Coupling direct numerical simulation with population balance modelling for predicting turbulent particle precipitation in a T-mixer

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What is particle precipitation?

What are in common in these products?

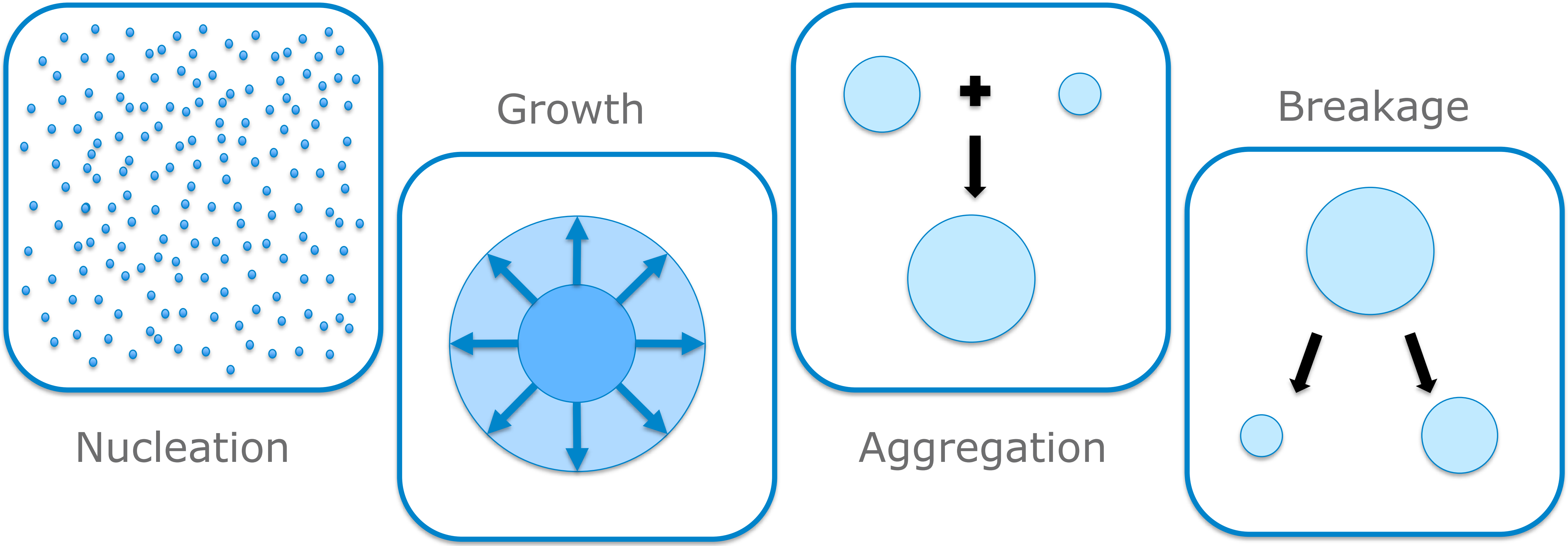
How are they made?

→ Precipitation: A Particulate Process

- Mixing reactants
- Supersaturated Solution
- Reaction between chemical species
- Phase change

General Particulate Processes

4 Mechanisms → Poly-disperse phase



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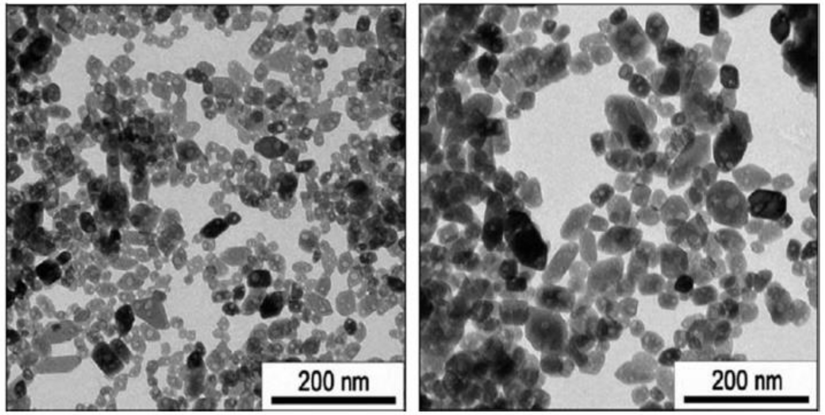
Particle size matters !

- The product quality usually correlates with the particle size distribution (PSD)

- Important quantity for manufacturers

- Interested in predicting and controlling the PSD



L.Metzger, et.al (2016)

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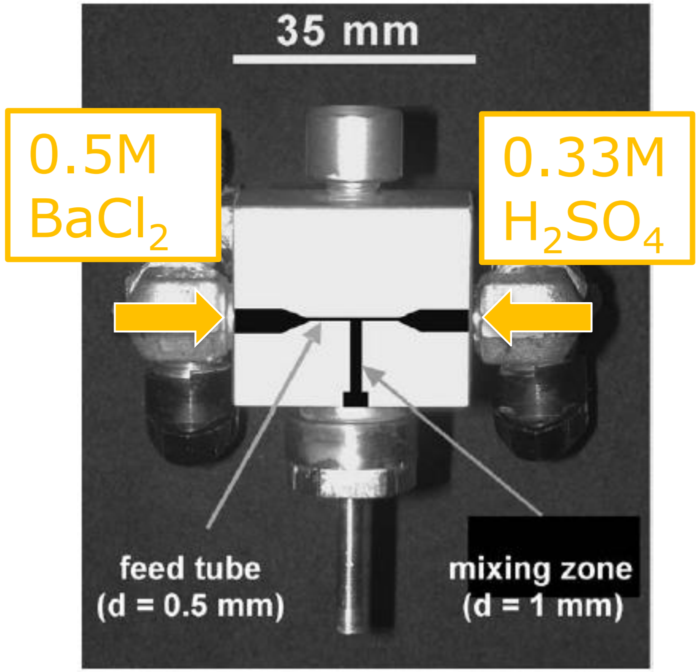
Challenges

- Precipitation is highly sensitive to the local supersaturation
  - Fast changing kinetics
- Fast reaction-precipitation
  - Small precipitation time scale
- Turbulent fluctuations alters the local composition
  - Small mixing length scale

- → PSD is changing rapidly in space and in time
  (especially when the flow time scale and kinetics are of similar order)
- → Local information can hardly be captured by experiments / resolved by simple numerical methods

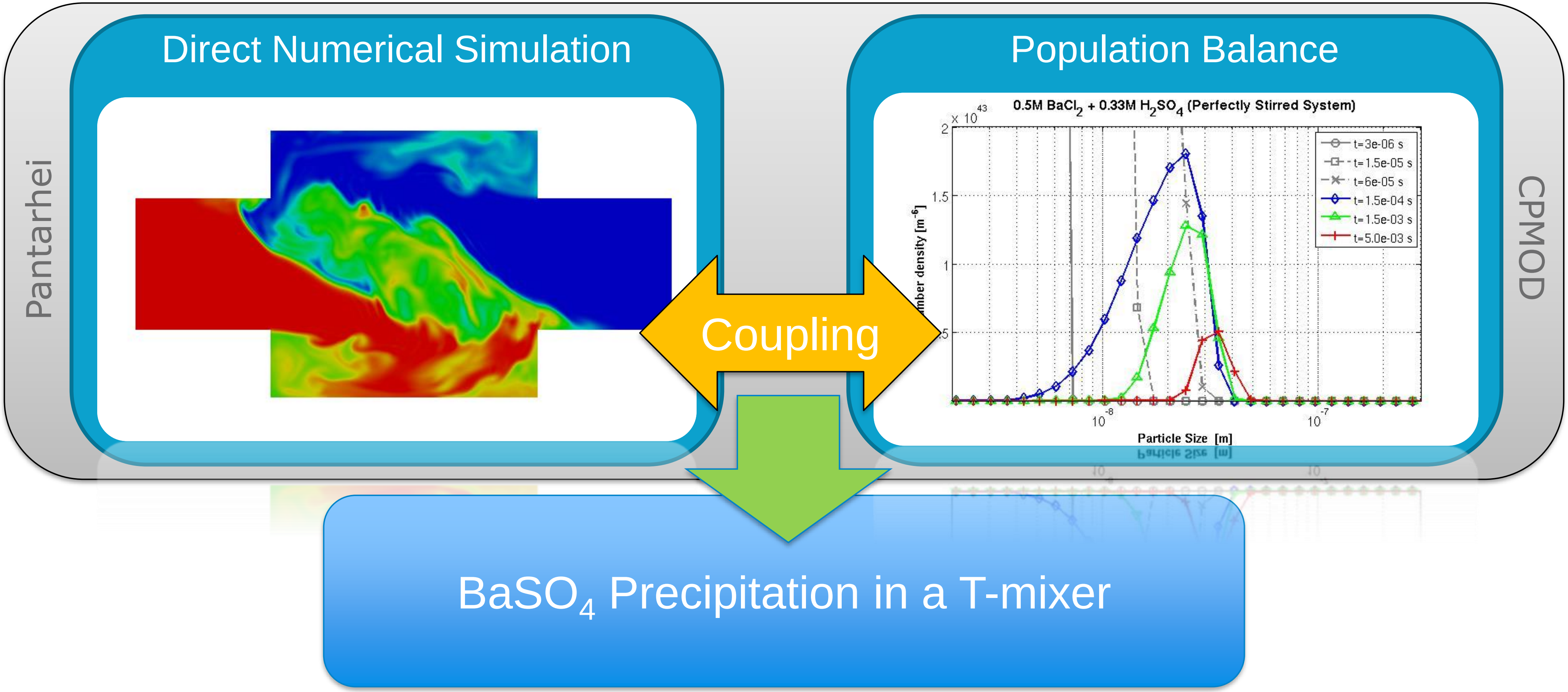
The motivation

- What are the effects of mixing on the precipitation process ?
- How the PSD is influenced by local effects ?
- Develop a coupled DNS-PBE approach for simulating particulate process in turbulent flow
- Case study: Apply the coupled DNS-PBE approach to the nano-particle precipitation of $BaSO_4$ in a T-mixer (H.-C. Schwarzer, 2004)



H.-C. Schwarzer, et. Al (2004)

What this project is about?



