Mass Transfer, Mixing and Chemical Reactions in Deformable Bubble Swarms

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Bubbly Flows

Many processes, both in nature and in industry, involve bubbly flows.

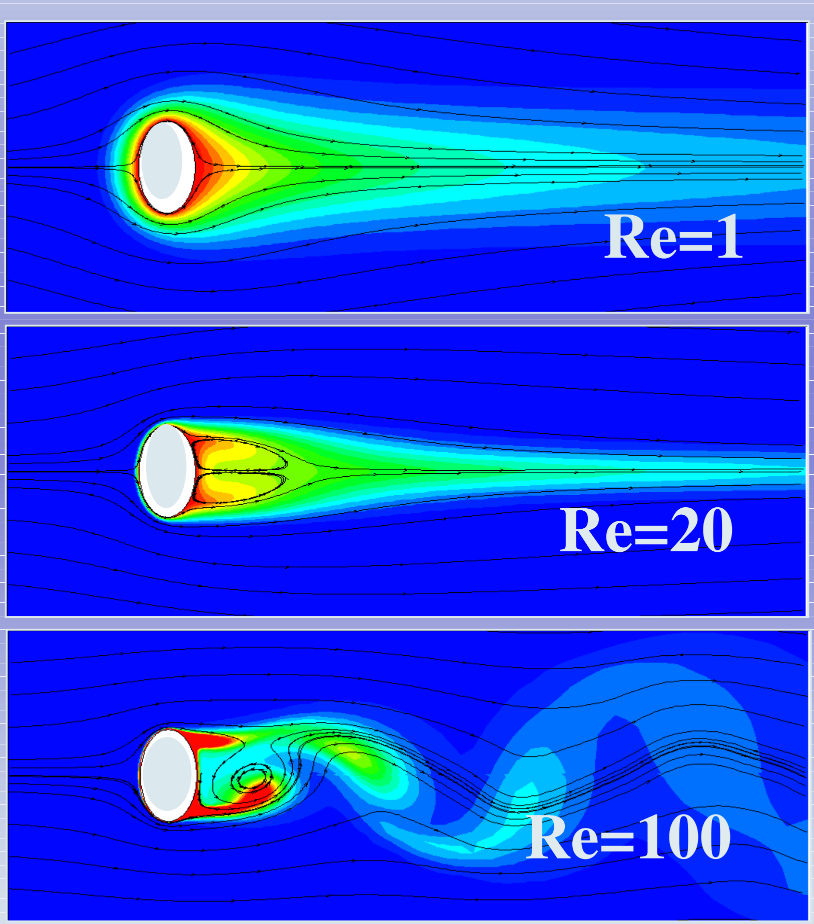
- Oxygen supply to biological systems.
  - Aeration of lakes.
  - Bio-reactors and fermentors.
  - Bio-remediation plants.
- Wet scrubbers.
  - Fly-ash and particulate pollutants removal.
- Bubble columns, jet-loop and air-lift reactors.
  - Hydrogenations, carbonylations.
  - Fischer-Tropsch synthesis.

Motivating Example

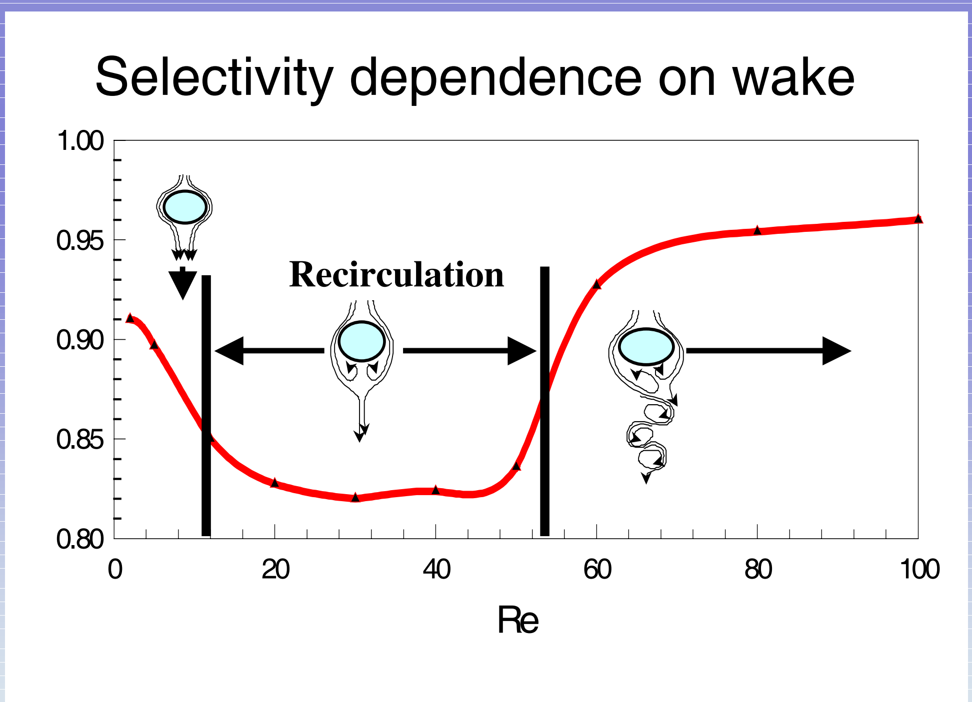
Khinast, ***AIChE J.***, **47**, 2001

$$A + H_2 \rightarrow R$$

$$R + H_2 \rightarrow S$$



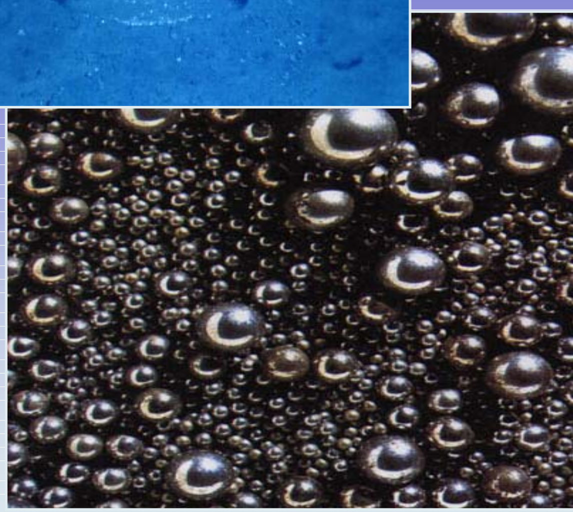
Dimensionless concentration profiles of dissolved gas.



Objectives

In realistic bubbly flows, bubble deformation and bubble-bubble interactions demand a numerical model, capable of simulating the simultaneous motion of multiple, freely deformable, reacting bubbles.

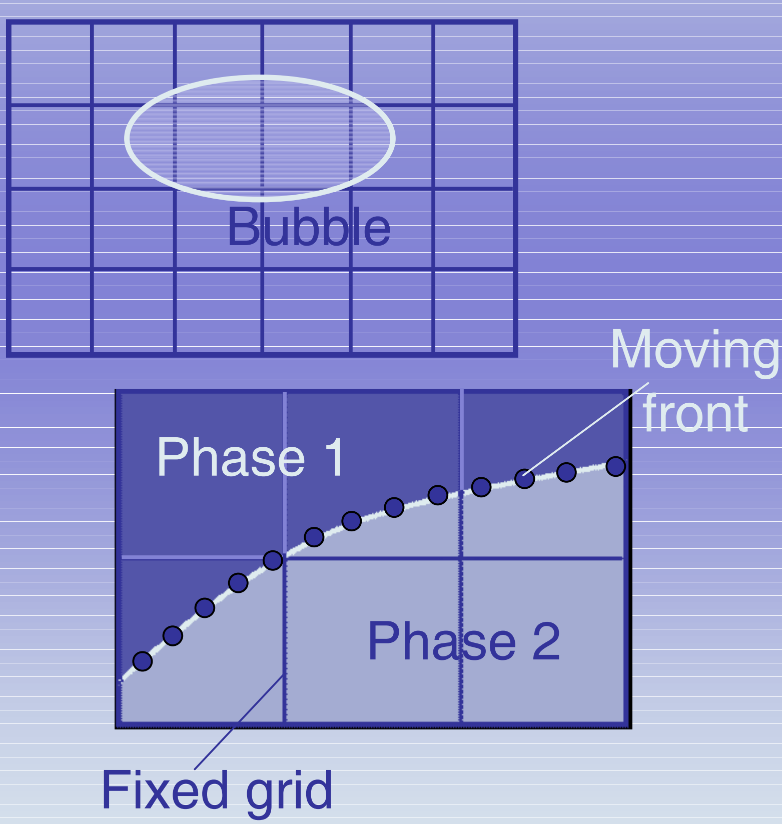
Transport in Bubbly Flows



Effects in Bubble Swarms:

- ❑ Mass and heat transfer
- ❑ Chemical reactions
- ❑ Bubble-bubble interactions
- ❑ Mixing
- ❑ Gas-liquid interaction
- ❑ Acoustic interactions
- ❑ Surfactant adsorption

Front-Capturing Method*



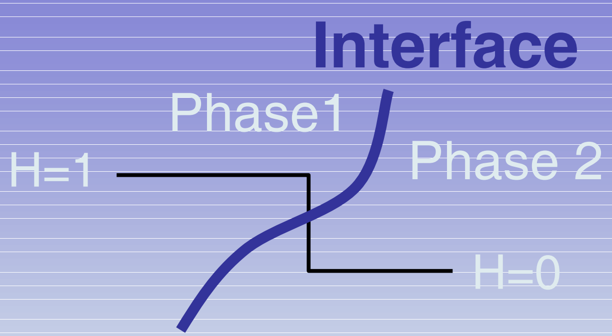
- The fluid flow is computed on a regular fixed grid.
- The location of the gas-liquid interface is tracked by a moving, deformable grid.

