DIRECT NUMERICAL SIMULATION OF MASS, MOMENTUM AND HEAT TRANSPORT IN FIXED BED CHEMICAL REACTORS

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GROUP MULTIPHASE REACTORS
EINDHOVEN UNIVERSITY OF TECHNOLOGY

IMPROOF WORKSHOP ON CFD ASSISTED PROCESS INTENSIFICATION

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Technische Universiteit **Eindhoven** University of Technology

Where innovation starts

INDUSTRIAL APPLICATIONS OF FIXED BED REACTORS

some important processes

dehydrogenation of lower alkanes to corresponding alkenes

dehydrogenation of lower alcohols to corresponding aldehydes

partial oxidation reactions (o-xylene to phtalic anhydride)

steam reforming of methane (synthesis gas production)

ammonia synthesis

ammoxidation of propylene (acrylonitril)

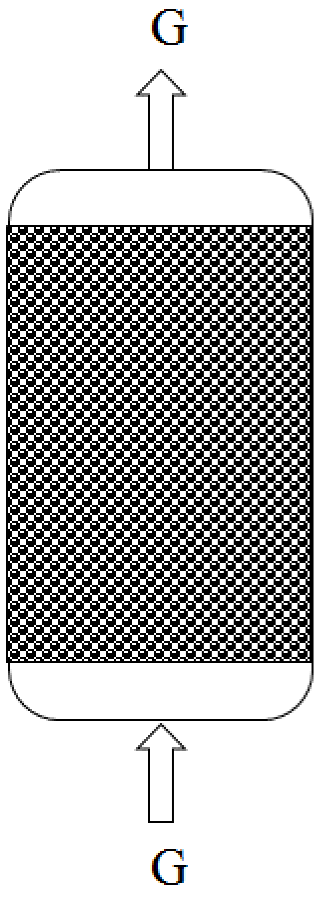
oxychlorination of ethylene (vinyl chloride)

alkylation of aromatic compounds (ethylbenzene)

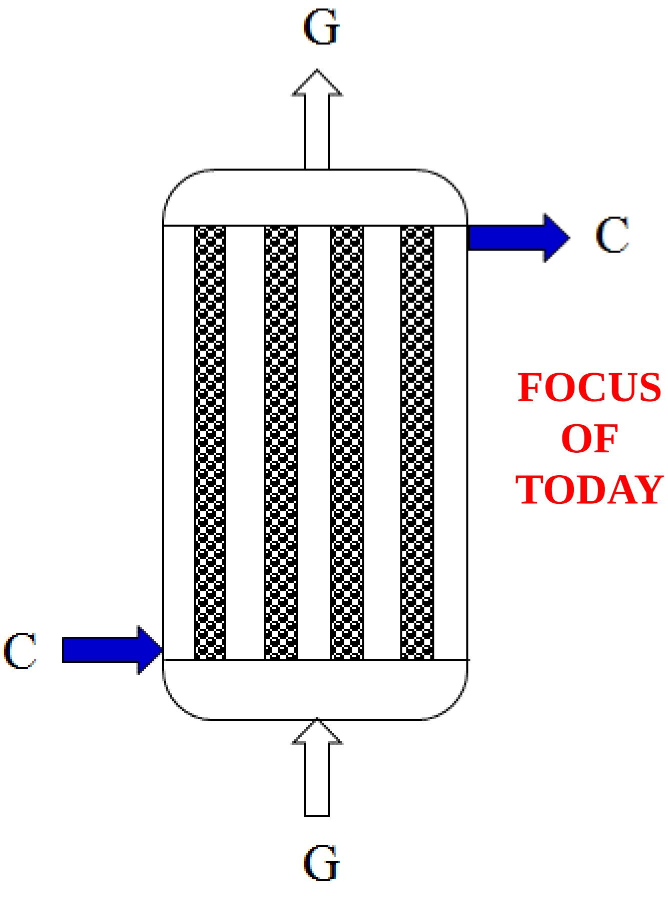
ammonia oxidation in nitric acid synthesis

INDUSTRIAL APPLICATIONS OF FIXED BED REACTORS

two main types of fixed bed reactors



single adiabatic fixed bed commonly used for mildly exothermic reactions



FOCUS OF TODAY

multi-tubular fixed bed commonly used for highly exothermic reactions

MULTI-TUBULAR FIXED BED REACTORS
characteristics

- TYPICAL PROPERTIES

+ commercial reactors contain up to 20000 parallel tubes !

+ tube diameter: 0.04 m

+ tube length: 4.0 m

+ catalyst particle diameter: 4-5 mm

+ ratio of tube diameter to catalyst particle diameter: 8-10 !!!

low ratio of tube diameter to particle diameter leads to considerable deviation of plug flow condition

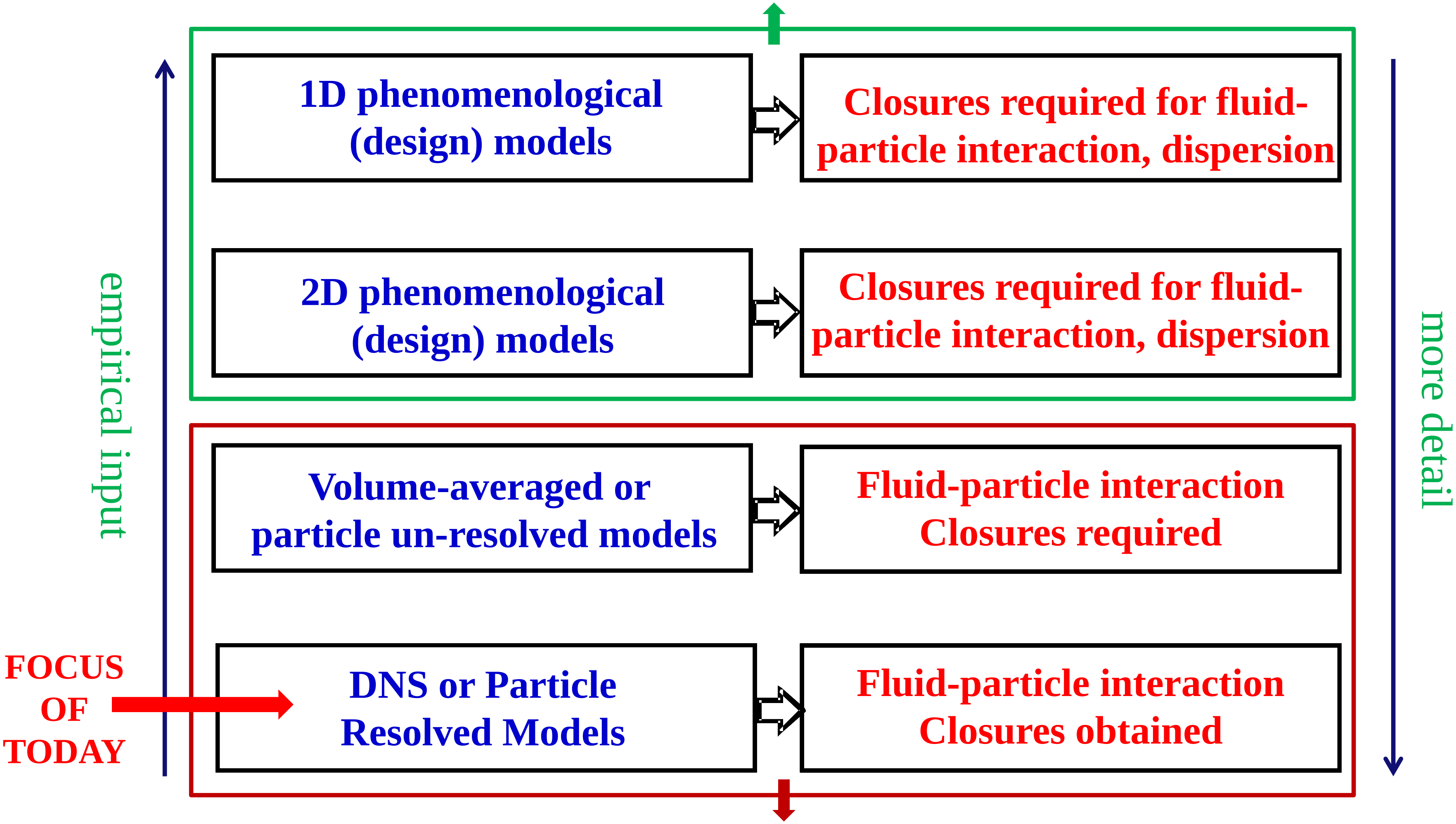
considerable effect on performance of multi-tubular fixed bed chemical reactor (conversion and selectivity)

even flow distribution over many parallel tubes is important and challenging to achieve

MODELLING OF FIXED BED REACTORS

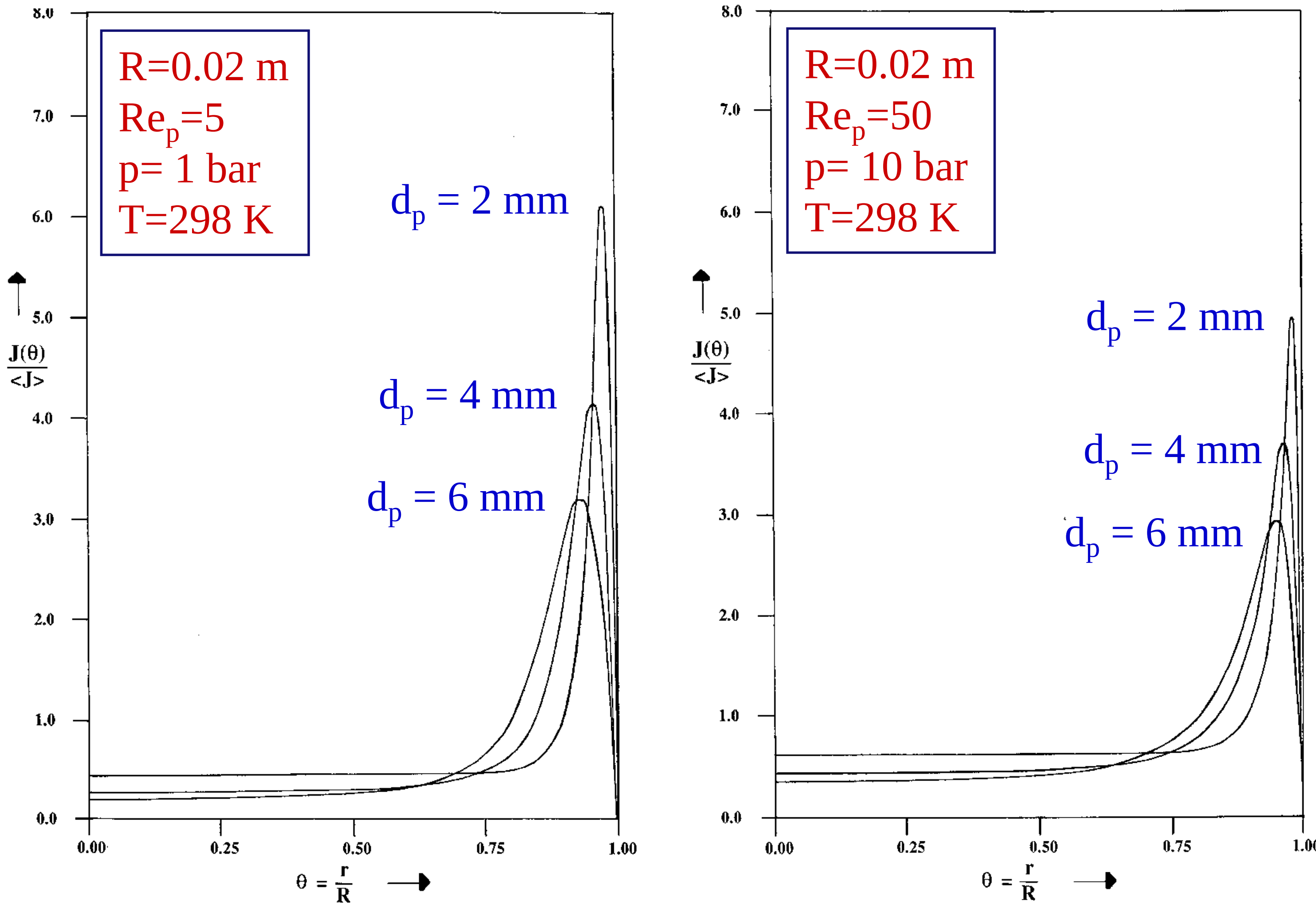
approaches

classical design models used in Chemical Reaction Engineering (CRE)



CFD models based on micro-balances for mass, momentum and heat

WALL-CHANNELING FROM VOLUME-AVERAGED MODEL
dimensionless radial profile of axial fluid mass flux



$$< J(\theta) >= 2\int_0^1 \theta J(\theta) d\theta \qquad J(\theta) = \varepsilon \rho_f u_z$$

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DNS BASED ON IMMERSED BOUNDARY METHOD (IBM)

- FEATURES

+ Eulerian grid + implicit boundary condition treatment at IB

- ADVANTAGES

+ all details of continuous phase flow field are captured
+ arbitrary shape of solid particles can be accounted for

- DISADVANTAGES

+ IBM simulations are CPU-demanding (especially in 3D)
+ limited to relatively small number of solid bodies (typically 10^3)

IBM BASED DNS MODEL
reactive systems

Main assumptions

constant physical properties of the fluid (gas) phase

first order exothermal chemical reaction
inside catalyst particles with uniform diameter

No reaction in the fluid (gas) phase

conjugate mass and heat transport

Arrhenius dependence of reaction rate constant

$$r_{A,s} = -kc_{A,s} = -k_0 \exp[-E_a / (RT_s)]c_{A,s}$$

radiative heat transport can be neglected

IBM BASED DNS MODEL
governing equations fluid phase

- CONTINUITY EQUATION

$$(\nabla \cdot \overline{u}) = 0$$

- MOMENTUM EQUATION

$$\rho\left[\frac{\partial}{\partial t}(\overline{u}) + (\nabla \cdot \overline{u}\overline{u})\right] = -\nabla p + \mu \nabla^2 \overline{u} + \rho \overline{g}$$

- THERMAL ENERGY EQUATION

$$\rho_f C_{p,f}\left[\frac{\partial T_f}{\partial t} + (\nabla \cdot \overline{u} T_f)\right] = \lambda_f \nabla^2 T_f$$

- SPECIES CONSERVATION EQUATION

$$\frac{\partial c_{A,f}}{\partial t} + (\nabla \cdot \overline{u} c_{A,f}) = D_{A,f} \nabla^2 c_{A,f} + r_{A,f}$$

IBM BASED DNS MODEL
governing equations “solids” or particle phase

- TRANSLATIONAL EQUATION OF MOTION

$$m_p \frac{d\overline{w}_p}{dt} = m_p \overline{g} + \overline{F}_{f \to s}$$

- ROTATIONAL EQUATION OF MOTION

$$I_p \frac{d\overline{\omega}_p}{dt} = \overline{T}_{f \to s}$$

equations of motion are only required for moving particles as encountered in gas-particle flows

- SPECIES AND THERMAL ENERGY EQUATIONS

$$\frac{\partial c_{A,s}}{\partial t} = D_{A,s} \nabla^2 c_{A,s} + r_{A,s}$$

$$\rho_s C_{p,s} \frac{\partial T_s}{\partial t} = \lambda_s \nabla^2 T_s + (-r_{A,s})(-\Delta H_r)$$

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IBM BASED DNS MODEL
closure equations

- FLUID-PARTICLE DRAG

$$\overline{F}_{f\to s} = -\iint_{S_p} (\tau_f \cdot \overline{n} + p\overline{n})dS \qquad \tau_f = -\mu\left((\nabla\overline{u}) + (\nabla\overline{u})^T\right)$$

separate evaluation of friction drag and pressure drag

- FLUID-PARTICLE TORQUE

$$\overline{T}_{f\to s} = -\iint_{S_p} (\overline{r} - \overline{r}_p) \times (\tau_f \cdot \overline{n} + p\overline{n})dS = -\iint_{S_p} (\overline{r} - \overline{r}_p) \times (\tau_f \cdot \overline{n})dS$$

- FLUID-PARTICLE HEAT AND MASS TRANSFER RATES

$$\Phi_{h,f\to s} = -\iint_{S_p} (\lambda_f \nabla T_f \cdot \overline{n})dS \qquad \Phi_{m,f\to s} = -\iint_{S_p} (D_{A,f} \nabla c_{A,f} \cdot \overline{n})dS$$

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IBM BASED DNS MODEL
numerical solution of fluid equations

- KEY FEATURES

+ explicit treatment of convection term

$$\left[\frac{|u_x|}{\Delta x} + \frac{|u_y|}{\Delta y} + \frac{|u_z|}{\Delta z} \right] \Delta t < 1$$

stability condition

+ implicit treatment of pressure gradient

+ implicit treatment of diffusion terms

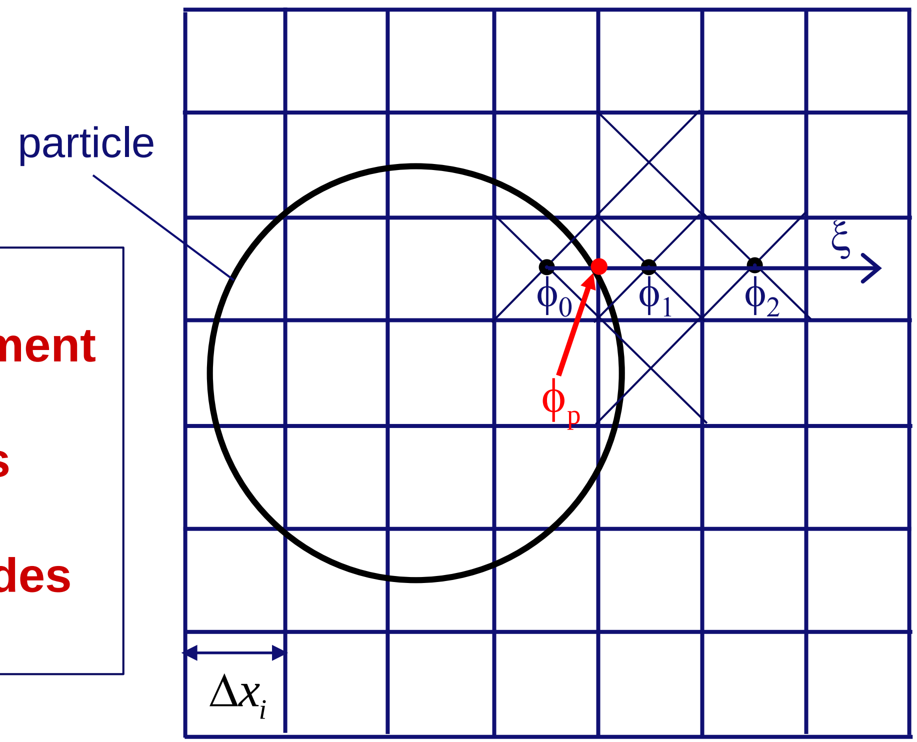
+ staggered computational mesh

+ sequential solution methodology

IBM BASED DNS MODEL
fluid-solid coupling for Dirichlet boundary conditions

- GENERIC FORM DISCRETE EQUATIONS FOR PROPERTY ϕ

$$a_c \phi_c = \sum_{nb} a_{nb} \phi_{nb} + b_c$$



$$\xi = \frac{x_i}{\Delta x_i}$$

Immersed Boundary treatment

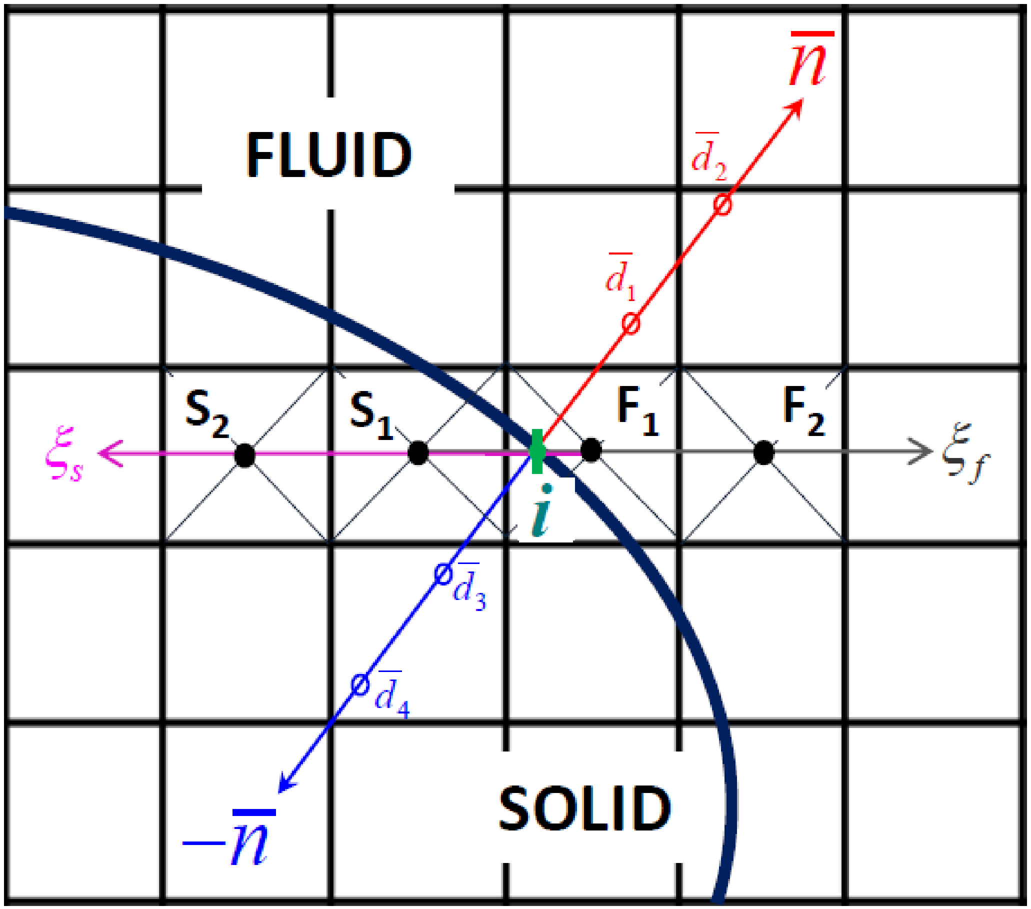
identify "solids" nodes

eliminate ϕ_0 at "solids" nodes

$$\phi_0 = -\frac{2\xi_s}{1-\xi_s}\phi_1 + \frac{\xi_s}{2-\xi_s}\phi_2 + \frac{2}{(1-\xi_s)(2-\xi_s)}\phi_p$$

