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

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PSYCHE WORKSHOP

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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)

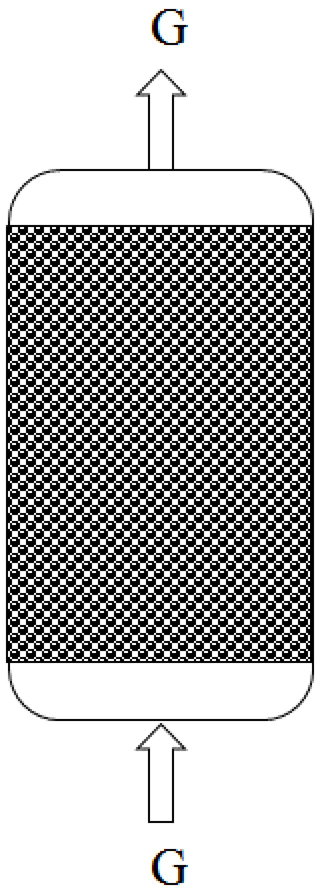
oxychlorination of ethylene (vinyl chloride)

alkylation of aromatic compounds (ethylbenzene)

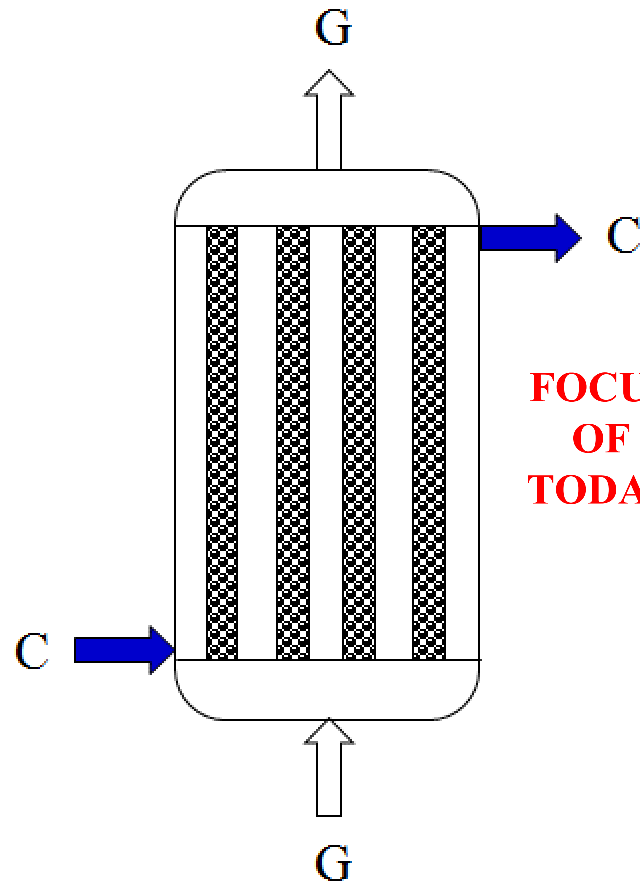
ammonia oxidation in nitric acid synthesis

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)

1D phenomenological (design) models → **Closures required for fluid-particle interaction, dispersion**

2D phenomenological (design) models → **Closures required for fluid-particle interaction, dispersion**

Volume-averaged or particle un-resolved models → **Fluid-particle interaction Closures required**

FOCUS OF TODAY → **DNS or Particle Resolved Models** → **Fluid-particle interaction Closures obtained**

