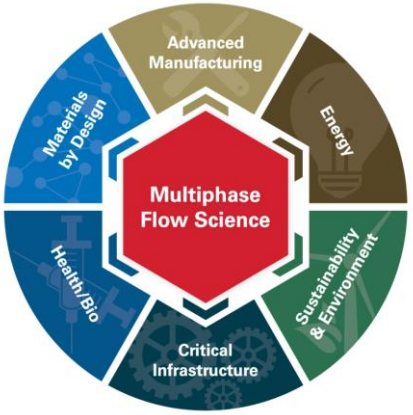

CoMFRE Multiphase Flow Research

IOWA STATE UNIVERSITY

Quadrature-based moment methods for turbulent mixing and gas-liquid flows

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EM2C Seminar – July 5th, 2017

Outline

1. Problems of interest
2. PDF transport models for turbulent reacting flows
3. Mixing, reaction and population balance in multi-inlet vortex reactor
4. Quadrature-based moment methods for bubbly flows
5. Conclusions

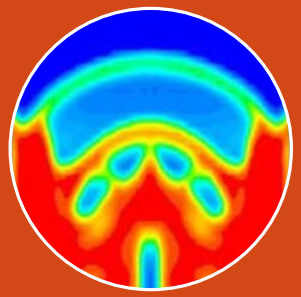
Families of problems of interest

Population balance equations

- Non-inertial particles ($\mathrm{St} \to 0$)
- Turbulent mixing
- Reacting flows

Gas-liquid systems

- Small inertia ($\mathrm{St} < 1$)
- Coalescence, breakup
- Free surfaces



Gas-solid systems

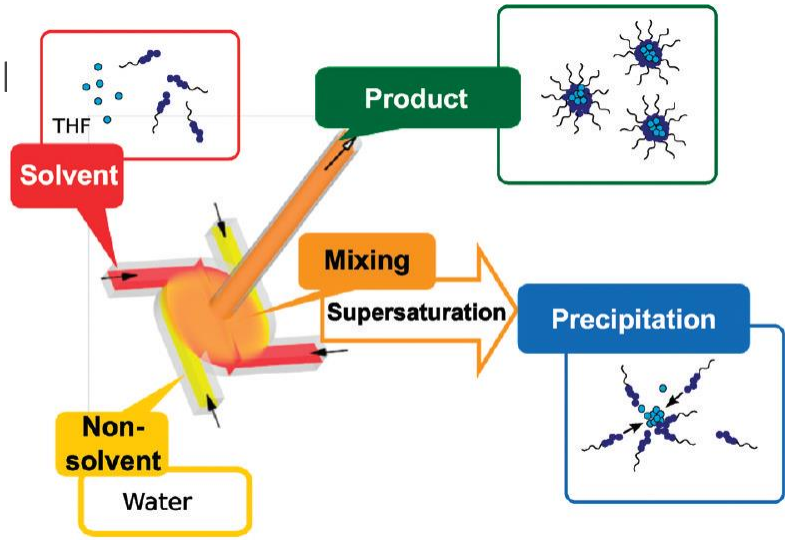
- Large particle inertia ($\mathrm{St} \gg 1$)
- Particle trajectory crossing
- Nearly packed regions

Multi inlet vortex reactor (MIVR)

The multi-inlet vortex reactor (MIVR) is used for flash nanoprecipitation to manufacture functional nanoparticles:

- **Electronics:** circuit printing, sensors.
- **Drug Delivery**: nano-sized micelle accumulate in tumors via the enhanced permeability and retention effect.
  - **Nanoparticles doped on hydrogels for cancer treatment**

The MIVR achieves fast mixing by inducing turbulent swirling flow from four inlet streams.

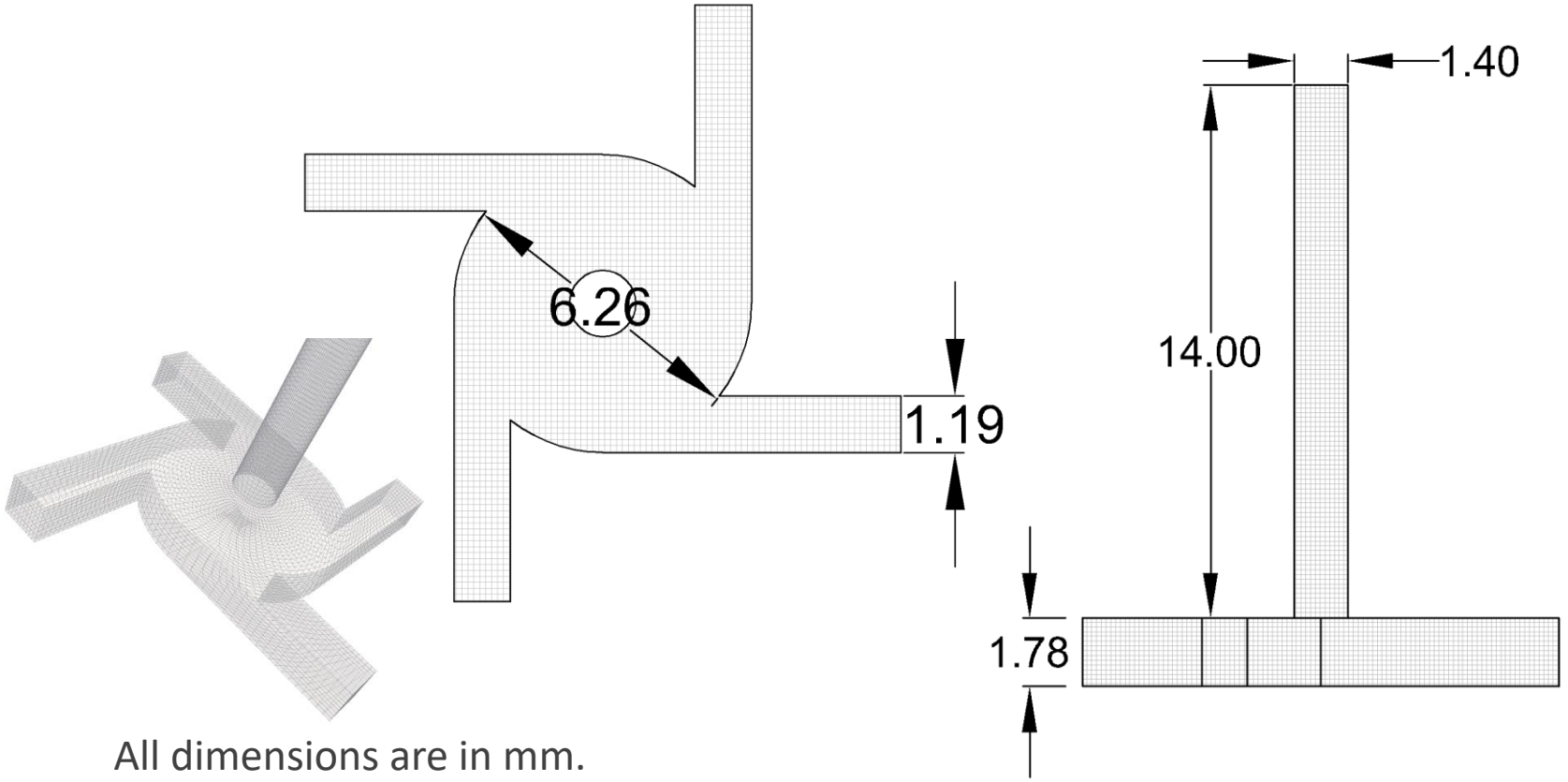


J. C. Cheng and R. O. Fox, "Kinetic Modeling of Nanoprecipitation using CFD Coupled with a Population Balance," Industrial & Engineering Chemistry Research, vol. 49, pp. 10651-10662, Nov 3 2010

Numerical models are needed to support the design and optimization of MIVRs. They need to predict:

