DYNAMICS OF A REACTIVE FALLING FILM AT LARGE PÉCLET NUMBERS

Serafim Kalliadasis, Philip Trevelyan
Department of Chemical Engineering, University of Leeds, Leeds, LS2 9JT, UK

Summary We study the dynamics of a vertically falling film in the presence of a first-order (exothermic or endothermic) chemical reaction. We extend the work by Trevelyan *et al.* [1] on the same problem to large heat/mass transport Péclet numbers and so we take into account the convective terms of the heat/mass transport equations. Our analysis is based on a long-wave expansion and integral-boundary-layer theory of the equations of motion and associated boundary conditions.

FORMULATION

Figure 1 shows the problem definition. We consider a thin viscous film falling down a vertical substrate. The ambient gas phase contains a species A that is absorbed into the liquid where it undergoes a simple first-order decay, $\mathrm{A} \rightarrow \mathrm{B} \pm \mathrm{heat}$. The reaction rate is taken to be temperature dependent so that heat is released into the film or absorbed from the film. Such heat generation gives rise to thermocapillary stresses, which in turn, affect the dynamics of the free surface.

The governing equations, namely equation of motion, energy and concentration equations are

$$u_x + v_y = 0 \tag{1a}$$

$$u_{yy} + 2 = 2\epsilon p_x + \epsilon Re(u_t + uu_x + vu_y) - \epsilon^2 u_{xx} \tag{1b}$$

$$\epsilon v_{yy} = 2p_y + \epsilon^2 Re(v_t + uv_x + vv_y) - \epsilon^3 v_{xx} \tag{1c}$$

$$a_{yy} = (1 + Daa)e^{Da\beta\phi T/(1+Da\phi T)} + \epsilon ReSc(a_t + ua_x + va_y) - \epsilon^2 a_{xx} \tag{1d}$$

$$T_{yy} = -(1 + Daa)e^{Da\beta\phi T/(1+Da\phi T)} + \epsilon RePr(T_t + uT_x + vT_y) - \epsilon^2 T_{xx} \tag{1e}$$

where ϵ the film parameter, Re, Pr and Sc the Reynolds, Prandtl and Schmidt numbers, respectively, and Da the Damköhler number. These equations are subject to the boundary conditions at $y = h(x, t)$

$$p = -\epsilon^2 We(1 - MaDa\phi T)N^{-\frac{3}{2}} h_{xx} + \epsilon N^{-1}(v_y - h_x u_y + \epsilon^2(h_x^2 u_x - h_x v_x)) \tag{2a}$$

$$u_y + \epsilon^2(v_x + 2h_x(v_y - u_x) - h_x^2 u_y) - \epsilon^4 h_x^2 v_x = -2\epsilon WeMaDa\phi(T_x + h_x T_y)N^{\frac{1}{2}} \tag{2b}$$

$$a = 0 \tag{2c}$$

$$T_y - \epsilon^2 h_x T_x = 0 \tag{2d}$$

$$h_t + uh_x = v \tag{2e}$$

and the wall boundary conditions at $y = 0$

$$u = v = 0, \quad a_y = 0, \quad T = 0 \tag{3}$$

where $N = 1 + \epsilon^2 h_x^2$. The thermocapillary effect is modelled by using a linear approximation for the surface tension, $\sigma = \sigma_0 - \gamma(T - T_0)$ with σ_0 the surface tension at the reference temperature T_0 and $\gamma > 0$ for typical liquids. $We = \sigma_0/\rho g h_0^2$ is the Weber number and $Ma = \gamma T_0/\sigma_0$ is the Marangoni number.

A long-wave expansion (LWE) of the above equations for $\epsilon \ll 1$ leads to a single highly nonlinear evolution equation for the film thickness h. Here we extend the previous study by Trevelyan *et al.* on the same problem to the case of large Péclet numbers. As a consequence, the convective effects in both heat and mass transport processes are retained in our LWE formulation.

We construct the solitary wave bifurcation diagrams for a wide range of the pertinent parameters. In all cases the bifurcation diagrams exhibit two branches (a lower one and an upper one). We demonstrate the existence of non-dissipative solitary waves even away from criticality. Time-dependent computations of the fully nonlinear evolution equation show that for large times the interface approaches a train of solitary pulses similar to the lower branch solitary waves. For a given Reynolds numbers and sufficiently small Prandtl and Schmidt numbers the interface is characterized by an irregular row of solitary pulses which interact indefinitely with each other. This complex spatio-temporal behavior is similar to the large-time evolution of a vertically falling film in the absence of Marangoni effects. However, for a sufficiently large Schmidt number or a sufficiently large Prandtl number we observe an intriguing pattern formation process characterized by a regular train of non-dissipative solitary waves (see Fig. 2). This highly ordered state persists indefinitely. Hence, the convective effects associated with the chemical reaction induce a high degree of organization not observed before in falling film studies.