empirical input

more detail

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

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

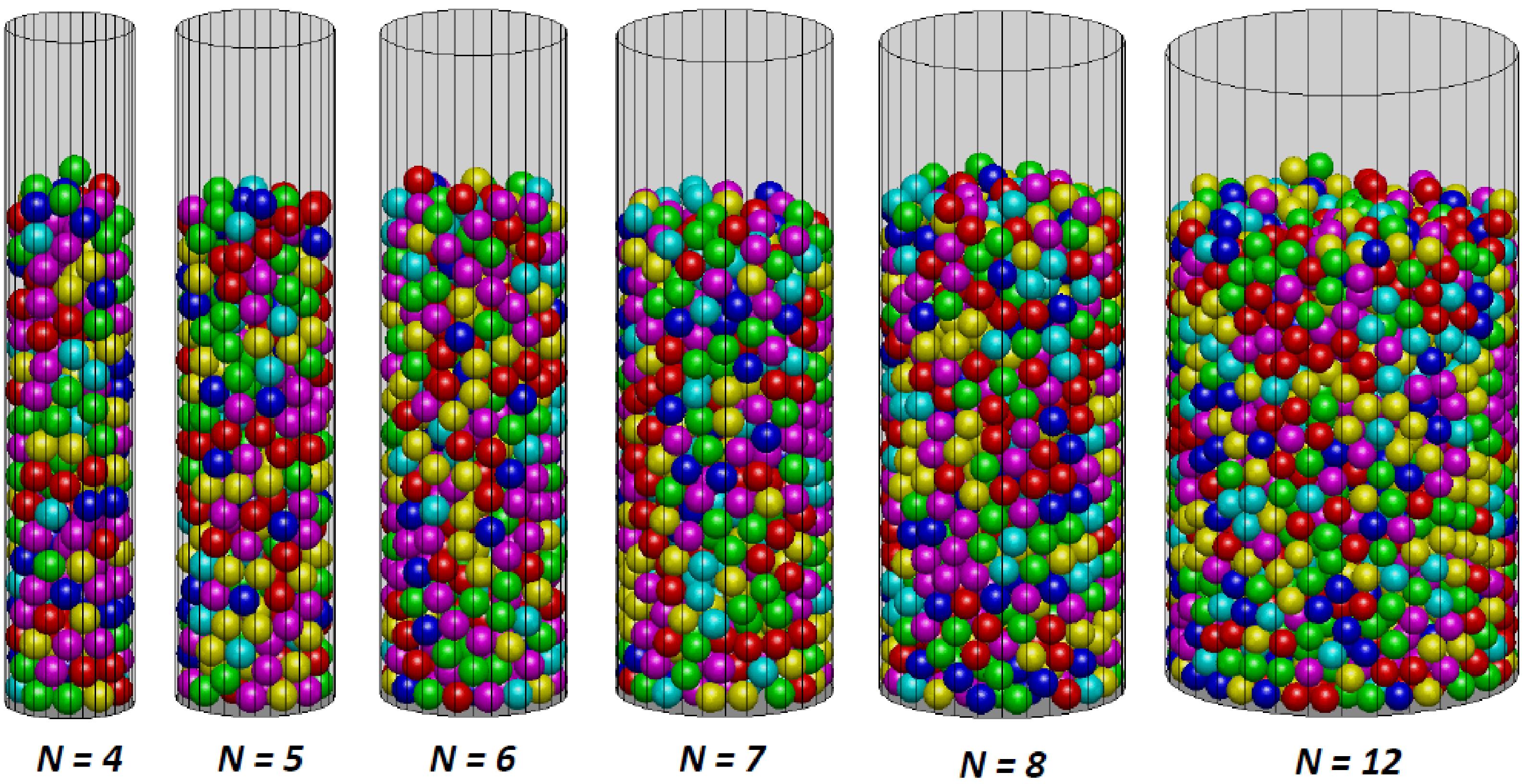
Arrhenius dependence of reaction rate constant

