

Direct Numerical Simulation of Fluid Interfaces Influenced by Soluble Surfactant



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Center of
Smart Interfaces

Motivation

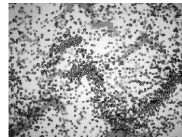
Bubbly Flows in Chemical Engineering



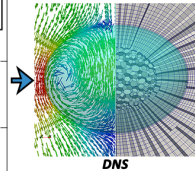
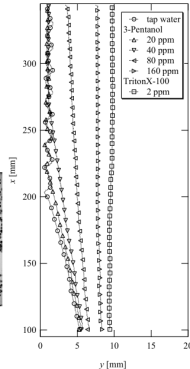
Phenol production, INEOS Phenol, Gladbeck, Germany.



Bubbly flow in tap water



Bubbly flow in 3-Penthanol
solution (40 ppm)



Takagi et. al, *Fluid Dyn. Res.* (2009)

Modeling Concepts

Validation

Results

Outlook



Modeling Concepts I

Mathematical model: Governing Equation Overview

► **Velocity & Pressure fields:** two-phase Navier-Stokes equations

- ▷ transmission conditions at $\Sigma \Rightarrow$ Marangoni term:

$$[[p\mathbf{I} - \mathbb{S}_{\text{visc}}]] \cdot \mathbf{n}_{\Sigma} = \sigma(c_i^{\Sigma}) \kappa_{\Sigma} \mathbf{n}_{\Sigma} + \nabla_{\Sigma} \sigma(c_i^{\Sigma})$$

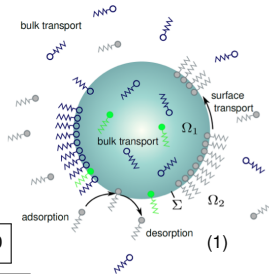
► **Surfactant transport:**

$$\text{In } \Omega \setminus \Sigma(t) : \quad \partial_t c_i + \nabla \cdot (c_i \mathbf{v} + \mathbf{j}_i) = 0$$

$$\text{On } \Sigma(t) : \quad \partial_t^{\Sigma} c_i^{\Sigma} + \nabla_{\Sigma} \cdot (c_i^{\Sigma} \mathbf{v}^{\Sigma} + \mathbf{j}_i^{\Sigma}) = s_i^{\Sigma} \quad (1)$$

with the transmission conditions:

$$s_i^{\Sigma} + [[\mathbf{j}_i \cdot \mathbf{n}_{\Sigma}]] = 0, \quad \text{dilute concentrations } c_i \Rightarrow \mathbf{j}_i = -D_i \nabla c_i \quad (2)$$



Note 1. ∂_t^{Σ} is the Thomas derivative, i.e. the derivative following the normal motion of the interface.

Note 2. Surface divergence contains a **curvature** term, $\kappa_{\Sigma} = \nabla_{\Sigma} \cdot (-\mathbf{n}_{\Sigma})$:

$$\nabla_{\Sigma} \cdot (c_i^{\Sigma} \mathbf{v}^{\Sigma}) = \nabla_{\Sigma} \cdot (c_i^{\Sigma} \mathbf{v}_{||}^{\Sigma}) - c_i^{\Sigma} \kappa_{\Sigma} V_{\Sigma}$$



- ▶ Diffusion-controlled sorption (fast) → sorption process much faster than diffusive transport

$$s_i^{\text{ads}}(c_{i|\Sigma}, c_i^\Sigma) = s_i^{\text{des}}(c_i^\Sigma) \Rightarrow c_i^\Sigma = f(c_{i|\Sigma}), \quad (4)$$

where $f(c_{i|\Sigma})$ is the **adsorption isotherm**.

Then:

$$s_{i,\text{fast}}^\Sigma = - \llbracket \mathbf{j}_i \cdot \mathbf{n}_\Sigma \rrbracket. \quad (5)$$

- ▶ Kinetically-controlled sorption (slow) → diffusive transport faster than sorption process (kinetic barrier).