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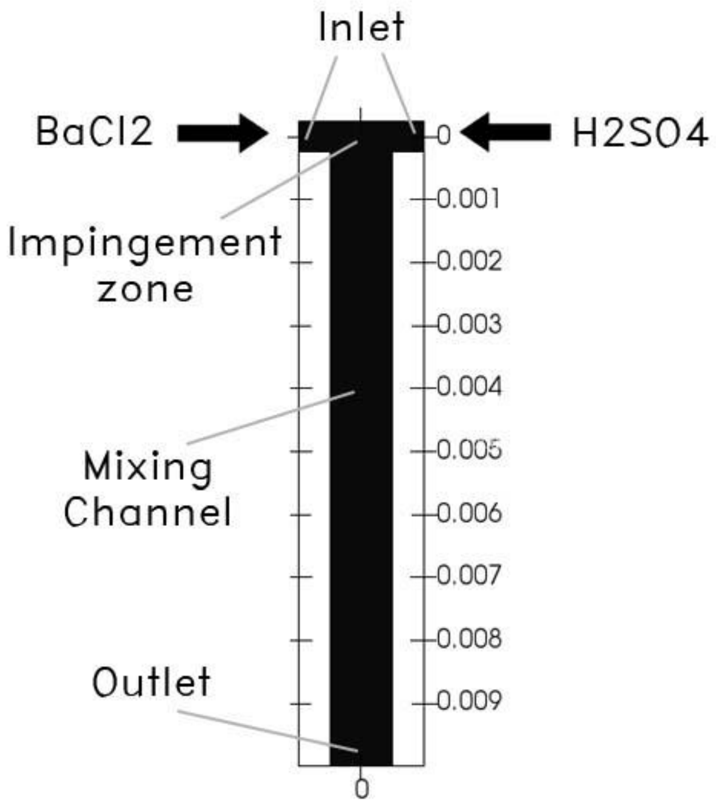
Direct numerical simulation (DNS)

- N-S equations: $$\frac{\partial \boldsymbol{u}}{\partial t} + \boldsymbol{u} \cdot \nabla \boldsymbol{u} = -\nabla P + \upsilon \nabla^2 \boldsymbol{u}$$
- Transport for ions: $$\frac{\partial C_i}{\partial t} + \boldsymbol{u} \cdot \nabla C_i = \frac{\upsilon}{Sc} \nabla^2 C_i + r_i$$
- Fraction step
- BDF3 for time advancement
- Implicit diffusion and explicit convection

- Cartesian mesh
- 21 million cells
- Flow field is fully resolved to Kolmogorov scale

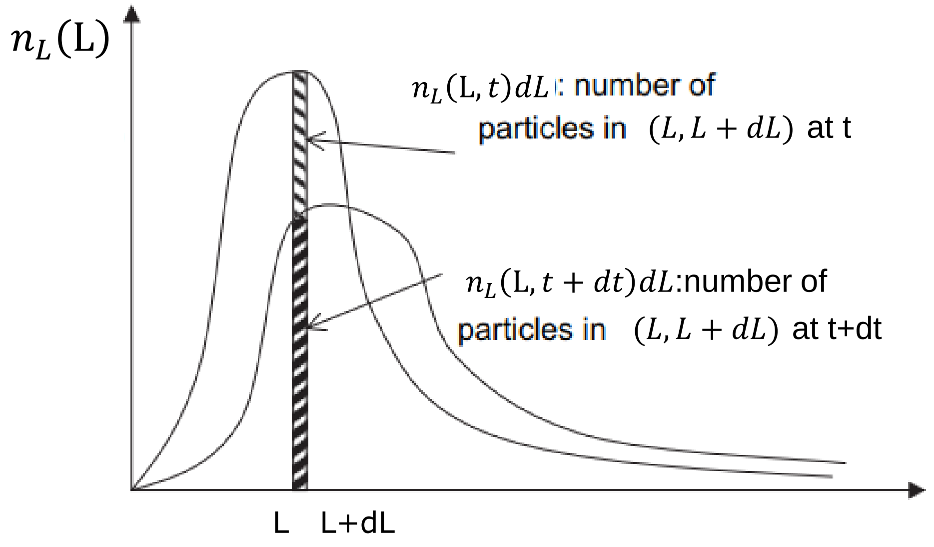


Population Balance Modelling

- Particle number
  - Discretised into intervals
  - Number density

$$n_L(L) = \frac{dN(L)}{VdL}$$

Evolution of Number density distribution in time



S. Rigopoulos (2010)

  - General form of Population Balance Equation (PBE) for particulate processes

$$\underbrace{\frac{\partial n_L}{\partial t}}_{\text{Transient term}} + \underbrace{\frac{\partial G \cdot n_L}{\partial L}}_{\text{Growth term}} = \underbrace{B_0}_{\text{Nucleation term}} + \underbrace{B_{aggl}}_{\text{Generation term due to aggregation}} + \underbrace{B_{disr}}_{\text{Generation term due to breakage}} - \underbrace{D_{aggl}}_{\text{Destruction term due to aggregation}} - \underbrace{D_{disr}}_{\text{Destruction term due to breakage}}$$

N	Total number of particles
n_L	Number density
L	Size interval (Class)
B_0	Nucleation term
B	Generation term
D	Destruction term
G	Growth rate

In the current case study, the system is dominated by nucleation and growth only

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Transported PBE

- Coupling with the flow
- Assume particles as tracer

Convection term

$$\frac{\partial n_L}{\partial t} + \boldsymbol{u} \cdot \nabla n_L + \frac{\partial G \cdot n_L}{\partial L} = \frac{\upsilon}{Sc} \nabla^2 n_L + B_0$$

Diffusion term

- PBE is solved locally in each cell together with the DNS

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$$\overbrace{\frac{\partial n_j}{\partial t} + \boldsymbol{u}\cdot\nabla n_j}^{\text{Number transport}} = \frac{\upsilon}{\mathrm{Sc}}\nabla^2 n_j + \overbrace{B_0 - \frac{\partial G\cdot n_j}{\partial L}}^{\text{Source term}}$$

Nucleation

Growth

Semi-empirical relations

- The flow field is captured by DNS, which can be considered as fully resolved
- The only modelling terms are the growth and nucleation rates
- These kinetics depends on the supersaturation (driving force) and are non-linear

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The kinetics

$$\frac{\partial n_j}{\partial t} + \boldsymbol{u}\cdot\nabla n_j = \frac{\upsilon}{\mathrm{Sc}}\nabla^2 n_j + \boldsymbol{B_0} - \frac{\partial \boldsymbol{G}\cdot n_j}{\partial L}$$

Supersaturation

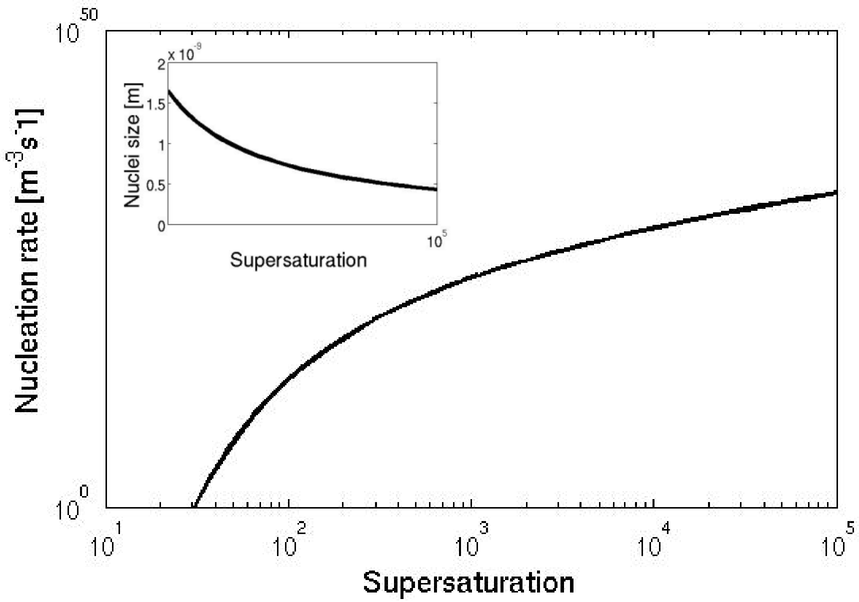
$$S = \gamma_{\pm}\sqrt{\frac{C_{Ba^{2+}}\, C_{SO4^{2-}}}{K_{sp}}}$$

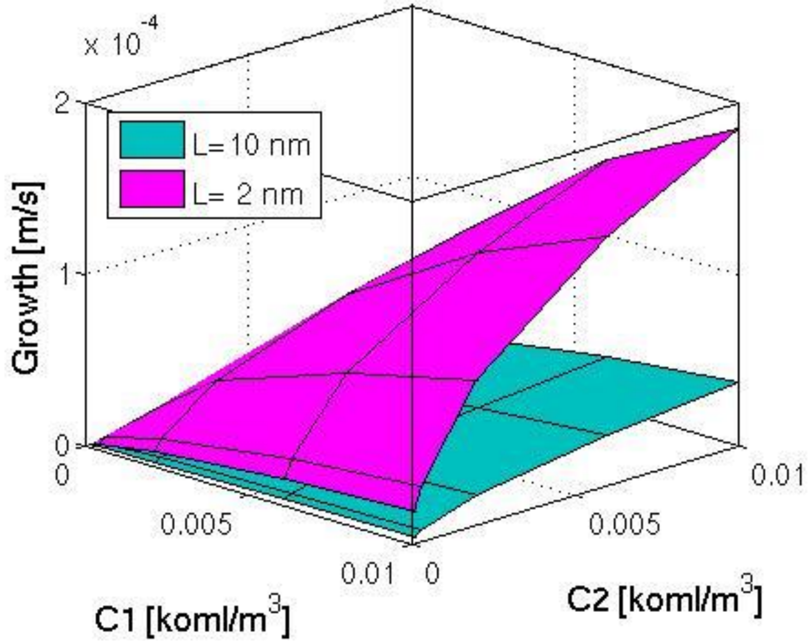
Nucleation rate

$$B_0 = 1.5D\left(\sqrt{K_{sp}}\,\boldsymbol{S}N_A\right)^{\frac{7}{3}}\sqrt{\frac{\gamma}{k_B T}}\,V_m \exp\left(-\frac{16\pi}{3}\left(\frac{\gamma}{k_B T}\right)^3 \frac{V_m^2}{(\upsilon \ln \boldsymbol{S})^2}\right)$$

growth rate

$$G = 2\,\frac{Sh\, D_\alpha \sqrt{K_{SP}}\, M_w}{\rho_{BaSO4}}\,\frac{\boldsymbol{S}-1}{\boldsymbol{L}}$$





