Micro-fluidized bed membrane reactors: Experimental investigation into hydrodynamics and gas back-mixing

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TU/e Technische Universiteit Eindhoven University of Technology

Where innovation starts

Introduction

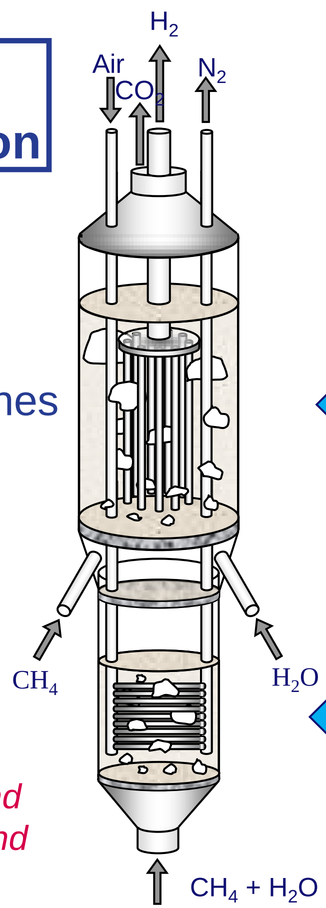
Membrane fluidized beds for pure hydrogen production

Concept 1: CH_4 combustion

H_2 extraction via Pd-membranes

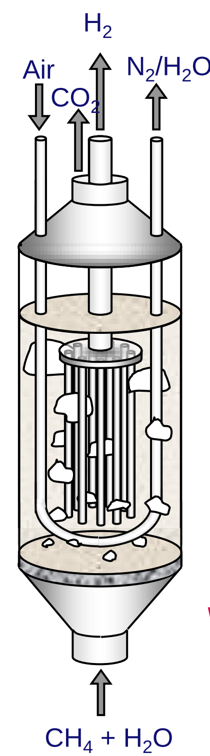
O_2 addition via perovskite membranes

Kuipers, Patil and Van Sint Annaland (2004) US Patent 10/893466



Reforming/shift ~650 °C

CPO ~1000 °C



Concept 2: H_2 combustion

Pd-membranes for H_2 extraction

for product + for energy production

Van Sint Annaland, Nijmeijer, Assink and Kramer (2006) European Patent 06116419

⇒ + integrated CO_2 capture
Complete process integration

Introduction: Fluidized bed membrane reactor (FBMR)

Fluidized bed membrane reactor

Pd-membrane assembly

Patil et al., 2005

Limitations:

- Low permeation rate of H_2 via Pd-membranes
- Mass transfer from bubble-to-emulsion phase

Micro-structured membrane-assisted fluidised bed reactor

Maximization of membrane area

Complete process integration

Optimal energy & mass transfer efficiency

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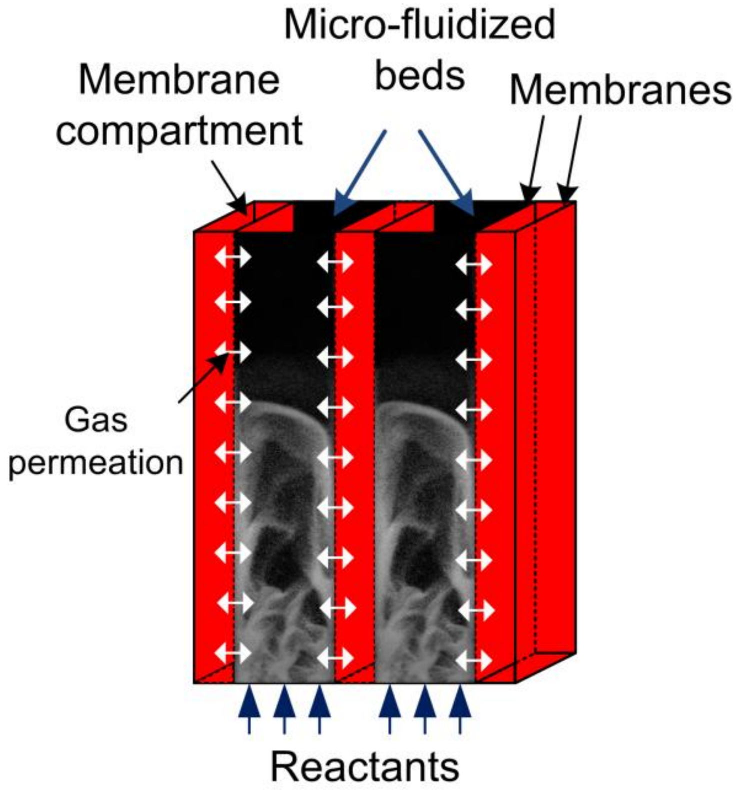
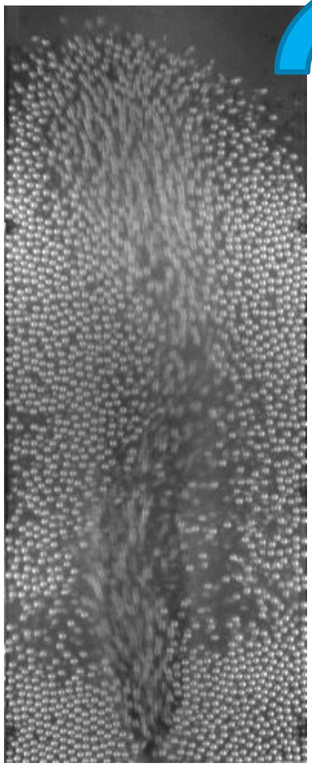
Micro-fluidized bed membrane reactors



- **Length scale between membrane walls?**
- **Influence of gas permeation on the hydrodynamics?**
- **Influence of gas permeation on the gas back-mixing characteristics?**

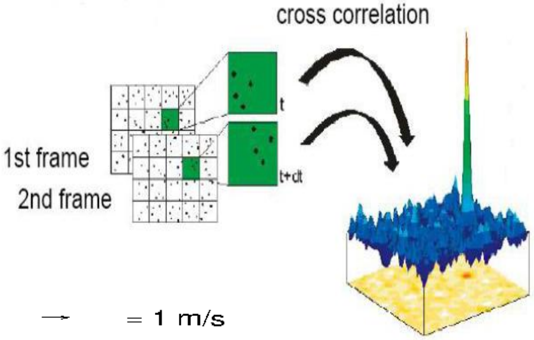
Hydrodynamics: Solid velocity using Particle Image Velocimetry (PIV)

PIV setup with visual high speed camera (2016x2016px@1600Hz)

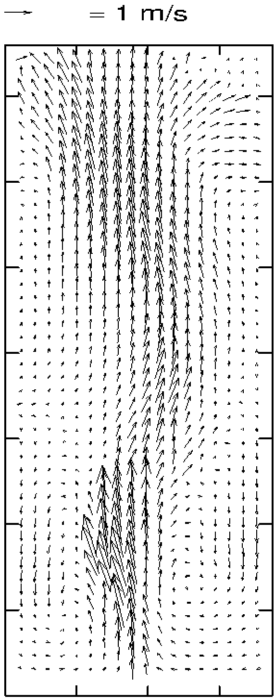


Double frame with small time delay (Δt = 500μs)

