estabilidad de flujos centro-anulares es de fundamental importancia científica y práctica. La interface entre dos fluidos puede llegar a inestabilizarse por varios mecanismos físicos. La tensión superficial es uno de esos mecanismos de importancia práctica. Incluimos en nuestro modelo los efectos de surfactantes insolubles. Derivamos nuestro problema completo considerando la ecuación de transporte de surfactantes.

Buscamos una solución asintótica del problema cuando el ánulo es fino comparado con el radio del fluido central y para ondas que son del orden del radio del tubo. Obtenemos del análisis asintótico empalmado un sistema de dos ecuaciones diferenciales parciales no lineales acopladas para la amplitud interfacial y la concentración de surfactantes sobre la interface.

**Palabras y frases clave:** flujo centro-anular, surfactantes, tensión interfacial.

## 1 Introduction

Two immiscible fluids, generally, arrange themselves such that the less viscous one is in the region of high shear (Joseph and Renardy [18], 1993). When a fluid in a capillary tube is displaced by another, a layer of the first fluid is left behind coating the tube walls (Taylor [34], 1961). This is called a **core-annular flow**. In general, core-annular flows are parallel flows of immiscible liquids in a cylinder; one fluid flows through the cylinder core and the other ones move in successive annuli that surround the core fluid. Core-annular flow occurs, for example, during liquid-liquid displacements in porous media (Edwards, Brenner, and Wasan [4], 1991) when a wetting layer is present, and in lung airways where internal airway surface is coated with a thin liquid lining (Halpern and Grotberg [10], 1992 and [11], 1993).

The case of lubricated pipelining is an important technological application of interest to the oil industry, where the annular liquid (water) lubricates the motion of the core liquid (viscous oil). There are several flow regimes in horizontal pipes including, stratified flow with heavy fluid below, bamboo waves, oil bubbles and slugs in water, water in oil with(out) emulsions, and an annulus of water surrounding a concentric oil core.

Stein ([33], 1978), and Oliemans, Ooms, Wu and Duÿvestin ([24], 1985) have developed experiments on water-lubricated pipelining. Advances of economically acceptable pipelines has been developed by The Shell Oil Company. About ten years ago, Maraven of PdVSA (Petróleos de Venezuela Sociedad Anónima) implemented a 60 kilometer pipeline for the transportation of water-lubricated heavy oil.

There are a number of studies concerned with core-annular flows when the interfaces are free of surfactant. For the case of no flow, studies of a long cylindrical thread of a viscous liquid suspended in a different unbounded fluid were developed by Tomotika ([35], 1935). Goren ([9], 1962) studied the linear stability of an annular film coating a wire or the inner surface of a cylinder,

when the ambient or core fluid is inviscid. His results indicate that the film is unstable to infinitesimal sinusoidal disturbances.

Hammond ([12], 1983) developed a nonlinear analysis based on lubrication theory for the adjustment of a thin annular film under surface tension. He suggests that an initial sinusoidal disturbance of the interface may lead to the breakup of the film in the form of axisymmetric droplets or 'lenses' of the annular liquid separated by the core fluid.

For the more general case of two co-flowing fluids with a core-annular configuration, the linear theory of stability has been studied by several people. Hickox ([13], 1971), Joseph, Renardy, and Renardy ([15], 1983 and [16], 1984), Smith ([29], 1989), Russo and Steen ([26], 1989), Hu and Joseph ([14], 1989), and Chen, Bai, and Joseph ([1], 1990) are some of them.

Georgiou, Maldarelli, Papageorgiou, and Rumschitzki ([8], 1992) analyzed the linear stability of a vertical, perfectly concentric core-annular flow in the limit when the film is much thinner than the core. Using asymptotic expansions, they developed a new theory for the linear stability of the wetting layer in low-capillary-number liquid-liquid displacements.

Kouris and Tsamopoulos ([21], 2001) studied the linear stability of core-annular flow of two immiscible fluids in a periodically constricted tube.

Dynamics of core-annular flows in which effects of nonlinearity are kept can be described by nonlinear stability theories. Frenkel, Babchin, Levich, Shlang, and Sivashinsky ([7], 1987) studied two fluids in a straight tube with the annular one being thin. Both fluids have equal properties and only interfacial tension acts between them. They derived the Kuramoto-Sivashinsky equation which includes both stabilizing and destabilizing terms related to the interfacial tension, leading to growth of the initial disturbances. Frenkel ([6], 1988) considered the wavelength to be long compared to the annular thickness but of the order of the core radius. He developed a modified Kuramoto-Sivashinsky equation for slow flow and discussed how the extra terms in his equation could alter the behavior of the Kuramoto-Sivashinsky equation. Papageorgiou, Maldarelli, and Rumschitzky ([25], 1990) investigated the weakly nonlinear evolution of thin films (wavelength long compared to the annular thickness). They studied the core contribution by searching in a larger set of core flow regimes. They conclude that viscosity stratification greatly increases the likelihood of regular nonlinear traveling waves.

Kerchman ([20], 1995) modeled the problem of oil in the annular region using strongly nonlinear theory. A modified Kuramoto-Sivashinsky equation was derived with additional dispersive terms. By solving this last equation he found a large variety of solutions in the dynamics, from chaos to quasi-steady waves. Coward, Papageorgiou, and Smyrlis ([2], 1995) examined the

case when the pressure gradient is modulated by time harmonic oscillations. Viscosity stratification and interfacial tension are present. They developed a weakly nonlinear asymptotic approximation valid for thin annular films.

Kouris and Tsamopoulos ([22], 2001) studied the nonlinear dynamics of a concentric, two-phase flow of immiscible fluids in a cylindrical tube, when the more viscous fluid is in the core for any thickness of the film. Also, in Kouris and Tsamopoulos ([23], 2002) they studied the nonlinear dynamics of a concentric, two-phase flow of immiscible fluids in a cylindrical tube, with the less viscous fluid in the core.