- Fluid flow
- Mixing
- Precipitation and particle size evolution

Reference multi-inlet vortex reactor

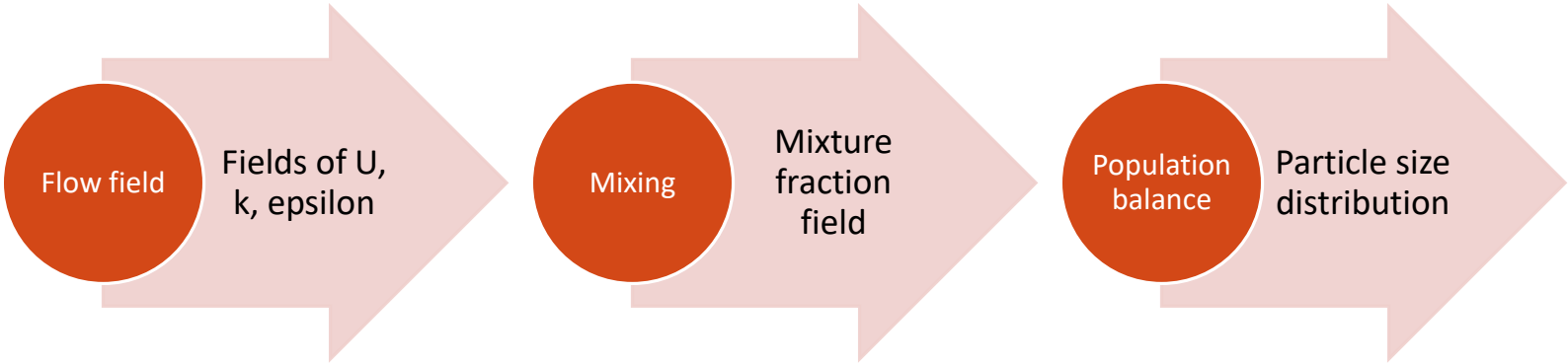


All dimensions are in mm.

Y. Shi, J. C. Cheng, R. O. Fox and M. G. Olsen, "Measurements of turbulence in a microscale multi-inlet vortex nanoprecipitation reactor" *Journal of Micromechanics and Microengineering,* vol. 23, 075005(10pp), 2013.

Simulation workflow

A sequential workflow is used to investigate the MIVR:

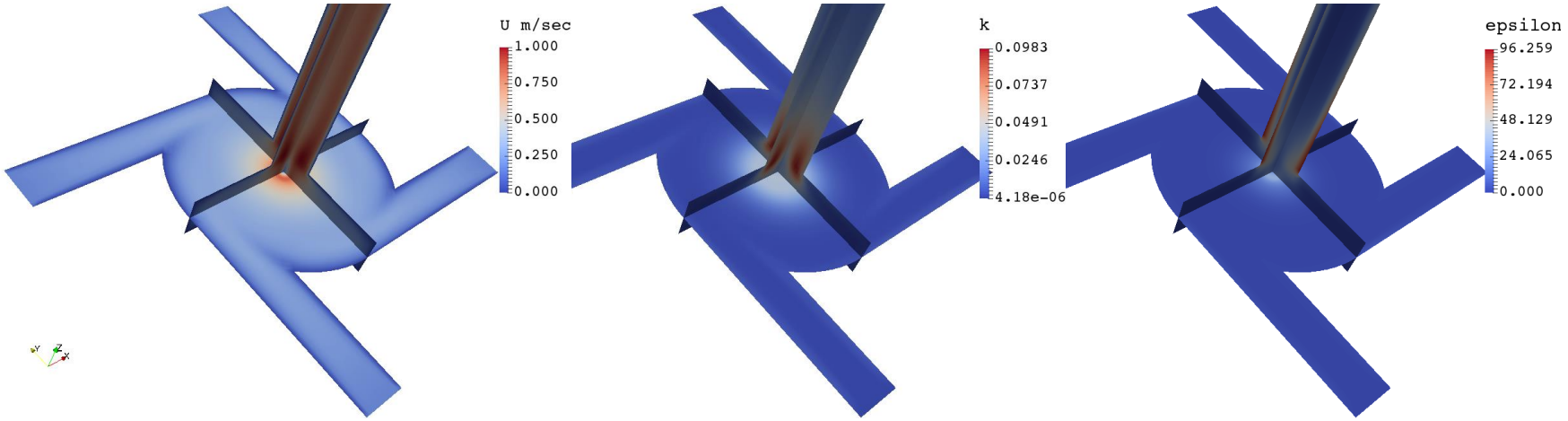


Assumptions

- Particles have small Stokes number and do not affect the fluid motion
- Flash nano-precipitation does not significantly affect mixing

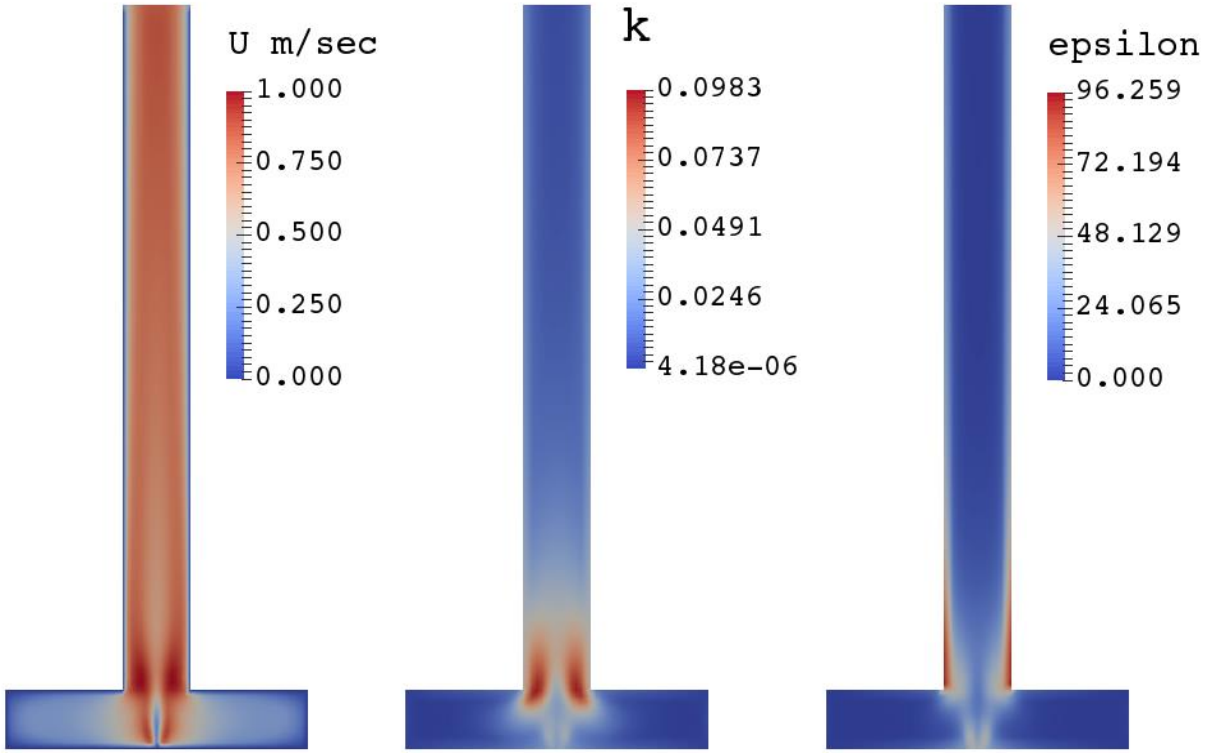
Flow field

- Incompressible steady-state RANS or DDES simulation (simpleFoam solver in OpenFOAM® with LRR model or pimpleFoam with customized DDES)
- MIVR generates swirling flow, with higher turbulent intensity in the center of the mixing chamber



Steady state solution of the velocity (left), turbulent kinetic (middle) and turbulent dissipation (right) of the flow.

Flow field and turbulent quantities



Steady state solution of the velocity (left), turbulent kinetic (middle) and turbulent dissipation (right) of the flow.