Then:

$$s_{i,\text{slow}}^\Sigma = s_i^{\text{ads}}(c_{i|\Sigma}, c_i^\Sigma) - s_i^{\text{des}}(c_i^\Sigma). \quad (6)$$

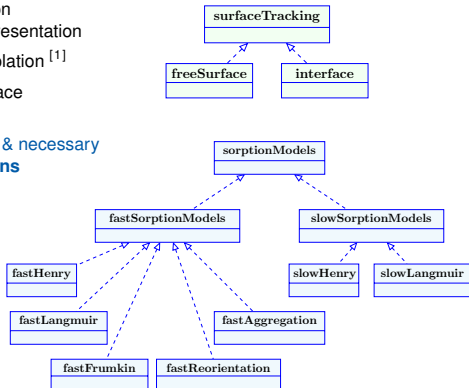


► Hydrodynamics

- Arbitrary Lagrangian Eulerian (**ALE**) formulation of governing equations + **Sharp interface** representation
- **PISO** algorithm + corrected Rhie-Chow interpolation ^[1]
- **Moving mesh support** based on Discrete Space Conservation Law ^[2] (D_SCL)
- re-organization of the **surfaceTracking** library & necessary changes in the **interfacial boundary conditions**

► Surfactant Transport:

- Transport processes bulk \Rightarrow **FV Method**
- Transport processes interface \Rightarrow **FA Method** + correction in *gaussFaGrad*
- Dedicated **sorption library**



[1] Z. Tuković and H. Jasak, "A moving mesh Finite Volume Interface Tracking method for surface tension dominated interfacial fluid flow," *Computers & Fluids* 55 (2012).

[2] I. Demirdžić and M. Perić, "Space conservation law in Finite Volume calculations of fluid flow," *International Journal for Numerical Methods in Fluids* 8/9 (1988).

Modeling Concepts IV

Numerical Model: Algorithm Overview

► Surfactant transport and adsorption

- **sorptionModels** library ^[3] with *runTimeSelectionTable* for slow/fast adsorption processes

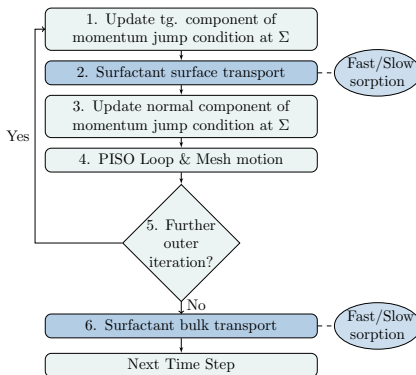
Step.2 Source term for c^Σ eq.

- **slow:** $s^\Sigma = s^{\text{ads}}(c|_\Sigma, c^\Sigma) - s^{\text{des}}(c^\Sigma)$
- **fast:** $s^\Sigma = -D(\mathbf{n} \cdot \nabla c)_\Sigma$

Step.6 Boundary condition for c eq.

- **slow:** $\mathbf{n} \cdot \nabla c = -s^\Sigma/D$
- **fast:** $c|_\Sigma = f^{-1}(c^\Sigma)$

- **Local** update of the surface tension: $\sigma(c^\Sigma)$



[3] C. Pesci et al., "Finite Volume/Finite Area Interface Tracking method for two-phase flows with fluid interfaces influenced by surfactant," in M. T. Rahni, M. Karbaschi, and R. Miller (eds.), *Progress in colloid and interface science* (CRC Press, Taylor & Francis Group, 2015).