value of ϕ at the immersed boundary

IBM BASED DNS MODEL
fluid-solid coupling with intra-particle scalar transport



thermal boundary conditions at the fluid-solid interface (similar for mass transport)

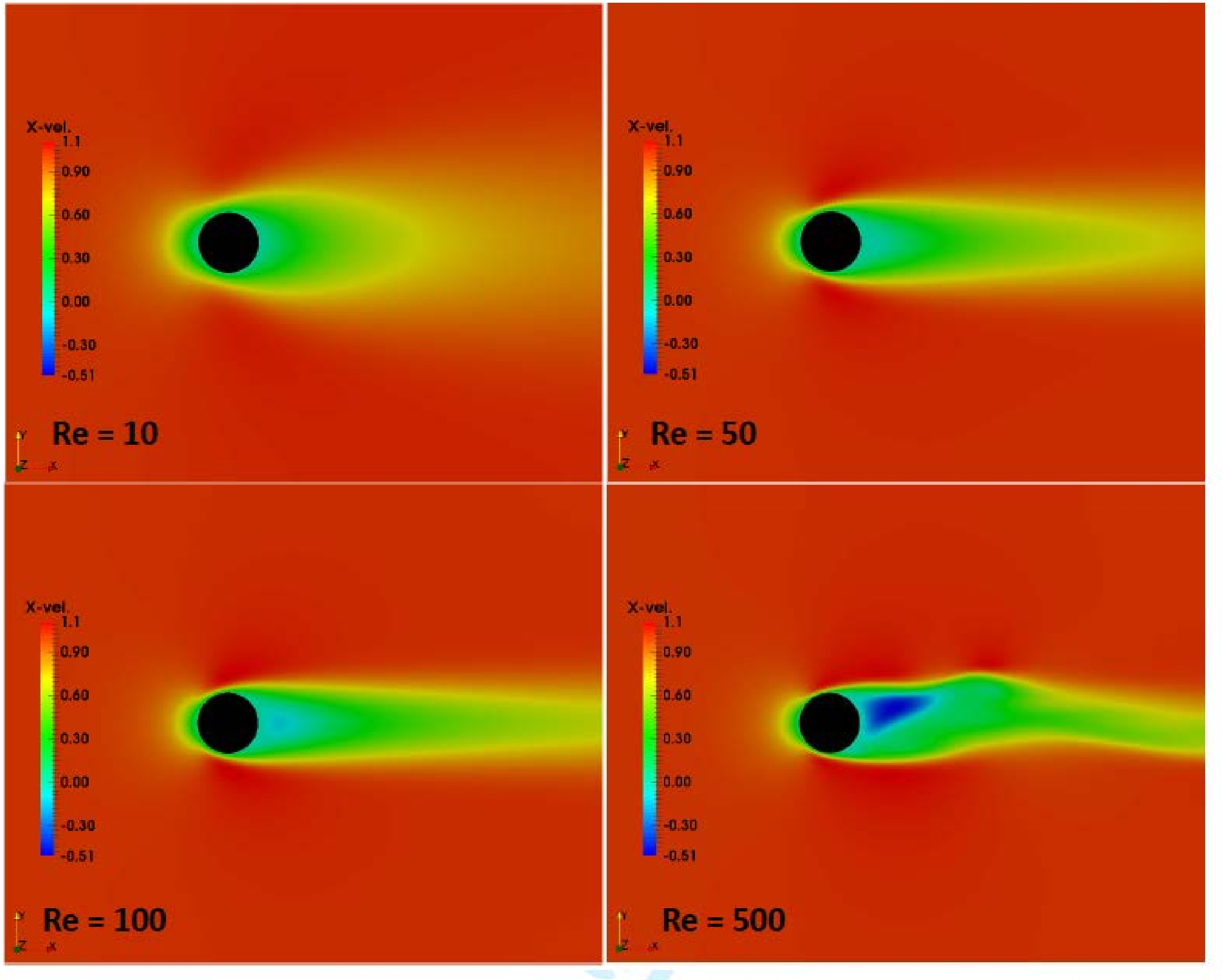
$$T_f = T_s$$

$$-\lambda_f \frac{\partial T_f}{\partial n} = -\lambda_s \frac{\partial T_s}{\partial n}$$

Das et al., 2017, CES

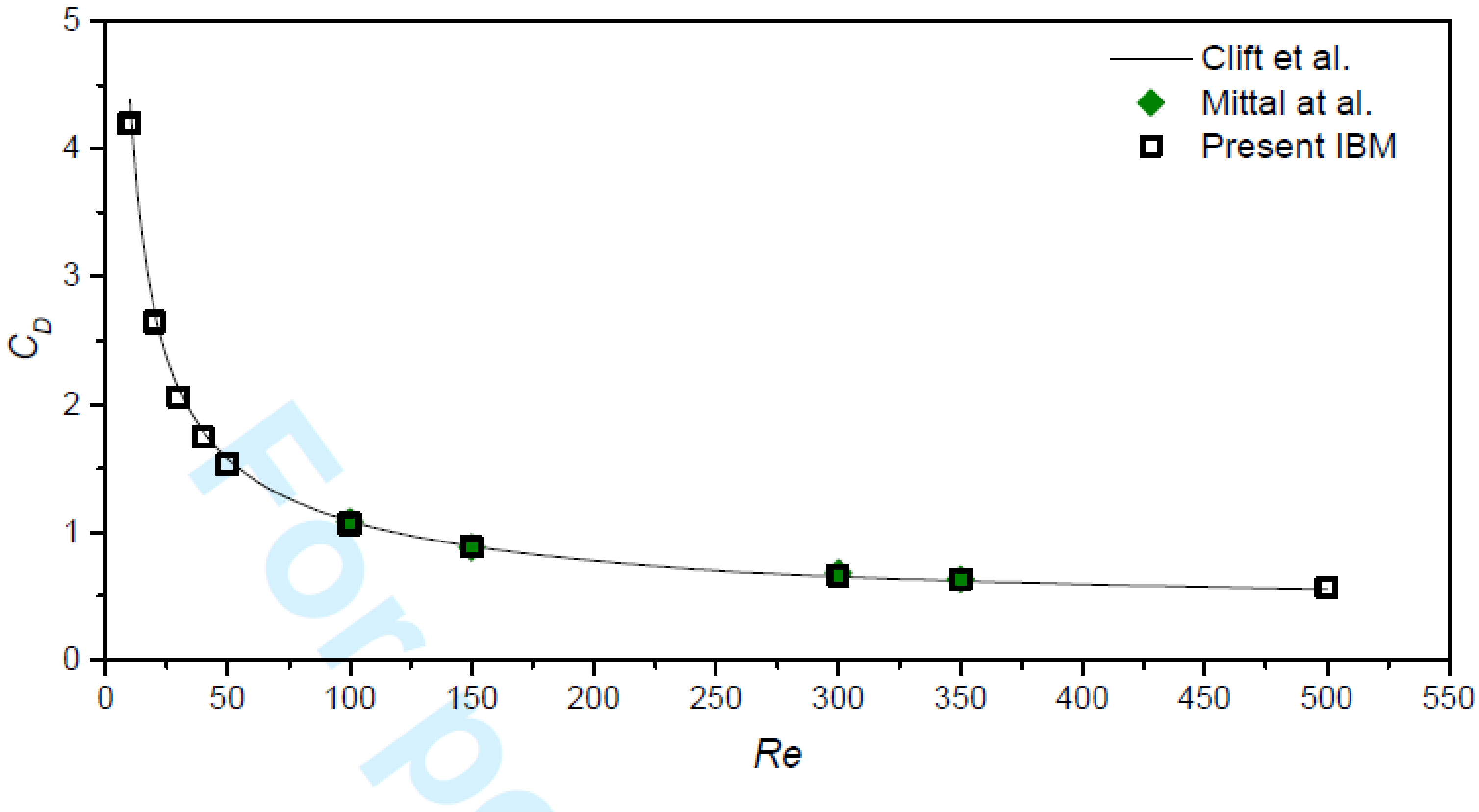
IBM BASED DNS MODEL

verification: flow past a stationary sphere



Das et al., 2016, AIChEJ

IBM BASED DNS MODEL
verification: flow past a stationary sphere



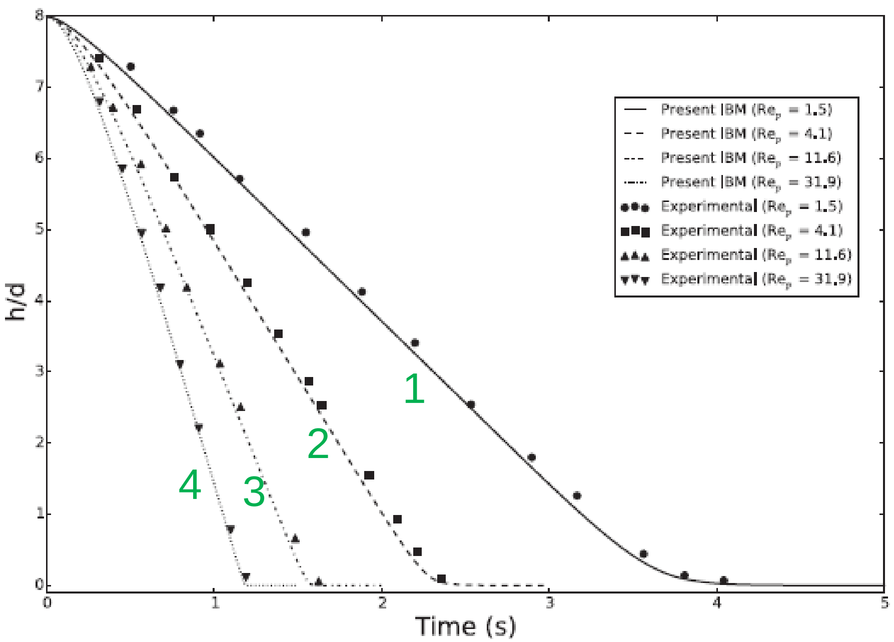
Das et al., 2016, AIChEJ

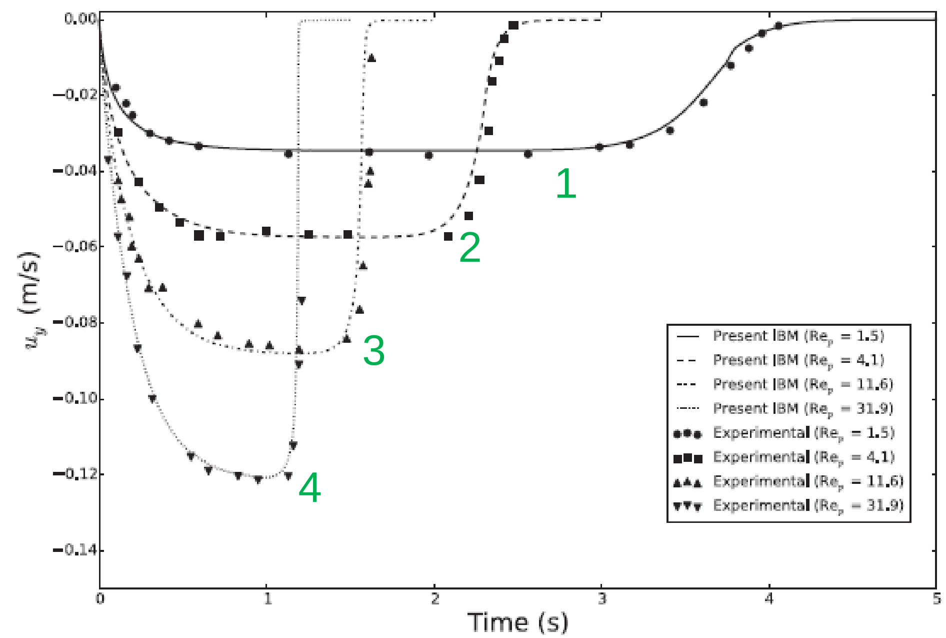
IBM BASED DNS MODEL
sedimentation of a single particle

Parameter	Case 1	Case 2	Case 3	Case 4
ρ_f	970	965	962	960
μ_f	0.373	0.212	0.113	0.058
$\mathbf{x}_p$	$(2.67D_p, 3.33D_p, 3.33D_p)$			
Ω	$10.67D_p \times 6.67D_p \times 6.67D_p$			
$\mathbf{g}$	$(9.81, 0, 0)$			
Re_p	1.5	4.1	11.6	31.9

Ten Cate et al. (exp., 2002)

$$\frac{D_p}{h} = 15.0$$

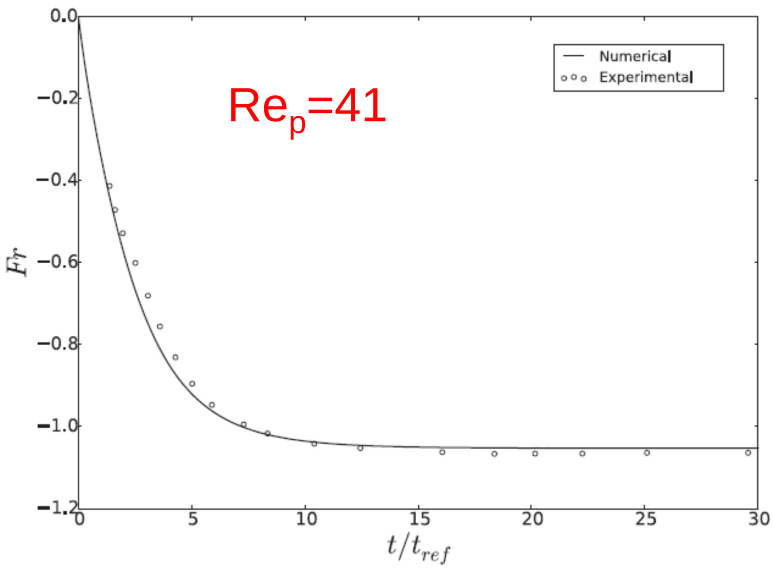


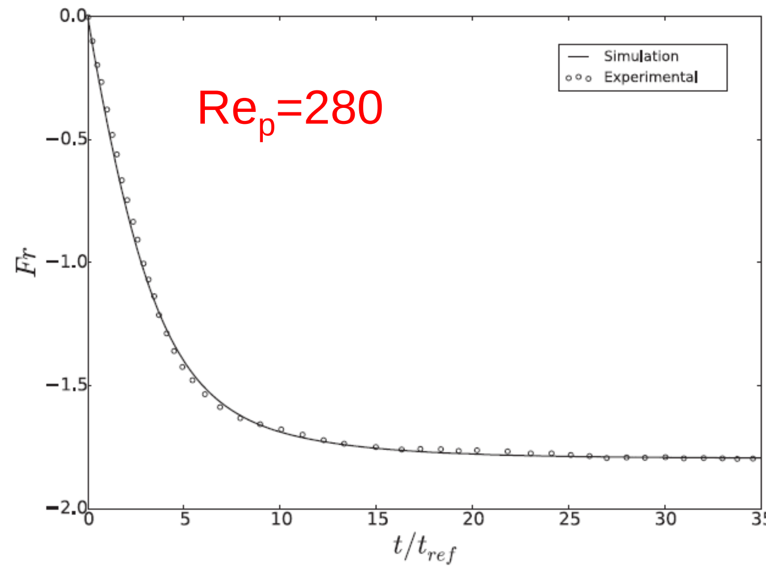


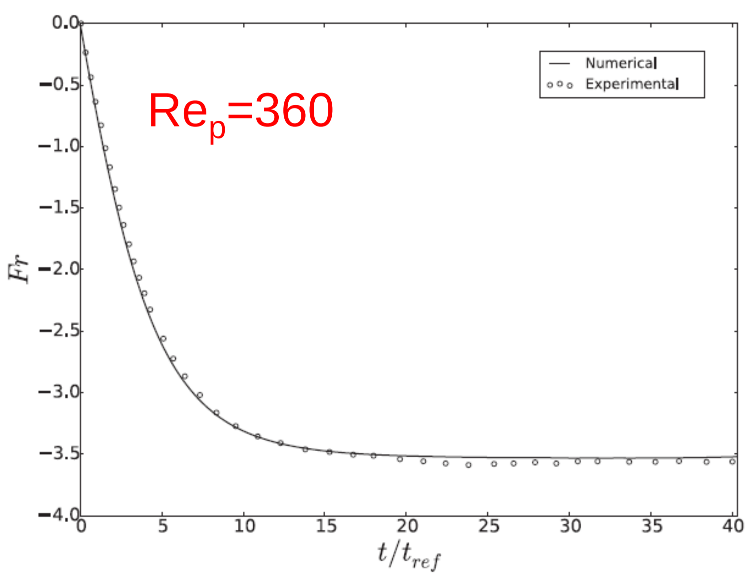
Maitri et al., 2018, CAF

IBM BASED DNS MODEL

sedimentation of a single particle







Parameter	Case 1	Case 2	Case 3
ν_f	0.00543	0.00268	0.00104
ρ_p/ρ_f	2.56	7.71	2.56
$\mathbf{g}$	(9.81, 0, 0)		

