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## THE DYNAMICS OF MISCIBLE INTERFACES: A SPACE FLIGHT EXPERIMENT

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### ABSTRACT

Experiments as well as accompanying simulations are described that serve in preparation of a space flight experiment to study the dynamics of miscible interfaces. The investigation specifically addresses the importance of both nonsolenoidal effects as well as nonconventional Korteweg stresses in flows that give rise to steep but finite concentration gradients.

The investigation focuses on the flow in which a less viscous fluid displaces one of higher viscosity and different density within a narrow capillary tube. The fluids are miscible in all proportions. An intruding finger forms that occupies a fraction of the total tube diameter. Depending on the flow conditions, as expressed by the Peclet number, a dimensionless viscosity ratio, and a gravity parameter, this fraction can vary between approximately 0.9 and 0.2. For large  $Pe$  values, a quasi-steady finger forms, which persists for a time of  $O(Pe)$  before it starts to decay, and Poiseuille flow and Taylor dispersion are approached asymptotically. Depending on the specific flow conditions, we observe a variety of topologically different streamline patterns, among them some that leak fluid from the finger tip. For small  $Pe$  values, the flow decays from the start and asymptotically reaches Taylor dispersion after a time of  $O(Pe)$ .

Comparisons between experiments and numerical simulations based on the 'conventional' assumption of solenoidal velocity fields and without Korteweg stresses yield poor agreement as far as the  $Pe$  value is concerned that distinguishes these two regimes. As one possibility, we attribute this lack of agreement to the disregard of these terms. An attempt is made to use scaling arguments in order to evaluate the importance of the Korteweg stresses and of the assumption of solenoidality. While these effects should be strongest in absolute terms when steep concentration fronts exist, i.e. at large  $Pe$ , they may be relatively most important at lower values of  $Pe$ . We subsequently compare these conventional simulations to more complete simulations that account for nonvanishing divergence as well as Korteweg stresses. While the exact value of the relevant stress coefficients are not known, ballpark numbers do exist, and their use in the simulations indicates that these stresses may indeed be important. We plan to evaluate these issues in detail by means of comparing a space experiment with corresponding simulations, in order to extract more accurate Korteweg stress coefficients, and to confirm or deny the importance of such stresses.

# **Dynamics of Miscible Interfaces**

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- **Introduction and background**
  - **effects of non-vanishing divergence**
  - **non-conventional stresses**
- **Principle experiment: Miscible flow in a capillary tube**
- **Simulations**
- **Preliminary conclusions**

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## **Introduction and background**

- **Many applications give rise to steep concentration gradients in the ‘miscible interface’ region that separates miscible liquids:**
  - **mixing devices**
  - **chemical reactors**
  - **materials processing applications**
  - **biology and biomedical applications**
  - **enhanced oil recovery**
  - **....**
- **Conventional analysis of such flows is based on**
  - **divergence free velocity field**
  - **Stokes or Navier-Stokes equations with standard stress tensor**
- **How accurate is this approach?**

**a) Divergence effects:**

\* **continuity equation**  $\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \vec{u}) = 0$

→  $\frac{D\rho}{Dt} + \rho \nabla \cdot \vec{u} = 0$

\* **conventional assumption**

$$\frac{D\rho}{Dt} = 0 \rightarrow \nabla \cdot \vec{u} = 0$$

**but: when the density of a fluid particle can change as a result of diffusion**

$$\frac{D\rho}{Dt} \neq 0 \rightarrow \nabla \cdot \vec{u} \neq 0$$

**this also leads to modifications in the conventional Stokes or Navier Stokes equations**

**→ How important are these divergence effects in real world applications?**

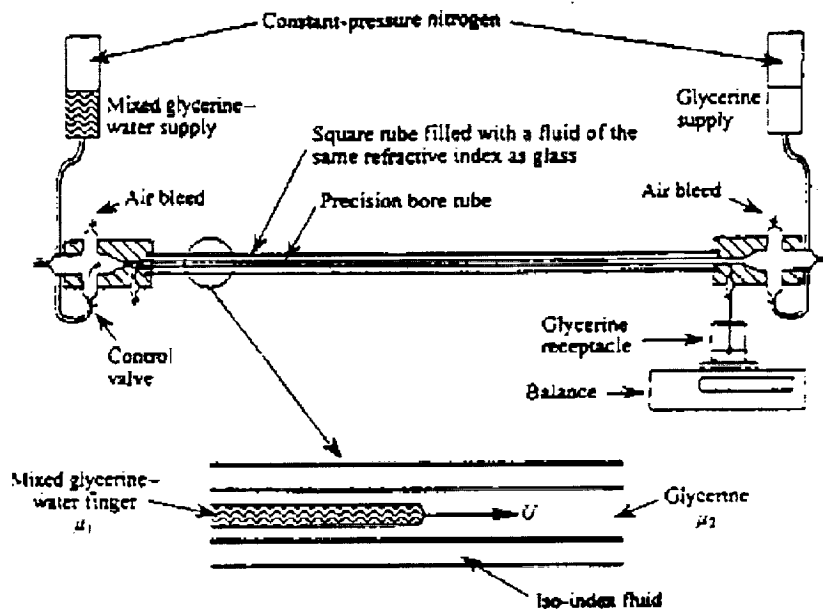
**b) Non-conventional stresses:**

- **Korteweg (1901): additional stresses could potentially be important in regions of large concentration gradients ('Korteweg stresses')**
  