Modeling Concepts

Validation

Results

Outlook

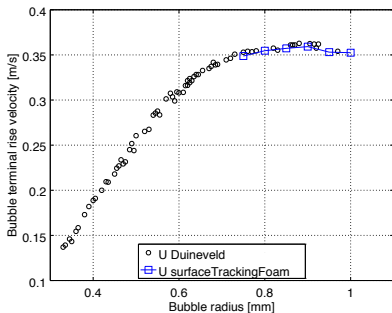


Validation I

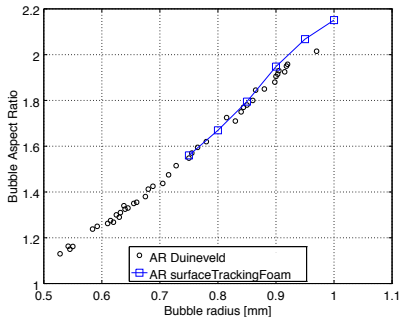
Hydrodynamics

Air bubbles rising into pure water, $T = 293\text{K}$

$$\rho_A = 998.3\text{kg/m}^3, \mu_A = 1 \cdot 10^{-3}\text{kg}/(\text{ms}), \rho_B = 1.205\text{kg/m}^3, \mu_B = 1.82 \cdot 10^{-5}\text{kg}/(\text{ms})$$



(a) Terminal rise velocity.



(b) Aspect ratio.

Fig. 1: Simulation results compared to experimental ones from Duineveld [4].

[4] P. C. Duineveld, "The rise velocity and shape of bubbles in pure water at high Reynolds number," *Journal of Fluid Mechanics* 292 (1995).

Validation II

Sorption source term: slow adsorption

Semi-analytical solution ^[5,6] of $\partial_t c = \frac{D}{r^2} \partial_r (r^2 \partial_r c)$, $\partial_t c^\Sigma = k_{\text{ads}} c|_{r=r_b}$

for short times or for infinite domains: $c(t, r)$, $c^\Sigma(t) = \frac{r_b c_0 \text{Bi}^{\text{S}}}{c_{\infty}^\Sigma} \cdot \sum_{k=1}^{\infty} \frac{C_k}{\omega_k^2} \left(1 - e^{-\omega_k^2 t}\right)$

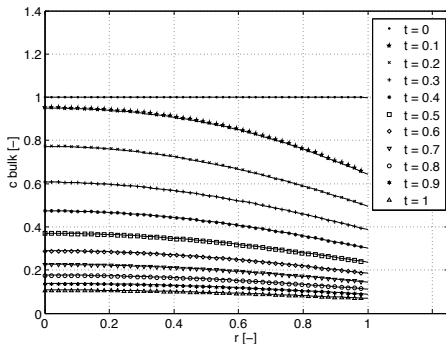
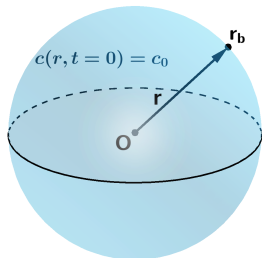


Fig. 2: $c(r)$ for different time instances.

- [5] M. Muradoglu and G. Tryggvason, "A front-tracking method for computation of interfacial flows with soluble surfactants," *Journal of Computational Physics* 227/4 (2008).
- [6] Y. A. Cengel and M. A. Boles, *Thermodynamics: An Engineering Approach* (McGraw-Hill College, 2001).

Validation III

Sorption source term: fast adsorption

Semi-analytical solution^[7] of $\partial_t c = \frac{D}{r^2} \partial_r (r^2 \partial_r c)$, $\partial_t c^\Sigma = -(n \cdot \nabla c)|_{r=r_B}$

for short times or for infinite domains: $c(t, r)$, $c^\Sigma(t) = \frac{r_b(c_0 - c_\infty)}{c_\Sigma^\infty} \cdot \sum_{k=1}^{\infty} \beta(k) \cdot (1 - e^{-\omega_k^2 t})$,

with $\beta(k) = C_k(\sin \omega_k - \omega_k \cos \omega_k) / \omega_k^2$.