Mordant and Pinton (exp., 2000)

$$\frac{D_p}{h} = 16.7 \qquad Fr = \frac{u_p}{\sqrt{gD_p}}$$

Maitri et al., 2018, CAF

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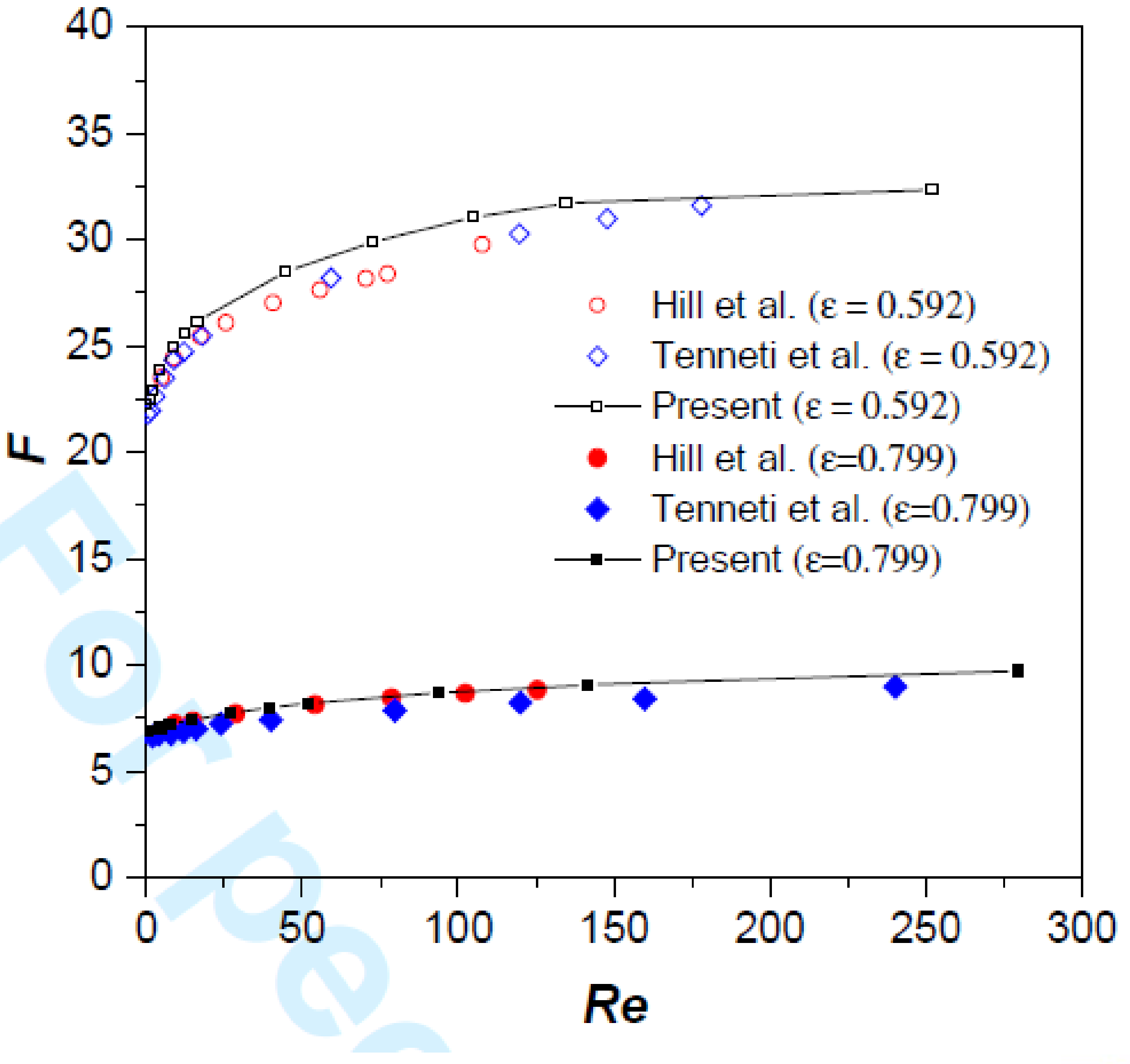
IBM BASED DNS MODEL

verification: flow past stationary arrays of spheres



Das et al., 2016, AIChEJ

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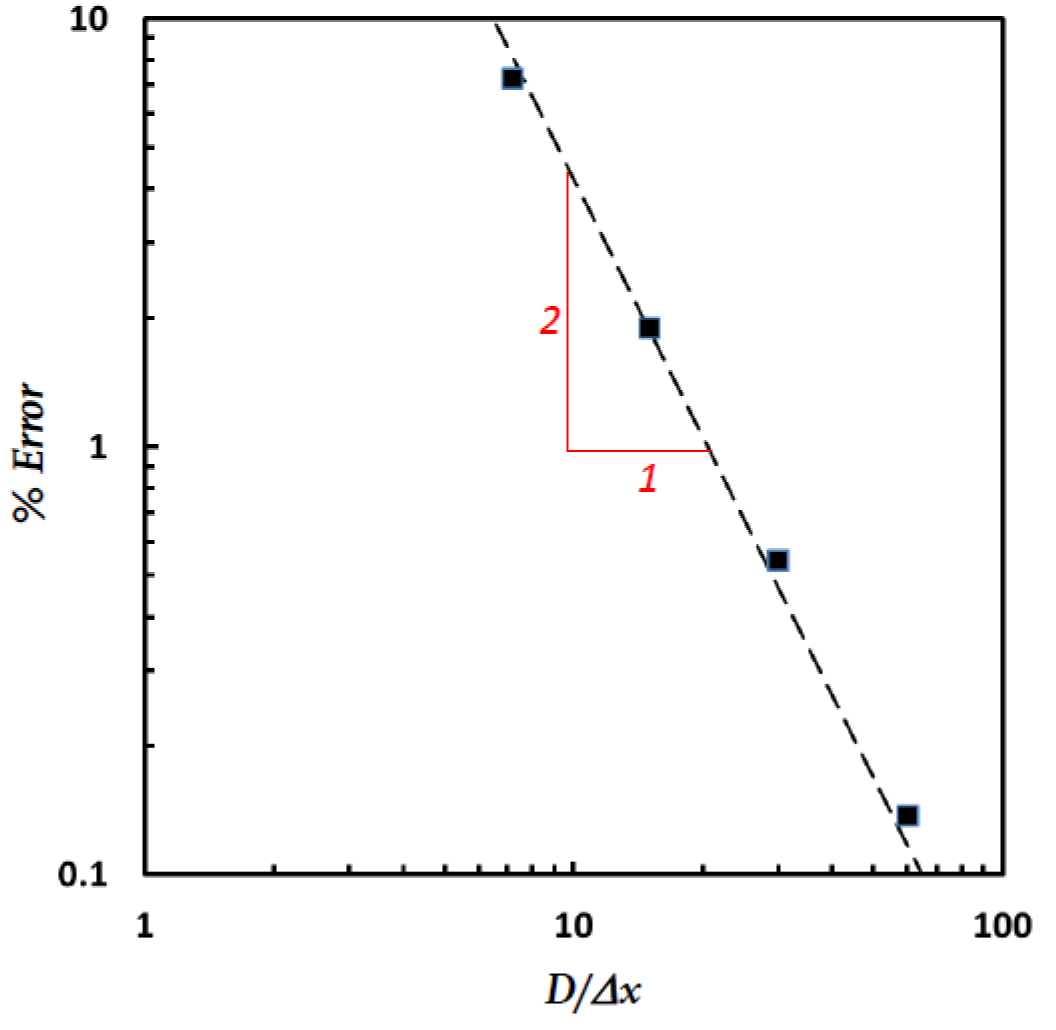
IBM BASED DNS MODEL

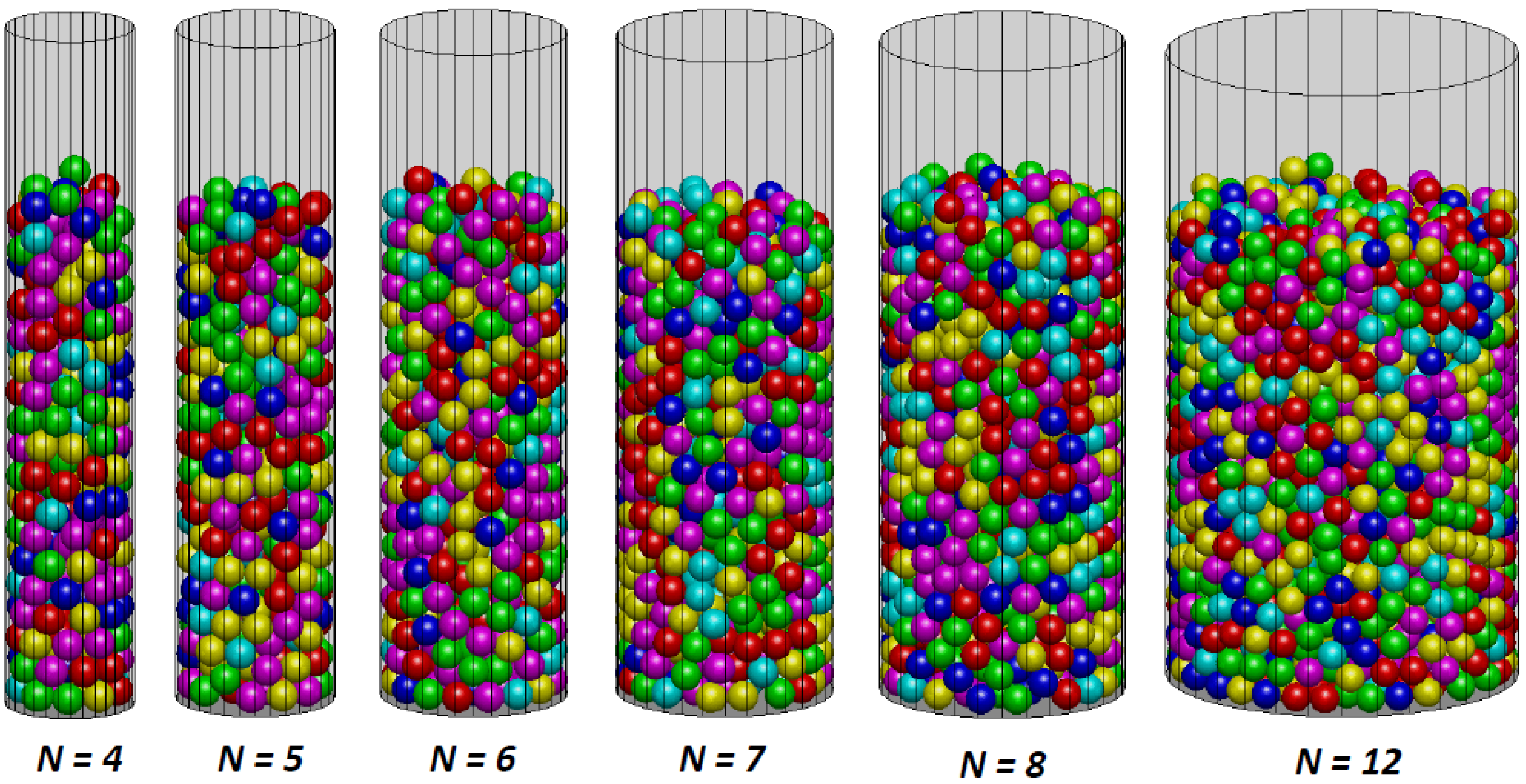
flow past a stationary arrays of spheres: grid convergence



Das et al., 2016, AIChEJ

IBM BASED DNS MODEL

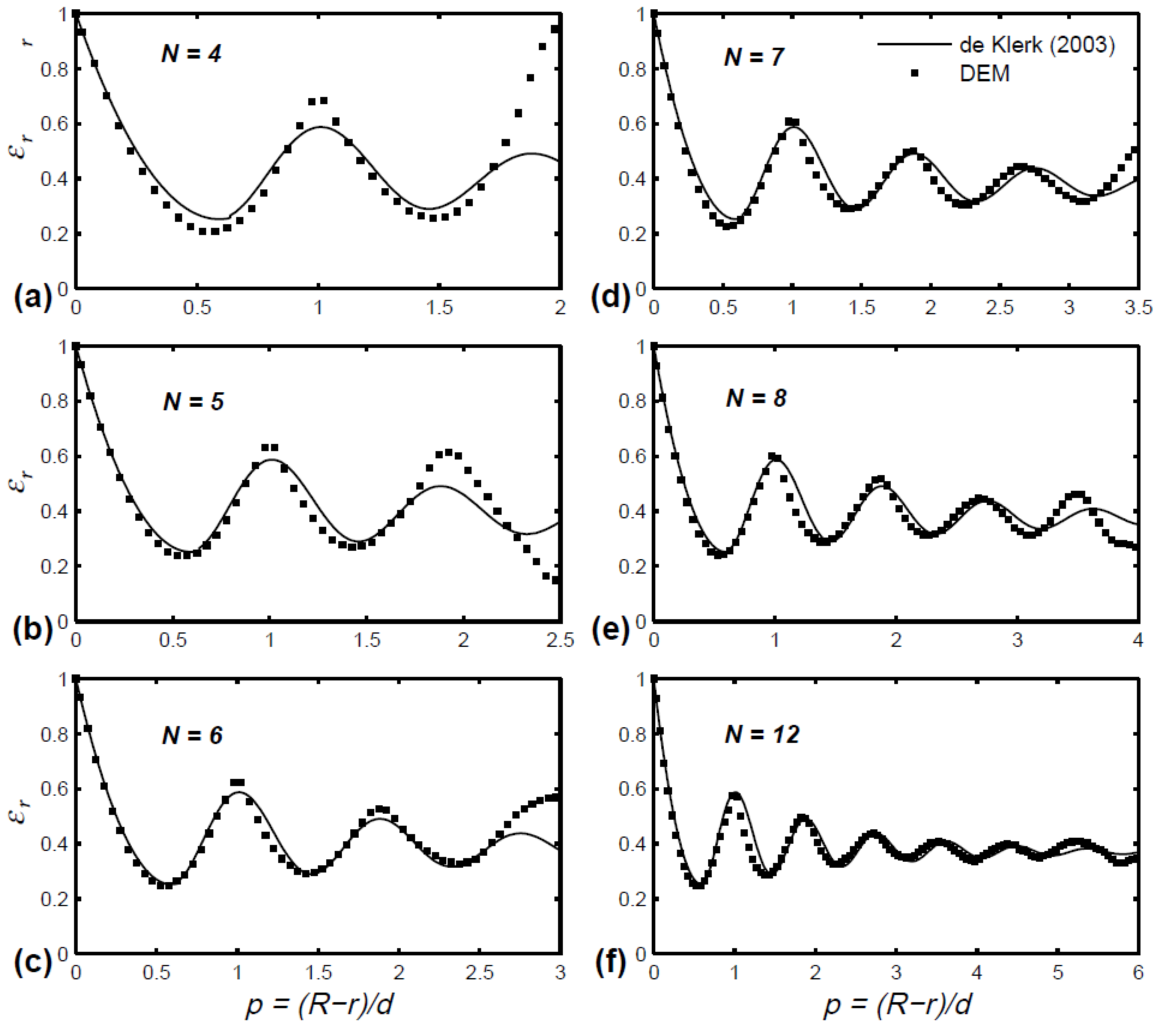
transport phenomena in packed bed reactors: DEM generated beds



Das et al., 2017, CES

IBM BASED DNS MODEL

transport phenomena in packed bed reactors: porosity profiles



Das et al., 2017, CES

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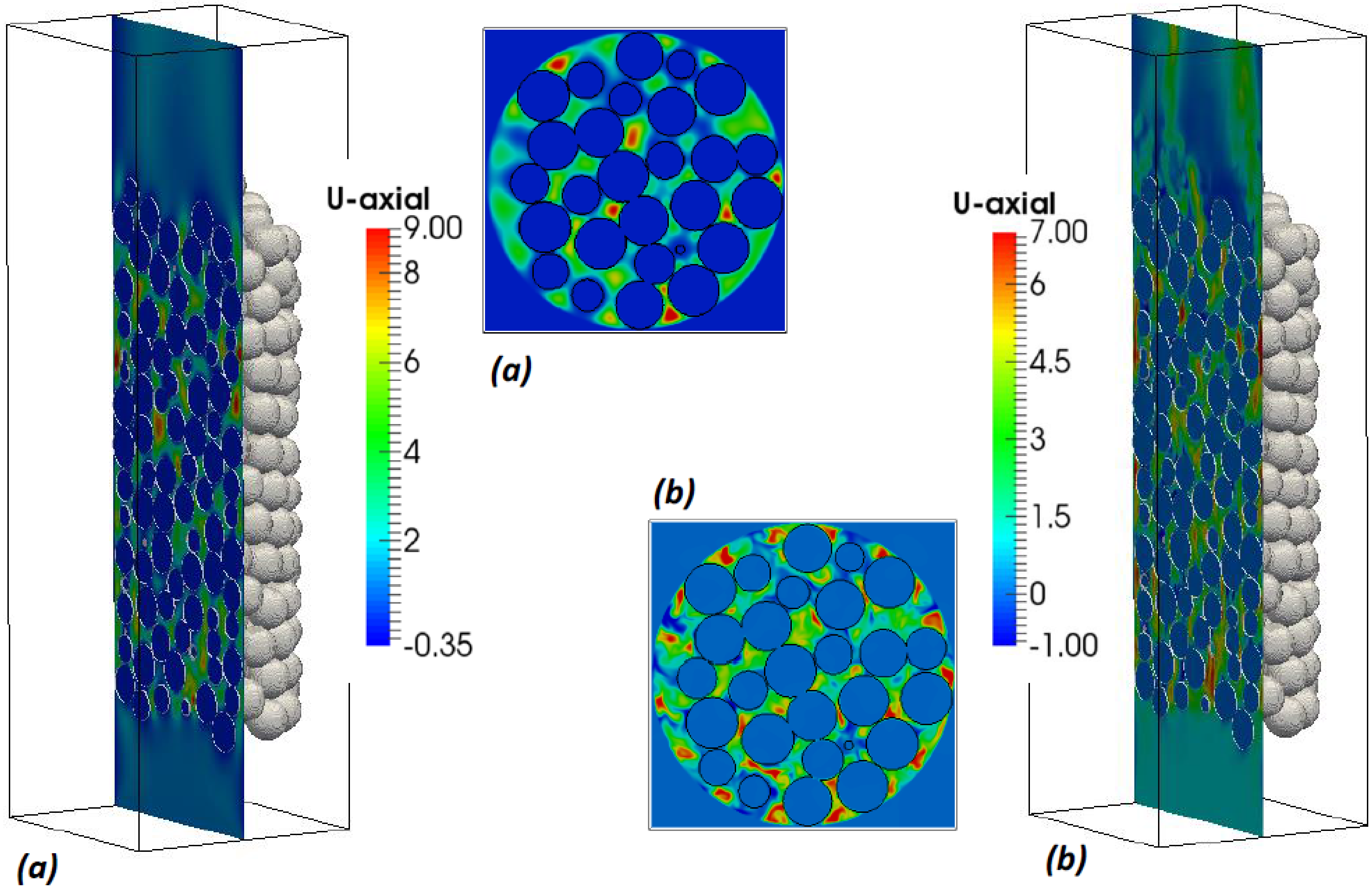
transport phenomena in packed bed reactors: velocity profiles (N=6)



