CFD for the Study of the Multiphase Flow and Mass Transfer in a Stirred Tank Reactor for the Screening of Shaped Catalysts

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The Company

IFP *Energies Nouvelles*

Lyon Site

Public research and training institute with international scope, covering the fields of energy, transports and the environment

Transposing research to industry, technological innovation is central to all its activities

Public interest missions:

- **providing solutions to face society's challenges in terms of energy and the climate change, promoting a sustainable use of energy sources**
- **creating wealth and employment by supporting the French and European economic activity, and its competitiveness in related industrial sectors**

Evaluation of commercially shaped catalysts

Petrochemical, Refining and New Biological Green industrial Processes

Beads

Extrudates

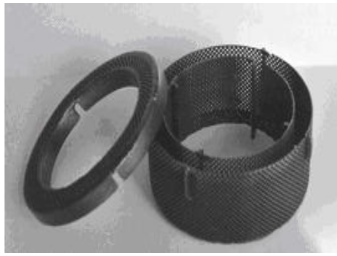
Particles (1-3 mm)

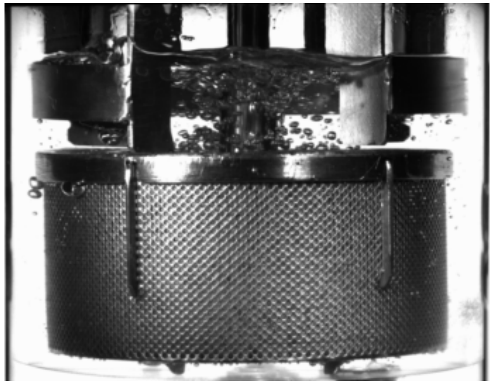
Catalysts are evaluated/tested in its commercial shape to maintain the spatial distribution of its active sites

Evaluation of commercially shaped catalysts

Possible choice at IFPEN:
Stirred reactor with stationary catalytic basket


Hollow self-inducing impeller

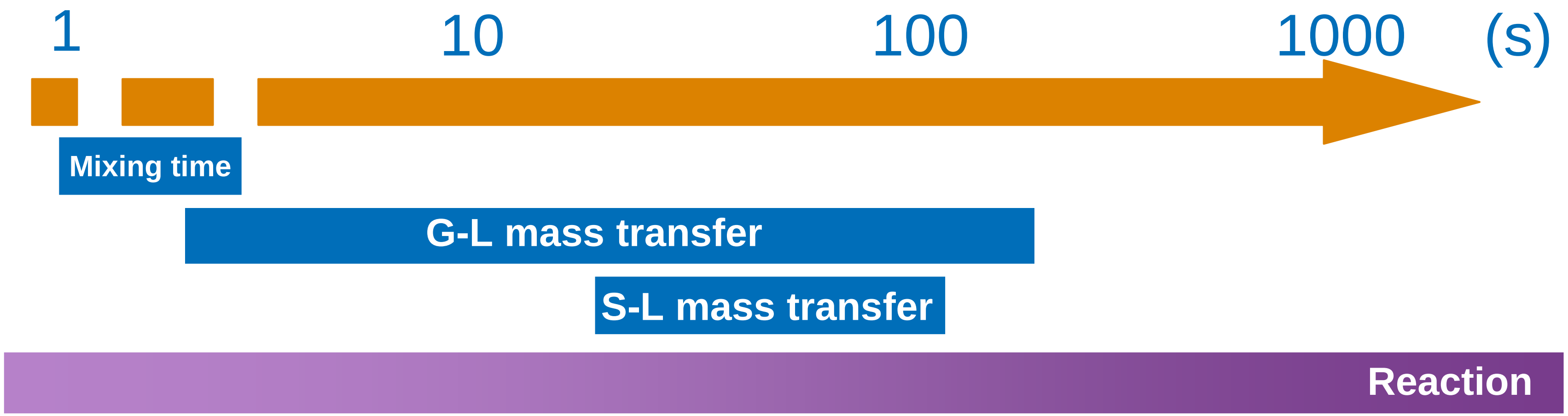

Catalytic basket



- Lab-scale baffled stirred tank reactor (STR)
  - Volume = 300 cm^3
- Stationary catalytic basket
  - Perforated metal walls
  - Support the catalyst particles (fixed bed)
  - Gap between basket walls = 9 mm
  - Diam. catalyst = 1 – 3 mm
- Self-inducing radial flow impeller
  - Hollow shaft with orifices
  - Recirculation of gas in a closed loop without the need for additional equipment
  - Mixing of the gas/liquid phases
  - D=20-25 mm, 800-2000 rpm

Highly complex gas-liquid turbulent flow (2 000 < Re < 20 000)

Characteristic time scales[1]



- Constant development of catalysts with higher performance
- Faster reactions
- Mass transport limitations between the phases can compromise kinetic tests

[1] - Braga, M., 2013. Etude des phénomènes de transfert et de l'hydrodynamique dans des réacteurs agités à panier catalytique (Study of transport phenomena and hydrodynamics in stirred reactors with a catalytic basket). Université Claude Bernard Lyon 1, Lyon, France.

General objectives of the project

- Having deeper knowledge on the hydrodynamics of STRs with stationary catalytic baskets
  - Complex multiphase flow
  - Difficult to observe experimentally the flow inside the catalytic basket
  - Only a few papers exist in the literature (for similar, but different, setups)
- Knowing the conditions for which the reaction kinetics is the controlling rate for fast reactions (no mixing or mass transfer limitations).
- Propose more efficient and robust reactor/basket designs:
  - More homogeneous dispersion of the gas in the liquid phase
  - Optimize gas absorption rate into the liquid phase (↑ $k_L a$)
  - Increase velocity of the liquid through the catalyst porous medium (↑ k_S)