- **Mathematical form of these stress terms?**
  
- **Suggestions by Davis (1988), Joseph et al. (1996)**
  - **how can the mathematical form be validated?**
  
  - **what are the signs and numerical values of the stress coefficients?**
  
  - **estimates by Smith et al. (1981), Davis (1988), Petitjeans and Maxworthy(1996):**  
**~1 dyn/cm, but time-dependent**

**Indications that these stresses may be important in many applications, e.g.:**

- **Joseph and Renardy (1993): drop of glycerin in water (miscible), drop shape is similar to that of immiscible drops**
- **Hu and Joseph (1992): miscible displacement in a Hele-Shaw cell, evidence of an effective surface tension?**
- **Petitjeans and Maxworthy (1996), Chen and Meiburg (1996): displacement of glycerin by glycerin/water mixture in a capillary tube: discrepancy between flow visualization and simulations based on Stokes equation and non-divergent velocity field**
- **additional attempts to extract 'effective surface tension': Kurowski and Misbah (1994), Petitjeans (1996), Petitjeans and Kurowski (1997)**

## Principle experiment: Miscible flow in a capillary tube



**Experiments: Petitjeans & Maxworthy (1996)**

**'Conventional' simulations: Chen & Meiburg (1996)**

**Main feature:**

**A finger of the injected, less viscous fluid propagates along the centerline of the tube, leaving behind a film of the resident fluid on the wall.**

## Experimental and numerical observations:

- **Miscible displacements: no truly steady state, eventually Taylor dispersion is approached**
- **Large Pe: a quasisteady state evolves for the finger.**

**Tip velocity and fraction of more viscous fluid left on the wall can be measured. They depend on**

\* **Peclet number**  $Pe = \frac{Ud}{D}$

\* **viscosity ratio**  $At = \frac{\mu_2 - \mu_1}{\mu_2 + \mu_1}$

\* **density ratio**  $F = g \frac{\Delta\rho}{\rho} \frac{d^2}{\nu U}$

- **Small Pe: no quasisteady finger forms. Finger quickly decays.**

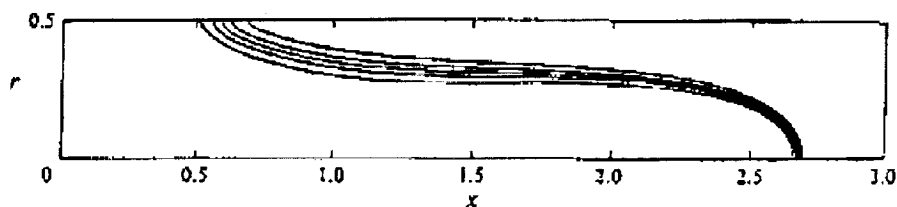


## Preliminary simulations

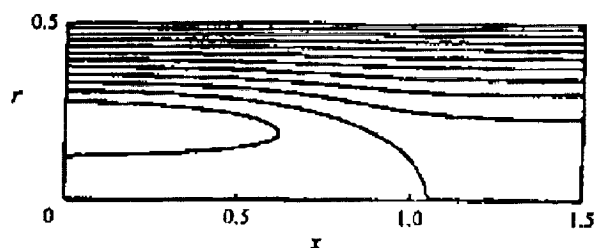
from Chen and Meiburg (1996), non-divergent,  
conventional Stokes eqns. ( $Pe=1,600$ ,  $At\sim 100$ ,  $F=0$ )

'standard' case:

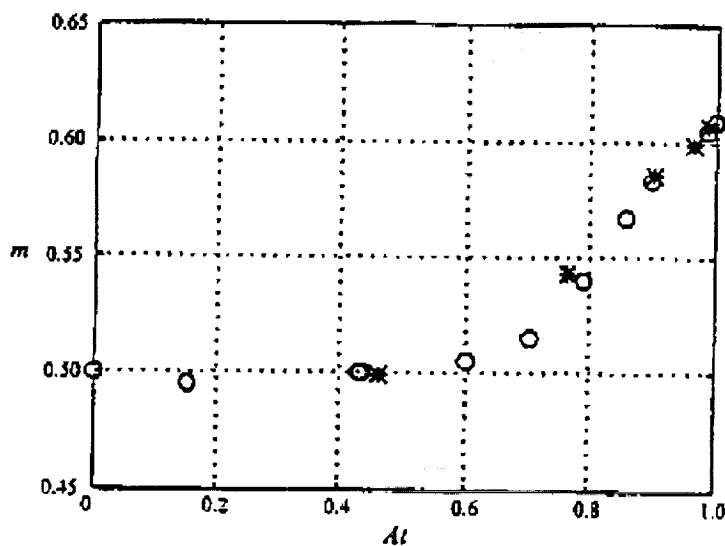
concentration



streamlines



thickness of fluid film left on the wall:



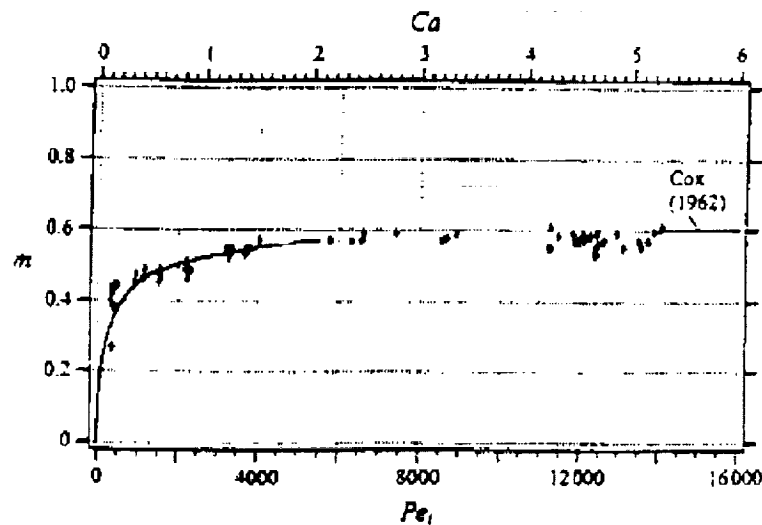
o exp.  
\* sim.

good agreement experiments/simulations

## Experiment (Petitjeans and Maxworthy 1996):

measure fraction of more viscous fluid left behind

on the wall as function of  $Pe$

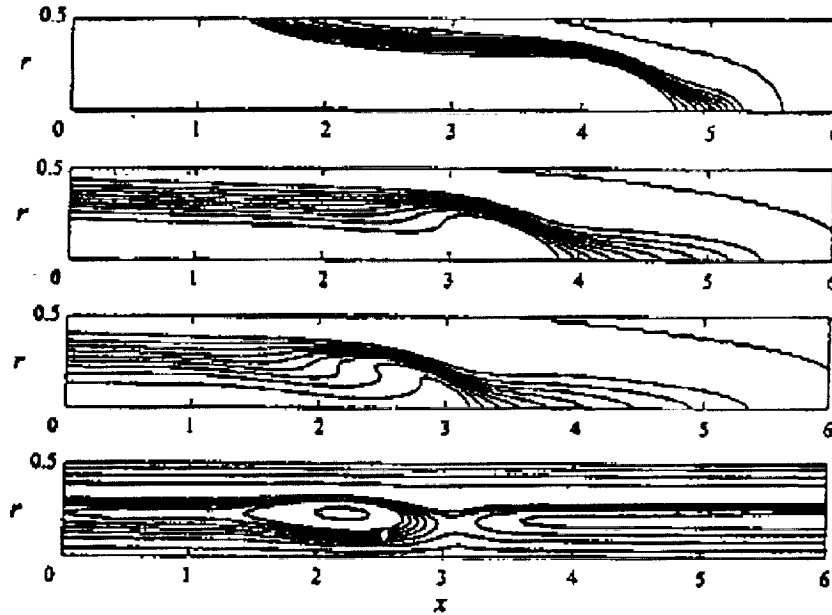


Comparison with immiscible data of Taylor (1961)