Precipitation timescales

$$\tau_{\mathrm{nuc}} = \frac{\int n(L)dL}{B_0}$$

$$\tau_{\mathrm{G}} = \frac{k_v \int L^3 n(L)dL}{k_a \int G(L)L^2 n(L)dL}$$

Discretisation on the transported PBE

$$\frac{\partial n_L(j)}{\partial \mathrm{t}} + \boldsymbol{u} \cdot \nabla n_L(j) = \frac{v}{\mathrm{Sc}} \nabla^2 n_L(j) + B_0 \delta(L_j - L_{nuc}) - \frac{\partial G_j \cdot n_L(j)}{\partial L_j}$$

- Fractional step method
- BDF3 for time advancement
- Explicit convection and diffusion terms are extrapolated with 3rd order scheme
- TVD scheme for growth discretization

- Particle size range is discretised into 40 intervals (j = 1 - 40) with an exponential grid

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Flow field & Mixing

- Re = 1135

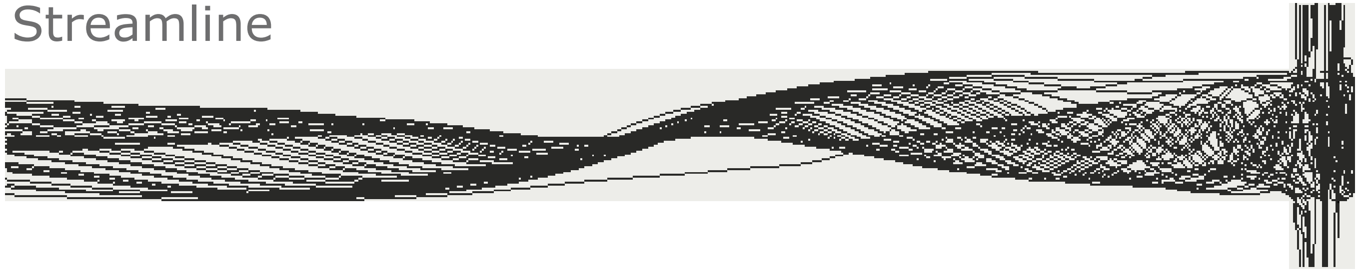
The flow in T-mixer can be considered as turbulent when Re>400 (Telib, et al., 2004)

- Helical Pattern

- Intense mixing at impingement zone

- Fastest mixing time scale in the order of 10^{-5}

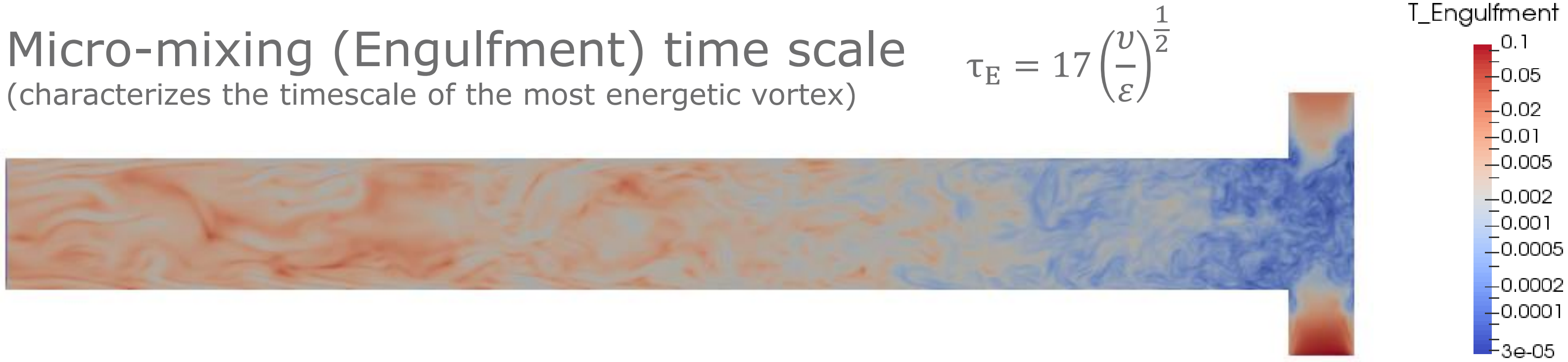
Streamline



Micro-mixing (Engulfment) time scale

(characterizes the timescale of the most energetic vortex)

$$\tau_E = 17\left(\frac{\upsilon}{\varepsilon}\right)^{\frac{1}{2}}$$



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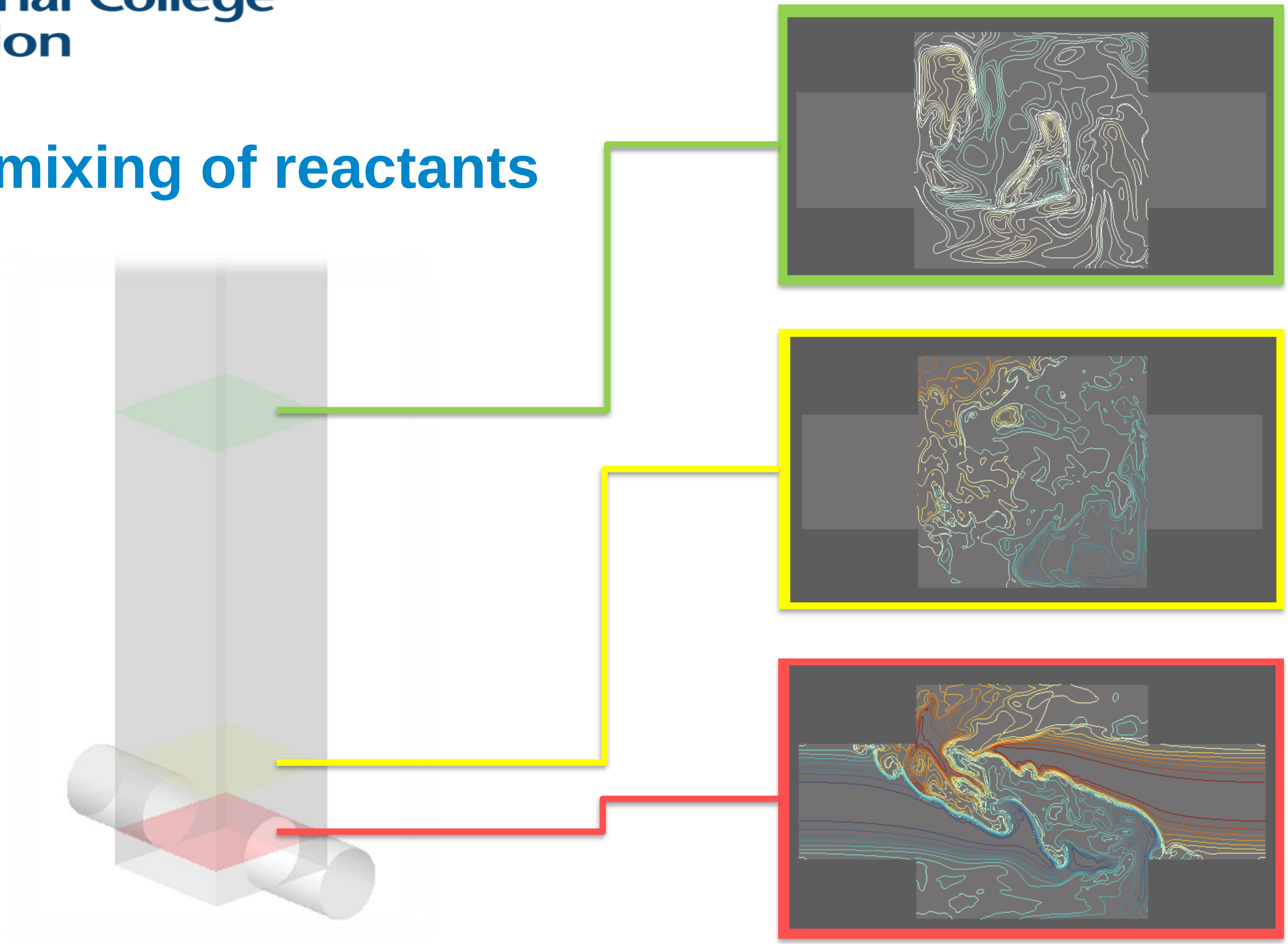
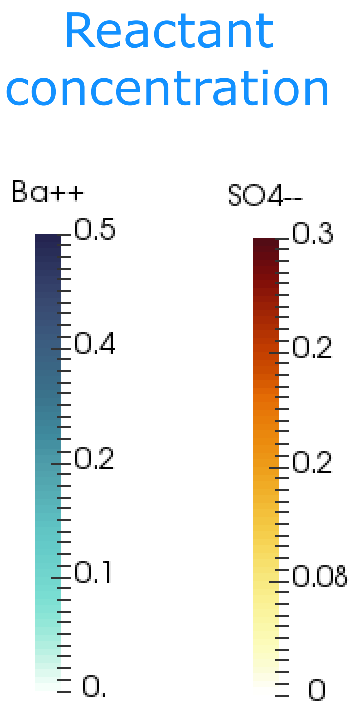
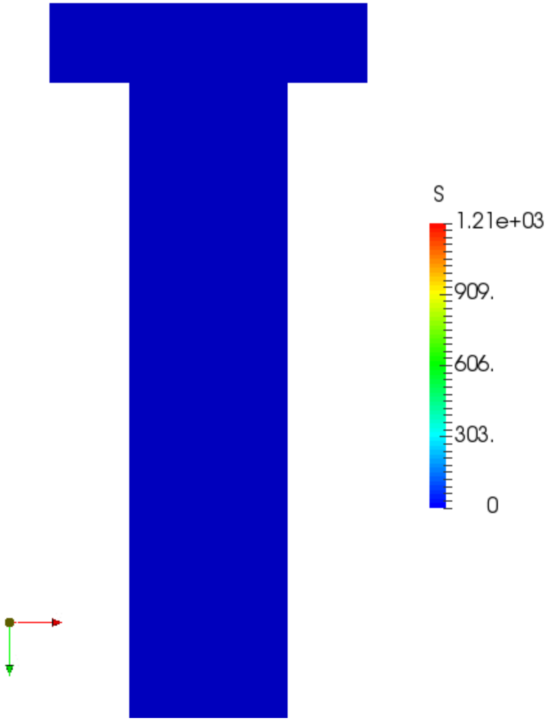
The mixing of reactants