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

radiative heat transport can be neglected

IBM BASED DNS MODEL

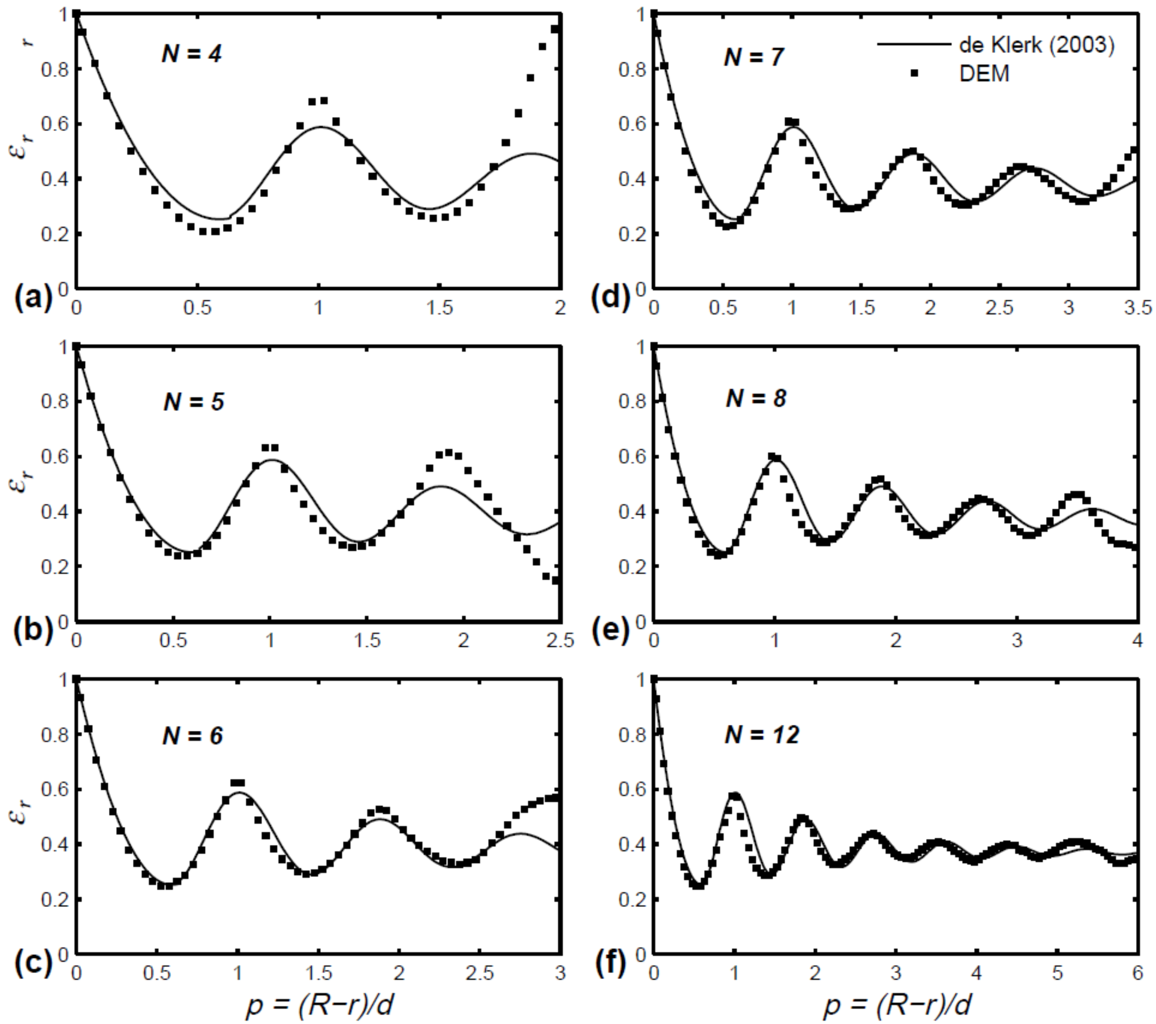
transport phenomena in packed bed reactors: DEM generated beds



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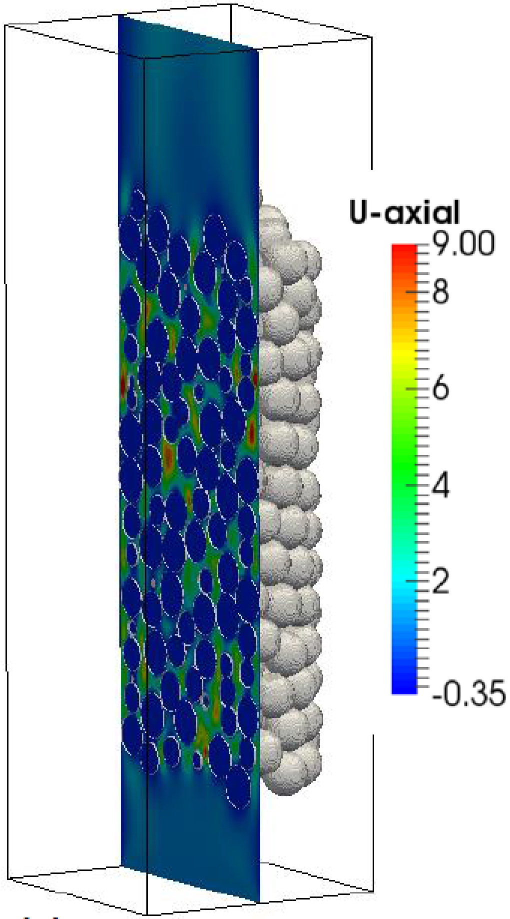
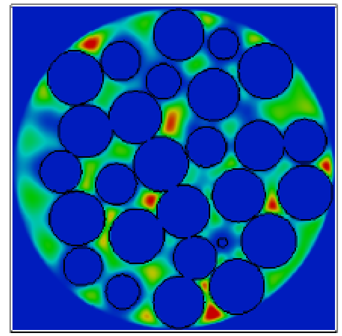
transport phenomena in packed bed reactors: velocity profiles (N=6)



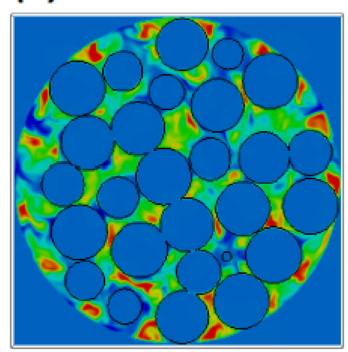
(a)