**** Grétar Tryggvason, WPI***

Tryggvason *et al.*, *J. of Comp. Phys.*, **169**, 2001

Model Equations

- A single set of conservation equations are solved for both phases.
- Since material properties, such as density and viscosity are discontinuous across the interface, all variables are written in terms of generalized functions.
- The equations are solved on the fixed grid and the front is tracked by a moving front composed of markers.



$$\rho(x,y,t) = \rho_1 H(x,y,t) + \rho_0 (1 - H(x,y,t))$$

$$\nabla\rho = \rho_1 \nabla H - \rho_0 \nabla H = (\rho_1 - \rho_0)\nabla H = (\rho_0 - \rho_1)\oint \delta(x - x')\delta(y - y')\mathbf{n}' ds'$$

Model Equations

- Momentum:

$$\frac{\partial \rho \mathbf{u}}{\partial t} + \nabla \cdot \rho \mathbf{u}\mathbf{u} = -\nabla P + \rho \mathbf{f} + \mu\left(\nabla \mathbf{u} + \nabla^T \mathbf{u}\right) + \underbrace{\int \sigma \kappa' \mathbf{n}' \delta^{\beta}\left(\mathbf{x} - \mathbf{x}'\right) ds'}$$

- Mass:

$$\frac{\partial \rho}{\partial t} + \nabla \cdot \rho \mathbf{u} = 0$$

σ - surface tension

β- dimension

κ- curvature

n – vector normal to the front

- Transport of species i:

$$\frac{\partial c_i}{\partial t} = -\mathbf{u} \cdot \nabla c_i + D\nabla^2 c_i + \sum_{j=1,m} r_{ij} \qquad i = 1, n$$

Material Transport

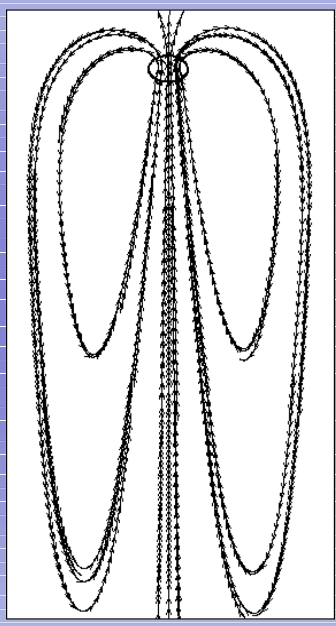
- In order to resolve the steep concentration gradients near the bubble, the resolution required for the mass transport calculations greatly exceeds the one required for the flow computations.

- The grids for the shown hydrodynamic computations have **120,000 (200x600)** cells. The mass transport grids have over **1,000,000 (600x1800) cells**.

- The velocities at each fine grid point, needed for the resolution of the convective fluxes were interpolated using a high-order continuity-preserving scheme.

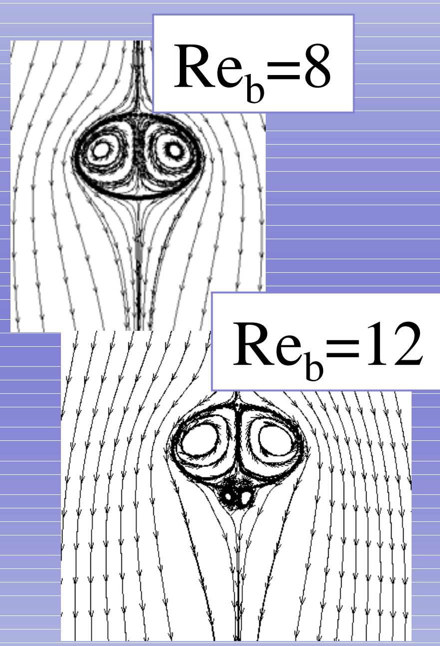
  - Details on the scheme can be found in Koynov *et al.*, *AIChE J.*, 2005.

Single Bubble
(hydrodynamics)

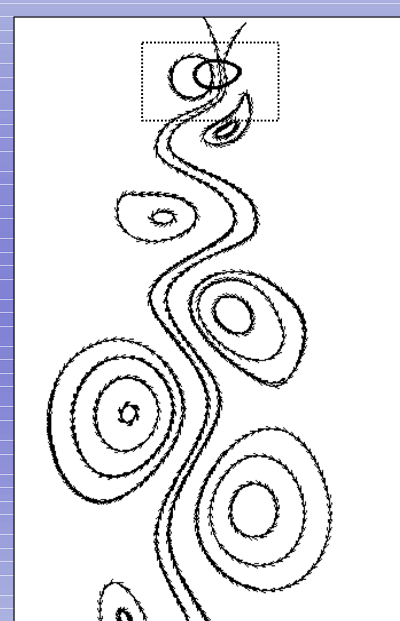


Re_b=8

- Closed, steady wake



- Internal circulation

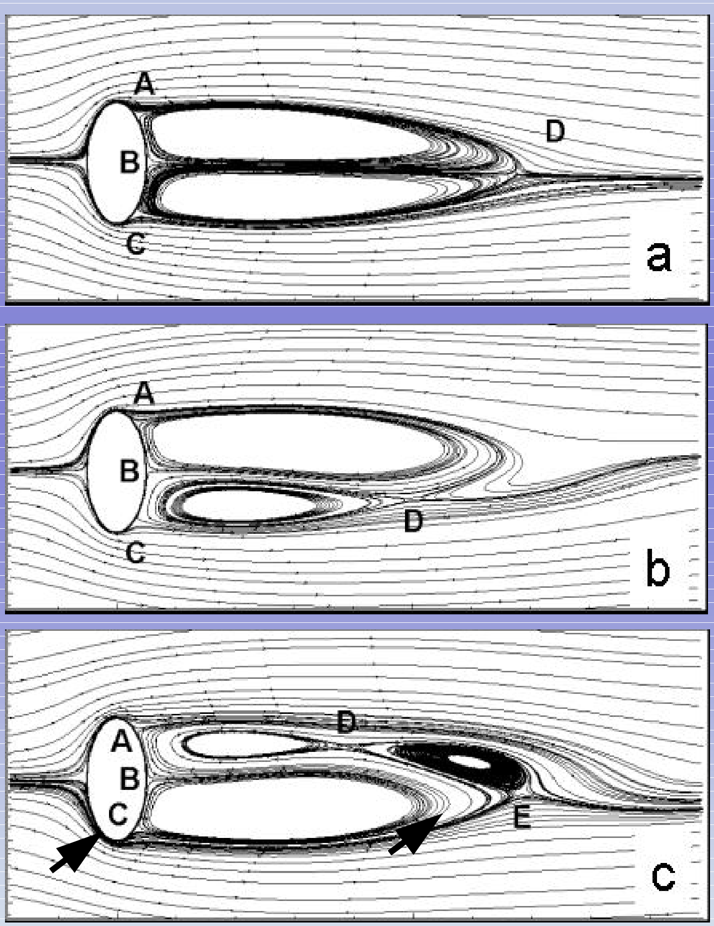


Re_b=68

- Vortex-shedding wake

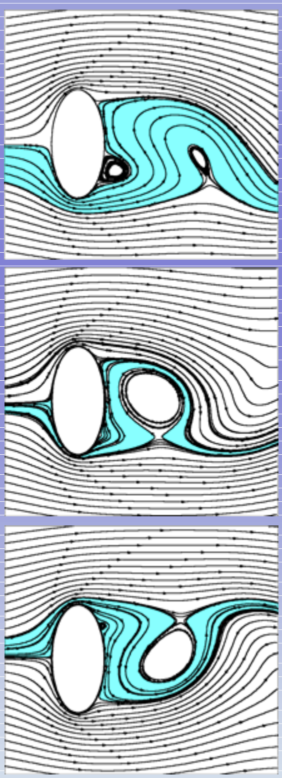
- Deformable shape
- Serpentine trajectory at higher Re.

Wake Transport



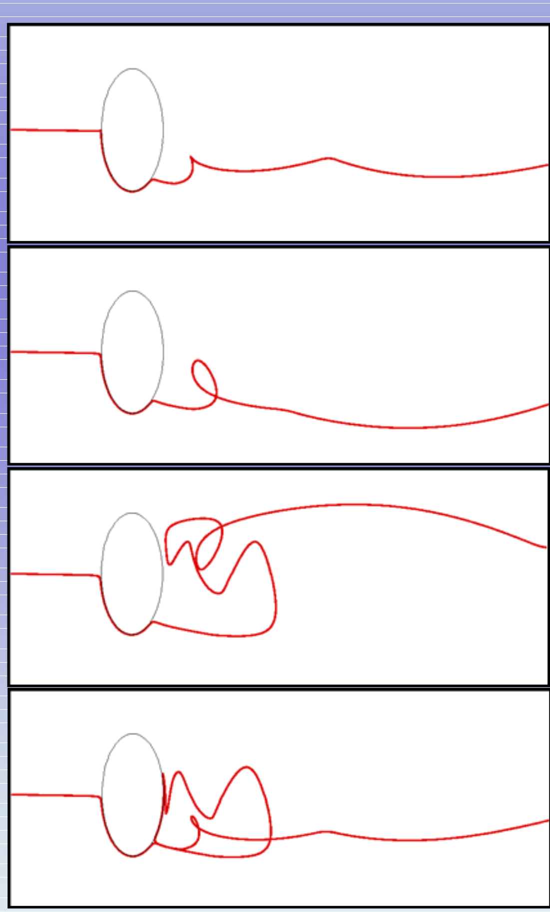
- The two recirculation zones lose stability.
- One zone grows larger than the other.
- The larger one pinches in two, shedding a vortex and creating a hyperbolic /elliptical pair.

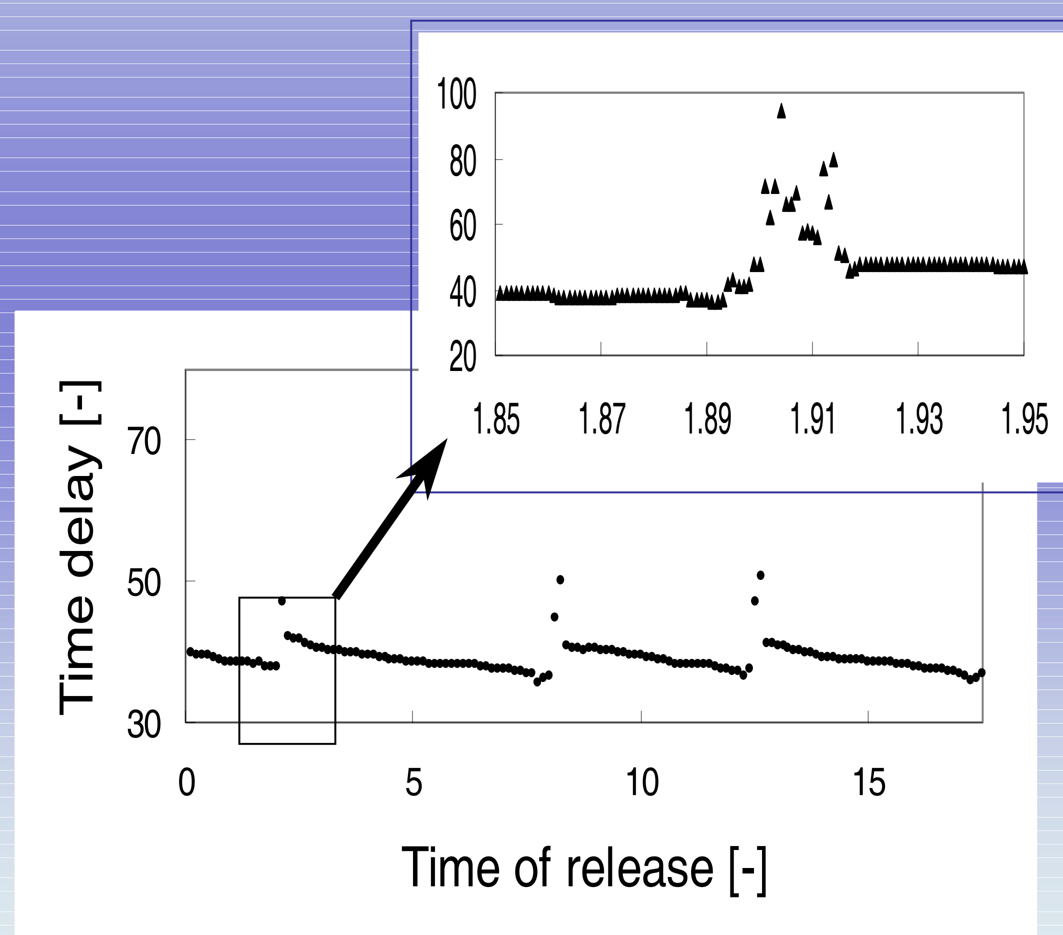
Channel formation



Hamiltonian Scattering

Massless particles injected in the steady asymptotic region upwind of the bubble, at different times. Re=100.



