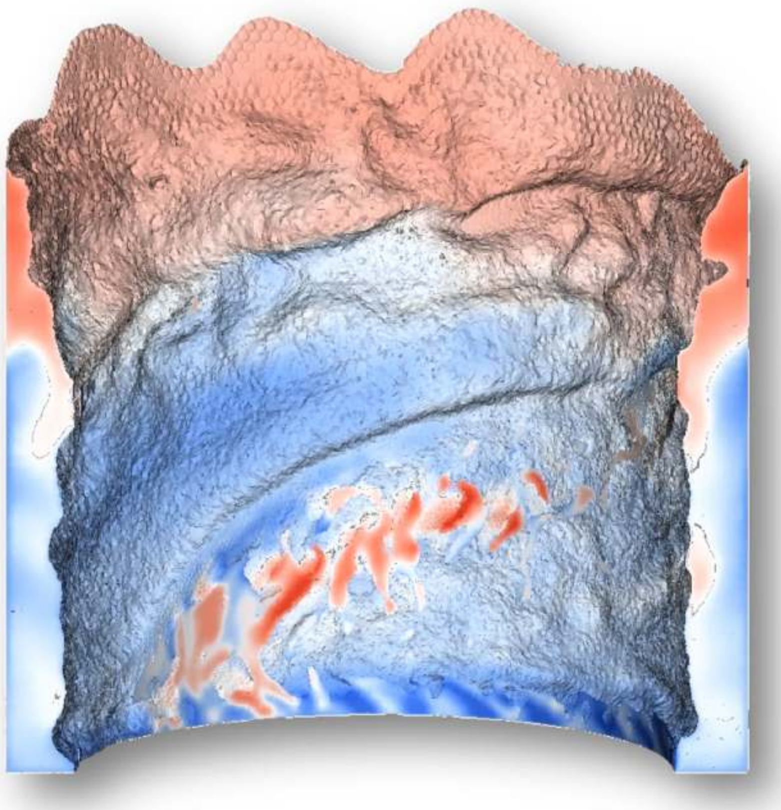
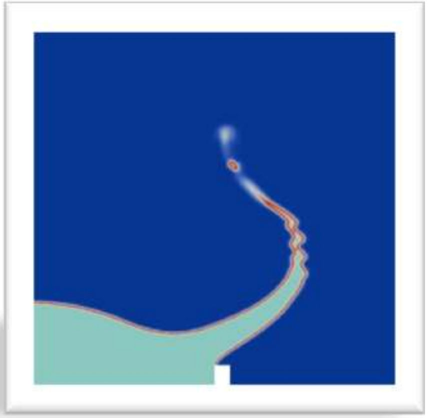
Hybrid Multiphase CFD Solver for Coupled Dispersed/Segregated Flows in Liquid-Liquid Extraction



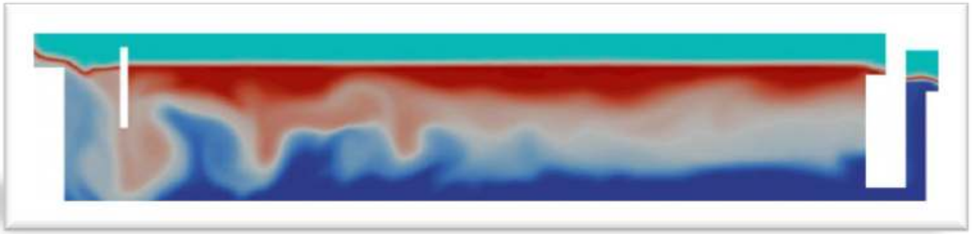
Kent E. Wardle
Chemical Sciences and Engineering Division
Argonne National Laboratory
kwardle@anl.gov

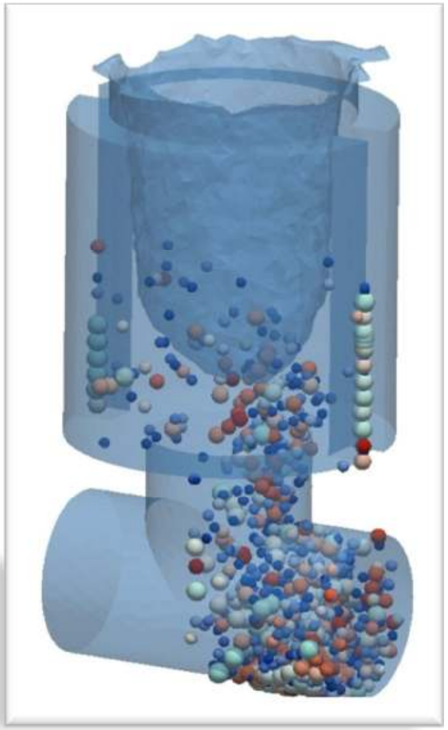
Presented at Mixing XXIII, 19 June 2012

CFD Challenges: Turbulent, Multiphase Flow



- CFD of multiphase systems tends to be regime-dependent
  - Segregated/free surface flow
    - Sharp interface capturing: Volume of Fluid (VOF) →
      - Others: Level-set, mesh-based interface tracking
    - Single mixture momentum equation, valid volume fraction = 0:1
  - Dispersed flow
    - Eulerian-Eulerian multiphase ↓
      - Momentum equation per phase, interphase transport terms
      - Fixed or variable droplet size



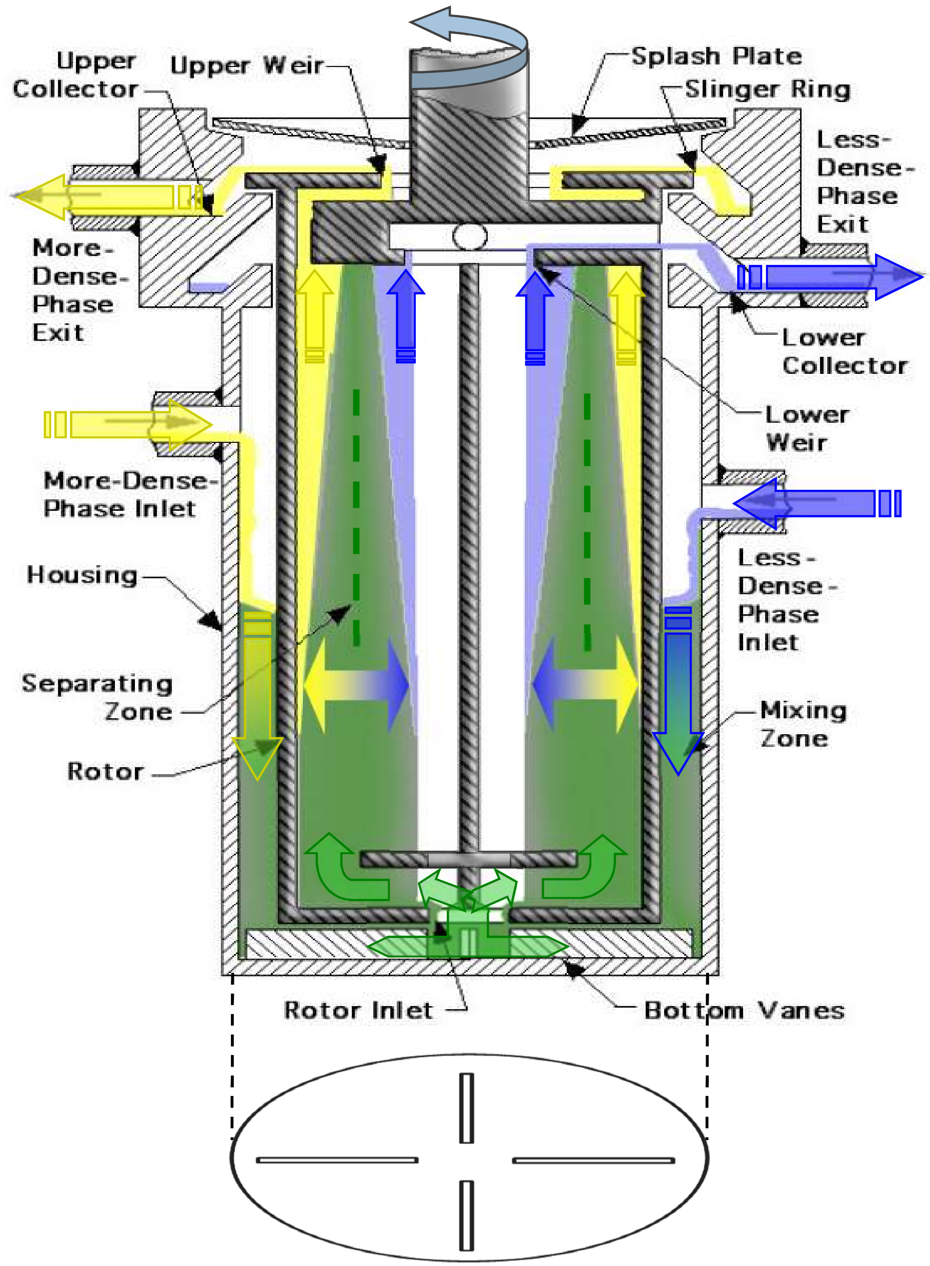


    - Lagrangian particle tracking →
      - Typically limited to low volume fractions (<10%)
- Flows in liquid-liquid extraction span multiple regimes from fully-segregated to fully-dispersed (and then fully-segregated again...)

Annular Centrifugal Contactor

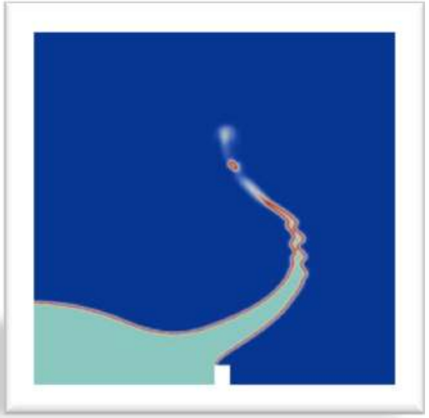
Solvent Extraction Equipment

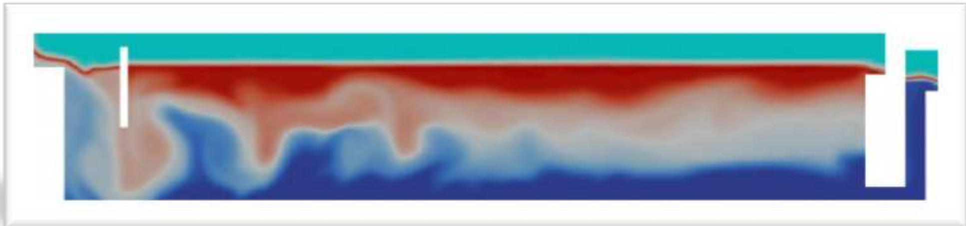
- Packed Columns
- Pulsed Columns
- Mixer Settlers
- **Centrifugal Contactors**
  - Small size
    - Physical footprint
    - Nuclear criticality
  - Short residence time
    - Low process hold-up
    - Less solvent degradation
  - High extraction efficiency
  - High throughput
  - Quick start-up/shut-down
    - Maintain conc. profile