In general, the presence of even minute amounts of surfactant on a fluid-fluid interface can have a substantial effect on the evolution of the interface (Edwards, Brenner, and Wasan [4], 1991). Insoluble surfactants are large molecules possessing a dipolar structure formed of hydrophobic (i.e. water-repelling) and hydrophilic (i.e. water-attracting) segments; in this way, insoluble surfactants are distributed on interfaces separating aqueous and nonaqueous phases as water and oil. Surfactants influence the interfacial dynamics in two ways. Firstly, most types of surfactant reduce the interfacial tension, i.e. the surface tension in a surfactant coated interface is lower than that for a clean interface, with the interfacial tension correspondingly lower over regions of the interface with higher surfactant concentration. Secondly, the presence of a gradient in surfactant concentration introduces a Marangoni force. This is a force along the interface which is directed from regions of high surfactant concentration (i.e. low surface tension) to regions of low surfactant concentration (i.e. high surface tension). In general, the Marangoni force acts to oppose any external flow which promotes build-up or excess of surfactant along the interface.

Some theoretical studies of deforming drops under the effects of surfactants were developed by Flumerfelt ([5], 1980). Siegel ([28], 1999) employed a simple plane flow model to examine the deformation of a bubble in strain type flows and under the influence of surfactants.

The stability of core-annular flows in the presence of surfactant has received little attention. Most of the work on the effects of surfactant in core-annular flow have been motivated by applications to pulmonary fluid dynamics. The lung airways are internally coated by a thin film of a liquid forming a liquid-air interface. The interfacial tension tries to minimize the interfacial area. Thus, the coating liquid may cause closing off of the tiny airways by the formation of a meniscus during exhalation. Biological surfactant tends to reduce the interfacial tension by decreasing the attractive force between molecules of the film. A role of surfactant then, is to have a stabilizing effect which prevents collapses and keeps airways open.

Here, we want to explore the influence of surfactant in a core-annular flow when the core liquid is surrounded by another annular liquid. We assume the surfactant to be insoluble in the film and the core. This, physically, corresponds to surfactant that has a very low solubility in both the film and core fluids. So, the surfactant remains at the fluid-fluid interface.

In this work, we employ a long wave asymptotic analysis to carefully derive a coupled nonlinear system of equations. The nonlinear system derived is a forced Kuramoto-Sivashinky equation, the forcing arising from the Marangoni effect.

Section 2 describes the mathematical model, governing equations, and basic flow. Section 3 presents the asymptotic analysis leading to the evolution equations. Section 4 re-scales the evolution equations to canonical form and presents the linear analysis of a particular case.

## 2 Mathematical model, governing equations, and basic flow

Our problem consists of an annular liquid film (fluid 2),  $-\infty < z < \infty$ , surrounding an infinitely long cylindrical fluid core (fluid 1). Fluid 1 is of undisturbed radius  $R_1$  and viscosity  $\mu_1$ . The viscosity of fluid 2 is  $\mu_2$  and the tube is of radius  $R_2$ . Here, we take the densities of the film and core fluids to be the same and equal to  $\rho$ . Hence, gravitational effects are neglected (Hammond [12], 1983, Hu and Joseph [14], 1989, and Joseph and Renardy [18], 1993); gravity does not appreciably change the shape of the interface if the Bond number  $B_0 = \frac{\rho g a^2}{\sigma}$  is small. The flow is driven by a constant pressure gradient  $\nabla p = -F \underline{e}_z$ , where  $\underline{e}_z = (0, 0, 1)$  and  $F > 0$ . Insoluble surfactants are present on the fluid interface; we denote the surfactant concentration (in units of mass of surfactant per unit of interfacial area) by  $\Gamma^*$ .

The interfacial tension  $\sigma$  and the surfactant concentration  $\Gamma$  are related by the linear expression

$$\sigma(\Gamma) = \sigma_o(1 - \beta\Gamma), \quad (1)$$

where  $\beta = \frac{R_g T \Gamma_\infty}{\sigma_o}$  and  $\sigma_o$  is the interfacial tension of the clean interface,  $R_g$  is the ideal gas constant,  $T$  is the temperature,  $\Gamma_\infty$  is the maximum packing concentration that the interface can support, and  $\Gamma$  is the dimensionless surfactant concentration  $\Gamma = \frac{\Gamma^*}{\Gamma_\infty}$ . Our asymptotic solution is developed for small surfactant variations about a uniform state.

We use cylindrical polar coordinates  $\vec{x} = (r, \theta, z)$  with associated velocity components  $\vec{u}_1 = (u_1, v_1, w_1)$  for the fluid core and  $\vec{u}_2 = (u_2, v_2, w_2)$  for the fluid film. The interface between the fluids is denoted by  $r = S(z, \theta, t)$ . Our problem is axisymmetric and the dimensional interface  $S(z, t)$  can be written as

$$S(z, t) = R_1(1 + \delta H), \quad (2)$$

where  $R_1$  is the undisturbed core radius, and  $\delta$  is a dimensionless amplitude.

For the interface evolution, we start from the **Navier-Stokes** and **Continuity** equations for axisymmetric flows. We require a **no-slip condition** at the pipe wall  $\vec{u}_2 = 0$ , **continuity of velocity** at the interface  $\vec{u}_1 = \vec{u}_2$ , and the **kinematic condition** holding at the interface  $r = S(z, t)$  (Joseph and Renardy [17], 1993). We also require **Normal Stress Balance** and **Tangential Stress Balance** at the interface.

We start from the convective-diffusion equation for **surfactant transport** to obtain the surfactant concentration evolution (Wong, Rumschitzki, and Maldarelli [37], 1996), using results from tensor analysis (Rutherford [27], 1989, Wheeler and McFadden [36], 1994, and Kas-Danouche [19], 2002).

