A phase-field method for numerical simulation of interfacial two-phase flows using OpenFOAM

Holger Marschall[1], Xuan Cai[2,*], Martin Wörner[3], Olaf Deutschmann[2,3]

[1] Mathematical Modeling and Analysis (MMA), Thermo-Fluids & Interfaces, Technische Universität Darmstadt
[2] Institute of Technical Chemistry and Polymer Chemistry (ITCP), Karlsruhe Institute of Technology (KIT)
[3] Institute of Catalysis Research and Technology (IKFT), Karlsruhe Institute of Technology (KIT)
* Present address: Robert Bosch GmbH, Stuttgart

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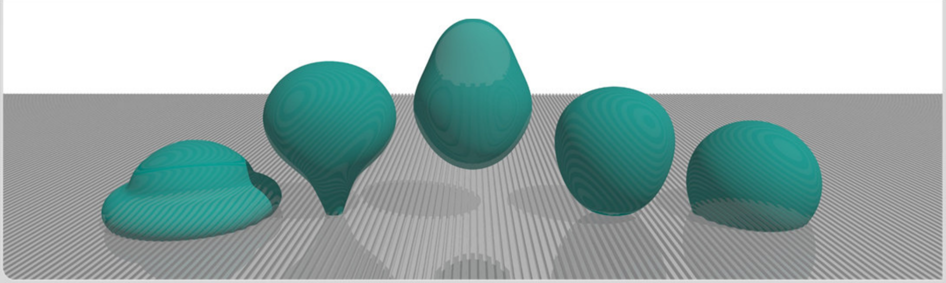
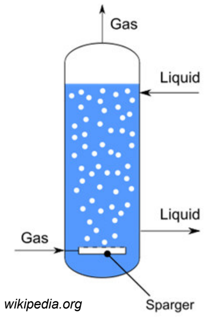


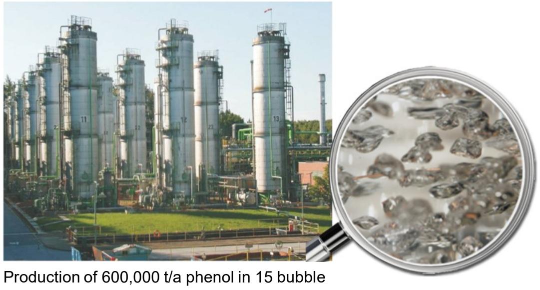
Outline

- Introduction
  - Examples for multiphase flows in chemical process engineering
  - Why phase-field method for interface resolving simulations?
- Mathematical approach
  - Phase-field method for gas-liquid flows
  - Numerical implementation in OpenFOAM®
  - Characterization of “spurious currents”
- Sample applications related to process engineering
  - Cutting of a rising bubble by a horizontal wire
  - Drop impact on a solid surface
- Summary and outlook

Bubble column reactors



wikipedia.org



Production of 600,000 t/a phenol in 15 bubble columns by INEOS Phenol, Gladbeck, Germany

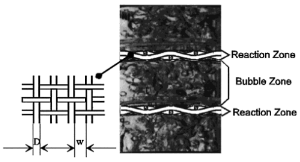


Figure 1. Bubble column staged with structured fibrous catalysts.

- Reactor internals serve to fragment bubbles and increase interfacial area
- Wire meshes serve as catalyst support
- **Does the interaction between bubbles and mesh depend on wire wettability?**

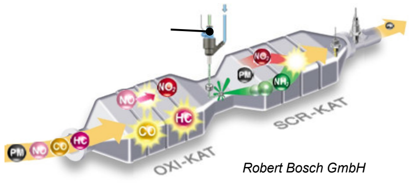
🕮 V. Höller et al., *Ind. Eng. Chem. Res.* **40** (2001) 1575–1579

Diesel exhaust gas aftertreatment

- **Selective catalytic reduction (SCR):** converting nitrogen oxides (No_x) with the help of ammonia (NH_3) to harmless nitrogen and water

$$2\,NH_3 + NO + NO_2 \rightarrow 2\,N_2 + 3\,H_2O$$

- Ammonia is provided by spray injection of urea-water-solution (AdBlue®)



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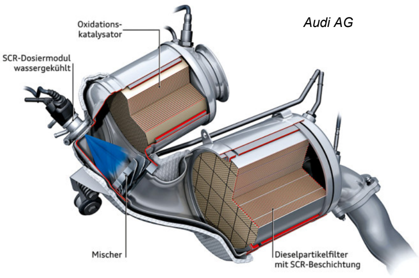
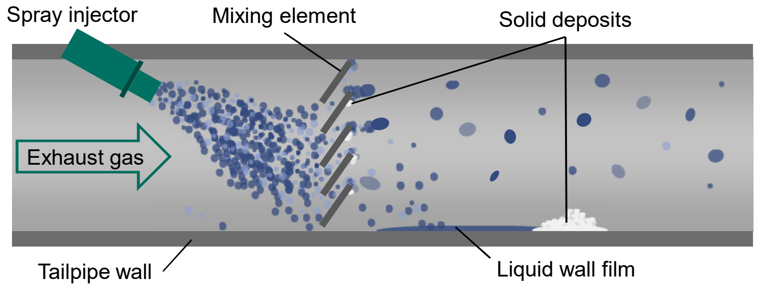
$$(NH_2)_2CO\ (l) \Rightarrow NH_3\ (g) + HNCO\ (g)$$
$$HNCO\ (g) + H_2O\ (g) \Rightarrow NH_3\ (g) + CO_2\ (g)$$