Re_d=1 Re_d=500

Das et al., 2017, CES

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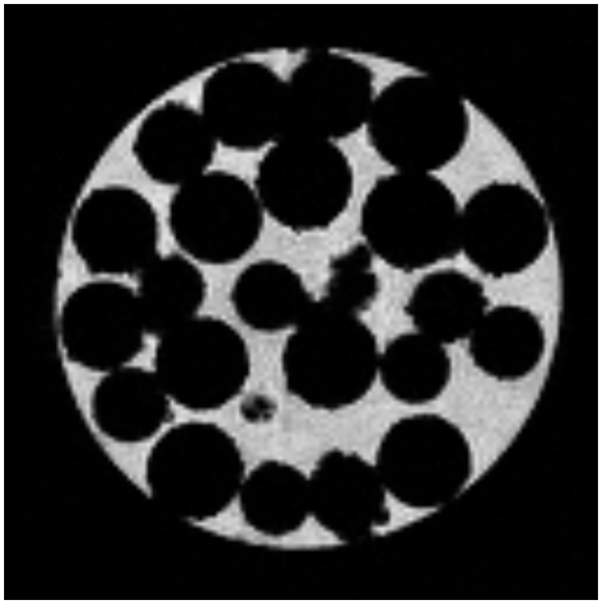
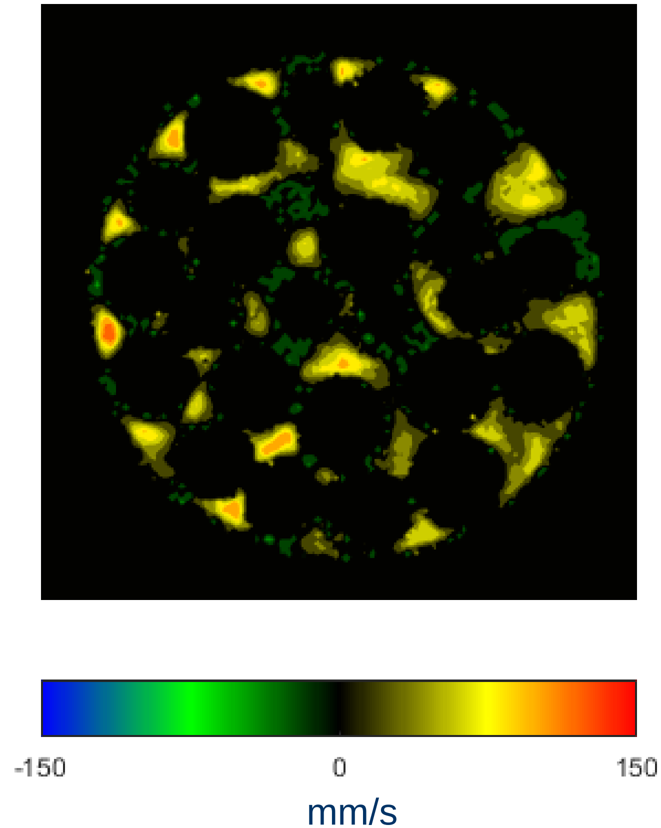
IBM BASED DNS MODEL

comparison with experiment (MRI flow imaging)





phase fractions (left) and axial velocity map (right) for a packed bed of spheres with a diameter of 4 mm

Lovreglio et al., 2018, AIChEJ

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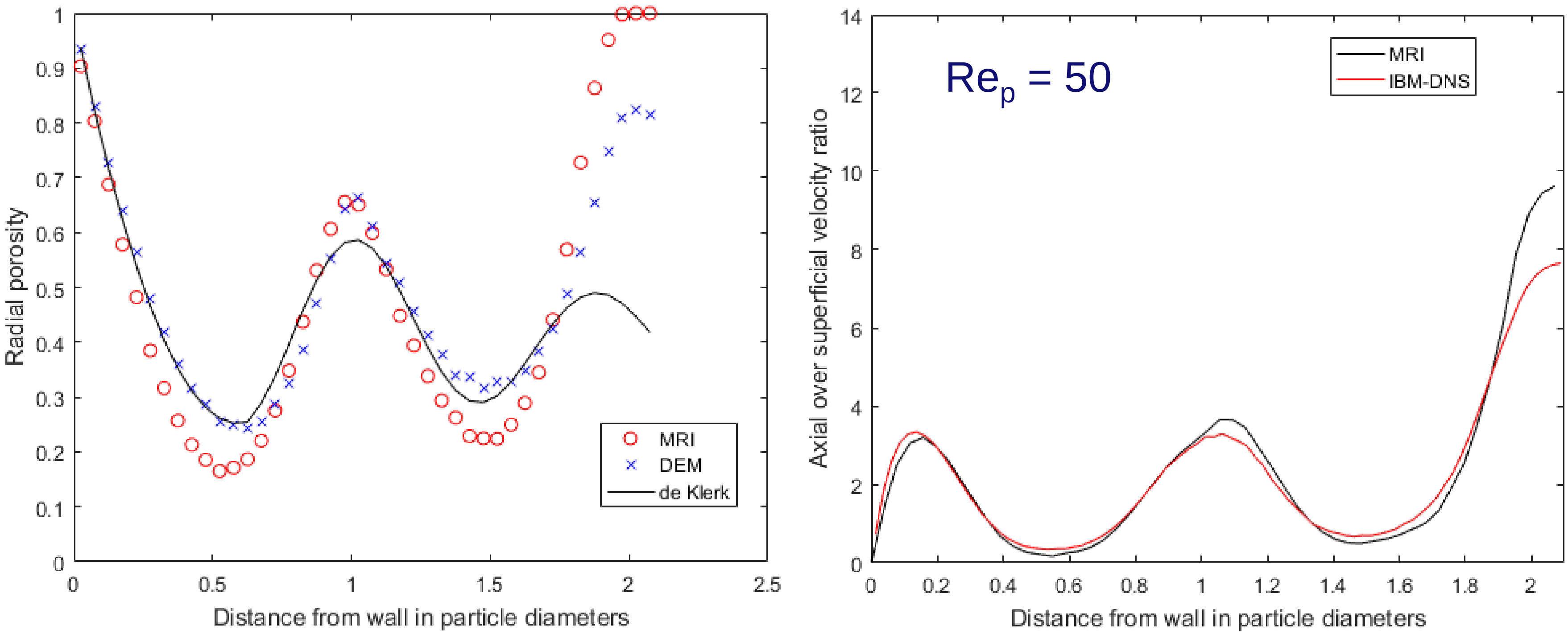
comparison with experiment (MRI flow imaging)



radial porosity profile (left) and axial velocity profile (right)
for a packed bed of spheres with a diameter of 5 mm

Lovreglio et al., 2018, AIChEJ

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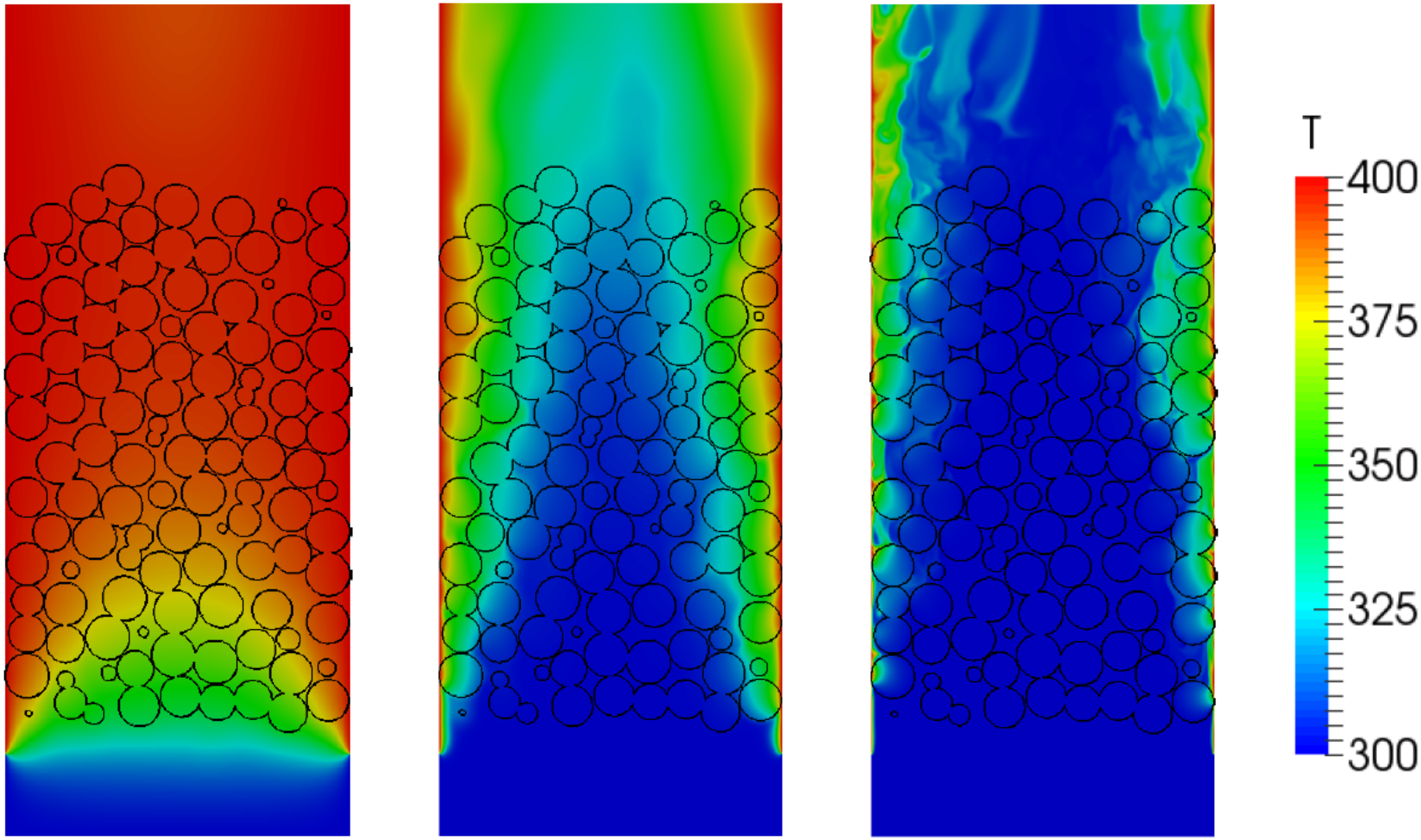
transport phenomena in packed bed reactors: temperature profiles



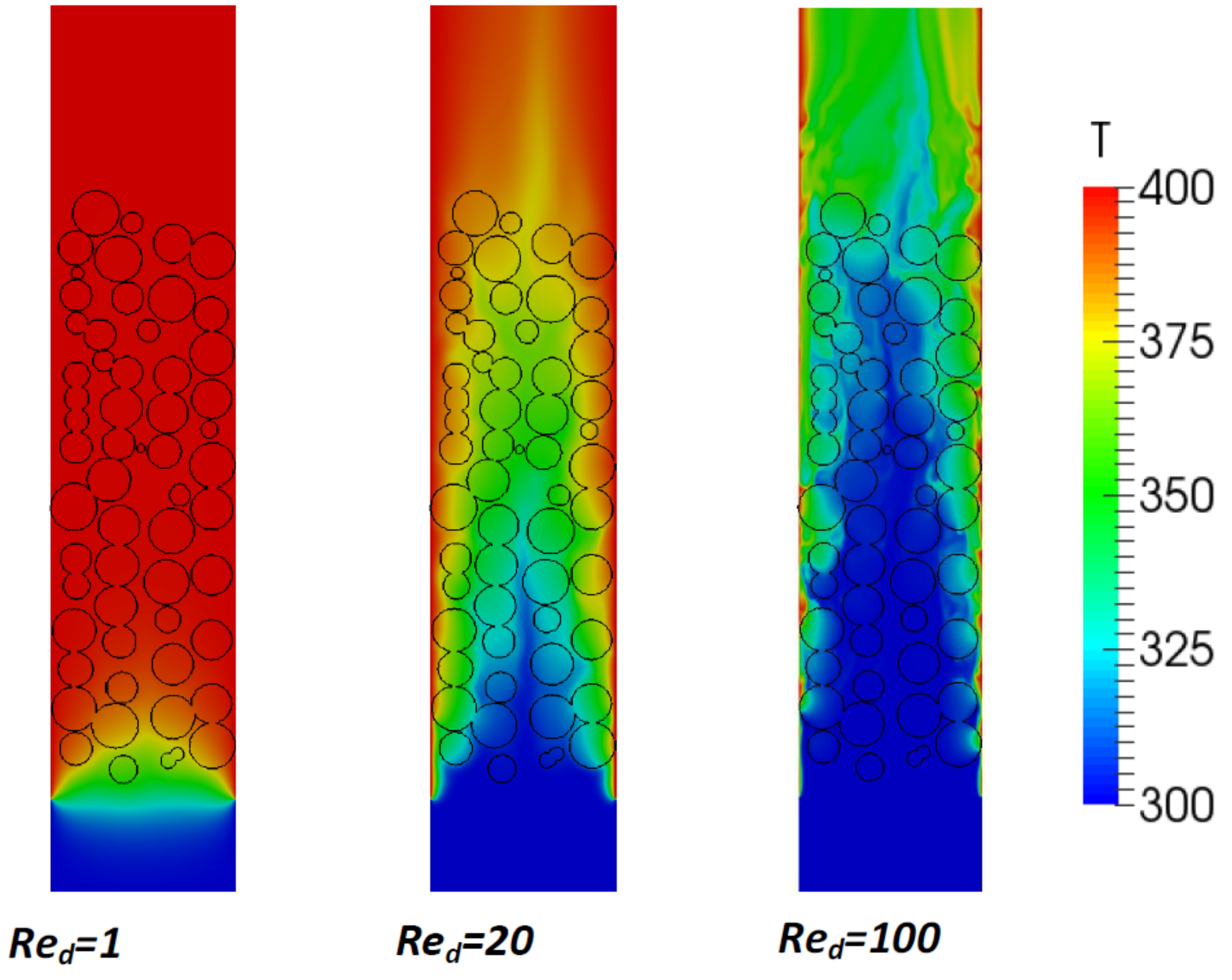
$Re_d=1$ $Re_d=20$ $Re_d=100$

conjugate heat transport in fluid and solid phase

Das et al., 2017, CES

IBM BASED DNS MODEL

transport phenomena in packed bed reactors: temperature profiles

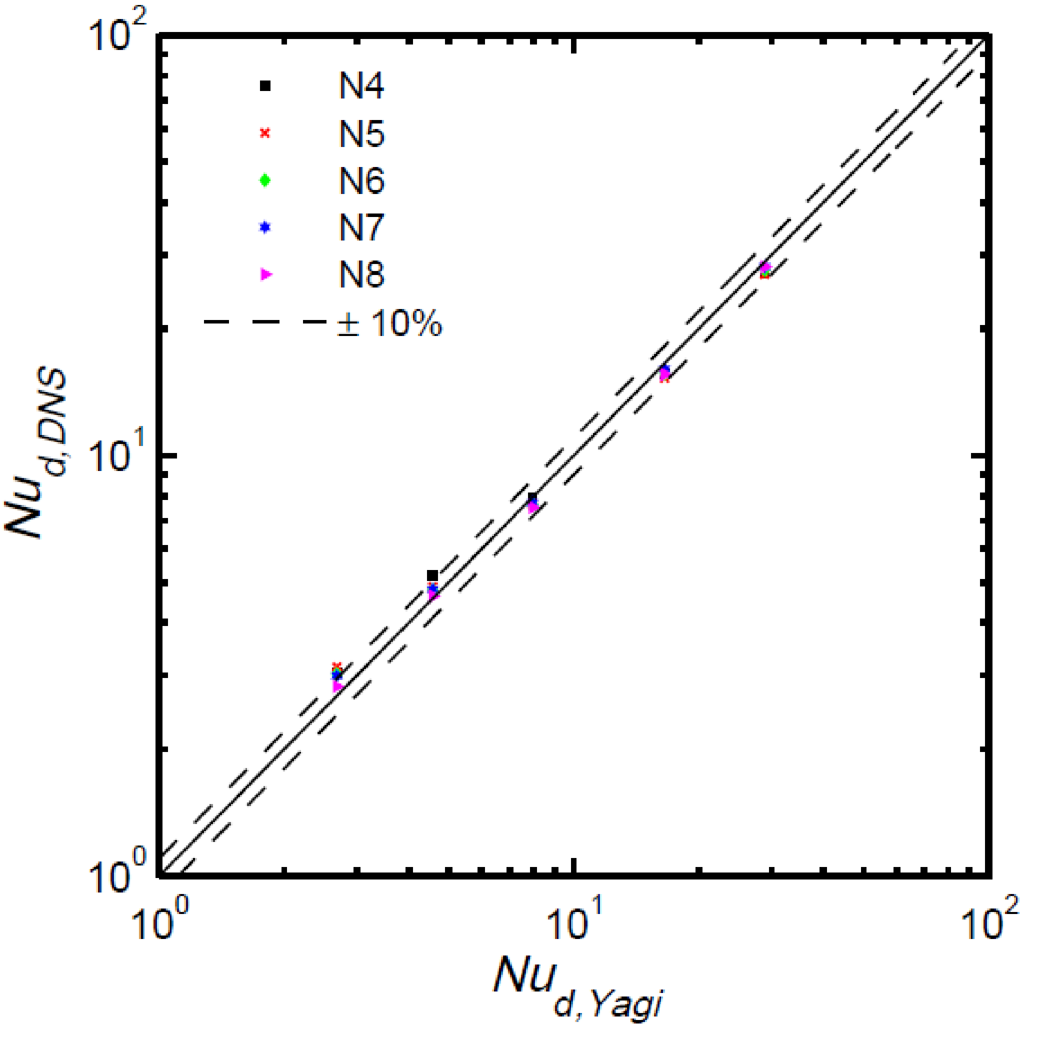


conjugate heat transport in
fluid and solid phase

Das et al., 2017, CES

IBM BASED DNS MODEL

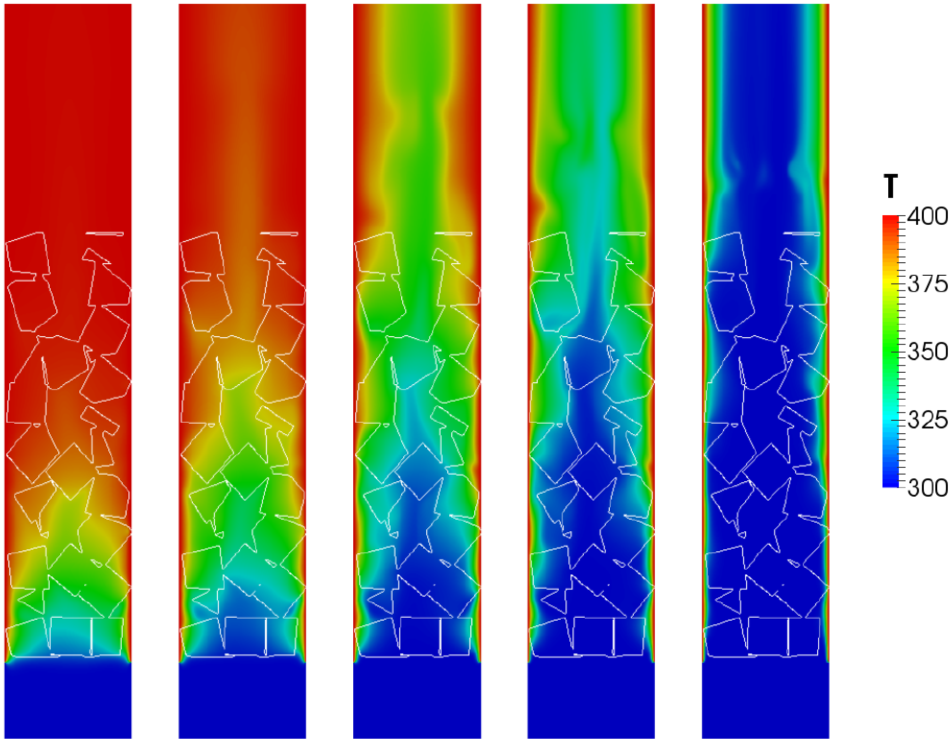
transport phenomena in packed bed reactors: wall-to-bed heat transfer