Discrete solid displacement



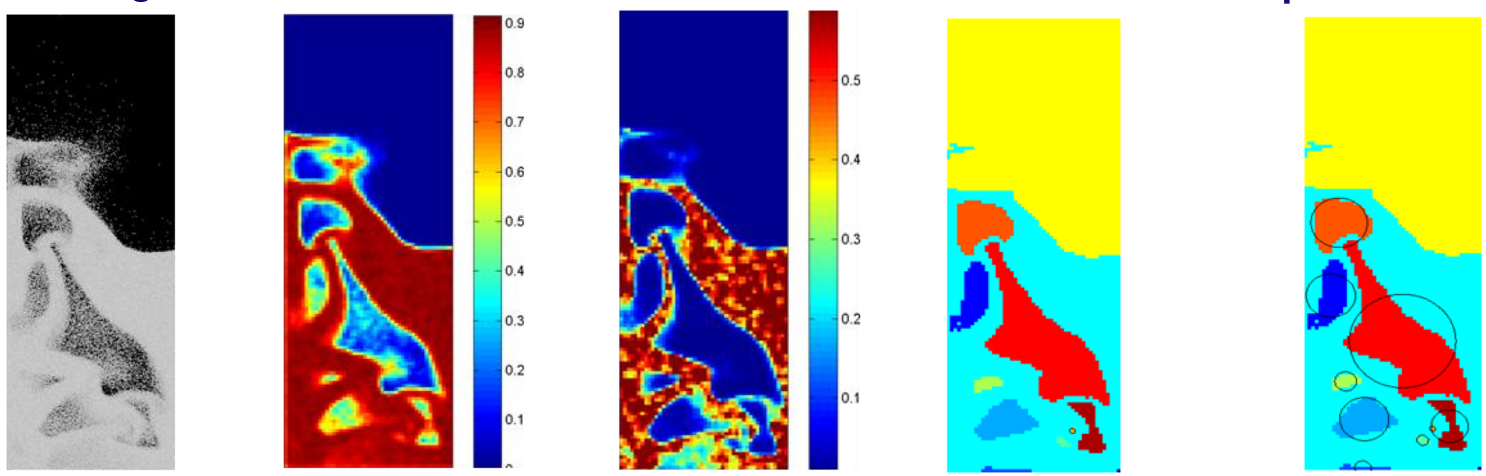
$$\mathbf{v}_p(\mathbf{x},t) = \frac{\mathbf{s}(\mathbf{x},t)}{M\Delta t}$$

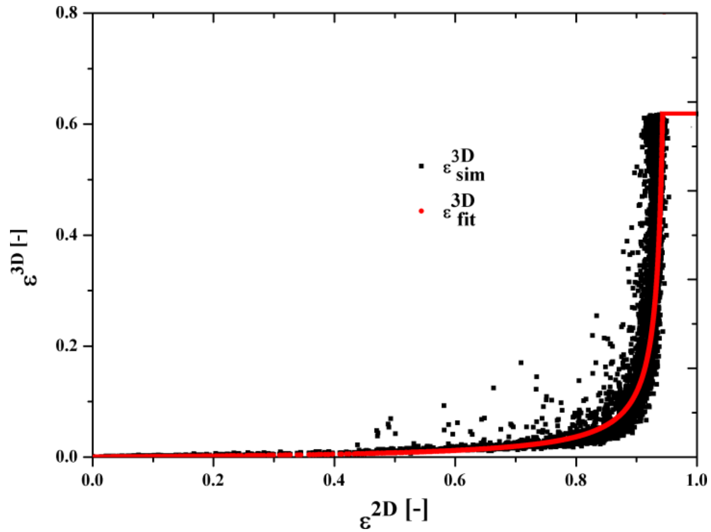


Solid flow field

Hydrodynamics: Digital Image Analysis (DIA)

VIS image | ε^{2D} | ε^{3D} | Bubble detection | Equi. Bubble diameter



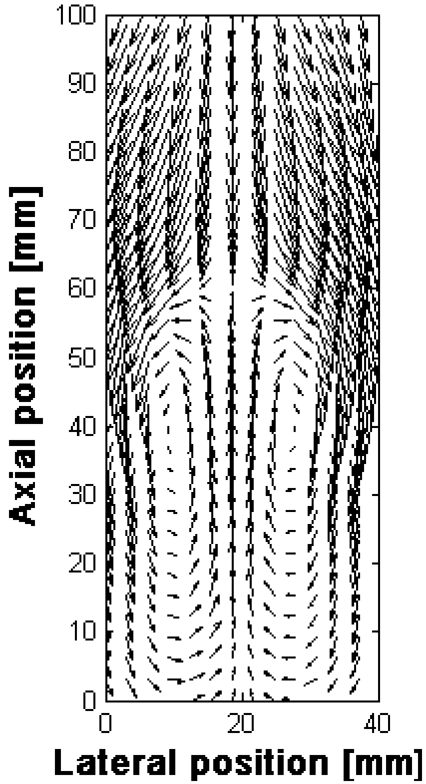


$\varepsilon_{,3D}$ obtained from DPM simulation

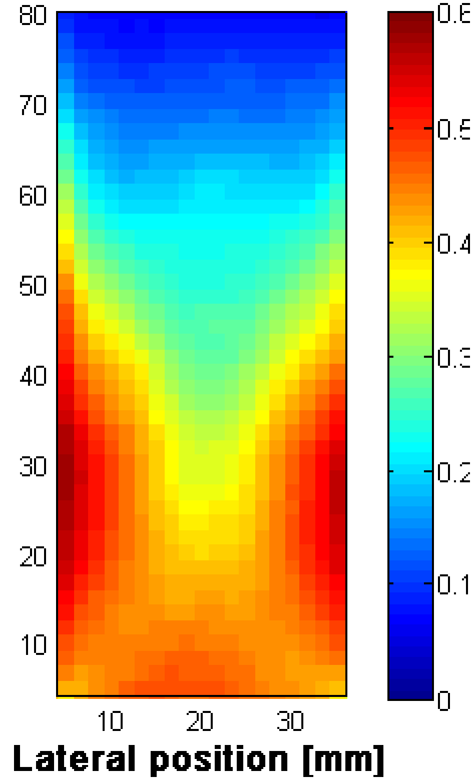
$$\varepsilon^{3D,cal} = \begin{cases} A\dfrac{\varepsilon^{2D,cal}}{B-\varepsilon^{2D,cal}} \quad if \quad \varepsilon^{2D} < \dfrac{0.6B}{0.6+A} \\ 0.6 \quad if \quad \varepsilon^{2D} > \dfrac{0.6B}{0.6+A} \end{cases}$$