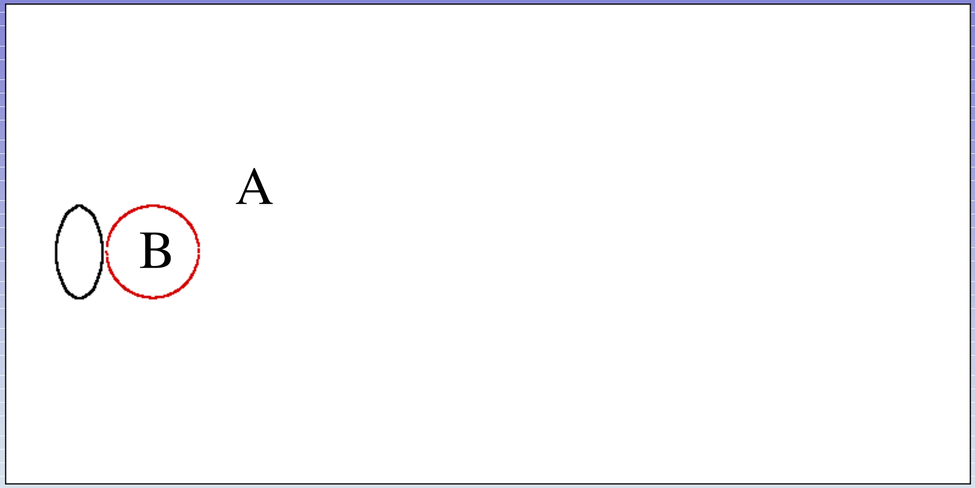


Front Evolution

Closed, steady wake.

No recirculation.

Re=5

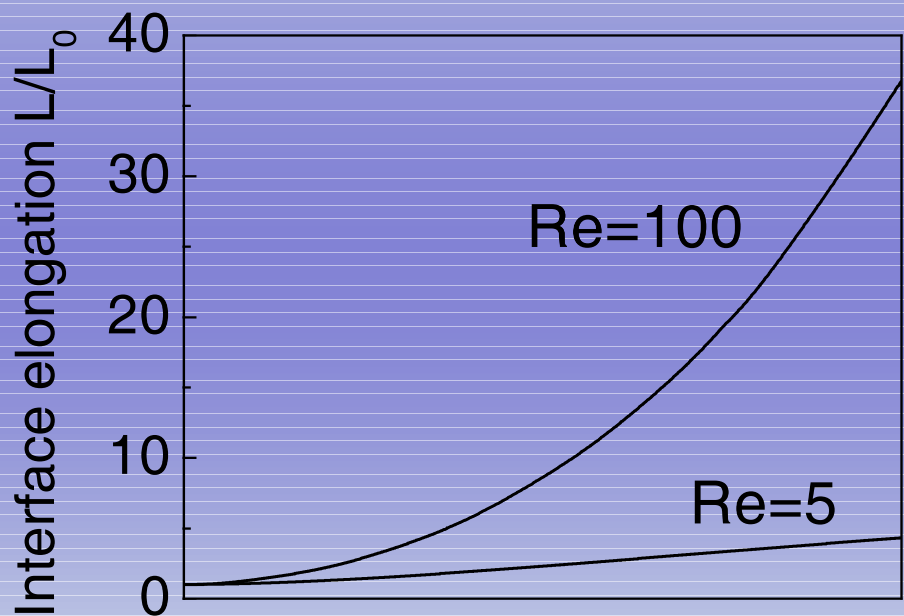


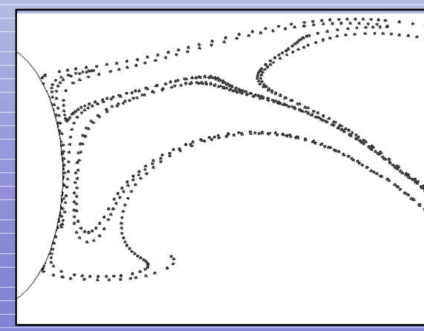
Open, unsteady wake.

Vortex-shedding.

Re=100

Front Evolution





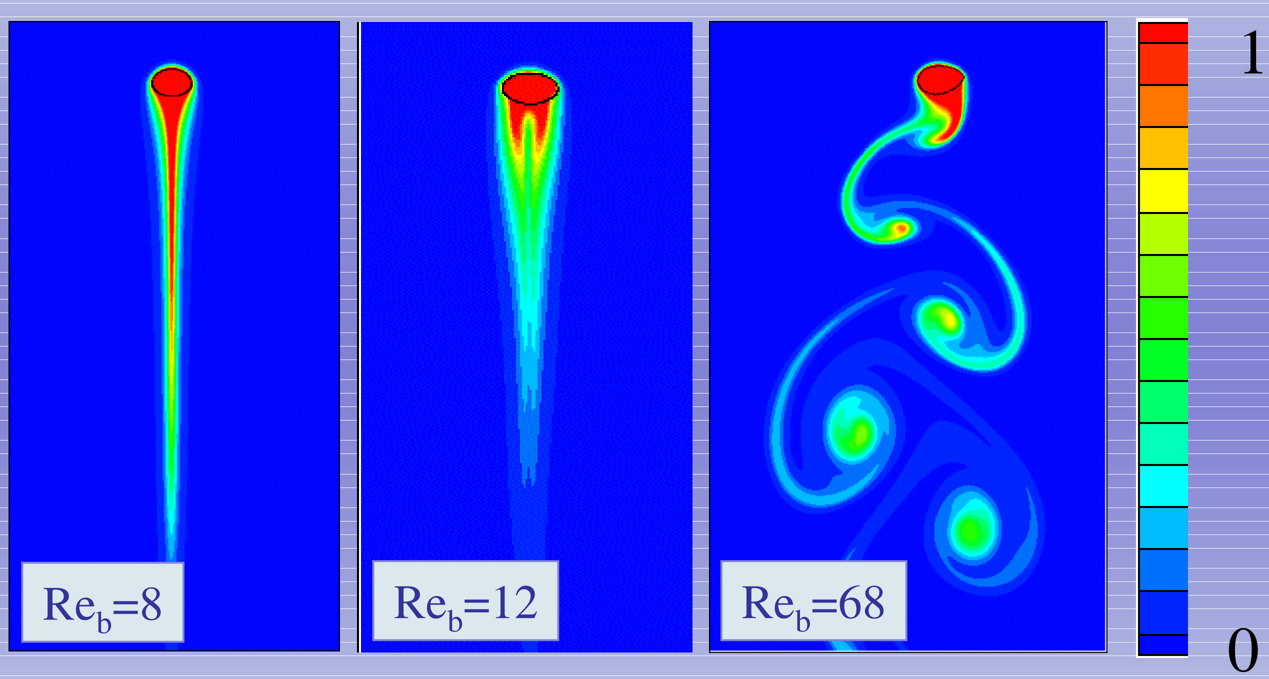
Zoom-in of the wake region for Re=100.

Due to stretching and folding, the length of the interface will grow **exponentially faster** in the vortex shedding regime than in a steady wake regime.

Koynov and Khinast, ***Chem. Eng. Sci.***, 2004

Single Bubble

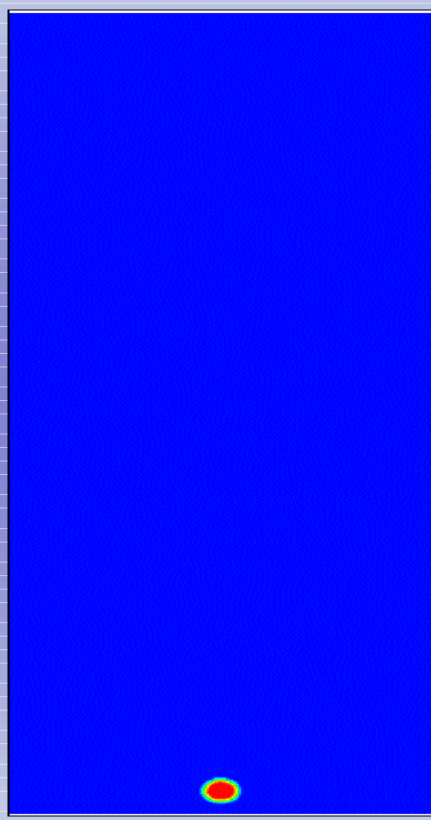
(mass transfer)