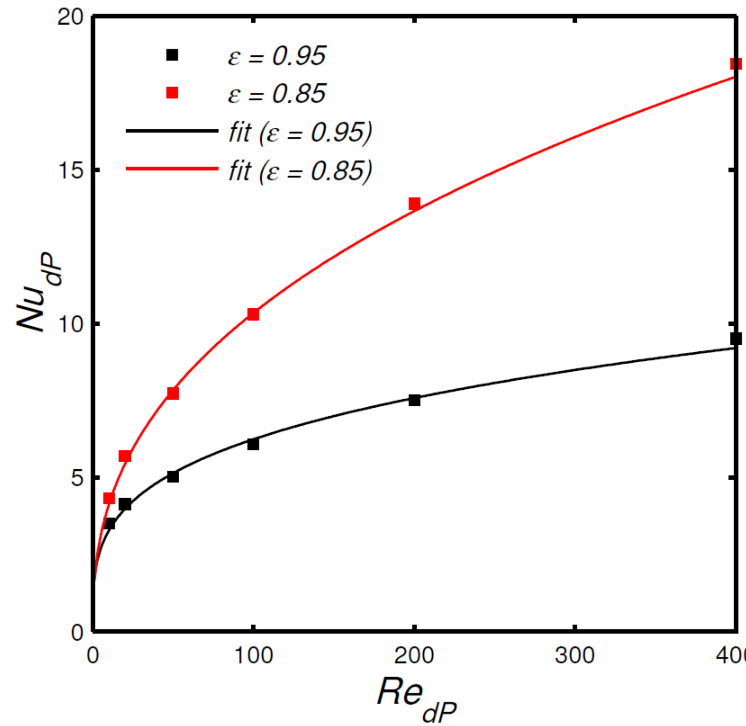
Das et al., 2017, CES

IBM BASED DNS MODEL

wall-to-bed heat transfer in fixed beds with non-spherical particles consisting of solid foam (porous particles)



$\mathrm{Re}_{d_p} = 10$ $\mathrm{Re}_{d_p} = 20$ $\mathrm{Re}_{d_p} = 50$ $\mathrm{Re}_{d_p} = 100$ $\mathrm{Re}_{d_p} = 400$



cubic porous particles (P9: 9x9x9 mm) consisting of solid foam are packed in 30 mm diameter (C30) tube using DEM followed by fully resolved IBM simulations to obtain the wall-to-bed heat transfer coeficients versus particle Reyolds number

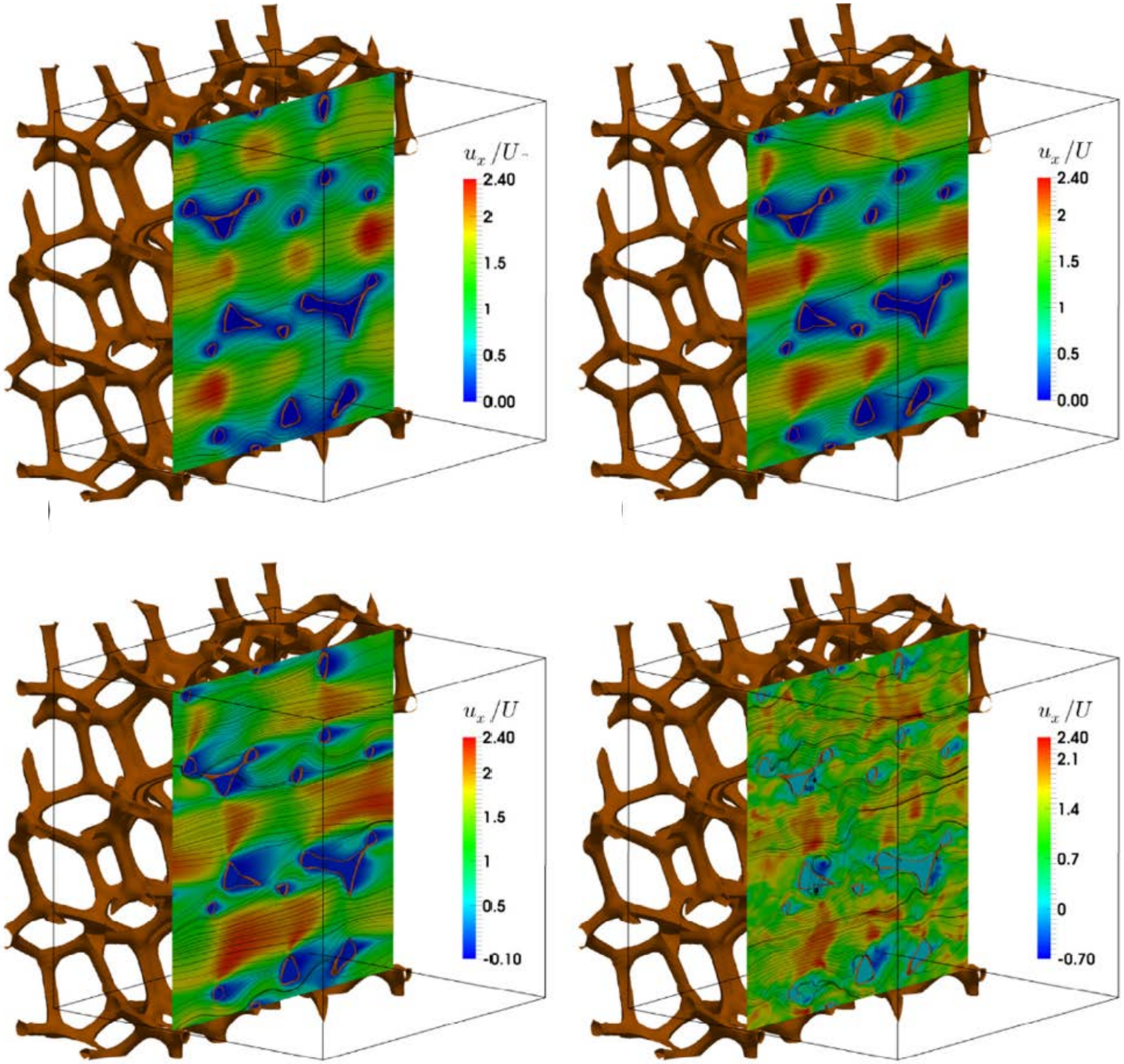
micro-scale closures obtained from fully-resolved simulations (next two slides)

IBM BASED DNS MODEL

micro-scale closure development: hydrodynamics

Re = 0.01

Re = 10



Re = 10

Re = 0.01

Das et al., 2018, CES

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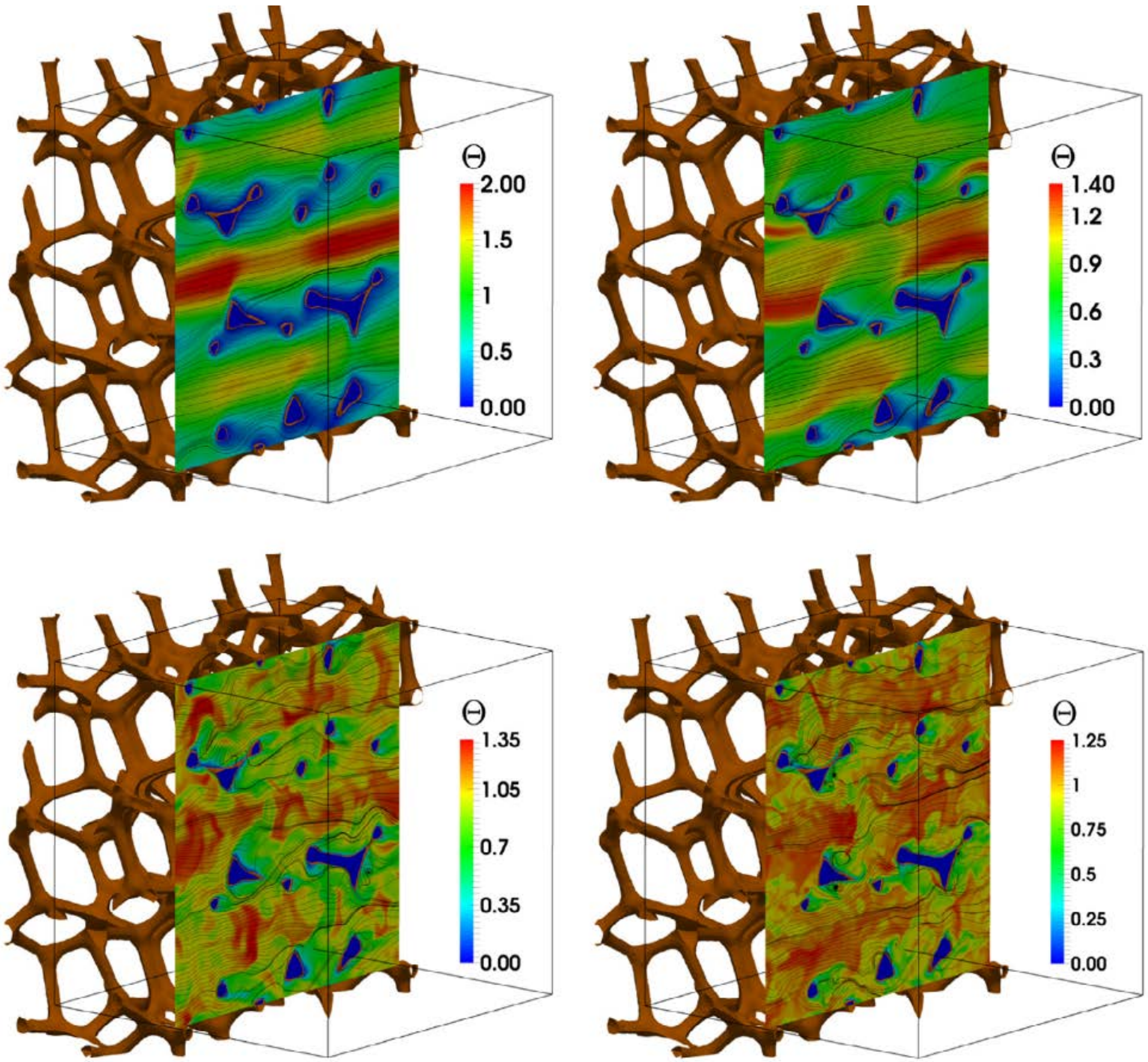
micro-scale closure development: heat transfer

$\mathrm{Re} = 10$

$\mathrm{Re} = 50$



$$\Theta = \frac{T(x,y,z) - T_s}{\langle T_c(x) \rangle - T_s}$$

$\mathrm{Re} = 200$

$\mathrm{Re} = 500$

Das et al., 2018, CES

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IBM BASED DNS MODEL

fluid-particle (spheres) heat transfer: comparison with empirical correlation

Gunn, International J. Heat and Mass Transfer (1978)

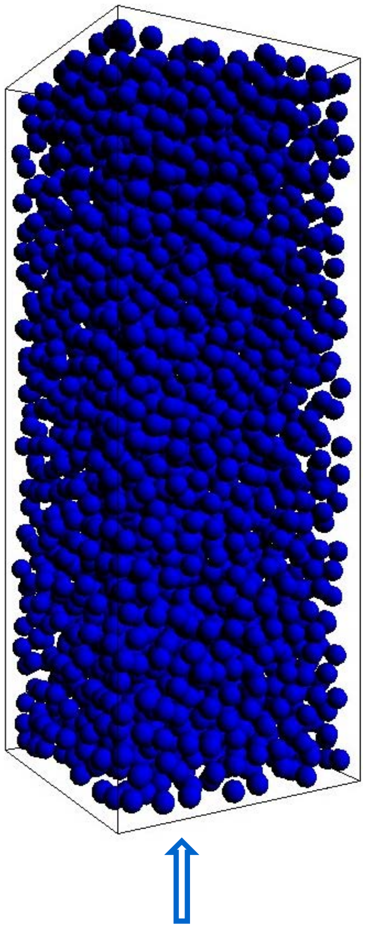
$$Nu_p = \left(7 - 10\varepsilon_b + 5\varepsilon_b^2\right)\left(1 + 0.7\,\mathrm{Re}_p^{0.2}\,\mathrm{Pr}^{1/3}\right) + \left(1.33 - 2.4\varepsilon_b + 1.2\varepsilon_b^2\right)\mathrm{Re}_p^{0.7}\,\mathrm{Pr}^{1/3}$$

$$Sh_p = \left(7 - 10\varepsilon_b + 5\varepsilon_b^2\right)\left(1 + 0.7\,\mathrm{Re}_p^{0.2}\,Sc^{1/3}\right) + \left(1.33 - 2.4\varepsilon_b + 1.2\varepsilon_b^2\right)\mathrm{Re}_p^{0.7}\,Sc^{1/3}$$

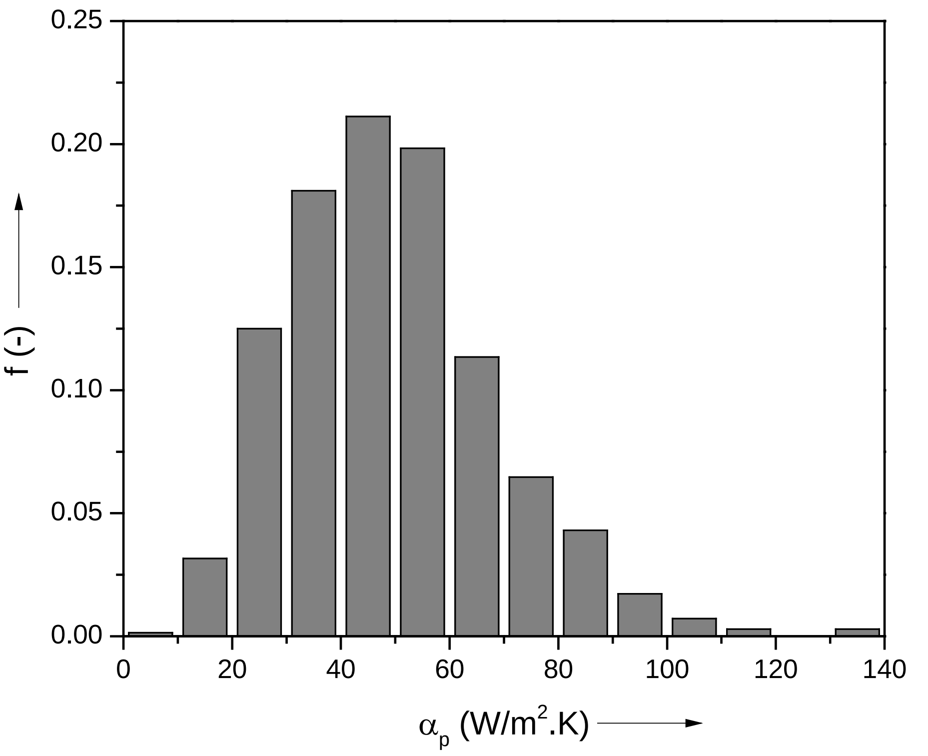
Re_p	α_p (W/(m^2.K)) DNS	α_p (W/(m^2.K)) Gunn (1978)	k_m (m/s) DNS	k_m (m/s) Gunn (1978)
120	25.23	26.87	0.0219	0.0228
180	30.28	31.91	0.0263	0.0272
240	34.33	36.36	0.0298	0.0310

IBM BASED DNS MODEL

distribution of fluid-particle heat transfer coefficient at Re_p=60 for 3000 spherical particles



particles kept at a constant temperature, cold fluid entering from bottom of the column
solidity equals 0.30



broad distribution of α_p prevails due to existence of preferential pathways of the fluid percolating through the stationary array

Deen & Kuipers, 2014, CES

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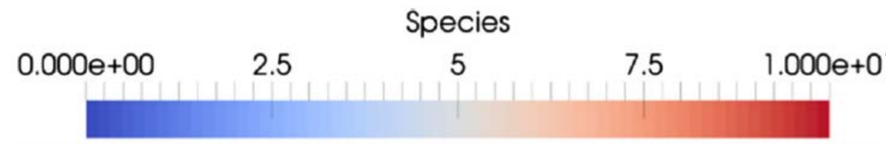
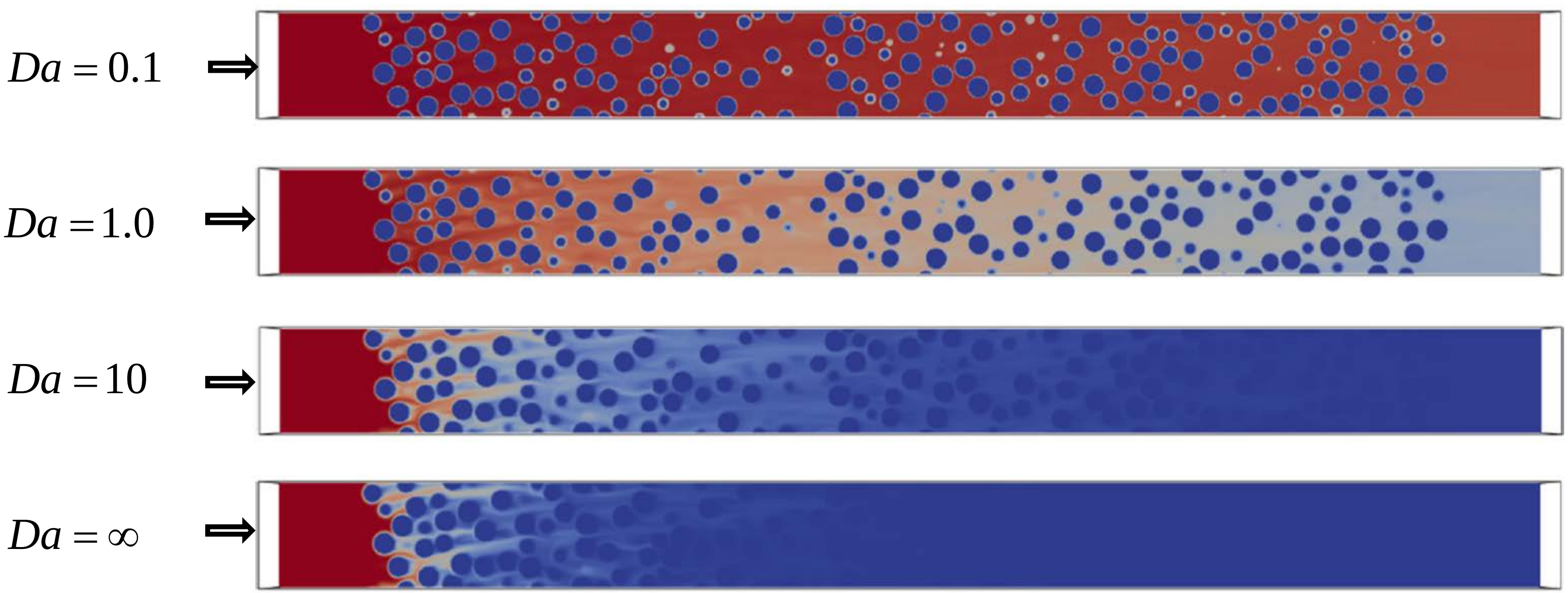
IBM BASED DNS MODEL

interplay between transport phenomena and chemical reaction

$$\alpha\phi_B + \beta\frac{\partial\phi_B}{\partial n} = f \quad \Longrightarrow \quad f = 0 \qquad \beta = D_f \qquad Da = \frac{\alpha R_p}{D_f}$$

