Computer Automated Radioactive Particle Tracking (CARPT) and Gamma Ray Computed Tomography (CT) for Opaque Multiphase Flows

Power Point Presentation with Notes

NERL-DOE Workshop on Multiphase Flow Research

Morgantown, WV, June 6-7, 2006

Being unable to attend this important workshop, I would like to share with attendees some of my thoughts and suggestions via this presentation.

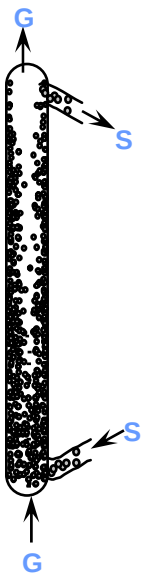
Understanding multiphase flows on all scales, from nano to very large equipment scale, and being able to model them quantitatively is essential for a myriad of technologies, including generation of liquid fuels and energy from novel sources. While both accurate experimentation and mathematical models are needed on all scales and for all types of flows (e.g., gas-solid, gas-liquid, liquid solid, gas-liquid-solid, gas-liquid-liquid-solid, etc) I will focus here on a subset of problems that deal with quantification of fluid dynamics in multiphase reactor systems. This requires the development of codes that can effectively handle large systems of complex geometry, the improved understanding and better physical models of inter-phase interaction and turbulence, and the experimental validation of these codes.

In our Chemical Reaction Engineering Laboratory (CREL) at Washington University (WUSTL) we have developed and implemented two unique facilities for determination of velocity and holdup (volume fraction) fields in opaque systems of large volume fraction of dispersed phase. Our Computer Automated Radioactive Particle Tracking (CARPT) and Gamma Ray Computed Tomography (CT) have been used successfully to map bubble columns, stirred tanks, fluidized beds, etc.

The enclosed power point slides and notes (please read the document in Notes format) illustrate some of the successful uses of these techniques and continued remaining challenges for which we hope to attract collaborators from the workshop attendees.

Please see our last slide 33 for areas in which we seek partners for collaboration

Computer Automated Radioactive Particle Tracking (CARPT) and Gamma Ray Computed Tomography (CT) for Opaque Multiphase Flows



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NETL Workshop on Multiphase Flow Research

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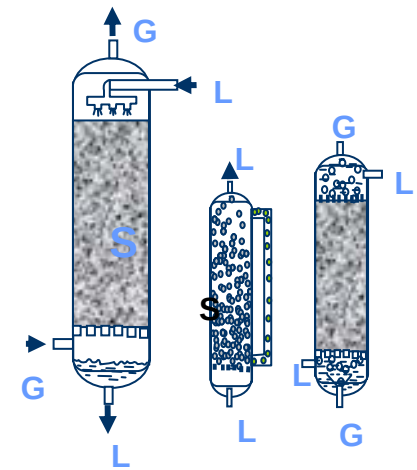
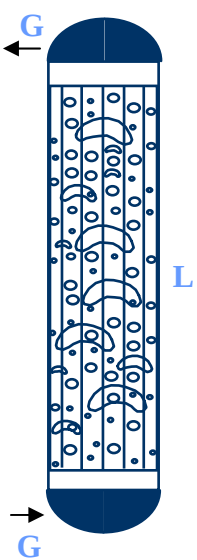
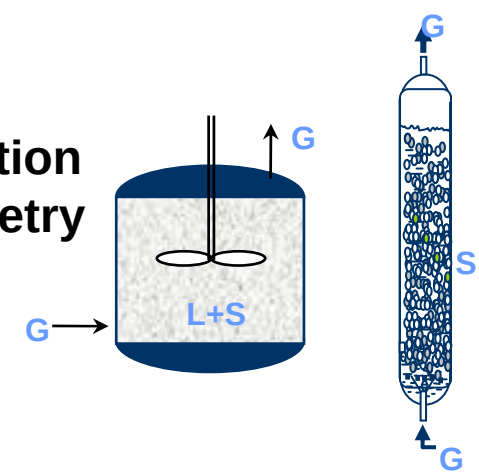


Outline

- Importance of multiphase reactors and flows
- Flow pattern and phase distribution determination
  - Conventional tracer technique and densitometry
  - Particle tracking and tomography
  - Computational fluid dynamics (CFD)
- Improved engineering models
- Conclusions





Chaouki, Larachi, Dudukovic, I & EC Res., 36(11), 4476-4503 (1997)
Dudukovic, Mills, Larachi, Catalysis Reviews, 44(1), 123-246 (2002)

Energy

Coal, oil, gas, nuclear power plants

Synthesis & Natural Gas Conversion

MeOH, DME, MTBE, Paraffins, Olefins, Higher alcohols, ….

Petroleum Refining

HDS, HDN, HDM, Dewaxing, Fuels, Aromatics, Olefins, ...

Value of Shipments:

$US 637,877 Million

Uses of Multiphase Reactor Technology

Bulk Chemicals

Aldehydes, Alcohols, Amines, Acids, Esters, LAB's, Inorg Acids, ...

Polymer Manufacture

Polycarbonates, PPO, Polyolefins, Specialty plastics

Fine Chemicals & Pharmaceuticals

Ag Chem, Dyes, Fragrances, Flavors, Nutraceuticals,...

Biomass Conversion

Syngas, Methanol, Ethanol, Oils, High Value Added Products

Environmental Remediation

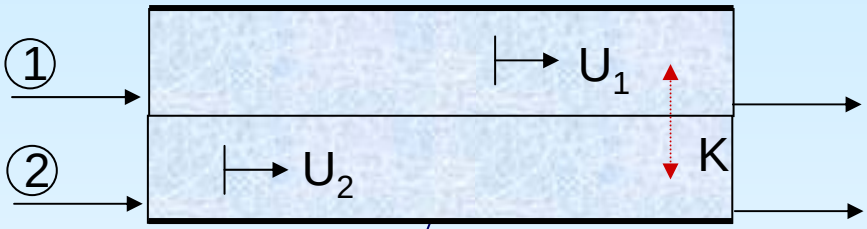
De-NOx, De-SOx, HCFC's, DPA, "Green" Processes ..

ADVANCES IN MULTIPHASE REACTORS REQUIRE:
Flow Mapping and Modeling of Opaque Multiphase Systems

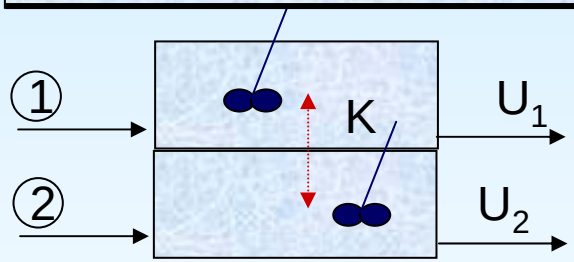
REACTOR SCALE MODELS FOR CONTACTING OF TWO MOVING PHASES

Ideal Reactor Concepts:

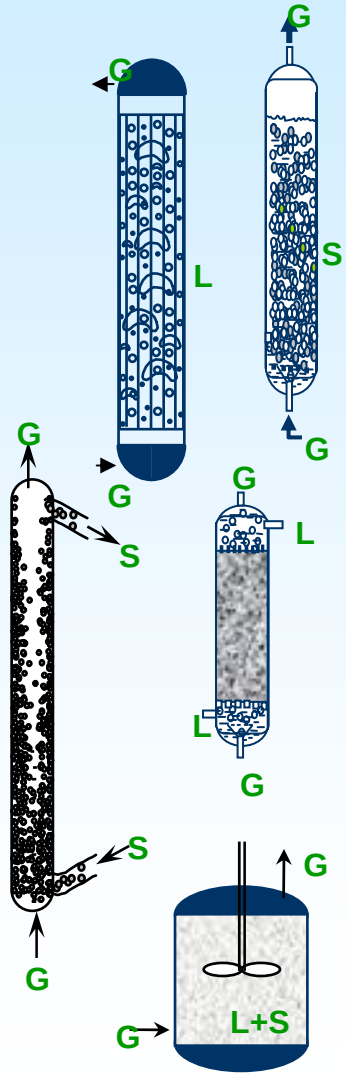
A) Plug Flow (PFR)



B) Stirred Tank (CSTR)



C) Axial Dispersion Model



D) Need More Accurate Flow & Mixing Description Via

Phenomenological models based on:

1) CFD Models (Euler-Euler Formulation)
2) Experimental Validation: Holdup Distribution and Velocity Field

Dudukovic, AICHE Symposium Ser., 321, 30-50 (1999)

Dudukovic, Larachi, Mills, Catalysis Reviews (2002), 44(1), 123-246

Photons of Visible light fail to pass through opaque objects

High energy gamma ray can pass through opaque objects

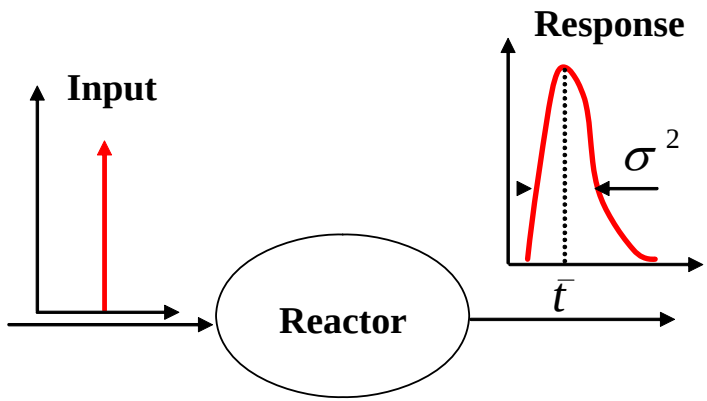
This concept is used:

- To determine chordal densities via gamma ray densitometry and global flow patterns (RTD) by radioactive tracer studies
- To quantify phase distributions with the aid of Computer Tomography
- To monitor the motion of a single radioactive particle which mimics the density and flow behavior of a particular phase in order to obtain velocity fields and mixing patterns

Radioactive Techniques in Reactor Model Development

Classic Methods for trouble shooting and "blackbox" model development

Tracer impulse response

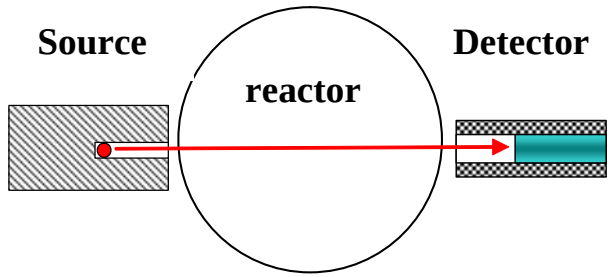


$\bar{t}$ → Mean holdup

σ^2 → Dispersion coefficient

Match dispersion model or CSTR in series model or some other compartmental model to observed response

Gamma Ray Densitometry



Line averaged holdup

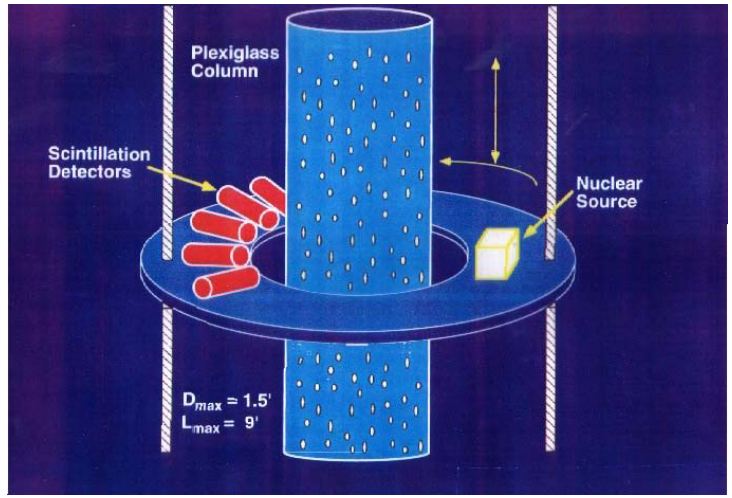
From a number of line measurements obtain an approximate assessment of density and phase holdup distribution

Modern methods for CFD validation and flow and mixing model development.

Tomography and single particle tracking.

Computed Tomography (CT)

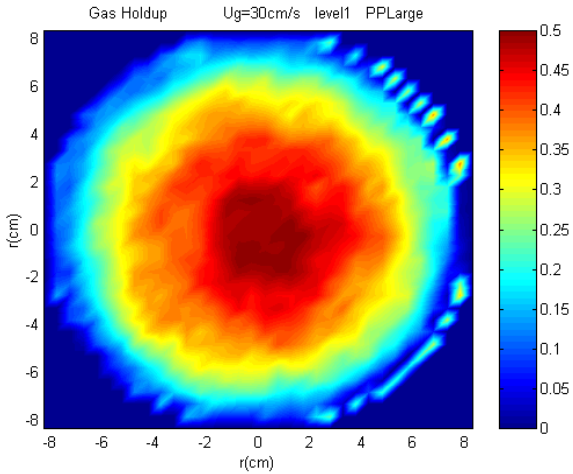
Single source CT is a technique for measurement of the cross-sectional density distribution of two phase flow by measuring the attenuation distribution in **two** phase systems (e.g. G-L, ...).

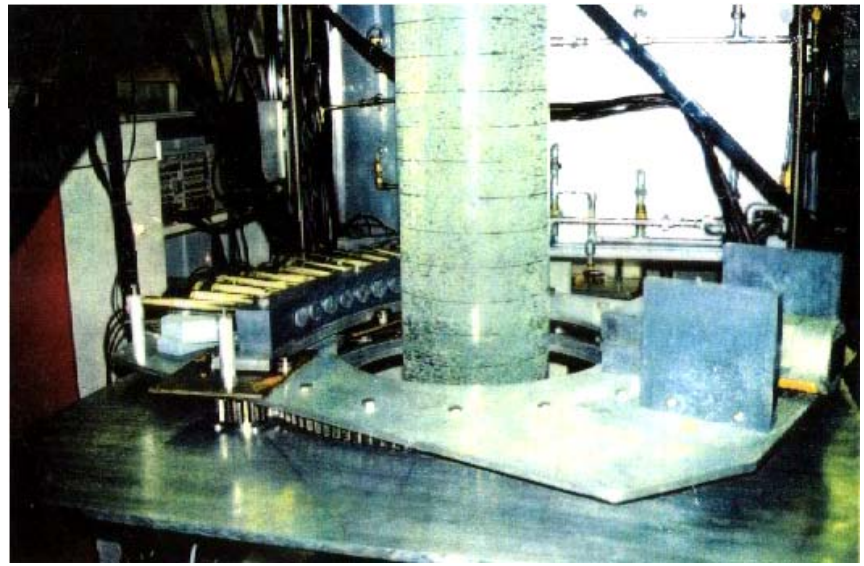


$$A = -\ln\frac{I}{I_0} = \sum_l (\rho\mu)_{eff,ij} l_{ij}$$

$$(\rho\mu)_{eff,ij} = \sum_K (\rho\mu)_{K,ij}\,\varepsilon_{K,ij}$$

Experimental Result





Kumar (1994), Kumar et al., (1995)