Strategy

Computational Model

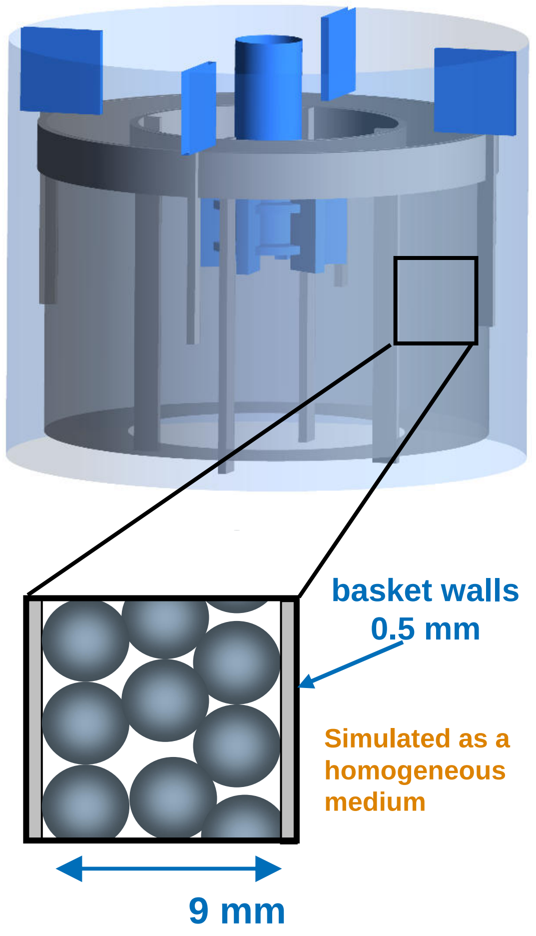
- Development of a "simple" parametric 3D Computational Fluid Dynamics (CFD) model of the reactor, describing the most significant aspects of:
  - Turbulent mixing
  - Flow through the porous media (catalyst particles in the basket)
  - Gas dispersion from the self inducing impeller

- Computational working tool, to be used internally, for the selection of correct operating conditions and for the design of new reactors, reducing the need for experimentation

Ensure regimes of kinetic control

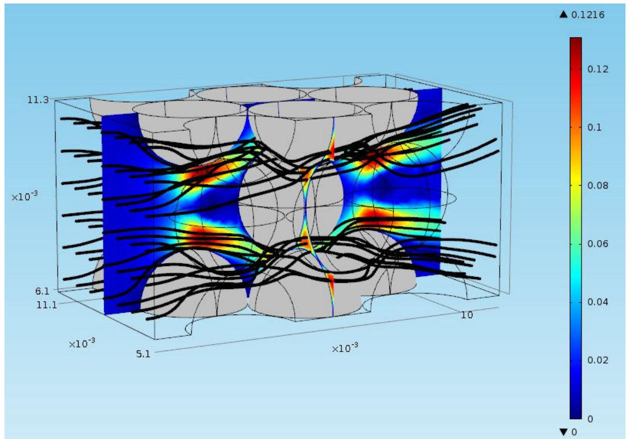
Solid-Liquid Mass transfer

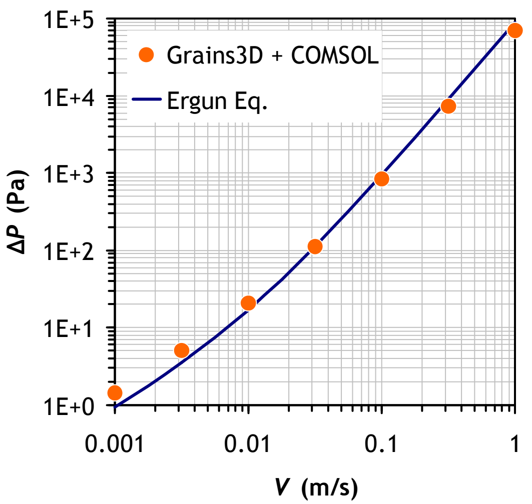
S-L CFD Model



- ANSYS Fluent
- Steady state
- Multiple Reference Frame approach to deal with impeller motion
  - *D* = 19 mm
  - *N* = 800-2000 rpm
- Working fluid: Heptane
- Reynolds Averaged Navier-Stokes equations
- Standard *k*-*ε* turbulence model to deal with the closure problem
- Standard Wall Functions
- Homogeneous porous media model inside the basket (momentum source terms from Ergun equation).
  - Particles of 3 mm
  - Porosity 47%
- Porous jump model to simulate the perforated basket walls

Homogeneous Porous Medium
Hypothesis verification





- A Discrete Elements Method code (GRAINS 3D)[2] was used to simulate a random packing of particles representative of the packing inside the basket
- Estimation of the pressure drop across the shallow packed bed inside the basket from CFD simulations at the particle scale using COMSOL Multiphysics

The numerical results support the use of the Ergun equation to model the pressure drop inside the basket

[2] Wachs A., Girolami L., Vinay G., Ferrer G., *Powder Technology* 224 (2012) 374–389

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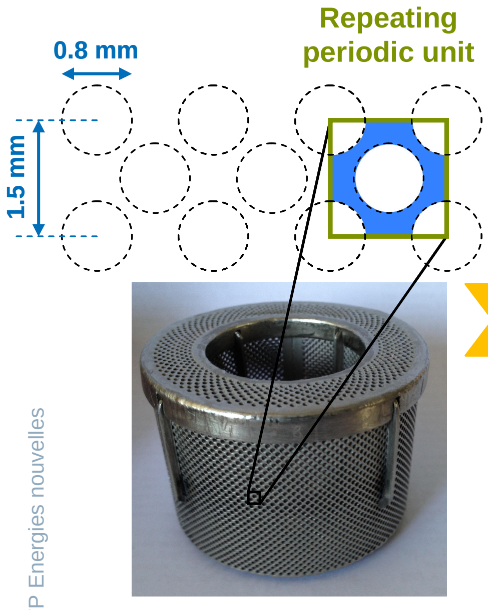
Grid of the catalytic basket