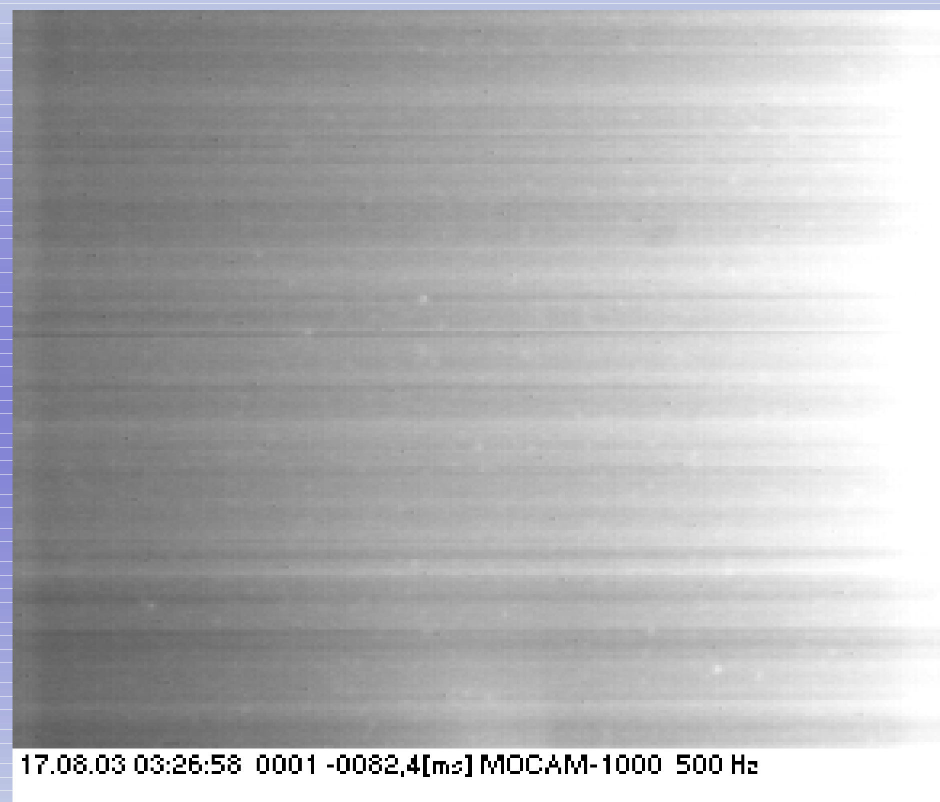


- Closed, steady wake
- Steady, recirculating wake
- Vortex-shedding wake

Sc=400; Eö=3.125,

Single Bubble
(mass transfer)





Model results

Experimental results (courtesy of **M. Schlüter** *et al. – Institut für Umweltverfahrenstechnik,* **Bremen, Germany**)

Sc=400; Eö=3.125,

Koynov *et al., submitted to Nature, 2005*

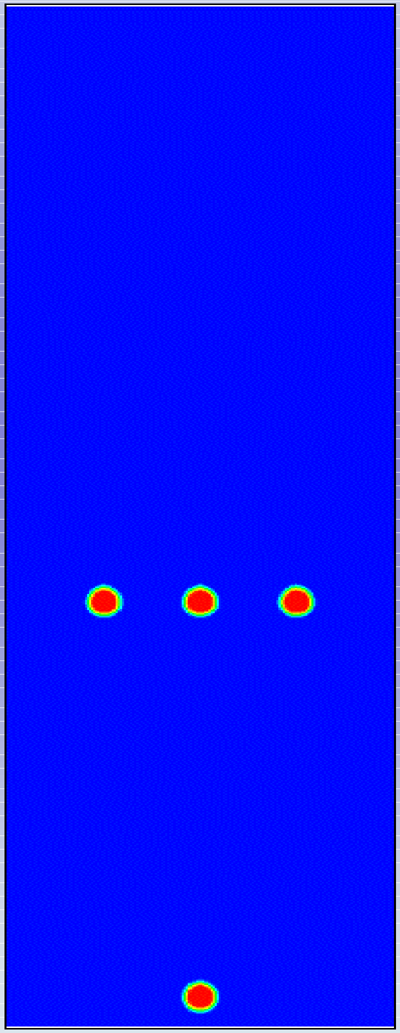
Bubble Swarm
(mass transfer)

Swarm of 4 bubbles

r=3.5

Sc=400; Eö=3.125,

Mo=$1.2 \cdot 10^{-3}$

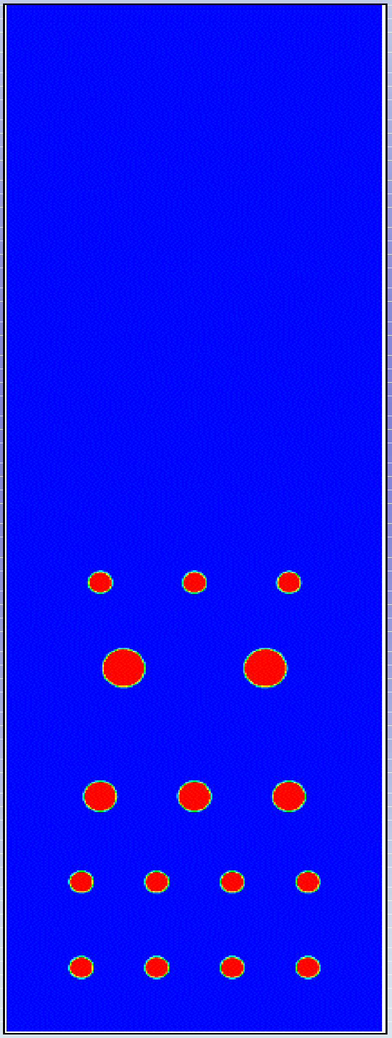


Swarm of 16 bubbles

$r_1:r_2:r_3=2.5:3.5:4.5$

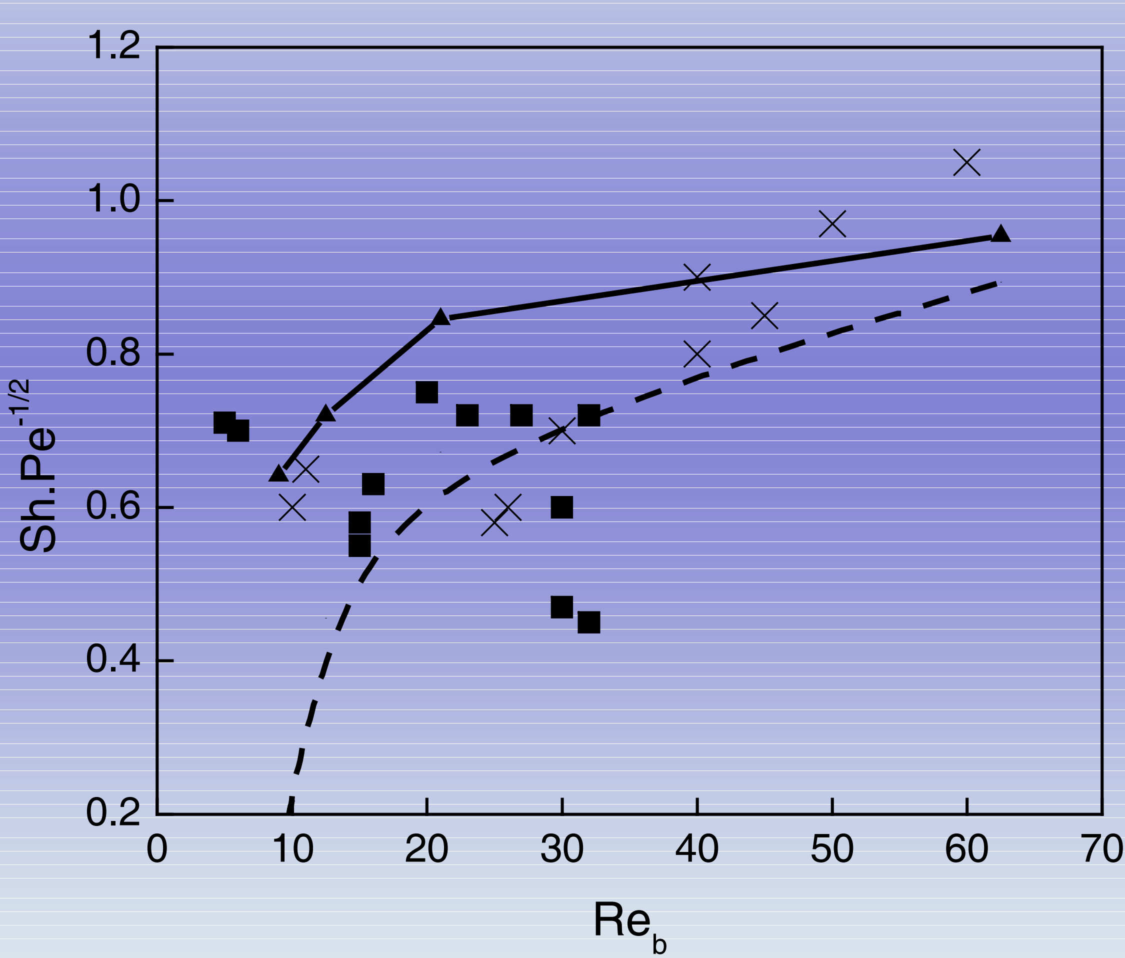
Sc=400; Eö=3.125,

Mo=$3.1 \cdot 10^{-4}$



Koynov *et al.*, *AIChE J.*, 2005.

Mass Transfer

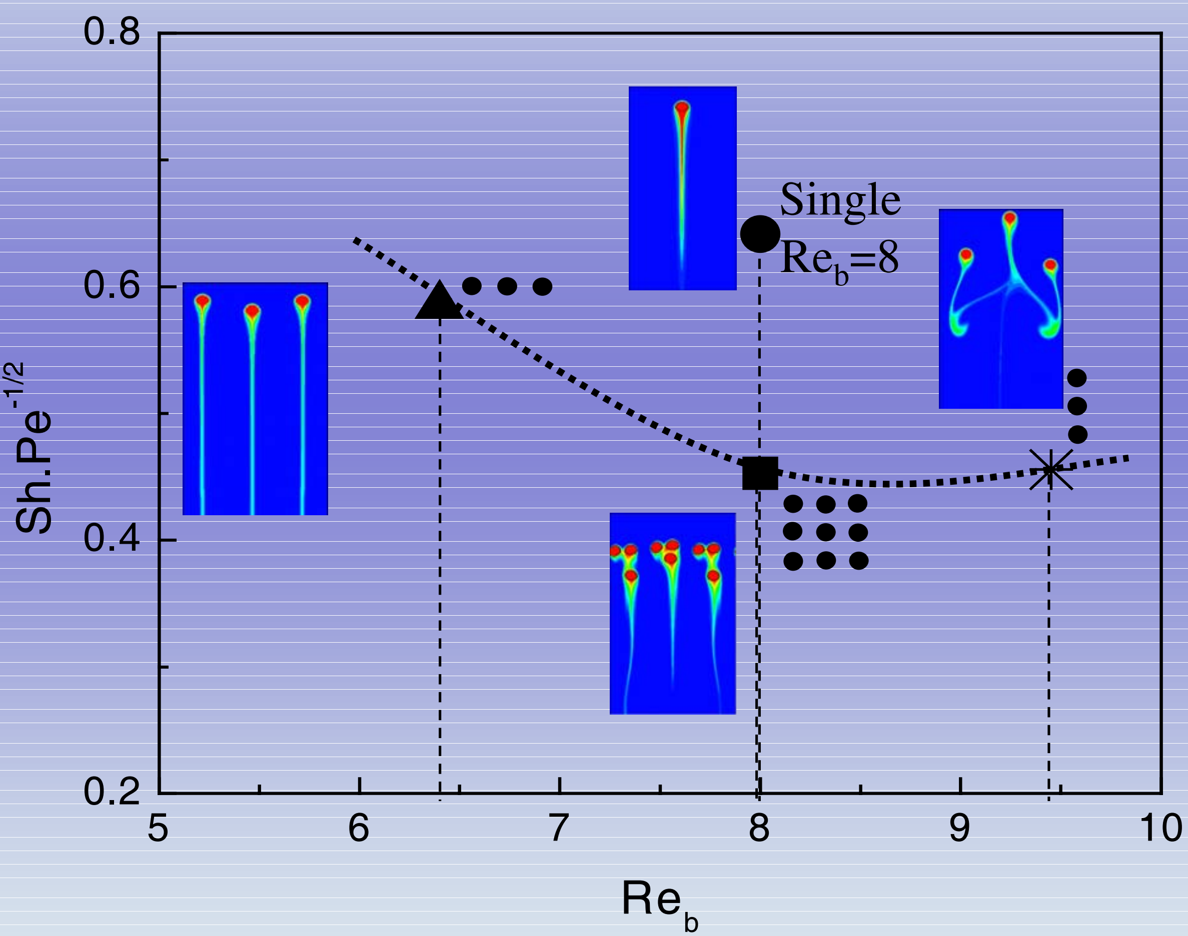


Mass transfer rates as a function of the Reynolds number.