Let us non-dimensionalize lengths, selecting the base core radius  $R_1$ , velocities are non-dimensionalized by the centerline velocity  $W_0$ , time by  $R_1/W_0$ , interfacial tension by the surface tension  $\sigma_0$  in the absence of surfactants, which is called the ‘clean’ surface tension, and pressure by  $\rho W_0^2$ , where  $\rho$  is the density of the fluids.

For the Navier-Stokes equations the nondimensionalization introduces the **Reynolds numbers** ( $Re_i$ ),  $i = 1$  for the core fluid and  $i = 2$  for the film fluid, defined by  $Re_i = \rho W_0 R_1 / \mu_i$  corresponding to the relative importance of the inertial and viscous forces acting on unit volume of the fluid  $i$ . The nondimensionalization of the surfactant transport equation produces the **Peclet number** ( $Pe$ ) which defines the transport ratio between convection and diffusion and is given by  $Pe = \frac{W_0 R_1}{D_s}$ . In the normal stress balance, the nondimensionalization leads to a **surface tension parameter**  $J = \frac{\sigma_0 R_1}{\rho \nu_1^2}$ . The

**Capillary number** ( $C_a$ ) and **viscosity ratio**  $m$  arise in the dimensionless tangential stress balance. The capillary number is given by  $C_a = \frac{\mu_1 W_0}{\sigma_0}$ . It measures the relative ratio between the base flow velocity and the capillary velocity. The viscosity ratio is given by  $m = \frac{\mu_2}{\mu_1}$ , the ratio of the film fluid viscosity to the core fluid viscosity. Note that the capillary number can be expressed in terms of  $Re_1$  and  $J$  as  $C_a = \frac{Re_1}{J}$  and the viscosity ratio in terms

of  $Re_1$  and  $Re_2$  as  $m = \frac{Re_1}{Re_2}$ .

The dimensionless Navier-Stokes equations and the continuity equation are

$$(u_i)_t + u_i(u_i)_r + w_i(u_i)_z = -(p_i)_r + \frac{1}{Re_i} \left[ \nabla^2 u_i - \frac{u_i}{r^2} \right], \quad (3)$$

$$(w_i)_t + u_i(w_i)_r + w_i(w_i)_z = -(p_i)_z + \frac{1}{Re_i} \nabla^2 w_i, \quad (4)$$

$$(u_i)_r + \frac{1}{r} u_i + (w_i)_z = 0, \quad (5)$$

where  $i = 1, 2$  for core and film respectively. The dimensionless surfactant equation is

$$\begin{aligned} \frac{\partial \Gamma}{\partial t} &- \frac{\dot{S}S'}{1+(S')^2} \frac{\partial \Gamma}{\partial z} + \frac{1}{S\sqrt{1+(S')^2}} \left\{ \frac{\partial}{\partial z} \left[ \frac{S\Gamma}{\sqrt{1+(S')^2}} (w + S'u) \right] \right\} \\ &- \frac{1}{Pe} \frac{1}{S\sqrt{1+(S')^2}} \frac{\partial}{\partial z} \left( \frac{S}{\sqrt{1+(S')^2}} \frac{\partial \Gamma}{\partial z} \right) \\ &+ \frac{\Gamma}{S(1+(S')^2)} \left[ 1 - \frac{SS''}{1+(S')^2} \right] (-S'w + u) = 0. \end{aligned} \quad (6)$$

The no-slip condition at the pipe wall is

$$u_2 = w_2 = 0 \quad \text{at } r = \frac{R_2}{R_1},$$

the continuity of velocities is

$$\{u_i\}_1^2 = 0, \quad \{w_i\}_1^2 = 0 \quad \text{on } r = S(z, t),$$

and the Kinematic condition is

$$u = \frac{\partial S}{\partial t} + w \frac{\partial S}{\partial z} = S_t + wS'.$$

The dimensionless normal stress balance is

$$\begin{aligned} \left\{ p(1+(S')^2) - \frac{2}{Re_i} [(S')^2 w_z - S'(u_z + w_r) + u_r] \right\}_1^2 = \\ \frac{J(1-\beta\Gamma)}{Re_1^2} \left\{ S'' - \frac{1}{S} [1+(S')^2] \right\} [1+(S')^2]^{-\frac{1}{2}}, \end{aligned} \quad (7)$$

and the dimensionless tangential stress balance is

$$\{m_i [2S'(u_r - w_z) + [1 - (S')^2](u_z + w_r)]\}_1^2 = -\frac{\beta\Gamma_z}{C_a}[1 + (S')^2]^{\frac{1}{2}}, \quad (8)$$

where  $m_1 = 1$  and  $m_2 = m$ .

We begin our stability analysis by finding the dimensionless basic state driven by a constant pressure gradient  $p_z = -\frac{FR_1}{\rho W_0^2}$  and using the definition of the Reynolds number,  $Re_i = \frac{\rho w_0 R_1}{\mu_i}$

$$w_1 = -\frac{1}{4\mu_1} \frac{FR_1^2}{W_0}(r^2 - 1) - \frac{1}{4\mu_2} \frac{FR_1^2}{W_0}(1 - a^2), \quad (9)$$

$$w_2 = -\frac{1}{4\mu_2} \frac{FR_1^2}{W_0}(r^2 - a^2). \quad (10)$$

The dimensionless centerline velocity is  $w_1(r = 0) = 1$ , and the dimensional centerline velocity is

$$W_0 = \frac{1}{4\mu_1\mu_2} [(\mu_2 - \mu_1)R_1^2 + \mu_1 R_2^2].$$

Substitution of  $W_0$  in (9) and (10) leads to a closed form of  $w_1$  and  $w_2$  in terms of  $r$ ,  $a$ , and  $m$

$$w_1 = 1 - \frac{mr^2}{a^2 + m - 1}, \quad 0 \leq r \leq 1, \quad (11)$$

$$w_2 = -\frac{r^2 - a^2}{a^2 + m - 1}, \quad 1 \leq r \leq a, \quad (12)$$

where  $a = R_2/R_1$  and  $m = \mu_2/\mu_1$ .

