# Oscillatory flow of a two-fluid system under reduced gravity

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### **Summary**

A system of two fluids having different thermo-physical properties is considered. In a rectangular domain with differently heated side walls a discrete interface exists separating both fluids. In any case a Marangoni flow is generated by temperature differences along that interface. On earth buoyancy driven flow interacts with the thermocapillary (Marangoni) flow showing a rather complicated flow pattern. This configuration roughly describes the situation in a polymerization experiment where a monomer (fluid 2) is converted into a polymer (fluid 1). It is a step toward understanding how transient and residual g can affect polymerization and other reactive systems with sharp concentration gradients. If the experiment is done in space the convective influences can be reduced considerably but a new problem arises because there is no real zero gravity in a space station. A certain residual g level or transient impulses will affect the flow behavior permanently. In a numerical study it can be shown that a bifurcation problem exists for the given geometry, the set of equations and boundary conditions. Starting from the zero gravity case the transient simulations show that depending on the amplitude and duration of a small gravity pulse oscillatory modes are possible. The order of magnitude of the corresponding flow velocities can exceed the zero-gravity ones considerably. As a consequence of these results it should be well known which residual g can be tolerated in order to estimate its influence on a certain fluid flow experiment.

## Introduction

It is well known that temperature and/or concentration gradients perpendicular or parallel to a fluid/fluid interface can lead to convection [1]. If the gradient is perpendicular to the interface the Marangoni instability can occur [2]. There are critical conditions for the onset of convection characterized by the dimensionless Marangoni number. The Marangoni number is the analog to the Rayleigh number but in which the driving force is the variation in the interfacial tension instead of density differences. Like with the buoyancy-driven case, any lateral temperature gradient will cause convection. If the gradient is along the interface the surface-tension-induced convection will always occur. This situation is analogous to buoyancy-driven convection. For a density gradient along the gravitational vector there must be a critical value of the Rayleigh number for the onset of convection. If the fluid is heated from one side, convection will always occur. If both phenomena interact, like in the following considerations, a complex fluid flow may arise. The experiment is sketched in fig 1. As an approximation we treat that problem as immiscible. In the real problem the fluids are miscible with an effective interfacial tension (EIT) and because of diffusion the EIT is transient. However, we assume that the effective interfacial tension is a

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function of temperature in the same way as interfacial tension for immiscible fluids [3] [4] [5]. Fig. 1 shows a sharp transition zone between the polymer and the monomer - which are miscible - created by photopolymerization in a rectangular cuvette  $3 \times 6 \times 1$  cm. We illuminate the reactor with UV light using masks that generate a known interfacial profile between polymer and monomer.

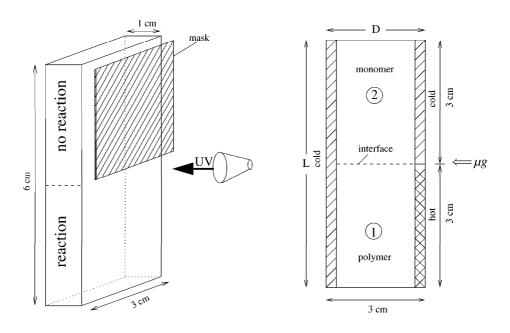


Fig. 1: Sketch of the experiment

Fig. 2: Computational domain

In fact the samples will consist of three regions. The upper region is the monomer (fluid 2), the lower region is the polymer (fluid 1). Between both there is a transition zone. In a limiting case for our numerical studies we assume the latter to be of zero width having in mind that this assumption would be "the worst case" with the maximum flow velocities.

In a microgravity environment buoyancy driven flow is reduced considerably but can not be ignored completely because of a remaining g-level and random movement in a space station. Therefore the question has to be answered what maximum g-level is tolerable for an experiment in space. Using a numerical method this answer can be given in advance.

### **Mathematical model**

A steady-state 2D model is employed to calculate the temperature distribution and the fluid flow in both parts of the domain (see fig 2.). The mathematical model for the thermal and velocity field governed by the Navier-Stokes equations with the Boussinesq approximation, the continuity equation and the energy equations reads as follows.