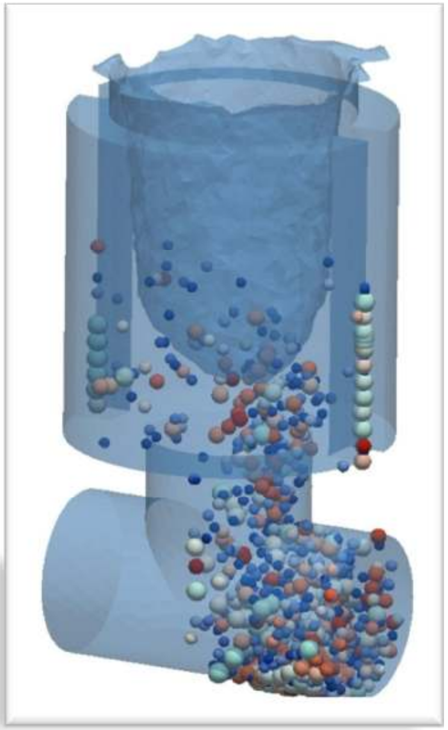
CFD Challenges: Turbulent, Multiphase Flow

- CFD of multiphase systems tends to be regime-dependent
  - Segregated/free surface flow
    - Sharp interface capturing: Volume of Fluid (VOF) →
      - Others: Level-set, mesh-based interface tracking
    - Single mixture momentum equation, valid volume fraction = 0:1
  - Dispersed flow
    - Eulerian-Eulerian multiphase ↓
      - Momentum equation per phase, interphase transport terms
      - Fixed or variable droplet size

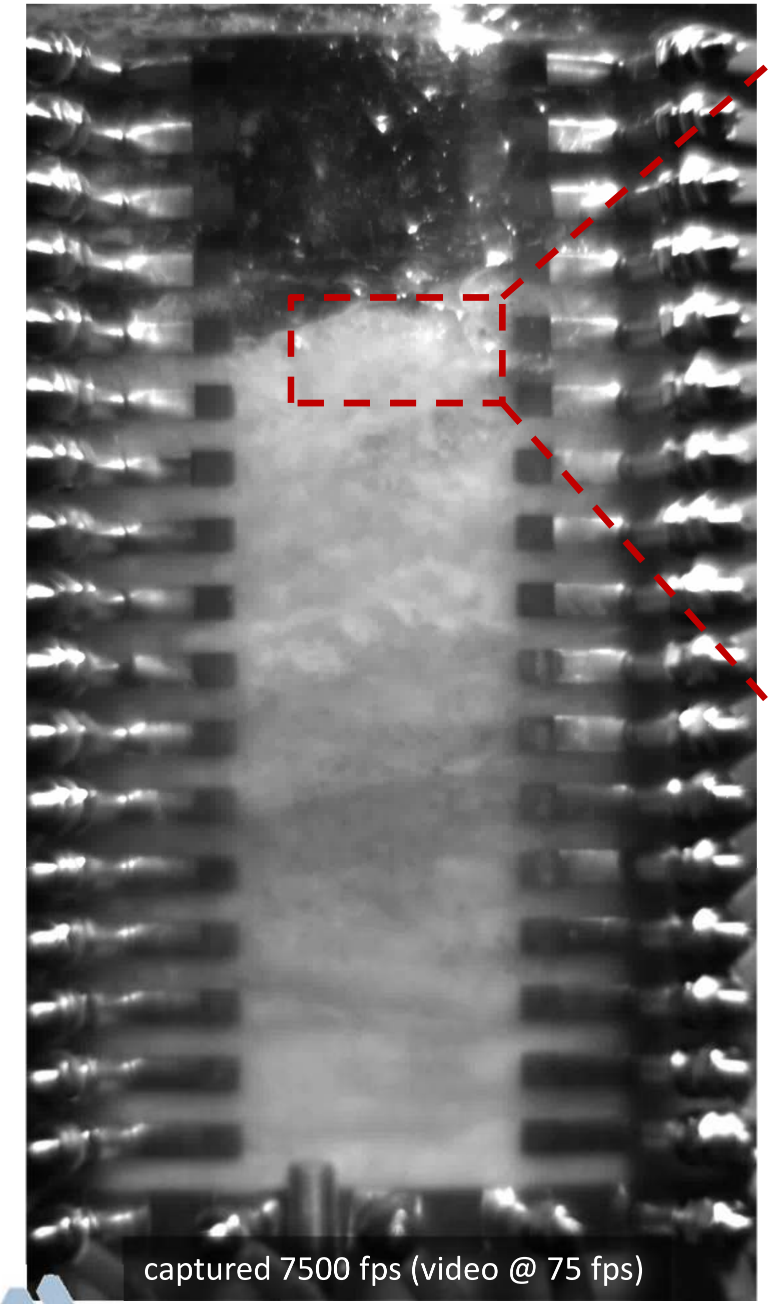


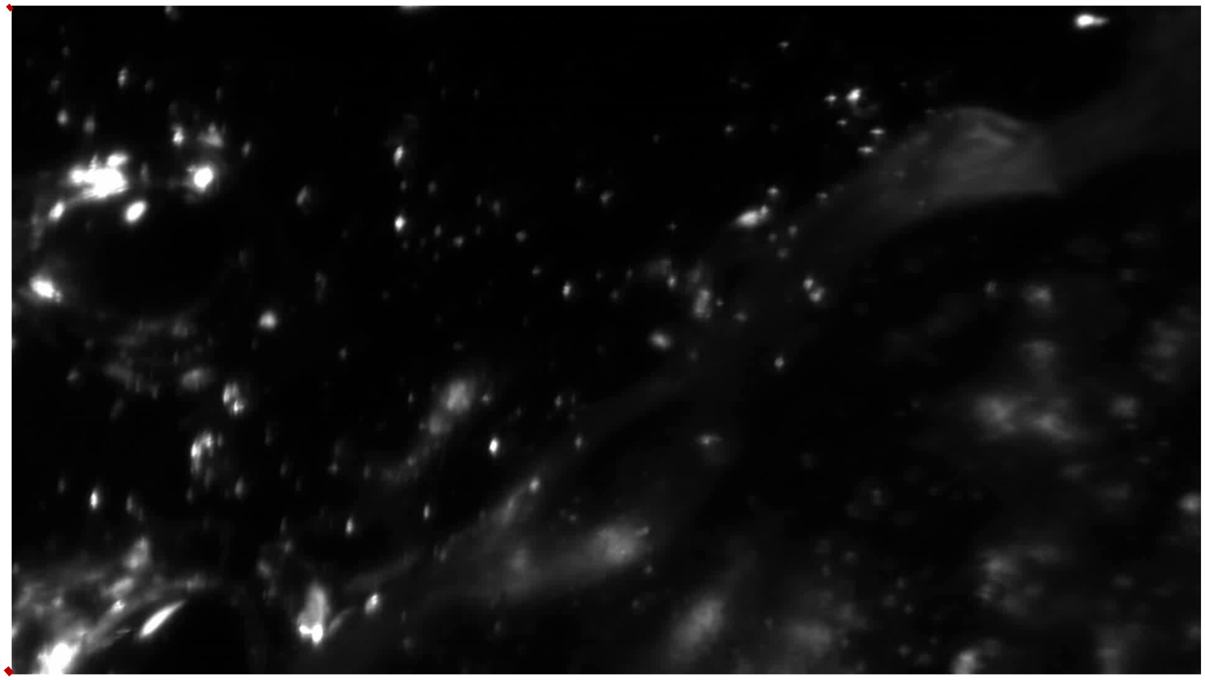


    - Lagrangian particle tracking →
      - Typically limited to volume fractions of <10%
- ***Centrifugal contactor flow spans multiple regimes***
  - Free surface flow in the annular region
  - Liquid-liquid dispersed phase flow throughout
  - Free surface flow in the rotor, weirs, and collector rings



“Let’s see you try and simulate *this*…” [Summer 2004]



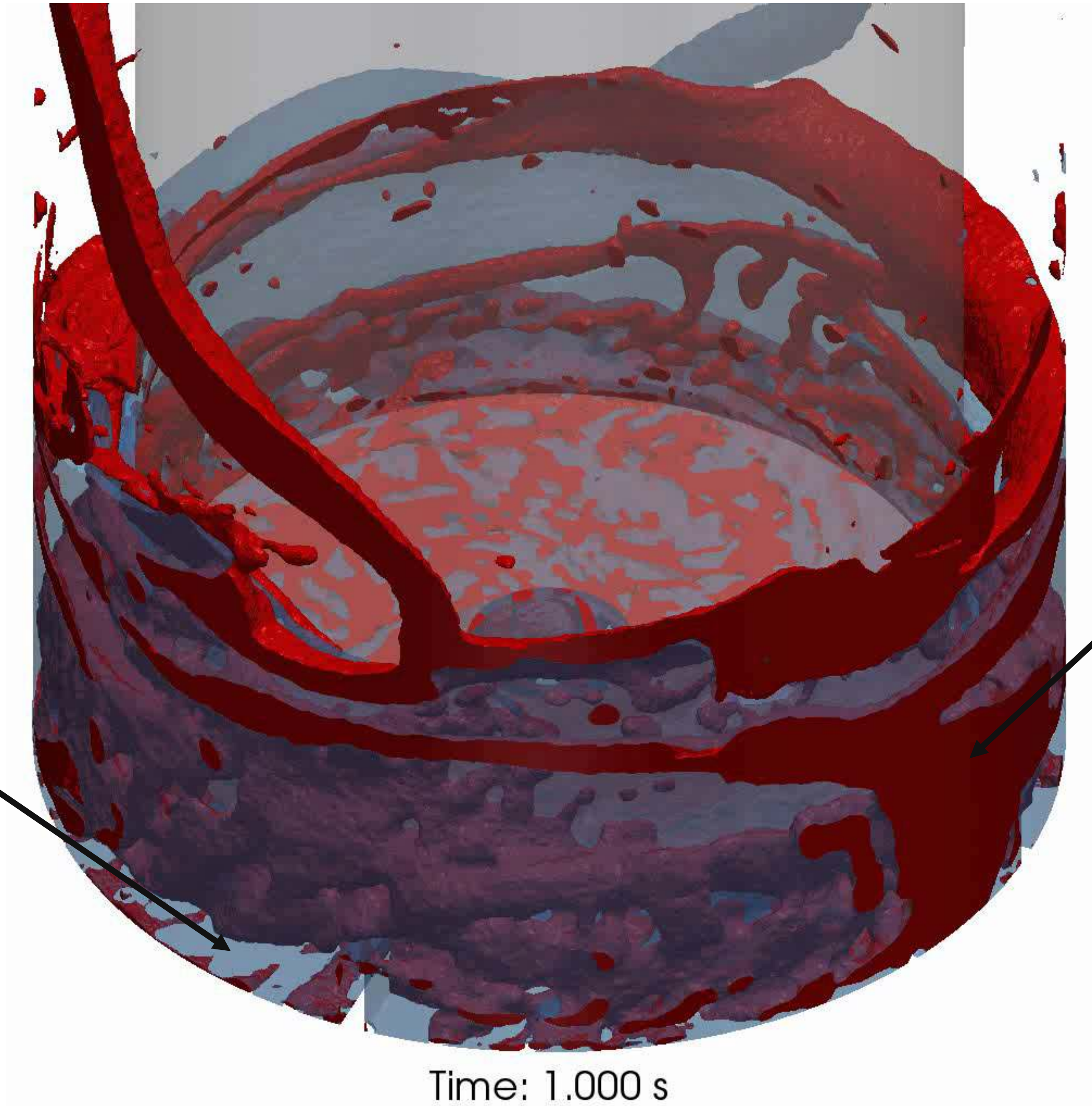


captured 7500 fps (video @ 30 fps)