The difference in pressures of the basic core-annular flow comes from the normal stress balance

$$p_2 - p_1 = -\frac{J(1 - \beta\Gamma_0)}{Re_1^2} = -\frac{\sigma_0(1 - \beta\Gamma_0)}{\rho W_0^2 R_1}, \quad (13)$$

where  $p_1$  and  $p_2$  are the basic core and film pressures, respectively.



### 3 Derivation of the evolution equations

Now, we derive an asymptotic solution considering the smallness of the thickness of the film (relative to the core). A coupled system of leading order evolution equations is obtained. One equation describes the spatio-temporal evolution of the interface between the core and the film, and the other describes the evolution of the concentration of surfactant at the interface. The core radius (in the nondimensional undisturbed state) is 1 and the distance from the wall to the interface is  $\varepsilon = a - 1$ , where  $a = R_2/R_1$ .

We proceed asymptotically with  $\varepsilon \ll 1$ . Let us consider deformation of the interface to heights of order  $\delta$ , where  $\delta \ll \varepsilon$ . (This corresponds to a weakly nonlinear theory; in the absence of a background flow, we can deal with  $\delta \sim \varepsilon$ .) Thus, the disturbed interface, can be expressed as  $S(z, t) = 1 + \delta H(z, t)$  and the dimensionless film thickness is  $\varepsilon - \delta H(z, t)$ .

To separate the radial scales in the film and core, a local variable is introduced in the film region by  $r = a - \varepsilon y$ , where  $y$  is 0 at the pipe wall and  $1 - \frac{\delta}{\varepsilon} H(z, t)$  at the interface.

#### 3.1 Derivation of the Interface Evolution Equation

Balance of terms in the scaled continuity equation for small  $\varepsilon$  provides an estimate for  $u_2$  to be equal to  $\varepsilon$  times the order of  $w_2$ . Similarly, from the continuity equation in the core,  $u_1$  and  $w_1$  must be of the same order.

Suppose  $p_1 = \bar{p}_1 + \tilde{p}_1$  and  $p_2 = \bar{p}_2 + \tilde{p}_2$ , where  $\bar{\quad}$  and  $\tilde{\quad}$  indicate base and perturbed states, respectively. From the Navier-Stokes equation (4) in the film and in the core, and supposing  $(\bar{p}_2)_z \sim 1$  and  $(\bar{p}_1)_z \sim 1$ , we find that balancing viscous terms with the perturbation pressure gradient (this is essentially a lubrication approximation) gives

$$\tilde{p}_{2z} \sim \frac{1}{Re_2 \varepsilon^2} (\text{order of } w_2) \quad \text{and} \quad \tilde{p}_{1z} \sim \frac{1}{Re_1} (\text{order of } w_1),$$

respectively. Continuity of axial velocity at the interface yield order of  $w_1 =$  order of  $w_2$ . From the normal stress balance (7), we obtain the order of the perturbation pressure to be  $\tilde{p}_2 \sim \frac{J\delta}{Re_1^2}$ . This scaling indicates that we are considering capillary driven motions which arise from pressure changes due to surface tension. Therefore,

$$w_2 \sim \frac{\varepsilon^2 \delta J Re_2}{Re_1^2} \quad \text{and} \quad u_2 \sim \frac{\varepsilon^3 \delta J Re_2}{Re_1^2}.$$

For the case of  $m = \frac{Re_1}{Re_2} \sim O(1)$ , the film perturbation velocities can be estimated to be

$$w_2 \sim \frac{\varepsilon^2 \delta J}{Re_1} \quad \text{and} \quad u_2 \sim \frac{\varepsilon^3 \delta J}{Re_1}. \quad (14)$$

The difference in basic axial velocity,  $w_1 - w_2$ , at the interface,  $r = S = 1 + \delta H$ , is

$$(w_1 - w_2)|_{r=1+\delta H} = \frac{2(1-m)\delta H + O(\delta^2)}{m + 2\varepsilon - \varepsilon^2} \sim O(\delta). \quad (15)$$

Physically, this arises from viscosity stratification. We already know, from (14), that the axial velocity perturbation in the film is of  $O(\varepsilon^2 \delta J / (Re_1))$ , implying that the core contribution must be of  $O(\delta)$ , to ensure continuity of velocities.

From the tangential stress balance (8), the dominant term in the film is the radial derivative of the axial film velocity,  $w_{2,r}$ , which is of  $O(\varepsilon \delta J / (Re_1))$  and the core contribution is of  $O(\delta)$ . So, we consider various regimes.

**Regime 1. Film and core do not couple (film contribution dominates over the core contribution):**

If the film contribution dominates over the core contribution; i.e.  $\varepsilon J \gg Re_1$  and  $C_a \ll \varepsilon$ , then the core influence is not introduced into the dynamics of the problem to leading order. This decoupling is a result of the core contribution in the tangential stress balance equation (8) being of lower order than the corresponding film contribution. The kinematic condition (2)

$$u = S_t + (\bar{w} + \tilde{w})S_z,$$

where  $\bar{w}$  represents the base state axial velocity and  $\tilde{w}$  represents the perturbed axial velocity, taken in a frame of reference traveling with speed  $\bar{w}(r = 1; \varepsilon) \sim O(\varepsilon)$ , provides an estimate for  $\delta$ , the interfacial amplitude. This comes from balancing  $u$  and the convective term on the right hand side, as well as allowing for unsteadiness on a new long time scale. We find

$$\delta = \frac{\varepsilon^3 J}{Re_1} \gg \varepsilon^2,$$

since  $\frac{\varepsilon J}{Re_1} \gg 1$ . The size of  $\delta$  depends on the magnitude of  $\frac{\varepsilon J}{Re_1}$  and the evolution ranges from highly nonlinear regimes,  $\delta \sim \varepsilon$ , leading to Hammond type equations (Hammond [12], 1983), or weakly nonlinear regimes leading

to the Kuramoto-Sivashinsky equation (Smyrlis and Papageorgiou [30], 1991 ;[31], 1996 and [32], 1998). We are interested in the case when film and core couple, and in what follows, we describe these delicate scalings in full detail.