$$\frac{\partial u_i}{\partial t} + u_i \frac{\partial u_i}{\partial x} + v_i \frac{\partial u_i}{\partial y} = -\frac{\partial p_i}{\partial x} + \frac{\partial^2 u_i}{\partial x^2} + \frac{\partial^2 u_i}{\partial y^2}$$

$$\frac{\partial v_i}{\partial t} + u_i \frac{\partial v_i}{\partial x} + v_i \frac{\partial v_i}{\partial y} = -\frac{\partial p_i}{\partial y} + \frac{\partial^2 v_i}{\partial x^2} + \frac{\partial^2 v_i}{\partial y^2} - g \beta_i T_i$$

$$\frac{\partial u_i}{\partial x} + \frac{\partial v_i}{\partial y} = 0$$

$$\frac{\partial T_i}{\partial t} + u \frac{\partial T_i}{\partial x} + v_i \frac{\partial T_i}{\partial y} = \kappa_i \left( \frac{\partial^2 T_i}{\partial x^2} + \frac{\partial^2 T_i}{\partial y^2} \right) \qquad i = 1, 2$$

The velocities  $u_i, v_i$ , the temperature  $T_i$ , and the pressure  $p_i$  are the unknown variables. Using appropriate quantities for scaling like  $x := \frac{x}{L}$  for the length,  $u_i := \frac{u_i L}{v_1}$  for the velocities and  $T_i := \frac{T_i}{T_0}$  for the temperature the following non-dimensional numbers exist.

$$Pr = \frac{\mathbf{v}_1}{\mathbf{\kappa}_1}, \quad Ma = \frac{\left|\frac{d\sigma}{dT}\right|T_0L}{\mu_1\mathbf{v}_1}, \quad Gr = \frac{g\beta_1T_0L^3}{\mathbf{v}_1^2}, \quad Ar = \frac{D}{L}, \quad Dr = \frac{\rho_2}{\rho_1}, \quad Vr = \frac{\mathbf{v}_2}{\mathbf{v}_1}$$

The boundary conditions are prescribed temperatures at the cold ( $T_i = 0$ ) and hot ( $T_1 = T_0$ ) wall, adiabatic upper and lower walls and no slip for the fluid flow at all walls. At the interface we have continuous heat and momentum fluxes. In particular the equilibrium of tangential stress holds.

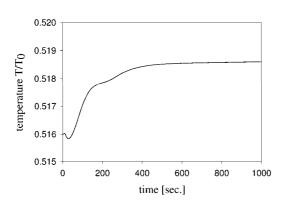
$$\frac{\partial u_1}{\partial y} - \frac{\partial u_2}{\partial y} = Ma \frac{\partial T_1}{\partial x}$$

Here L is the domain length, D the domain width, g the gravity vector. The properties of the polymer (1) and monomer (2) are  $v_i$  - the kinematic viscosity,  $\kappa_i$  - thermal diffusivity,  $\beta_i$  - linear expansion coefficient,  $\rho_i$  - density. The solution depends on the Prandtl number Pr, the Marangoni number Ma, the Grashoff number Gr, the aspect ratio Ar, the density ratio Dr and the viscosity ratio Vr. Because of only small differences of specific heat, heat conductivity and linear expansion for polymer and monomer the corresponding ratios have been taken as equal one.

The set of equations is discretized in standard Galerkin formulation of the finite-element method [6]. A graded mesh of second order finite elements was carefully designed in order to obtain a sufficiently accurate approximation of the solution. The nonlinear discrete equations are solved using a Newton-Raphson method combined with a frontal solver for the linear system arising from this method. An adaptive Gear scheme for time-stepping is used. Depending on the complexity of the flow structure up to 20000 finite elements with about 150000 degrees of freedom are required. The FEM code is running in parallel under MPI (Message Passing Interface). This has been done by implementing the MUMPS solver (MUltifrontal Massively Parallel sparse direct Solver) [7].

#### Results

For the numerical simulations the geometry of fig. 2 has been used under the assumption that the interface is not deformed. Typical parameter values for the polymer/monomer system under consideration at 1g are Pr=400, Ma=20, Gr=67000, Dr=0.91 and Vr=0.034. In a sequence of steady state and transient calculations several solutions of the problem are obtained for different g-levels. It is well known that in multiple-bounded fluid layers different convective modes are possible. This is especially true if the corresponding fluids have different viscosities like a gas/fluid system [8].



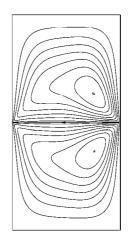


Fig. 3: Time plot of temperature at mid interface position for  $10^{-5}g$ 

Fig. 4: Streamlines for  $10^{-5}g$  in the monomer (above) and polymer (below)

In the case of strong interactions of surface tension and buoyancy a Hopf bifurcation can occur [9] generating an oscillatory convection. In a materials processing application the influence of lateral walls, i.e. the aspect ratio has been investigated [10]. However, in all these examples the temperature gradients orientation is initially perpendicular to the interface, and 1g buoyancy is in effect.