Audi AG

Challenges in NH_3 treatment

- Solid deposit formation by incomplete drop evaporation



- How can the contact of droplets with the tailpipe wall be minimized in order to avoid film formation?
- *<u>Approach of group:</u> Study fundamental gas-liquid flow phenomena by interface resolving numerical simulations*

Simulation challenges

- Topological changes
  - Breakup/coalescence of bubbles/droplets
  - Liquid film formation
- Wetting behavior
  - Conflict between contact line motion and no-slip boundary condition
- Dominance of surface tension forces at small scales
  - Numerical artifact of “spurious currents”
- *Phase-field method offers conceptual advantages as compared to sharp-interface methods*

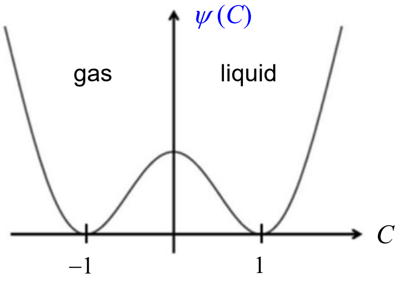
Phase-field method

- Free energy of two-phase mixture (m = mixture) [J]

$$\mathcal{F}_\mathrm{m} = \int_\Omega f_\mathrm{m}(C,\nabla C)\,\mathrm{d}\Omega$$

$$f_\mathrm{m}(C,\nabla C) = \underbrace{\frac{\lambda}{\varepsilon^2}\psi(C)}_{\text{bulk energy}} + \underbrace{\frac{\lambda}{2}\left|\nabla C\right|^2}_{\text{gradient energy}}$$

$$\underbrace{\psi(C) = \frac{1}{4}(1-C^2)^2}_{\text{Ginzburg-Landau double-well potential}}$$



  - C = order parameter [-]
  - ε = capillary width [m]
  - λ = mixing energy density [J/m]
  - f_m= free energy density [J/m^3]

Equilibrium profile and surface tension

- Bulk chemical potential [J/m^3]

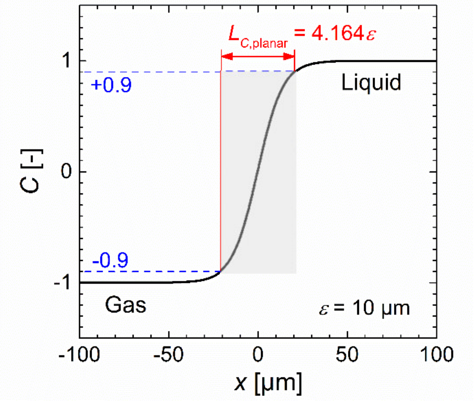
$$\phi_\mathrm{m} = \frac{\delta\mathcal{F}}{\delta C} = \lambda\varepsilon^{-2}\underbrace{(C^3 - C)}_{=\psi'} - \lambda\nabla^2 C$$

- 1D equilibrium profile

$$C(x) = \tanh\left(\frac{x}{\sqrt{2}\varepsilon}\right)$$

- Interfacial tension

$$\sigma = \int_{-\infty}^{\infty} \lambda \left(\frac{\mathrm{d}C}{\mathrm{d}x}\right)^2 \mathrm{d}x = \frac{\sqrt{8}}{3}\frac{\lambda}{\varepsilon}$$



Wetting boundary condition

- Free energy at solid wall [J]

$$\mathcal{F}_{\mathrm{w}} = \int_{\partial\Omega_{\mathrm{w}}} f_{\mathrm{w}}(C)\,\mathrm{d}A$$

- Free energy density of wall (static contact angle θ_{s})

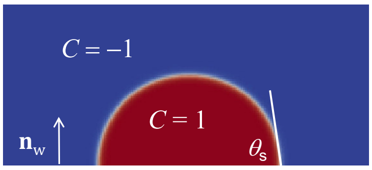
$$f_{\mathrm{w}}(C) = -\frac{\sigma}{4}(3C - C^3)\cos\theta_s + \frac{\sigma_{\mathrm{w1}} + \sigma_{\mathrm{w2}}}{2} \qquad \underbrace{\sigma_{\mathrm{w2}} - \sigma_{\mathrm{w1}} = \sigma\cos\theta_{\mathrm{s}}}_{\text{Young-Dupre equation}}$$