**Regime 2. Film and core couple:**

If the film contribution and core contribution balance, then  $\varepsilon J \sim Re_1$  and  $C_a \sim \varepsilon$ . In this regime, we consider two cases. One case with **moderate surface tension** ( $J \sim O(1)$ ) and **slow moving core** ( $Re_1 \sim O(\varepsilon)$ ). Another case with **strong surface tension** ( $J \sim O(1/\varepsilon)$ ) and **moderate core flow** ( $Re_1 \sim O(1)$ ).

The kinematic condition using the film variables

$$u_2 = S_t + (\bar{w}_2 + \tilde{w}_2)S_z, \quad (16)$$

where  $\bar{w}_2$  and  $\tilde{w}_2$  represent the base state and perturbed axial velocities of the film, respectively, has to be balanced; but, the term  $u_2$  is of  $O(\varepsilon^2\delta)$ ,  $S_t$  is of order  $\delta$  times order of the time scale  $t$ , and the term  $(\bar{w}_2 + \tilde{w}_2)S_z$  is of  $O(\varepsilon\delta)$ , so, we can not balance them. Using the Galilean transformation defining a system of coordinates traveling with speed  $\bar{w}_{2\varepsilon}$ ,

$$\frac{\partial}{\partial t} \longrightarrow -\bar{w}_{2\varepsilon} \frac{\partial}{\partial z} + \frac{\partial}{\partial \tilde{t}}. \quad (17)$$

where  $\bar{w}_2|_{r=1+\delta H} = \bar{w}_{2\varepsilon} + \bar{w}_{2\delta}$ , with  $\bar{w}_{2\varepsilon} \sim O(\varepsilon)$  and  $\bar{w}_{2\delta} \sim O(\delta)$ , we achieve a balance in (16). So, plugging (17) in the kinematic condition (16) and balancing all the terms and introducing a new time variable  $\tau$  we conclude that  $\delta = \varepsilon^2$  and  $\tau = \delta t$ .

Consider the case  $J \sim O(1)$  and  $Re_1 \sim O(\varepsilon)$  of the regime 2. Then, in the film

$$u_2 = \varepsilon^4 \tilde{u}_2 + O(\varepsilon^5) \quad (18)$$

$$w_2 = \bar{w}_2 + \varepsilon^3 \tilde{w}_2 + O(\varepsilon^4) \quad (19)$$

$$p_2 = \bar{p}_2 + \tilde{p}_0 + \varepsilon \tilde{p}_2 + \dots \quad (20)$$

and in the core

$$u_1 = \varepsilon^2 \tilde{u}_1 + O(\varepsilon^3) \quad (21)$$

$$w_1 = \bar{w}_1 + \varepsilon^2 \tilde{w}_1 + O(\varepsilon^3) \quad (22)$$

$$p_1 = \bar{p}_1 + \varepsilon \tilde{p}_1 + \dots \quad (23)$$

Set  $Re_1 = \lambda\varepsilon$ , where  $\lambda \sim O(1)$ . Substituting  $u_2$ ,  $w_2$  and,  $p_2$  into the Navier-Stokes equations and using  $Re_1 = mRe_2$ , we obtain to leading order from

(4)

$$\tilde{p}_{0z} - \frac{m}{\lambda} \tilde{w}_{2yy} = 0. \quad (24)$$

From (3) keeping leading order terms, we obtain  $\tilde{p}_{0y} = 0$  and keeping leading order terms in the continuity equation (5) yields  $\tilde{w}_{2z} = \tilde{u}_{2y}$ .

The fact that  $\tilde{p}_0$  is not a function of  $y$ , allows us to integrate (24) twice to get

$$\tilde{w}_2 = \frac{\lambda}{m} \left( \frac{1}{2} \tilde{p}_{0z} y^2 + A(z, t) y \right), \quad (25)$$

where the no-slip condition  $\tilde{w}_2(y = 0, z, t) = 0$  has been used. Now, we differentiate  $\tilde{w}_2$  with respect to  $z$  and then integrate the resulting equation over  $y$  to obtain an expression for  $\tilde{u}_2$

$$\tilde{u}_2 = \frac{\lambda}{m} \left( \frac{1}{6} \tilde{p}_{0zz} y^3 + \frac{1}{2} A_z(z, t) y^2 \right), \quad (26)$$

where the no-slip condition  $\tilde{u}_2(y = 0, z, t) = 0$  has been used again. Next, we use the normal stress balance (7) and tangential stress balance (8), to obtain, to leading order,

$$\tilde{p}_0 = \frac{J}{\lambda^2} (H + H_{zz}) \quad (27)$$

and

$$m \tilde{w}_{2y}(1, z, t) + \tilde{u}_{1z}(1, z, t) + \tilde{w}_{1r}(1, z, t) = \frac{\beta}{C_a \varepsilon^2} \Gamma_z. \quad (28)$$