5-inch rotor (CINC-V5)
curved mixing vane geom (CV)
total flow of 10LPM @ O/A =3
1800 RPM

(videos from MotionPro Y7 at 7500 fps)

Three-phase Water-Oil-Air Annular Mixing Simulation Using VOF-type Solver (multiphaseInterFoam, OF-1.5.x)



water is transparent blue

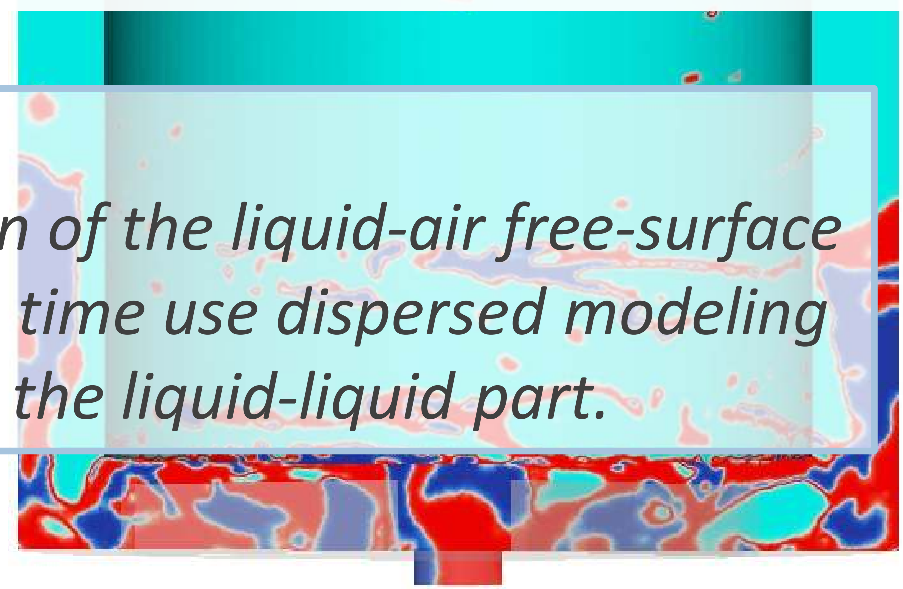
oil is red

Three-phase Water-Oil-Air Annular Mixing Simulation Using VOF-type Solver (`multiphaseInterFoam`, OF-1.5.x)

Only 'large' droplets are resolved (~1mm)

- Actual droplet size, ~25 µm
- ~5 µm mesh (Δx, ~50x smaller)
  - N ~ 1×10^{11} cells
  - Δt ~ 1×10^{-7} s
  - Cr limit, as Δx ↓, Δt ↓

$$Cr = \frac{u\Delta t}{\Delta x} \approx 0.25$$



WHAT WE NEED:

A tool that can capture the motion of the liquid-air free-surface accurately (VOF), but at the same time use dispersed modeling methods (Eulerian multi-fluid) for the liquid-liquid part.

Coupled Multi-fluid—VOF Model Equations

Momentum equations for each phase k:

$$\frac{\partial(\rho_k \alpha_k \vec{u}_k)}{\partial t} + (\rho_k \alpha_k \vec{u}_k \cdot \nabla)\vec{u}_k = -\alpha_k \nabla p + \nabla \cdot (\mu_k \alpha_k \nabla \vec{u}_k) + \rho_k \alpha_k \vec{g} + \vec{F}_{D,k} + \vec{F}_{s,k}$$

drag force ($\vec{F}_{D,k}$)

surface tension force ($\vec{F}_{s,k}$)

Volume fraction transport:

$$\frac{\partial \alpha_k}{\partial t} + \vec{u}_k \cdot \nabla \alpha_k + \nabla \cdot (\vec{u}_c \alpha_k (1 - \alpha_k)) = 0$$

Interface compression velocity [1]:

$$\vec{u}_c = C_\alpha \, |\vec{u}| \frac{\nabla \alpha}{|\nabla \alpha|}$$

$$C_\alpha = \begin{cases} 0, & \text{no interface sharpening} \\ 1, & \text{interface sharpening active} \end{cases}$$

[1] Weller, H.G., A New Approach to VOF-based Interface Capturing Methods for Incompressible and Compressible Flow. Technical Report. OpenCFD (2008).

Coupled Multi-fluid—VOF Model Equations

Momentum equations for each phase k:

$$\frac{\partial(\rho_k \alpha_k \vec{u}_k)}{\partial t} + (\rho_k \alpha_k \vec{u}_k \cdot \nabla)\vec{u}_k = -\alpha_k \nabla p + \nabla \cdot (\mu_k \alpha_k \nabla \vec{u}_k) + \rho_k \alpha_k \vec{g} + \vec{F}_{D,k} + \vec{F}_{s,k}$$

drag force

face

orce

Working solver for arbitrary number of phases released as part of OpenFOAM 2.1.(1) as `multiphaseEulerFoam` along with some tutorial cases. Download at www.openfoam.com

$|\nabla \alpha|$

$$C_\alpha = \begin{cases} \text{0, no interface sharpening} \\ \text{1, interface sharpening active} \end{cases}$$

[1] Weller, H.G., A New Approach to VOF-based Interface Capturing Methods for Incompressible and Compressible Flow. Technical Report. OpenCFD (2008).

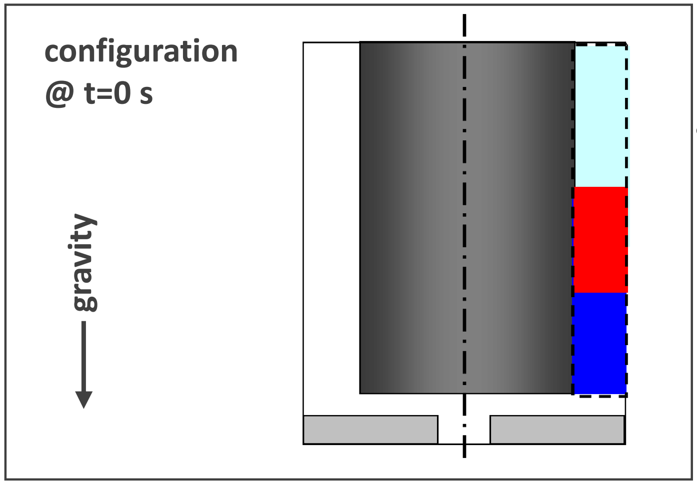
Three-phase (Water-Oil-Air) Examples

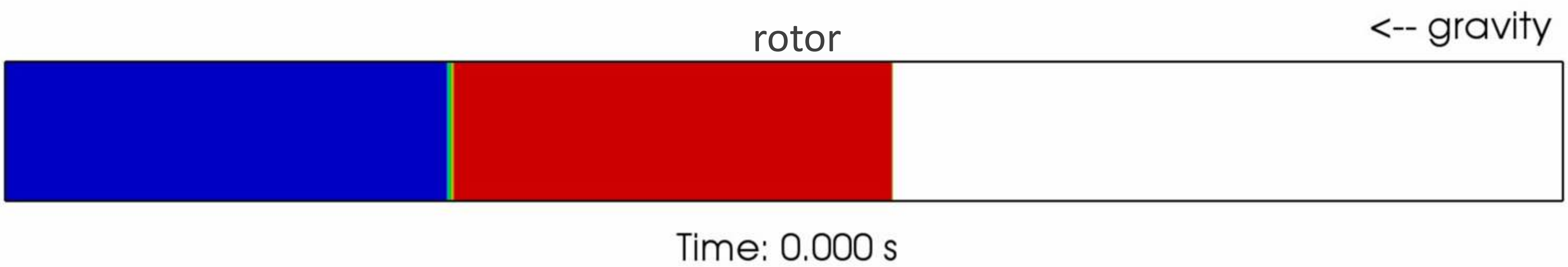
- No cAlpha switching -- interface compression applied for air/water & air/oil interfaces only
- Water/oil interface treated with multi-fluid model w/ interphase drag model of Schiller-Naumann
- Fixed droplet sizes [2]
  - $d_{water} = d_{oil} = 0.150$ mm
  - $d_{air} = 1$ mm

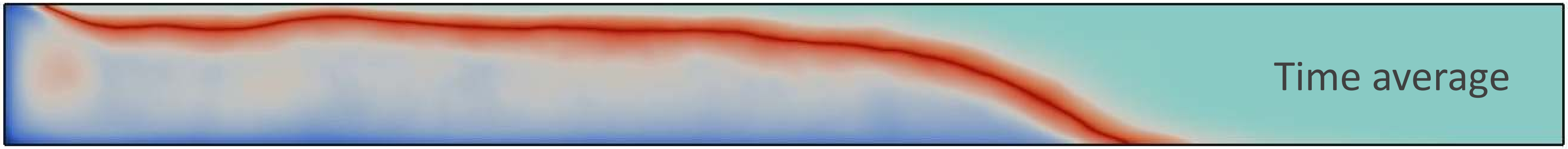
[2] Padial-Collins et al. *Sep. Sci. Technol.* **41**, 1001–1023 (2006).

2D Annular Mixer (axisymmetric)

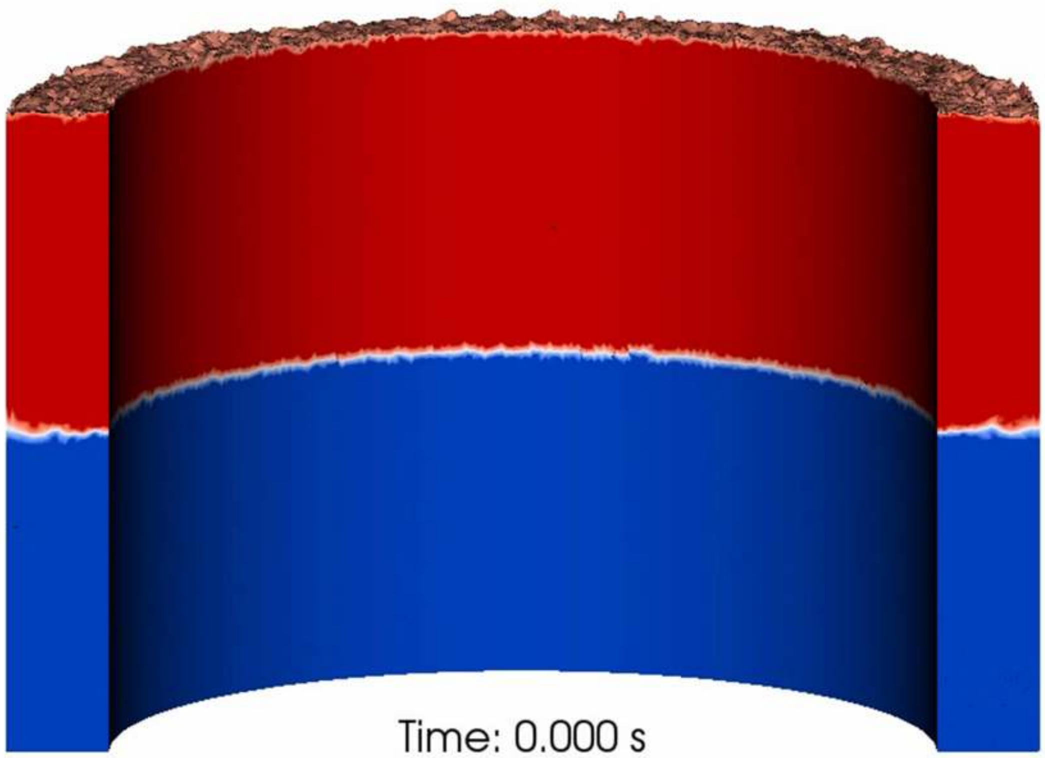
- CINC-V2 dimensions
  - 5.04 cm (2”) rotor
  - 0.63 cm (0.25”) gap
- 3600 RPM
- Three phase water-oil-air