Reactive mixing modeling

We consider the evolution equation for the joint composition PDF $f(\boldsymbol{\phi}, \mathbf{x}, t)$:

$$\overbrace{\frac{\partial f_\phi}{\partial t}}^{\text{rate of change}} + \overbrace{\langle U_i \rangle \frac{\partial f_\phi}{\partial x_i}}^{\text{macromixing}} - \overbrace{\frac{\partial}{\partial x_i}\left(\Gamma_T \frac{\partial f_\phi}{\partial x_i}\right)}^{\text{mesomixing}} = -\frac{\partial}{\partial \psi_i}\left[\left(\frac{\varepsilon_\phi}{\langle \phi'^2 \rangle}(\langle \phi_i \rangle - \psi_i) + S_i(\psi)\right) f_\phi\right]$$

Closed by gradient-diffusion model

where

- ϕ is the passive scalar
- $\langle \phi'^2 \rangle$ is the scalar variance
- ε_ϕ is the dissipation factor
- $\langle U_i \rangle$ is the mean velocity
- Γ_T is the turbulent diffusivity
- t is time

Interaction by Exchange with the Mean mixing model (Fox, 2003)

Source term due to reaction

Chemical kinetics

We consider a simple case of two competitive consecutive reactions

A typical test-case to study mixing (Bourne reactions)

$$A + B \xrightarrow{k_1} R$$ Fast reaction

$$B + R \xrightarrow{k_2} S$$ Slower reaction

where

- R is the desired product
- S is the byproduct
- $\frac{k_1}{k_2} = O(10^3)$

Rewrite PDF in terms of

- Mixture fraction ξ
- Reaction progress variables Y_1, Y_2

$$f(\boldsymbol{\phi}, \mathbf{x}, t) \rightarrow f(\xi, Y_1, Y_2, \mathbf{x}, t)$$

$$\xi_{st} = \frac{c_{A_0}}{c_{A_0} + c_{B_0}}$$

$$c_A = c_{A_0}[1 - \xi - (1 - \xi_{\mathrm{st}})Y_1]$$
$$c_B = c_{B_0}[\xi - \xi_{\mathrm{st}}(Y_1 + Y_2)]$$
$$c_R = c_{B_0}\xi_{\mathrm{st}}(Y_1 - Y_2)$$
$$c_S = c_{B_0}\xi_{\mathrm{st}}Y_2$$

Quadrature-based moment methods

The evolution equation for the joint composition PDF of mixture fraction, progress variable one and two $f(\xi, Y_1, Y_2;\ \mathbf{x}, t)$:

$$\frac{\partial f}{\partial t} + \langle U_i \rangle \frac{\partial f}{\partial x_i} - \frac{\partial}{\partial x_i}\left(\Gamma_T \frac{\partial f}{\partial x_i}\right) = -\frac{\partial}{\partial \xi}\left[\left(\frac{\varepsilon_\xi}{\langle \xi'^2 \rangle}(\langle \xi \rangle - \xi) + S(\xi)\right) f\right]$$
$$-\frac{\partial}{\partial Y_1}\left[\left(\frac{\varepsilon_{Y_1}}{\langle Y_1'^2 \rangle}(\langle Y_1 \rangle - Y_1) + S(Y_1)\right) f\right]$$
$$-\frac{\partial}{\partial Y_2}\left[\left(\frac{\varepsilon_{Y_2}}{\langle Y_2'^2 \rangle}(\langle Y_2 \rangle - Y_2) + S(Y_2)\right) f\right]$$

Apply moment definition

$$M_{njk} = \int \xi^n Y_1^j Y_2^k f \mathrm{d}\xi \mathrm{d}Y_1 \mathrm{d}Y_2$$

Finite set of moment transport equations

$$\frac{\partial M_{njk}}{\partial t} + \nabla \cdot \left(\mathbf{U} M_{njk}\right) - \nabla \cdot \left(\Gamma_T \nabla M_{njk}\right) = h$$

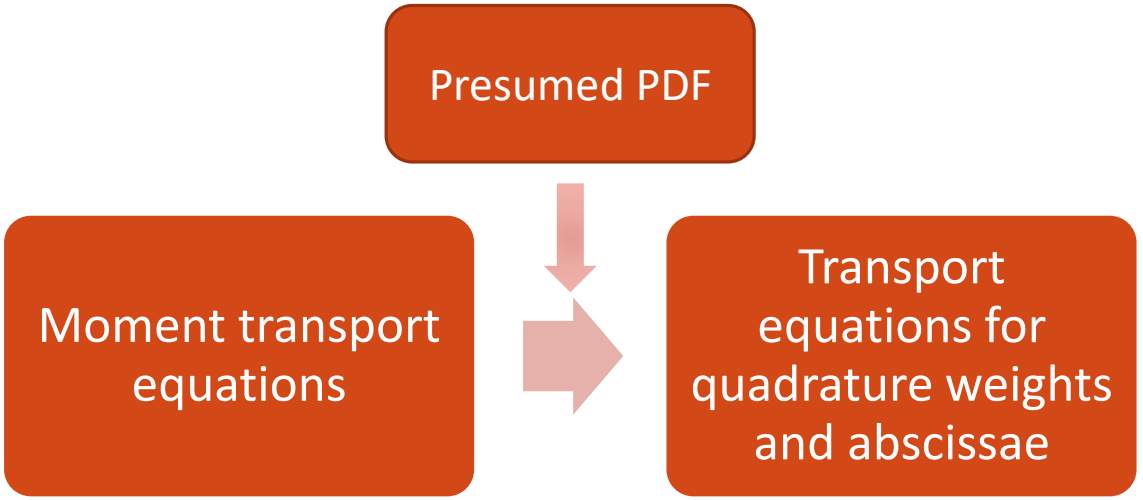
Closure problem

Direct quadrature method of moments - DQMOM

The PDF is presumed to have the form:

$$f(\phi, x, t) = \sum_{\alpha=1}^{N} w_\alpha \delta(\phi - \phi_\alpha)$$

where w_α and ϕ_α are quadrature weights and abscissae obtained from the transported moments by means of a moment inversion algorithm



Challenges with DQMOM

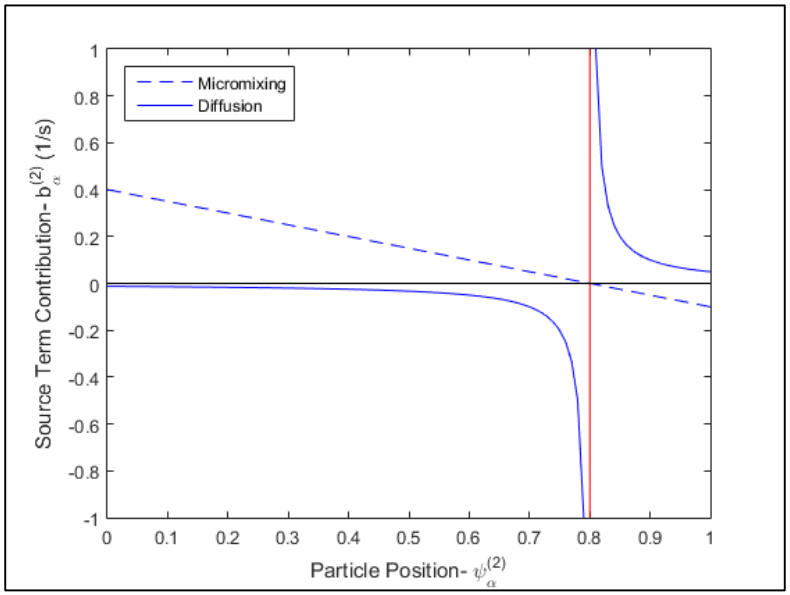
Singularity in the correction terms

- Numerical instability if untreated
- Need of smoothing functions (Akroyd et al., Chem. Eng. Sci, 2010)
- Removal of correction terms (but it leads to incorrect predictions of the variance of the mixture fraction)

Transports non-conservative quantities (weights and abscissae)

CQMOM:

- Avoids singularities
- Transports conserved quantities (moments) rather than non-conservative quantities



J. Akroyd, A. J. Smith, L. R. McGlashan, and M. Kraft, "Numerical investigation of DQMoM-IEM as a turbulent reaction closure," Chemical Engineering Science, vol. 65, pp. 1915-1924, Mar 15 2010.