Consider  $\bar{w}_2$  at the interface

$$\bar{w}_2|_{r=1+\varepsilon^2 H} \sim \frac{(2+\varepsilon)\varepsilon}{m+2\varepsilon+\varepsilon^2} - \frac{2\varepsilon^2 H}{m+2\varepsilon+\varepsilon^2}. \quad (29)$$

where  $\bar{w}_{2\varepsilon} = \frac{(2+\varepsilon)\varepsilon}{m+2\varepsilon+\varepsilon^2}$ . From the kinematic condition (16) and using the Galilean transformation (17), we obtain to leading order

$$\tilde{u}_2 = H_\tau - \frac{2}{m} H H_z. \quad (30)$$

Now, consider  $\bar{w}_1$  at the interface

$$\bar{w}_1|_{r=1+\varepsilon^2 H} \sim \frac{(2+\varepsilon)\varepsilon}{m+2\varepsilon+\varepsilon^2} - \frac{2m\varepsilon^2 H}{m+2\varepsilon+\varepsilon^2} \quad (31)$$

and due to continuity of velocities at the interface, we obtain from (31) and (29),

$$-2H + \tilde{w}_1|_{r=1+\varepsilon^2 H} = -\frac{2}{m}H. \quad (32)$$

Therefore, we conclude from continuity of axial velocities,  $w_1 = w_2$ , and radial velocities,  $u_1 = u_2$ , at the interface,  $r = 1$  to leading order, that

$$\tilde{w}_1|_{r=1} = 2H \left(1 - \frac{1}{m}\right) \quad \text{and} \quad \tilde{u}_1|_{r=1} = 0. \quad (33)$$

$A(z, \tau)$  still unknown. Solving the core problem, we can use the normal stress balance (7) to find  $A(z, \tau)$ . Substitution of the core variables into the governing equations (3), (4), and (5), and considering  $Re_1 = \lambda\varepsilon$ , gives the following leading order core problem

$$\lambda\tilde{p}_{1r} = \nabla^2\tilde{u}_1 - \frac{\tilde{u}_1}{r^2} \quad (34)$$

$$\lambda\tilde{p}_{1z} = \nabla^2\tilde{w}_1 \quad (35)$$

$$\frac{1}{r}(r\tilde{u}_1)_r + \tilde{w}_{1z} = 0.$$

Let us introduce the streamfunction  $\psi$  as

$$\tilde{u}_1 = -\frac{1}{r}\psi_z \quad \text{and} \quad \tilde{w}_1 = \frac{1}{r}\psi_r. \quad (36)$$

Thus,

$$\tilde{u}_{1z} = -\frac{1}{r}\psi_{zz} \quad (37)$$

$$\tilde{w}_{1r} = -\frac{1}{r^2}\psi_r + \frac{1}{r}\psi_{rr}. \quad (38)$$

Differentiating (25) with respect to  $y$ , gives

$$\tilde{w}_{2y} = \frac{\lambda}{m}(y\tilde{p}_{0z} + A(z, t)). \quad (39)$$

Since  $C_a = \frac{Re_1}{J}$ , we have, in this regime,  $C_a \sim \varepsilon$ . Considering  $\beta = \varepsilon^3\beta_0$  and  $C_a = \varepsilon\bar{C}_a$ , and substituting (37), (38), (39) into the tangential stress balance equation (28), yields

$$\lambda\tilde{p}_{0z} + \lambda A(z, \tau) = \psi_{zz} + \psi_r - \psi_{rr} + \frac{\beta_0}{\bar{C}_a}\Gamma_z. \quad (40)$$

The choice  $\beta = \varepsilon^3 \beta_0$  is made in order to keep Marangoni effects in the leading order evolution equations.

On the other hand, substituting (36) into (34) and (35), differentiating (34) with respect to  $z$  and (35) with respect to  $r$ , and eliminating  $\hat{p}_1$ , we obtain the creeping flow equation

$$\left( \frac{\partial^2}{\partial r^2} - \frac{1}{r} \frac{\partial}{\partial r} + \frac{\partial^2}{\partial z^2} \right)^2 \psi = 0. \quad (41)$$

This is consistent with the fact that  $Re_1$  is order  $\varepsilon$ . The solution of the creeping flow equation (41) is most easily accomplished in Fourier space, and is

$$\hat{\psi} = C_1(k)rI_1(kr) + C_2(k)r^2I_0(kr), \quad (42)$$

where  $\hat{\psi} = \int_{-\infty}^{\infty} \psi(r, z)e^{-ikz} dz$  is the Fourier transform of  $\psi$  and  $I_0, I_1$  are the modified Bessel functions of order zero and one, respectively.  $C_1(k)$  and  $C_2(k)$  are two functions independent from  $r$  and  $z$ , to be found.

Differentiating (42) with respect to  $r$  and plugging  $\hat{\psi}_r$  and  $\hat{\psi}_{rr}$  into the Fourier transform of (40), we find that

$$\hat{A} = -\frac{2k}{\lambda}(kC_1 + C_2)I_1(k) - \frac{2C_2k^2}{\lambda}I_0(k) + \frac{ik\beta_0}{\lambda\bar{C}_a}\hat{\Gamma} - ik\hat{p}_0 \quad (43)$$

Taking the Fourier transform of (26) and plugging (43) into it, we have:

$$\begin{aligned} \hat{u}_2 &= \frac{\lambda k^2}{3m} \left( 1 - \frac{1}{3}y \right) y^2 \hat{p}_0 - \frac{ik^2(C_1k + C_2)}{m} y^2 I_1(k) \\ &- \frac{ik^3 C_2}{m} y^2 I_0(k) - \frac{k^2 \beta_0}{2m\bar{C}_a} y^2 \hat{\Gamma}. \end{aligned} \quad (44)$$

In order to find  $C_1(k)$  and  $C_2(k)$ , we consider the Fourier transform of (36)

$$\hat{\psi} = \frac{ir}{k} \int_{-\infty}^{\infty} \tilde{u}_1 e^{-ikz} dz \quad \text{and} \quad \hat{\psi}_r = r \int_{-\infty}^{\infty} \tilde{w}_1 e^{-ikz} dz. \quad (45)$$