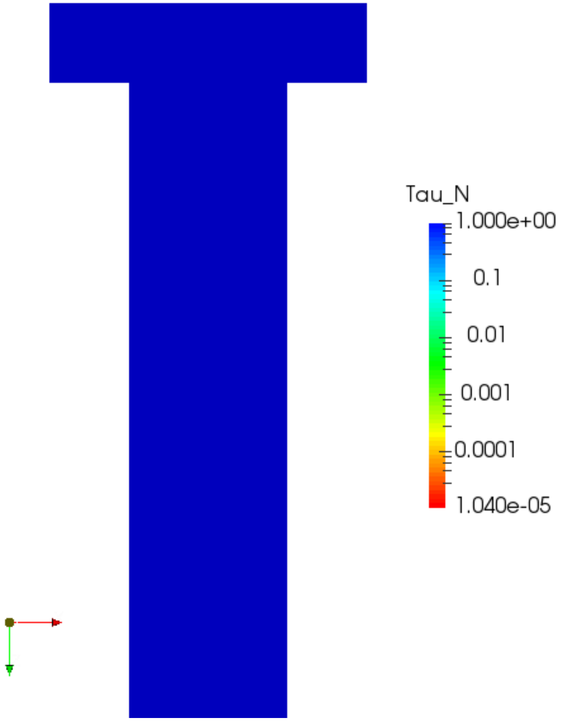


The local kinetics

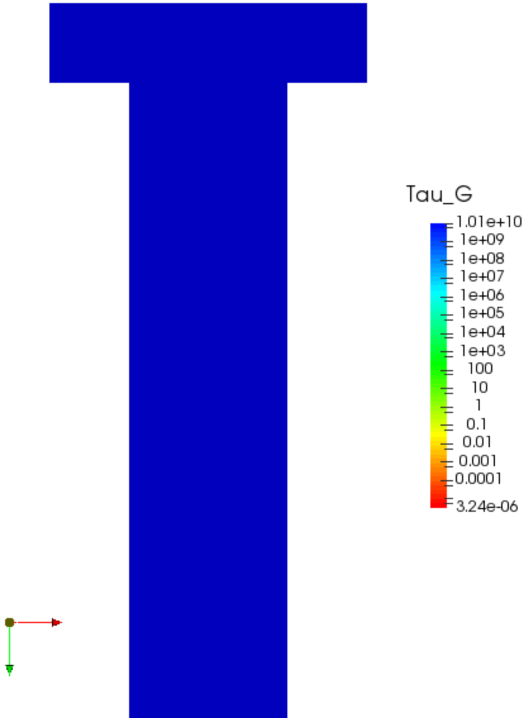
Supersaturation



Nucleation timescale



Growth timescale



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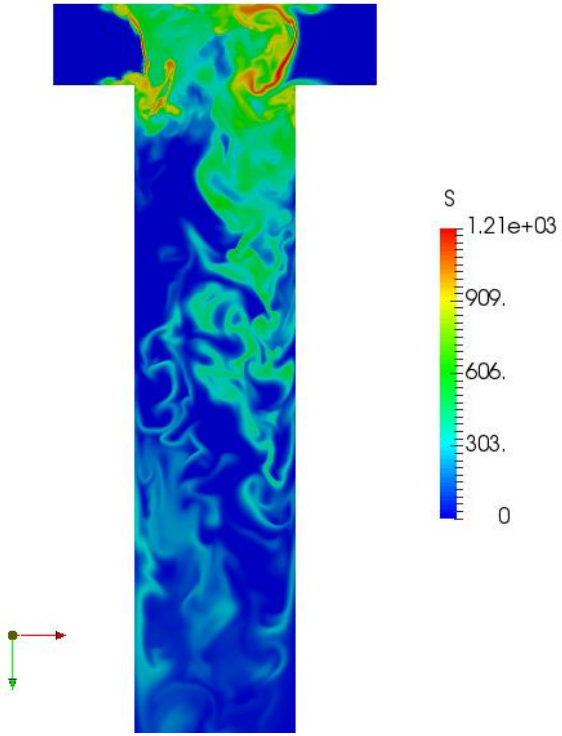
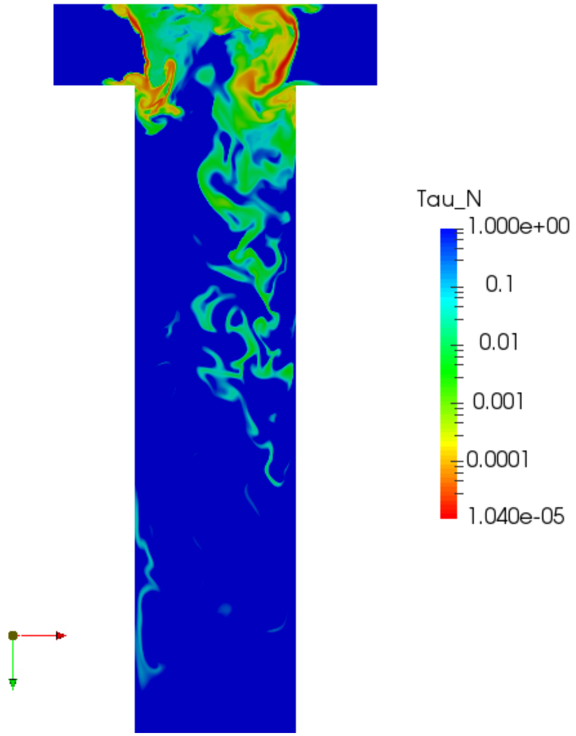
The local kinetics

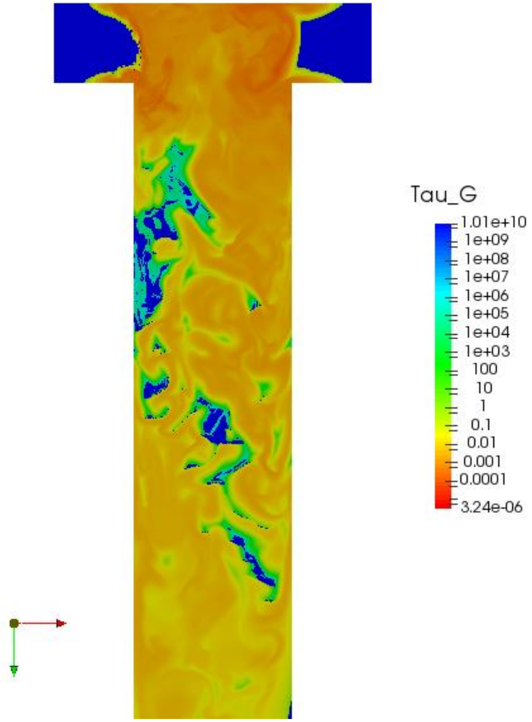
Supersaturation



Nucleation timescale



Growth timescale



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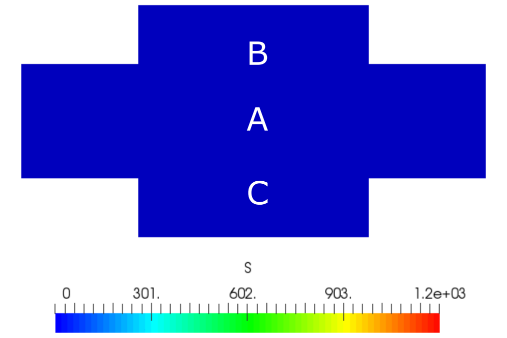
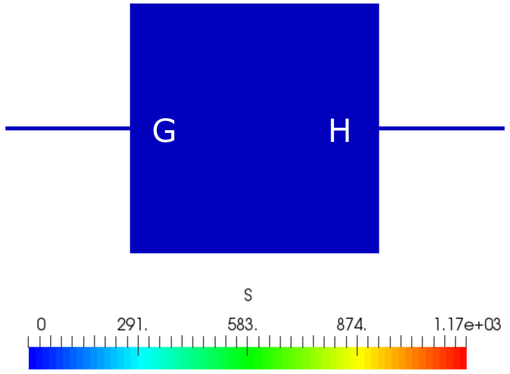
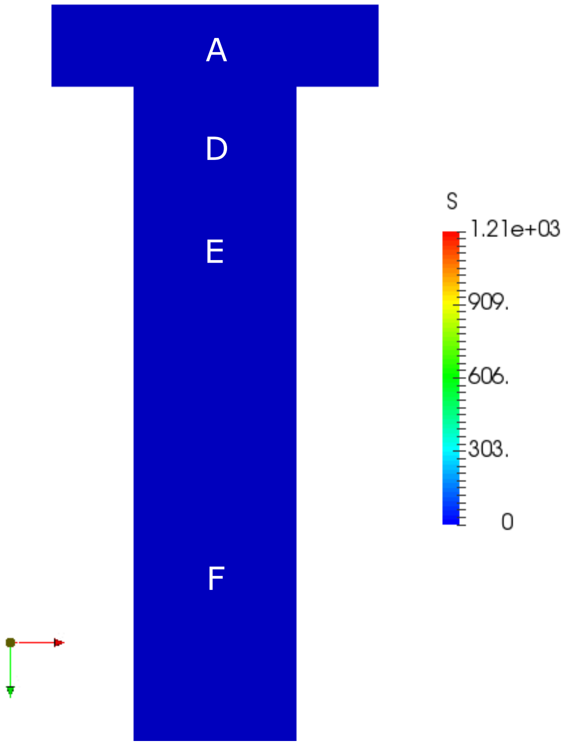
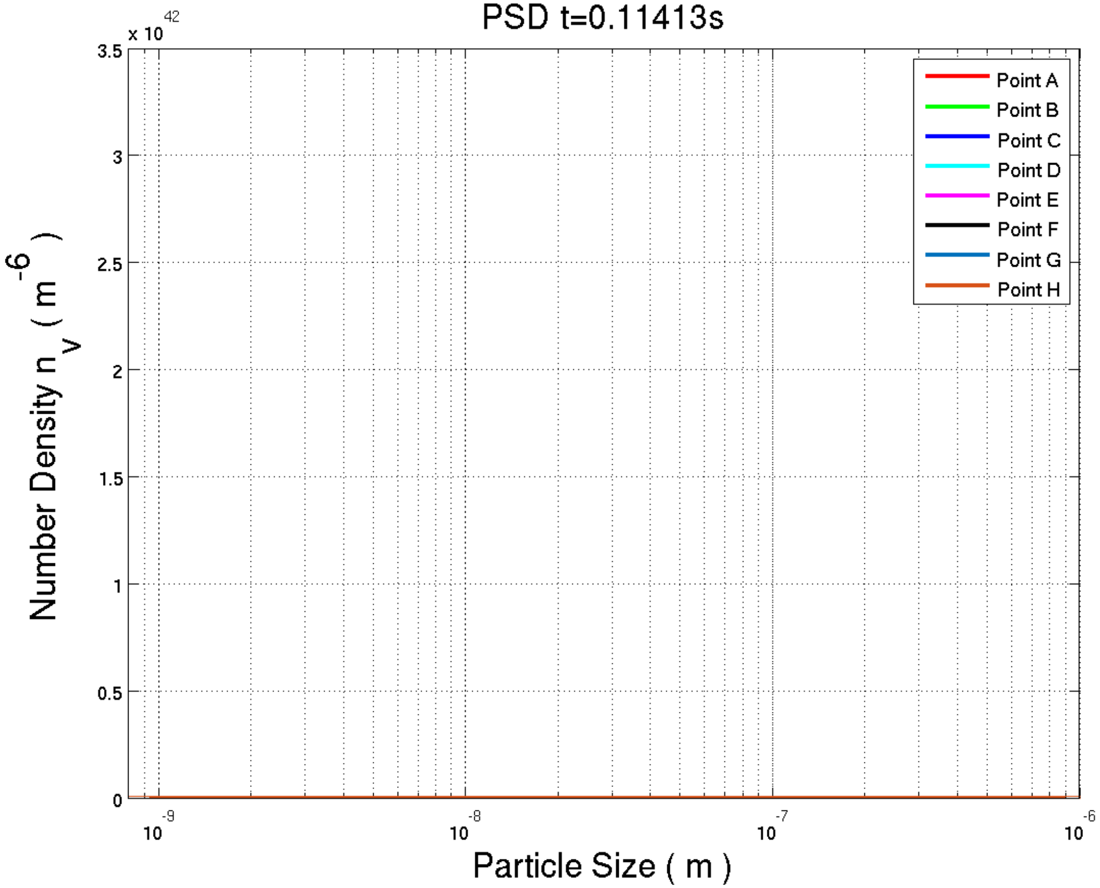
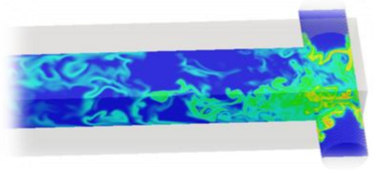
The PSD