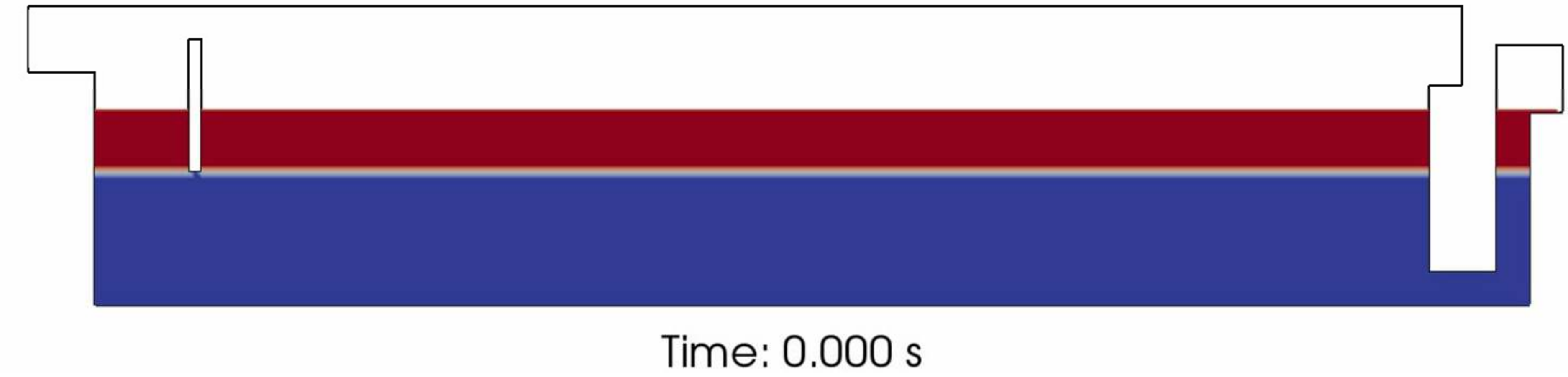


3D Annular Mixer

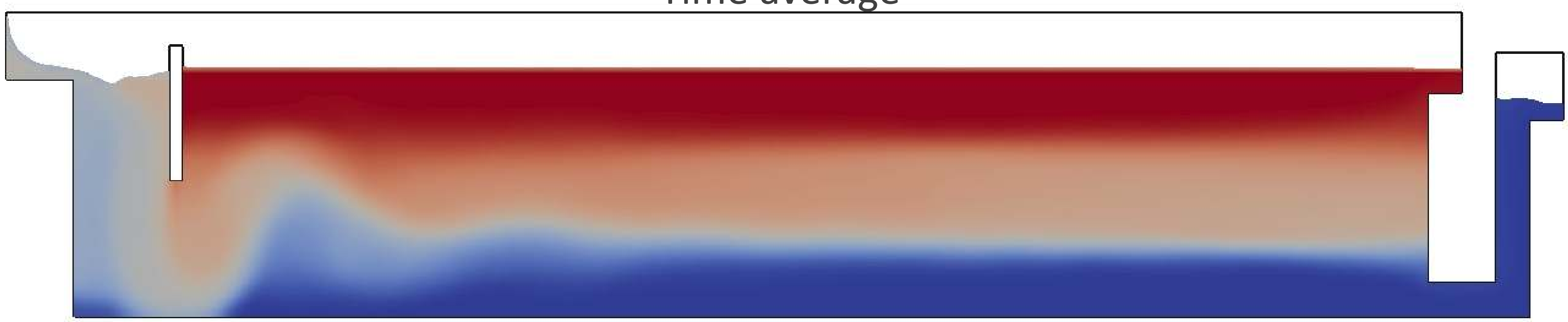


Three-phase 2D Horizontal Settler (Water-Oil-Air)

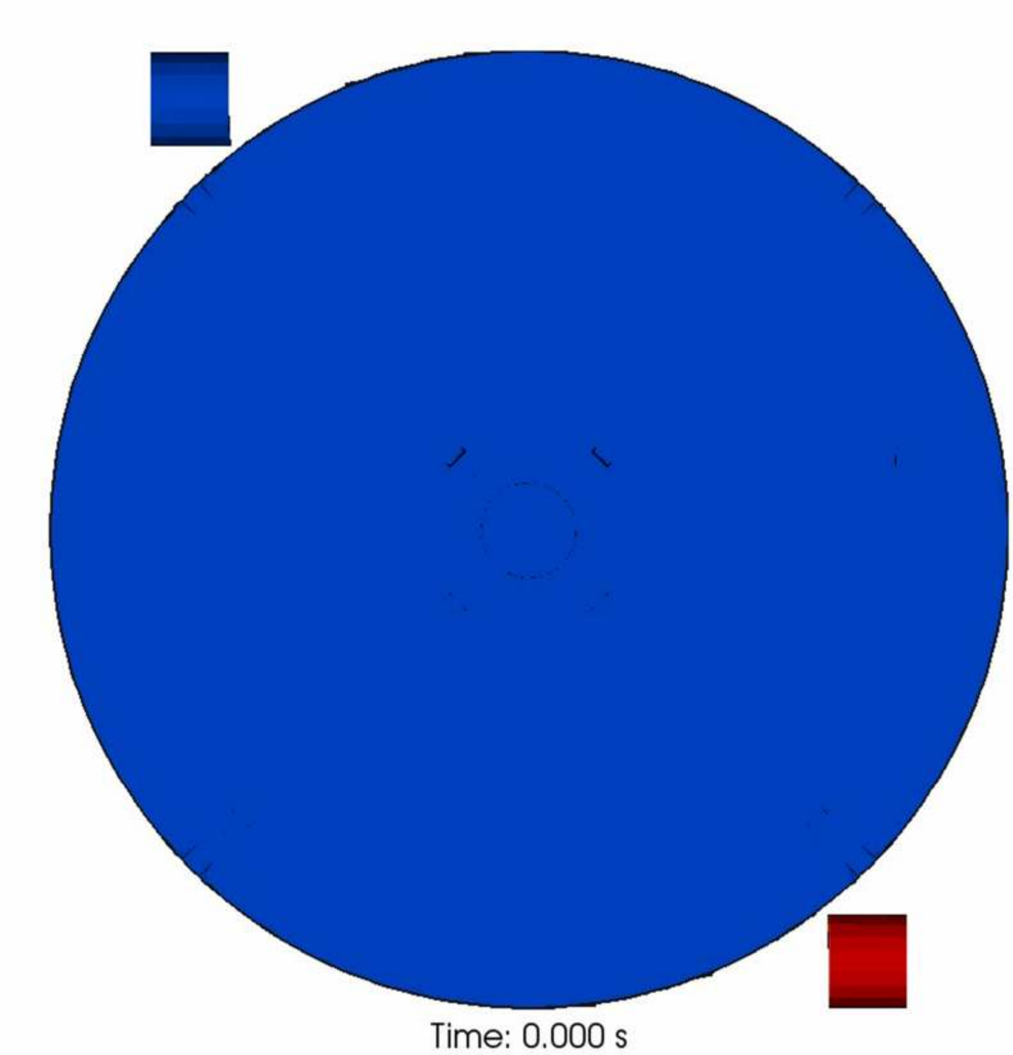
- Inlet flow 50:50 homogenous dispersion (O/A = 1)
- gravity = 2g

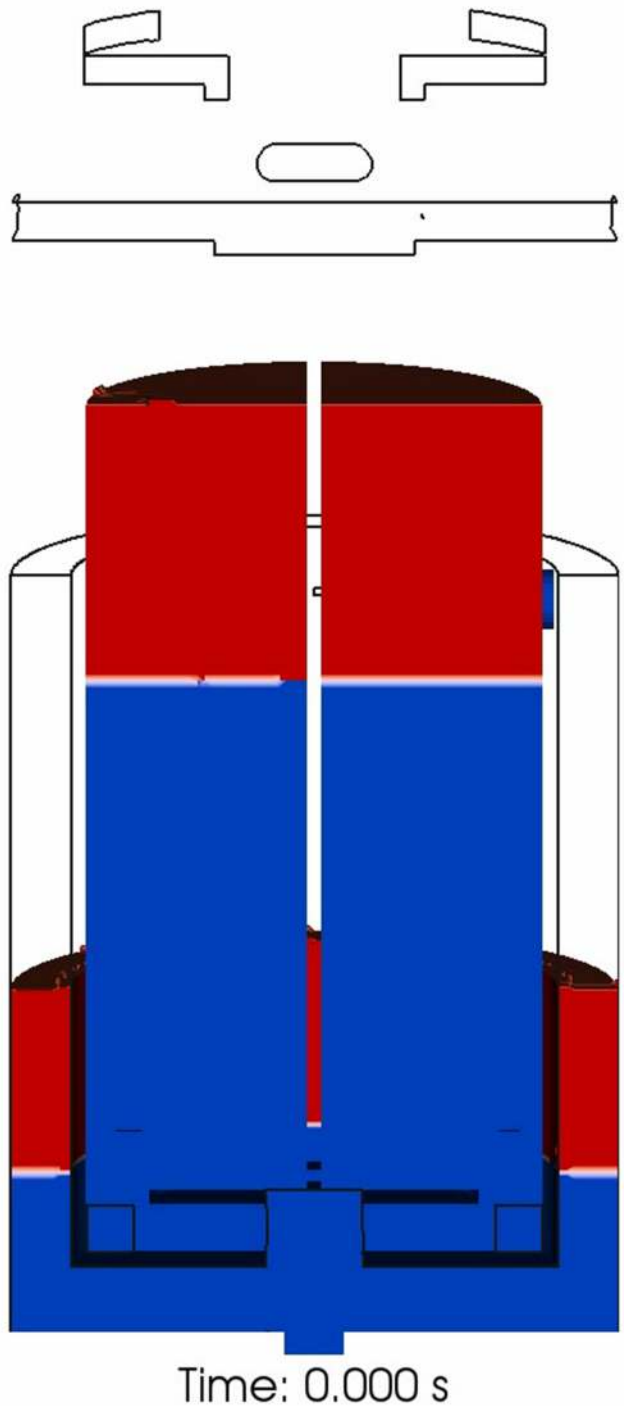


Time average



Coupled Mixing/Separation Zone
Three-phase (water-oil-air)





Variable Droplet Size and Interfacial Area Estimation (preliminary)

~ PROFESSOR KRESTA'S ASSIGNMENT ~

Hinze, J. O., Fundamentals of the hydrodynamic mechanism of splitting in dispersion process, *AIChE J.* 1, 289-295 (1955).