- Wall chemical potential [J/m^2]

$$\phi_{\mathrm{w}} = \frac{\delta\mathcal{F}_{\mathrm{w}}}{\delta C} = \lambda\mathbf{n}_{\mathrm{w}}\cdot\nabla C - \frac{3}{4}\sigma(1 - C^2)\cos\theta_{\mathrm{s}}$$

- Equilibrium condition

$$\mathbf{n}_{\mathrm{w}}\cdot\nabla C = \partial_{n,\mathrm{w}}C = \frac{1 - C^2}{\sqrt{2}\varepsilon}\cos\theta_{\mathrm{s}}$$



Cahn-Hilliard equation

- Convective Cahn-Hilliard equation

$$\partial_t C + \nabla\cdot(C\,\mathbf{u}) = M\nabla^2\phi_{\mathrm{m}} \qquad \phi_{\mathrm{m}} = \lambda\varepsilon^{-2}(C^3 - C) - \lambda\nabla^2 C$$

  - $\mathbf{u}$ = velocity field [m/s]
  - M = constant mobility parameter [m^{3}s/kg]
  - Fourth order derivatives
- Boundary conditions
  - Zero gradient of chemical potential $\partial_n\phi_{\mathrm{m}} = 0$
  - Wettability condition at wall $\partial_{n,\mathrm{w}}C = 2^{-1/2}(1 - C^2)\varepsilon^{-1}\cos\theta_{\mathrm{s}}$
- No-slip condition for velocity field
  - Diffusive mechanism for motion of the contact line

$$\mathbf{u}_{\mathrm{w}} = \mathbf{0} \quad\rightarrow\quad \partial_t C = M\nabla^2\phi_{\mathrm{m}}$$

📖 J.W. Cahn, J.E. Hilliard, *J. Chem. Phys.* **28** (1957) 258–267

Single field Navier-Stokes equations

- Two incompressible Newtonian fluids

$$\nabla\cdot\mathbf{u}=0$$

$$\partial_t(\rho_C\mathbf{u})+\nabla\cdot(\rho_C\mathbf{u}\otimes\mathbf{u})=-\nabla p+\nabla\cdot\left[\mu_C\left(\nabla\mathbf{u}+(\nabla\mathbf{u})^{\mathrm{T}}\right)\right]+\rho_C\mathbf{g}+\mathbf{f}_\sigma$$

$$\rho_C=\frac{1+C}{2}\rho_{\mathrm{L}}+\frac{1-C}{2}\rho_{\mathrm{G}},\qquad \mu_C=\frac{1+C}{2}\mu_{\mathrm{L}}+\frac{1-C}{2}\mu_{\mathrm{G}},\qquad \mathbf{f}_\sigma=-C\nabla\phi_{\mathrm{m}}$$

- Determining the phase field parameters ε, λ, M
  - Cahn number $Cn=\varepsilon/L=\mathcal{O}(10^{-2})$
    - L = characteristic macroscopic length scale (e.g. bubble diameter)
  - Mixing energy parameter $\lambda=3\varepsilon\sigma/\sqrt{8}$
  - Mobility parameter $M=\chi\varepsilon^2$
    - Proportionality factor χ [m·s/kg] $\chi=\mathcal{O}(10^{-1}-10^{1})$

📖 D. Jacqmin, *J. Comput. Phys.* **155** (1999) 96–127
📖 J. Kim, *Commun. Comput. Phys.* **12** (2012) 613–661

Computer code *phaseFieldFoam*

- Code development
  - Dr. Holger Marschall (TU Darmstadt)
  - Dr. Xuan Cai (KIT, now at Bosch)
  - …
- Implementation in Open∇FOAM
  - **C++ toolbox** for the development of customized numerical solvers for solution of problems in continuum physics including **computational fluid mechanics** (CFD)
  - foam-extend-1.6, foam-extend-3.2, foam-extend-4.0
- Validation and application for various test cases
  - typical mesh resolution $h=\varepsilon/2\rightarrow 8$ cells in diffuse interface

📖 X. Cai, H. Marschall, M. Wörner, O. Deutschmann, *Chem. Eng. Technol.* **38** (2015) 1985–1992
📖 X. Cai, M. Wörner, H. Marschall, O. Deutschmann, *Catalysis Today* **273** (2016) 151–160
📖 X. Cai, M. Wörner, H. Marschall, O. Deutschmann, *Emission Control Science and Technology* **3** (2017) 289–301

Numerical method

- Finite volume method on general grids
- Cahn-Hilliard or Allen-Cahn approach
- Relative density flux term in NS equation for better volume conservation at high density ratios
  - similar to Ding et al. (2007) and Abels et al. (2012)
- Segregated or coupled solution of CH-NS equations
- PISO algorithm for pressure-velocity coupling
- 2nd order schemes in space and time
- Code shall be released to the public after forthcoming publication (Marschall et al. 2019)