Evaluating  $\hat{\psi}$  and  $\hat{\psi}_r$  at the interface and using (33), we obtain, taking the leading order terms,

$$\hat{\psi}(r=1) = 0 \quad \text{and} \quad \hat{\psi}_r(r=1) = 2 \left( 1 - \frac{1}{m} \right) \hat{H}, \quad (46)$$

respectively. Thus, we find that  $C_1(k) = -I_0(k)F(k)\hat{H}(z, t)$  and  $C_2(k) = I_1(k)F(k)\hat{H}(z, t)$ , where

$$F(k) = \frac{2(1 - \frac{1}{m})}{kI_1^2(k) - kI_0^2(k) + 2I_0(k)I_1(k)}. \tag{47}$$

Evaluating (44) at  $r = y = 1$  (the undisturbed interface), we obtain

$$\hat{u}_2|_{y=1} = \frac{\lambda k^2}{3m} \hat{p}_0 - \frac{2ik^2 I_1^2(k)}{m(kI_1^2 - kI_0^2 + 2I_0 I_1)} \left(1 - \frac{1}{m}\right) \hat{H} - \frac{k^2 \beta_0}{2m\bar{C}_a} \hat{\Gamma}. \tag{48}$$

Let us define  $d = kI_1^2(k) - kI_0^2(k) + 2I_0(k)I_1(k)$  and  $N(k) = \frac{k^2 I_1^2(k)}{d}$ . On the other hand, we know that  $k^2 \hat{p}_0 = -\widehat{\tilde{p}_{0zz}}$  and  $k^2 \hat{\Gamma} = -\widehat{\Gamma_{zz}}$ . Thus,

$$\hat{u}_2|_{y=1} = -\frac{\lambda}{3m} \widehat{\tilde{p}_{0zz}} - \frac{2i}{m} \left(1 - \frac{1}{m}\right) N(k) \hat{H} + \frac{\beta_0}{2m\bar{C}_a} \widehat{\Gamma_{zz}}. \tag{49}$$

Applying inverse Fourier transform and substituting (27) and (30) into the last equation, we find the **interface evolution equation**

$$\begin{aligned} H_\tau &- \frac{2}{m} H H_z + \frac{i}{m\pi} \left(1 - \frac{1}{m}\right) \int_{-\infty}^{\infty} N(k) \int_{-\infty}^{\infty} H(z, \tau) e^{ik(z-\bar{z})} d\bar{z} dk \\ &+ \frac{J}{3m\lambda} (H + H_{zz})_{zz} - \frac{\beta_0}{2m\bar{C}_a} \Gamma_{zz} = 0. \end{aligned} \tag{50}$$

The intergral term represents the influence of viscosity stratification, and when  $m = 1$ , that term disappears. Note that the equation (50) (without the surfactant diffusion and integral terms) is known as the Kuramoto-Sivashinsky equation (Frenkel, Babchin, Levich, Shlang, and Sivashinsky [7], 1987; Papageorgiou, Maldarelli, and Rumschitzki [25], 1990 and Smyrlis and Papageorgiou [31], 1996).

### 3.2 Derivation of the Concentration of Surfactant Evolution Equation

Consider the non-dimensional concentration of surfactant equation (6). Suppose  $Pe \sim O(1/\varepsilon^2)$  and substitute (18) and (19) into (6). Evaluation of  $\bar{w}_2$  at  $r = 1 + \varepsilon^2 H$ , gives, neglecting high order terms,

$$\Gamma_t + \bar{w}_{2\varepsilon} \Gamma_z - \frac{2\varepsilon^2}{m} (H\Gamma)_z - \frac{\varepsilon^2}{Pe} \Gamma_{zz} = 0, \tag{51}$$

where  $\tilde{P}e = \varepsilon^2 Pe \sim O(1)$ . Using the Galilean transformation (17), we finally obtain, dropping  $\sim$  symbol, the **concentration of surfactant evolution equation**

$$\Gamma_\tau - \frac{2}{m}(H\Gamma)_z - \frac{1}{Pe}\Gamma_{zz} = 0. \quad (52)$$

This coupled system, (50) and (52), of evolution equations constitutes an initial value problem for  $H$  and  $\Gamma$ , which has to be addressed numerically, in general.

## 4 Re-scaling to Canonical Form and Analytical Properties

### 4.1 Re-scaling to canonical form

We want to go from domains of length  $2L$  to domains of length  $2\pi$ . We redefine  $H \rightarrow \alpha H$ ,  $\Gamma \rightarrow \gamma \Gamma$ , and  $\frac{\partial}{\partial \tau} \rightarrow \beta \frac{\partial}{\partial t}$ , where  $\alpha$ ,  $\beta$ , and  $\gamma$  are chosen to make as many coefficients as possible in the nonlinear equations equal to one.

Thus, we obtain the re-scaled coupled system of the interface and concentration of surfactant evolution equations

$$\begin{aligned} H_t + HH_z &+ i \frac{3\lambda}{\pi\nu J} \left(1 - \frac{1}{m}\right) \int_{-\infty}^{\infty} N(\sqrt{\nu}\tilde{k}) \int_{-\infty}^{\infty} H(z,t) e^{i\tilde{k}(z-\tilde{z})} d\tilde{z} d\tilde{k} \\ &+ (H + \nu H_{zz})_{zz} = -\Gamma_{zz} \end{aligned} \quad (53)$$

and

$$\Gamma_t + (H\Gamma)_z = \frac{3m\lambda}{PeJ}\Gamma_{zz}. \quad (54)$$

### 4.2 Analytical Properties

In this section, we start calculating the canonical form of the perturbed axial velocity  $W_2$ . Next, we show that the volume of fluid and the amount of surfactants are conserved. We devote the last part of this section to the linear analysis of the system (53) and (54), when ( $m = 1$ )

$$H_\tau + HH_z + H_{zz} + \nu H_{zzzz} = -\Gamma_{zz} \quad (55)$$

$$\Gamma_\tau + (H\Gamma)_z = \eta \Gamma_{zz}, \quad (56)$$

where  $\eta = \frac{3\lambda}{PeJ}$ .