Conditional quadrature method of moments

We rewrite the joint composition PDF in terms of conditional PDFs:

$$f(\xi, Y_1, Y_2) = f(Y_2|\xi, Y_1) f(\xi, Y_1) = f(Y_2|\xi, Y_1) f(Y_1|\xi) f(\xi)$$

we then consider the conditional moments

$$\left\langle Y_1^j \right\rangle (\xi) \stackrel{\text{def}}{=} \int Y_1^j f(Y_1|\xi) \mathrm{d}Y_1$$

$$\left\langle Y_2^k \right\rangle (\xi, Y_1) \stackrel{\text{def}}{=} \int Y_2^k f(Y_2|\xi, Y_1) \mathrm{d}Y_2$$

and we represent the moments of PDF as

$$M_{njk}(\mathbf{x}, t) = \iiint \xi^n Y_1^j Y_2^k f(\xi, Y_1, Y_2; \mathbf{x}, t) \mathrm{d}\xi \ \mathrm{d}Y_1 \ \mathrm{d}Y_2$$

Conditional quadrature method of moments

We represent the PDF as (assuming one node for Y_1 and Y_2):

$$f(\xi, Y_1, Y_2; \mathbf{x}, t) = \sum_{\alpha=1}^{\mathrm{N}} w_\alpha \delta(\xi - \xi_\alpha)\delta\left(Y_1 - Y_{1_\alpha}\right)\delta\left(Y_2 - Y_{2_\alpha}\right)$$

and we find the moments of PDF as:

$$M_{njk}(\mathbf{x}, t) = \sum_{\alpha=1}^{\mathrm{N}} w_\alpha \xi_\alpha^n Y_{1_\alpha}^j Y_{2_\alpha}^k$$

We consider:

- Two quadrature nodes for the ξ direction
- One quadrature node for the Y_1 direction
- One quadrature node for the Y_2 direction

$M_{000}, M_{100}, M_{200}, M_{300}$

M_{010}, M_{110}

M_{001}, M_{011}

Moment transport equations in CQMOM

$$\frac{\partial M_{njk}}{\partial t} + \nabla \cdot \left(\mathbf{U} M_{njk}\right) - \nabla \cdot \left(\Gamma_T \nabla M_{njk}\right)$$

$$= \frac{n\varepsilon_\xi}{\langle \xi'^2 \rangle}\left(M_{n-1jk} M_{100} - M_{njk}\right)$$

$$+ \sum_{\alpha=1}^{N} w_\alpha R_{1\alpha}\left(j \xi_\alpha^n Y_{1_\alpha}^{j-1} Y_{2_\alpha}^{k}\right) + \sum_{\alpha=1}^{N} w_\alpha R_{2\alpha}\left(k \xi_\alpha^n Y_{1_\alpha}^{j} Y_{2_\alpha}^{k-1}\right)$$

Scale similarity

$$\frac{\varepsilon_\xi}{\langle \xi'^2 \rangle} = C_\xi \frac{\varepsilon}{k}$$

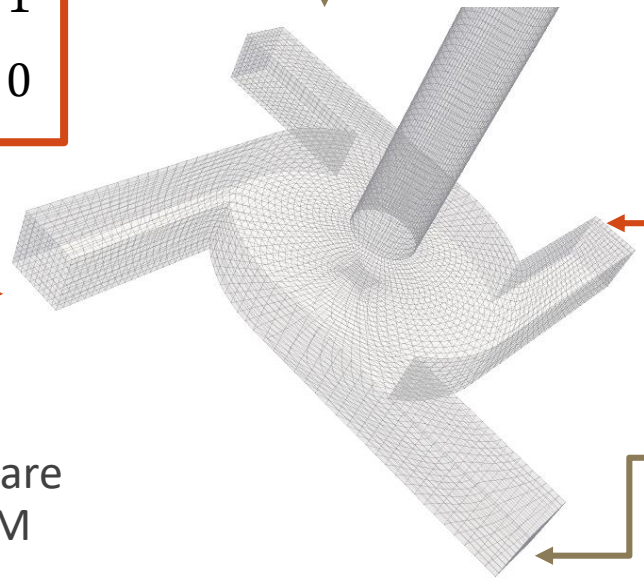
$$R_{1\alpha}(\xi_\alpha, Y_{1\alpha}, Y_{2\alpha}) = \xi_{st} k_1 c_{B_0}\left(\frac{1-\xi_\alpha}{1-\xi_{st}} - Y_{1\alpha}\right)\left(\frac{\xi_\alpha}{\xi_{st}} - Y_{1\alpha} - Y_{2\alpha}\right)$$

$$R_{2\alpha}(\xi_\alpha, Y_{1\alpha}, Y_{2\alpha}) = \xi_{st} k_2 c_{B_0}(Y_{1\alpha} - Y_{2\alpha})\left(\frac{\xi_\alpha}{\xi_{st}} - Y_{1\alpha} - Y_{2\alpha}\right)$$

Mixing - Simulation setup

Two opposing inlet streams for solvent

$$M_{000} = 1 \quad M_{100} = 1$$
$$M_{200} = 1 \quad M_{300} = 1$$
$$M_{010} = 0 \quad M_{110} = 1$$
$$M_{001} = 0 \quad M_{101} = 0$$



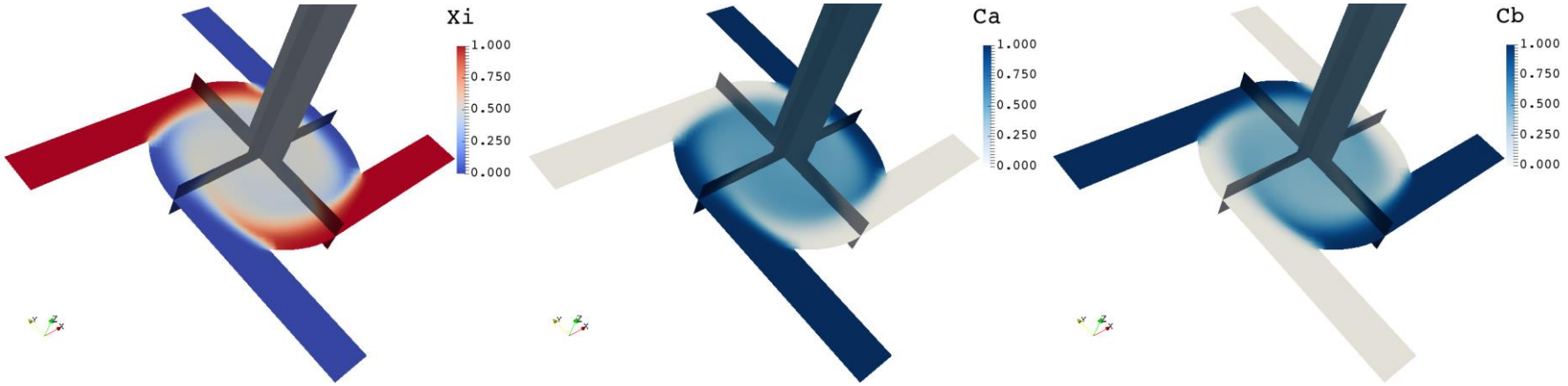
Two opposing inlet streams for nonsolvent

$$M_{000} = 1 \quad M_{100} = 0$$
$$M_{200} = 0 \quad M_{300} = 0$$
$$M_{010} = 0 \quad M_{110} = 1$$
$$M_{001} = 0 \quad M_{101} = 0$$

Mixing and reaction fields are obtained using OpenQBMM (www.openqbmm.org)

Pure mixing (no reaction)