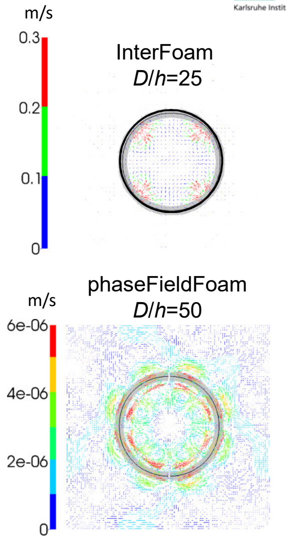
📖 H. Ding, P.D.M. Spelt, C. Shu, *J. Comput. Phys.* **226** (2007) 2078–2095

📖 H. Abels, H. Garcke, G. Grün, *Math. Mod. Meth. Appl. S.* **22** (2012) 1150013

📖 H. Marschall et al., Conservative and bounded finite volume discretization of diffuse interface models on general grids with application to dynamic wetting, in preparation (2019)

Spurious currents (SC)

- Test problem
  - Bubble or drop in absence of gravity
  - Pressure gradient and surface tension forces are in equilibrium
  - Both phases are at rest
- Numerical simulations
  - Numerical inaccuracies induce unphysical artificial flows U_{sc}
  - Diameter ↓ ⇒ U_{sc} ↑
  - Comparison phaseFieldFoam (PFF) with volume-of-fluid method (InterFoam, IF) for submillimeter air bubble (D=500µm) in water



h = mesh size

📖 F. Jamshidi et al., *Comp. Phys. Commun.* **236** (2019) 72–85

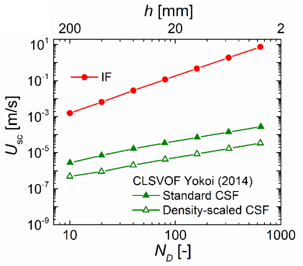
SC under mesh refinement

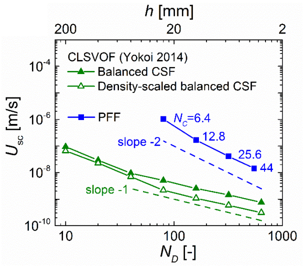
- Continuum surface force (CSF) model*
  - Curvature (κ) convergence with mesh refinement is hard to achieve for sharp interface methods

$$\mathbf{f}_\sigma = \sigma\kappa\delta_h\mathbf{n}_{\text{int}}$$

- Phase field method
  - Free energy formulation does not require curvature computation
  - Convergence of U_{sc} under mesh refinement without special measures

$$\mathbf{f}_\sigma = -C\nabla\phi_{\text{m}}$$





- *J.U. Brackbill, D.B. Kothe, C. Zemach, *J. Comput. Phys.* **100** (1992) 335–354
- K. Yokoi, *J. Comput. Phys.* **278** (2014) 221–228
- F. Jamshidi, H. Heimel, M. Hasert, X. Cai, O. Deutschmann, H. Marschall, M. Wörner, *Comp. Phys. Commun.* **236** (2019) 72–85

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Wire-induced bubble cutting

- Wire meshes as catalyst support
- Interaction between bubble and wire
  - Understand bubble cutting process
  - Does it depend on wire wettability?
- Own experiments by P. Rohlfs (2018)
  - Set-up similar to Segers (2015)
  - Viscous water-glycerol solution
  - Wires with different wettability
    - Glass, stainless steel, Teflon
  - Wire/cylinder diameter d_{cyl} = 3 – 5 mm
  - Bubble volume V_B = 50 – 1000 µL

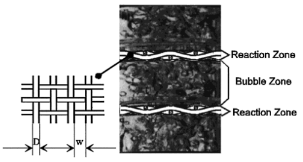
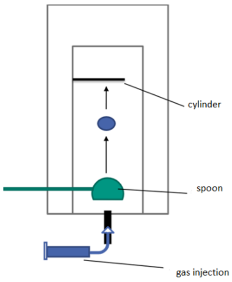


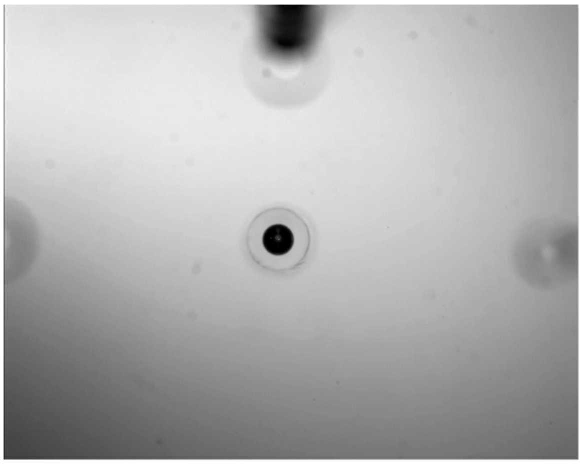
Figure 1. Bubble column staged with structured fibrous catalysts.