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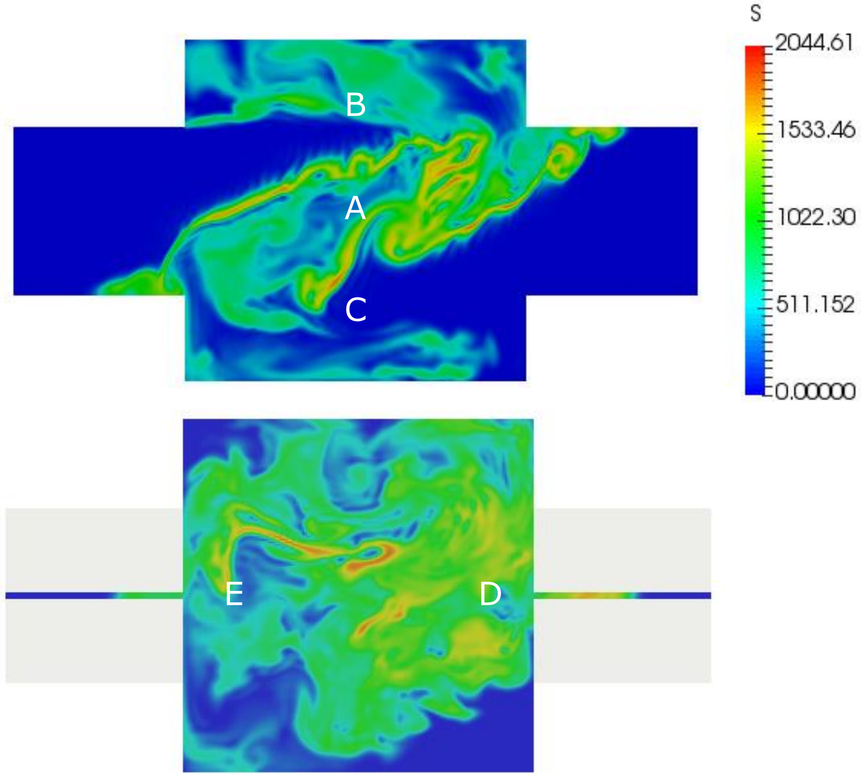
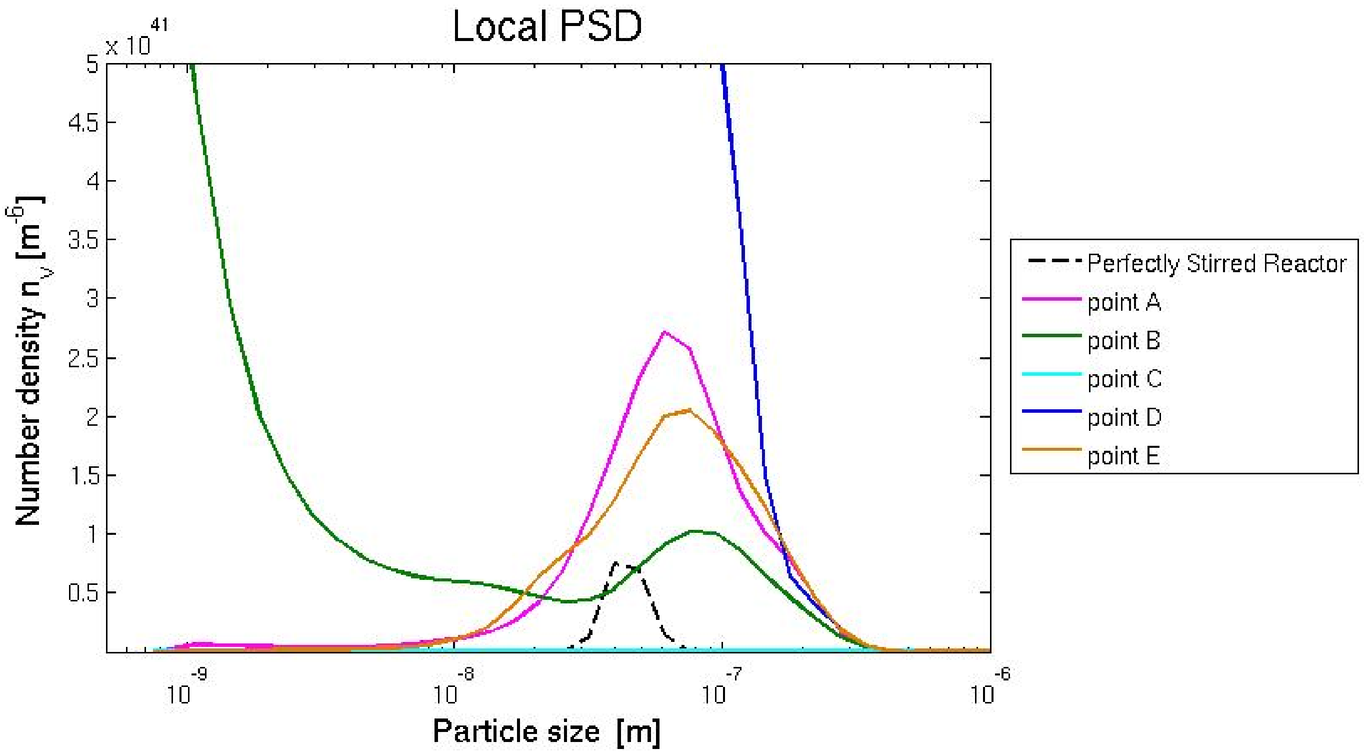
The PSD (cont'd)





Conclusion

- DNS – PBE coupled approach on $BaSO_4$ precipitation
- Local variation in the reactant leads to different local dominating mechanism
- The impingement zone is the most critical region and it determine the resultant PSD

Acknowledgement

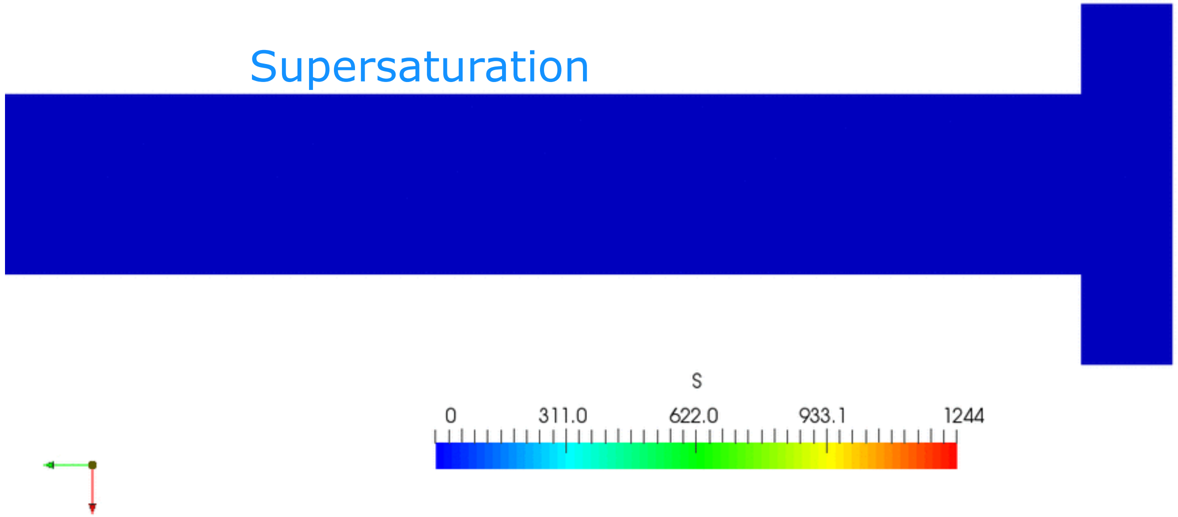
- This work used the ARCHER UK National Supercomputer Service (http://www.archer.ac.uk) and was supported by the UK Consortium on Turbulent Reacting Flows [EP/K026801/1].