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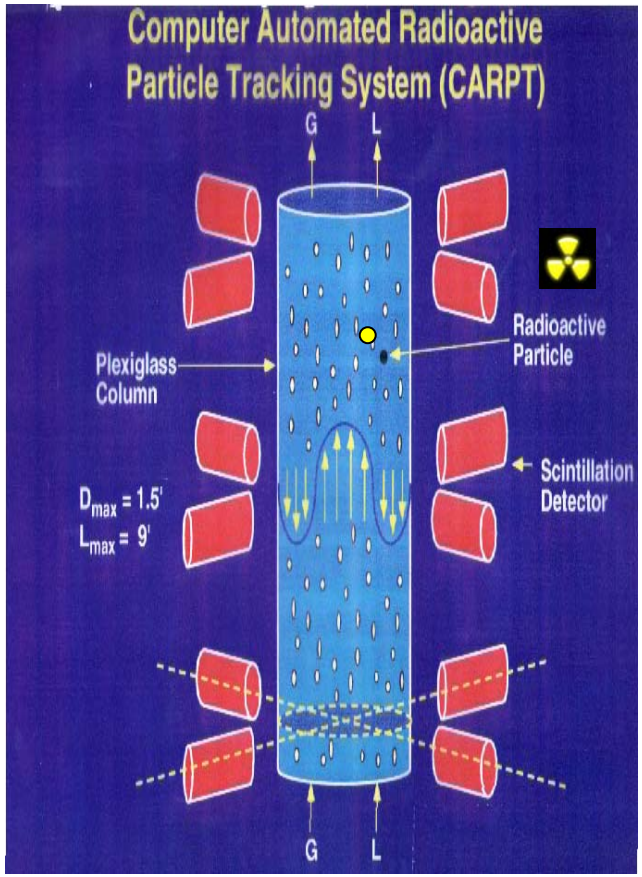
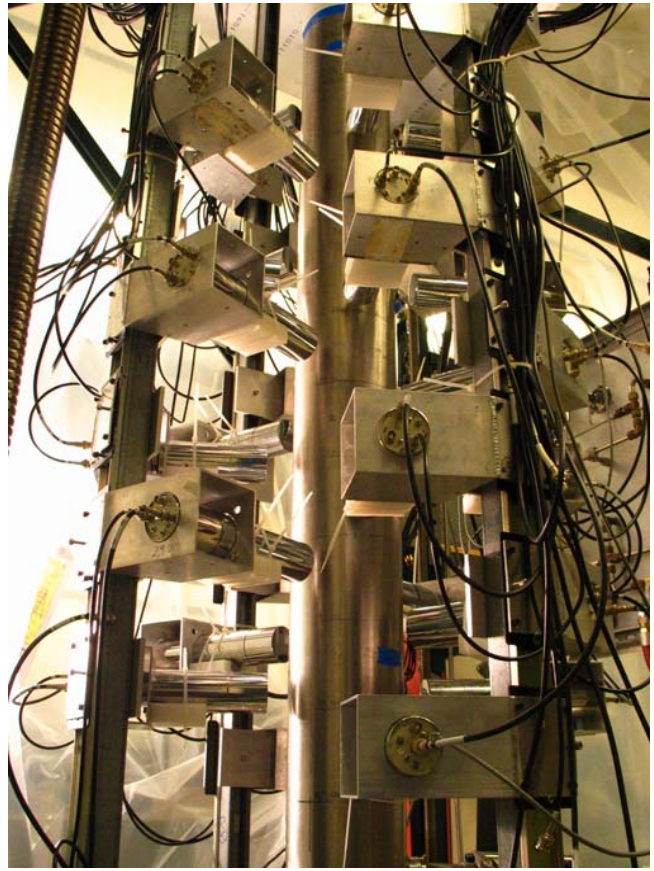
Radioactive Particle Tracking

Lin et al. (1985), Moslemian (1986), Devanathan (1990), Devanathan et al. (1991), Chaouki, Larachi and Dudukovic (1997)



CARPT (RPT) Schematic



Experimental Setup

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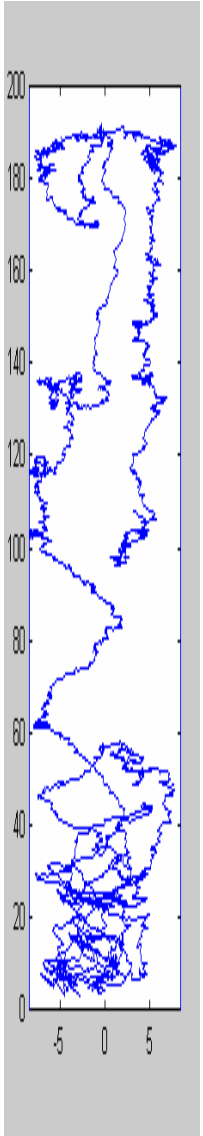
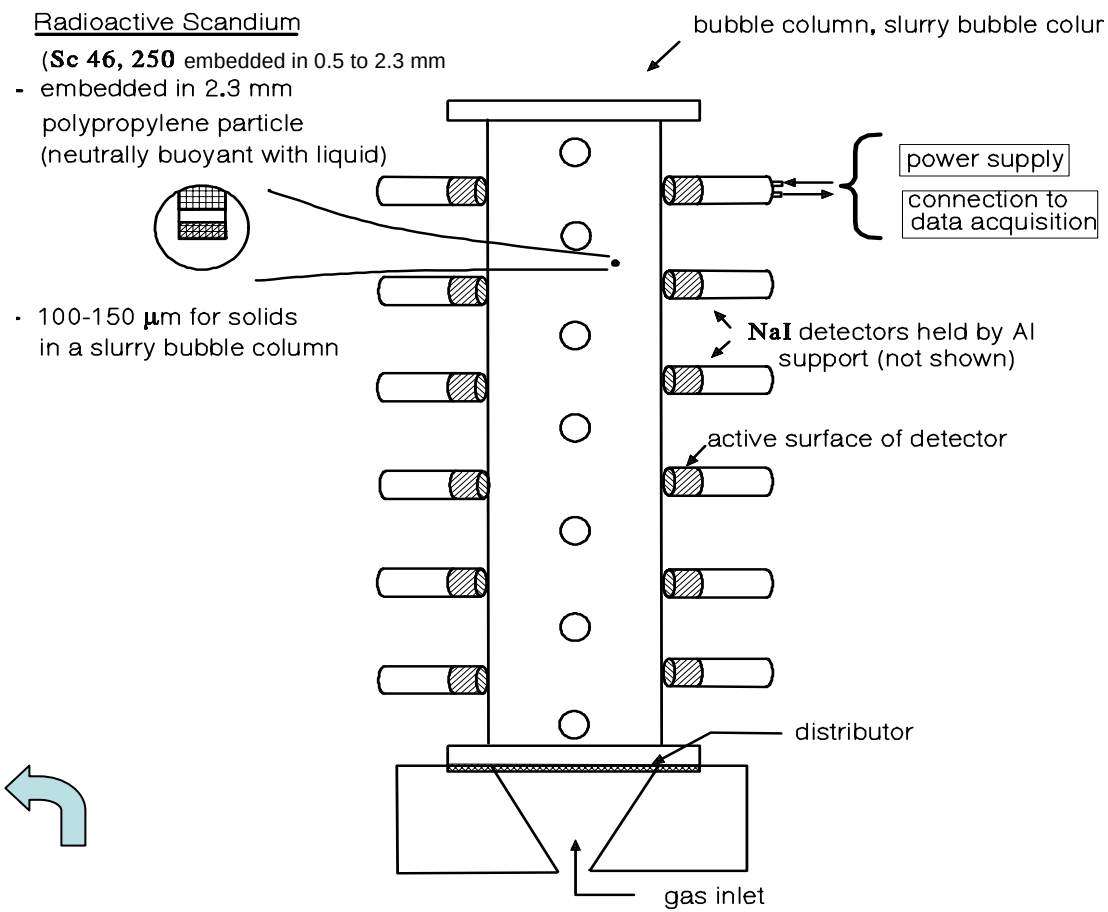
Computer Automated Radioactive Particle Tracking (CARPT)



1.In-situ calibration

2. Particle Tracking



The tracer particle Lagrangian trajectory

Counts from Detectors (*t*)

↓ +

Distance - Count Map

↓

Regression / Monte-Carlo Search

↓

Instantaneous Positions
(*x, y, z, t*)

↓

Filter

↓

Filtered Instantaneous
Positions (*x, y, z, t*)

↓

Time-Difference Between
Successive Locations

Instantaneous Velocities
(*x, y, z, t*)

↓

Ensemble (Time) Average

↓

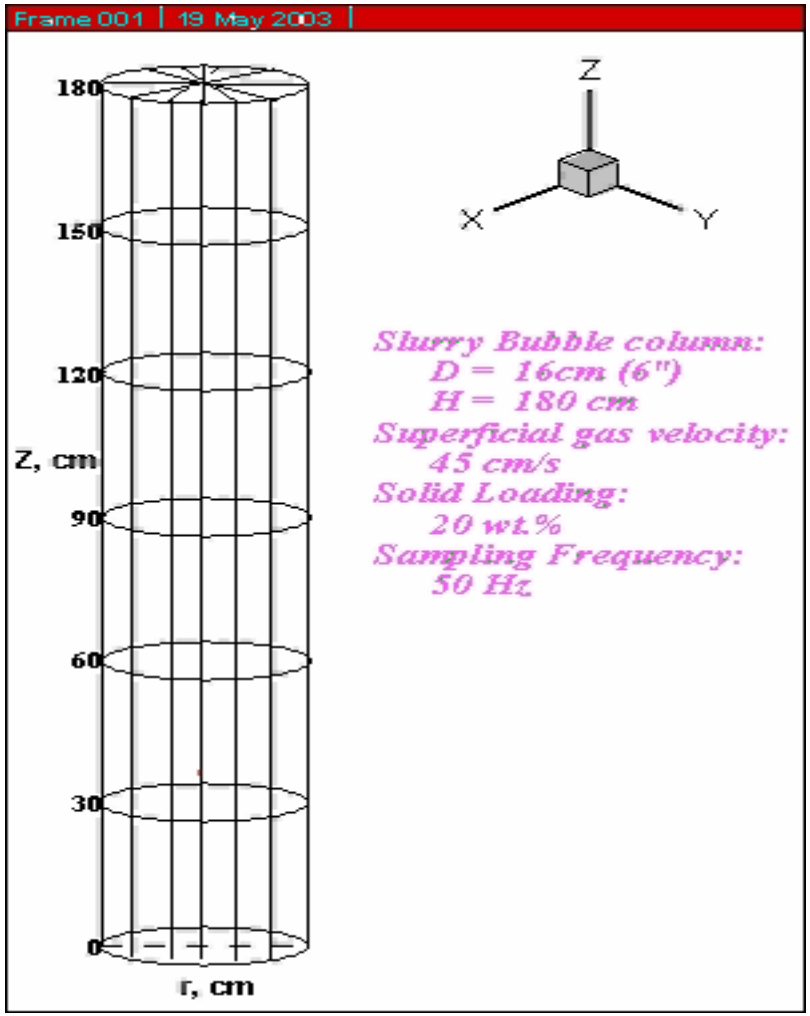
Mean Velocities
(*x, y, z*)

Fluctuating
Velocities
(*x, y, z, t*)

Turbulent Parameters,
Reynolds Stresses,
TKE, Eddy diffusivities,
etc.

Moslemian (1986);
Devanathan (1990); Degaleesan (1996);
Chaouki, Larachi, Dudukovic (1997);

Particle tracking in multiphase systems



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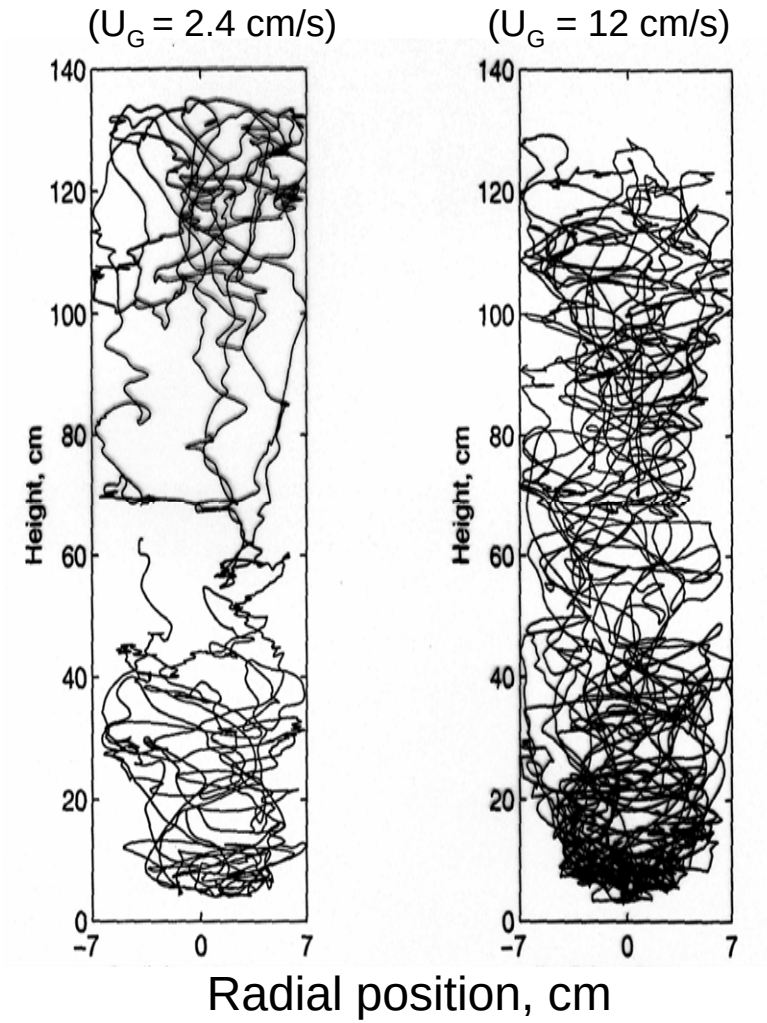
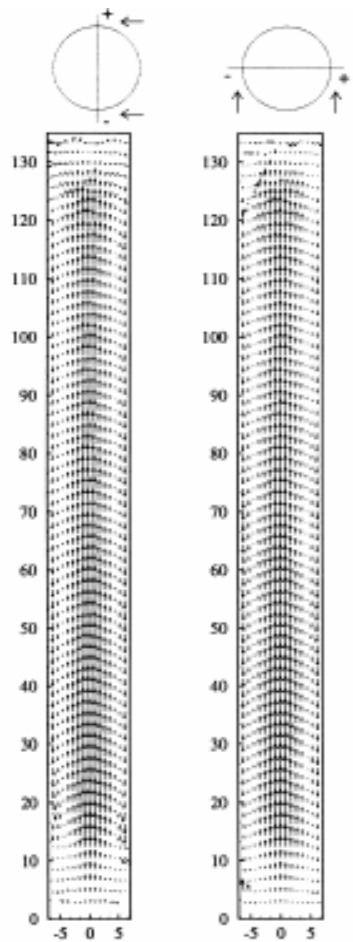
Example of information gained from particle tracking

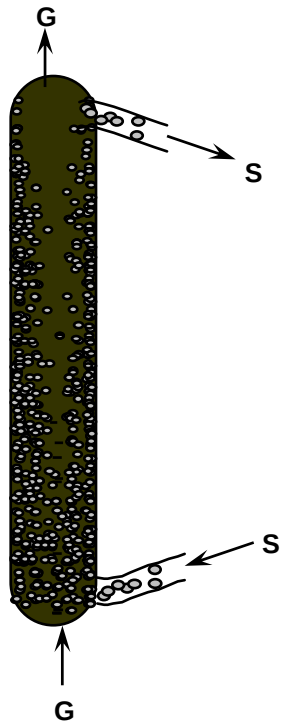


Portion of Particle Lagrangian Trajectory from CARPT in a 6” Bubble Column

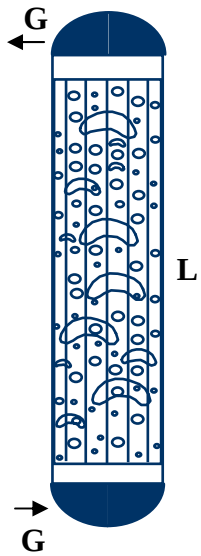


Ensemble Averaged Velocity Vectors

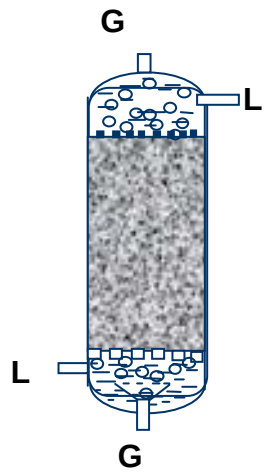
Systems to which these techniques have been applied in the past