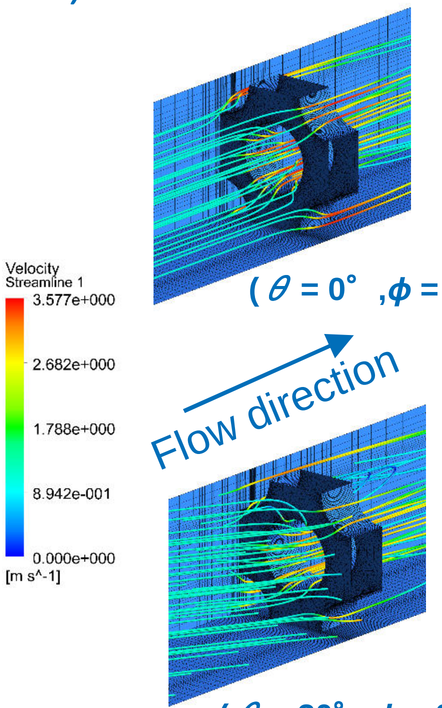
Perforated Plate Model

Catalytic basket walls simulated as a semi-permeable zero-thickness surface with pressure drop (porous jump model)

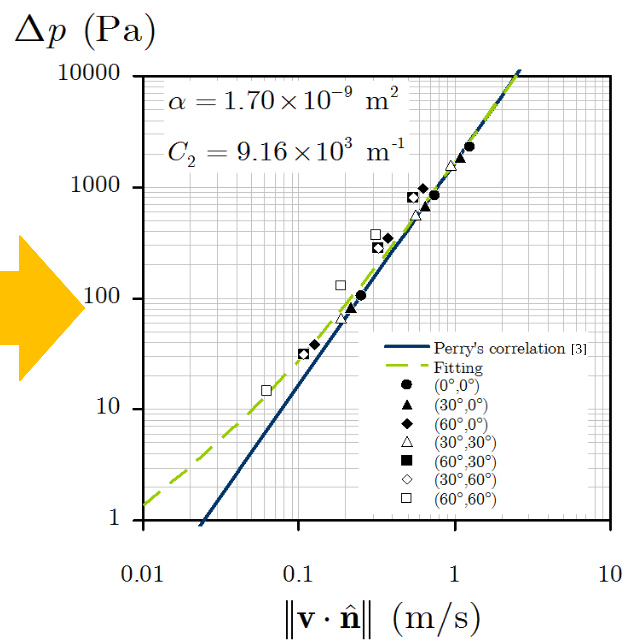
$$\frac{\Delta p}{\Delta x} = -\left(\frac{\mu}{\alpha}(\mathbf{v}\cdot\mathbf{n}) + \frac{1}{2}\rho C_2 (\mathbf{v}\cdot\mathbf{n})^2\right)$$



Empty catalytic basket



Simulations of the flow through a repeating periodic unit of the perforated plate
Different velocity angles of attack