- ▲ - our simulations;
- ■ and ✕ - data from Redfield and Houghton (1965)*;
- Dashed line – modified Boussinesq equation.

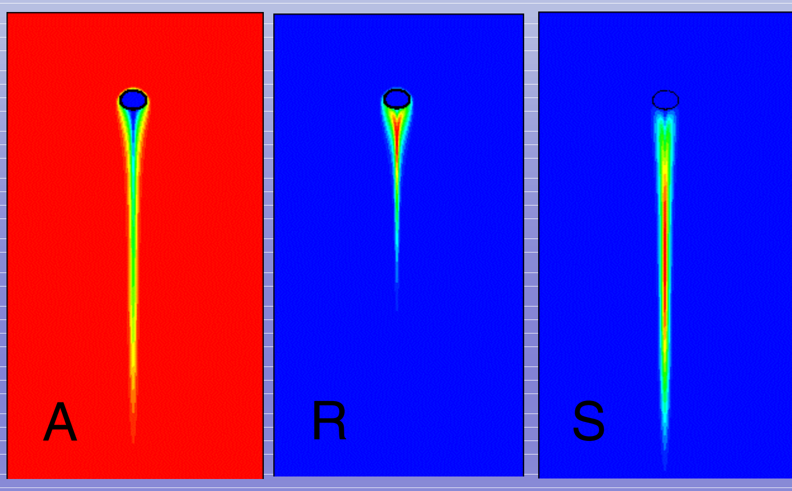
*Redfield and Houghton, *Chem. Eng. Sci.*, **20**, 2001

Mass Transfer
(swarms)



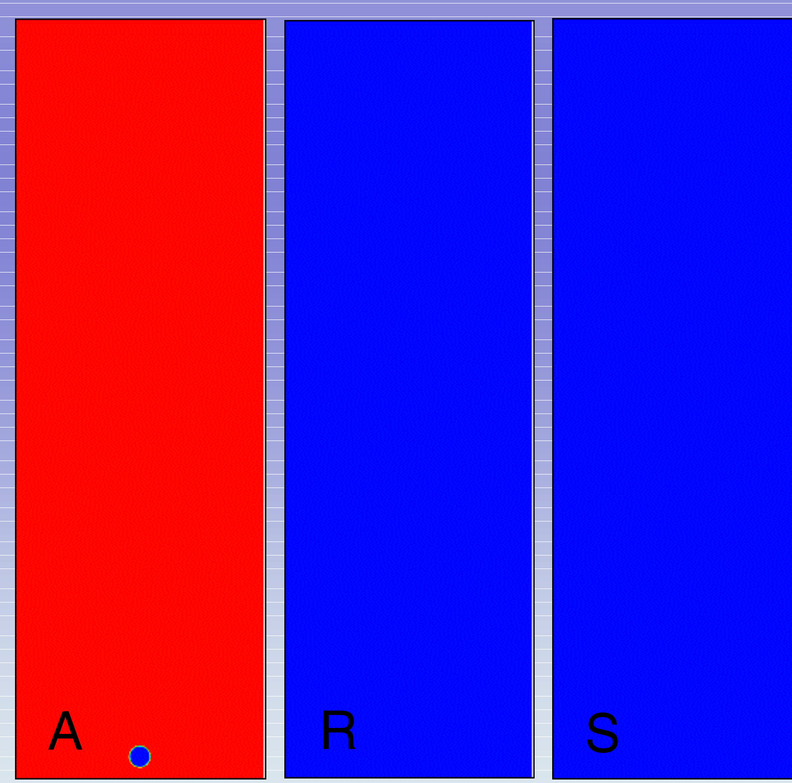
Wake effects lead to lower mass transfer in bubble clusters.

Chemical Reactions
(single bubble)



Re$_b$=8
(steady wake)

Re$_b$=68
(vortex-shedding wake)



$$A + B_{(g)} \rightarrow R$$

$$R + A \rightarrow S$$

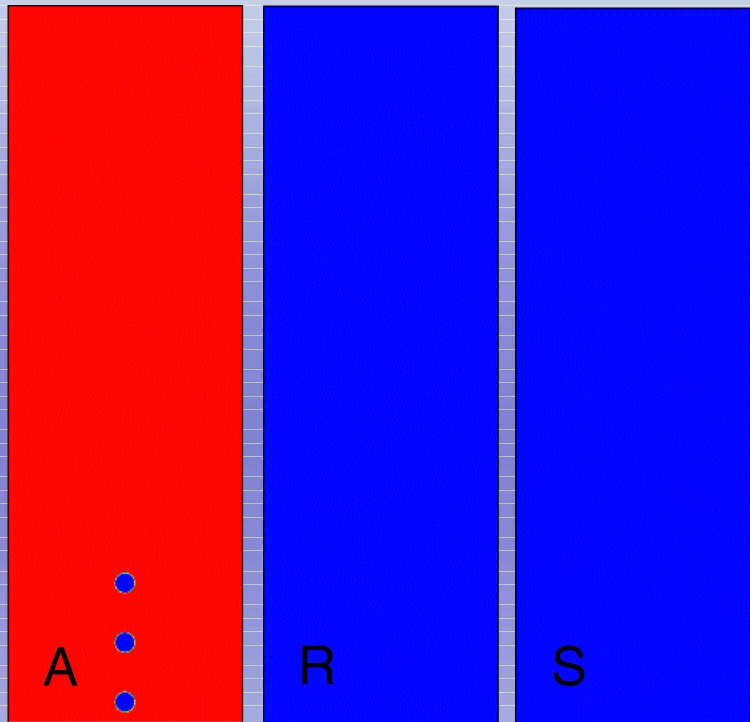
Da_1=0.25
Da_1/Da_2=10
Sc=400; Eö=3.125,

Chemical Reactions

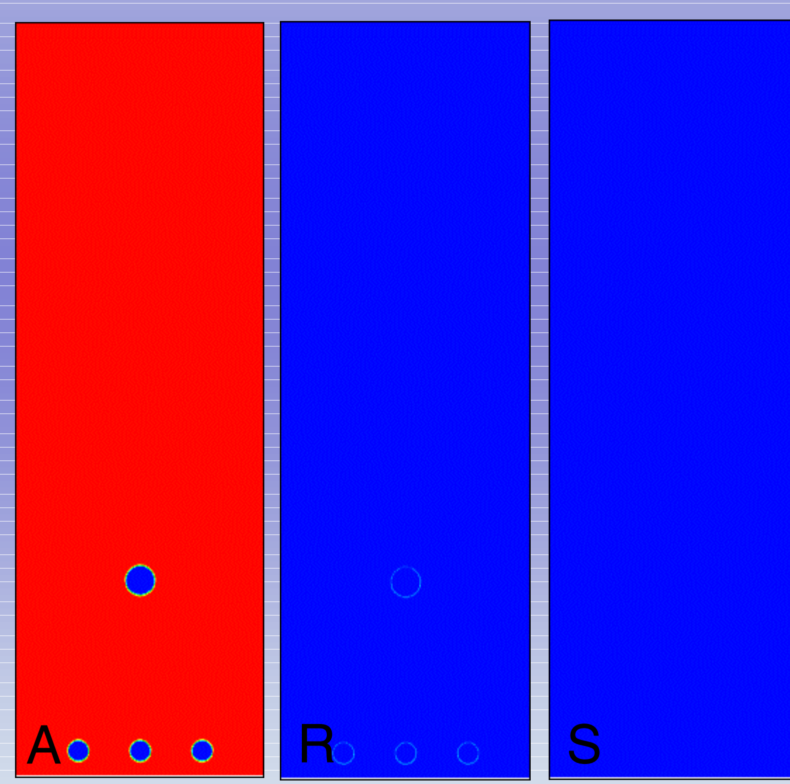
(swarms)