and Cox (1962) suggests value for an effective

surface tension coefficient

## formation of spike:



## concentration field

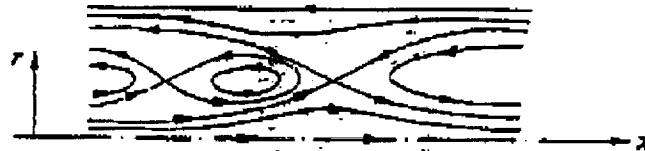
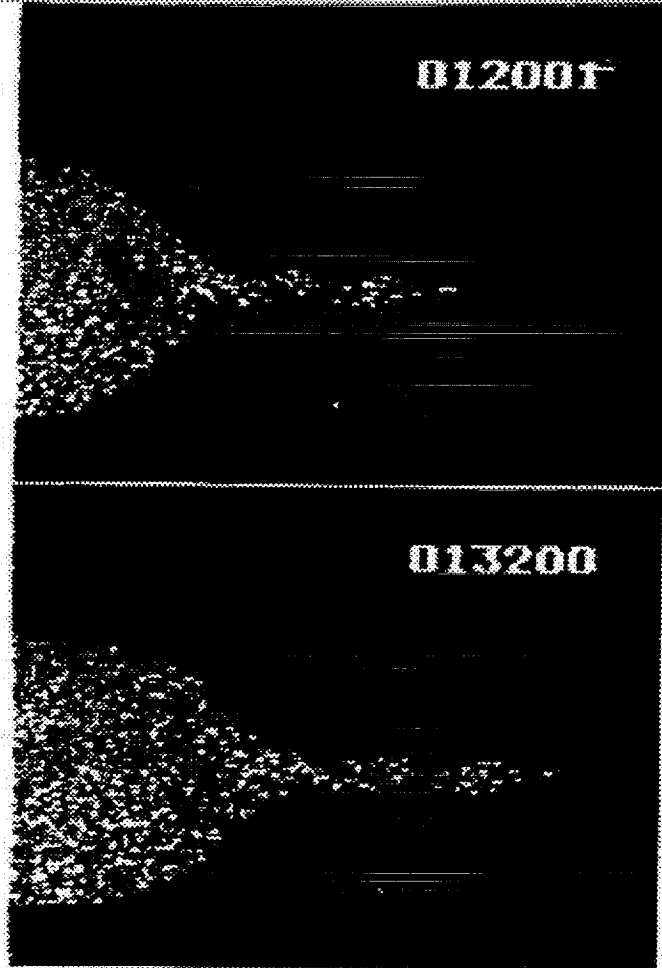


FIGURE 16. (a) Unsteady evolution of the concentration field for  $R = 3$ ,  $F = -49.78$ , and  $Pe = 1600$ . Times are  $t = 4, 12$ , and  $18$ . Also shown is the streamline pattern in the reference frame moving with the  $c = 0.5$  contour at  $t = 18$ , as well as a sketch of its topology (b). A closed, toroidal recirculation region forms that leads to a 'pinch-off'-like phenomenon near the finger tip.

## streamline topology



- **Low Pe: discrepancy between experiments and numerical simulations: Experiments show quasisteady finger at substantially lower values of Pe than the numerical simulations**

### **Indication of nonconventional stresses?**

**Note: these stresses should be largest when concentration gradients are steep (large Pe), but they could be *relatively* more important at lower values of Pe.**

preliminary simulations to evaluate the influence of divergence and Korteweg stresses:

\* perform simulations based on Joseph's suggestions for the form of the governing equations:

- split velocity field into solenoidal part and divergent part

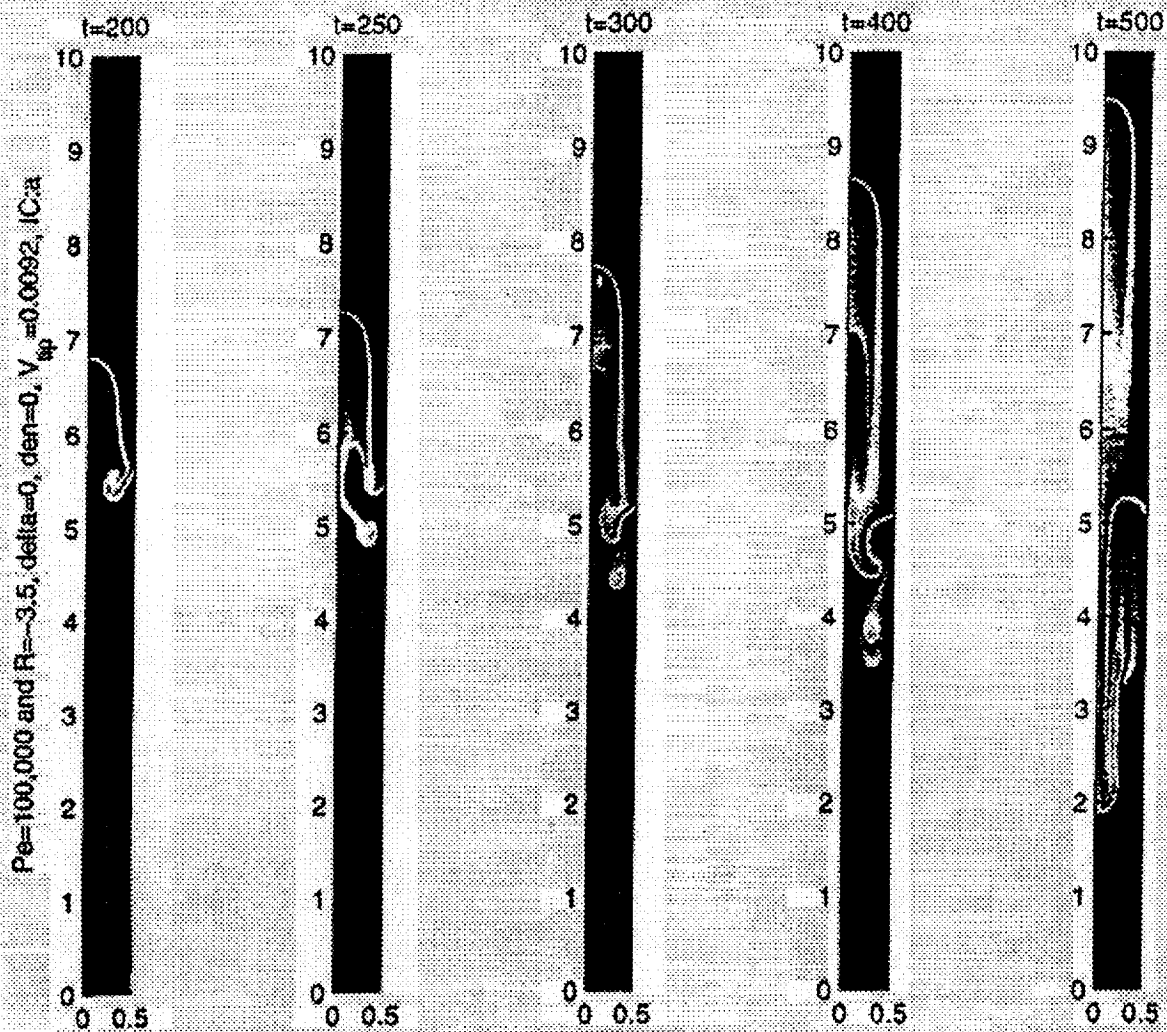
- postulate Korteweg stress terms with coefficients of unknown size