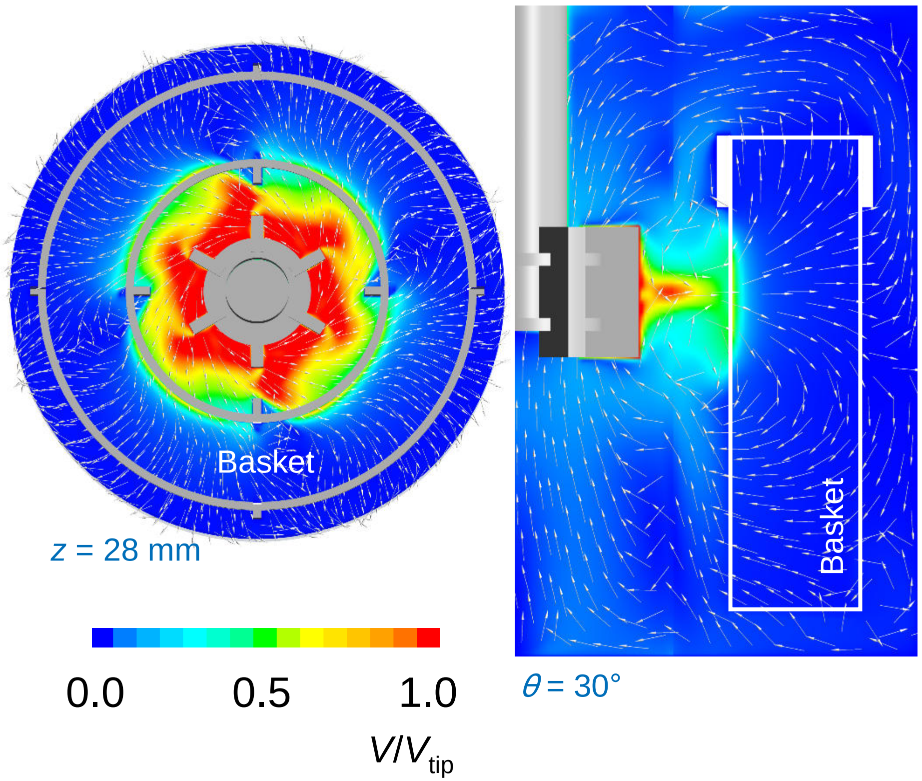


Parameters α and C2 from fitting to CFD results

[3] Perry's Chemical Engineer's Handbook, 14-30

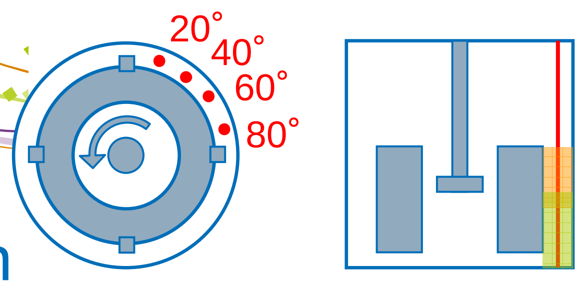
Flow field N = 2000 rpm

Top view **Side view**

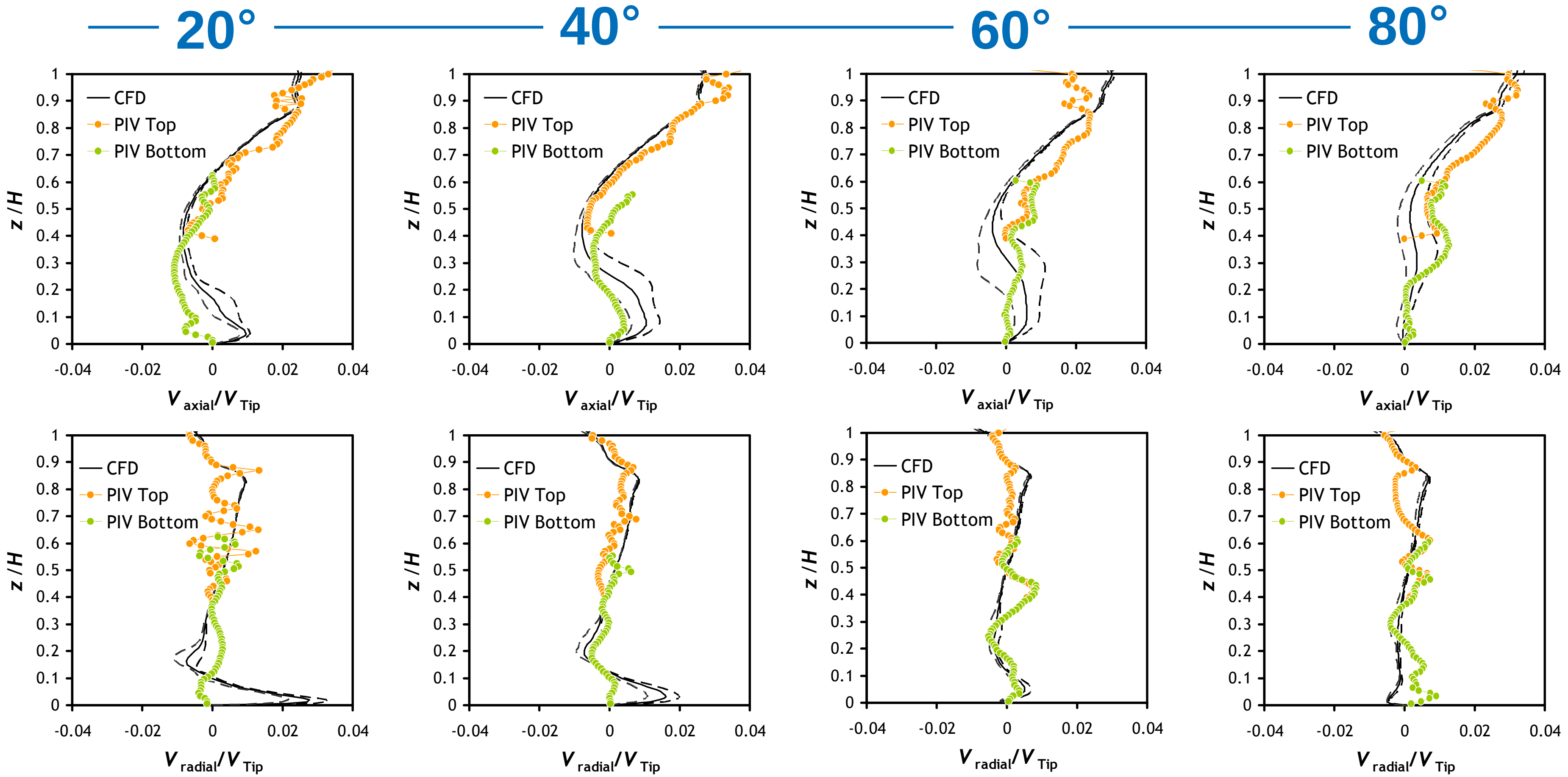


- Flow in the basket is not homogeneous
- Fluid pumped by the impeller blades traverses the basket radially at the height of the impeller
- Porous media promotes a rapid dissipation of velocity gradients by spreading the pumped jet in the axial direction
- Fluid reenters the basket at the bottom

Particle Image Velocimetry (PIV) Validation



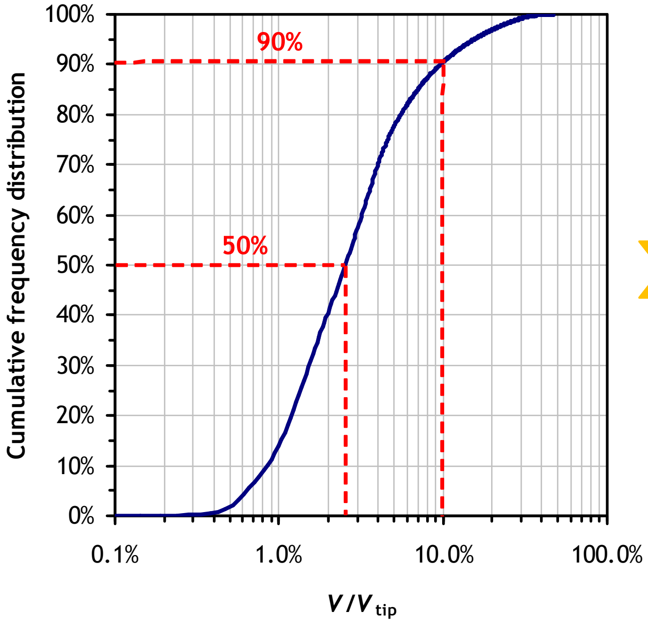
2.25 mm from the wall



Black discontinuous lines: range for the 8 impeller/baffle orientations in CFD

S-L mass transfer coefficient

Velocity distribution in the catalyst volume can be used to estimate the solid-liquid mass transport rate