generic boundary condition at surface of a catalyst particles





only surface reaction considered here, no intra-particle transport

Lu et al., 2018, CES

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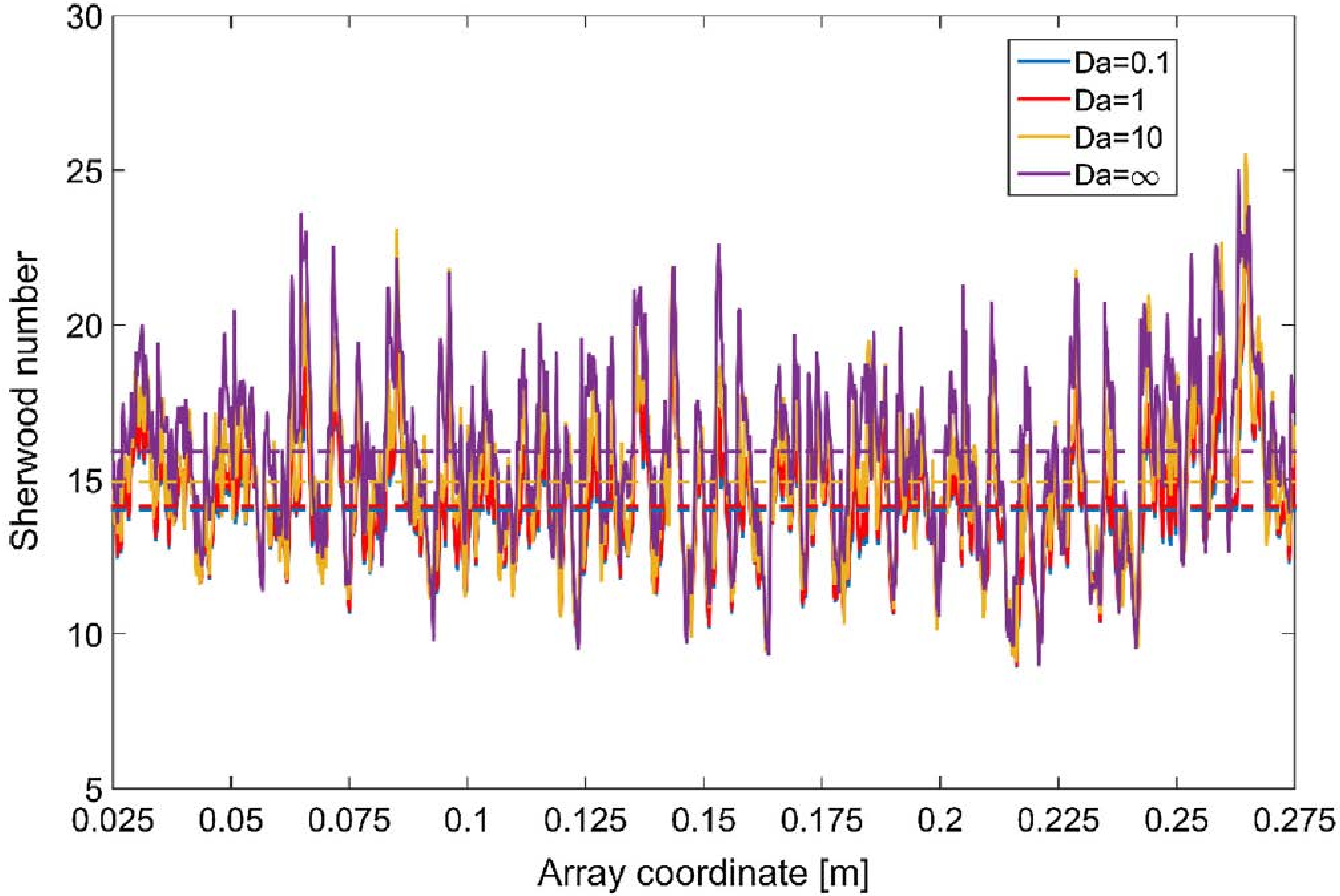
IBM BASED DNS MODEL

interplay between transport phenomena and chemical reaction



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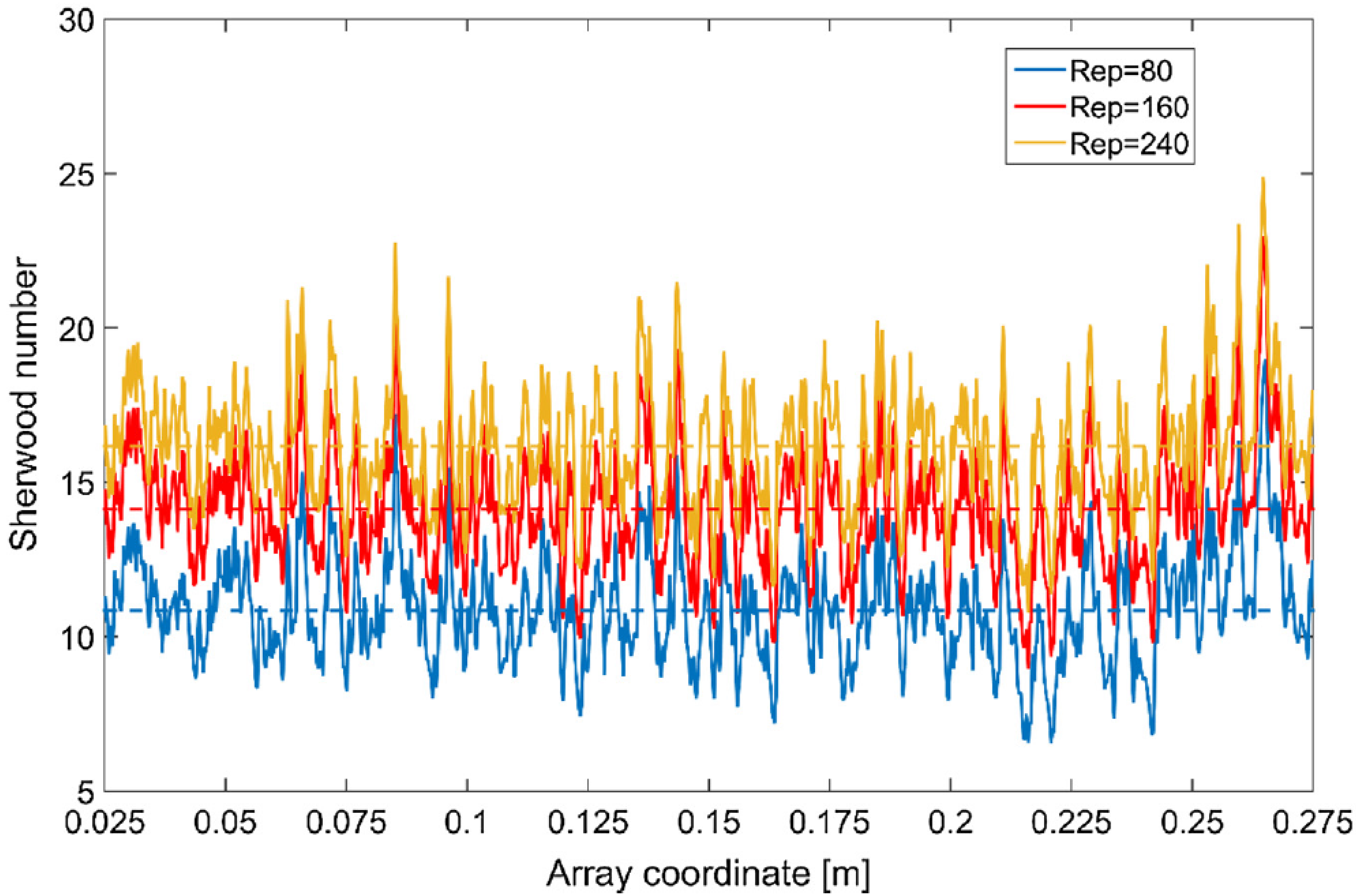
Lu et al., 2018, CES

IBM BASED DNS MODEL
interplay between transport phenomena and chemical reaction



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Lu et al., 2018, CES

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IBM BASED DNS MODEL

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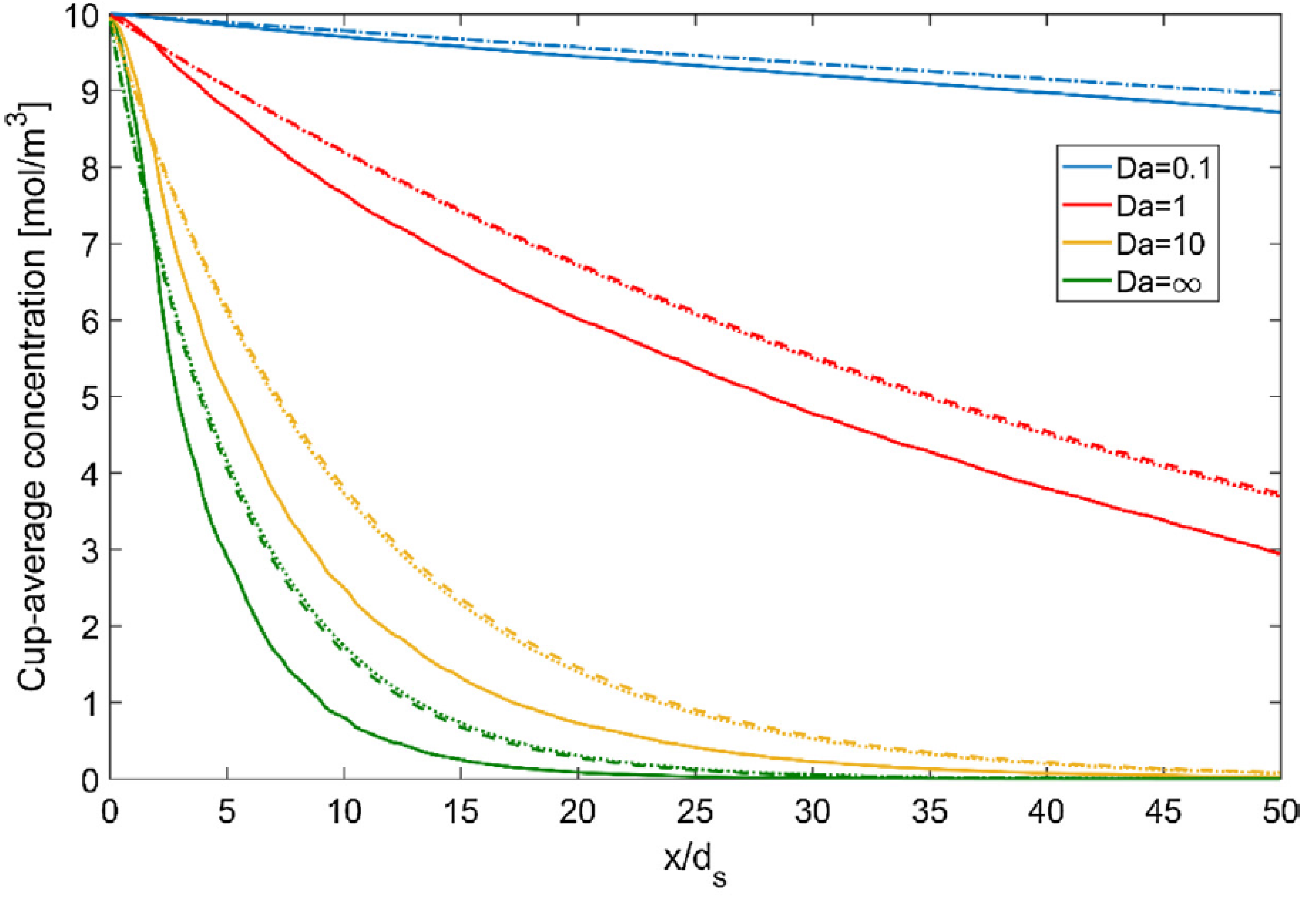
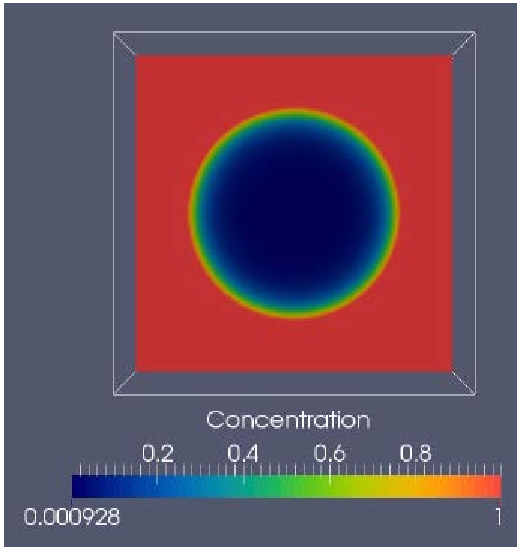
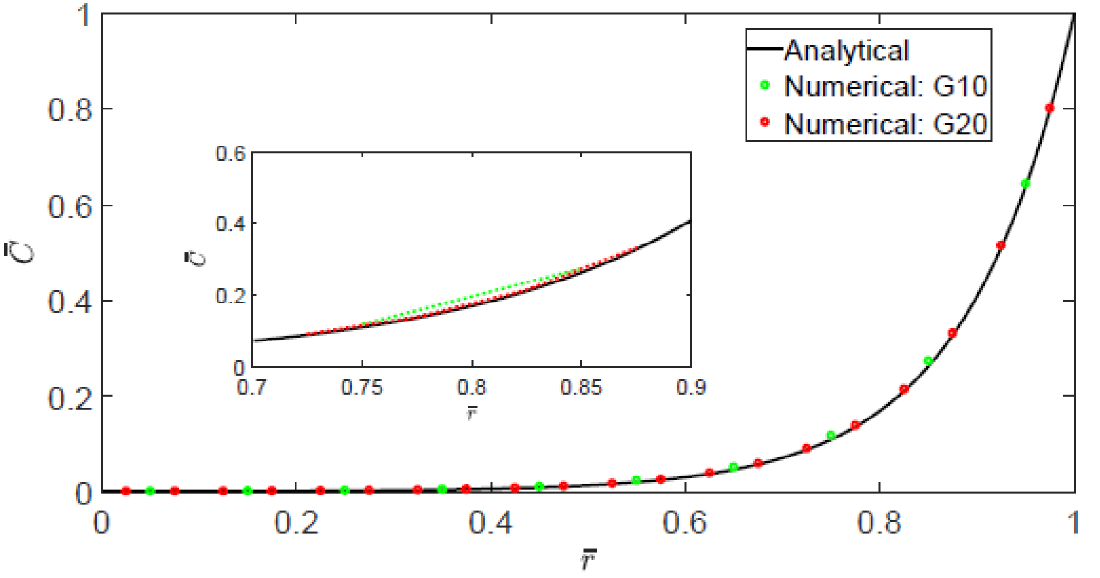


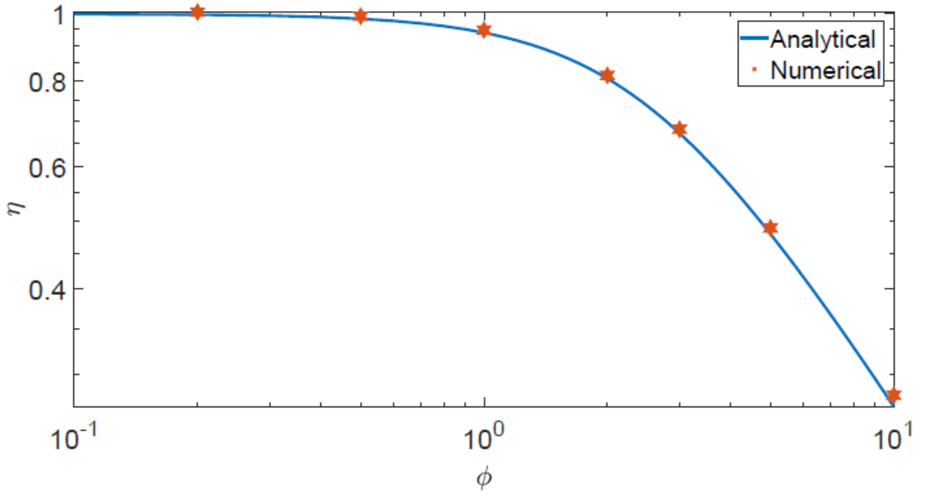
Lu et al., 2018, CES

IBM BASED DNS MODEL

isothermal reaction with diffusion limitation in single particle







effectiveness factor

$$\eta = \frac{3}{\phi^2}\left(\phi \coth(\phi) - 1\right)$$

Thiele modulus

$$\phi = R\sqrt{\frac{k}{D_s}}$$

Chandra et al., 2020, CEJ

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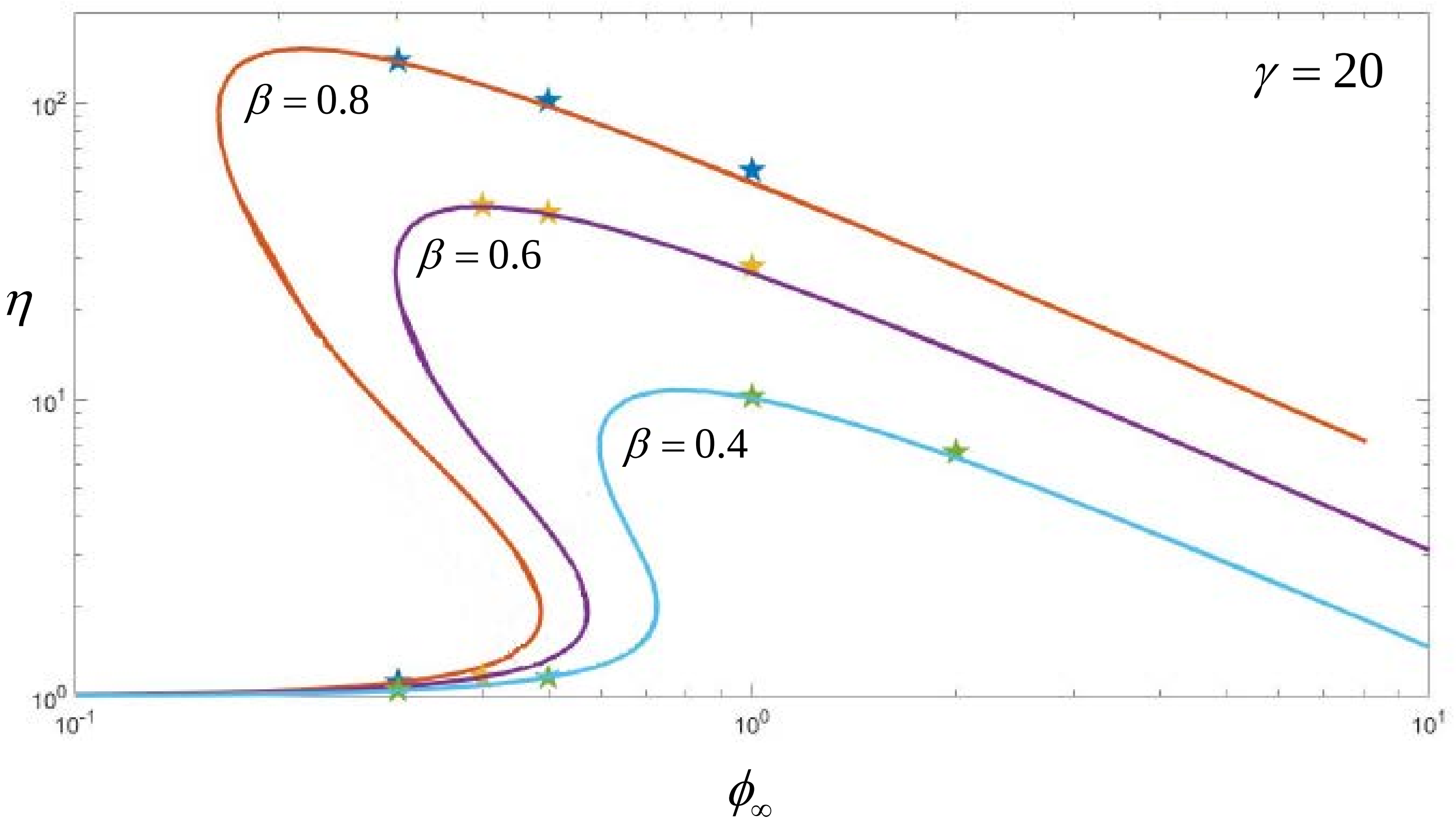
IBM BASED DNS MODEL