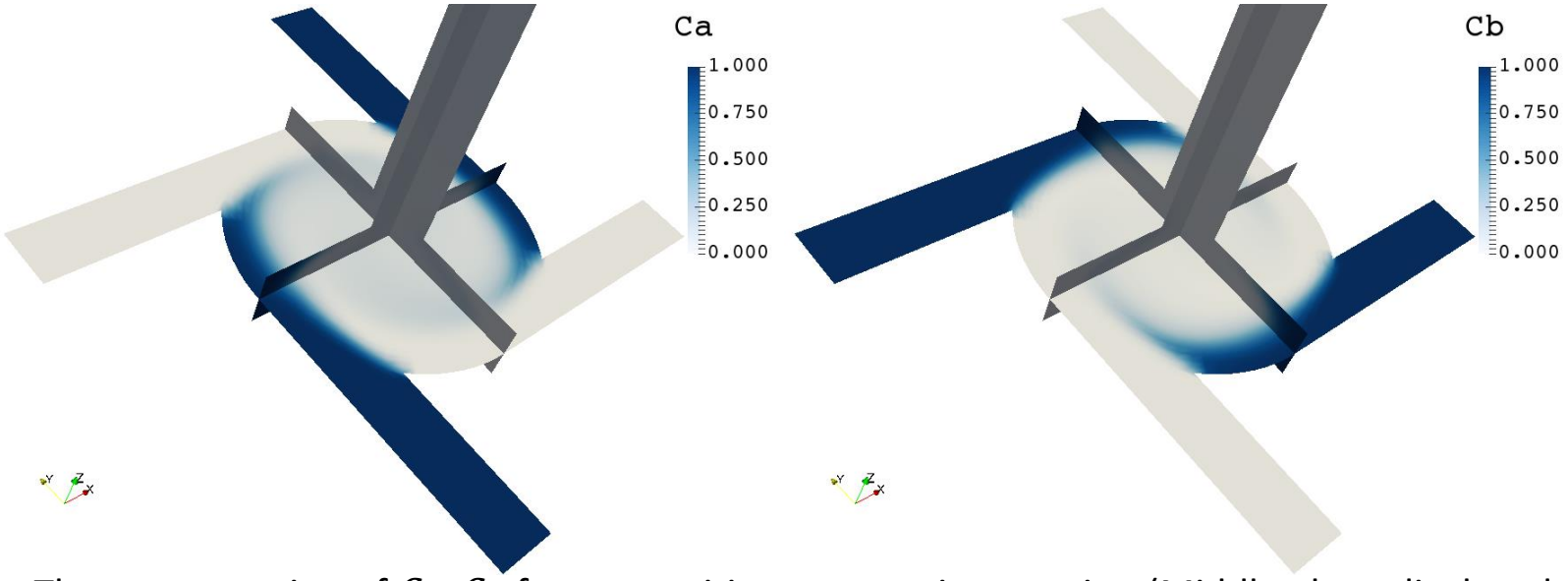
- Mixture fraction represents the mixing progress.
- Two streams have $\langle \xi \rangle = 0$ and two other have $\langle \xi \rangle = 1$.
- Through micromixing, mixture fraction relaxes to $\langle \xi \rangle = 0.5$.
- In FNP processes $\langle \xi \rangle$ is key, since the solvent and non-solvent should reach a specific ratio to precipitate.



The mixture fraction (left), concentration A (middle), and concentration B (right).

Mixing with competitive consecutive reaction

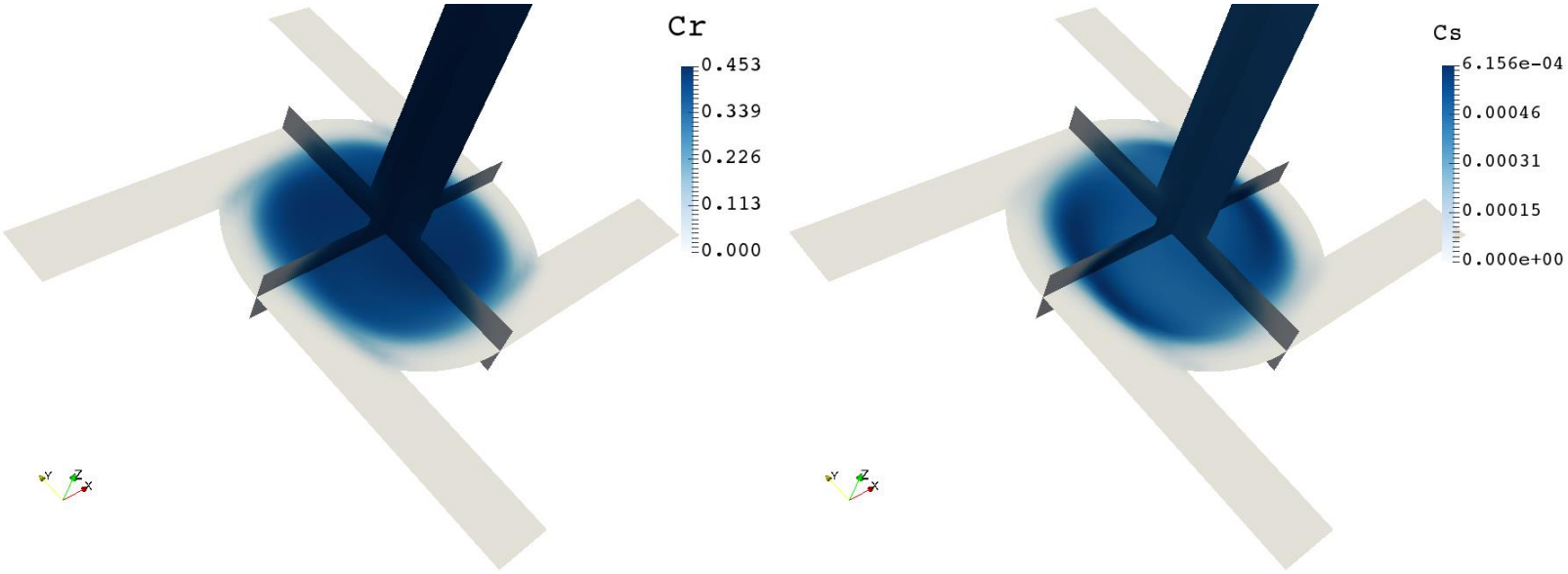
- Simulations show that mostly inside the mixing chamber, species B is consumed.
- The main reaction happens between A and B since the rate of their reaction is $O(1000)$ higher than the secondary reaction.



The concentration of C_A, C_B for competitive consecutive reaction (Middle plane displayed for the chamber).

Mixing with competitive consecutive reaction

- The main product R is mostly produced inside the chamber and moves toward the exit.
- The byproduct S is produced due to the presence of R and B, when A is absent (mixing limitation).



Concentration of product and bi-product for competitive consecutive reaction (Middle plane displayed for the chamber).

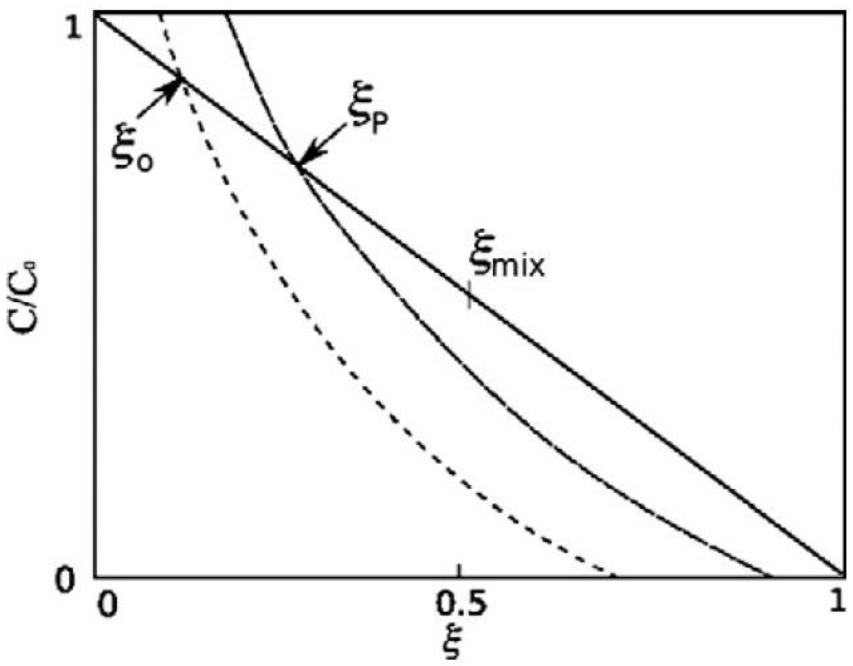
Flash nanoprecipitation solubility curve

The straight line indicates the mixing line between solvent and anti-solvent.