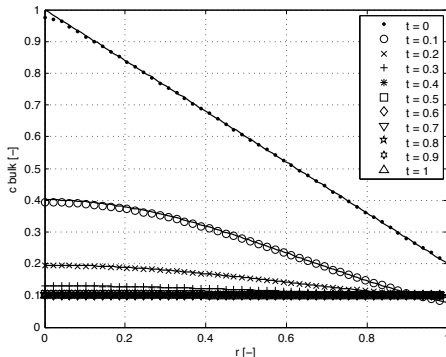
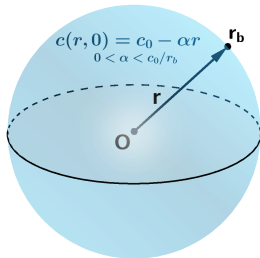


Fig. 3: $c(r)$ for different time instances.

[7] C. Pesci, H. Marschall and D. Bothe, unpublished work within the framework of the DFG priority program SPP 1506, 2015.

Modeling Concepts

Validation

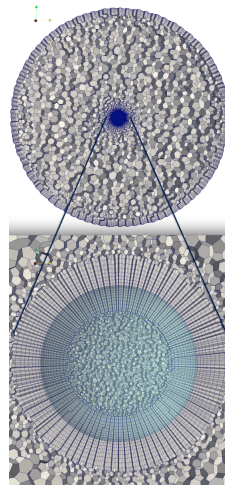
Results

Outlook

Results I

Case set-up

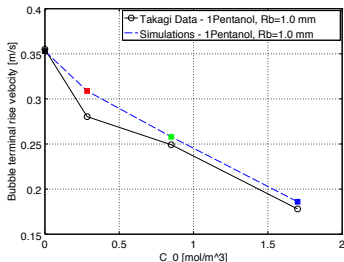
- ▶ Deformable air **bubble rising** in contaminated water with **soluble surfactant** (and **mass transfer**)
- ▶ **Moving reference frame** that follows the bubble
- ▶ **3D unstructured** polyhedral/prismatic mesh
- ▶ Initial shape of the bubble: sphere in a spherical domain
- ▶ Liquid phase \equiv A, gas phase \equiv B
- ▶ Bubble radius: $r_b = 0.65 \dots 1.0$ mm.
Outer radius: $r_d = 20 r_b$
- ▶ Initial condition for surfactant:
 - ▶ $c_i^A(t=0) = c_{i,0}$,
 - ▶ $c_i^B(t) = 0$,
 - ▶ $c_i^\Sigma(t=0) = 0$



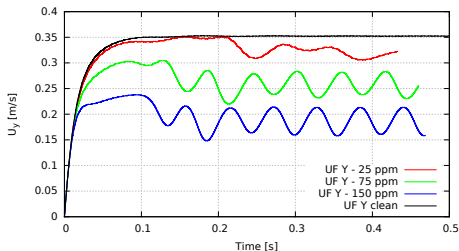
Results I

Surfactant Effects I

Air bubbles rising into contaminated water with 1-Pentanol, $r_B = 1.0\text{mm}$
Sorption model: *slow Langmuir*, $c_0 = [0, 25, 75, 150]\text{ppm}$



(a) Simulation results compared to experiments from Tagawa and Takagi [8].



(b) Transient rise velocity.

Fig. 4: Terminal rise velocities.

[8] Y. Tagawa, S. Takagi and Y. Matsumoto, "Surfactant effects on path instability of a rising bubble," *Journal of Fluid Mechanics* 378 (2014).

Results II

Surfactant Effects II

Air bubbles rising into contaminated water with 1-Penthanol, $r_B = 1.0\text{mm}$
Sorption model: *slow Langmuir*, $c_0 = 150\text{ppm}$