Buijtenen et al., 2010

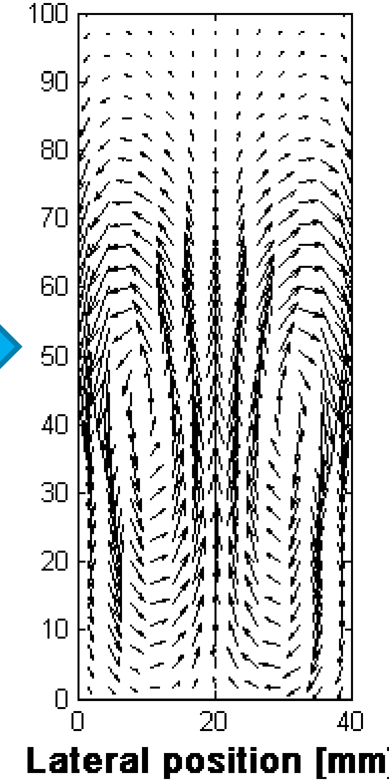
Coupled PIV/DIA



Solid flow field from PIV (non-invasively):
<u>Apparent solid flow!!</u>



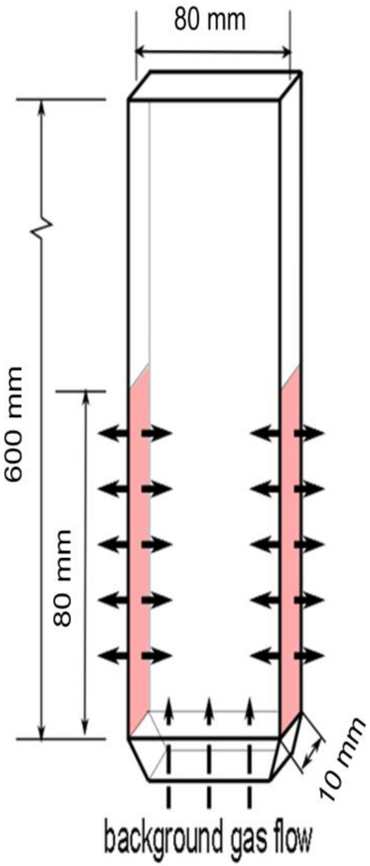
Solid holdup from DIA (non-invasively)

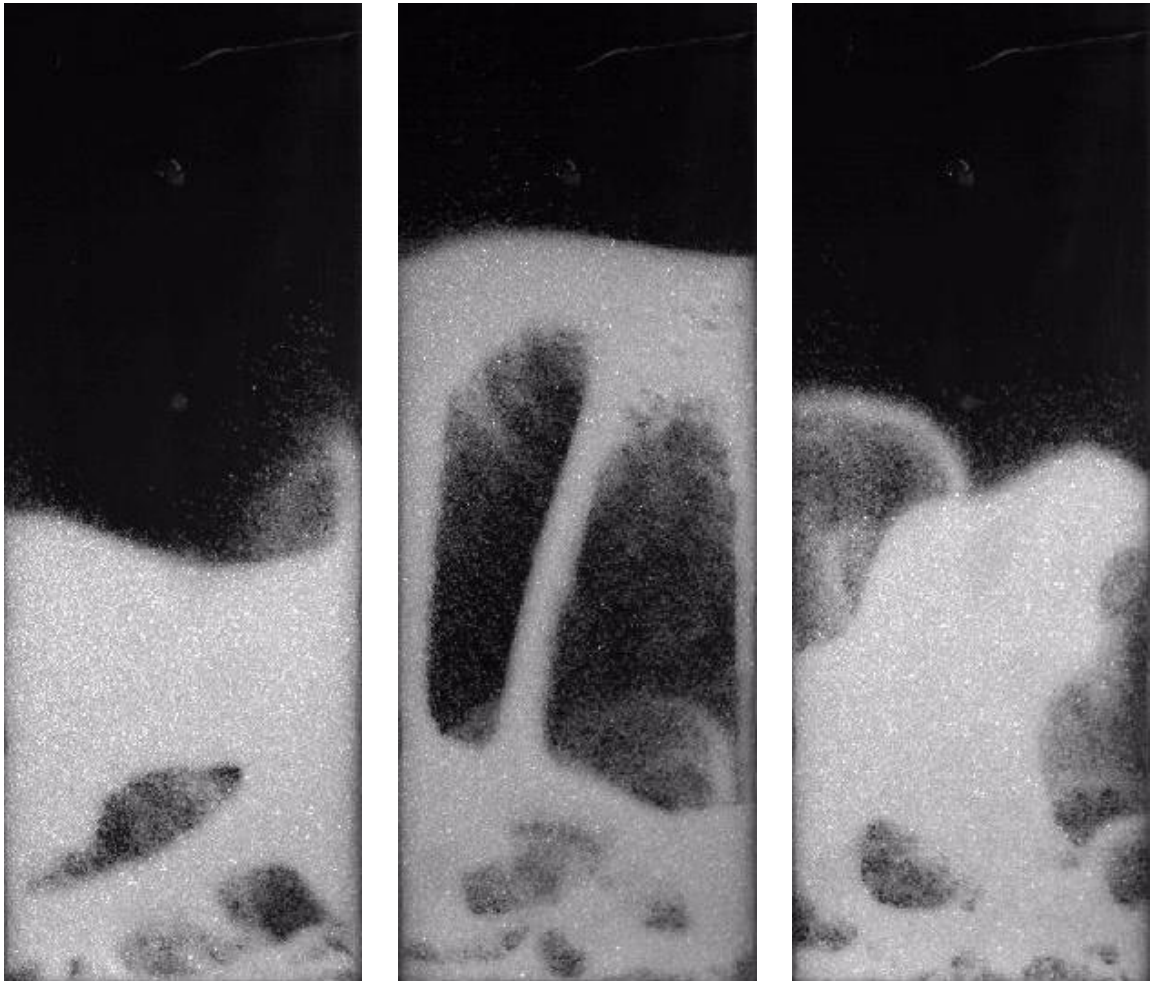


Corrected solid flow field:
Solid flux (non-invasively)

$$\langle \Phi_{V,p}(\mathbf{x},) \rangle = \frac{1}{N_t} \sum_{t}^{N_t} \left(\mathbf{v}_p(\mathbf{x},t) \cdot \varepsilon_p(\mathbf{x},t) \right)$$

Results: Solid circulation pattern (bubbling regime)





40% Extraction **Reference** **40% Addition**

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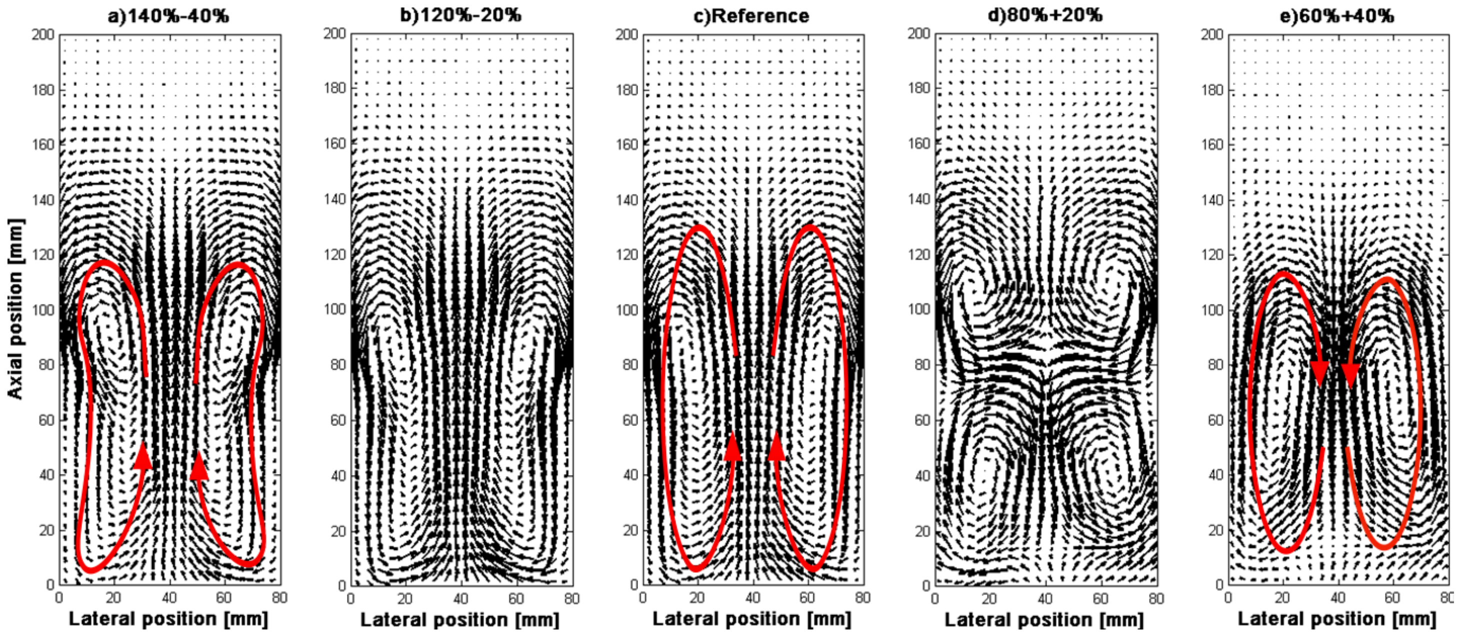
Results: Solid circulation pattern (bubbling regime)