Davies, J. T., A physical interpretation of drop sizes in homogenizers and agitated tanks, including the dispersion of viscous oils, *Chem. Eng. Sci.,* **42**, 1671-1676 (1987).
Davies, J. T., Drop sizes of emulsions related to turbulence energy dissipation rates, *Chem. Eng. Sci.,* **40**, 839-842 (1985).

Zhou, G. and **Kresta, S.M.**, Correlation of mean drop size and minimum drop size with the turbulence energy dissipation and the flow in an agitated tank. *Chem. Eng. Sci.*, **53**,2063 (1998).

Interfacial Area Prediction

- Critical quantity for mass transfer between phases, evaluation of mixing effectiveness
  - Separation of phases also depends on transport of smallest droplets
- Variety of available methods
  - Population Balance Equation
    - Classes Method
    - QMOM & variants
    - One-Group [3]
  - Interfacial Area Transport (IAT)
  - Lagrangian methods
- All require models/correlations for droplet (bubble) breakup and coalescence → *here lies the uncertainty*
  - Function of turbulence dissipation rate, ε
  - Some theoretical basis (balance of forces, frequency of collisions, etc.) but usually some number of adjustable parameters

[3] Drumm et al. Ind. Eng. Chem. Res. **49**, 3442-3451 (2010).

Particle Number Density

- Transport equation for the particle number density (N_d) based on d_{30}

$$\frac{\partial(\rho_d N_d)}{\partial t} + \nabla \cdot [\rho_d \vec{u}_d(d_{30}) N_d] = \rho_d S$$

$$S = [n_d - 1] g(d_{30}) N_d - \frac{1}{2} a(d_{30}, d_{30}) N_d^2$$

$$d_{30} = \sqrt[3]{\frac{6\alpha_d}{\pi N_d}}$$

where g and a are the breakage and coalescence kernels

- Breakage Rate
  (from Martinez-Bazan, 1999)

$$g(d) = \frac{\kappa \sqrt{\beta_0 (\varepsilon d)^{2/3} - 12(\sigma/\rho d)}}{d}$$

$$\kappa = 0.25$$

$$\beta_0 = 8.2$$

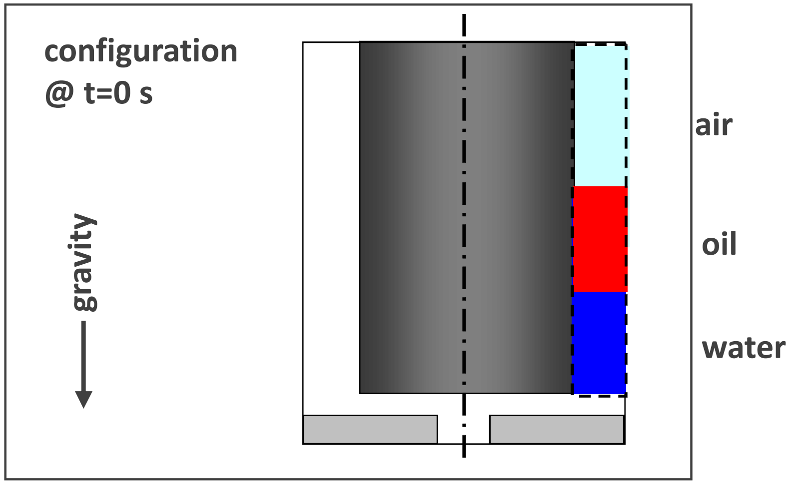
- Coalescence Rate
  (Prince and Blanch, 1990)

$$a(d_1, d_2) = h(d_1, d_2) \cdot \lambda(d_1, d_2)$$

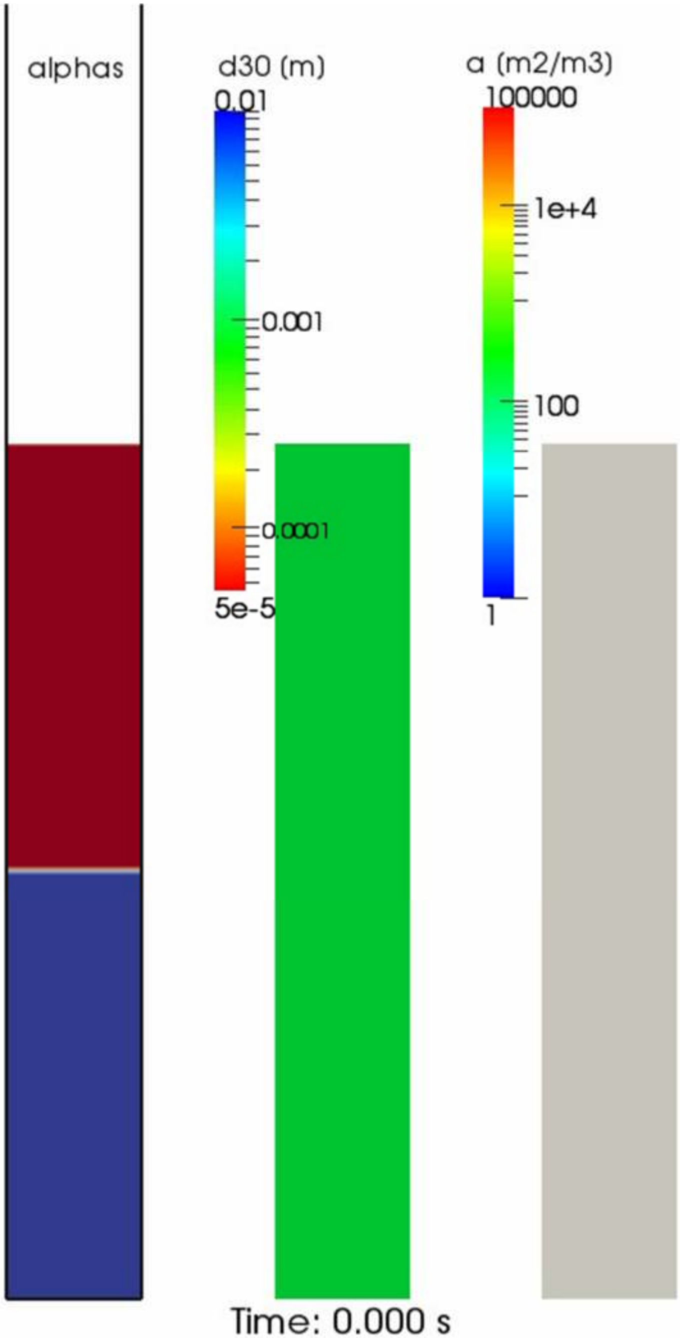
$$h(d_1, d_2) = 0.89\pi (d_1 + d_2)^2 \varepsilon^{1/3} \left(d_1^{2/3} + d_2^{2/3} \right)^{1/2}$$

$$\lambda(d_1, d_2) = exp\left(-\frac{\left(d_1 d_2 / \left(4(d_1 + d_2)\right)\right)^{5/6} \rho_c^{1/2} \varepsilon^{1/3}}{4\sigma^{1/2}} ln\left(\frac{h_0}{h_c}\right)\right)$$

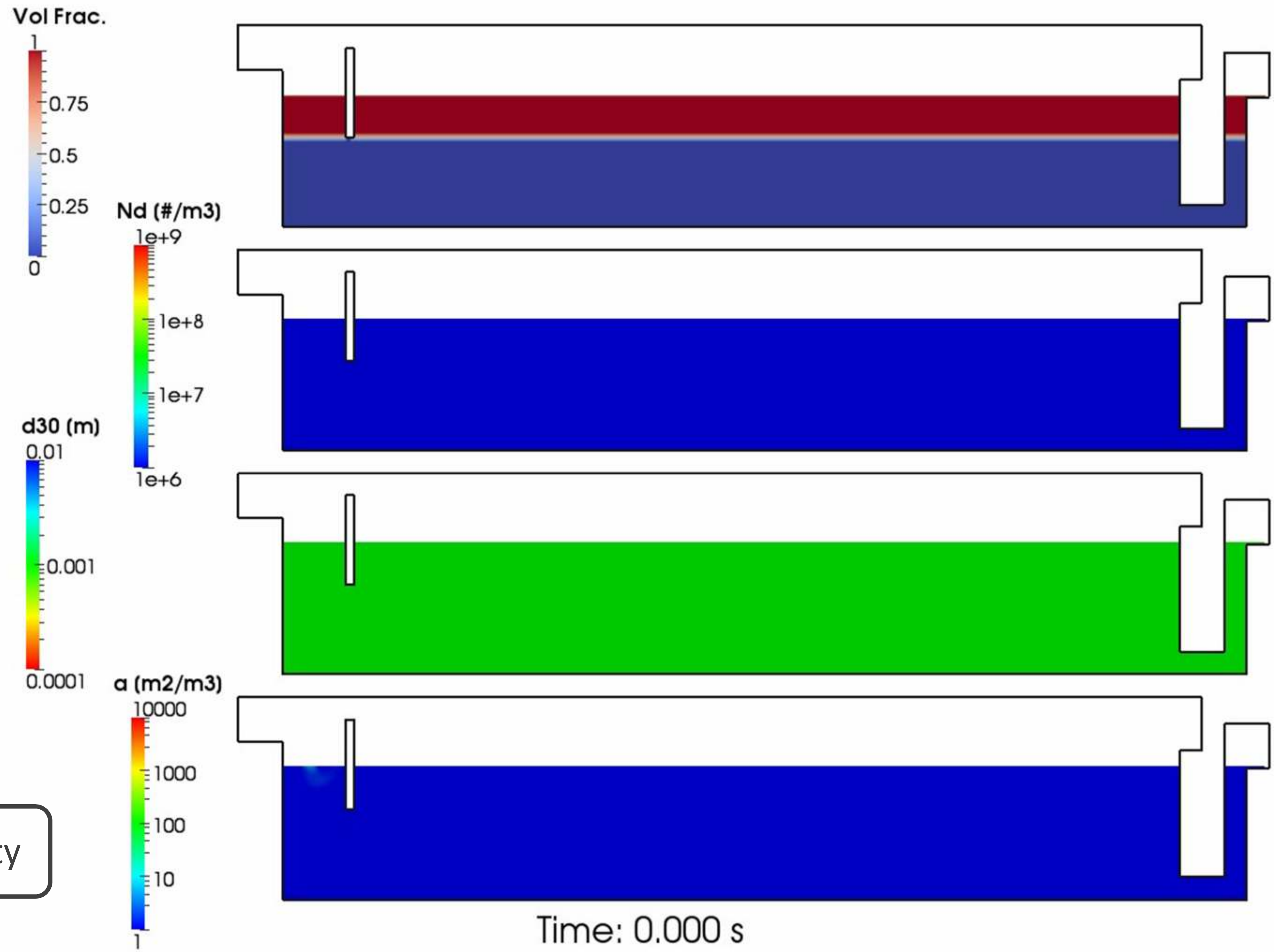
2D Annular Mixer (Water-Oil-Air)

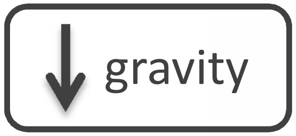


- Conditions
  - Water is dispersed phase
  - LES turbulence using Smagorinsky model
- Observations
  - Smallest droplets (largest specific interfacial area) when/where liquid contact rotor
  - Droplet size much larger than correlation predictions
  - Some problems with instabilities in Nd & a (resolved), 3D testing