Broad distribution of velocities inside the basket

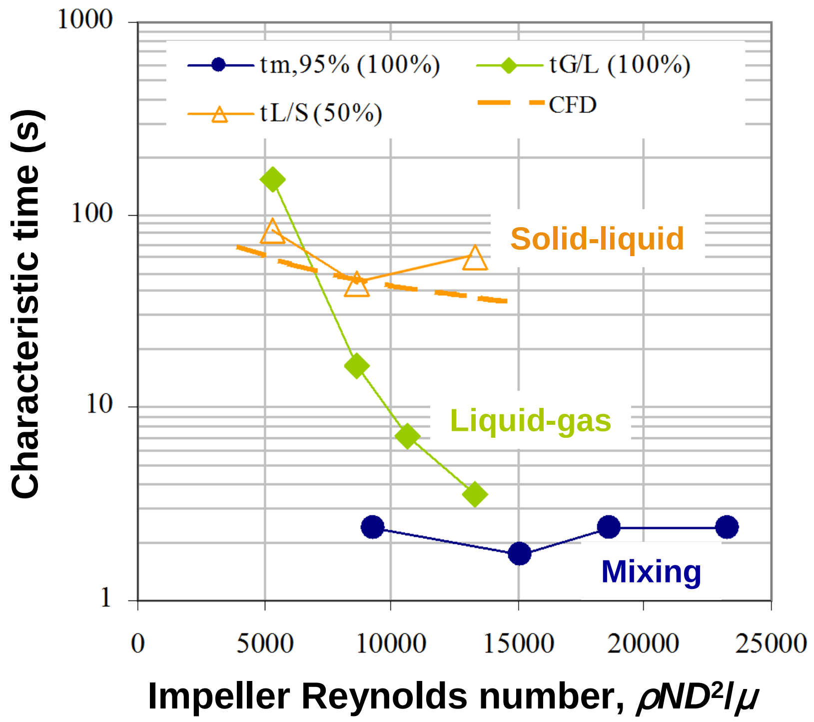
Ranz-Marshall correlation to estimate the spatial distribution of Sh

$$\mathrm{Sh}(\mathbf{x}) = 2 + 1.8\,\mathrm{Re_p}(\mathbf{x})^{0.5}\,\mathrm{Sc}^{0.33}$$

Average S-L mass transfer rate

$$\mathrm{Sh_{app}} = \frac{\langle k_S \rangle d_p}{D_m} = \frac{\iiint_{V_{catalyst}} \mathrm{Sh}(\mathbf{x})\,\mathrm{d}^3\mathbf{x}}{V_{catalyst}}$$

S-L mass transfer coefficient

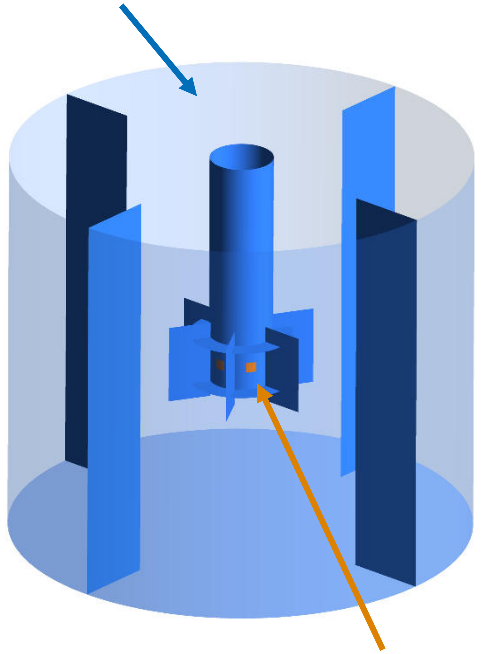


- CFD values show good agreement with experimental values reported in Braga's PhD thesis[1]
- Assumed simplification of homogeneous porous medium proven acceptable
- Ranz-Marshall correlation also proven suited for the determination of the mass transfer coefficient

[1] - Braga, M., 2013. Etude des phénomènes de transfert et de l'hydrodynamique dans des réacteurs agités à panier catalytique (Study of transport phenomena and hydrodynamics in stirred reactors with a catalytic basket). Université Claude Bernard Lyon 1, Lyon, France.

Gas-Liquid Mass transfer

G-L CFD Model



- ANSYS Fluent
- Steady state
- Eulerian-Eulerian multiphase flow model
  - Heptane-Hydrogen binary system
- Gas inlet flow rate as a function of impeller rotational speed [4]
- Multiple Reference Frame approach to deal with impeller motion
- Reynolds Averaged Navier-Stokes equations
- Per-phase Realizable *k-ε* turbulence model to deal with the closure problem
- Standard Wall Functions
- A turbulence dispersive force modeled as a dispersive term in the phases mass conservation equations (Sc_{turb}=0.75)
- Spatially invariant bubble diameter of 2 mm (experimental)
- Bubble drag coefficient from Schiller-Naumann correlation
  - Different drag correction correlations for turbulent flows have been tested

[4] – C. P. Fonte *et al.*, Chem Eng Technol (2014)

Drag Modification Correlation

- Different correlations exist for taking into account the apparent decrease of the bubbles rising time due to constant accelerations and decelerations by turbulent velocities fluctuations.

$$\frac{\overline{C_D}}{C_{D,0}} = 1 + 8.76\times10^{-4}\left(\frac{d_\mathrm{b}}{\lambda_\mathrm{K}}\right)^3$$

Brucato
(Brucato et al., 1998)

$$\frac{\overline{C_D}}{C_{D,0}} = 1 + 6.5\times10^{-6}\left(\frac{d_\mathrm{b}}{\lambda_\mathrm{K}}\right)^3$$

Modified Brucato
(Lane et al., 2000)

$$\frac{\overline{C_D}}{C_{D,0}} = \left(0.4\tanh\left(16\frac{\lambda_\mathrm{K}}{d_\mathrm{b}}-1\right)+0.6\right)^{-2}$$

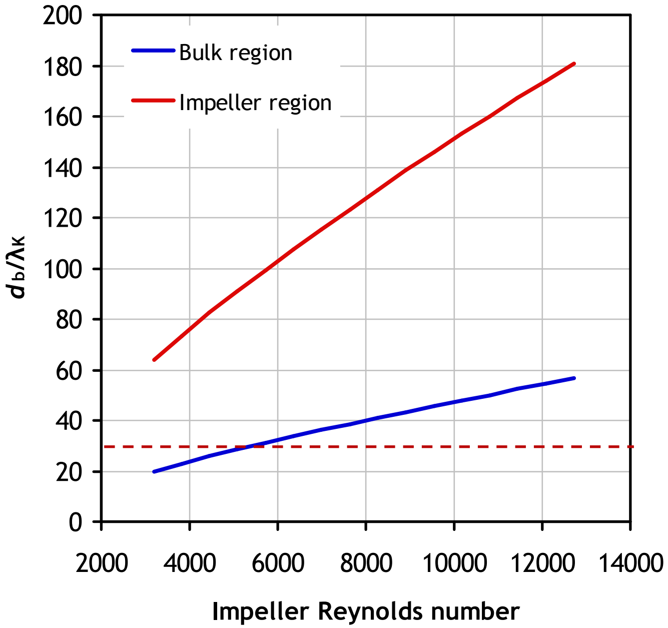
Pinelli
(Pinelli et al., 2001)

Different mechanism for bubbles and solid particles?