$$\begin{aligned} \frac{\partial \phi}{\partial t} + W_x \frac{\partial \phi}{\partial x} + W_r \frac{\partial \phi}{\partial r} &= D \nabla^2 \phi \\ \nabla(p + Q(\phi)) &= \nabla \cdot (2\mu D [\vec{u}] + \delta (\nabla \phi \otimes \nabla \phi)) + \rho g \\ Q(\phi) &= \frac{1}{3} \delta |\nabla \phi|^2 + \frac{2}{3} \xi \mu D \nabla^2 \phi - \frac{2}{3} \gamma \nabla^2 \phi \\ \frac{\mu}{\mu_1} &= e^{R\phi} \\ \vec{u} &= \vec{W} + \vec{u}_e \\ \nabla \cdot \vec{W} &= 0 \\ \vec{u}_e &= \xi D \nabla \phi \\ \rho &= \frac{\rho_1}{\rho_1 - \rho_2} - \phi \\ \xi &= \frac{\rho_1 - \rho_2}{\rho_1} \end{aligned}$$

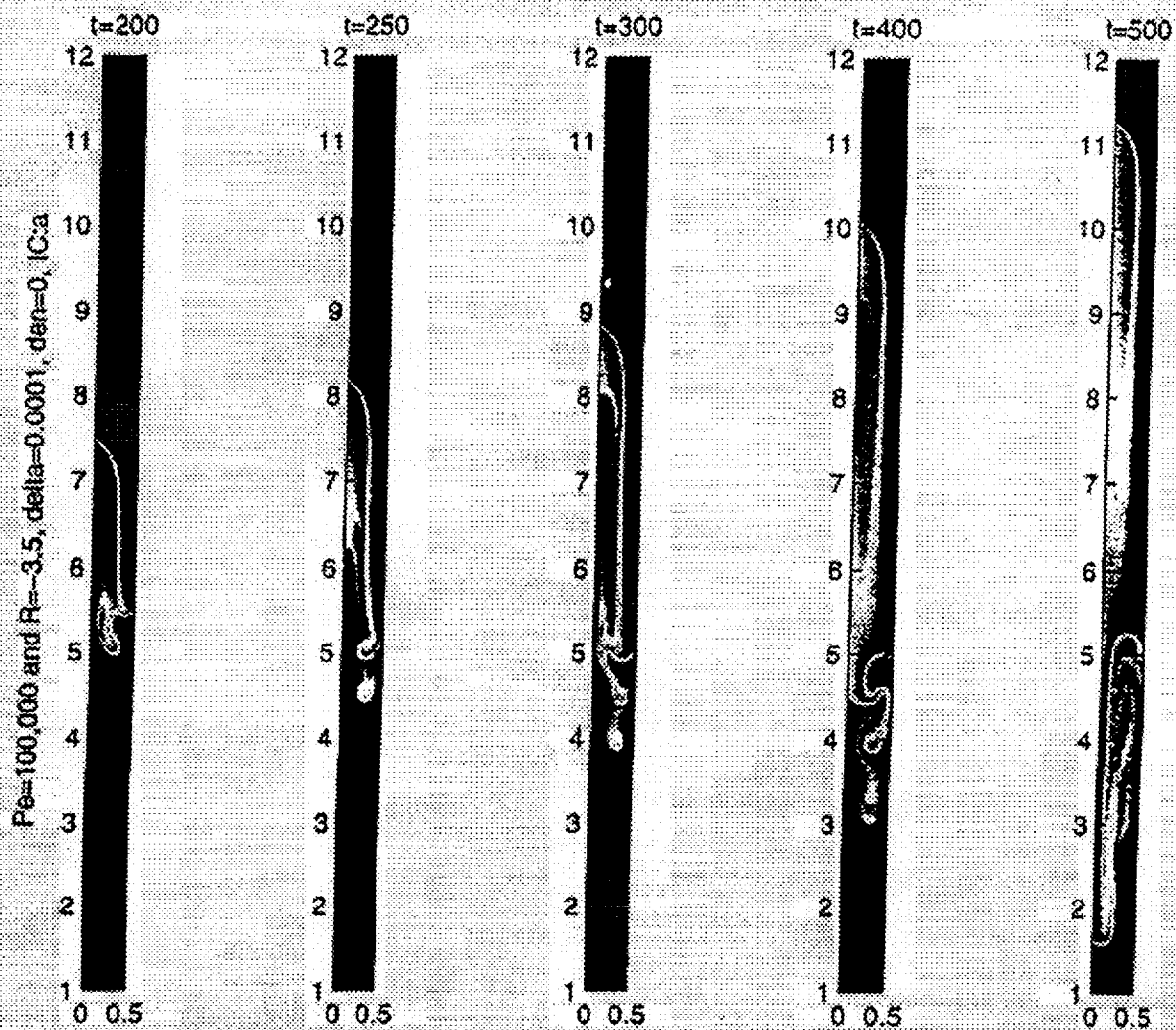
# empirical parameter variations:

## \* influence of Korteweg stresses:

- no such stresses

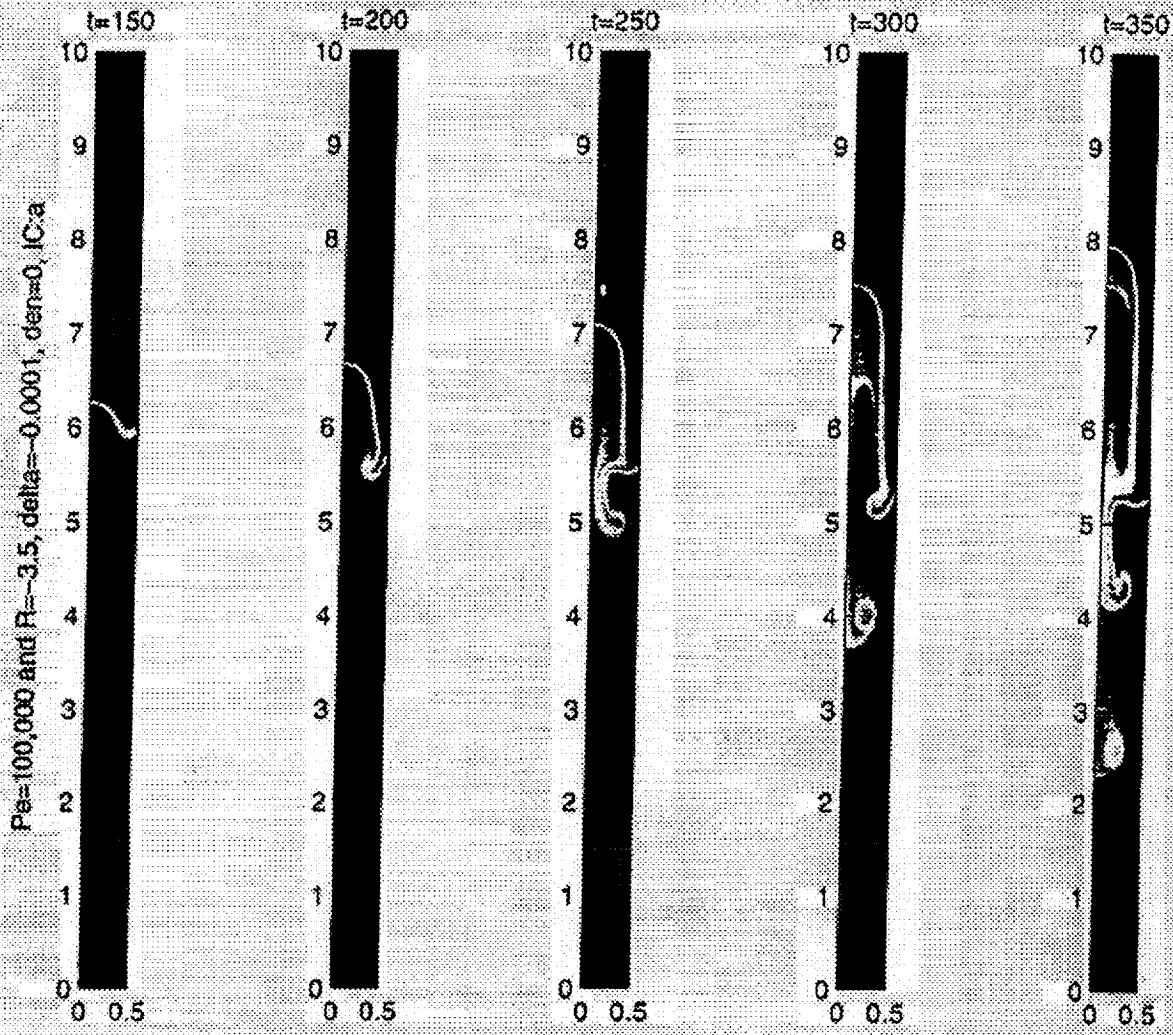


• small positive Korteweg stress coefficient 0.0001



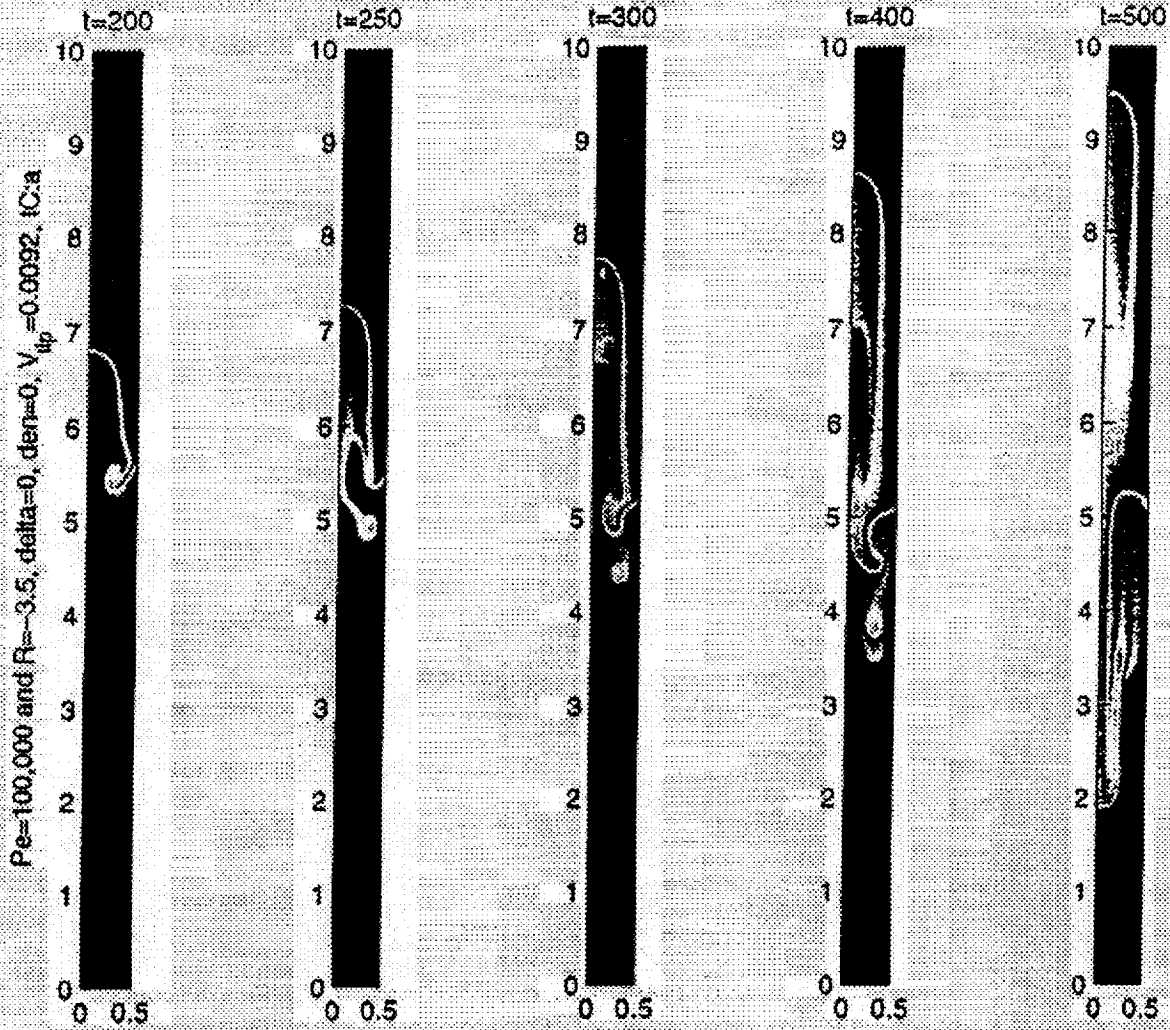


- small negative Korteweg stress coefficient -0.0001

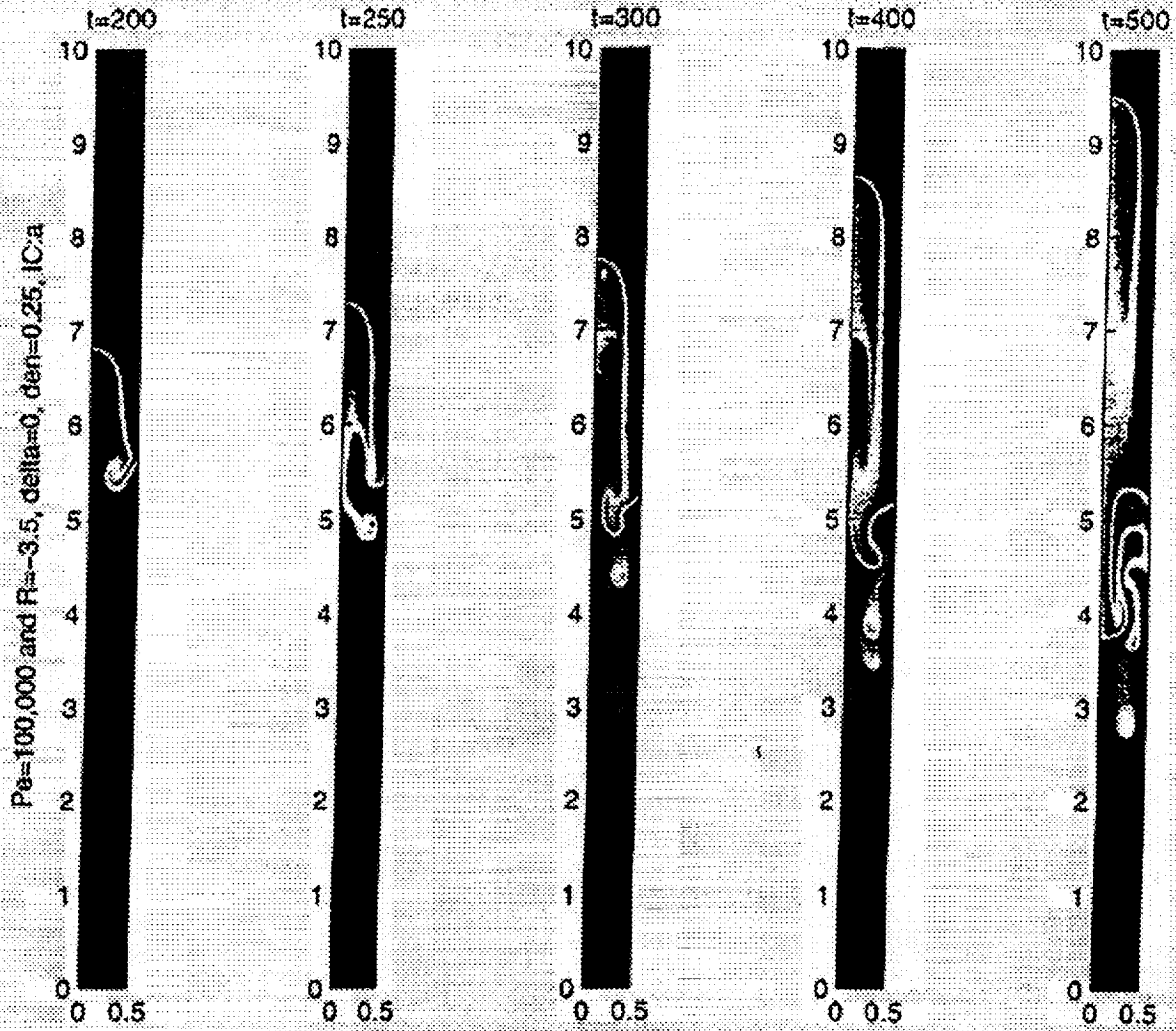


\* influence of divergence:

- no divergence



- density parameter 0.25



## **Possible space experiment**

**Need *detailed* comparison between experiments and numerical simulations at *low Pe***

- **can't use horizontal tube on the ground, because experimental flow is 3-d**
- **can't use vertical tube on the ground, because the experimental flow develops a 3-d instability at low flow rates (low  $Pe$ )**
- **have to go to microgravity environment in order to obtain an axisymmetric flow in the experiment**

### **Peliminary conclusions**

- **Fundamental questions regarding the validity of the ‘conventional’ continuity and Stokes/Navier Stokes equations in regions of steep concentration gradients**
- **mathematical formulation and magnitude of Korteweg stress coefficients are unknown, although there are suggestions**
- **there are indications from experiments and simulations that these stresses can be important**
- **plan to perform detailed comparison between microgravity experiment and numerical simulations in order to obtain more accurate information**