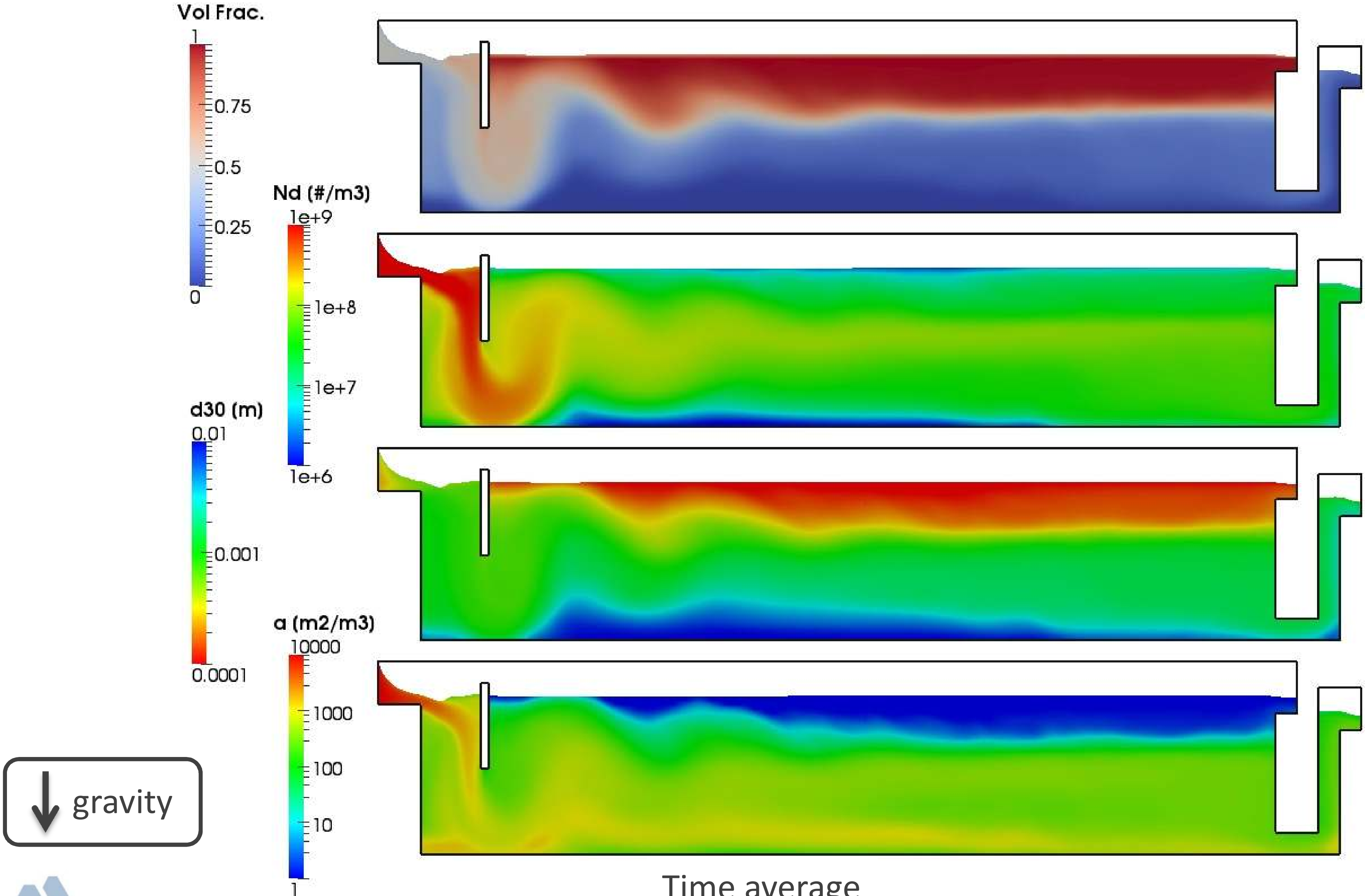


Three-phase 2D Horizontal Settler (Water-Oil-Air)





Three-phase 2D Horizontal Settler (Water-Oil-Air)

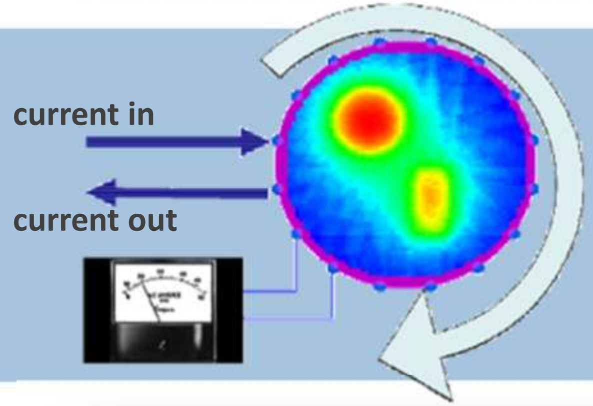


Time average

Companion Experimental Effort: Multiphase Measurements in Solvent Extraction Equipment for CFD Validation

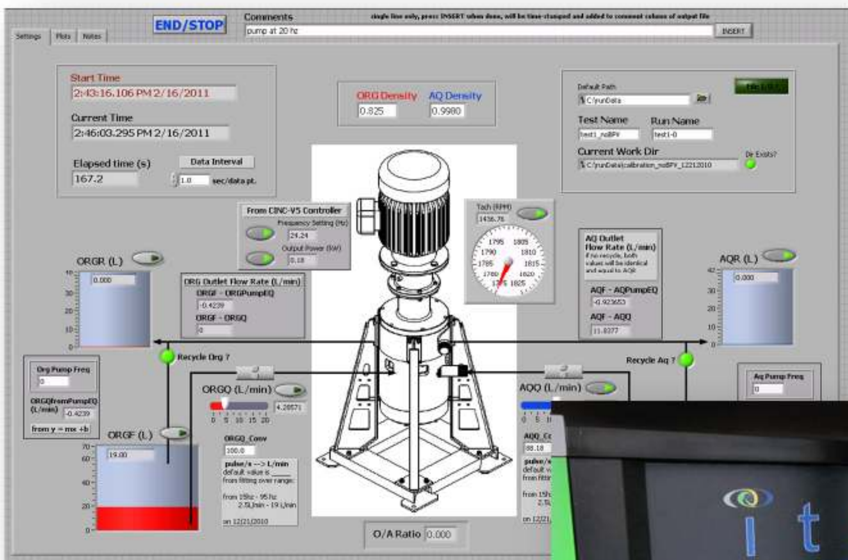
~ see poster for more ~

Contactor CFD Validation Using Electrical Resistance Tomography (ERT)

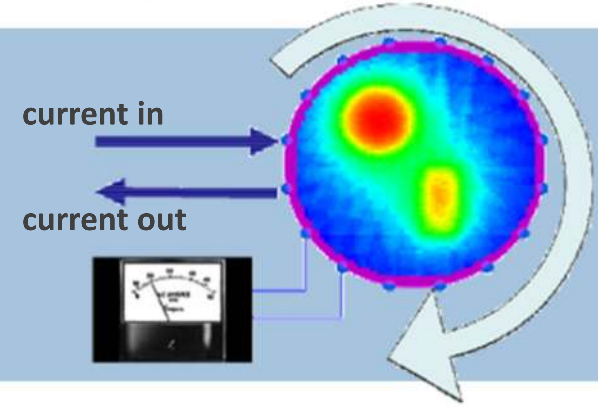


Contactor ERT (CERT) Facility

- Engineering-scale contactor (CINC V-5)
- Multiphase measurements using ERT
- HS-camera (Redlake X5plus, 500fps @ 4MP)