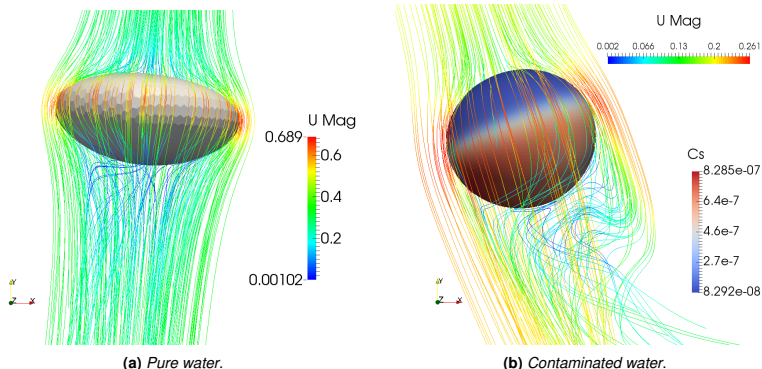
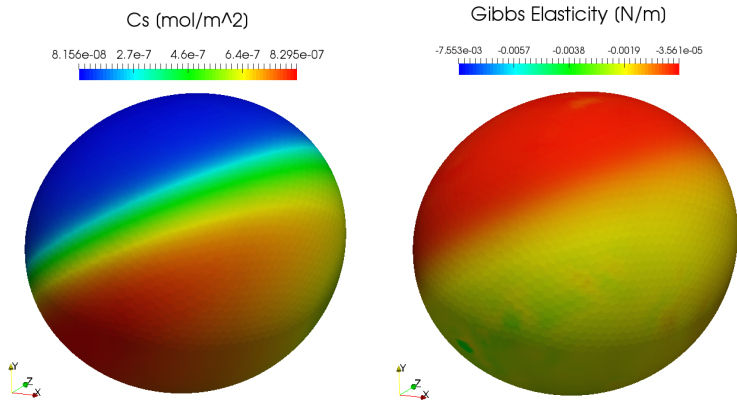


Fig. 5: Streamlines colored by the absolute velocity magnitude.

Results III

Surfactant Effects III

Gibbs elasticity: $E^\Sigma = \sigma'(c_\Sigma)c_\Sigma$



(a) Surfactant distribution on Σ .

(b) Gibbs elasticity.

Fig. 6: Bubble interface in contaminated water, local quantities.

Results IV

Surfactant Effects IV

Air bubbles rising into contaminated water with 1-Penthanol, $r_B = 1.0\text{mm}$
Sorption model: *slow Langmuir*, $c_0 = [25, 75, 150, 210]\text{ppm}$

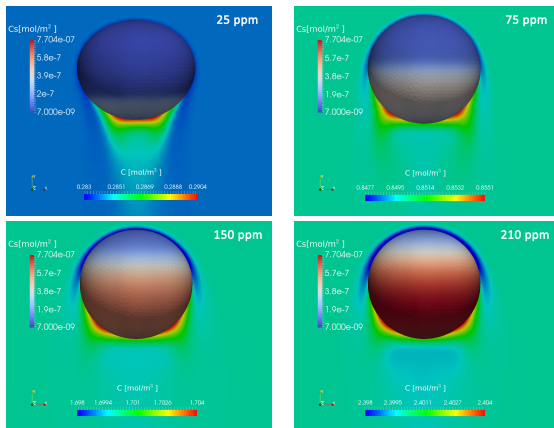


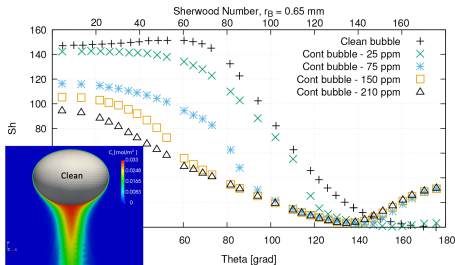
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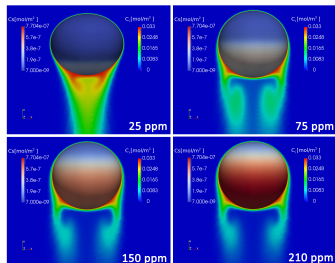
Results V

Mass transfer influenced by surfactant

Air bubbles rising into contaminated water with 1-Penthanol, $r_B = 1.0\text{mm}$
 Sorption model: *slow Langmuir*, $c_0 = [0, 25, 75, 150, 210]\text{ppm}$



(a) Local Sherwood number, $Sh_{loc} = \frac{\partial c / \partial \mathbf{n}}{(c|_{\Sigma} - c_{\infty}) / d_b}$.