- The authors acknowledge the IChemE's Fluid Mixing Processes Special Interest Group and Syngenta for their generous financial support.

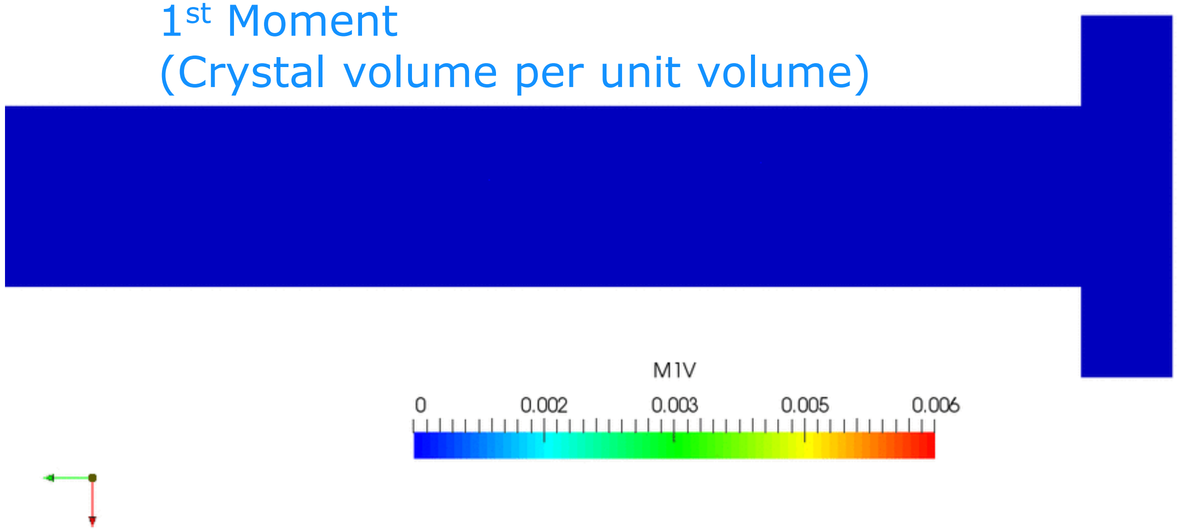
Thank you

Moment of distribution

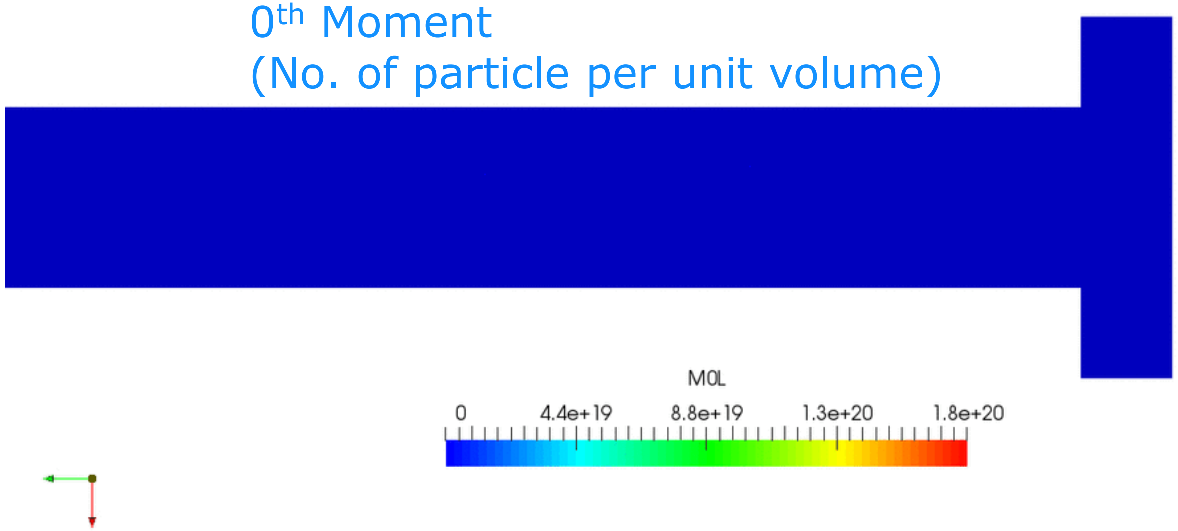
Supersaturation



1^{st} Moment
(Crystal volume per unit volume)



0^{th} Moment
(No. of particle per unit volume)



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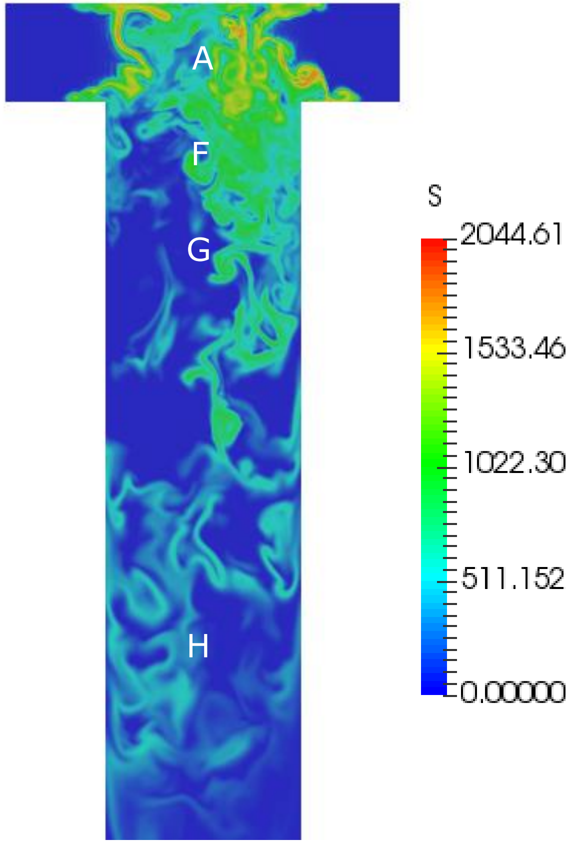
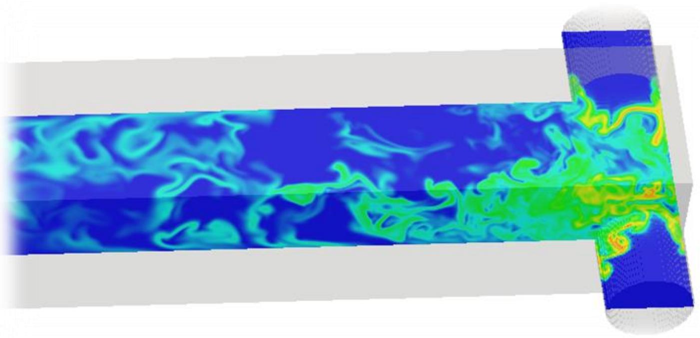
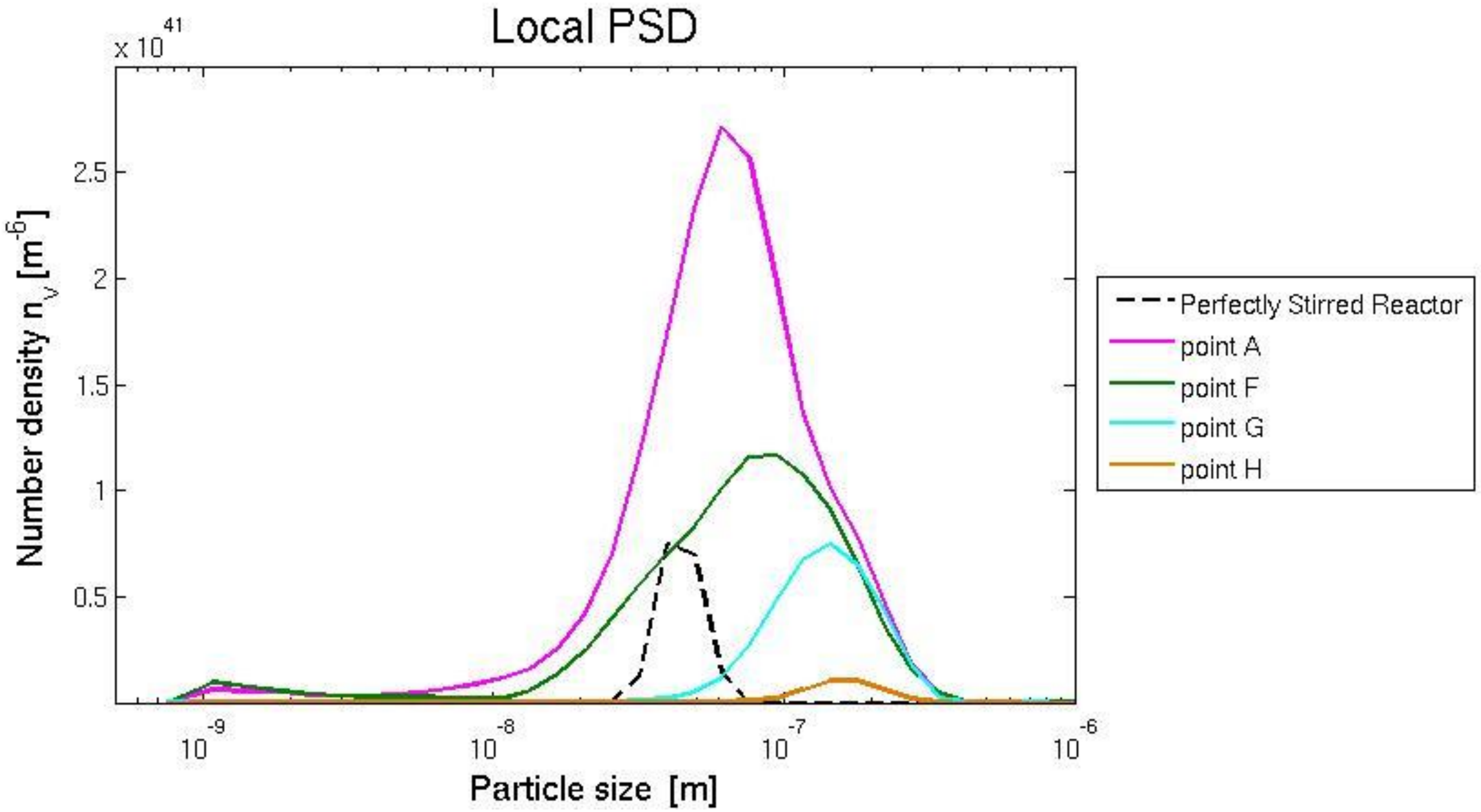
The PSD (cont'd)