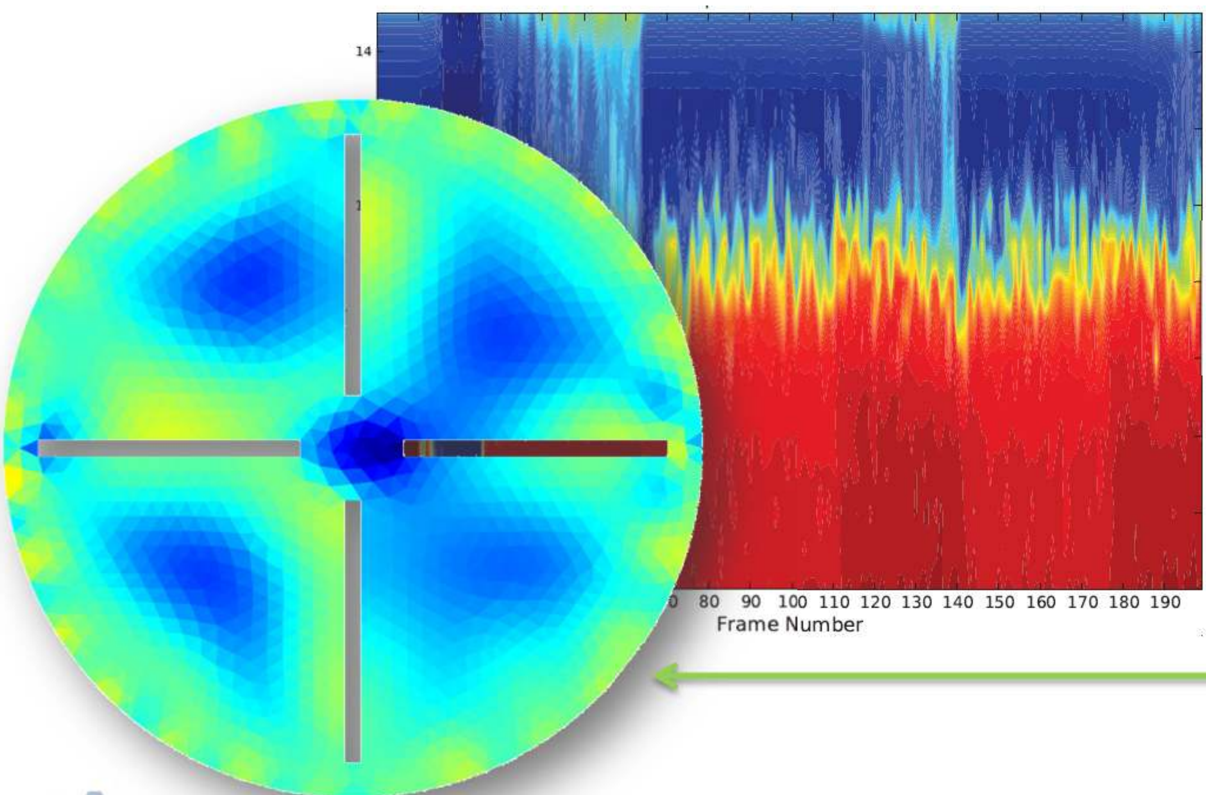


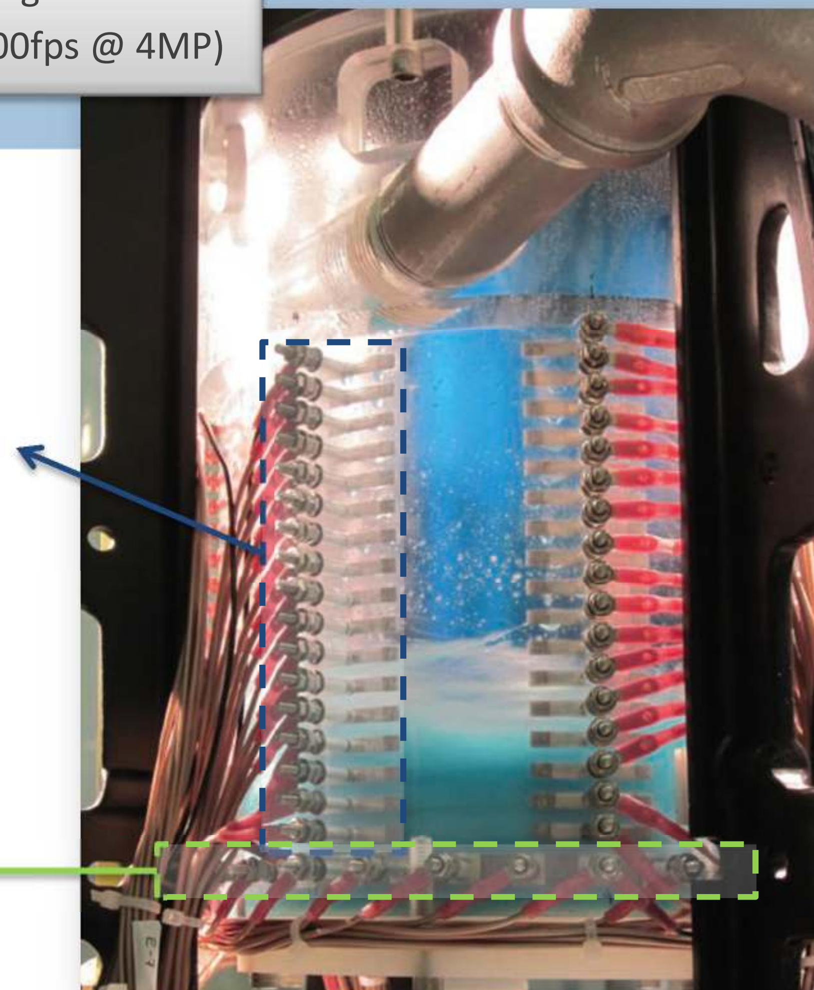
Contactor CFD Validation Using Electrical Resistance Tomography (ERT)



Contactor ERT (CERT) Facility

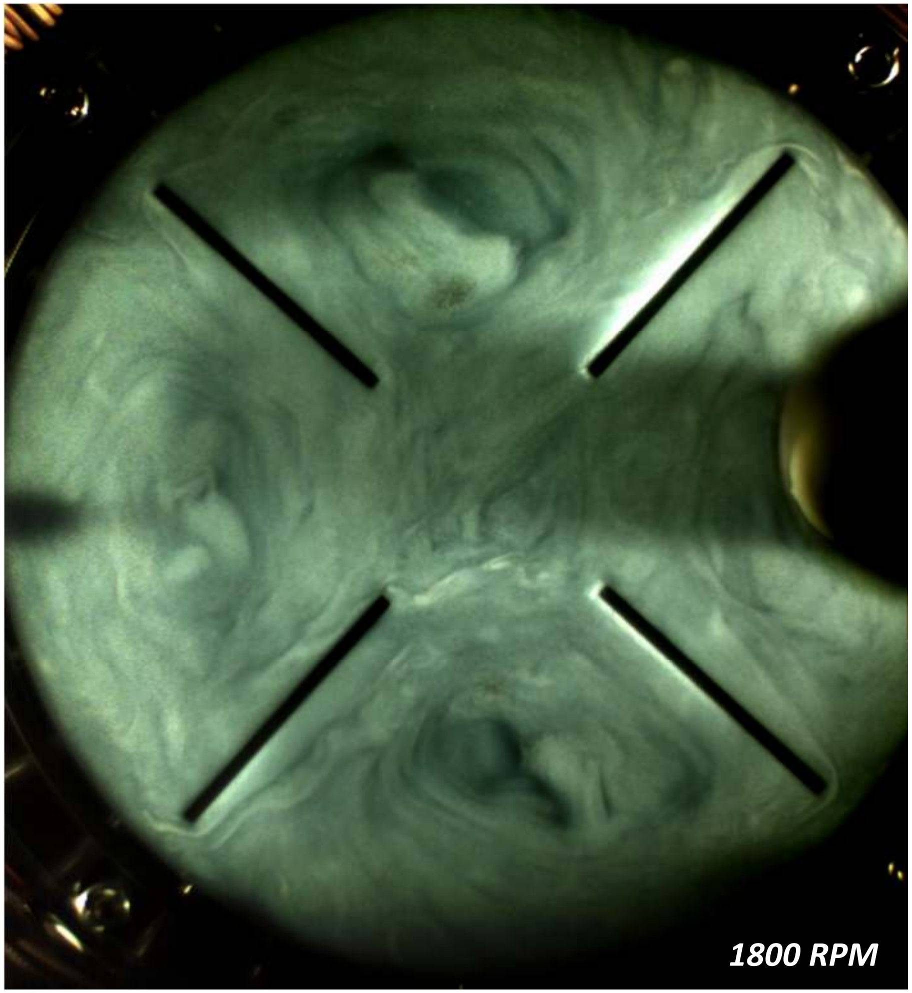
- Engineering-scale contactor (CINC V-5)
- Multiphase measurements using ERT
- HS-camera (Redlake X5plus, 500fps @ 4MP)

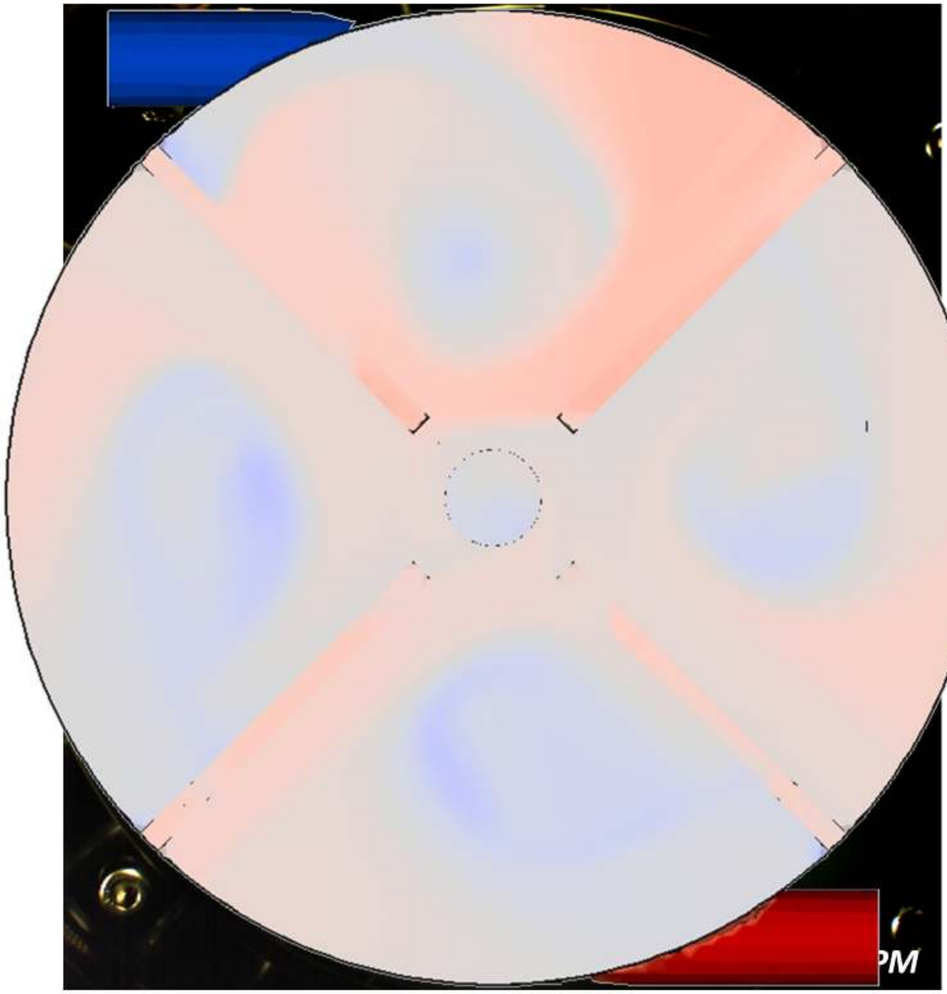




Liquid-Liquid Operation (O/A = 1)

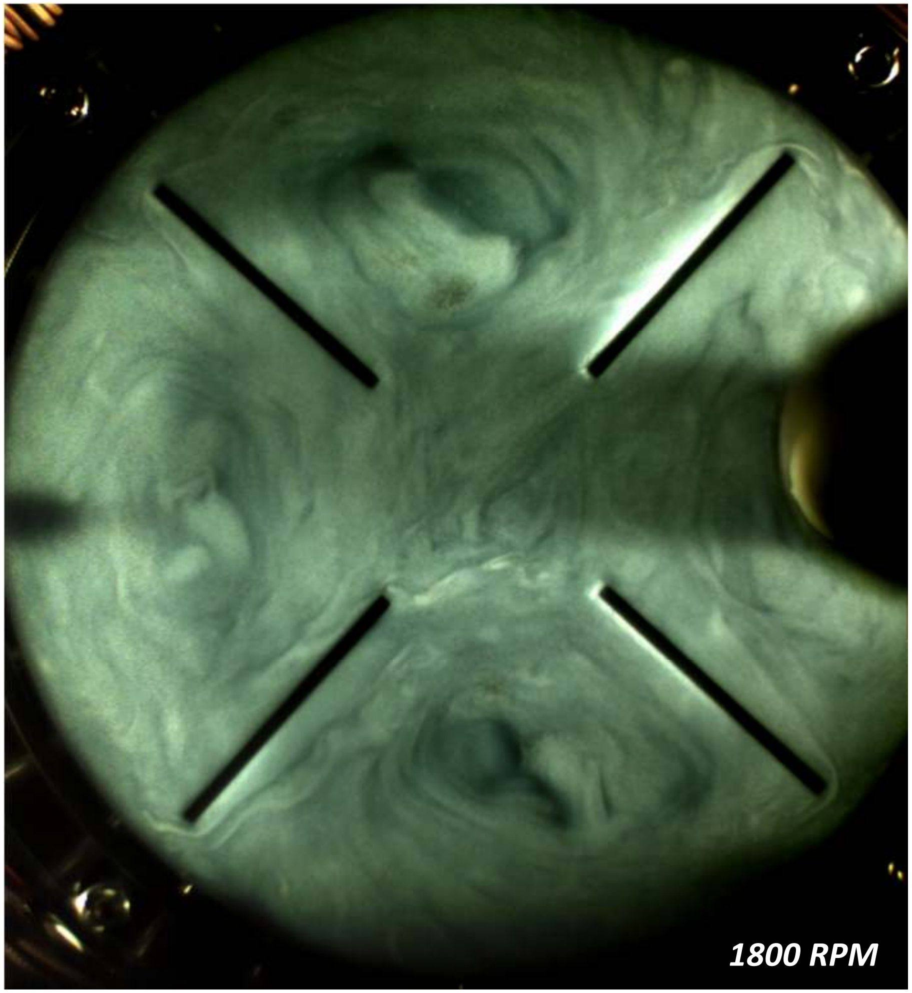
- No stable air voids between vanes as seen in the liquid-air only case
  - Aqueous rich regions?
- Fine turbulent structures and small droplet sizes