Drag Modification correlation

- Approx. 50% of energy dissipation occurs in the impeller region
- Dissipation rate is ~100x less in the bulk region

$$V_{\mathrm{impeller}} \sim D^3 \qquad \frac{\lambda_{\mathrm{K}}^{\mathrm{impeller}}}{D} \sim 1.2 N_{\mathrm{P}}^{-0.25}\,\mathrm{Re}_{\mathrm{imp}}^{-0.75} \qquad \lambda_{\mathrm{K}}^{\mathrm{bulk}} \sim 3\lambda_{\mathrm{K}}^{\mathrm{impeller}}$$



Bubbles of 2 mm of diameter

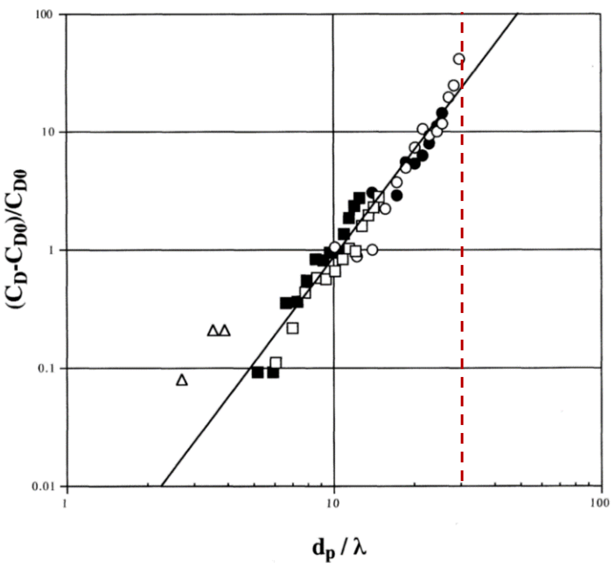


Fig. 11. Plot of normalized drag coefficients in turbulent media *vs* d_p/λ. Glass beads (△) 63–71 μm, (□) 212–250 μm, (○) 425–500 μm, silica (■) 180–212 μm, (●) 425–500 μm.

Values need to be extrapolated (high d_b/λ_K values)

Brucato et al., *Chem Eng Journal* (1998)

Drag Modification correlation

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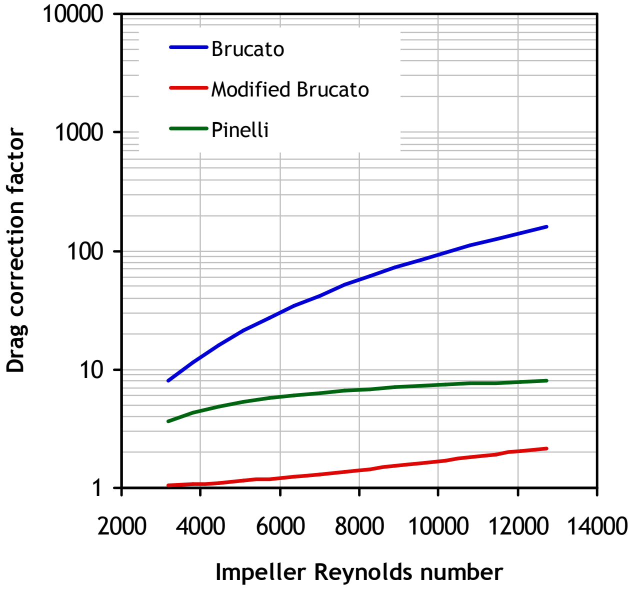
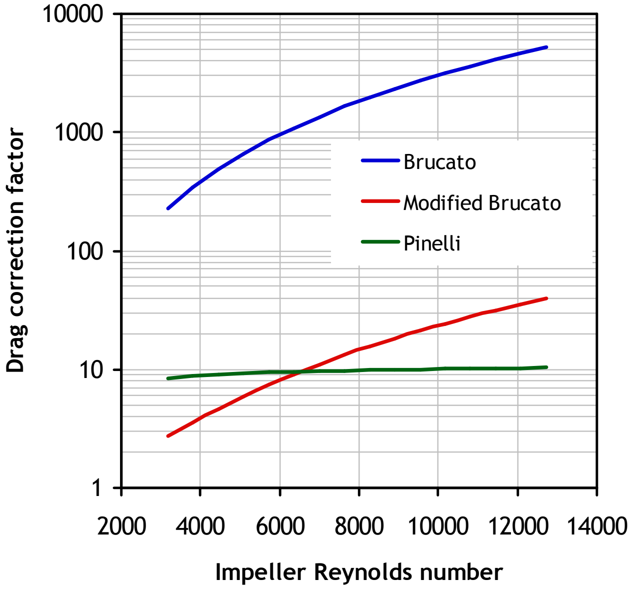
Bulk region