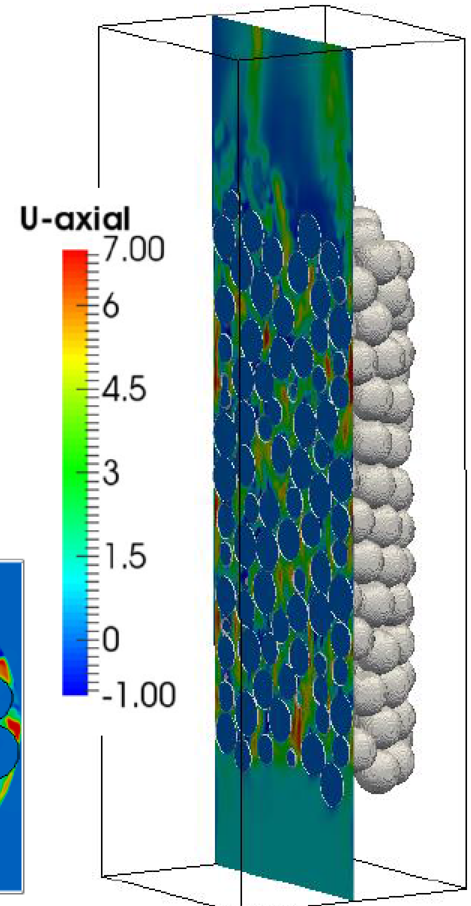
Re_d=1



(a)

(b)





(b)

Re_d=500

Das et al., 2017, CES

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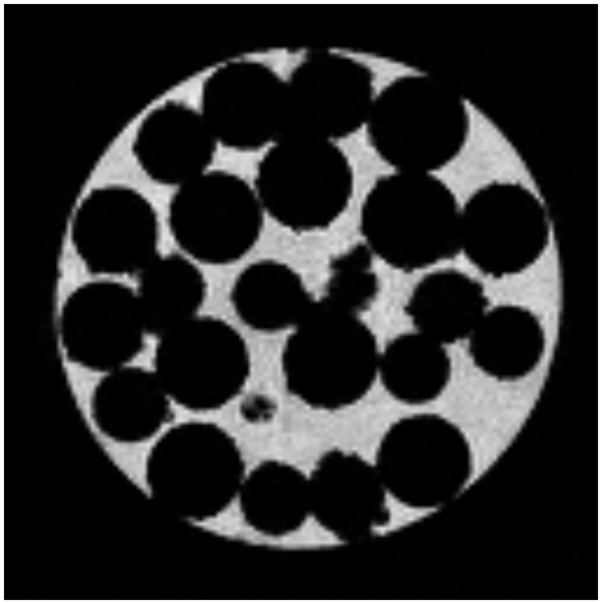
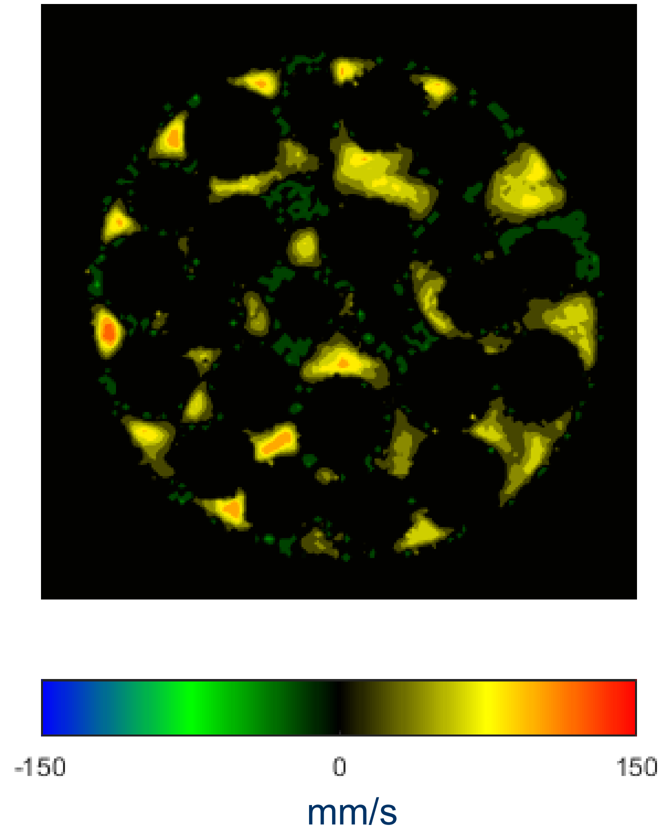
comparison with experiment (MRI flow imaging)