Solid flux:



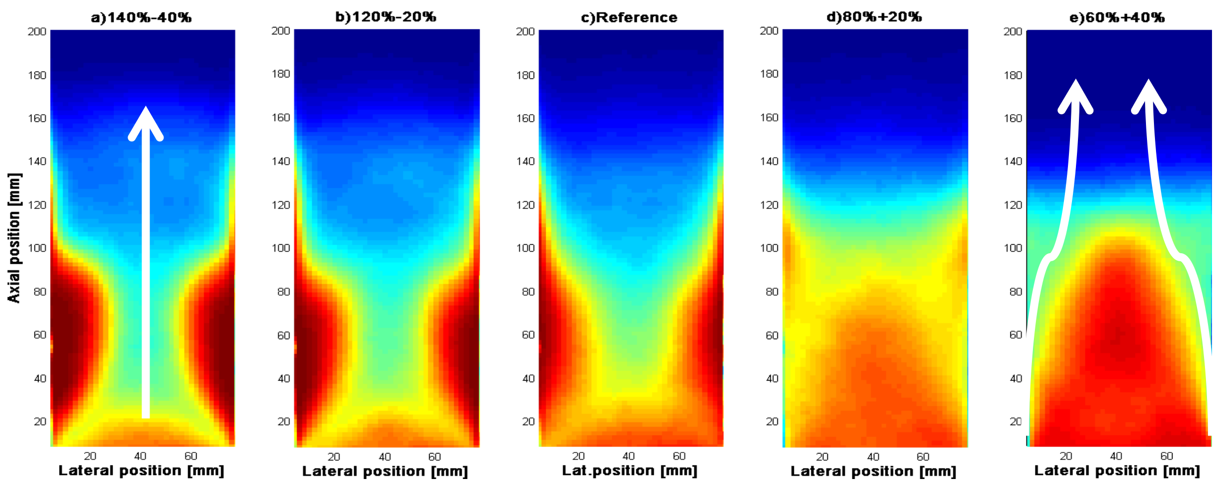
40% Extraction **20% Extraction** **Reference** **20% Addition** **40% Addition**

Ug,out = 0.75m/s

Dang et al., 2013 submitted to Chem.Eng.J.

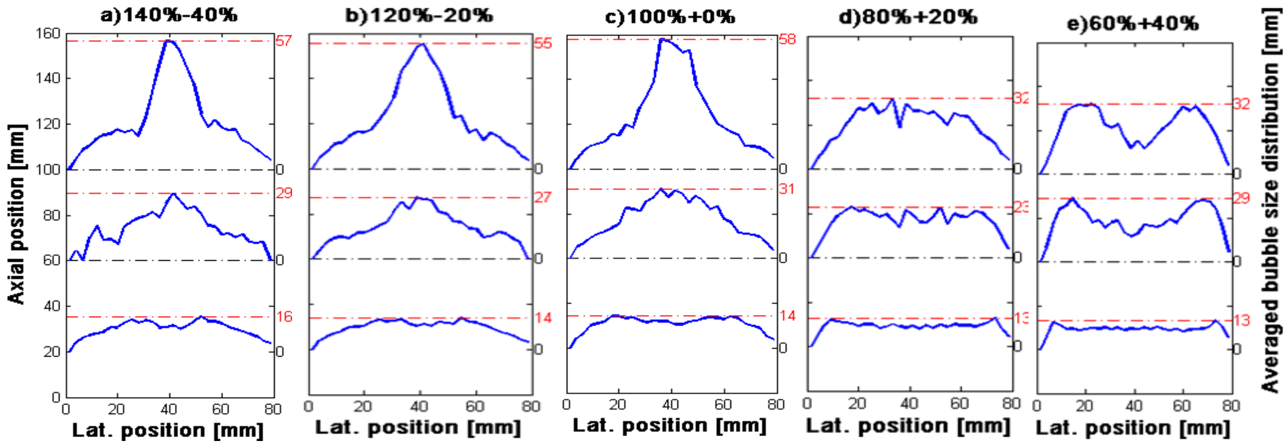
Results: Solid/bubble size distribution with influence of gas permeation

Solid holdup *Digital Image Analysis(PIV/DIA)*



$U_{g,out}$ = 0.75m/s

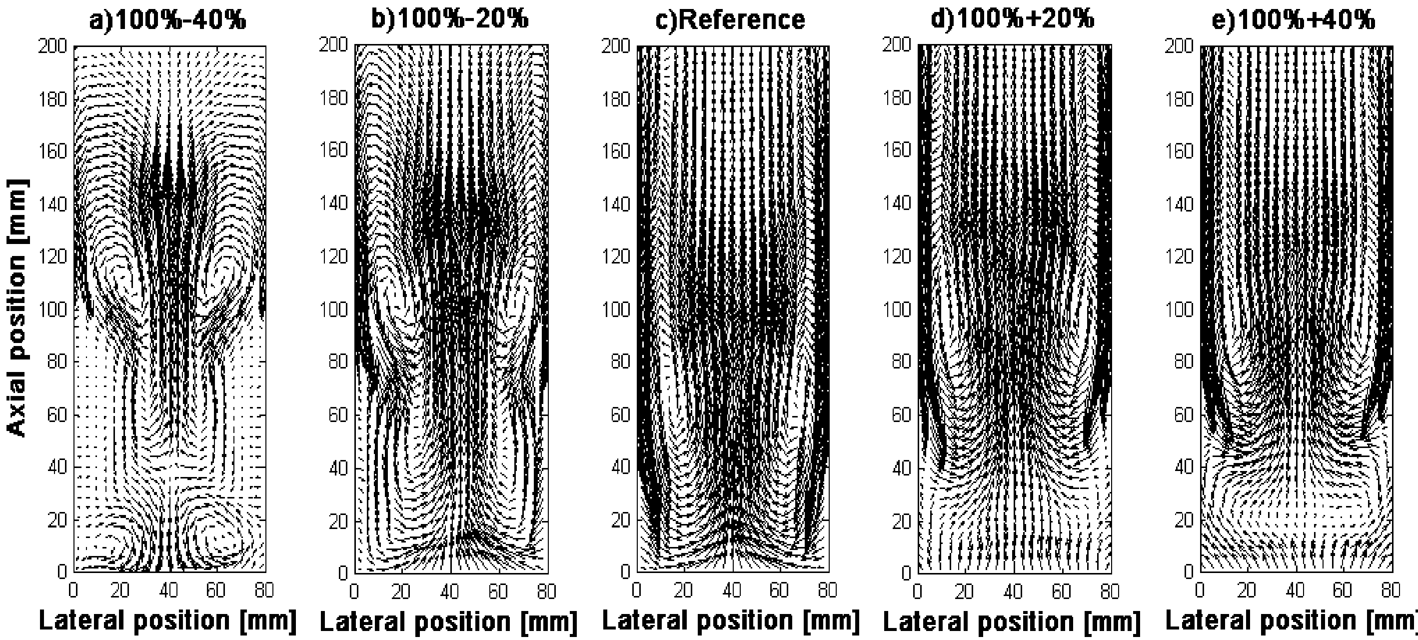
Bubble size distribution



40% Extraction **20% Extraction** **Reference** **20% Addition** **40% Addition**

Dang et al., 2013 submitted to Chem.Eng.J.