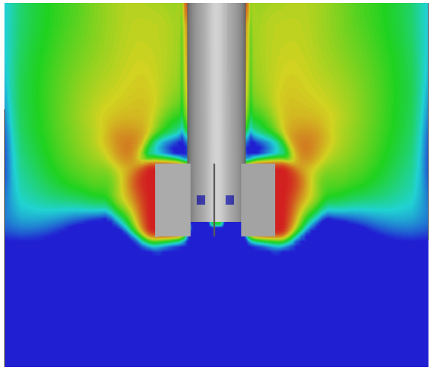
Impeller region



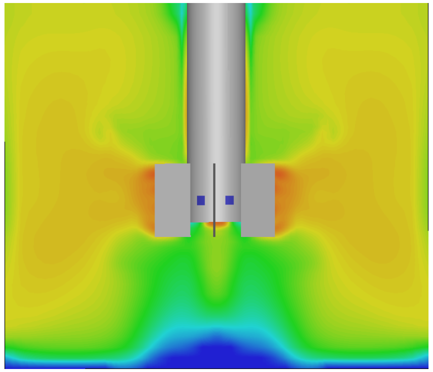
The correlations predict quite different correction factors

Bubbles of 2 mm of diameter

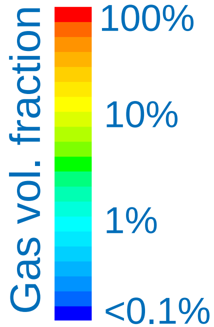
Drag Modification Correlation (N = 1600 rpm)

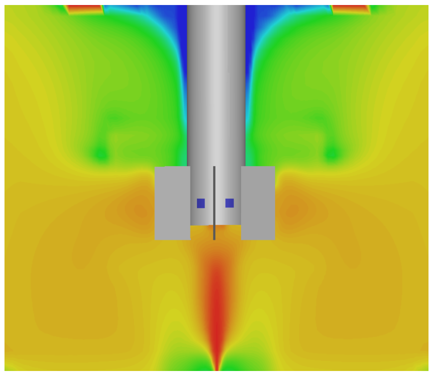


No modification

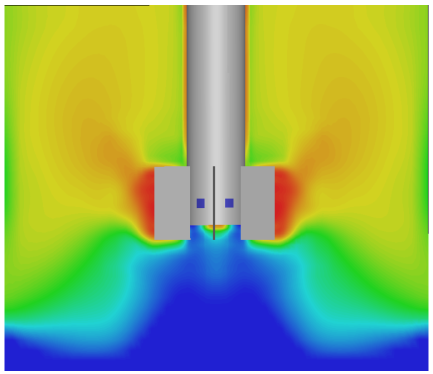


Pinelli

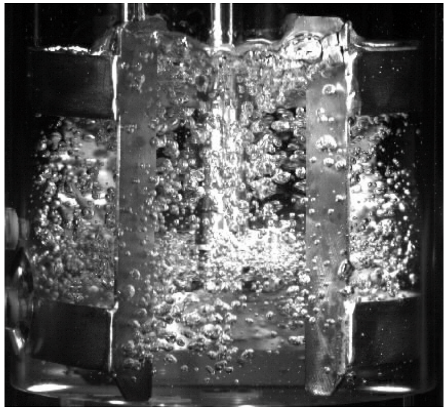




Brucato



Modified Brucato

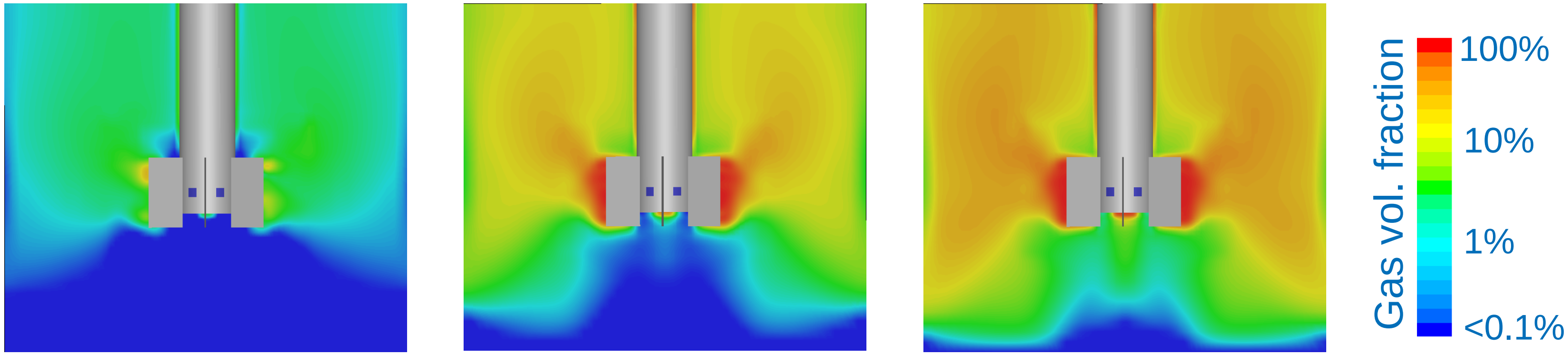


High speed camera

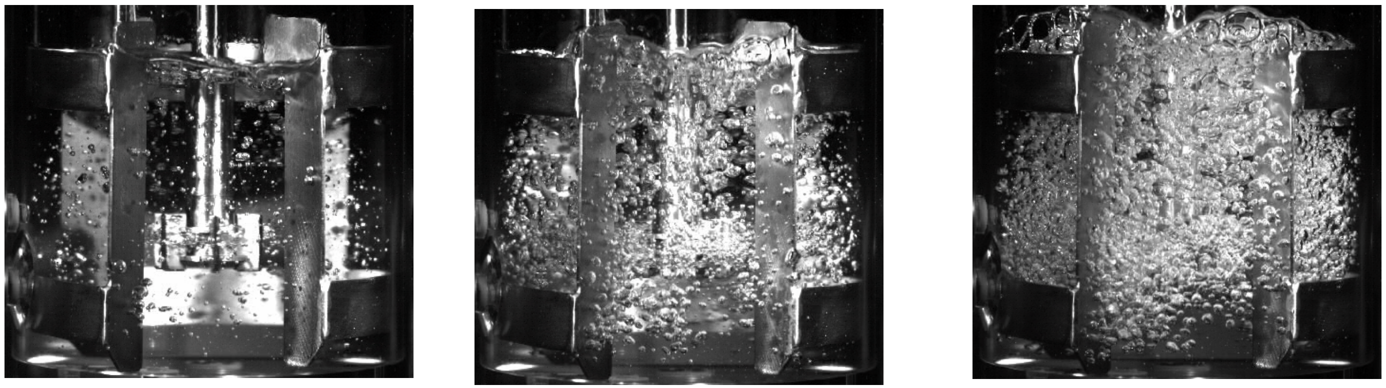
Gas phase distribution
(Modified Brucato)