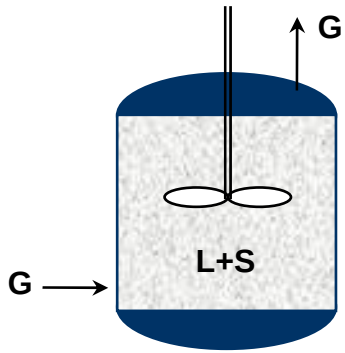
Risers



Bubble Column



Packed Beds



Stirred Tanks

Fluidized Beds

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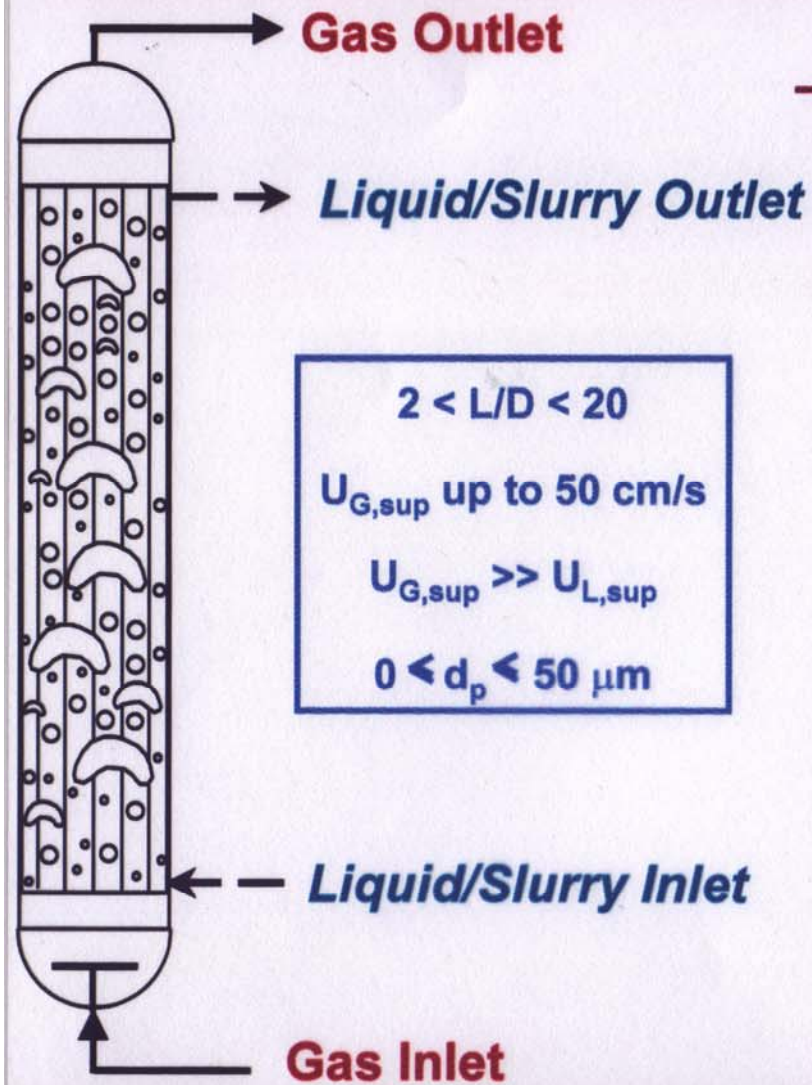
Bubble Column And Some Important Applications



$2 < L/D < 20$

$U_{G,sup}$ up to 50 cm/s

$U_{G,sup} >> U_{L,sup}$

$0 < d_p < 50\ \mu m$

	T, °C	P, atm
Partial oxidation of ethylene to acetaldehyde	130	3
Wet-air oxidation of sewage sludge	200-300	40-120
Oxidation of cumene to phenol	80-125	5-8
Hydrogenation of hydroxilamine	50-60	25-30
Conversion of natural gas to syngas	900	15-30
Methanol synthesis	220-250	50-100
Fischer-Tropsch synthesis	220-260	134-204
Hydroformylation (oxo) processes	160	50-100

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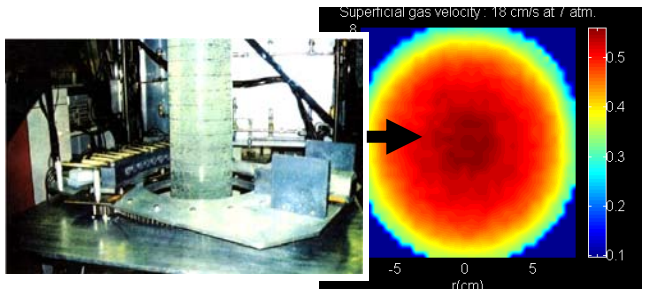
Bubble Column Example

CARPT-CT and other measurements are used to develop an appropriate phenomenological reactor flow and mixing model. CFD generated data are used to assess model parameters at pilot plant or plant conditions. Reactor flow and mixing model are coupled with the kinetic information.

Degaleesan et al., Chem. Eng. Sci., 51, 1967(1996); I&EC Research, 36,4670 (1997);

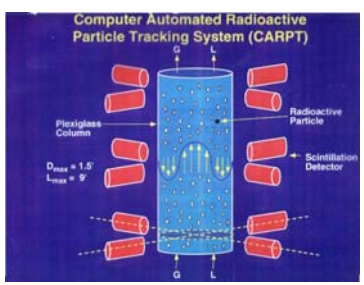
Gupta et al., Chem. Eng. Sci., 56, 1117 (2001)



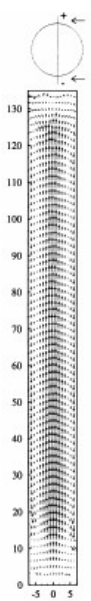
CT

CT SCAN

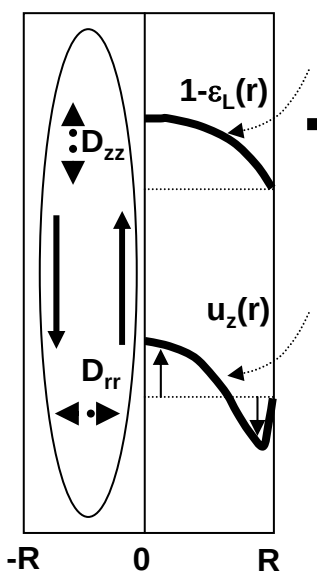
CFD + CARPT + CT

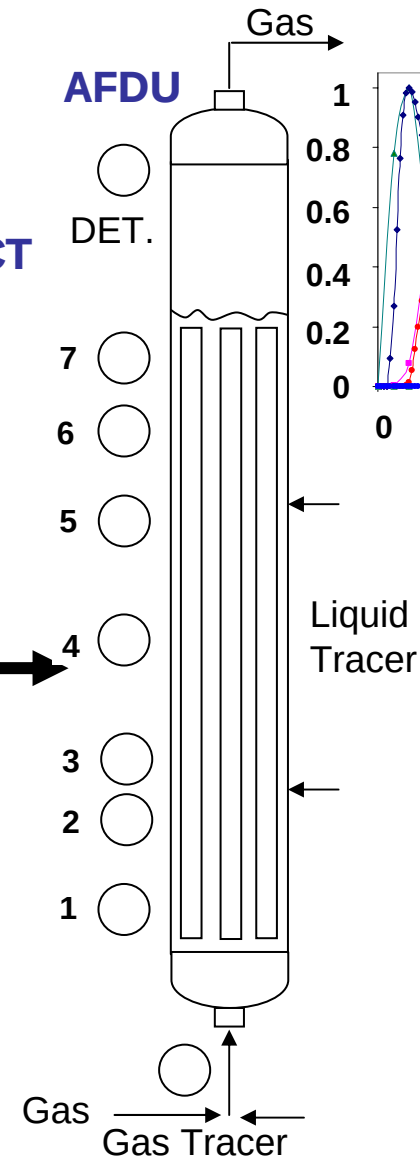


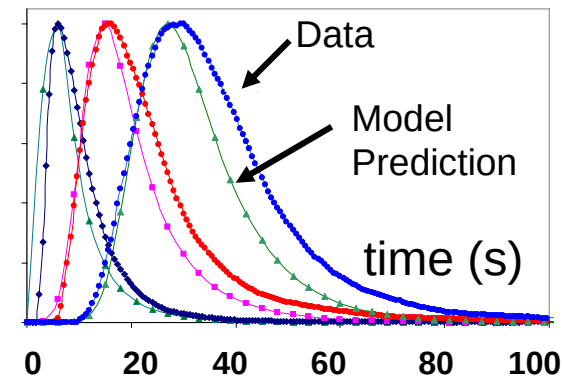
CARPT

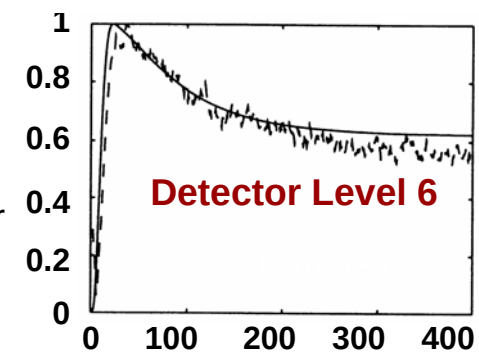


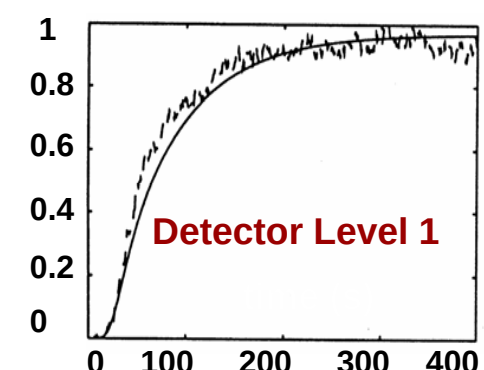
FLOW PATTERN











Ensemble Averaged Equations for Two-Phase Flow

Liquid Phase

$$\frac{\partial \varepsilon_c}{\partial t} + \nabla.(\varepsilon_c \mathbf{u}_c) = 0$$

$$\rho_c \varepsilon_c \left(\frac{\partial \mathbf{u}_c}{\partial t} + \mathbf{u}_c.\nabla \mathbf{u}_c \right) = \rho_c \varepsilon_c \mathbf{g} - \varepsilon_c \nabla p - (\mathbf{M}_d + \mathbf{M}_{vm}) + \nabla.(\varepsilon_c \boldsymbol{\sigma}_c) + \nabla.(\varepsilon_c \boldsymbol{\sigma}_c^b)$$

Gas Phase

$$\frac{\partial \varepsilon_d}{\partial t} + \nabla.(\varepsilon_d \mathbf{u}_d) = 0$$

$$\rho_d \varepsilon_d \left(\frac{\partial \mathbf{u}_d}{\partial t} + \mathbf{u}_d.\nabla \mathbf{u}_d \right) = \rho_d \varepsilon_d \mathbf{g} - \varepsilon_d \nabla p + (\mathbf{M}_d + \mathbf{M}_{vm})$$

CLOSURES

Inter-Phase Momentum Exchange

$$\mathbf{M}_{vm} = \frac{1}{2} \varepsilon_c \varepsilon_d C_{vm} \left(\frac{D\mathbf{u}_c}{Dt} - \frac{D\mathbf{u}_d}{Dt} \right)$$

$$C_{vm} = 1 + 3.32\varepsilon_d + O(\varepsilon_d^2)$$

$$\mathbf{M}_d = \frac{6\varepsilon_c \varepsilon_d}{\pi d_p^3} \mathbf{F}_d \; ; \qquad C_D = \max\left[\frac{24}{Re}\left(1 + 0.15 Re^{0.687}\right), f\,\frac{8}{3}\frac{Eo}{Eo+4} \right]$$

$$\mathbf{F}_d = \frac{1}{8} \rho_c \pi d_p^2 C_D |\mathbf{u}_c - \mathbf{u}_d| (\mathbf{u}_c - \mathbf{u}_d) \qquad f = \left\{ \frac{1 + 17.67\varepsilon_c^{9/7}}{18.67\varepsilon_c^{3/2}} \right\}^2$$

Stresses

$$\boldsymbol{\sigma}_c = \mu_c^* \left(\nabla \mathbf{u}_c + \nabla \mathbf{u}_c^T \right); \qquad \frac{\mu_c^*}{\mu_c} = 1 + \frac{5}{2}\varepsilon_d + O(\varepsilon_d)$$

$$\boldsymbol{\sigma}_c^b = -\rho_c \overline{\mathbf{u}_c' \mathbf{u}_c'}$$

$$\boldsymbol{\sigma}_c^b = \rho_c \nu_b^t \left(\nabla \mathbf{u}_c + \nabla \mathbf{u}_c^T \right); \qquad \nu_b^t = k_b \varepsilon_d d_p |\mathbf{u}_c - \mathbf{u}_d|$$

$$k_b = 1.2 \quad \text{(Empirical Constant)}$$

$$Eo \equiv \text{Eotvos Number} \equiv g\rho_c d_p^2 / \tau$$

$$Re \equiv \text{Bubble Reynolds Number} \equiv \rho_c d_p |\mathbf{u}_c - \mathbf{u}_d| / \mu_c$$

Input Parameter : Bubble Size, d_p

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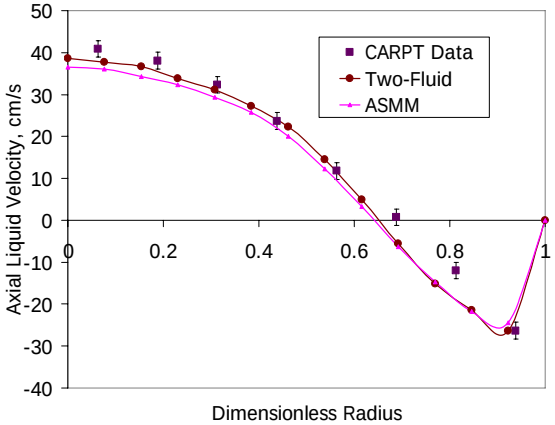
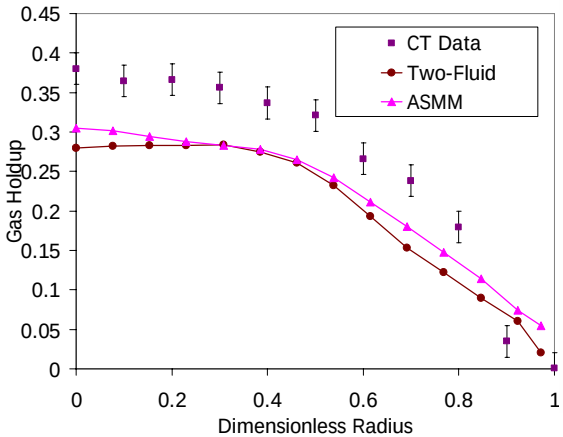
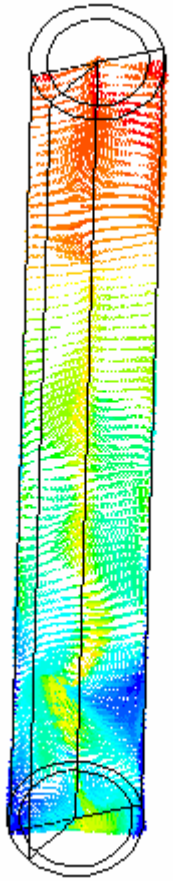
Two-Fluid CFD of 3D Bubble Columns Using FLUENT