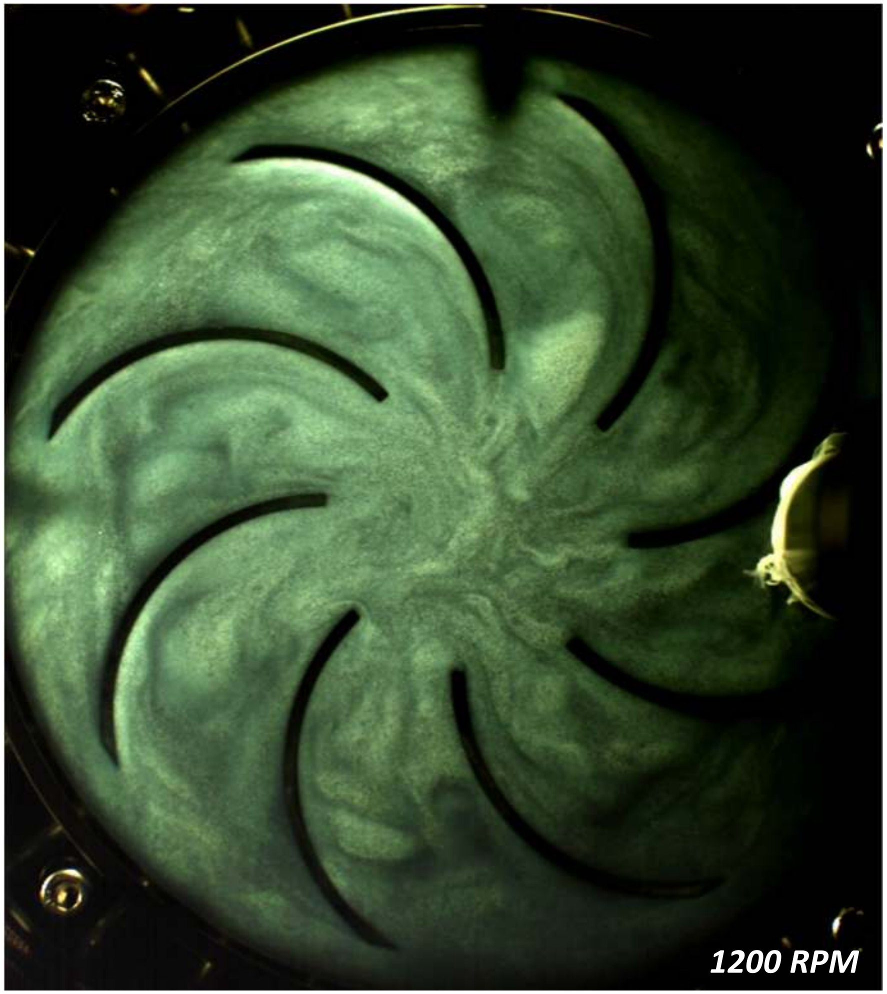




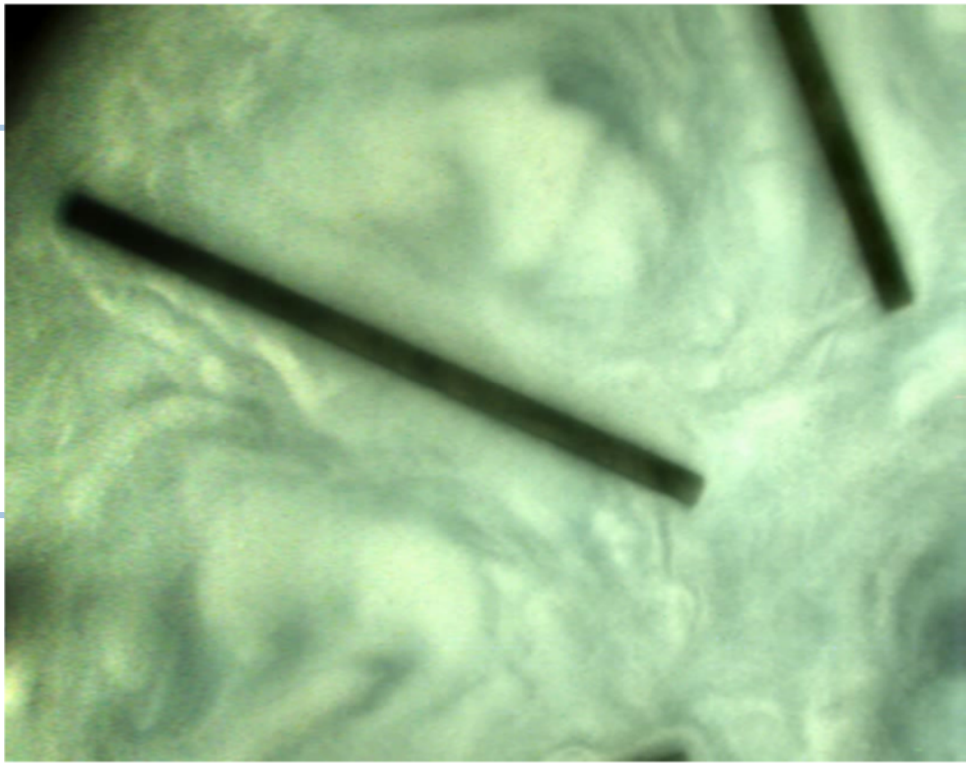
Liquid-Liquid Operation (O/A = 1)

- No stable air voids between vanes as seen in the liquid-air only case
  - Aqueous rich regions?
- Fine turbulent structures and small droplet sizes

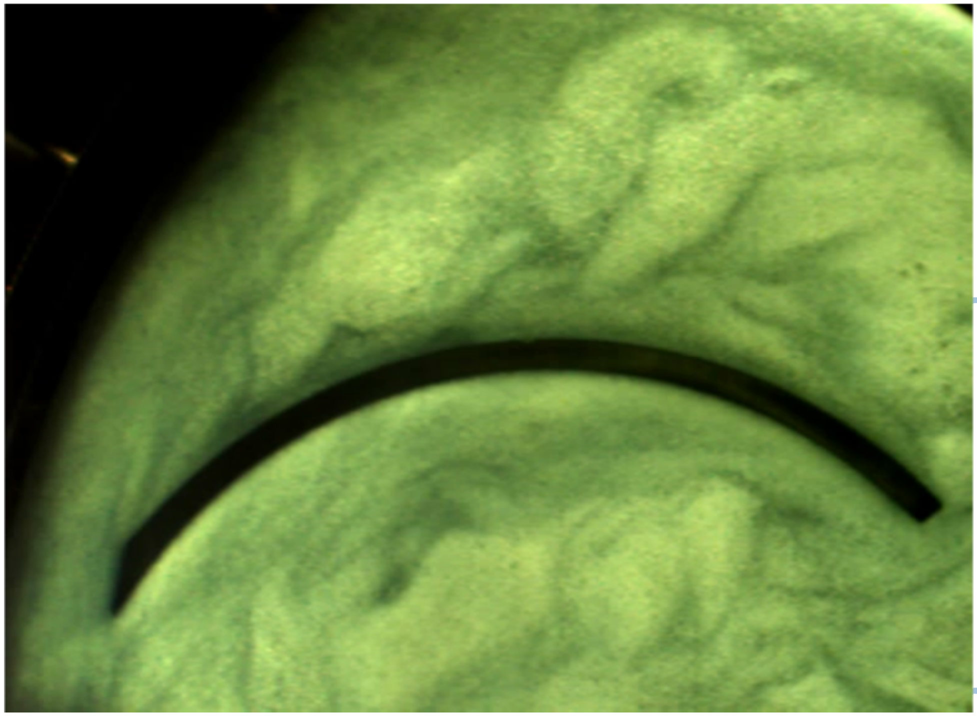




Flow Around Vanes



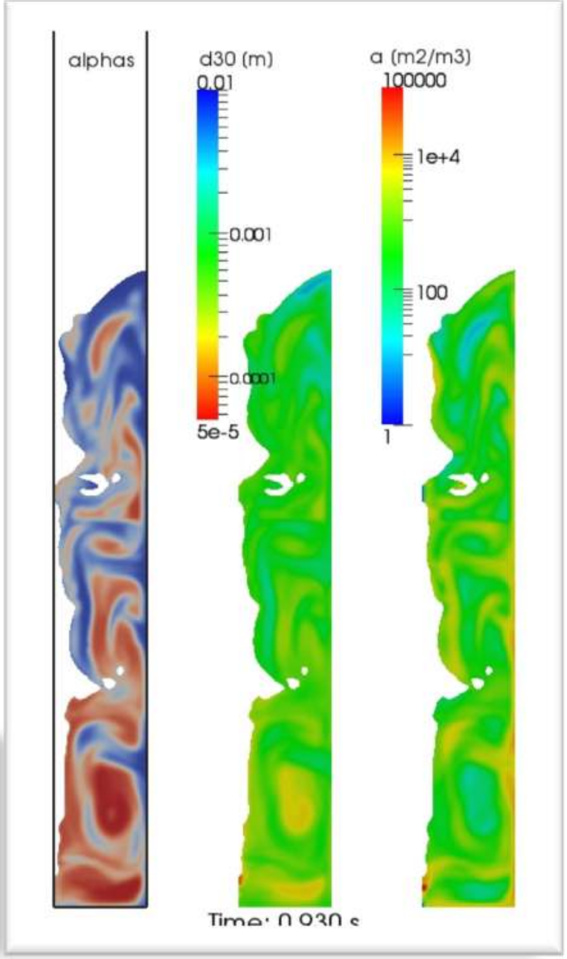
8 Straight Vanes
- 1800 RPM
- O/A = 3
- 1500 fps



8 Curved Vanes
- 1200 RPM
- O/A = 3
- 1000 fps

Summary

- Annular centrifugal contactor is a liquid-liquid extraction device with distinct advantages (e.g. compact, high-throughput) over traditional equipment—CFD offers a means of improving design and operation, supporting broader deployment
- Research focus to date has been on development of CFD tools for prediction of:
  - liquid-liquid mixing
  - stage efficiency
  - other-phase carry-over
- OpenFOAM provides open-source CFD framework for development of unique multiphase simulation capability
- Coupled Multi-fluid/VOF solver has been developed (and released)
  - Well tested for 2D mixing and separating systems; 3D testing for real contactor geometries is underway
  - Initial implementation of variable droplet size and interfacial area prediction using a reduced population balance method based on the droplet number density →
- Companion experimental effort aimed at validation of these multiphase CFD models [see poster]



Acknowledgments

- Funding from DOE-NE Fuel Cycle R&D-Separations Program
- Process Simulation and Equipment Design Group
  - Ralph Leonard (deceased)
  - Candido Pereira
- Henry Weller (OpenCFD-SGI)

Thank You

Argonne NATIONAL LABORATORY

Kent E. Wardle
Chemical Sciences and Engineering Division
Argonne National Laboratory
kwardle@anl.gov

This work was supported by the U.S. Department of Energy, Office of Nuclear Energy, under Contract DE-AC02-06CH11357.