At ξ_{mix}, the two streams are well-mixed.

On the solubility curve, the aggregation kernel "activates" above ξ_p (solubility limit of polymer).

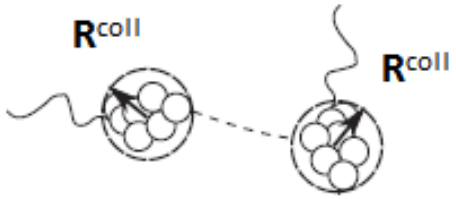
This behavior is described by a Heaviside function (i.e. $\Theta_p = \Theta(\xi-\xi_p)$).



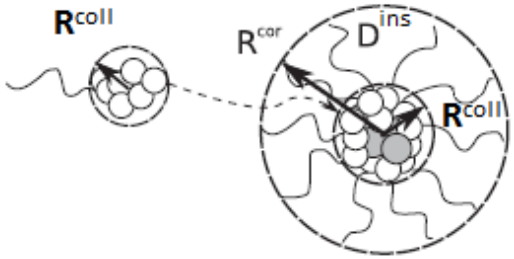
J. C. Cheng et al. "A competitive aggregation model for Flash NanoPrecipitation." *Journal of Colloid and Interface Science* ,vol. 351, pp. 330-342, Nov 15 2010.

Aggregation mechanisms

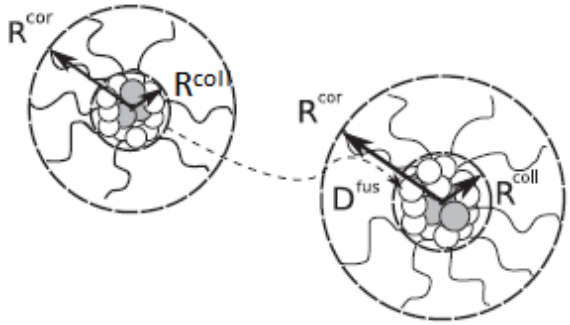
1) Free coupling



2) Unimer insertion



3) Large aggregate fusion



J. C. Cheng et al. “A competitive aggregation model for Flash NanoPrecipitation.” *Journal of Colloid and Interface Science* ,vol. 351, pp. 330-342, Nov 15 2010.

Aggregation kernels

Free coupling

$$\beta_{1,1}^{\text{free}} = 4\pi\Theta_p(D_1 + D_1)\left(R_1^{\text{coll}} + R_1^{\text{coll}}\right)$$

Unimer insertion

$$\beta_{p,1}^{\text{ins}} = 4\pi\Theta_p A_{1,p}^{\text{ins}} \frac{\left(R_p^{\text{coll}} + R_1^{\text{coll}}\right)\left(R_p^{\text{coll}} + R_p^{\text{cor}} + R_1^{\text{coll}}\right)\left(D_p + D_1\right)D_{p,1}^{\text{ins}}}{\left(D_p + D_1\right)R_p^{\text{cor}} + \left(R_p^{\text{coll}} + R_1^{\text{coll}}\right)D_{p,1}^{\text{ins}}}$$

Large aggregate fusion

$$\beta_{p,1}^{\text{fus}} = 4\pi\Theta_p A_{1,p}^{\text{fus}} \frac{\left(R_p^{\text{coll}} + R_i^{\text{coll}}\right)\left(R_p^{\text{coll}} + R_p^{\text{cor}} + R_i^{\text{coll}} + R_i^{\text{cor}}\right)\left(D_p + D_i\right)D_{p,i}^{fus}}{\left(D_p + D_1\right)\left(R_p^{\text{cor}} + R_i^{\text{cor}}\right) + \left(R_p^{\text{coll}} + R_1^{\text{coll}}\right)D_{p,i}^{\text{fus}}}$$

PBE for flash-nanoprecipitation in the MIVR

The evolution equation for the population of polymers:

$$\frac{\partial f}{\partial t} + \langle \mathbf{U} \rangle \frac{\partial f}{\partial \boldsymbol{x}} - \frac{\partial}{\partial \boldsymbol{x}} \left(\Gamma_t \frac{\partial f}{\partial \boldsymbol{x}} \right) = -\frac{\partial}{\partial \xi} \left[\frac{\varepsilon_\xi}{\langle \xi'^2 \rangle} (\langle \xi \rangle - \xi) f \right] - \frac{\partial}{\partial \underline{m}} \left[\frac{\varepsilon_\xi}{\langle \xi'^2 \rangle} (\langle \underline{m} \rangle - \underline{m}) f \right] - \frac{\partial}{\partial \underline{m}} \left[\underline{R}(\xi, \underline{m}) f \right]$$

where:

- $f(\xi, L; \mathbf{x}, t)$: joint PDF of mixture fraction and size
- $\underline{R}(\xi, \underline{m}) = \frac{d\underline{m}}{dt}$: change of moments in PBE given ξ
- $\underline{m}$: moment vector for PBE
- $\langle \mathbf{U} \rangle$: mean velocity
- Γ: diffusion coefficient

Apply moment definition

Finite set of moment transport equations

PBE for flash-nanoprecipitation in the MIVR

$$\underline{R}(\xi,\underline{m}) = \underbrace{\frac{L^2}{2}\int_0^L \frac{\beta\left((L^3-\lambda^3)^{1/3},\lambda\right)}{(L^3-\lambda^3)^{2/3}} f\left((L^3-\lambda^3)^{1/3};\mathbf{x},t\right) f(\lambda;\mathbf{x},t)\,d\lambda}_{\text{birth of particles due to aggregation}}$$

$$\underbrace{- f(L;\mathbf{x},t)\int_0^\infty \beta(L,\lambda) f(\lambda;\mathbf{x},t)\,d\lambda}_{\text{death of particles due to aggregation}}$$

Apply moment definition for PBE source terms

Aggregation kernel

$$\langle \underline{R}(\xi,\underline{m})\rangle = \frac{1}{2}\sum_{i=1}^{N} w_i \sum_{j=1}^{N} w_j (L_i^3 + L_j^3)^{k/3}\beta\left(L_i,L_j\right) - \sum_{i=1}^{N} w_i L_i^k \sum_{j=1}^{N} w_j \beta\left(L_i,L_j\right)$$

PBE for flash-nanoprecipitation in the MIVR

Moments of mixture fraction

$$\frac{\partial\langle\xi^k\rangle}{\partial t}+\nabla\cdot\left(\mathbf{U}\langle\xi^k\rangle\right)-\nabla\cdot\left(\Gamma_T\nabla\langle\xi^k\rangle\right)=\frac{k\varepsilon_\xi}{\langle\xi'^2\rangle}\left(\langle\xi^{k-1}\rangle\langle\xi\rangle-\langle\xi^k\rangle\right)$$

Scale similarity

$$\frac{\varepsilon_\xi}{\langle\xi'^2\rangle}=C_\xi\frac{\varepsilon}{k}$$

Conditional moments of PBE

$$\frac{\partial\langle\underline{m}\rangle}{\partial t}+\nabla\cdot\left(\mathbf{U}\langle\underline{m}\rangle\right)-\nabla\cdot\left(\Gamma_T\nabla\langle\underline{m}\rangle\right)=\langle\underline{R}(\xi,\underline{m})\rangle$$