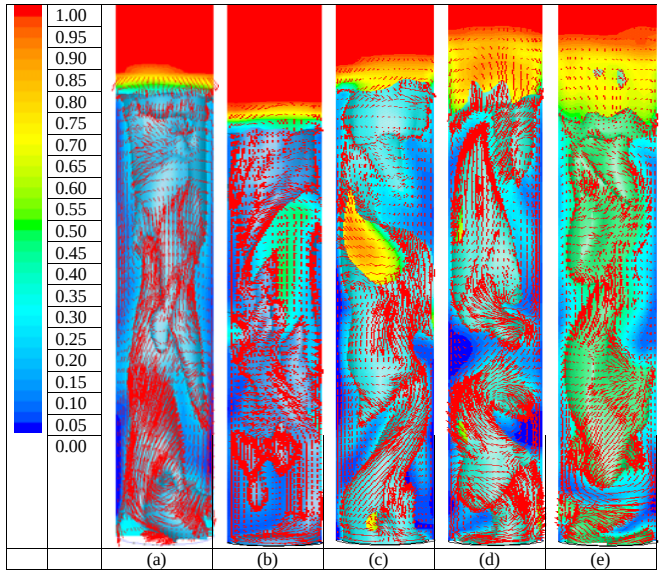
Sanyal et al., Chem. Eng. Sci., 55, 5071 (1999)



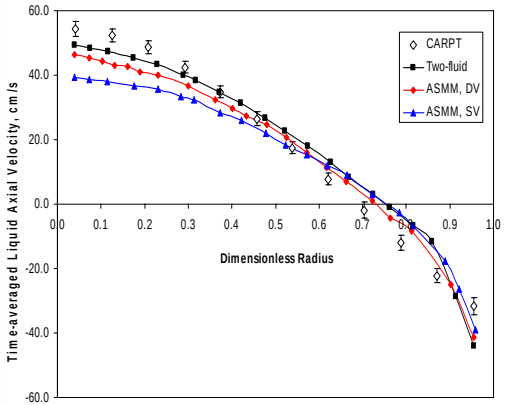
19 cm – ID
12 cm/s

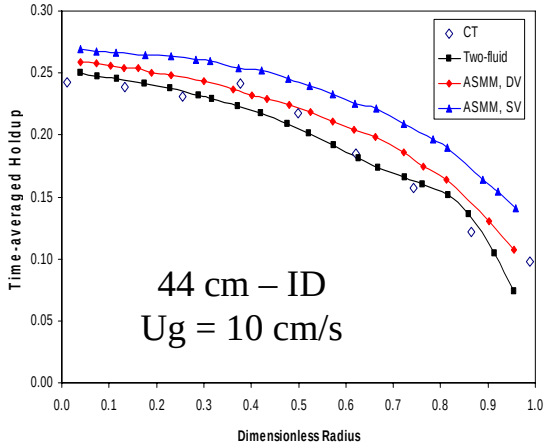






	(a)	(b)	(c)	(d)	(e)
Ug, cm/s	10	14	30	30	30
P, Bar	1	1	1	4	10





Multiphase k-ε

Implementation of Breakup and Coalescence Models ; Chen (2004)

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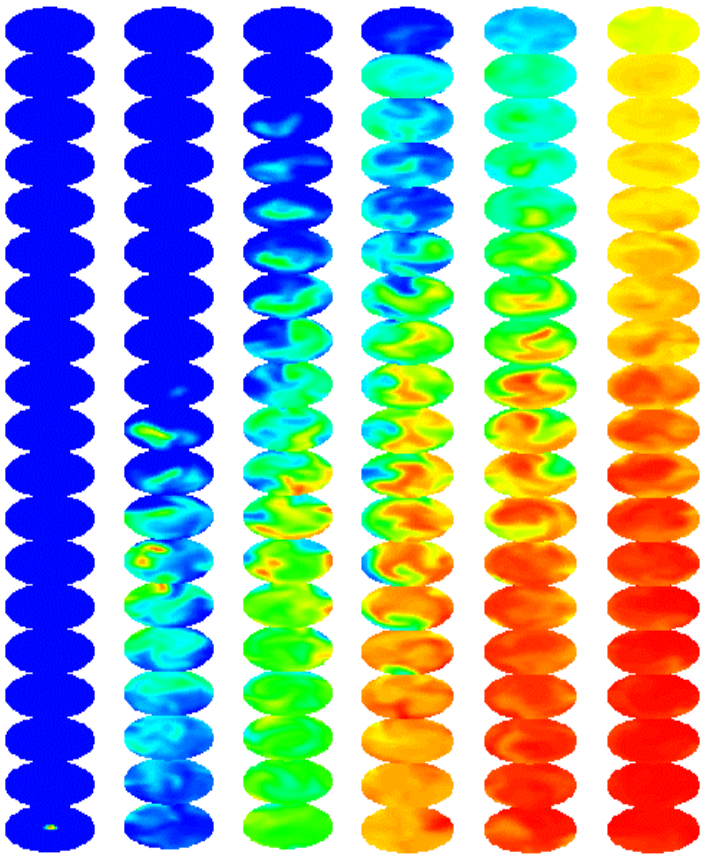
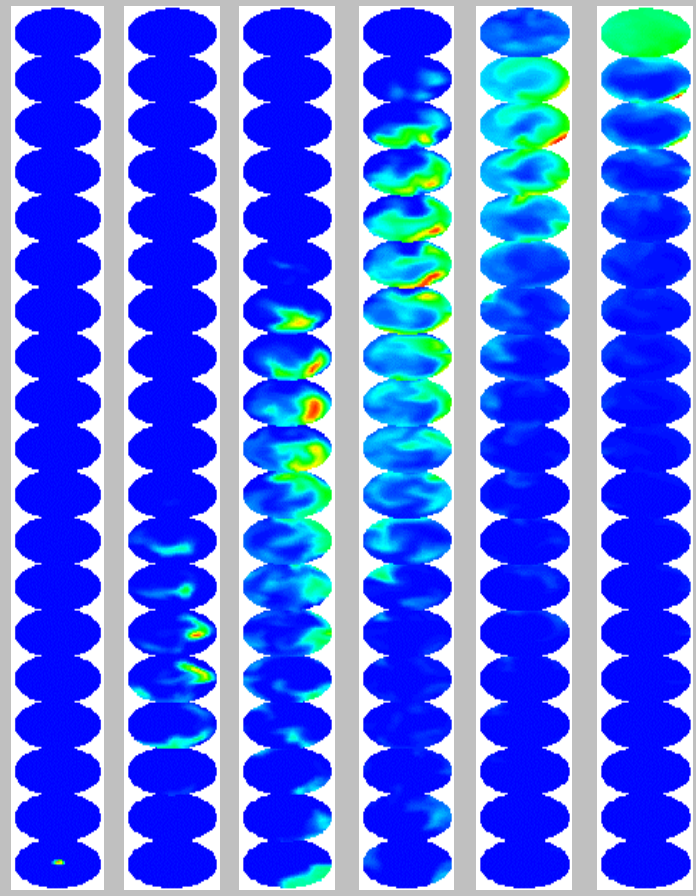
Time Evolution of the Liquid Tracer Concentration
D = 14-cm; U_g = 2.4 cm/s



Time, sec: 0 2 4 6 9 19

Time Evolution of the Gas Tracer Concentration
D = 14-cm; U_g = 2.4 cm/s



Time, sec: 0 1 2 3 5 7

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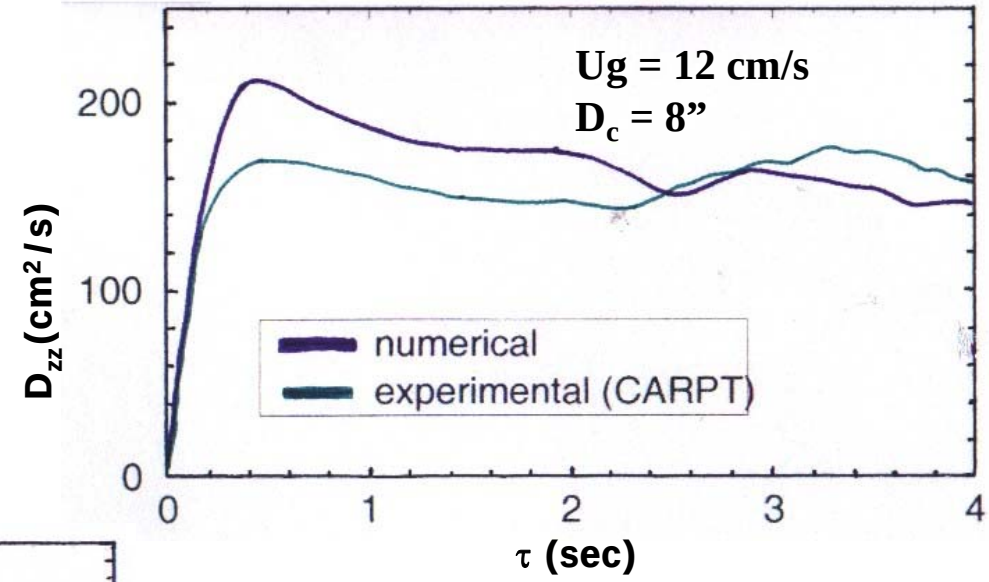
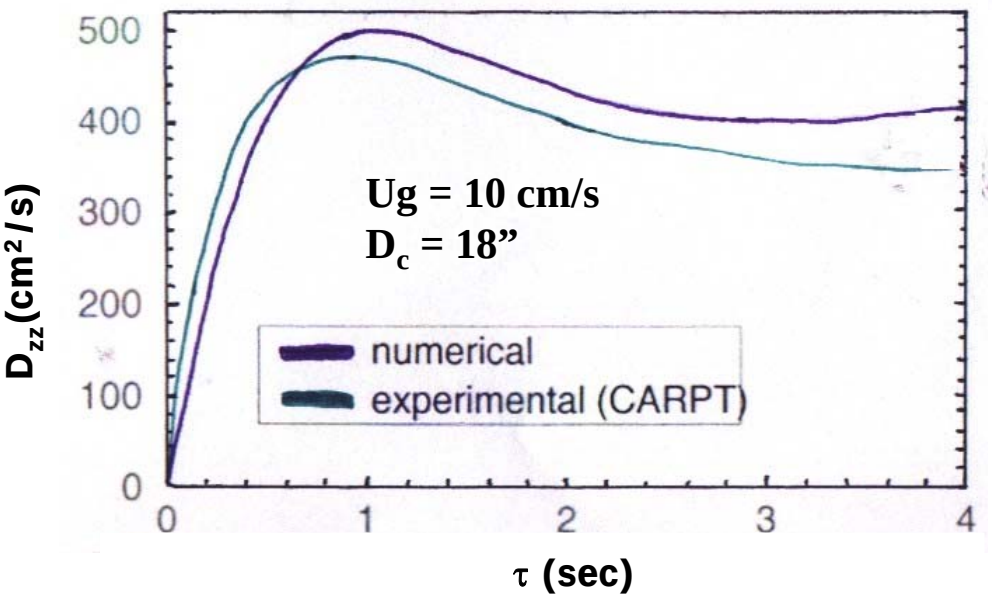
COMPARISON OF COMPUTED (CFDLIB) AND MEASURED D_{zz}





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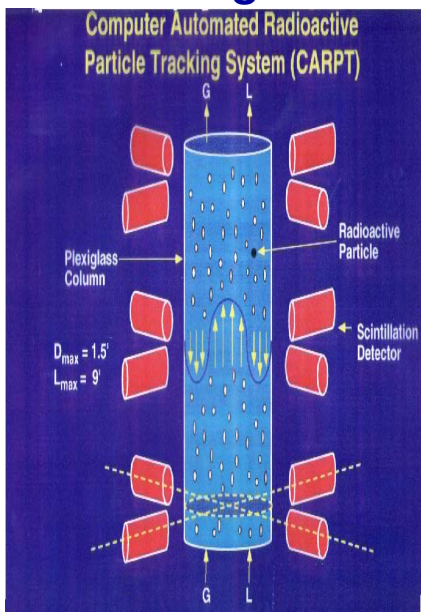
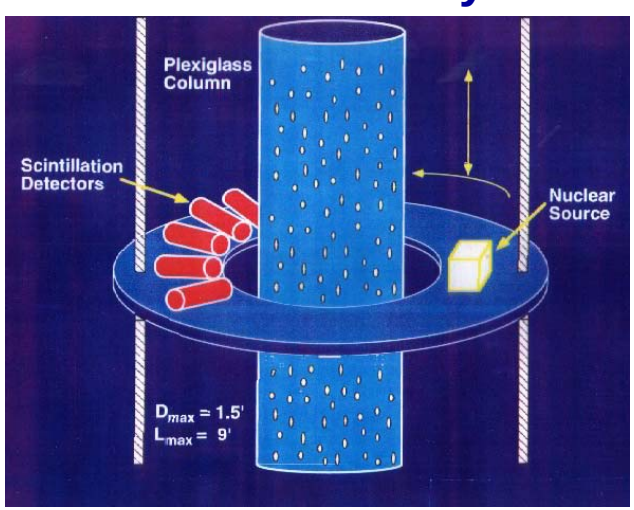
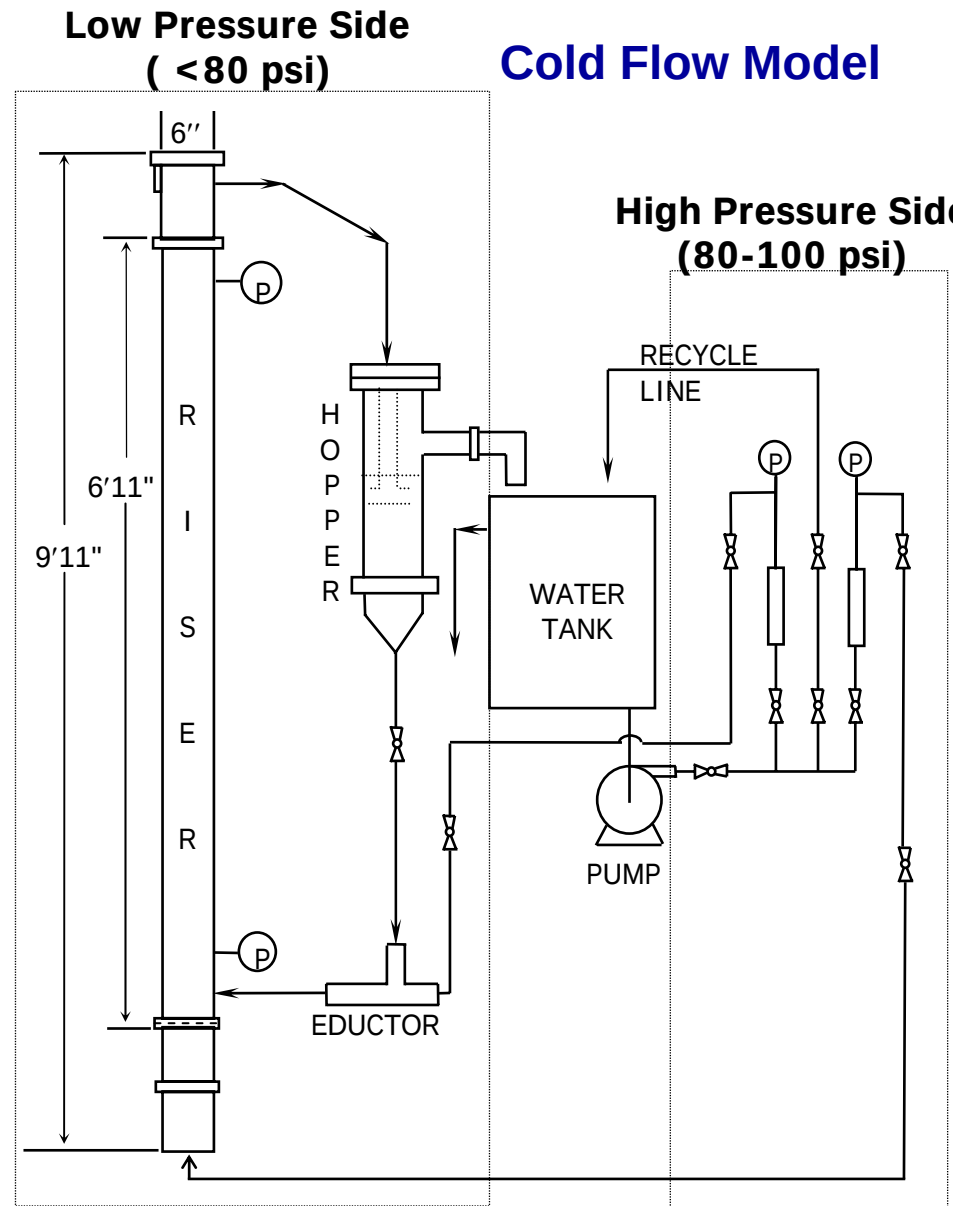
Radioactive Particle Tracking (CARPT) Provides Solids Velocity and Mixing Information