N = 1000 rpm | **N = 1600 rpm** | **N = 2000 rpm**

CFD

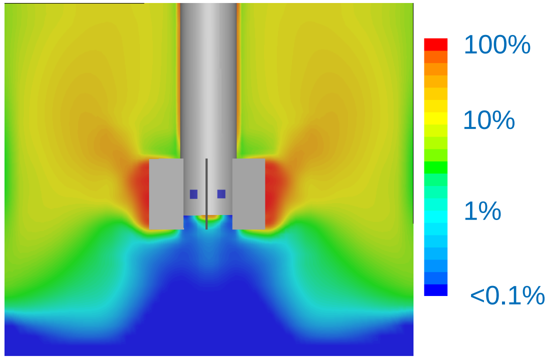


High speed camera



G-L mass transfer coefficient

Gas Volume Fraction



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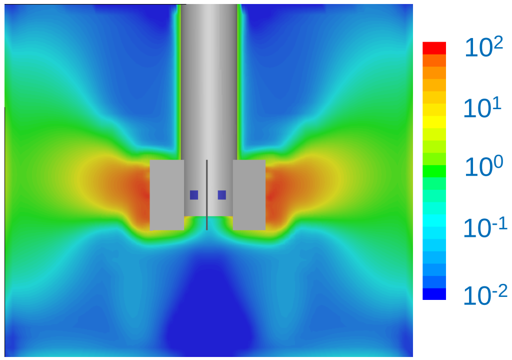
Turbulent Energy Dissipation Rate (m^2/s^3)



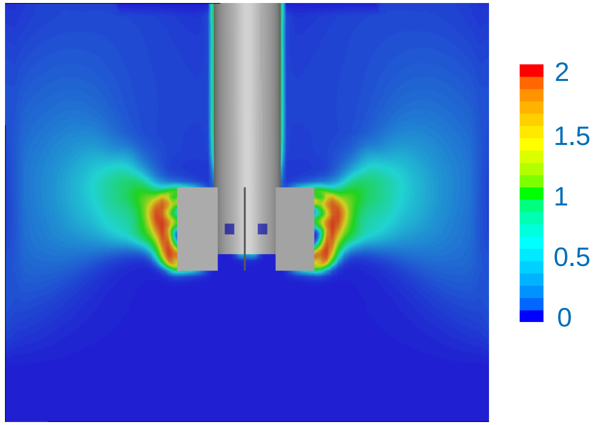
Danckwerts bubble surface renewal model

$$k_{\mathrm{L}} = 0.4\sqrt{D_{\mathrm{m}}}\left(\frac{\varepsilon_c}{\nu_c}\right)^{1/4}$$

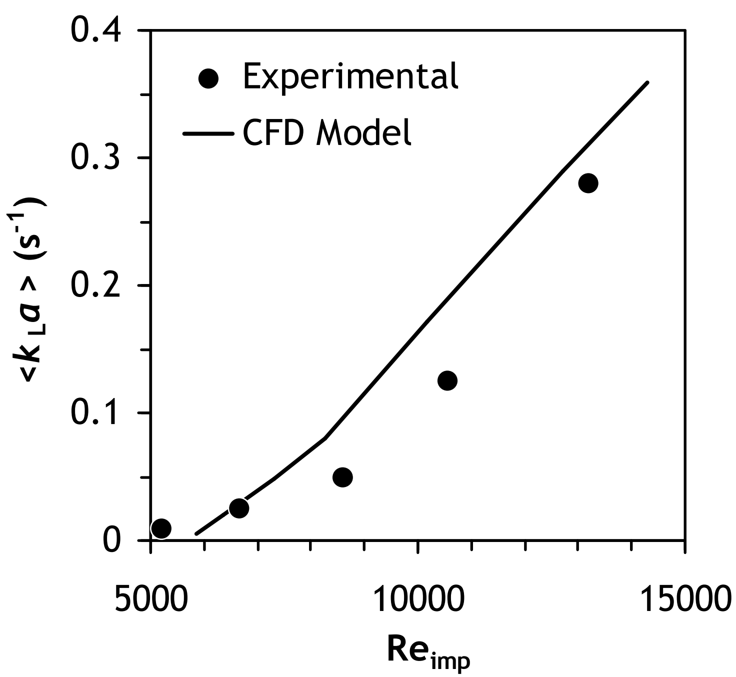
Specific surface area (symmetric model)

$$a = \frac{6}{d_{\mathrm{b}}}\alpha(1-\alpha)$$

Spatial distribution of $k_L a$ (s^{-1})



G-L mass transfer coefficient



G-L mass transfer coefficient as a function of the impeller rotational speed

- Global G-L mass transfer coefficients fairly well predicted by CFD in the studied range
- The curves trend is not the same, however
- Need for considering a bubble size distribution in the flow?
  - Multi-zonal bubble size model
  - Population balance modeling
- More accurate drag coefficient correction correlations?

Conclusions

- A CFD model has been proposed for simulating the flow in a STR for the testing of shaped catalysts
- Still a work in progress, but the developed CFD model is proving to be a valuable tool for the characterization of STR with stationary catalytic baskets
  - Defining maps of operational regimes for existing devices, reducing experimentation
  - Parametric design tool leading to intensified reactor designs
- Main difficulties have been due to dealing with phase interaction in turbulent multiphase flows
  - Available correlations for drag correction do not seem universal
  - Extrapolation of the correlations is needed
- Joint effort has been creating awareness and showing that the present testing reactor configurations and operational conditions may not be suited for measuring the kinetic constants of faster reactions
  - Limited by the mass transfer rate between the phases
  - Not always taken into consideration in laboratory tests

Thank you for your attention.

Questions? Suggestions?

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