Results: Solid circulation pattern (turbulent regime)



40% Extraction **20% Extraction** **Reference** **20% Addition** **40% Addition**

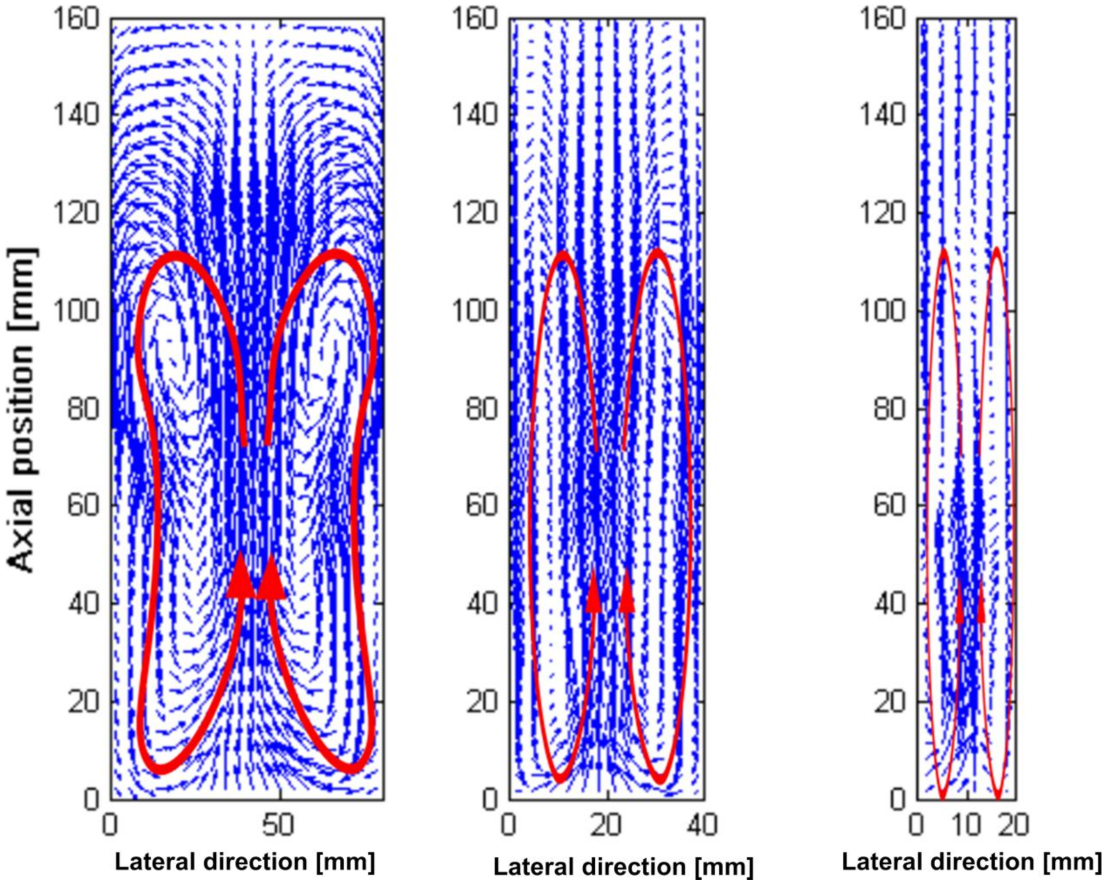
ρ_p = 2500 kg/m^3

d_p = 400 ÷ 600 μm

Ug_{inlet} = 2 m/s

Dang et al., 2013 submitted to Chem.Eng.J.

Results: Influence of reactor sizes



Smaller **reactor sizes** ***avoid*** **the densified zones and gas by-passing**

Dang et al., 2013 submitted to Chem.Eng.J.

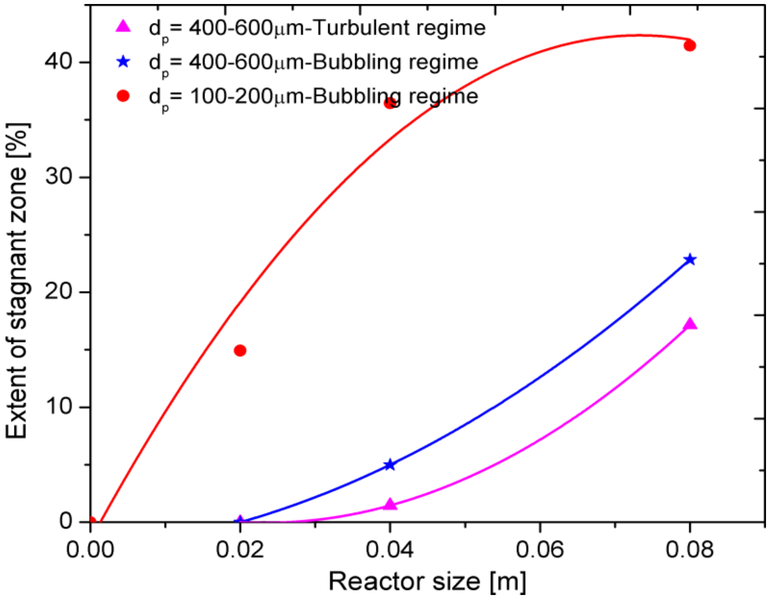
Results: densified zone creation for gas extraction

Extent of densified zone

Densified zone defined as:

$$E = \frac{\sum A_{densified}}{A_{Section}} \geq \alpha \varepsilon_{s,pack}$$

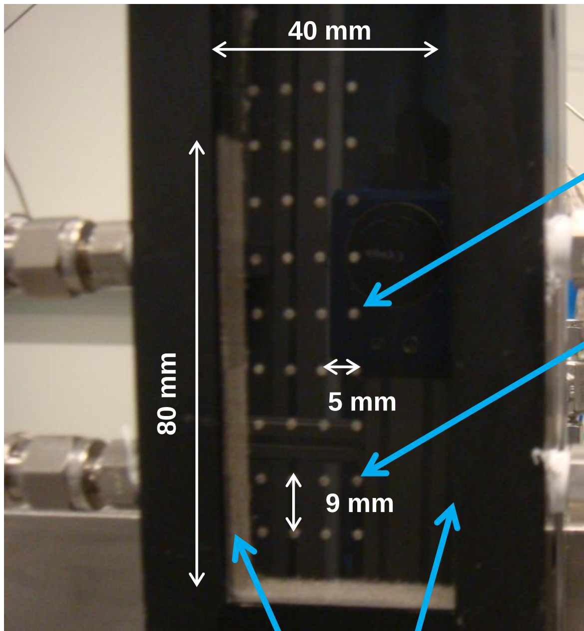
α : correction factor
ε: solid holdup in fixed beds