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Intensity of Segregation
(a measure of evenness)

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Computational cost

- Expensive but possible on HPC
- High resolution
- Will consider simplifying this to resolve zones with strong mixing only

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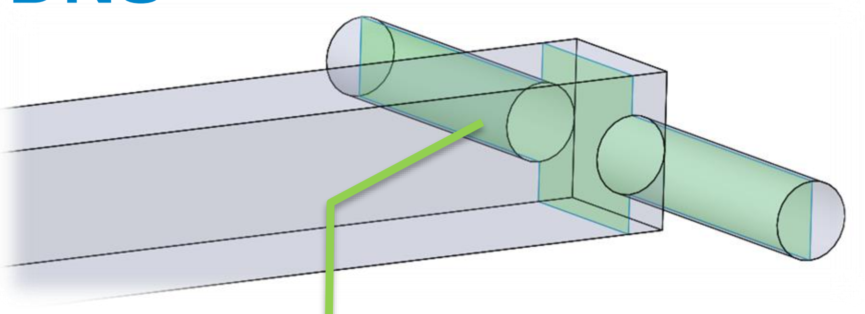
Intensity of segregation

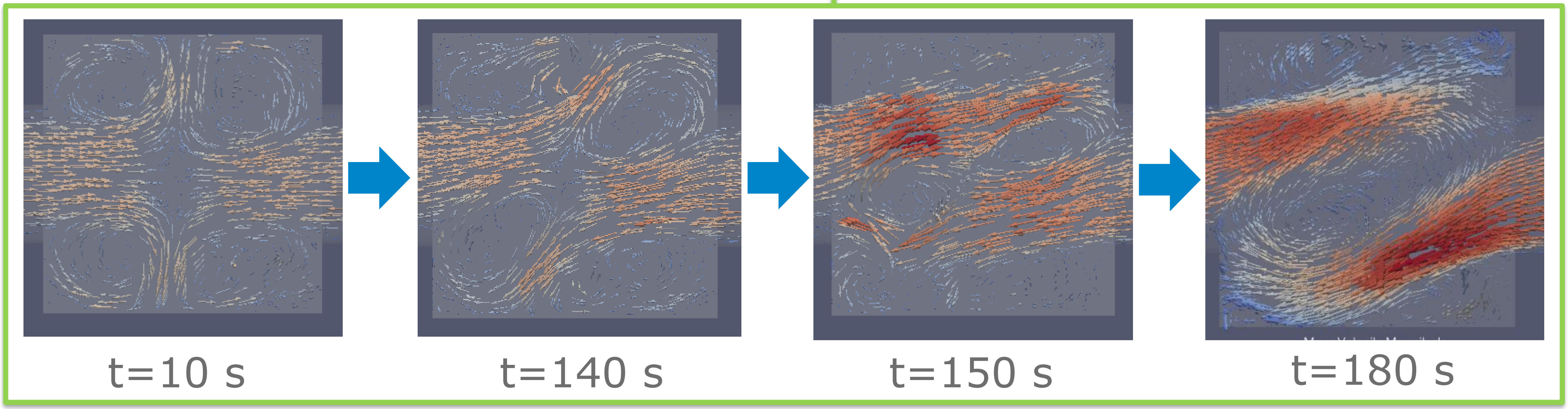
- A measure of evenness

- 1 - Fully segregated
- 0 – Homogeneously mixed

$$I = \frac{\sum_{m=1}^{M} (\overline{x_A} - x_{Am})^2}{M(\overline{x_A}(1 - \overline{x_A}))} = \frac{\overline{(\overline{x_A} - x_A)^2}}{\overline{x_A}(1 - \overline{x_A})}$$

Flow field obtained from DNS





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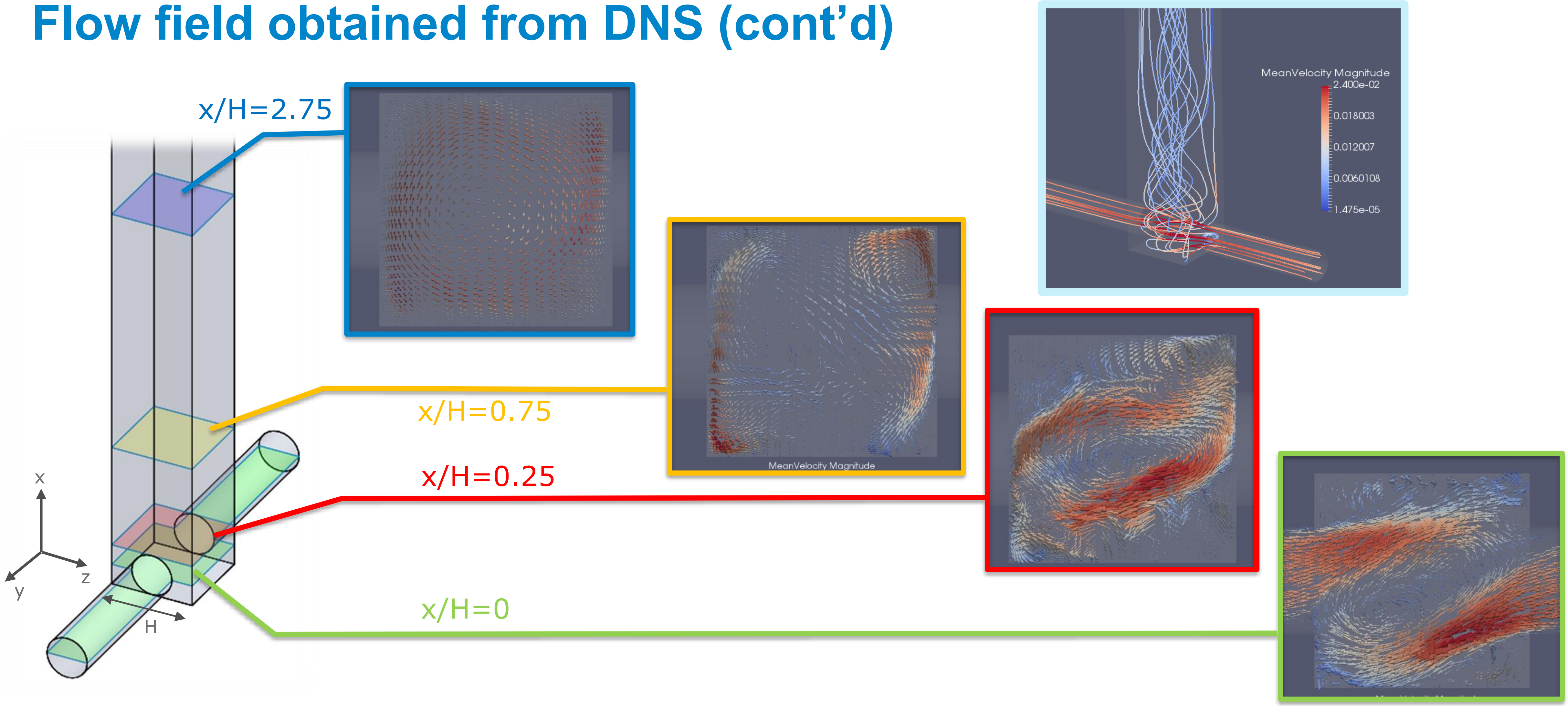
Flow field obtained from DNS (cont'd)



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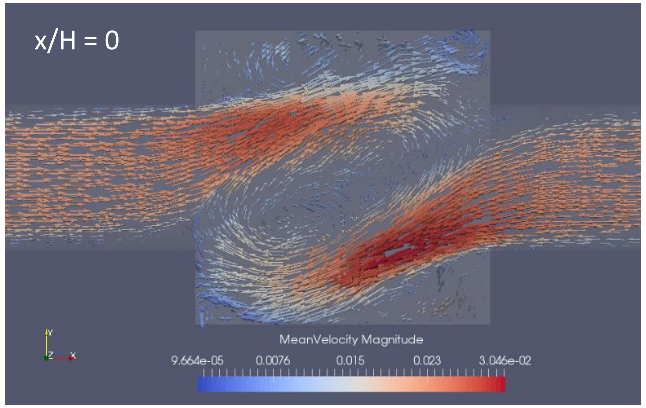
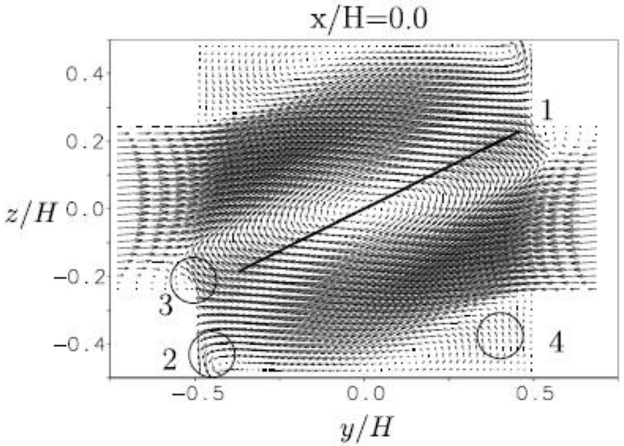
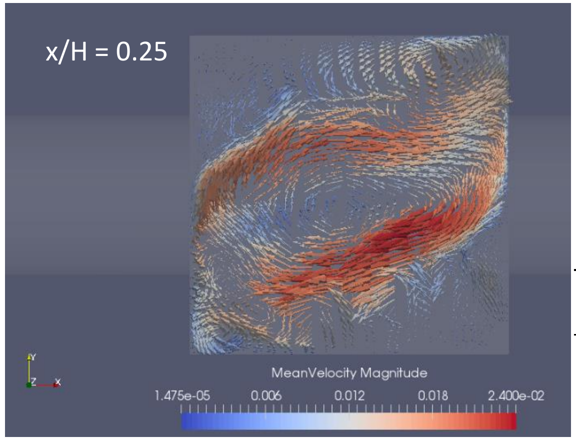
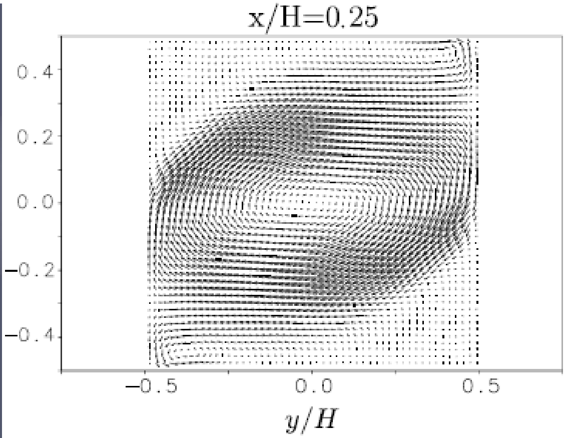
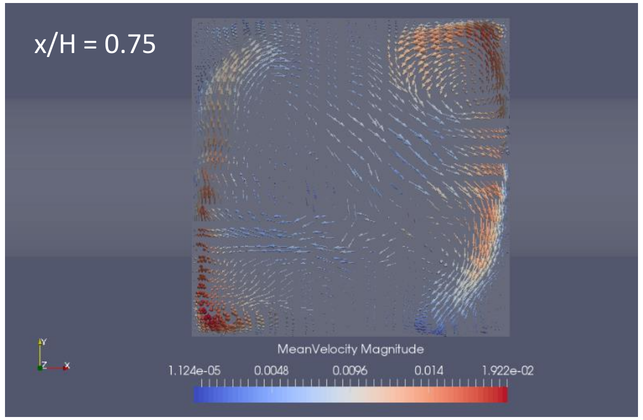
Compare with experimental PIV results[4]

