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Detailed numerical simulation of gas-liquid Taylor flow with heterogeneous chemical reaction

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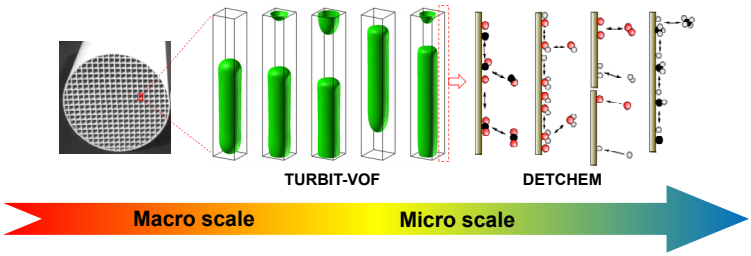
Objectives

- Scale-resolving simulation of reactive two-phase flows in monolith reactors (HA-E-0004 by Helmholtz Energy Alliance)
- The development of a computer code for detailed numerical simulations of heterogeneously catalyzed reactions in gas-liquid flows in a single channel
- Coupling two in-house computer codes
  - TURBIT-VOF[1] for the gas-liquid flows
  - DETCHEM[2] for the reaction kinetics



Mass transfer

- Validation of gas-liquid mass transfer with planar interface
  - Effective diffusivity model for multispecies diffusion
  - Interfacial diffusion model[1] for multiphase diffusion
- Example case: H_2-O_2 reaction-diffusion in water-vapor system with arbitrary reaction rate (k = 1 m/s)

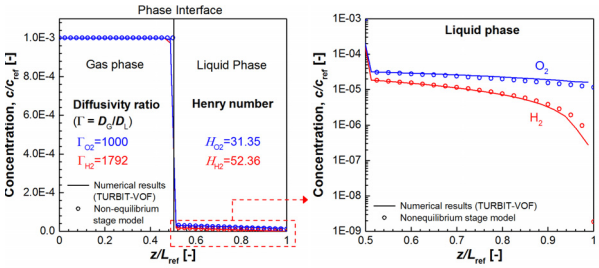


Figure 1. Species distribution for reaction-diffusion of hydrogen and oxygen in water-vapor system. Left: Entire domain, right: zoom-up for liquid area

Reaction

- Validation of reaction for nitrobenzene hydrogenation
  - One-step global reaction kinetics[3]

$$r = k' \frac{K_{NB} c_{NB}}{1 + K_{NB} c_{NB}} \quad \left(\mathrm{mol \cdot g_{Cat}^{-1} \cdot l^{-1} \cdot s^{-1}}\right)$$

k' modified rate constant $[\mathrm{mol \cdot g_{Cat}^{-1} \cdot l^{-1} \cdot s^{-1}}]$

K_{NB} equilibrium constant of adsorption of nitrobenzene $[\mathrm{l \cdot mol^{-1}}]$

c_{NB} concentration of nitrobenzene $[\mathrm{mol \cdot l^{-1}}]$

- Reproduce: nitrobenzene hydrogenation in batch reactor

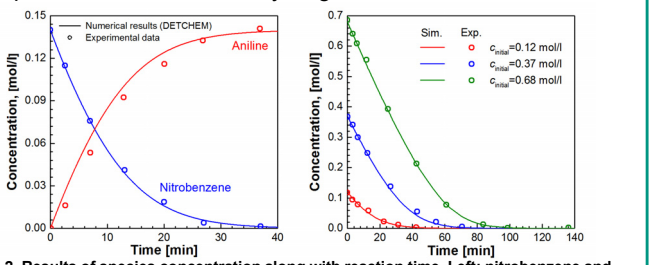
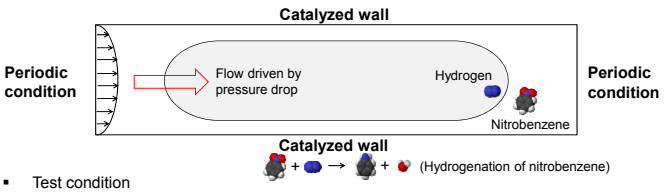


Figure 2. Results of species concentration along with reaction time. Left: nitrobenzene and aniline on Pt-supported catalyst, right: nitrobenzene with different initial concentration on Pd-supported catalyst, experimental data from [3]

Simulation of 2D Taylor flow for hydrogenation of nitrobenzene



- Test condition
  - Temperature: 323 K, pressure: 50 bar
- Properties

	Gas (Hydrogen)	Liquid (nitrobenzene)	Gas/liquid ratio
Density [kg/m³]	5.3	1175.8	222 (assumed to unity)
Viscosity [Pa-s]	7.4E-06	1.4E-3	194
Surface tension [N/m]	-	0.039	-
Diffusivity [m²/s] of hydrogen in	1.7E-4	2.5E-09	66723
Henry coefficient of hydrogen [-]	-	40.4	-

- Dimensionless number
  - Reynolds: 82, Schmidt: 491, Weber: 3.01

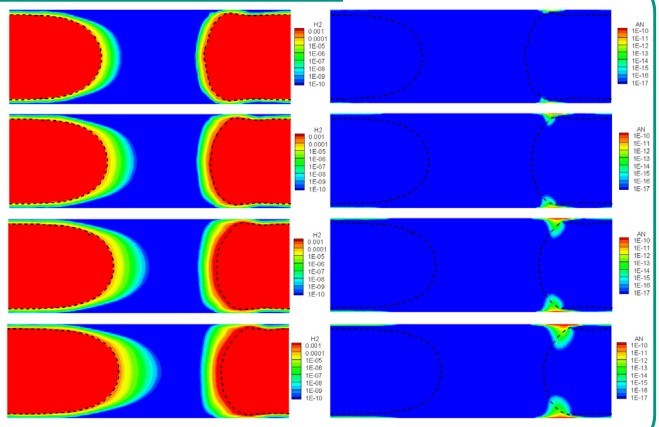


Figure 3. Concentration of hydrogen (reactant) and aniline (product) after 7, 14, 21 and 28 ms

Conclusions and outlook

- The development of solver for two-phase mass transfer with surface reaction is successfully accomplished with validation cases
- Most of hydrogen species reaching catalyzed wall is transported within liquid film region, and aniline is produced at the end of the bubble where liquid film thickness is narrowest
- Simulation with detailed kinetics and/or 3D Taylor flow will be further investigated in the next step

References

[1] Onea, A., Wörner, M. and Cacuci, D. G., Chem. Eng. Sci., 64 (2009) 1416-1435

[2] Deutschmann, O., Tischer, S., Kleditzsch, S., Janardhanan, V., Correa, C., Chatterjee, D., Mladenov, N., Minh, H. D. And Karadeniz, H., DETCHEM™ User Manual, 2012, http://www.detchem.com

[3] Höller, V., Wegricht, D., Yuranov, I., Kiwi-Minsker, L. And Renken A., Chem. Eng. Technol. 23 (2000) 3, 251-255