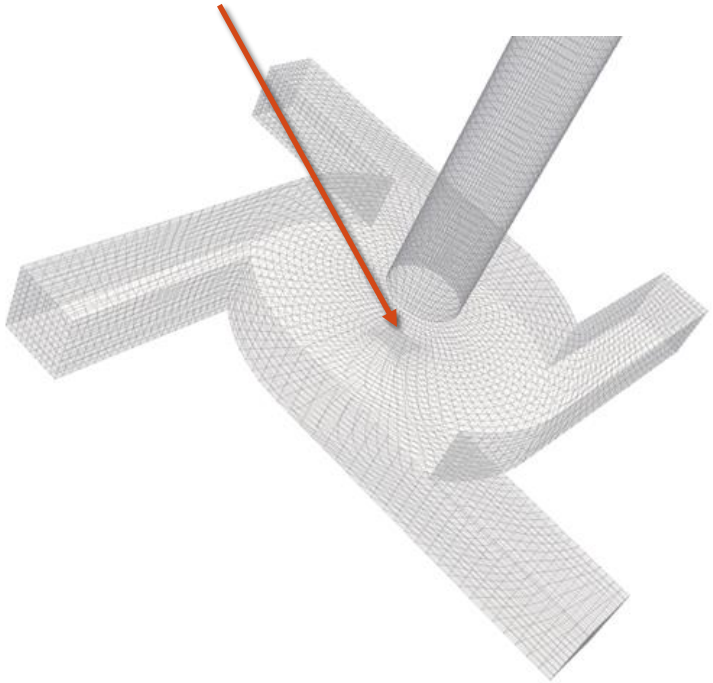
$$\frac{\partial\langle\xi\underline{m}\rangle}{\partial t}+\nabla\cdot\left(\mathbf{U}\langle\xi\underline{m}\rangle\right)-\nabla\cdot\left(\Gamma_T\nabla\langle\xi\underline{m}\rangle\right)=\frac{\varepsilon_\xi}{\langle\xi'^2\rangle}\left(\langle\xi\rangle\langle\underline{m}\rangle-\langle\xi\underline{m}\rangle\right)+\langle\xi\underline{R}(\xi,\underline{m})\rangle$$

Source term for conditional PBE moments

$$\begin{cases}\langle\underline{R}(\xi,\underline{m})\rangle=\sum_{\alpha=1}^{2}p_\alpha\underline{R}(\xi_\alpha,\underline{m}_\alpha)\\ \langle\underline{R}(\xi,\underline{m})\rangle=\sum_{\alpha=1}^{2}p_\alpha\xi_\alpha\underline{R}(\xi_\alpha,\underline{m}_\alpha)\end{cases}$$

PBE for FNP in MIVR – Predicted particle size

Particle size measured at the center of the mixing chamber



Mean particle size at the center of the mixing chamber

- Cheng et al.
  - 4.37 of initial polymer length
- Predicted in present work
  - 4.01 of initial polymer length

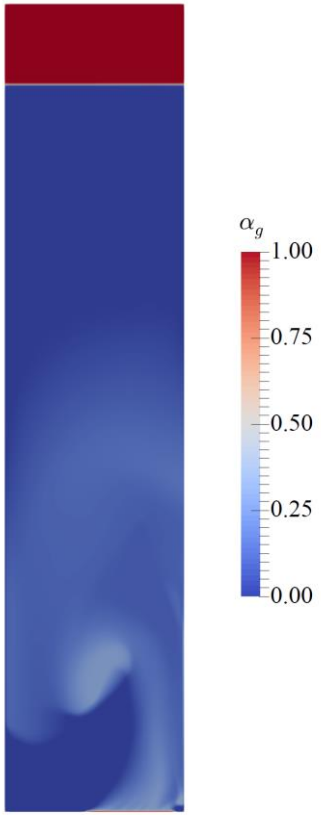
Satisfactory agreement

Gas-liquid flows

Properties

- Typically low Stokes number
- Wide range of volume fractions in the system
- Possible presence of free surface
- Several physical phenomena can affect the population of bubbles
  - Coalescence
  - Breakup
  - Cavitation
  - Boiling
  - Chemical processes

A reliable model should be able to account for these aspects and be numerically robust



Description of the gas phase

The gas phase can be
- Disperse
- Continuous

Disperse gas phase
- Generalized population balance equation
- $\frac{\partial f}{\partial t} + \mathbf{v} \cdot \frac{\partial f}{\partial \mathbf{x}} + \frac{\partial}{\partial \mathbf{v}} \cdot \left[(\mathbf{A} + \mathbf{g}) f - \mathbf{D} \cdot \frac{\partial \ln n}{\partial \mathbf{x}} f \right] = \mathbb{C}(\xi, \mathbf{v})$
- $f(t, \mathbf{x}, \xi, \mathbf{v})$: joint size-velocity NDF
- $n(t, \mathbf{x}, \xi) = \int f(t, \mathbf{x}, \xi, \mathbf{v}) \mathrm{d}\mathbf{v}$
- ξ: bubble mass
- $\mathbf{v}$: bubble velocity

Continuum gas phase
- Navier-Stokes equations

Liquid phase
- Navier-stokes: as in two-fluid approach.

A numerical challenge
- QBMM for the disperse regime
- NS for the continuous regime

How to make them work together?

Description of the gas phase [2]

We assume the NDF is monokinetic:

- One velocity for each bubble size
- $f(\xi, \mathbf{v}) = n(\xi)\delta(\mathbf{v} - \mathbf{U}(\xi))$
- $\mathbf{U}(\xi)$ bubble velocity conditioned on bubble mass

We reconstruct

- $n(\xi)$ from 2N moments

$$M_p = m_{p,0,0,0},\ p = 0, \dots, 2\mathrm{N} - 1 \text{ (Size moments)}$$

- $\mathbf{U}(\xi)$ from the N velocity moments

$$\mathcal{U}_p = \left(m_{p,1,0,0}, m_{p,0,1,0}, m_{p,0,0,1}\right), \qquad p = 0, \dots, \mathrm{N} - 1$$

A different velocity vector is associated to each particle size, to account for polycelerity

Solution process

Find quadrature approximation

Use M_p and $\mathcal{U}_p$ to find $w_\alpha, \xi_\alpha, \mathbf{U}_\alpha$

Advect moments due to "deviation velocity from the mean"

Define $\mathbf{V}_\alpha = \mathbf{U}_\alpha - \mathbf{U}_\mathrm{b}$ ($\mathbf{U}_\mathrm{b}$ = mean gas velocity)

Advect M_p and $\mathcal{U}_p$ based on $\mathbf{V}_\alpha$

Compute new mean volume fraction and velocity

Use a standard two-fluid model solver to account for mean transport

Update moments due to mean transport using limited velocity found at the previous step

Account for the effect of the force term on moments directly integrating velocity abscissae $\left(\frac{\mathrm{d}\mathbf{U}_\alpha}{\mathrm{d}t} = \mathbf{A}\right)$

Test cases

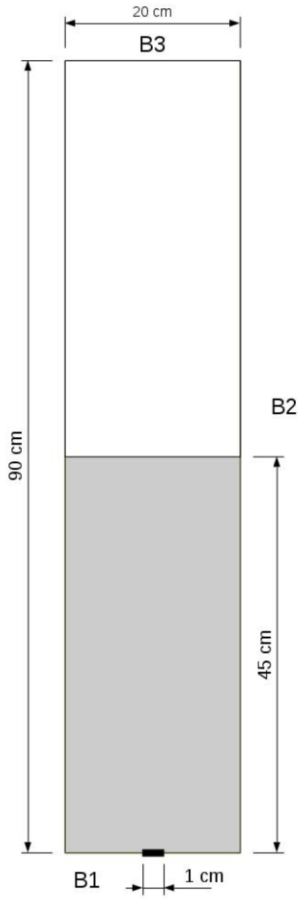
Case setup

- Pfleger et al. (1999)
- Monodisperse flow in a bubble column with central injection
- Bubble size: 2 mm
- Gas flow rate: 48 l/h