**4.2.1 Canonical form of the perturbed axial velocity  $W_2$ :**

Here, we calculate  $W_2$  which is the canonical form of  $\tilde{w}_2$ . Evaluating equation (25) at  $y = 1$  and considering  $m = 1$ , we know that

$$\tilde{w}_2|_{y=1} = \lambda \left( \frac{1}{2} \tilde{p}_{0z} + A(z, t) \right). \tag{57}$$

Since  $C_1 = C_2 = 0$  when  $m = 1$ , equation (43) becomes

$$\hat{A} = \frac{ik\beta_0}{\lambda C_a} \hat{\Gamma} - ik\hat{p}_0. \tag{58}$$

Applying the inverse Fourier transform and plugging it in (57), we obtain

$$\tilde{w}_2|_{y=1} = \lambda \left( -\frac{1}{2} \tilde{p}_{0z} + \frac{\beta_0}{\lambda C_a} \Gamma_z \right). \tag{59}$$

Taking the leading order of the equation (27) and plugging it in equation (59), yields

$$\tilde{w}_2 = -\frac{J}{2\lambda} (H + H_{zz})_z + \frac{\beta_0}{C_a} \Gamma_z. \tag{60}$$

Re-scaling  $\tilde{w}_2$  to canonical form  $W_2$ , we use the same re-scaled variables as in §4.1 and obtain

$$W_2 = -\frac{J^2\nu}{12\lambda^2} \left[ (H + \nu H_{zz})_z + \frac{4}{3} \Gamma_z \right]. \tag{61}$$

**4.2.2 Conserved quantities:**

We start considering the system (55) and (56), then

$$\frac{d}{d\tau} \left( \int_0^{2\pi} H dz \right) = \int_0^{2\pi} H_\tau dz \tag{62}$$

$$= - \int_0^{2\pi} (\Gamma_{zz} + HH_z + H_{zz} + \nu H_{zzzz}) dz = 0 \tag{63}$$

because of periodicity of  $H$  at the boundaries. Therefore,

$$\int_0^{2\pi} H dz = \text{constant} = 0 \tag{64}$$

if  $H$  has zero mean initially.

$$\frac{d}{d\tau} \left( \int_0^{2\pi} \Gamma dz \right) = \int_0^{2\pi} \Gamma_\tau dz \quad (65)$$

$$= - \int_0^{2\pi} (\eta \Gamma_{zz} - (H\Gamma)_z) dz \quad (66)$$

$$= -\Gamma_0 \int_0^{2\pi} H_z dz = 0 \quad (67)$$

because  $\Gamma = \Gamma_0$  is a constant and because of periodicity of  $H$  at the boundaries. Therefore,

$$\int_0^{2\pi} \Gamma dz = \text{constant} = \Gamma_0 \int_0^{2\pi} dz = 2\pi\Gamma_0. \quad (68)$$

### 4.2.3 Linear stability:

We consider the undisturbed state

$$H = 0, \quad \Gamma = \Gamma_0, \quad 0 < \Gamma_0 < 1 \quad (69)$$

and take normal modes (Drazin and Reid [3], 1999) in the form

$$H = \hat{H} e^{ikz + \omega t}, \quad \Gamma = \Gamma_0 + \hat{\Gamma} e^{ikz + \omega t}, \quad (70)$$

where  $k$  is the wave number and  $\omega$  is the growth rate. Substituting (70) in (55) and (56) and retaining only linear terms, we obtain

$$\begin{aligned} \omega \hat{H} - k^2 \hat{H} + k^4 \nu \hat{H} &= k^2 \hat{\Gamma} \\ \omega \hat{\Gamma} + ik \hat{H} \Gamma_0 &= -k^2 \eta \hat{\Gamma}. \end{aligned}$$

Grouping  $\hat{H}$  terms together and  $\hat{\Gamma}$  terms together, yields the system

$$\begin{aligned} (\omega - k^2 + \nu k^4) \hat{H} &= k^2 \hat{\Gamma} \\ ik \Gamma_0 \hat{H} &= -(\omega + \eta k^2) \hat{\Gamma}, \end{aligned}$$

which we solve to obtain

$$(\omega - k^2 + \nu k^4)(\omega + \eta k^2) = -ik^3 \Gamma_0. \quad (71)$$

Rewriting the last equation we obtain a quadratic equation for  $\omega$

$$\omega^2 + (\eta k^2 - k^2 + \nu k^4) \omega + \eta \nu k^6 - \eta k^4 + ik^3 \Gamma_0 = 0, \quad (72)$$

which gives two complex values of the growth rate  $\omega_1$  and  $\omega_2$

$$\omega_{1,2} = -\frac{1}{2}(\eta - 1 + \nu k^2)k^2 \pm \sqrt{\frac{1}{4}(\eta - 1 + \nu k^2)^2 k^4 - \eta k^4(\nu k^2 - 1) - ik^3 \Gamma_0}.$$

Next, we consider  $\Gamma_0 = 1$  and  $\eta = 1$  and compute the values of  $\omega_1$  and  $\omega_2$  for a range of values of the wave number  $k$ .

Considering the growth rate,  $Re(\omega_1)$ , we can see that it takes positive and negatives values. A cutoff wave number  $k_\nu$  exists for each  $\nu$  that we studied here. This indicates that for each  $\nu$ ,  $Re(\omega_1) < 0$  for  $k > k_\nu$ . On the other hand, all the values of the growth rate,  $Re(\omega_2)$ , are negative. Therefore, we can conclude that the solutions are linearly stable for wave numbers  $k$  bigger than  $k_\nu$ . This is consistent with the short wave stabilization supported by the Kuramoto-Sivashinsky equation.

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