$$A + B \rightarrow R$$

$$R + A \rightarrow S$$

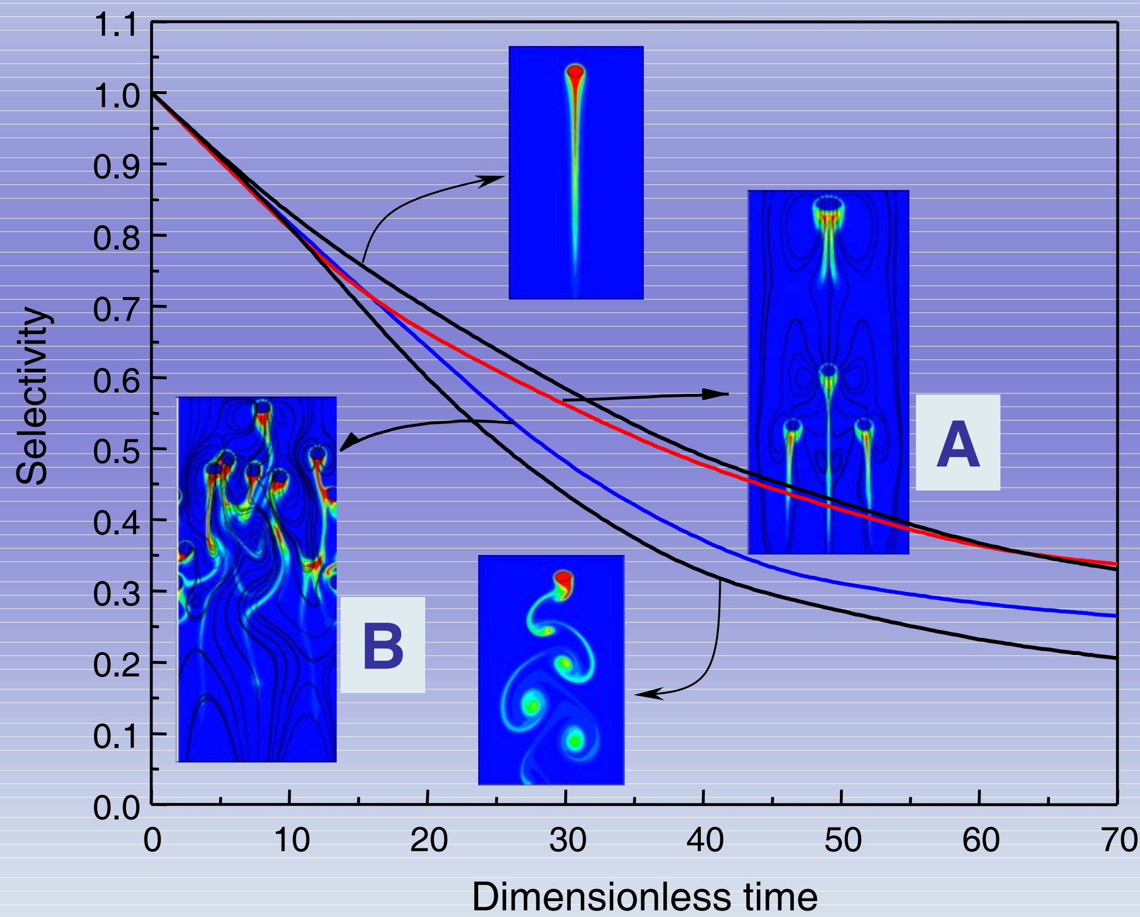


Same size bubbles



Smaller bubbles in the wake of a large bubble

Selectivity



The mixing in swarm A is inferior to the one in swarm B, due to the large separation between the bubbles, leading to a single bubble-like flow.

CDF in BRE

- In many biological systems, bubbly flows are used for the delivery of oxygen to liquid suspensions of cells. Examples include *bioreactors*, *fermentors* and *bioremediation plants*.

Modeling of Aerated Cell Suspensions

Considerations:

- Hypoxia: Suspended cells depend on the oxygen delivered by the bubbles for survival and insufficient supply can, over time, lead to cell damage and apoptosis.
- Membrane Rupture: The liquid flow will cause the suspended cells to deform. If the deformation exceeds some critical value, the cell membrane can rupture leading to cell lysis.
- Exposure to Shear: Prolonged exposure to shear stress (even at levels too low to cause membrane rupture) can lead to deterioration of the membrane properties and eventually to cell death through necrosis or apoptosis.

Modeling of Aerated Cell Suspensions

Model:

- Cells are modeled as neutrally buoyant Lagrangian tracers dispersed through the liquid phase surrounding the rising bubbles.

$$\dot{\mathbf{x}}_c = \sum_{i,j} w_{i,j} \mathbf{u}_{i,j}$$

$w_{i,j}$ – *weighing functions*

$\mathbf{u}_{i,j}$ – *velocity at grid point (i,j)*

- Simulations of coupled hydrodynamics, mass transfer, oxygen transport and depletion and cell advection are carried out for different bubbly flows.

Over 500,000 individual cells are tracked.

Modeling of Aerated Cell Suspensions

Born *et al., Biotech. and Bioeng.*, **40**, (1992)

Membrane Rupture:

If a cell is assumed to behave like a viscous drop, its deformation, caused by laminar shear will equal:

$$D = \xi\tau\frac{r_c}{\sigma}$$

The membrane tension caused by the deformation will, therefore, be

$$\sigma = \frac{\xi\tau r_c}{D}$$

r_c - cell radius
σ – membrane tension
τ – shear stress
ξ – deformation coefficient

If the cell tension σ exceeds a given bursting value, σ_b i.e.

$$\frac{\xi\tau r_c}{D_b} \geq \sigma_b$$

the cell will be destroyed.

To simulate biodiversity, bursting tensions of the cells obey a random Gaussian distribution

Modeling of Aerated Cell Suspensions

Hypoxia and Shear Related Accumulative Damage:

Prolonged exposure to shear as well as severe hypoxia can induce either necrotic or apoptotic cell death. Rate expressions correlating cumulative cell damage to local shear stresses and oxygen concentrations were derived, based on experimental data published in the literature (*e.g.* Abu-Reesh and Kargi, 1989; Charlier *et al.*, 2002 and Guarino *et al.*, 2004).

$$\frac{d\chi}{dt} = \left(a_c\tau^2 + b_c\tau + c_c\right) + H_c$$

a_c, b_c, c_c, d_c – fitting coefficients

χ – cell damage; R_c – consumption co

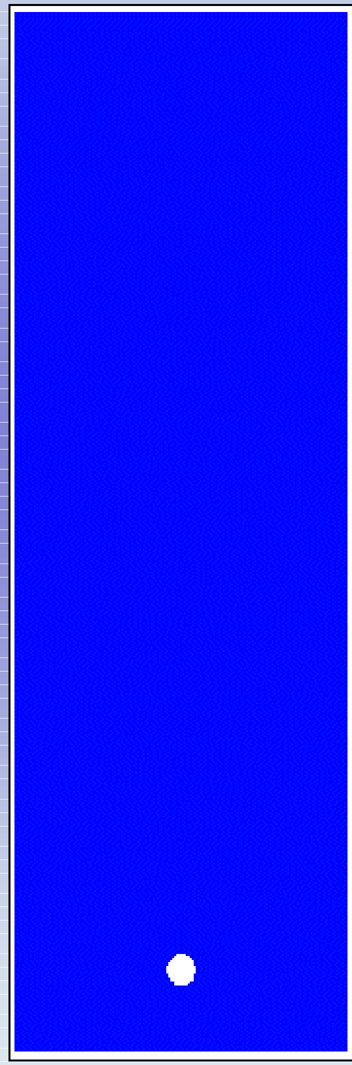
$$H_c = \begin{cases} 0, \text{ if } c_{O_2} > R_c \\ d_c \dfrac{c_{O_2} - R_c}{R_c} \quad \text{if } c_{O_2} < R_c \end{cases}$$

If the cell damage exceeds a certain critical value, the cell is destroyed.

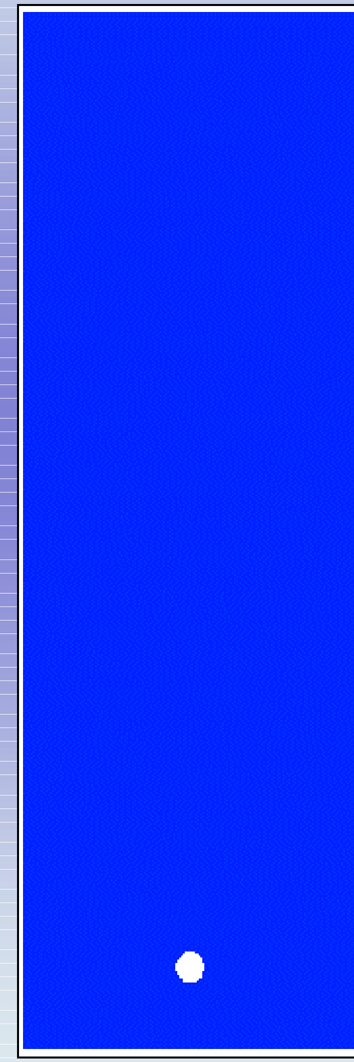
To simulate biodiversity, χ_c values obey a random Gaussian distribution