Computer Tomography (CT) Provides Solids Density Distribution



Cold Flow Model

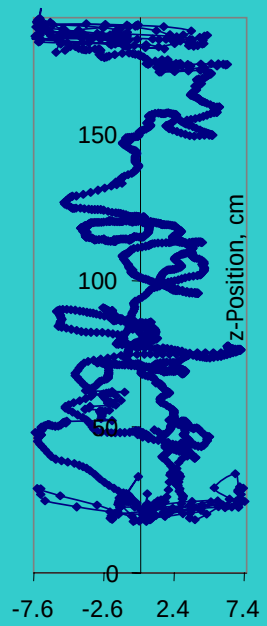


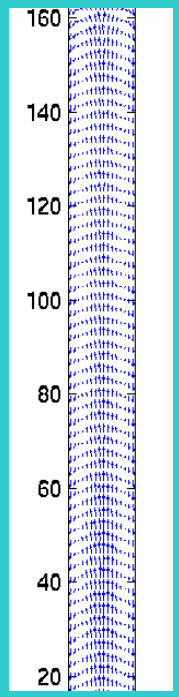
Tracer Studies Confirm Liquid In Plug Flow (N > 20)

$d_p = 2.5\ mm \quad \rho_p = 2.5\ g/cm^3 \quad L = 15, 20, 25\ cm/s \quad S/L = 0.1 \text{ to } 0.2$

(Devanathan, 1990; Kumar, 1994; Roy, 2000)

CARPT Results



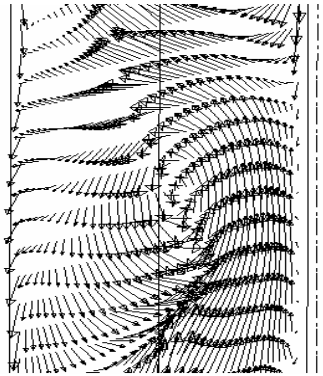


-505

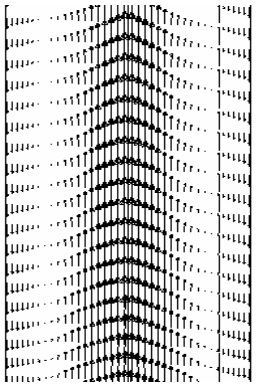
CFD Results

Trace over 38 s (1900 positions)

Z = 125 cm



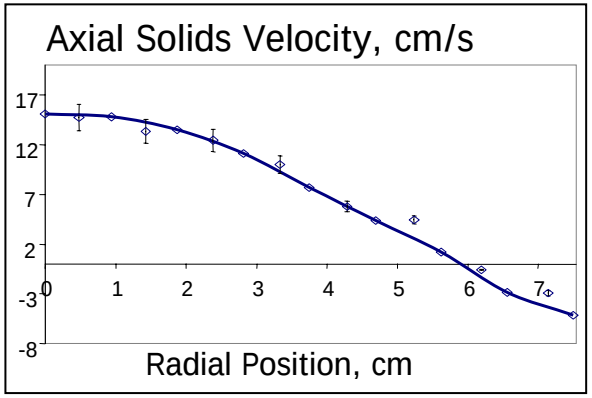
Z = 100 cm

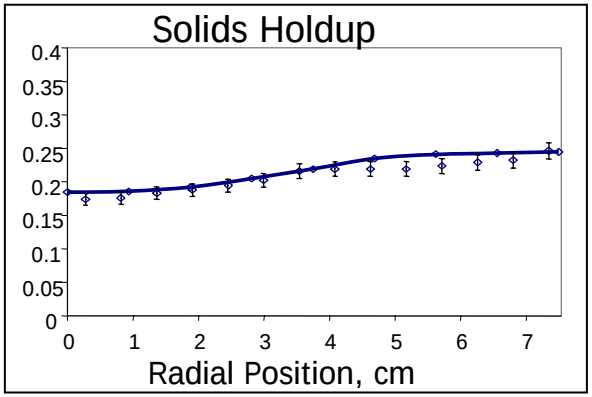


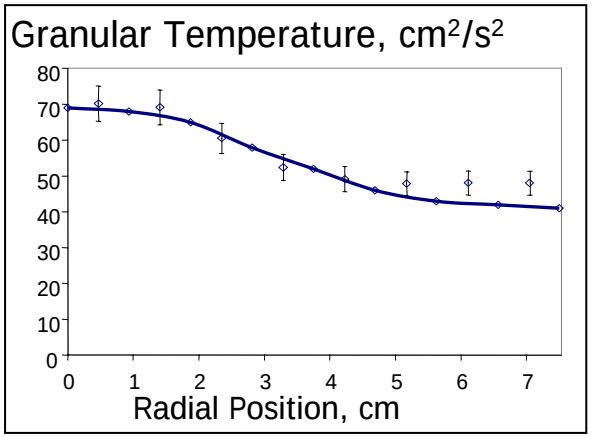
Time Average
(25 - 100 s)

t = 60 s

Comparison of CFD with Data

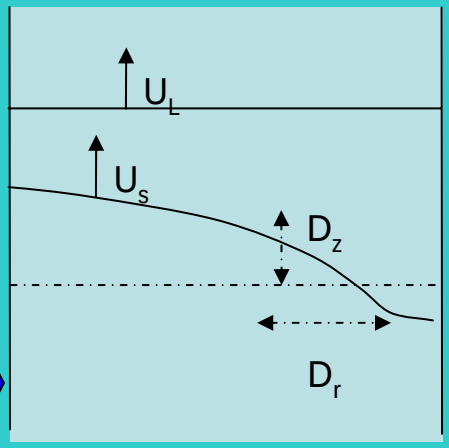






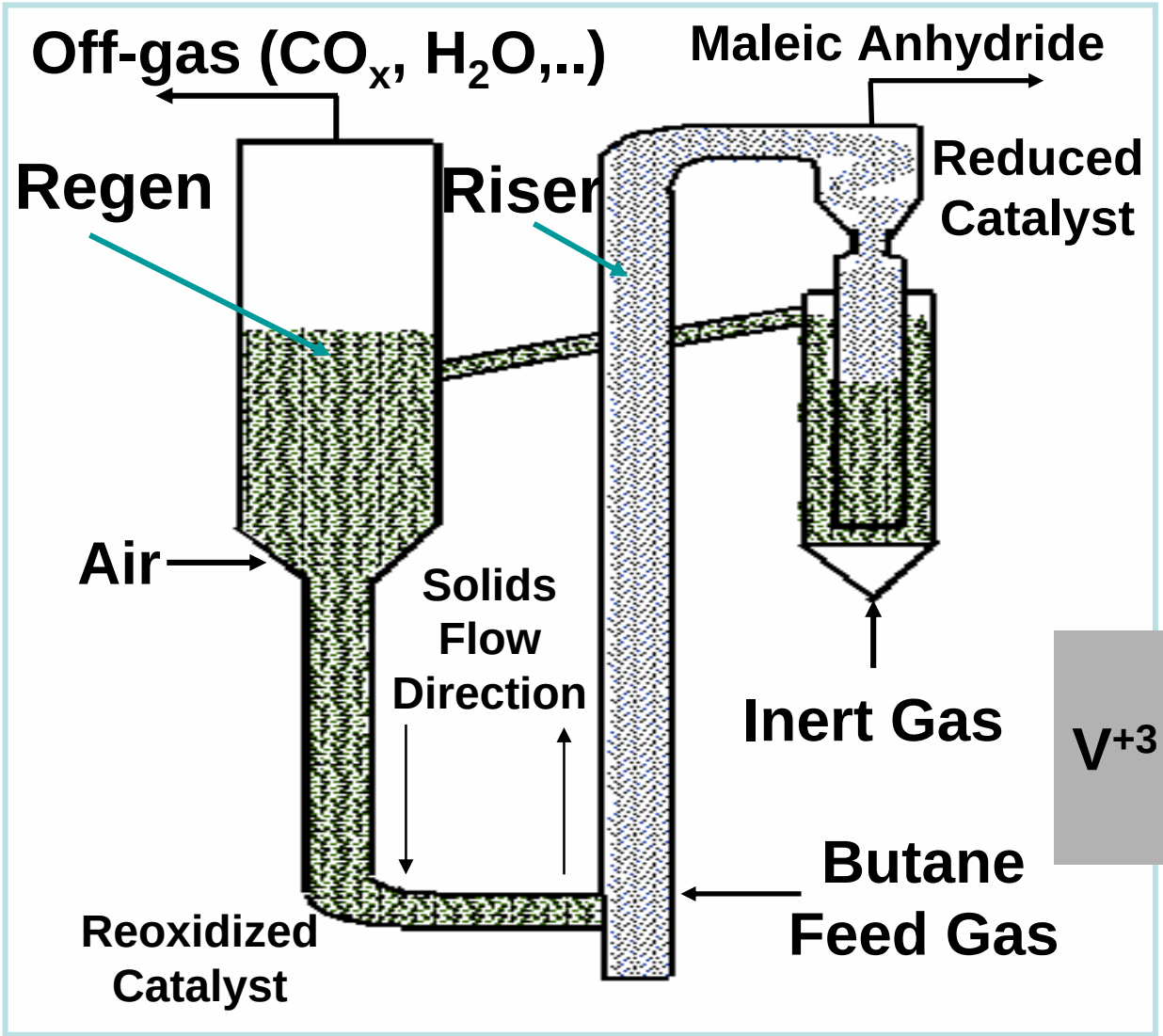
Roy and Dudukovic, I&EC Res., 40, 5440 (2001)

Final 2-D Convection Diffusion Reactor Model for the Riser

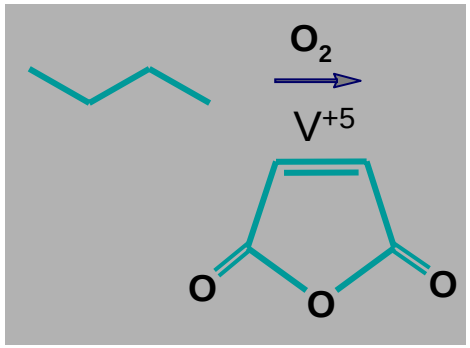


Ready for plant design, optimization and model based control

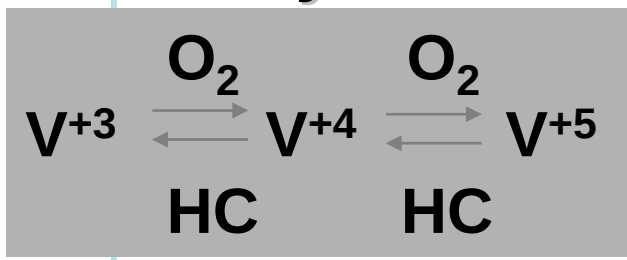
Circulating Fluid Bed (CFB) Reactor



Main Reaction



Catalyst Redox



Riser

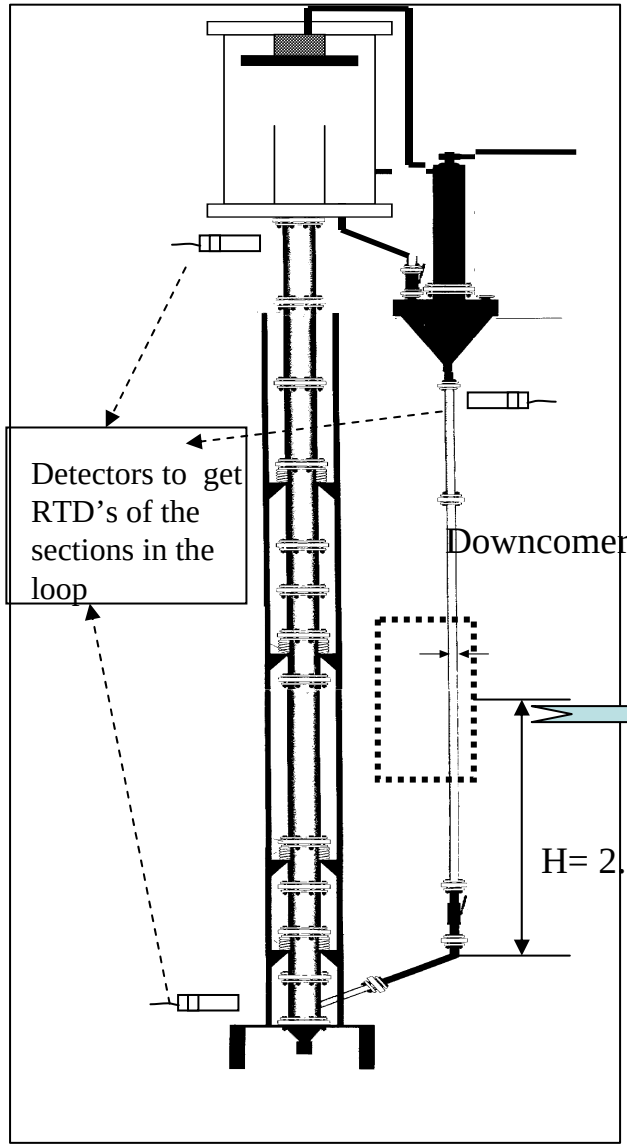
GAS-SOLID RISER

Gas-Solid Riser

Gas-Liquid or Liquid Fluidized Bed

CARPT Detectors on Gas-Solid Riser

OVERALL SOLIDS FLUX - TIME-OF-FLIGHT MEASUREMENTS



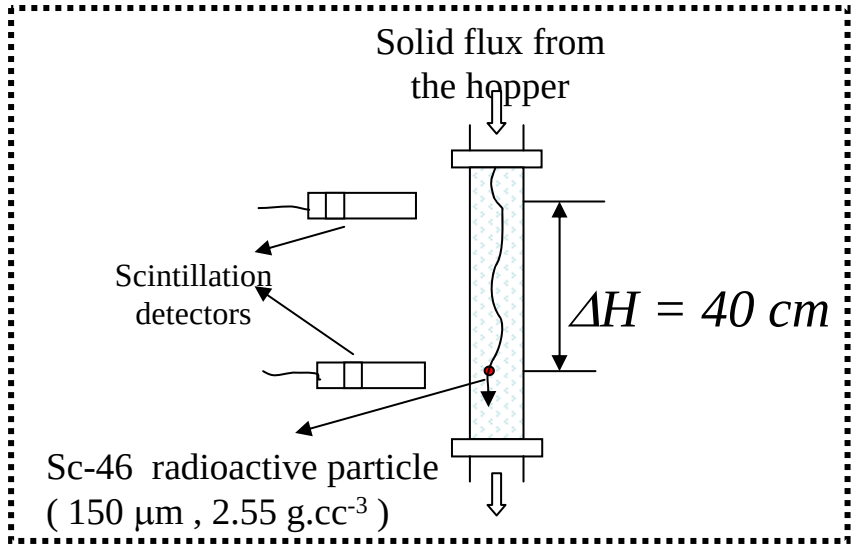
- Solids Mass Flux (G_s) in the downcomer is :