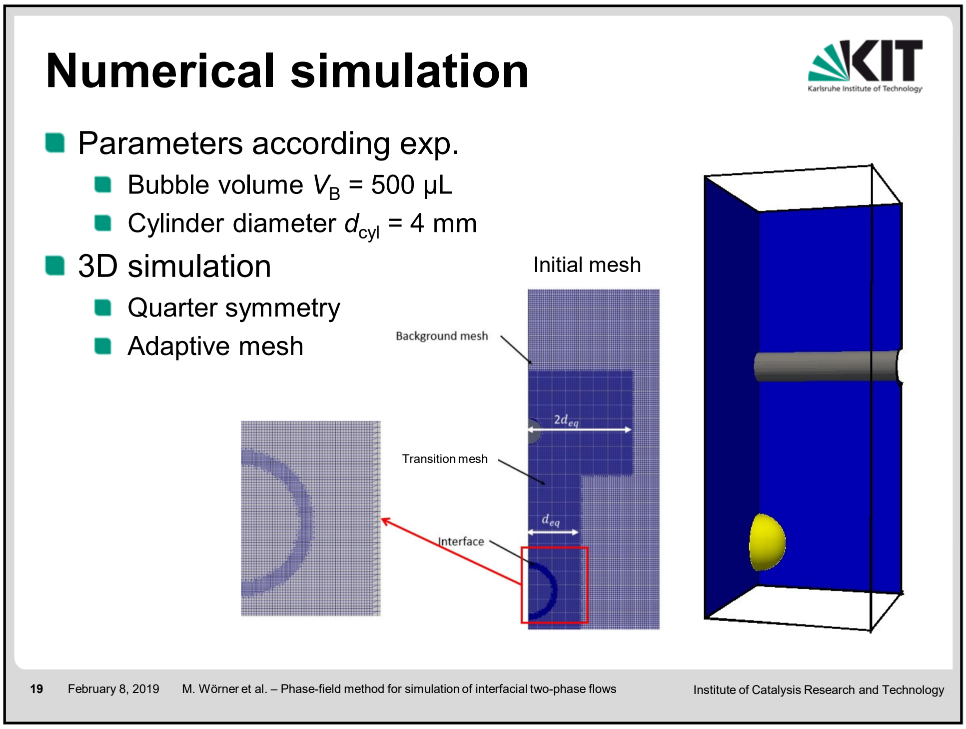
🕮 V. Höller, K. Radevic, L. Kiwi-Minsker, A. Renken, *Ind. Eng. Chem. Res.* **40** (2001) 1575–1579
🕮 Q. Segers, Cutting Bubbles using Wire-Mesh Structures - Direct Numerical Simulations, PhD thesis, TU Eindhoven 2015
🕮 P. Rohlfs, Einfluss des Benetzungsverhaltens auf die Blasenzerteilung an einem Zylinder, Bachelor thesis, KIT 2018

Bubble cutting in experiment

- Hollow glass cylinder d_{cyl} = 4 mm, V_B = 1000 µL



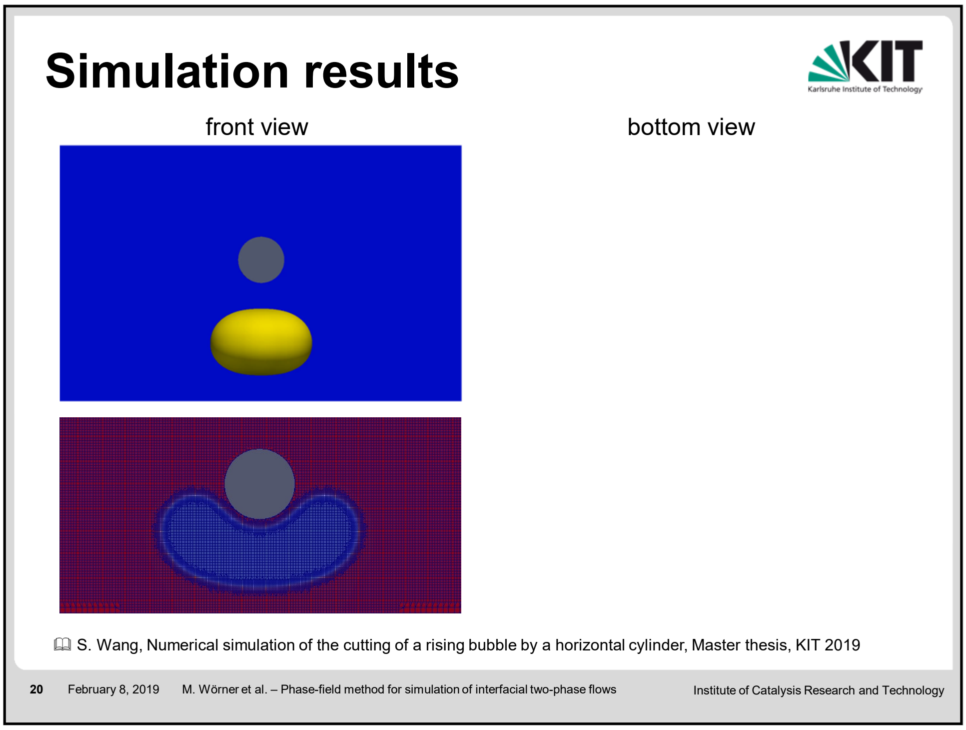
🕮 P. Rohls, Einfluss des Benetzungsverhaltens auf die Blasenzerteilung an einem Zylinder, Bachelor thesis, KIT 2018