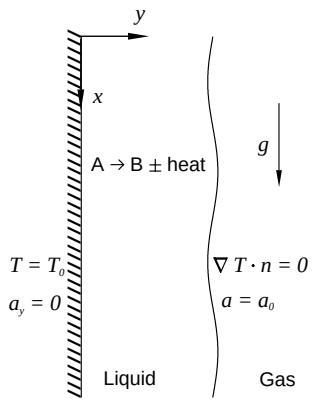

Figure 1. Sketch of the vertically falling film in the presence of a chemical reaction.

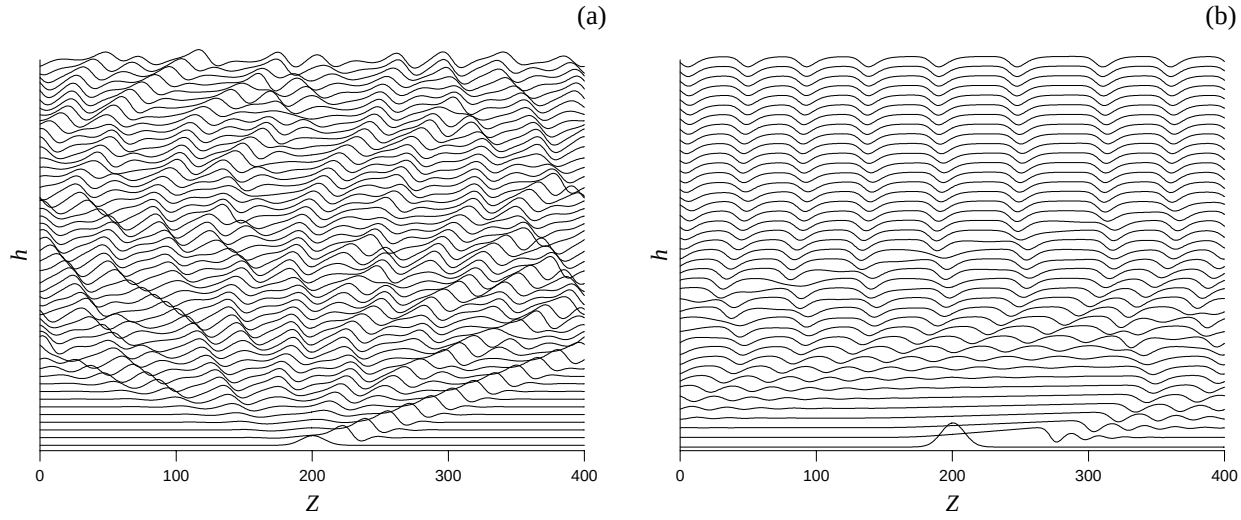

Figure 2. Time evolution of the interface in a moving frame Z for moderate (a) and large (b) heat/mass transport Péclet numbers.

Far from criticality and for moderate Reynolds numbers, the model equation of choise in the absence of Marangoni effects is the Shkadov integral-boundary-layer approximation [2]. However, the Shkadov approach predicts the critical Reynolds number with a 20% error. Here we extend this approximation to the reactive falling film problem and we show that a simple Galerkin projection for the momentum equation corrects the critical Reynolds number obtained from the Shkadov approach. In addition, we develop a hierarchy of IBL models based on high-order Galerkin projections for the concentration and temperature fields onto the set of polymonial test functions. Not only do these models correct the critical Reynolds number, but they also give close to criticality, with an appropriate gradient expansion, the full LWE. We also construct nonlinear solutions of the solitary wave type and we show that unlike LWE, our IBL models predict the continuing existence of solitary pulses for all Reynolds numbers.

CONCLUSIONS

We considered the dynamics of a reactive falling film. Our analysis was based on a long-wave expansion and integral-boundary-layer formulation. Particular emphasis was given to the solitary wave solution branches obtained from the different approaches. Our analysis also indicates that for sufficiently large Prandtl/Schmidt numbers the film can be excited in the form of non-dissipative solitary pulses which close to criticality assume the form of KdV solitons.

References

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[2] Shkadov, V.Ya.: Wave regimes in the flow of a thin layer of a viscous fluid under the action of gravity. *Izv. Akad. Nauk SSSR, Mekh. Zhidk i Gaza* **1**:43–50, (1967).