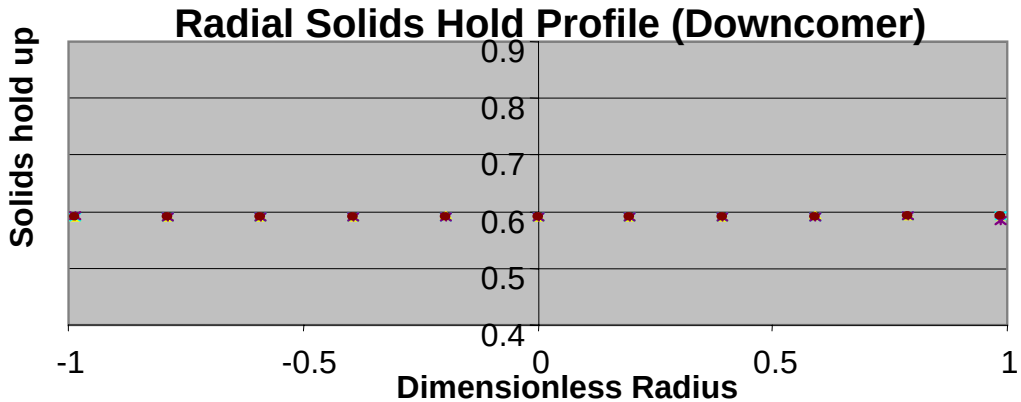
$$\langle G_s \rangle = \frac{\rho_s}{A}\left[\int_A \langle v_s \rangle \cdot \langle \varepsilon_s \rangle dA + \int_A \langle v_s' \varepsilon_s' \rangle dA \right] \approx \rho_s \cdot \langle v_s \rangle \cdot \langle \varepsilon_s \rangle$$

- Mean velocity can be calculated as

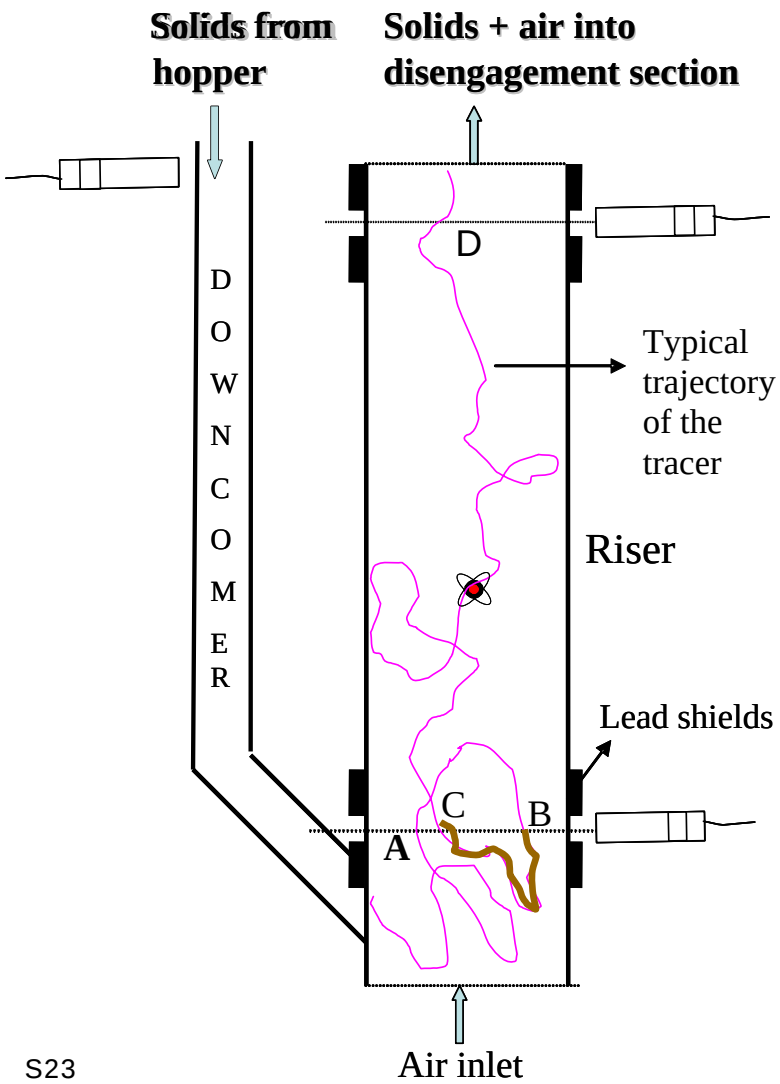
$$\langle v_s \rangle = \frac{\Delta H}{\langle t \rangle}$$

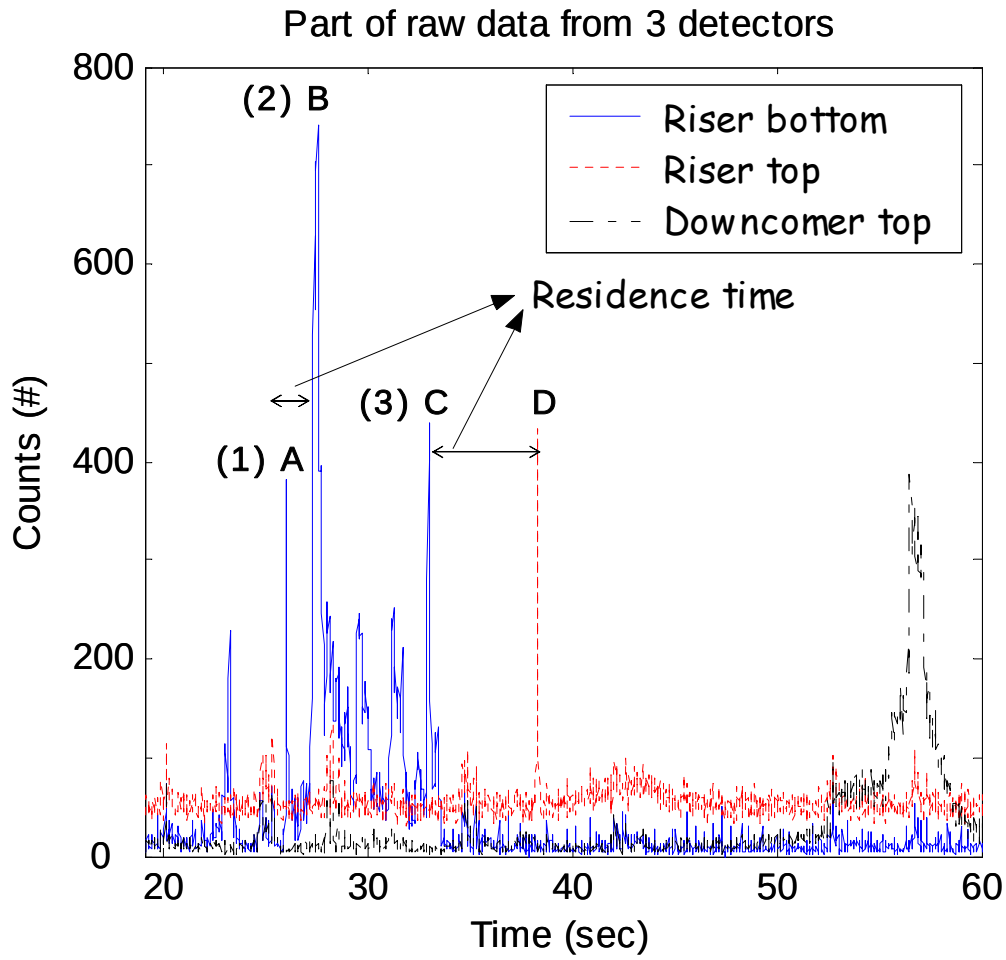
$\langle t \rangle$ average time of flight obtained for number of particle visits





Evaluation of Residence Time and First Passage Time Distributions from CARPT Experiments

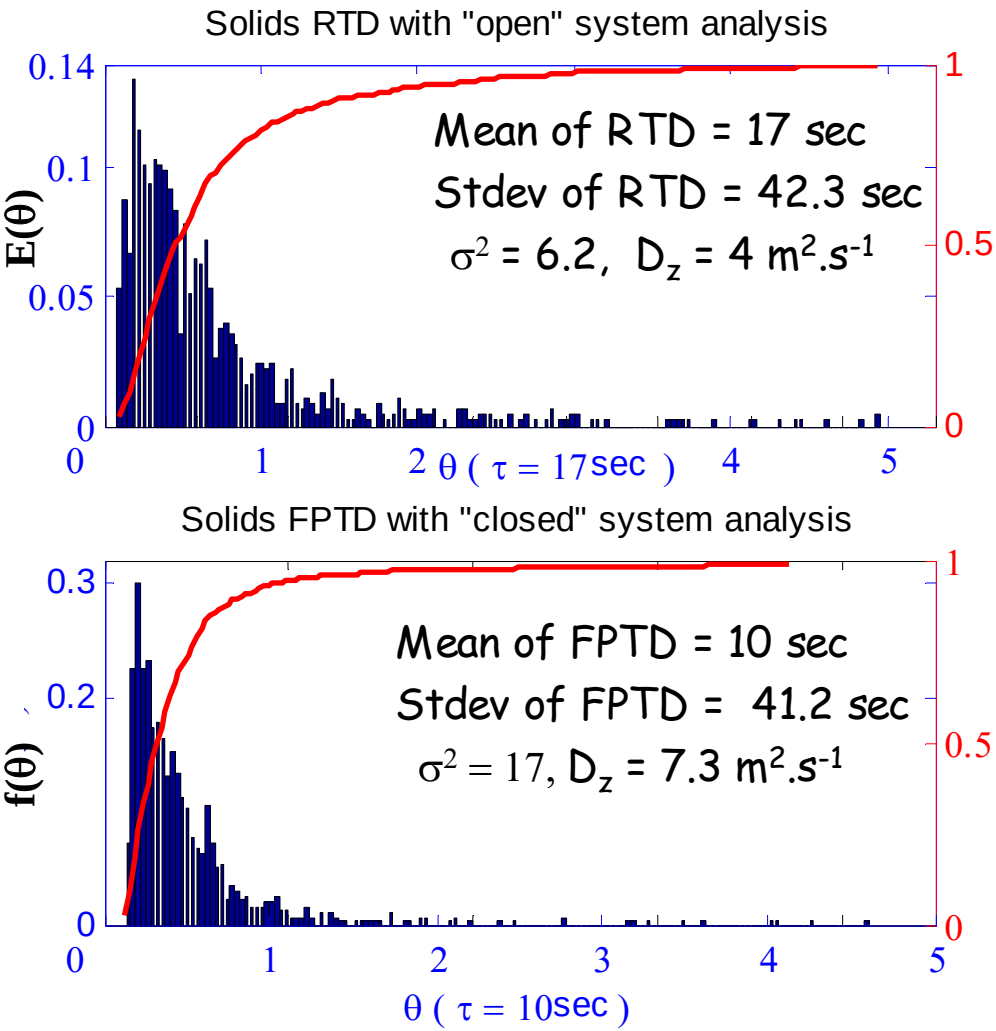




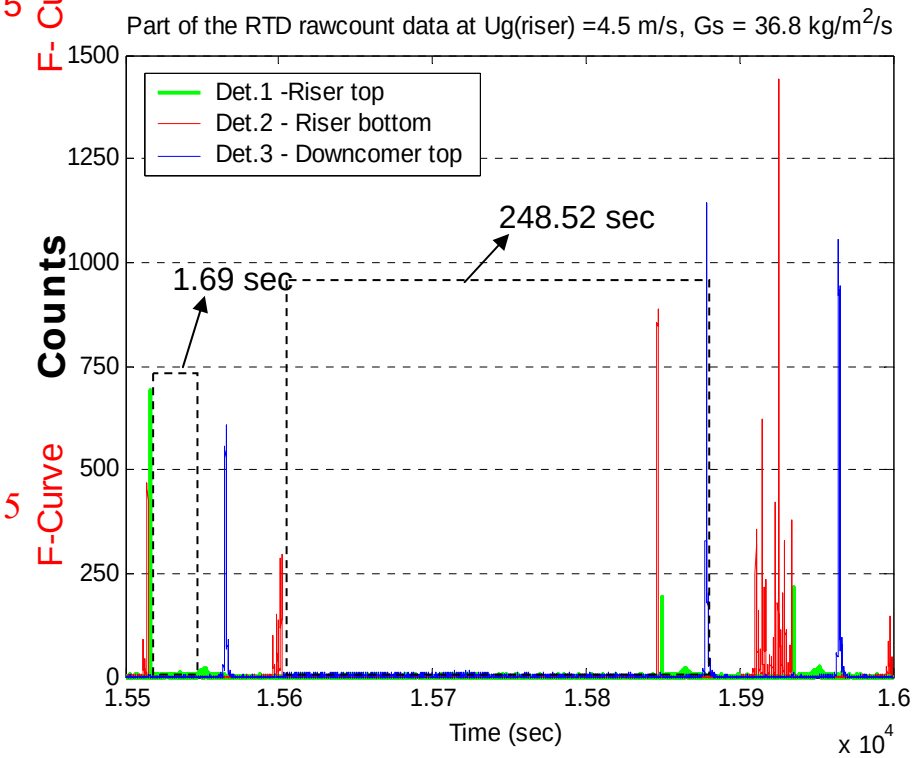
Time spent by the tracer between B-C should not be counted in the residence time

Solids RTD & FPTD Results – Dilute Phase Transport Regime

U_g^{riser} = 3.2 m.s^{-1} ; G_s = 33.7 kg.m^{-2}.s^{-1}; d_p = 150 μm; ρ_p = 2,550 kg/m^3



Very long solids internal recirculation



Bhusarapu (2004)