Numerical simulation

- Parameters according exp.
  - Bubble volume V_B = 500 µL
  - Cylinder diameter d_{cyl} = 4 mm
- 3D simulation
  - Quarter symmetry
  - Adaptive mesh

Initial mesh

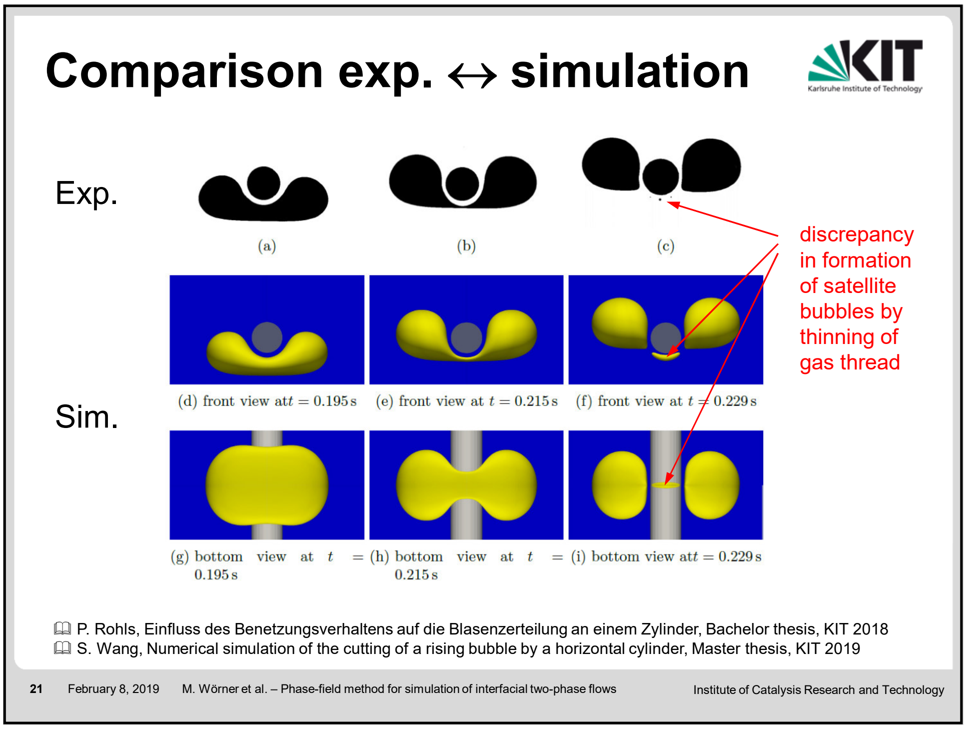


Simulation results

front view

bottom view

S. Wang, Numerical simulation of the cutting of a rising bubble by a horizontal cylinder, Master thesis, KIT 2019



Comparison exp. ↔ simulation

Exp.

(a) (b) (c)