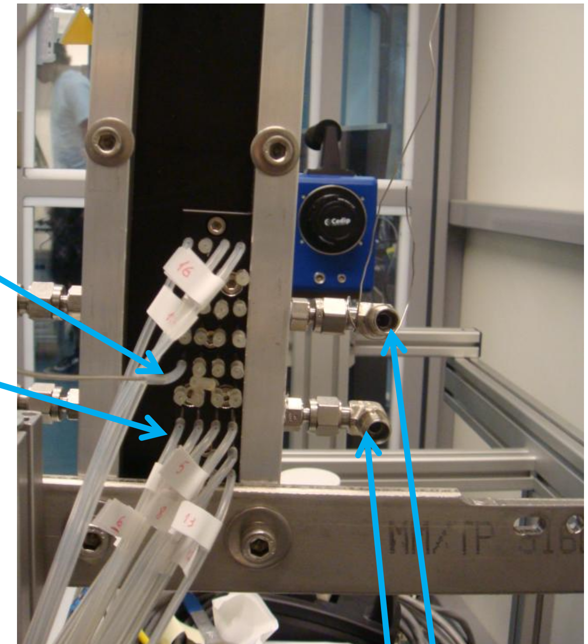
- **Smaller reactor → less densified zone → improve solid mixing**
- **Turbulent fluidization is more preferred than bubbling fluidization**
- **Relatively large particle size → better hydrodynamics**

Result: Gas back-mixing characteristics

Front side



Back side



Injection (upward)

Sampling

Membranes

Gas addition/extraction

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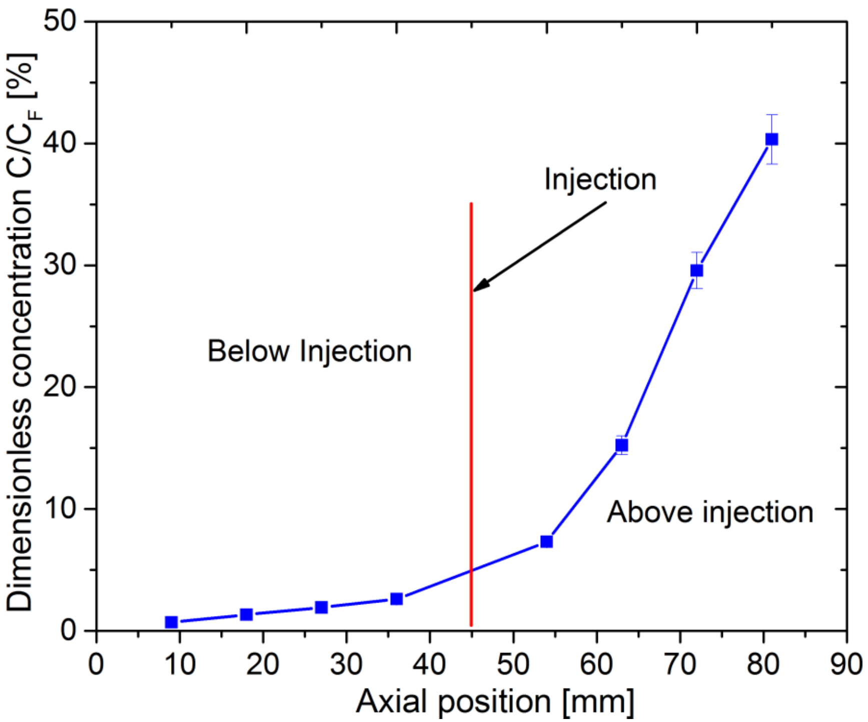
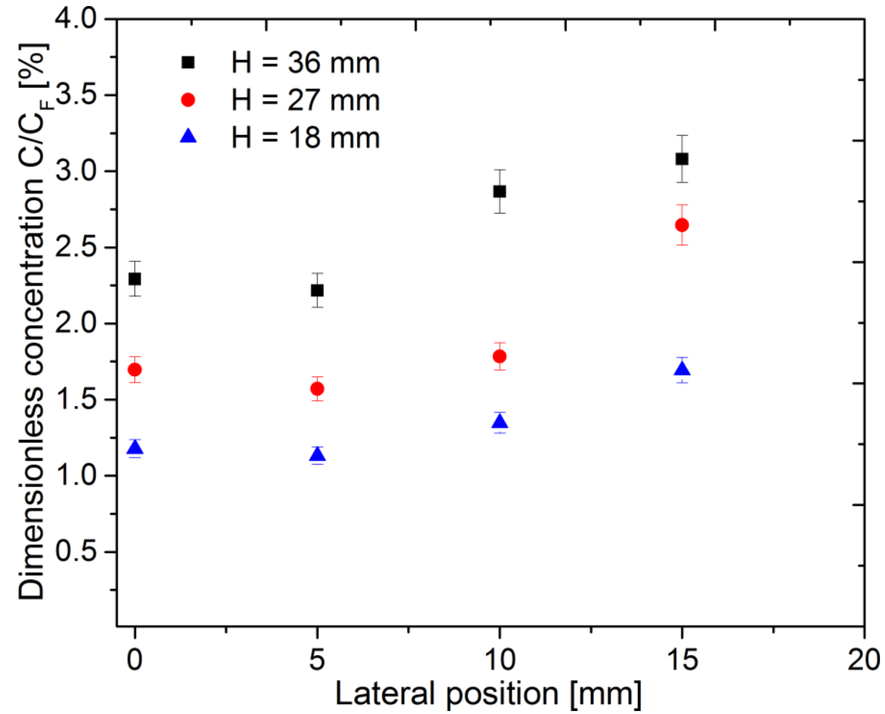
Gas back-mixing