In our current problem as mentioned above we are interested in the convective behavior under reduced gravity with the initial temperature gradient parallel to the interface. Therefore no critical conditions (trivial solution) exist and convection caused by thermocapillary and buoyancy forces sets in immediately.

Starting with  $10^{-5}g$  the problem has been solved by increasing the remaining g-level step by step until  $10^{-1}g$ . In all of these examples we performed a transient and steady state run as well. Fig. 3 shows the time plot of a characteristic temperature after suddenly jumping from zero to  $10^{-5}g$ . It turns out that only one steady-state solution exists for this slightly higher g-level. The corresponding streamline pattern (fig. 3) indicates an almost symmetric flow field dominated by surface tension induced convection. Increasing the residual g to  $10^{-4}g$  changes the result considerably in its nature. A transient calculation shows a non-damped oscillatory behavior (fig. 5) whereas the steady state problem has a different solution indicating a bifurcation since level  $10^{-5}g$ .

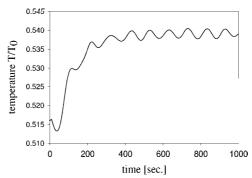


Fig. 5: Time plot of temperature at mid interface position for  $10^{-4}$ g

In this case the fluctuations of temperature and/or flow velocities are small and may not be recognized in an associated experiment.

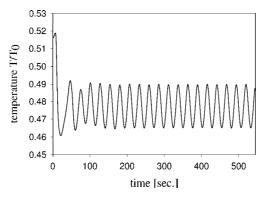
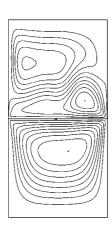


Fig. 6: Time plot of temperature at mid interface position for  $10^{-3}g$ 

Looking at the next step of residual gravitation  $10^{-3}g$  the oscillation amplitudes have increased dramatically (fig. 6) and the thermal and momentum interaction of both fluids has changed its character because the flow is now dominated by buoyancy. The more viscous fluid (polymer) rises along the hot wall and forms a typical bulk flow there. In the upper part (monomer) a competition takes place between surface tension and buoyancy and forms a complicated two-eddy flow field which is not stable in time. Fig. 7 shows that situation like a snapshot at a certain time position where the oscillation is fully developed. However, even in this case we found a steady state solution showing that the two branches of the bifurcation still exist.



The multilayer fluid flow interaction leading to transient fluctuations has already reported in several papers [8] [9] [10] in cases where g-vector and driving T-gradient have the same orientation. The current approach makes evident that also in situations where these orientations are different time-dependent solutions - even for reduced gravity - are possible.

Fig. 7: Streamlines for  $10^{-3}g$  in the monomer (above) and polymer (below)

#### Conclusions

The photopolymerization experiment under consideration can be done in space if the residual gravity does not exceed the level of  $10^{-5}g$ . Because the current numerical model does not reflect the real behavior of a miscible fluid where we can expect smaller driving forces a level of  $10^{-4}g$  may be acceptable. However, also in the case of a highly viscous fluid like a polymer convective effects should be considered even in space.

# Reference

- 1. Nagy, N.P., Sike, L., Pojman, J.A., Adv. Mat., 7 (1995) pp. 1038-1040.
- 2. Pojman, J.A., West W.W., J. Chem. Ed., 73 (1996) 35.
- 3. Joseph, D. D., Eur. J. Mech., B/Fluids, 9 (1990) pp. 565-596.
- 4. May, S. E., Maher, J. V., Phys. Rev. Ltts., 67 (1991) pp. 2013-2015.
- 5. Pojman, J., Volpert, V., Dumont, T., Chekanov, Y., Masere, J., Wilke, H., "Effective Interfacial Tension Induced Convection (EITIC) in miscible fluids", *AIAA* 2001-0764, 39th Aerospace Sciences Meeting, (2001).
- 6. Cliffe, K.A., ENTWIFE, www.sercoassurance.com/entwife/.
- 7. Amestoy, P.R., Duff, I.S. et al., www.enseeiht.fr/lima/apo/MUMPS/.
- 8. Golovin, A.A., Nepomnyashchy, A.A., Pismen, L.M., J. Fluid Mech., 341 (1997).
- 9. Johnson, D., Narayanan, R., Phil. Trans R. Soc. Lon. A, 356 (1998) pp. 885-898.
- 10. Narayanan, R., Johnson, D., CHAOS, 9 (1999) pp. 124-140.