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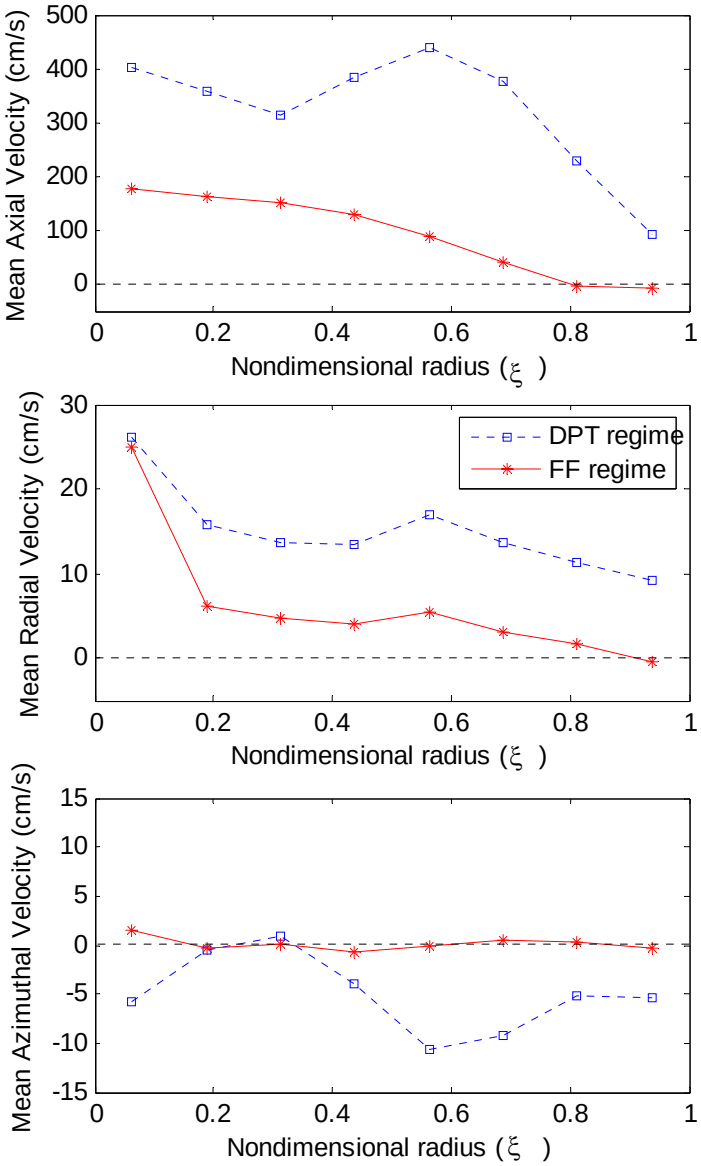
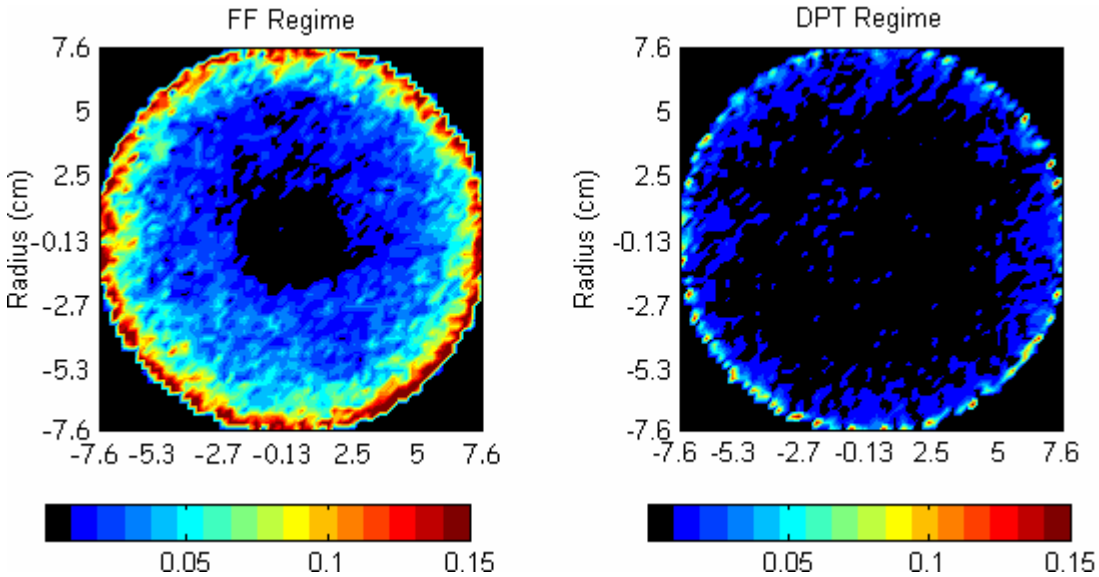
Mean Solids Velocity Field and Holdup – CARPT & CT



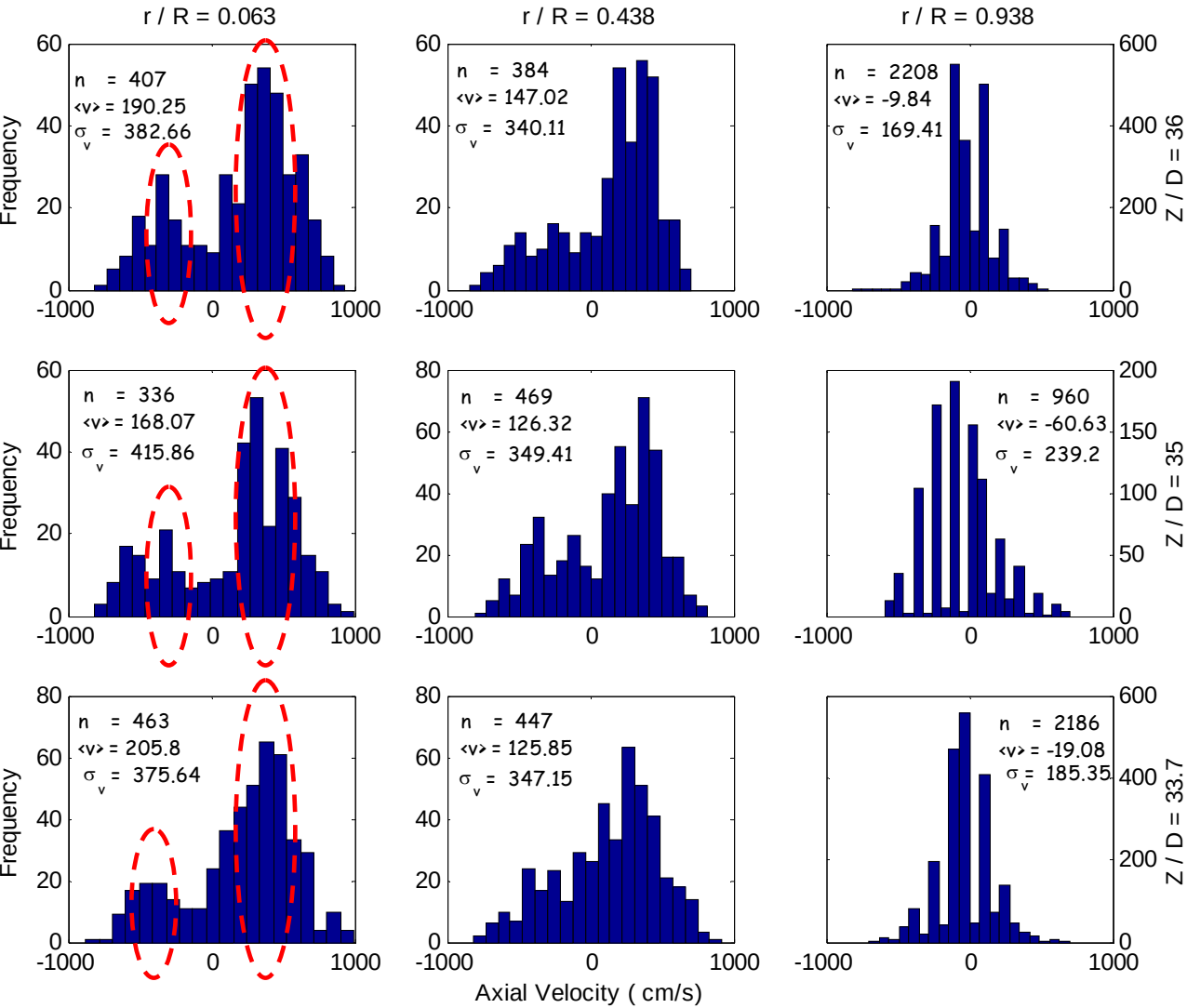
FF - U_g^{riser} = 3.2 m.s^{-1}; G_s = 26.6 kg.m^{-2}.s^{-1}

DPT - U_g^{riser} = 4.5 m.s^{-1}; G_s = 36.8 kg.m^{-2}.s^{-1}

Solids Holdup Tomograms



Axial Velocity PDFs – Spatial Variation, FF Regime



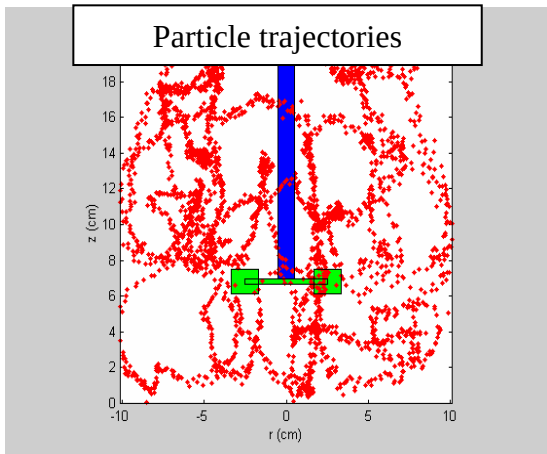
n - Particle Occurences in the voxel(#); <v> - Mean axial velocity(cm/s); σ_v - Standard deviation of axial velocity (cm/s)

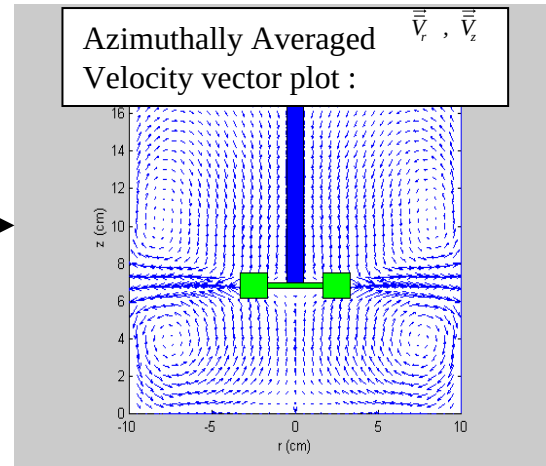
$U_g^{riser} = 3.2\ m.s^{-1}$

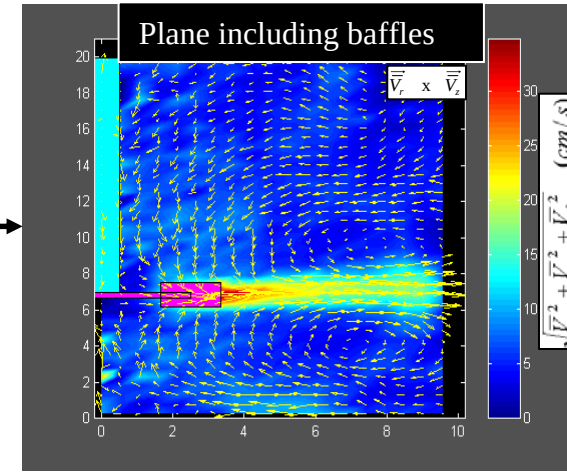
$G_s = 26.6\ kg.m^{-2}.s^{-1}$

Axial Velocity –

- Large radial gradients
- Negative near the wall
- Little axial variation in the zone
- In the core (near center) seems to have two prominent velocities
  - negative (downflow)
  - positive (upflow)

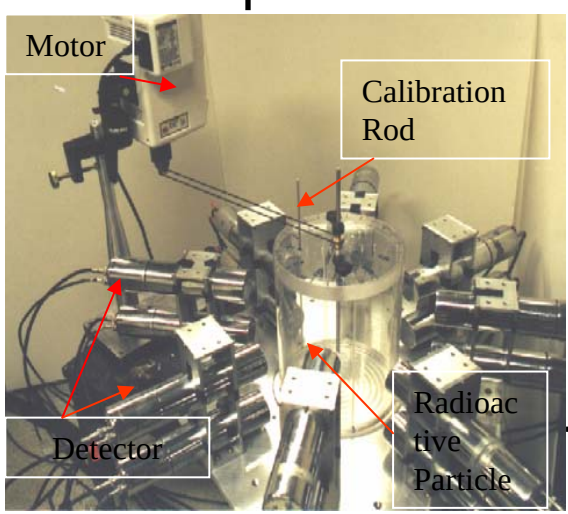


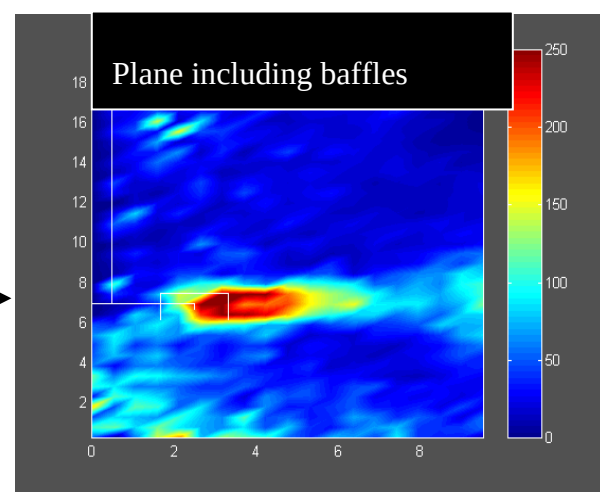


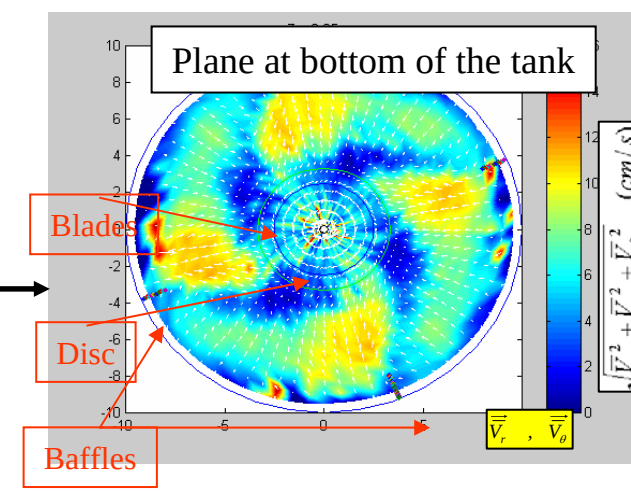


Single phase flow in STR :

results at a glance



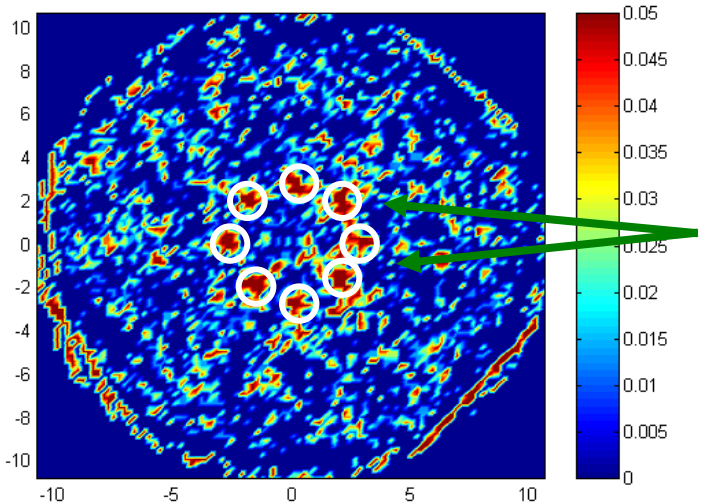




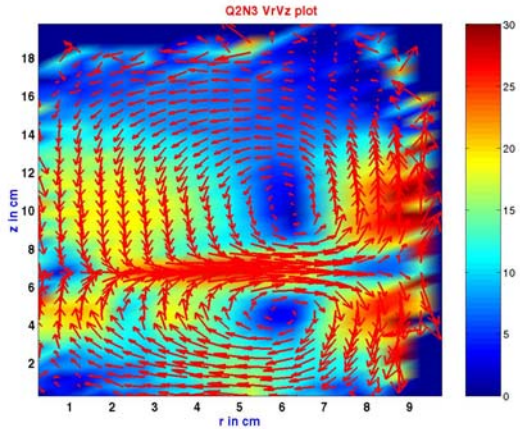
Rammohan et al., Chem. Eng. Research & Design (2001), 79(18), 831-844.