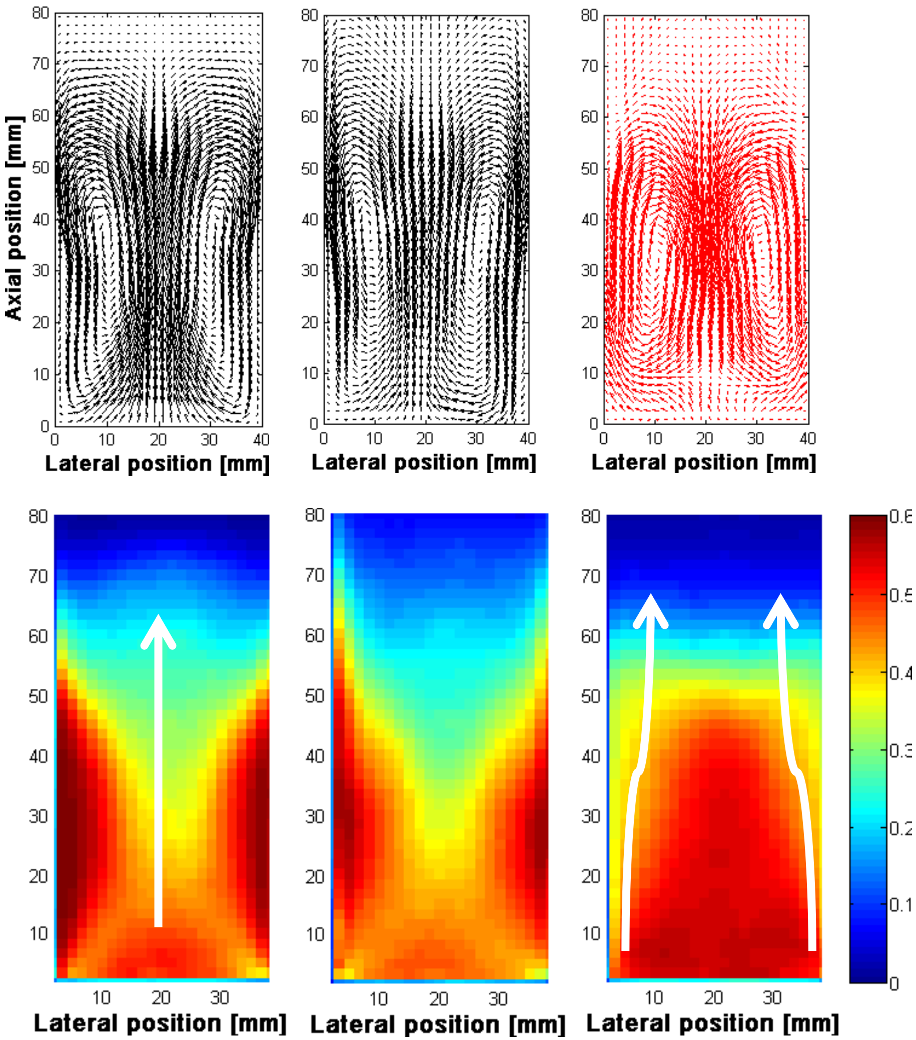




Below the injection point → tracer gas detected → finite gas back-mixing

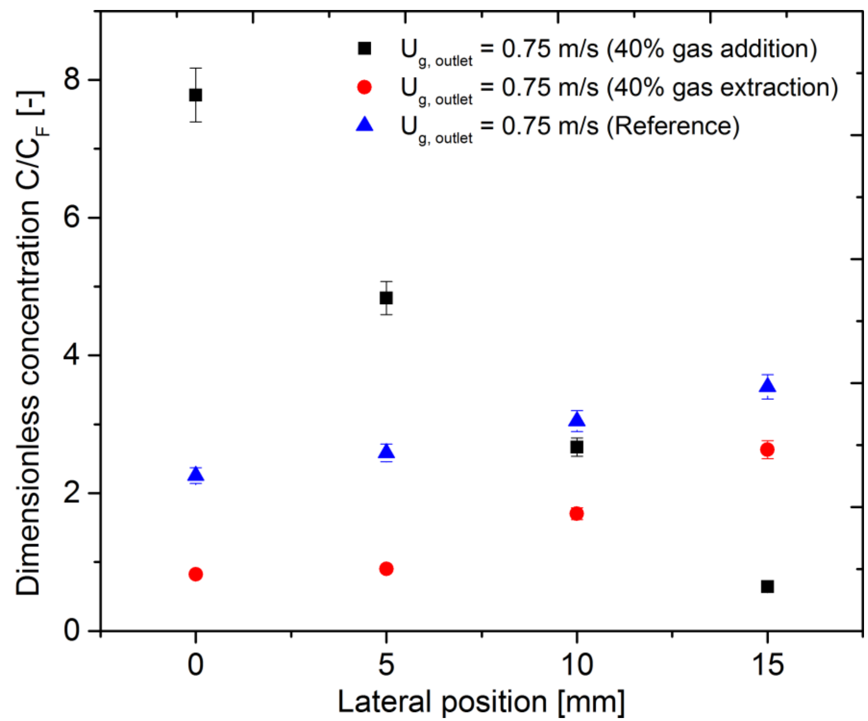
d_p = 400 ÷ 600 μm, ρ_p = 2500 kg/m^3
H_{bed} = 40 mm, H_{memb} = 40 mm
$U_{g,\ outlet}$ = 0.75 m/s

Gas back-mixing characteristics



40% Extraction **Reference** **40% Addition**

Gas back-mixing



d_p = 400 ÷ 600 µm, ρ_p = 2500 kg/m^3
H_{bed} = 40 mm, H_{memb} = 40 mm
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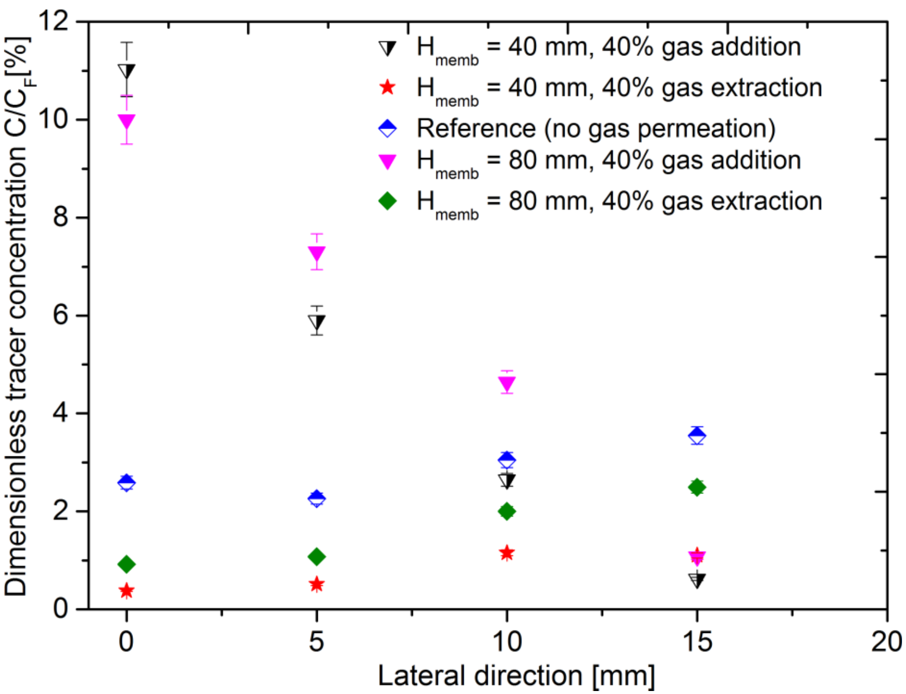
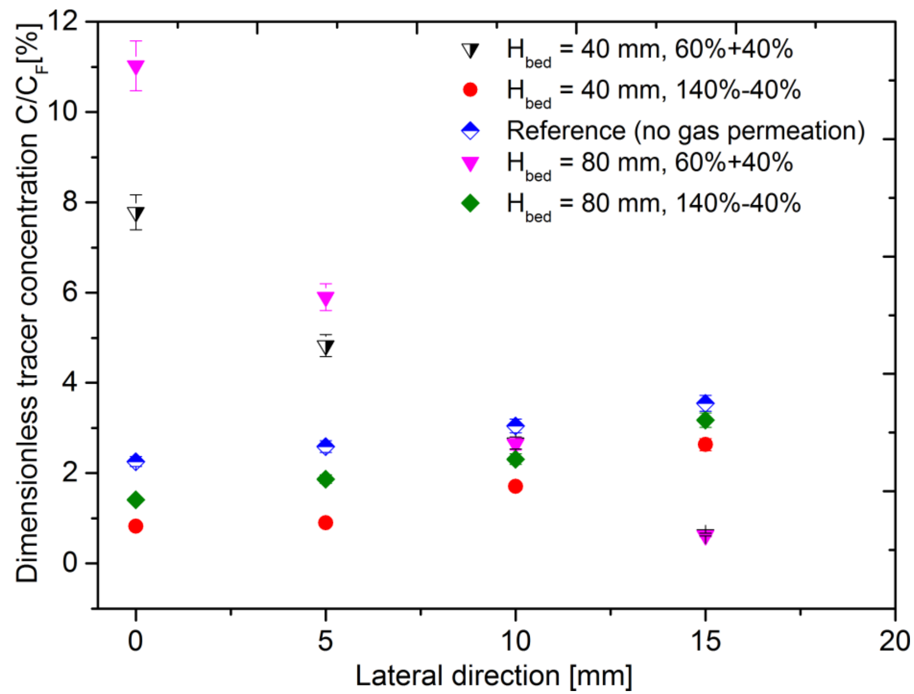
Dang et al., 2013 submitted to Catalysis Today