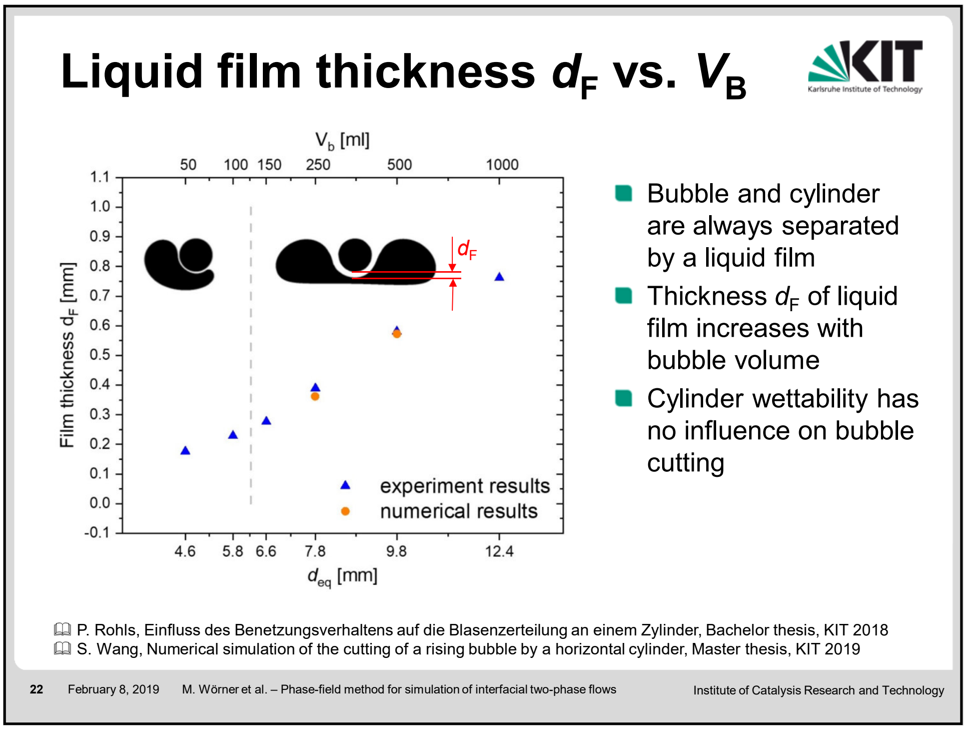
Sim.

(d) front view at $t = 0.195$ s (e) front view at $t = 0.215$ s (f) front view at $t = 0.229$ s

(g) bottom view at $t = 0.195$ s (h) bottom view at $t = 0.215$ s (i) bottom view at $t = 0.229$ s

discrepancy in formation of satellite bubbles by thinning of gas thread

🕮 P. Rohls, Einfluss des Benetzungsverhaltens auf die Blasenzerteilung an einem Zylinder, Bachelor thesis, KIT 2018
🕮 S. Wang, Numerical simulation of the cutting of a rising bubble by a horizontal cylinder, Master thesis, KIT 2019



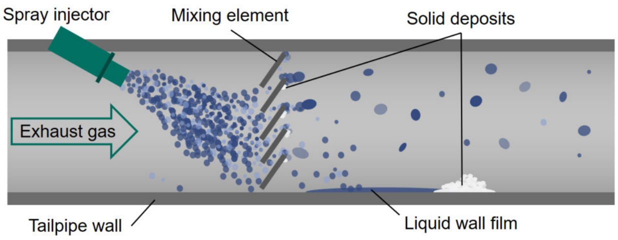
Liquid film thickness d_F vs. V_B

- Bubble and cylinder are always separated by a liquid film
- Thickness d_F of liquid film increases with bubble volume
- Cylinder wettability has no influence on bubble cutting

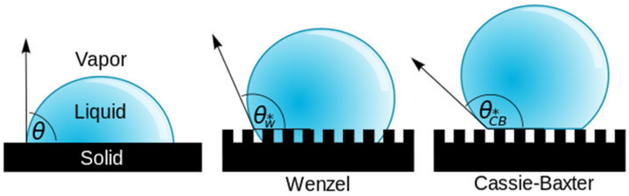
🕮 P. Rohls, Einfluss des Benetzungsverhaltens auf die Blasenzerteilung an einem Zylinder, Bachelor thesis, KIT 2018
🕮 S. Wang, Numerical simulation of the cutting of a rising bubble by a horizontal cylinder, Master thesis, KIT 2019

Drop impact on a solid surface

- How can the contact of droplets with a wall be minimized?

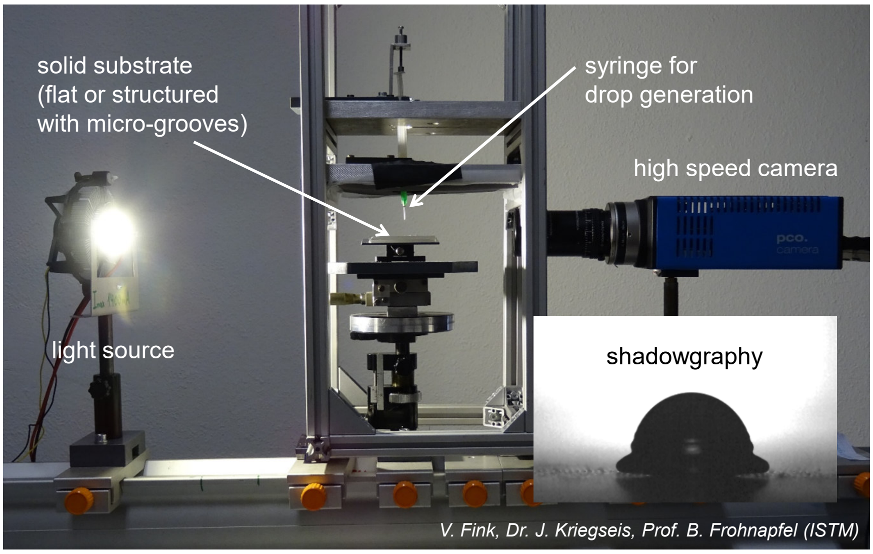


- Surface **roughness** reduces wettability / increases θ



Source: Wikipedia

Experimental setup



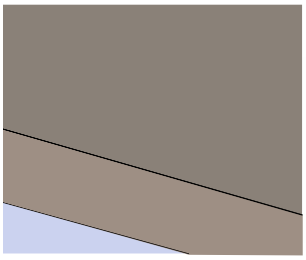
V. Fink, Dr. J. Kriegseis, Prof. B. Frohnapfel (ISTM)