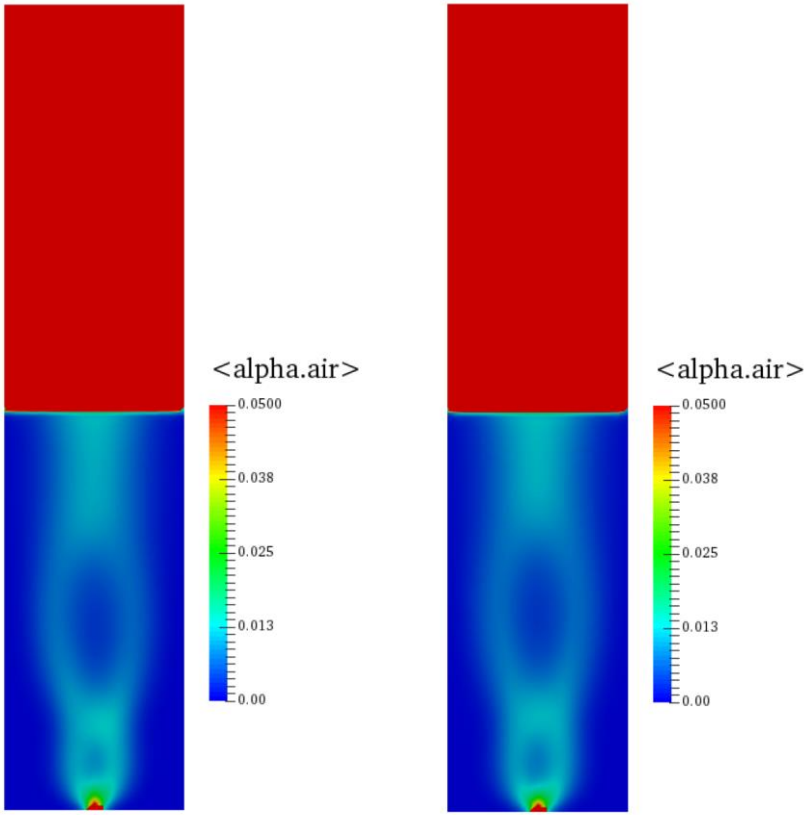
Objective

- Verify the QBMM model correctly degenerates in the two-fluid model in mono-disperse case
- Reference solver: twoPhaseEulerFoam (OpenFOAM-dev, OpenFOAM Foundation release).

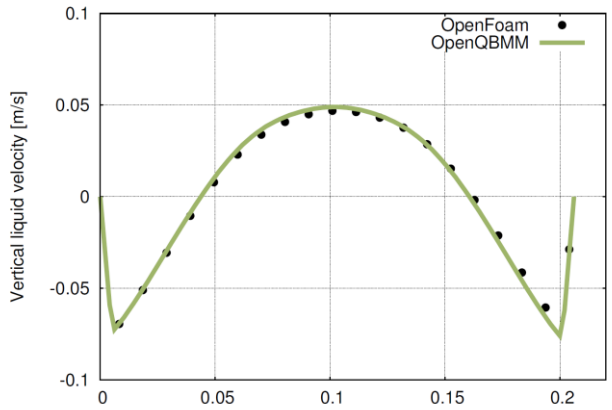
Pfleger, D., Gomez, S., Gilbert, N., Wagner, H.-G. 1999 Chem. Eng. Sci. 54, 5091 – 5099.



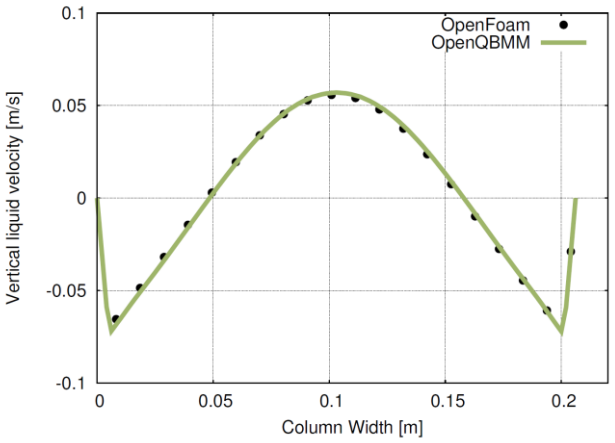
Monodisperse case



OpenQBMM twoPhaseEulerFoam



H = 0.13 m



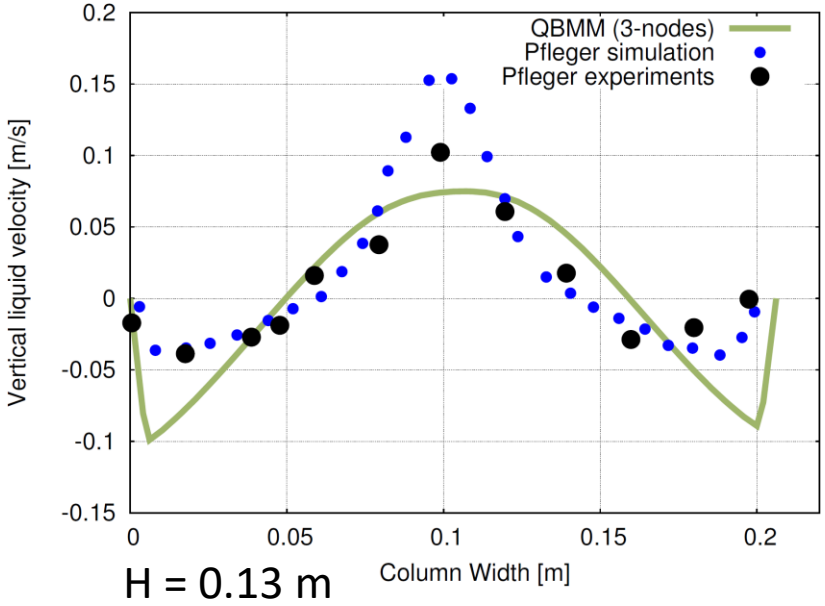
H = 0.37 m

Polydisperse case

Same bubble column as in the monodisperse case

Three bubble sizes:

- 1.0, 2.5, 4.0 mm
- Equal volume fractions and inlet velocities to match the mean bubble diameter in experiments



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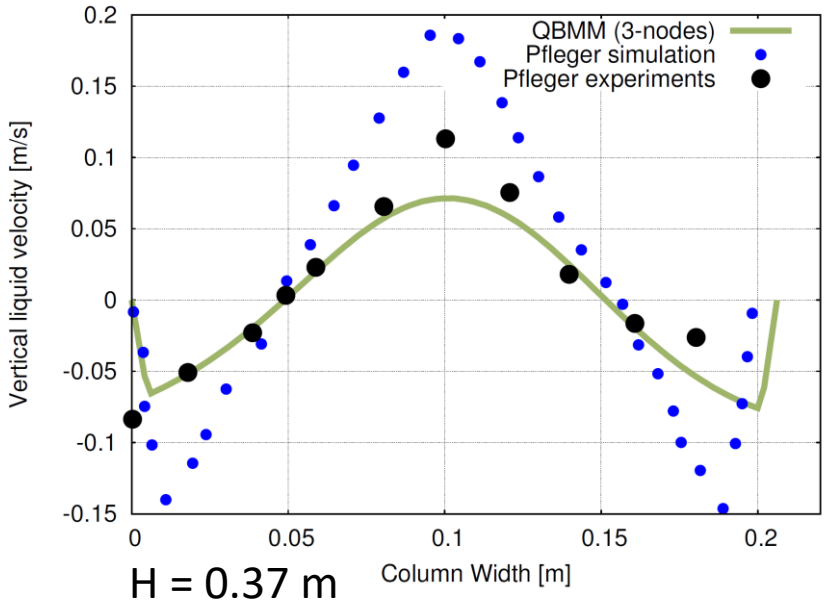

H = 0.37 m

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Thank you!