Gas back-mixing characteristics

Influence of membrane areas



Influence of total bed mass



- **Increase bed mass → increase gas back-mixing**
- **Membrane areas → vary the local gas back-mixing profiles**

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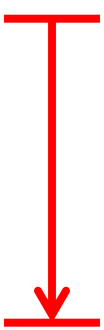
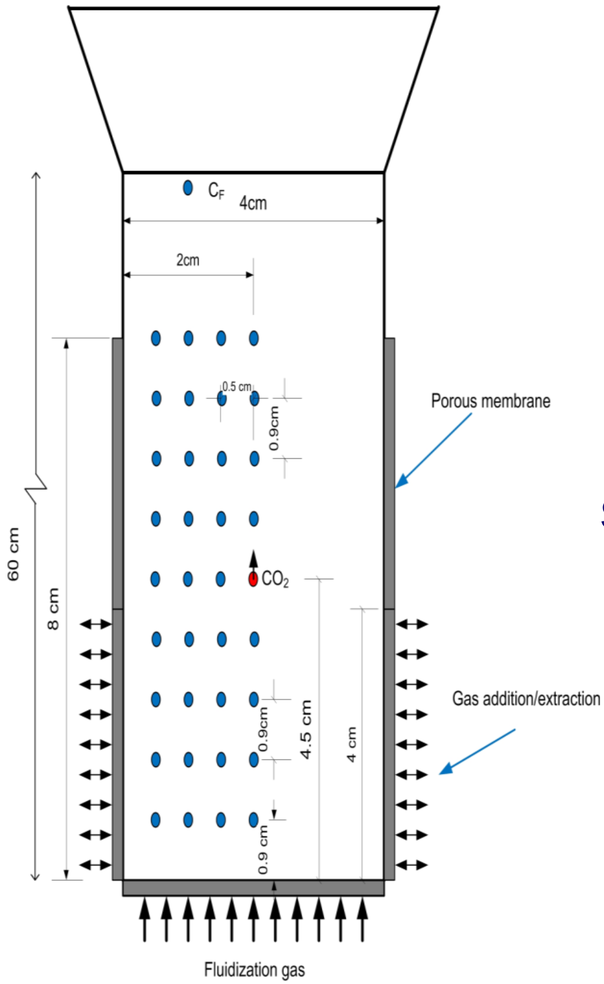
Gas back-mixing
Axial dispersion model

2D-steady state axial back-mixing model

z = 0, C=C$_F$



z = -∞, C=0



$$D_{a,g}\frac{\partial^2 C}{\partial z^2} - \frac{U_{g,z}}{\varepsilon_{g,z}}\frac{\partial C}{\partial z} = 0 \quad \textbf{(1)}$$

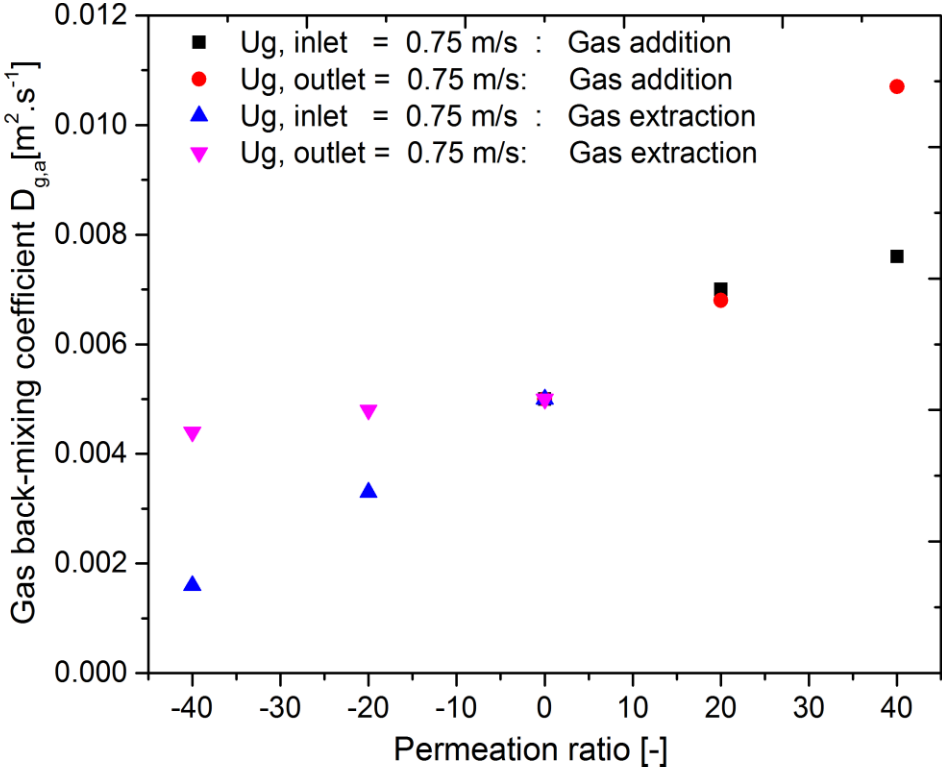
Boundary conditions:

Z = 0, C = C$_F$
Z = - ∞, C = 0

Solution of eqn.(1)

$$D_{a,g}\frac{1}{Z - z_i} = \frac{U_{g,z_i}}{\varepsilon_{z_i}}\frac{1}{\ln\left(\frac{C_{z_i}}{C_F}\right)}$$

Gas back-mixing coefficients



d_p = 400 ÷ 600 µm, ρ_p = 2500 kg/m³
H_{bed} = 40 mm, H_{memb} = 40 mm
$U_{g,\ outlet}$ = 0.75 m/s

- **Gas addition → increase gas back-mixing rate**
- **Gas extraction → Increase bed-to-membrane resistance → decrease back-mixing rate**

Dang et al., 2013 submitted to Catalysis Today

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Conclusions & outlook

Hydrodynamics:

- Gas extraction creases ***densified zones*** with low solid mixing and ***gas by-passing*** in the center
- Gas addition ***inverses solid circulation pattern*** with ***gas by -passing near the membrane walls***

→ Very careful to integrate membranes inside the fluidized beds

- Increase membrane areas/fluidization gas velocity → avoids densified zones & solid inversion
- Micro-fluidized bed membrane reactors → enhance reactor performance

Gas back-mixing

- Gas addition ***increases*** overall gas back-mixing rate
- Gas extraction ***increases mass resistance*** → ***decreases*** gas back-mixing rate

Outlook

- Study hydrodynamics and mass transfer operated under reactive conditions

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Acknowledgements

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Thank you for your attention!

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