Two phase flow in STR :

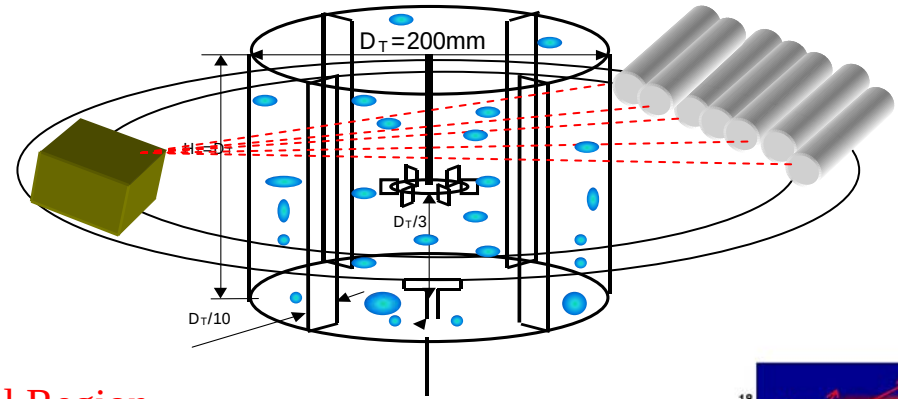
results at a glance



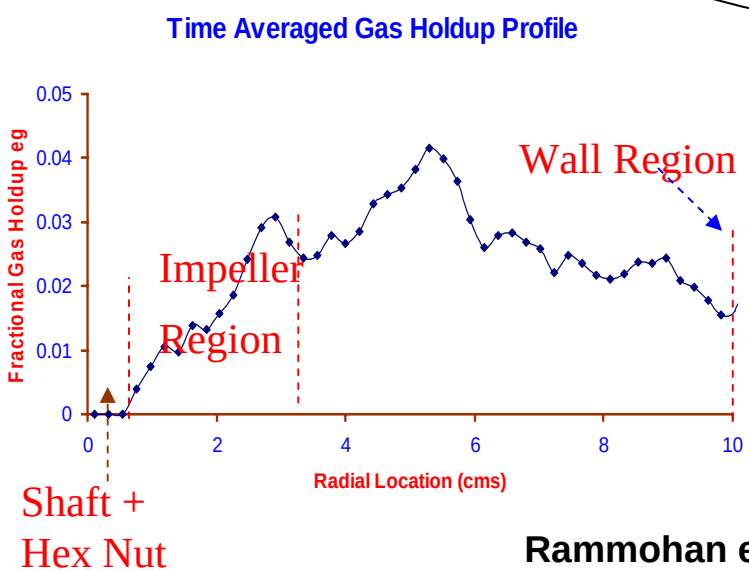
Gas Jets from Sparger with 8 holes

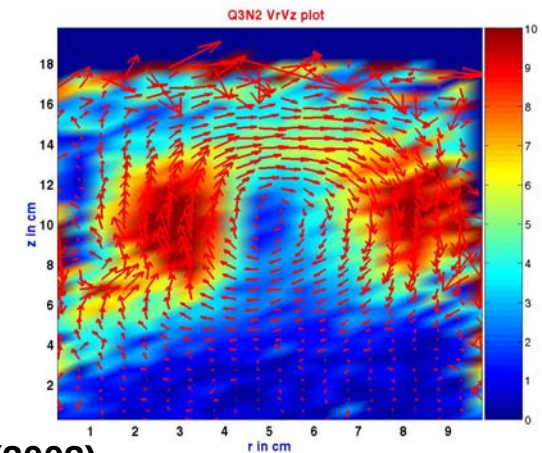


CT



CARPT





Rammohan et al. I&EC Research, 42, 2589 (2003)

Liquid Distribution in Study Trickle Bed Reactors - Experimental Setup

- **Exit Liquid Distribution**

DISTRIBUTOR

Single point
10mm diameter
liquid inlet

PACKING

6'' (14 cm)
Column

1<L/D<4

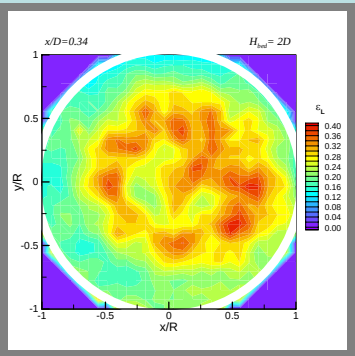
3mm diameter
glass beads

COLLECTING TRAY

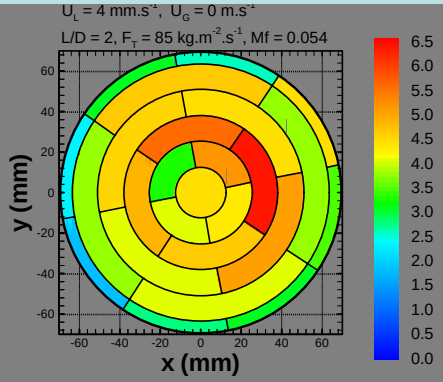
25 cross
sectional areas

- **Computed Tomography**

- **Computed Tomography Liquid holdup**



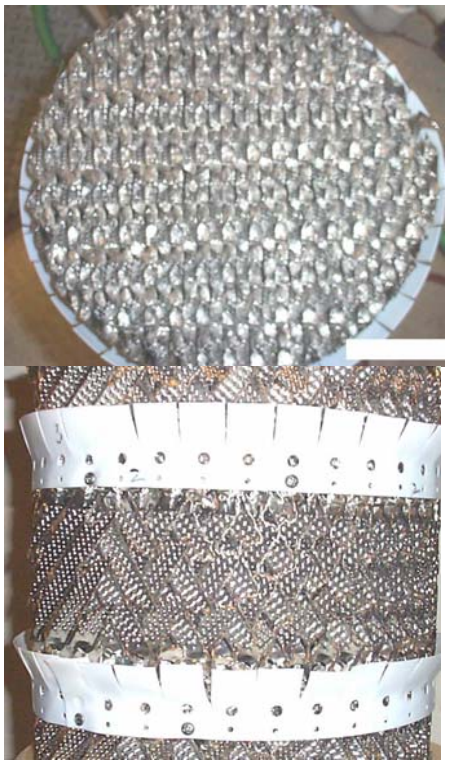
- **Exit Liquid Distribution**



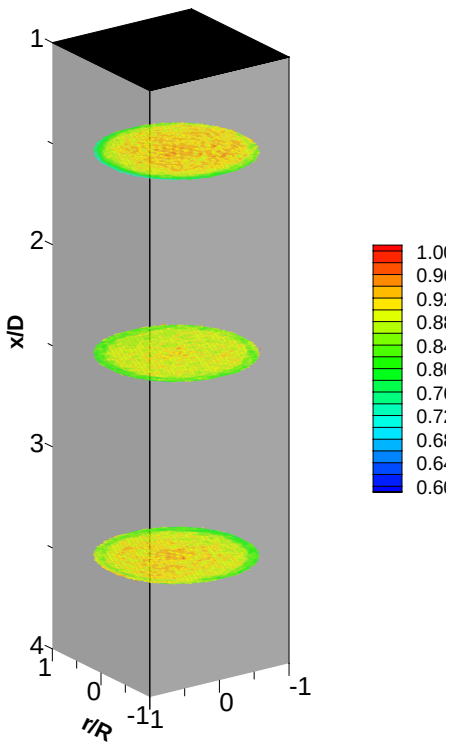
Cross-sectional liquid holdup and exit liquid distribution are compared in the region close to the reactor bottom. Figures show that results are in good qualitative agreement even though two different parameters (i.e. liquid holdup and exit liquid fluxes) are compared.

CT in Structured Packing under Counter-current flow

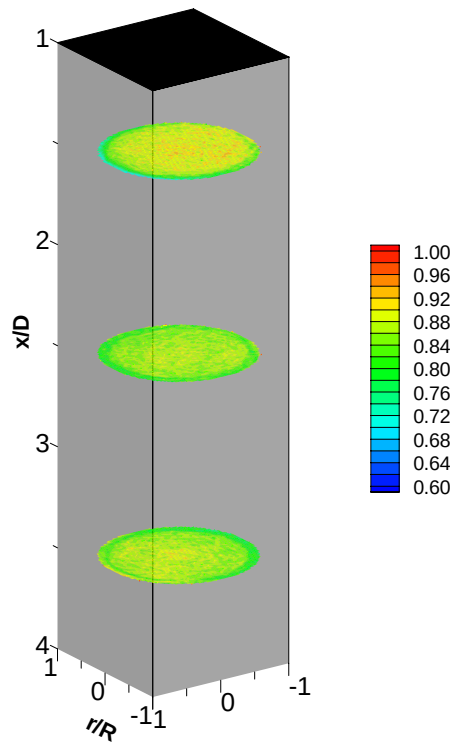
Results at a glance



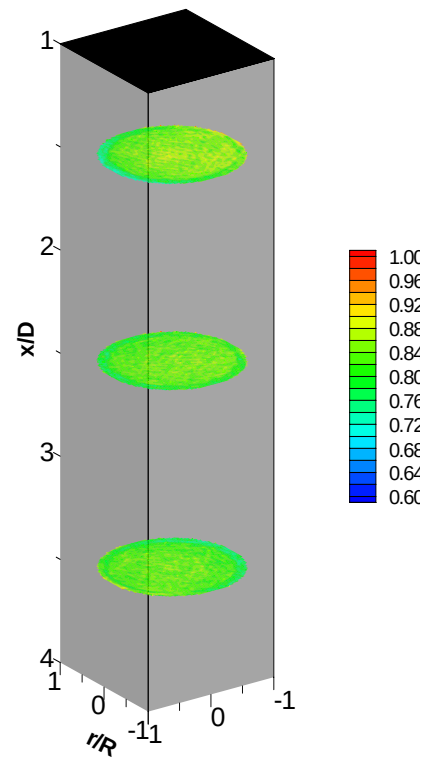
Corrugated Structured Packing



$V_l = 0.63$



$V_l = 1.3$ cm/sec



$V_l = 2.17$

Gas holdup profiles at ZERO gas flow in a 12 inch column

Conclusions

- Development of fundamentally based phenomenological models for reactors with two (three) moving phases is possible (e.g. bubble columns, riser, stirred tank, etc.)
- CARPT-CT provide a unique tool for evaluation of holdup and velocity distribution in these systems and for validation of CFD codes.
- CFD codes based on Euler-Euler interpenetrating fluid model with appropriate closures, upon validation, provide the means for effective calculation of reactor flow and mixing parameters.
- Phenomenological reactor models are capable of predicting tracer impulse responses. Thus they can predict reactor performance for linear kinetics exactly.
- Radioactive techniques have a major role to play in such model development.

Acknowledgement of Financial Support and Effort in Advancing Multiphase Reaction Engineering and Establishing Unique CARPT/CT Technologies

Department of Energy:	**DE-FC22 95 95051** **DE-FG22 95 P 95512**
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CREL Colleagues and Graduate Students:	**M.H. Al-Dahhan, J. Chen, S. Degaleesan, N. Devanathan, P. Gupta, A. Kemoun, B.C. Ong, Y. Pan, N. Rados, S. Roy, A. Rammohan, Y. Jiang, M. Khadilkar** **Y. Jiang, A. Kemoun, B.C. Ong, Y. Pan, N. Rados, S. Roy**
Special Thanks to:	**B.A. Toseland, Air Products and Chemicals** **M. Chang, ExxonMobil** **J. Sanyal, FLUENT, USA** **B. Kashiwa, CFDLib, Los Alamos** **V. Ranade, NCL, Pune, India**

Chemical Reaction Engineering Laboratory (CREL)
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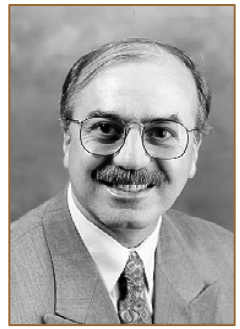
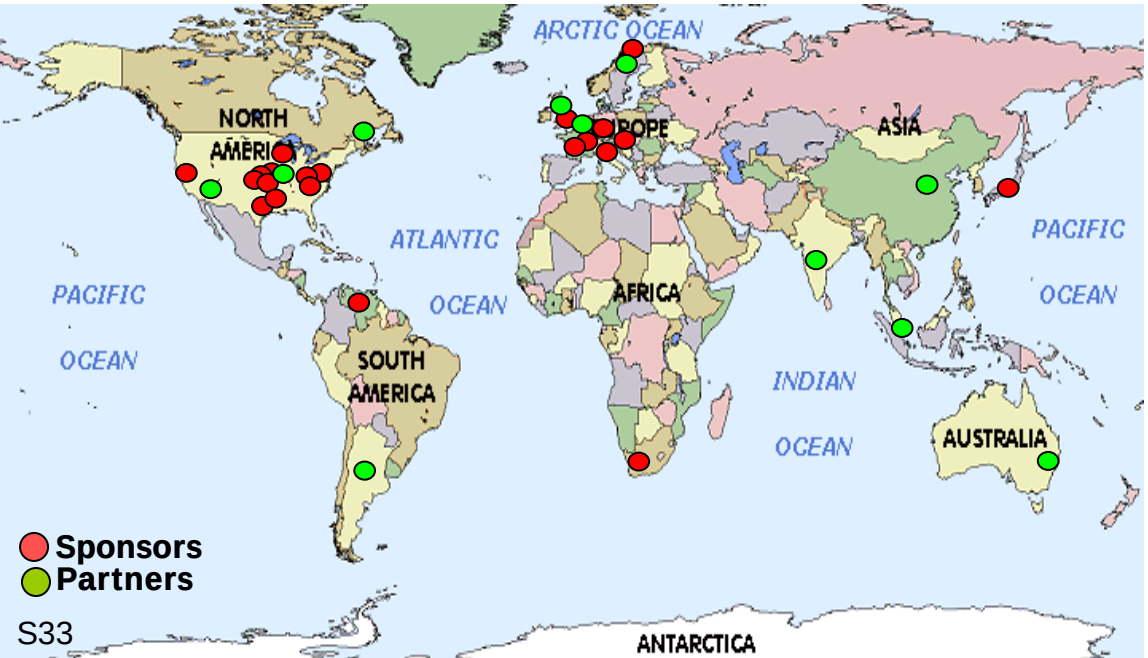
M. Dudukovic
Director



M. Al-Dahhan
Co-Director

Objectives

- Education and training of students
- Advancement of reaction engineering methodology
- Transfer of state-of-the-art reaction engineering to industrial practice



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