effectiveness factor versus Thiele modulus (single sphere)



$$\eta = \frac{\iiint_{V_s} k_0 \exp[-E_a/(RT_s)]c_{A,s}dV}{k_0 \exp[-E_a/(RT_\infty)]c_{A,\infty}V_s}$$

$$\beta = \frac{c_{A,\infty}(-\Delta H_r)D_{A,s}}{\lambda_s T_\infty}$$

$$\gamma = \frac{E_a}{RT_\infty}$$

Chandra et al., 2020, CEJ

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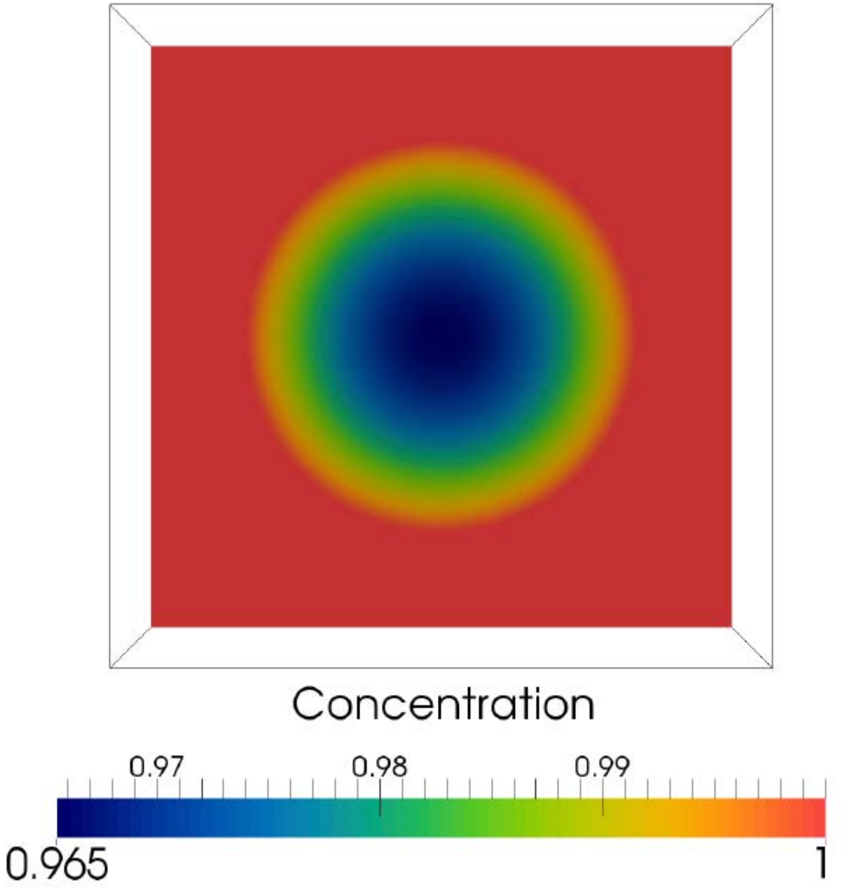
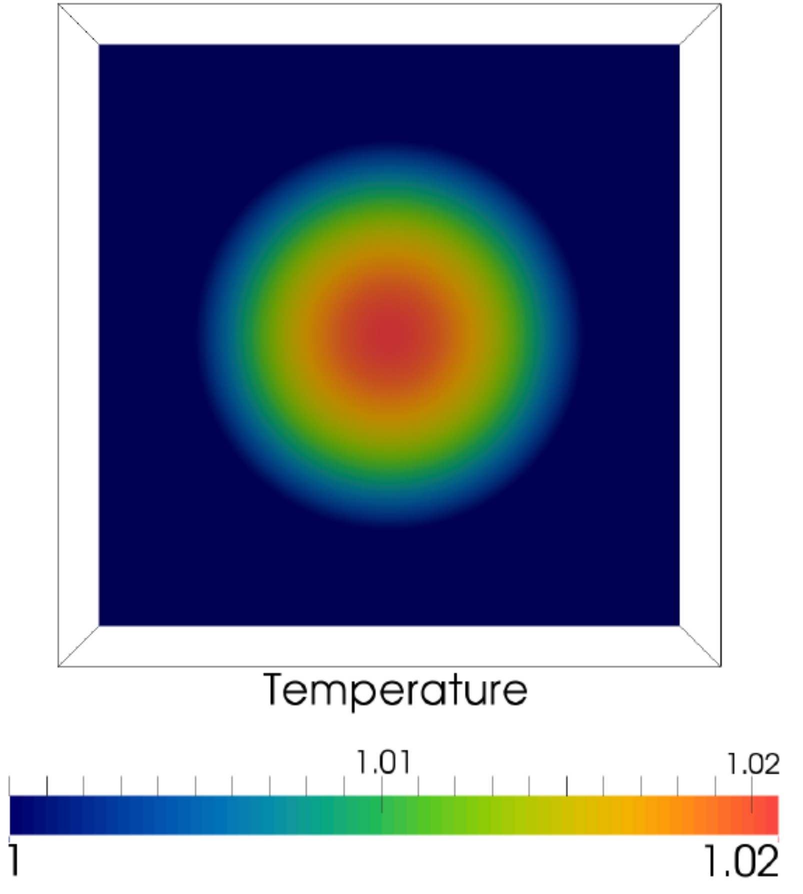
IBM BASED DNS MODEL
concentration and temperature distributions (single sphere)





lower stable steady state:

$$\phi_\infty = 0.4 \quad \gamma = 20 \quad \beta = 0.6 \quad \Rightarrow \quad \eta = 1.162$$

Chandra et al., 2020, CEJ

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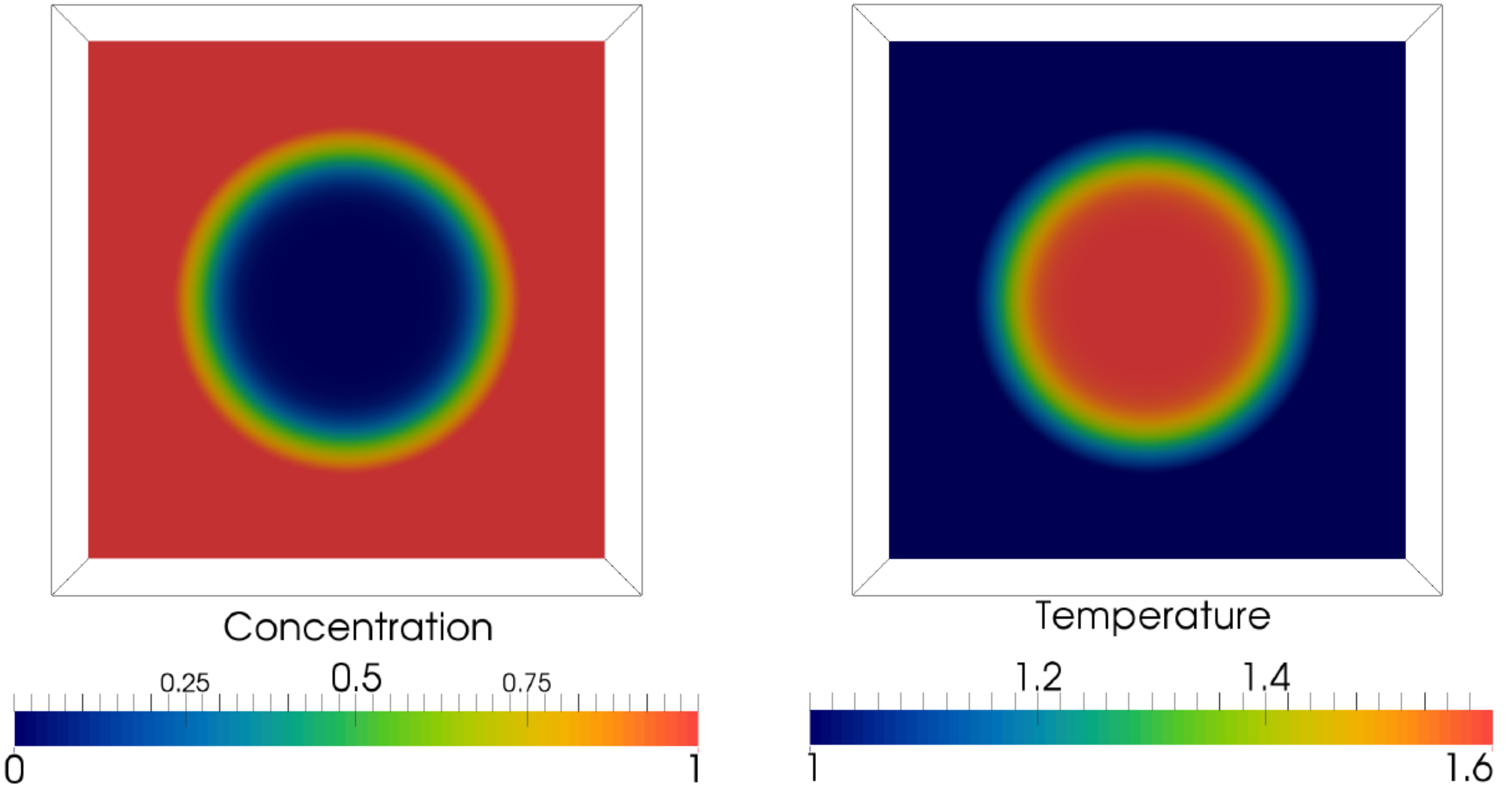
IBM BASED DNS MODEL

concentration and temperature distributions (single sphere)



higher stable steady state:

$$\phi_{\infty} = 0.4 \quad \gamma = 20 \quad \beta = 0.6 \quad \Rightarrow \quad \eta = 44.94$$

Chandra et al., 2020, CEJ

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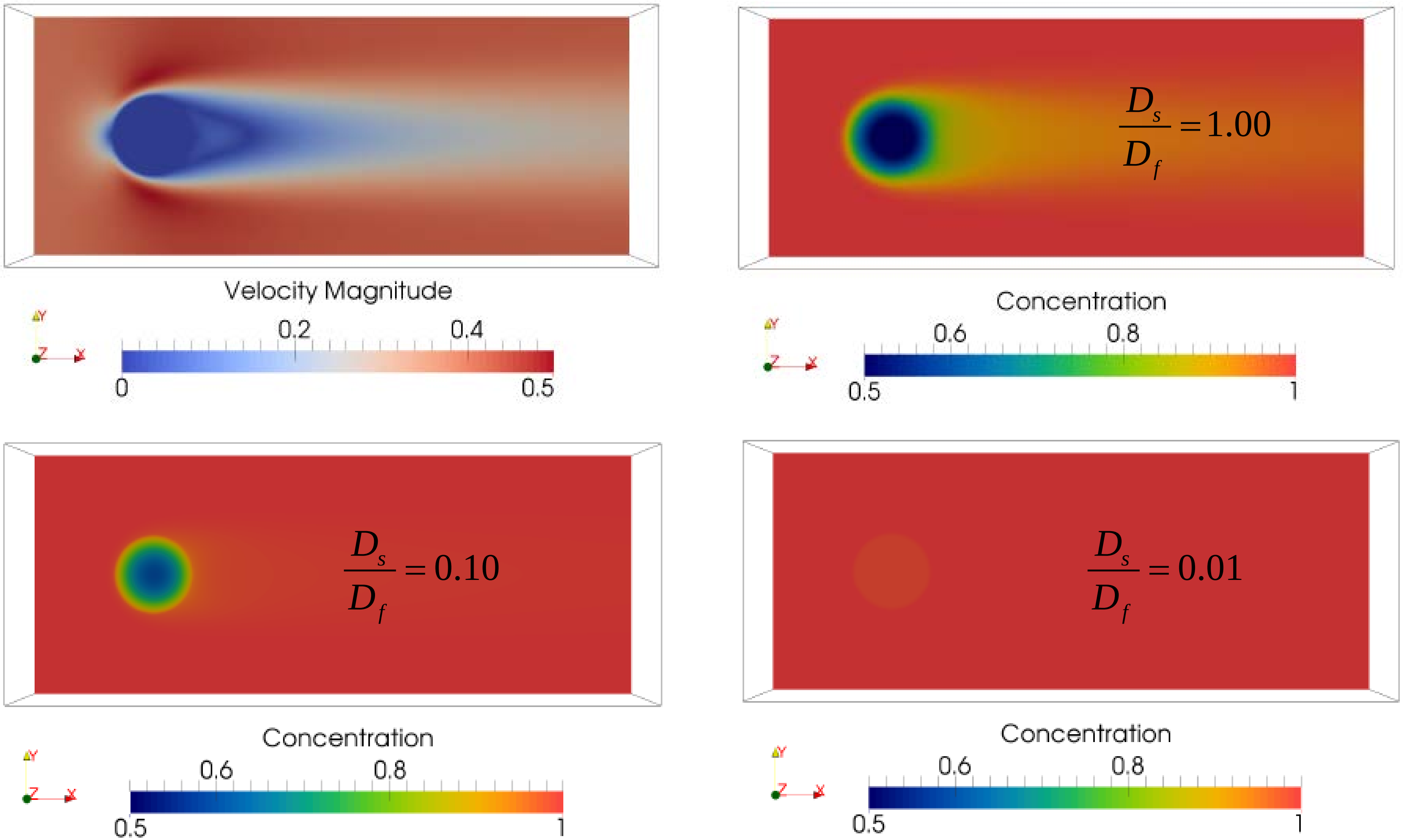
IBM BASED DNS MODEL

single sphere (Re_p=60 and ϕ=2.0): effect of ratio of diffusivities



Chandra et al., 2020, CEJ

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IBM BASED DNS MODEL

full bed simulations: values of nondimensional parameters

Reynolds number: $\mathrm{Re}_p = \dfrac{u_0 d_p}{\nu_f} = 100$

Diffusivity ratio: $\dfrac{D_f}{D_s} = 5$

Prandtl number: $\mathrm{Pr} = \dfrac{\mu_f C_{p,f}}{\lambda_f} = 1.0$

Ratio wall and inlet temperature: $\dfrac{T_w}{T_0} = 1.0$

Schmidt number: $Sc = \dfrac{\nu_f}{D_f} = 1.0$

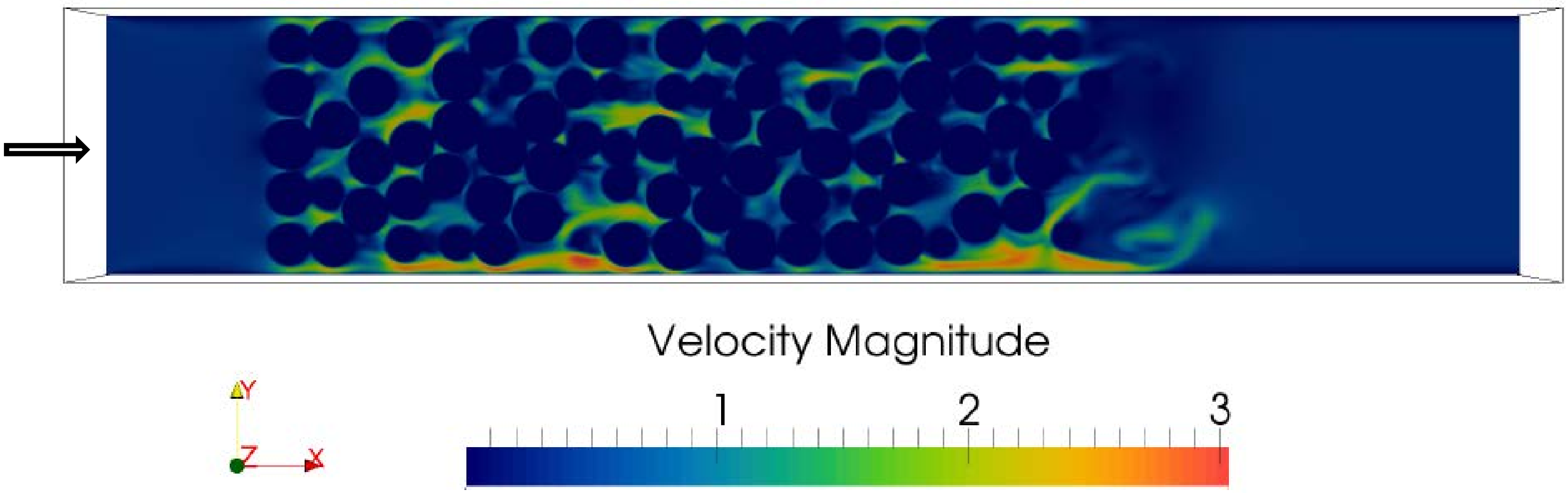
Arrhenius number: $\gamma = \dfrac{E_a}{RT_0} = 20.0$

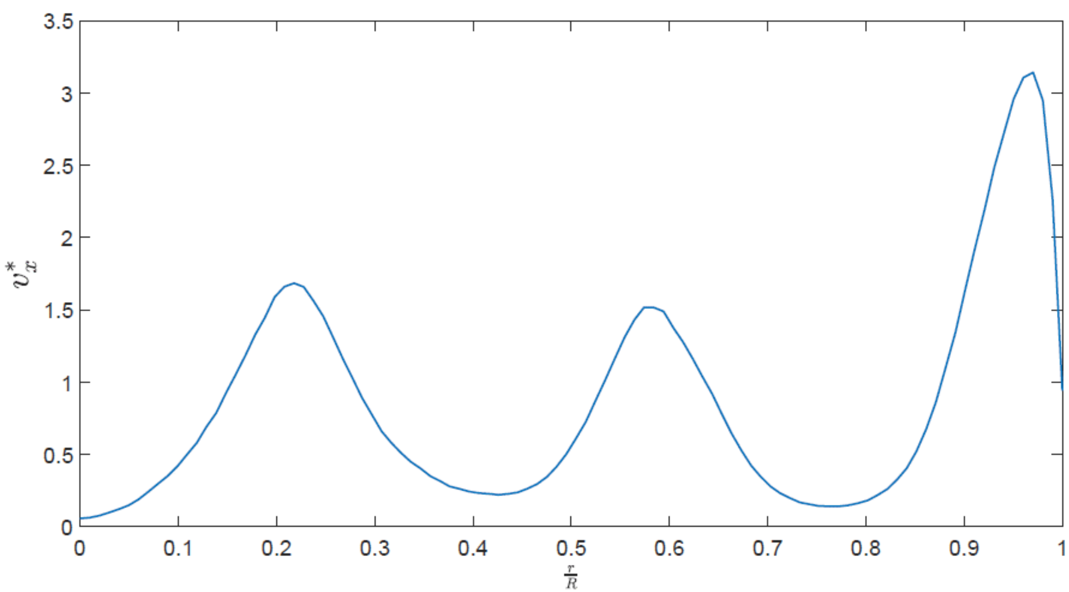
Thermal conductivity ratio: $\dfrac{\lambda_f}{\lambda_s} = 0.1$