Cell Damage –
Steady Wake

Hypoxia caused damage



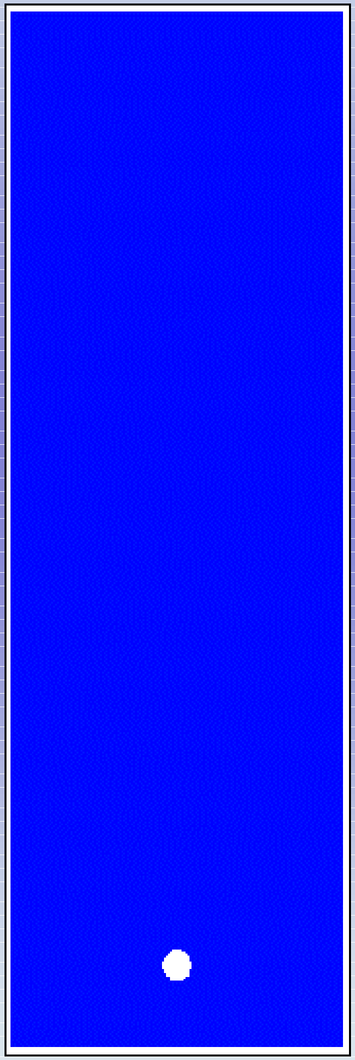
Shear caused damage



Re=8, Sc=400

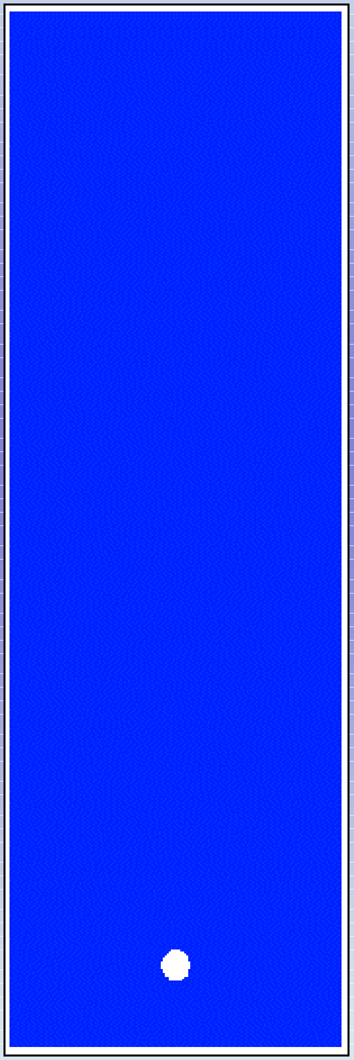
Cell Damage –
Vortex-Shedding Wake

Hypoxia caused damage

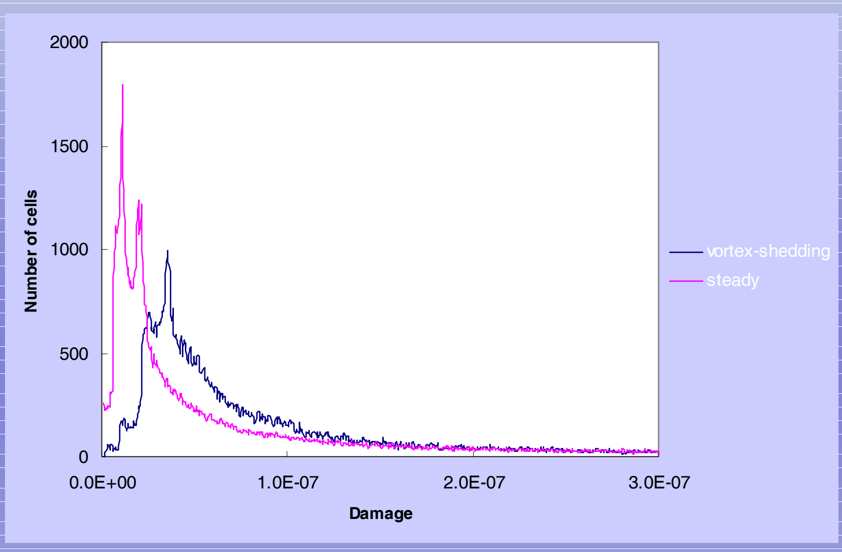


Re=68, Sc=400

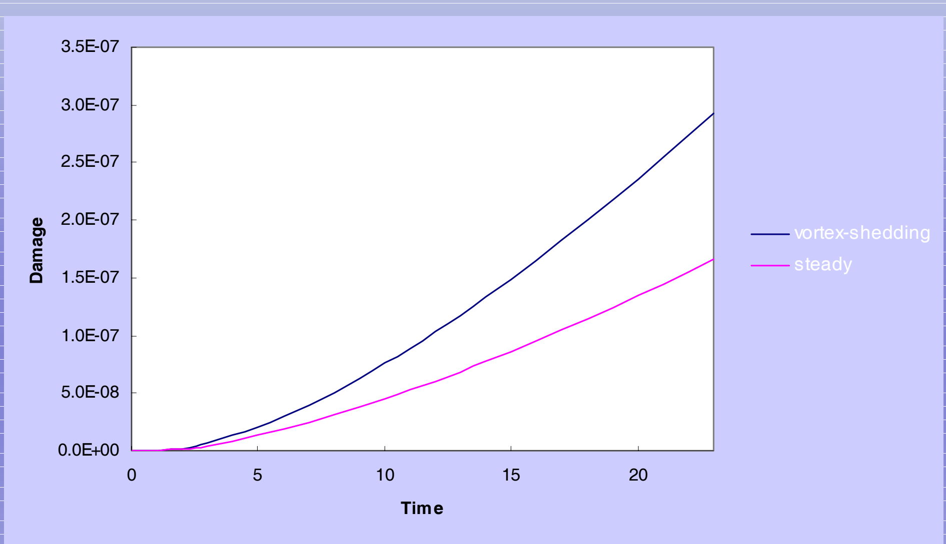
Shear caused damage



Shear Damage



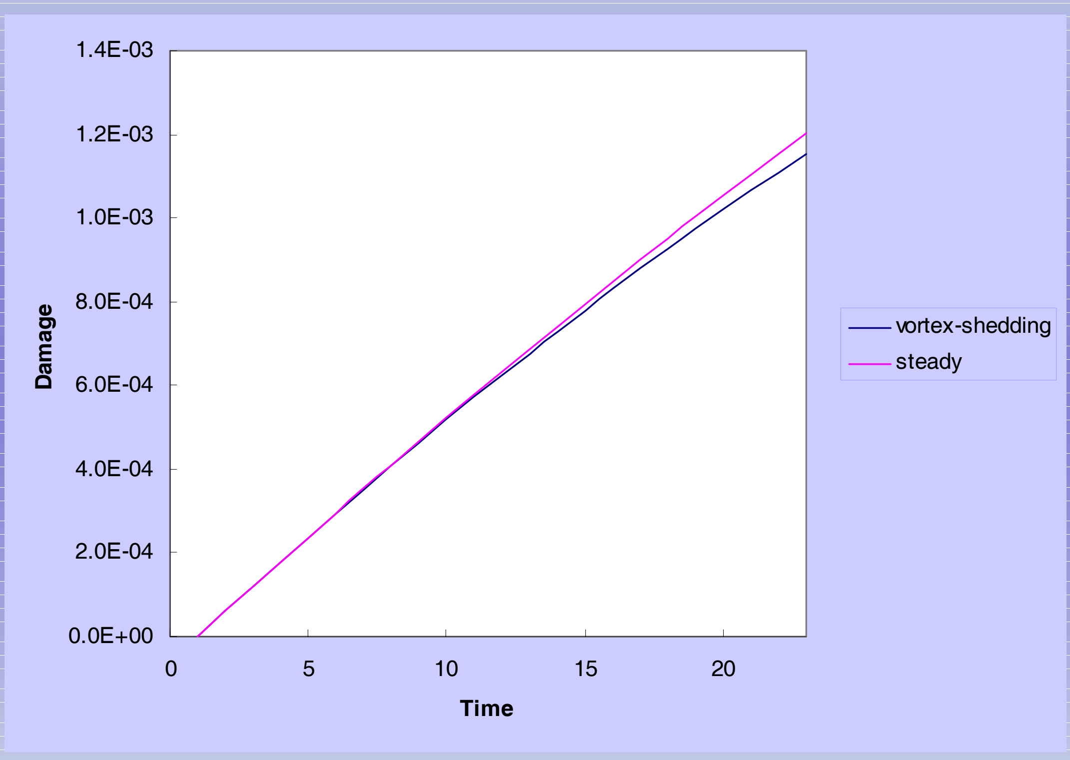
Damage distribution



Mean damage over time

Vortex-shedding results in flow-fields, characterized by higher shear and causing damage to more cells than the ones encountered in the steady wakes.

Hypoxia Damage



Mean damage over time
(due to hypoxia)

Better mixing in the vortex-shedding wake leads to better aeration and decreases the hypoxia-caused damage to the cells.

Conclusions

- A code has been developed allowing the simulations of the buoyancy-driven motion of clusters of deformable, reactive bubbles.
- Depending on flow parameters, such as density, viscosity, pressure and surface tension the dynamics of the flows can change dramatically.
- The different flow dynamics translate into different mixing and transport properties, which in turn can influence the product distribution of certain chemical reactions.
- In suspensions of cells, the differences in shear stresses and mixing rates can result in different rates of damage to the cells.

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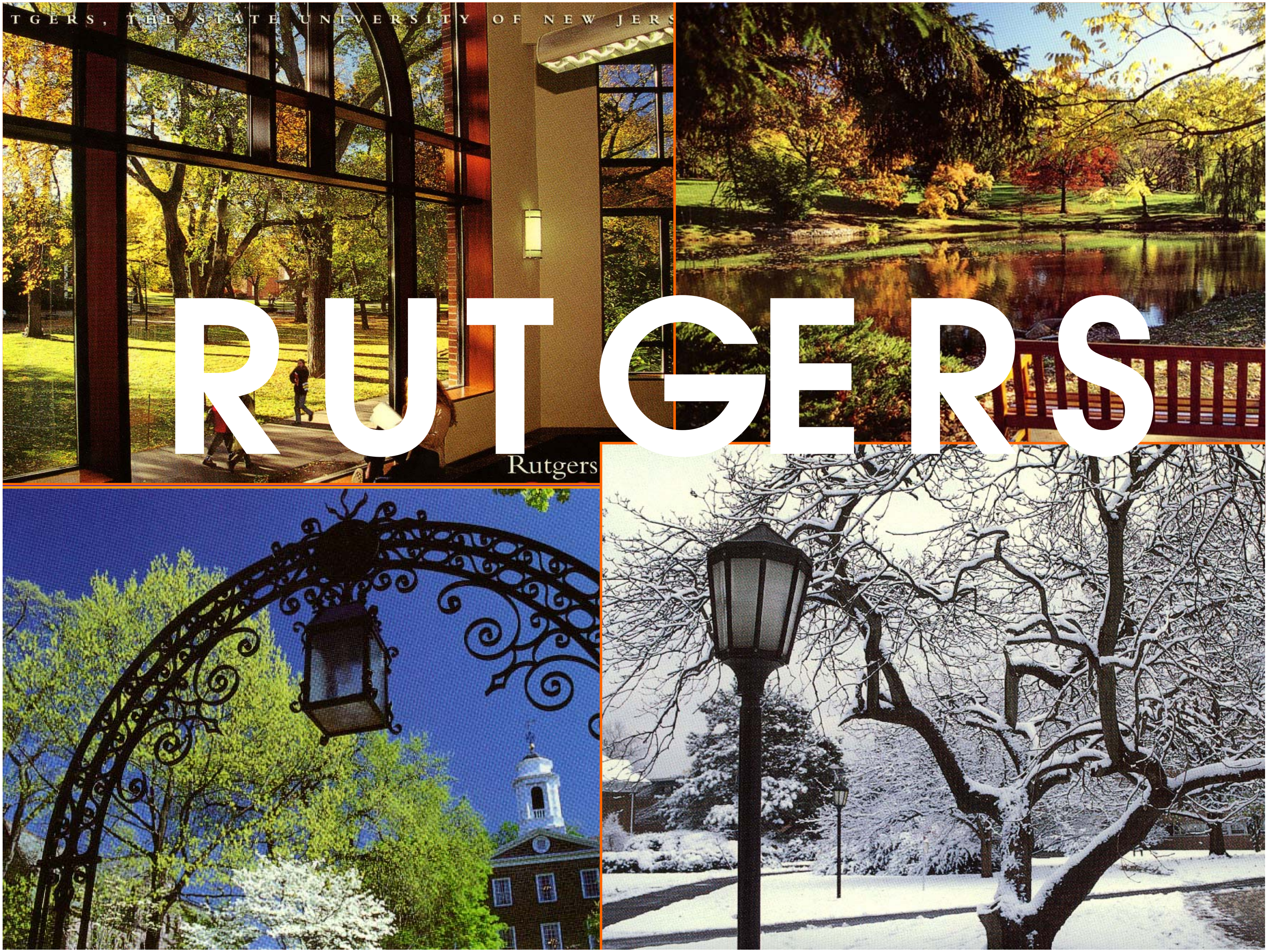
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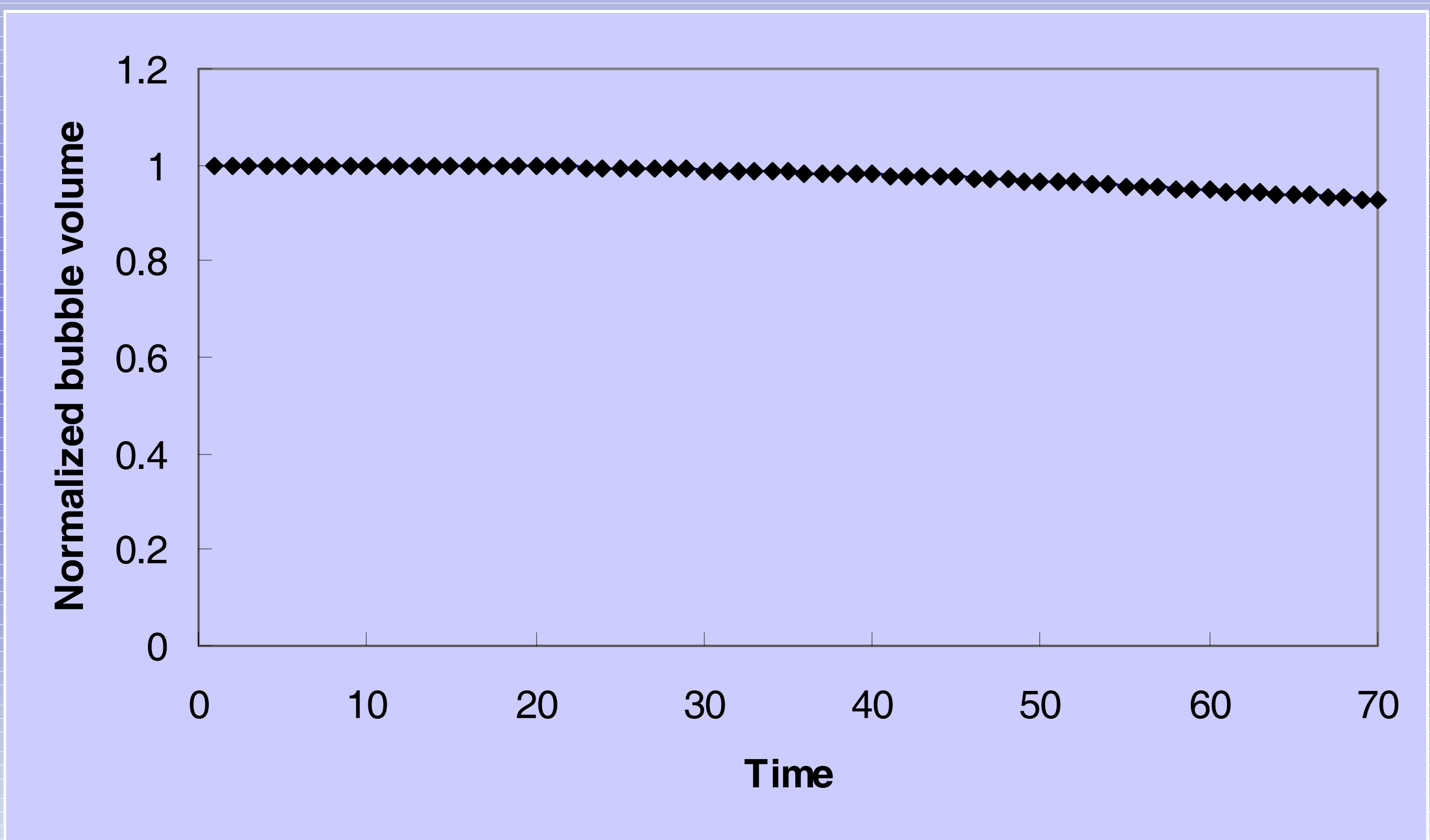
Loss of Volume