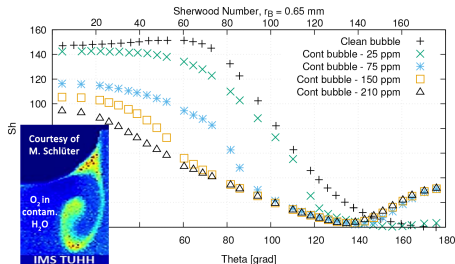
(b) Species concentration in the liquid phase, surface colored by c^{Σ} .

Fig. 8: Mass transfer under the influence of surfactant.

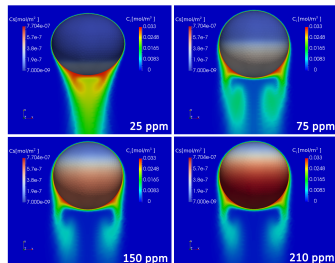
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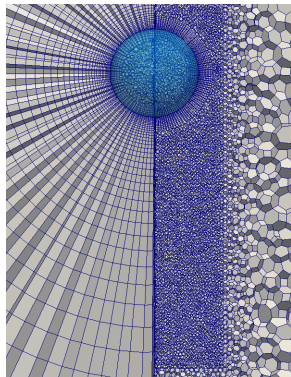
Outlook

► Physics:

- Further investigation of mass transfer under the influence of surfactants
 - Polyhedral meshes with refinement regions
 - Parameter studies: surfactants modelling & effects on mass transfer including hindrance ^[9]
- Taylor Bubble benchmark problem - Comparison with experiments by Prof. M. Schlüter's group (IMS, TUHH) ^[10]
- Partitioning phenomena ^[11]

► Numerics:

- Two-mesh approach (by P. Weber) for partitioning
- Improve parallelization
- Improve solver performances



[9] D. Bothe, "On the multi-physics of mass-transfer across fluid interfaces," *arXiv:1501.05610* (2015).

[10] S. Kastens et al., "Mass Transfer from Single Taylor Bubbles in Minichannels," *Chemical Engineering & Technology* 38/11 (2015).

[11] F. Ravera, M. Ferrari and L. Liggieri, "Adsorption and partitioning of surfactants in liquid-liquid systems," *Advances in Colloid and Interface Science* 88 (2000).

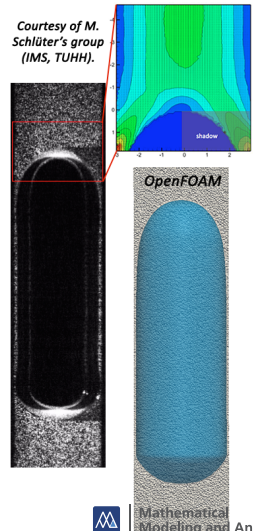
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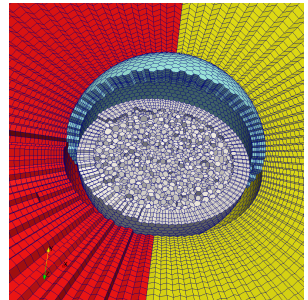


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- ▶ **H. Jasak** and **Ž. Tuković**, University of Zagreb for the software support.

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End ...



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Mathematical
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- ▷ **Bothe, D.**
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"The rise velocity and shape of bubbles in pure water at high Reynolds number."
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Journal of Fluid Mechanics 378 (2014): 124–142.

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"A moving mesh Finite Volume Interface Tracking method for surface tension dominated interfacial fluid flow."
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“A tensorial approach to computational continuum mechanics using object-oriented techniques.”

Computers in Physics 12 (1998): 620–631.