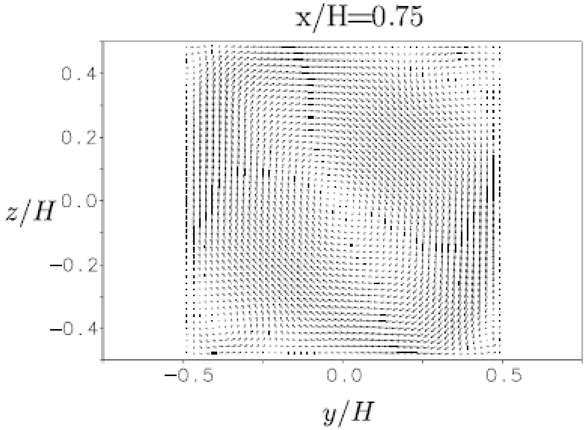


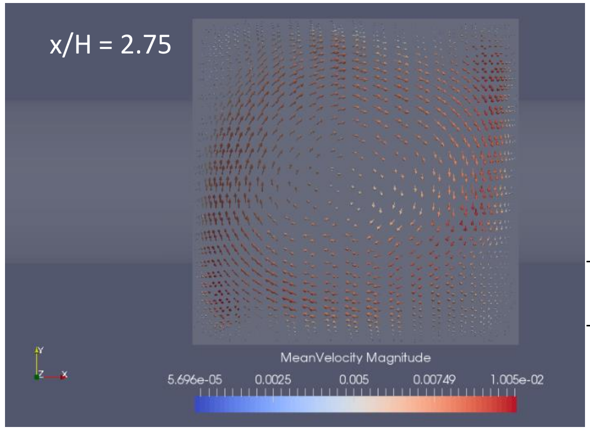


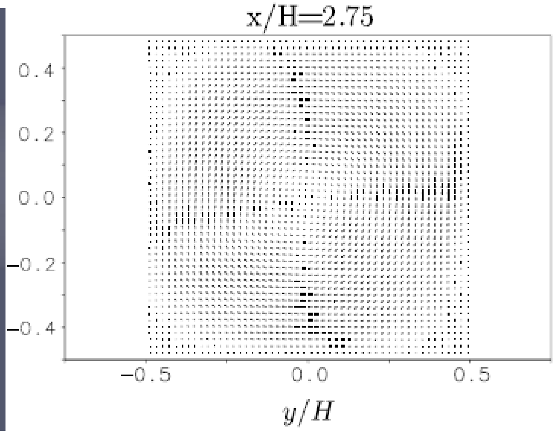












[4] F. Schwertfirm, J. Gradl, H. Schwarzer, W. Peukert and M. Manhart, “The low Reynolds number turbulent flow and mixing in a confined impinging jet reactor,” International Journal of Heat and Fluid Flow, vol. 28, pp. 1429-1442, 2007.

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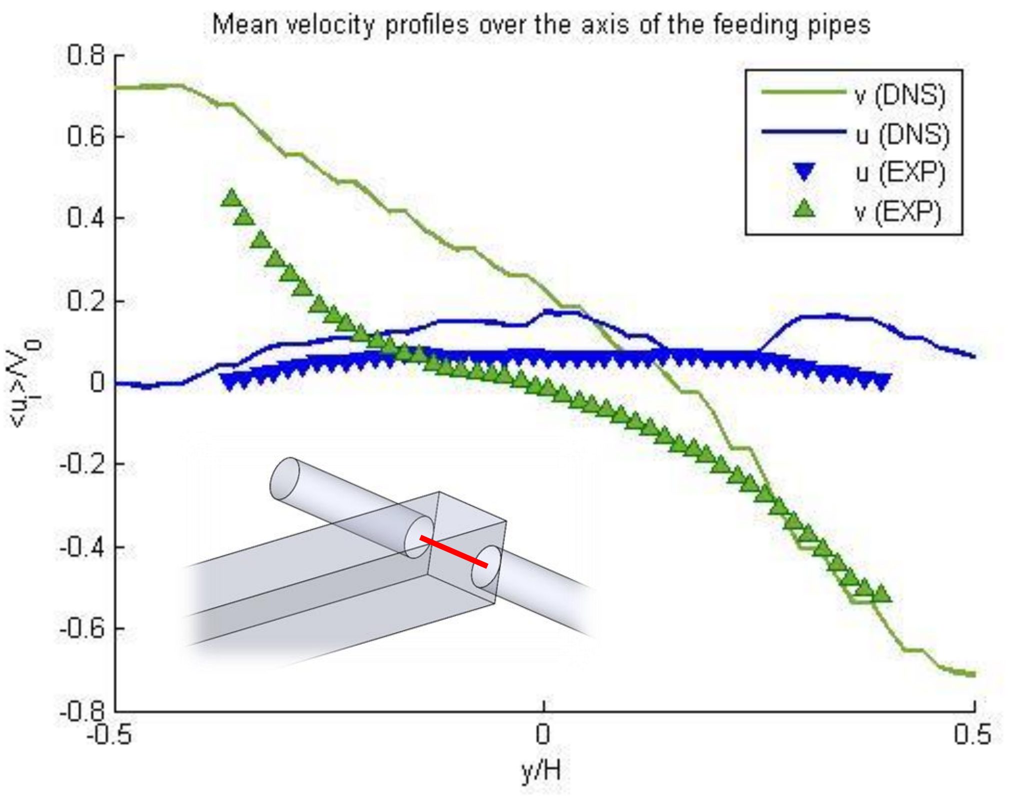
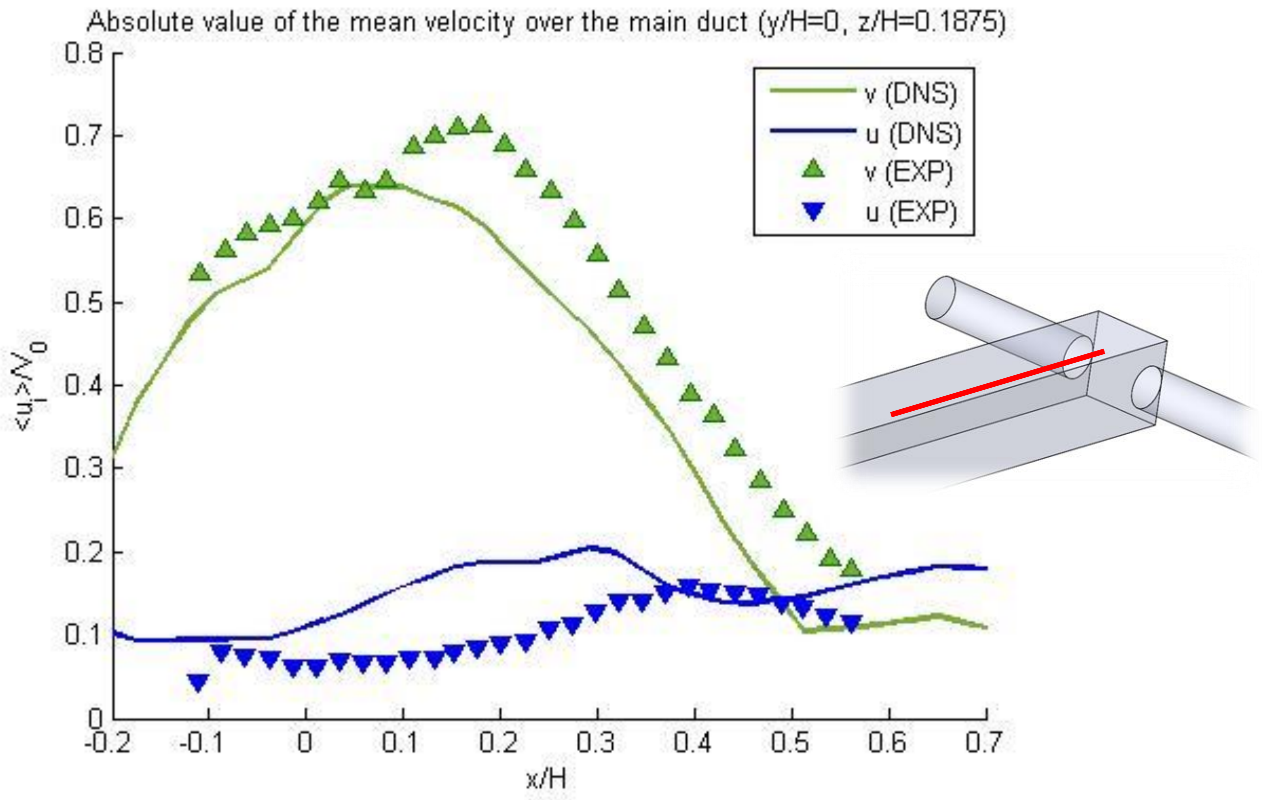
Velocity Profile
(preliminary comparison with experimental results[4])





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