Experiment – impact outcomes

- Hydrophobic PDMS substrate
  - Flat surface: θ = 100.3°, mean roughness depth 0.56 µm
- Water drop with diameter D_0 = 2.1 mm
- Drop impact velocity U_0 = 0.62 m/s (Re = 1300, We = 11)

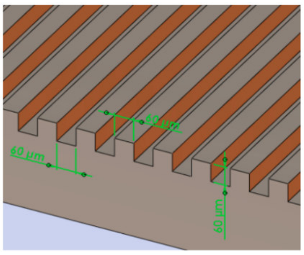
Flat Surface

Deposition



Surface with micro-grooves (60 µm)

Rebound



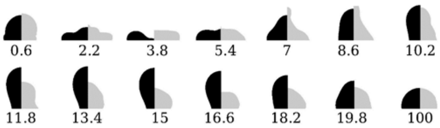
Can this distinct behaviour be reproduced numerically?

Flat surface – comparison

Simulation

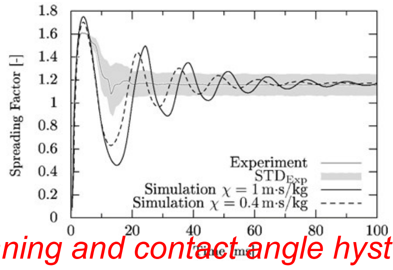
Experiment

- Instantaneous drop shape



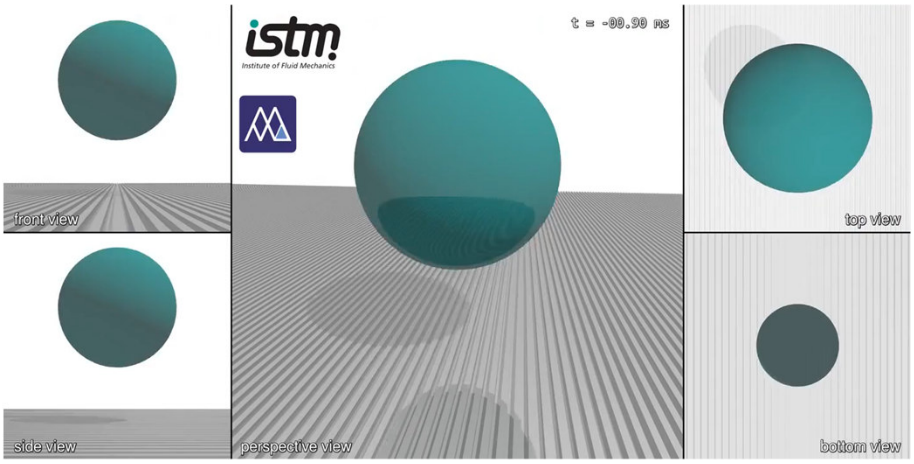
time in milliseconds

- Spreading factor



Pinning and contact angle hysteresis

Impact on <u>structured</u> surface (sim.)



📖 V. Fink, X. Cai, A. Stroh, R. Bernard, J. Kriegseis, B. Frohnapfel, H. Marschall, M. Wörner, *Int. J. Heat Fluid Flow* **70** (2018) 271–278

Summary and outlook

- Phase field method for simulation of gas-liquid flows
  - Coupled Cahn-Hilliard-Navier-Stokes equations
  - Implementation in OpenFOAM® (code *phaseFieldFoam*)
  - Comprehensive validation for various flow phenomena
- Achievements and current limitations
  - Method can well describe wetting phenomena ✓
  - Method can handle real density and viscosity ratios ✓
  - Difficulty to choose appropriate value for mobility 👓
  - Method globally conserves *C* but not phase volume 👓
  - Pinning and contact angle hysteresis 👓
  - Surface tension correction for curved interfaces 👓

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  - Boysen foundation (BOY-127)
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  - Patrick Rohlfs
  - Andrea Schillaci
  - Shuo Wang
  - Yanchen Wu

BOYSEN
INNOVATIONEN ABGASTECHNOLOGIE

SFB/Transregio 150
Turbulente, chemisch reagierende
Mehrphasenströmungen in Wandnähe

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  - Verena Fink (ISTM, now Miele)
  - Alexander Stroh (ISTM)
- Festo AG
  - Manuel Hasert
  - Hans Heimel