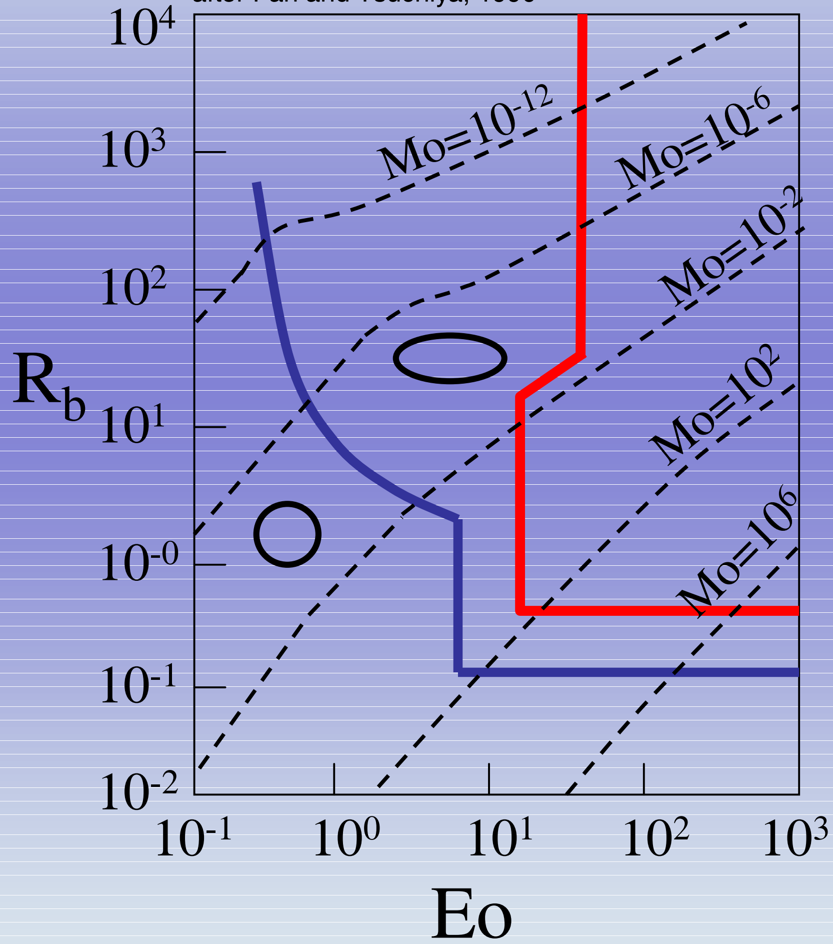


Bubble Shapes

Three dimensionless numbers determine bubble shape :

- ***Eötvös or Bond number Eo*:** Ratio of gravity to surface tension forces. When small, the surface tension dominates over the hydrostatic pressure difference from top to bubble bottom and the bubble keeps spherical. Otherwise, it tends to lower its height, becoming ellipsoidal.
- ***Morton number Mo:*** Ratio of viscous forces to surface tension forces at terminal velocity giving the relative tendency of the viscous forces to drag the bubble's lateral border backward in a spherical cap shape.
- ***Reynolds number R*:** Ratio of the inertial to viscous forces based on terminal velocity

after Fan and Tsuchiya, 1990



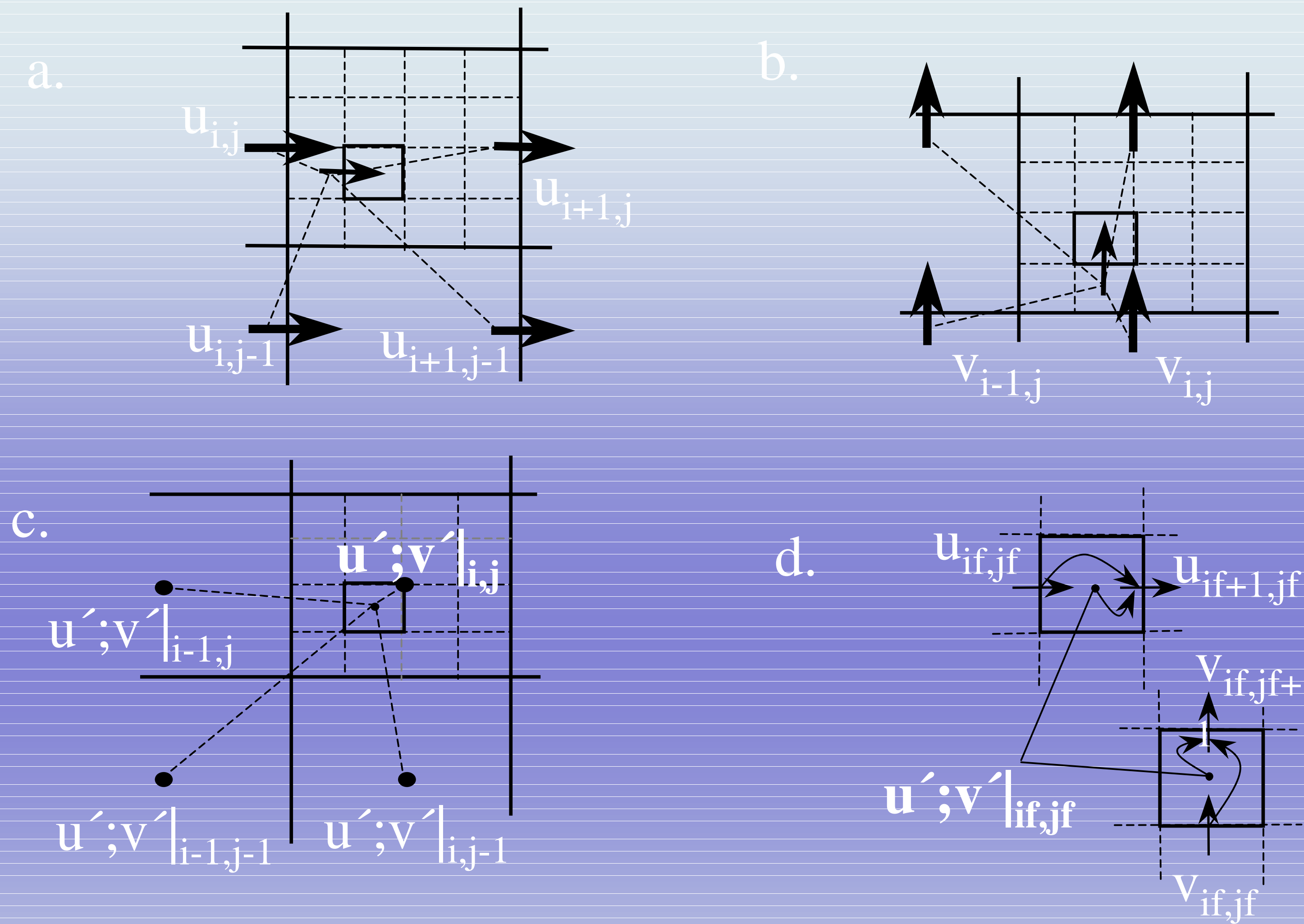


Figure 2. Interpolation of the velocities from the coarse onto the fine grid:
a.) interpolation of the velocity at the east face of the fine cell from the coarse values;
b.) interpolation of the velocity at the south face of the fine cell from the coarse values;
c.) interpolation of the velocity derivatives in the center of the fine cell from the coarse values;
d.) computation of the west and south face velocities.