Prater number: $\beta = \dfrac{c_0(-\Delta H_r)D_s}{\lambda_s T_0} = 0.02$

Chandra et al., 2020, CEJ

IBM BASED DNS MODEL
full bed simulations: flow field





radial profile of the azimuthally averaged superficial axial velocity component

Chandra et al., 2020, CEJ

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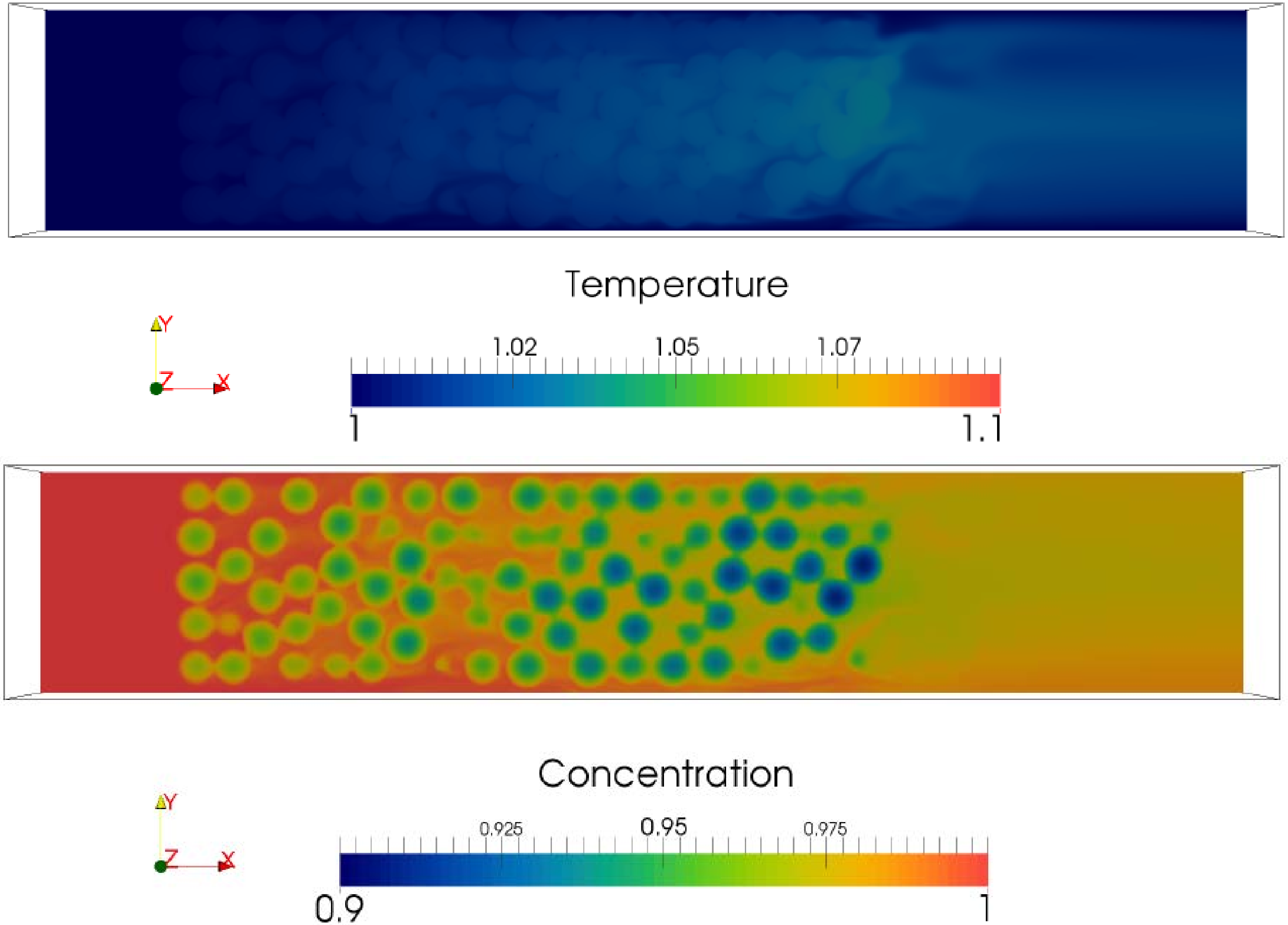
full bed simulations at ϕ_0=0.5: temperature and concentration distributions in central plane



Chandra et al., 2020, CEJ

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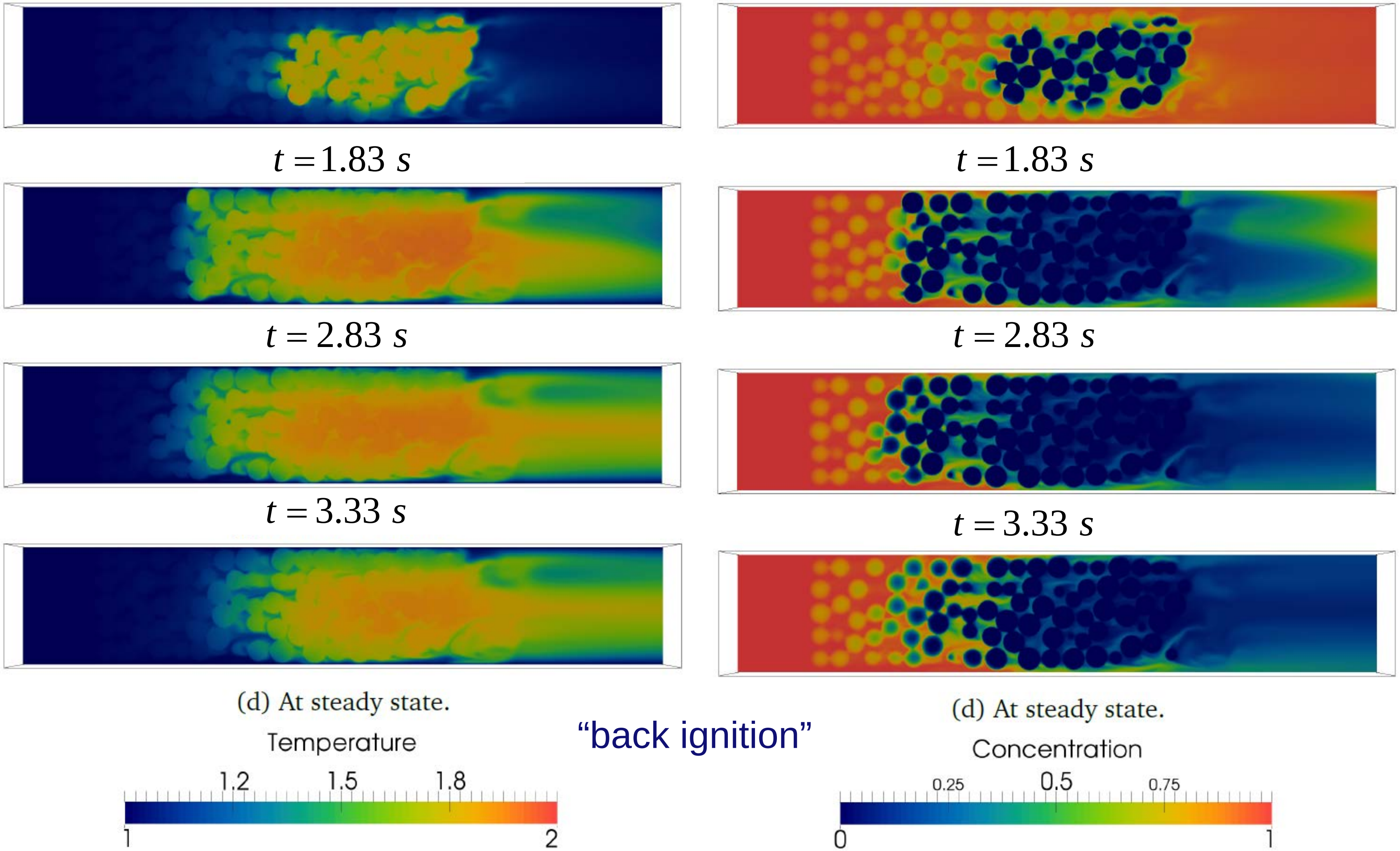
IBM BASED DNS MODEL

full bed simulations at ϕ_0=1.0: transient evolution of temperature and concentration distributions in central plane



"back ignition"

Chandra et al., 2020, CEJ

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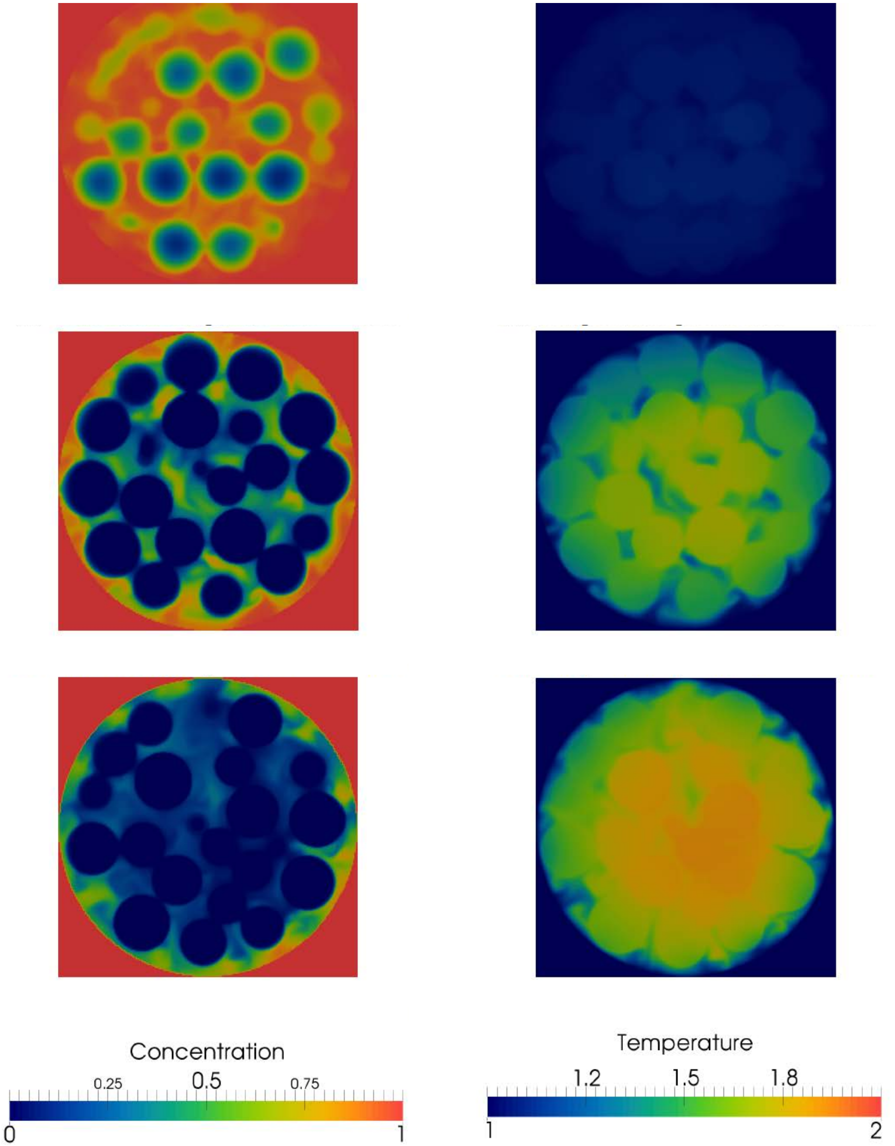
IBM BASED DNS MODEL

full bed simulations at ϕ_0=1.0: cross-sectional profiles of concentration and temperature

$$\frac{x}{L} = 0.25$$

$$\frac{x}{L} = 0.50$$

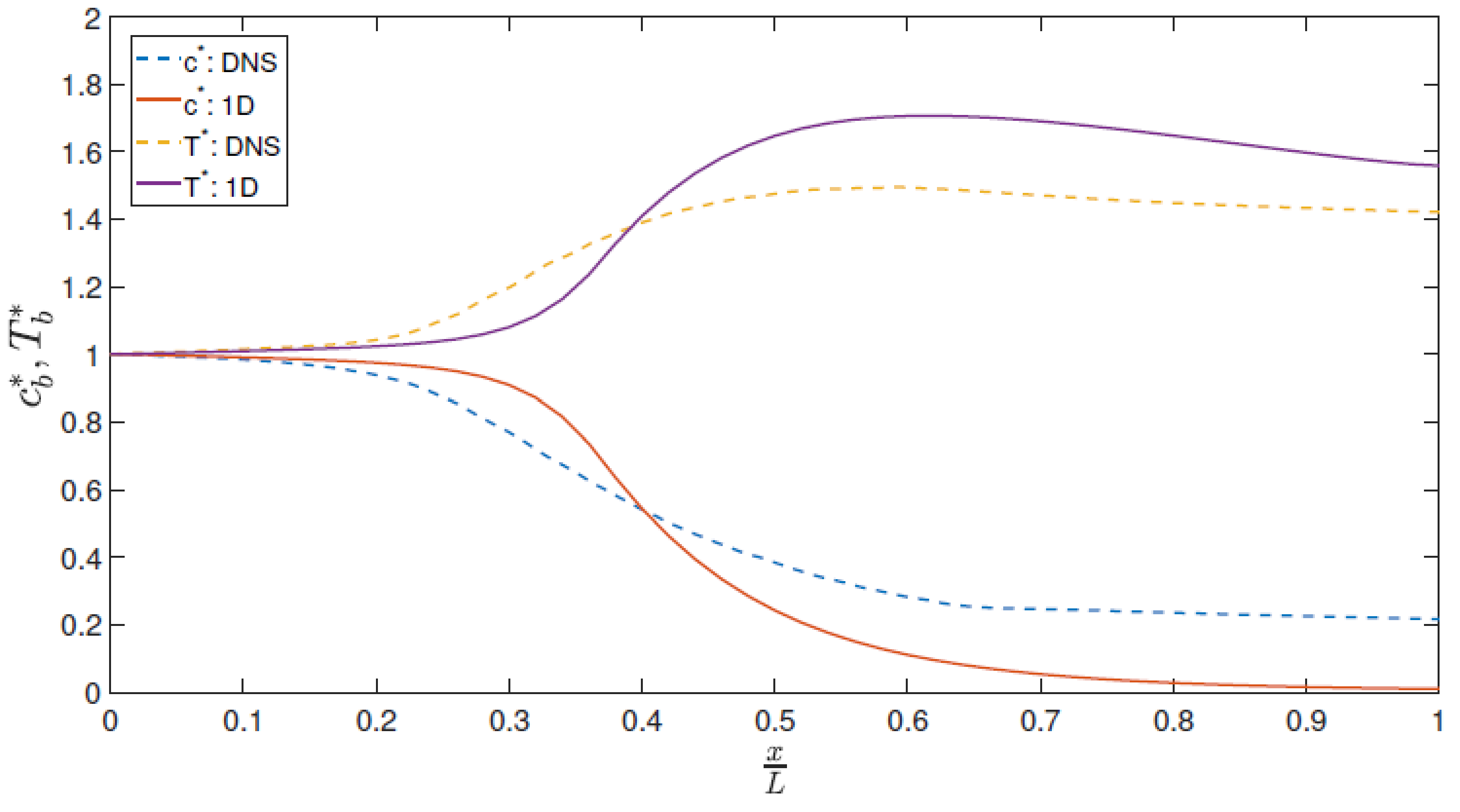
$$\frac{x}{L} = 0.75$$



Chandra et al., 2020, CEJ

IBM BASED DNS MODEL

full bed simulations at ϕ_0=1.0 and comparison with 1D heterogeneous model



1D heterogeneous model uses empirical closures for fluid-particle mass & heat transfer coefficients, heat & mass dispersion coefficients and wall-to-bed heat transfer coefficient

Chandra et al., 2020, CEJ

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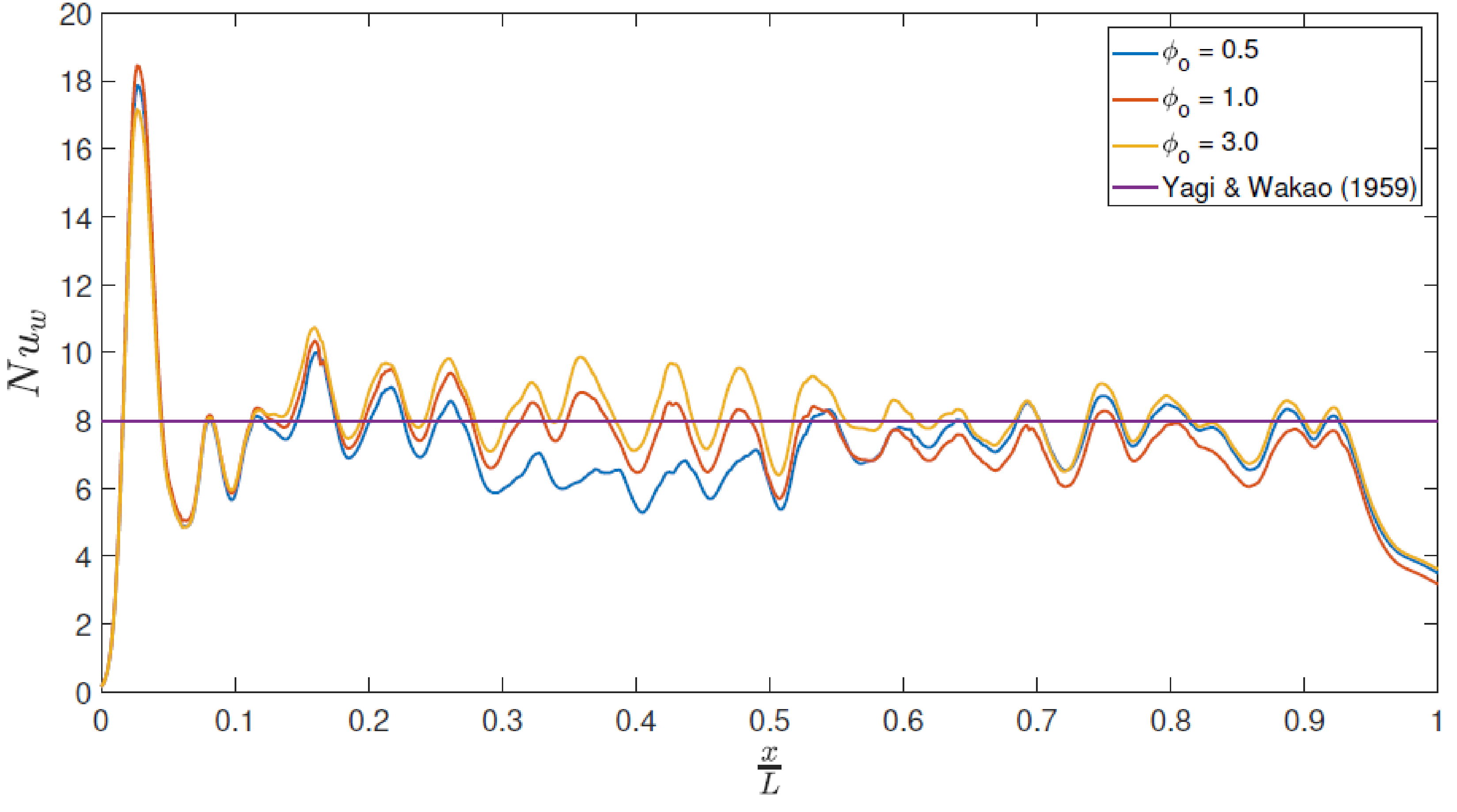
IBM BASED DNS MODEL

comparison of computed wall-to-bed heat transfer with Yagi & Wakao empirical correlation



Chandra et al., 2020, CEJ

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CONCLUSIONS

- DNS OF FIXED BED CHEMICAL REACTORS

  + powerful tool for advancing fundamental understanding

- EXPERIMENTAL VALIDATION

  + important role for non-invasive monitoring (MRI)

- MAJOR CHALLENGES

  + coupling to complex catalytic chemical reactions (MCEC, ARC CBBC)

  + closure development + improvements for phenomenological design models

  + imaging of multiphase flows with catalytic chemical reactions

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