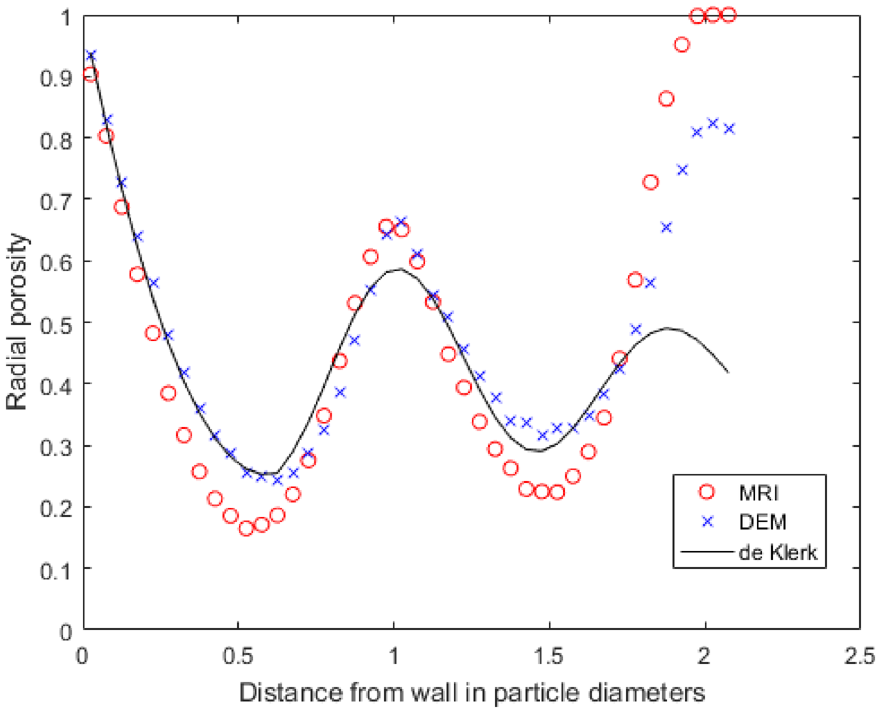


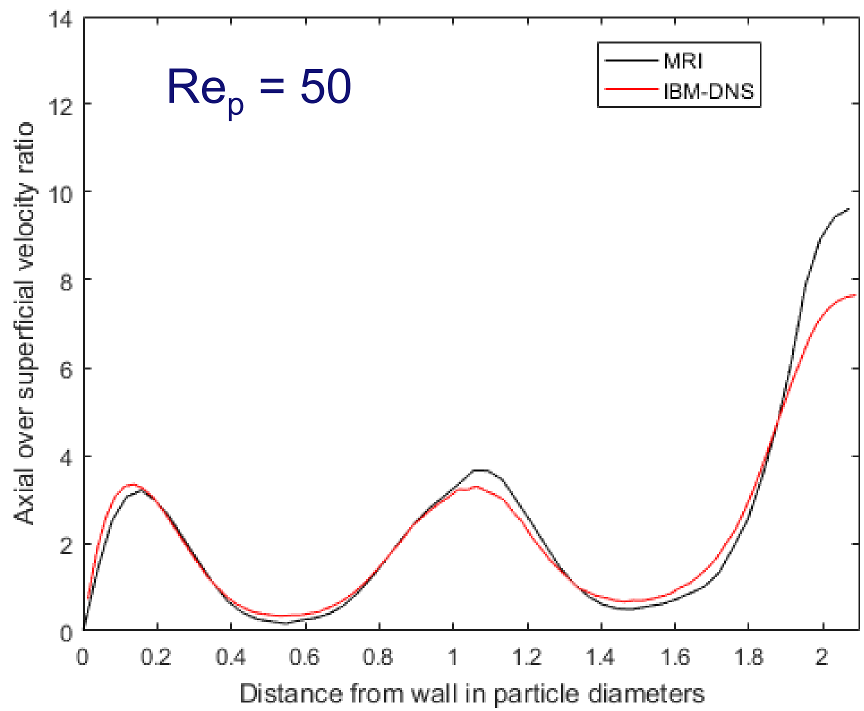
phase fractions (left) and axial velocity map (right) for a packed bed of spheres with a diameter of 4 mm

Lovreglio et al., 2018, AIChEJ

IBM BASED DNS MODEL

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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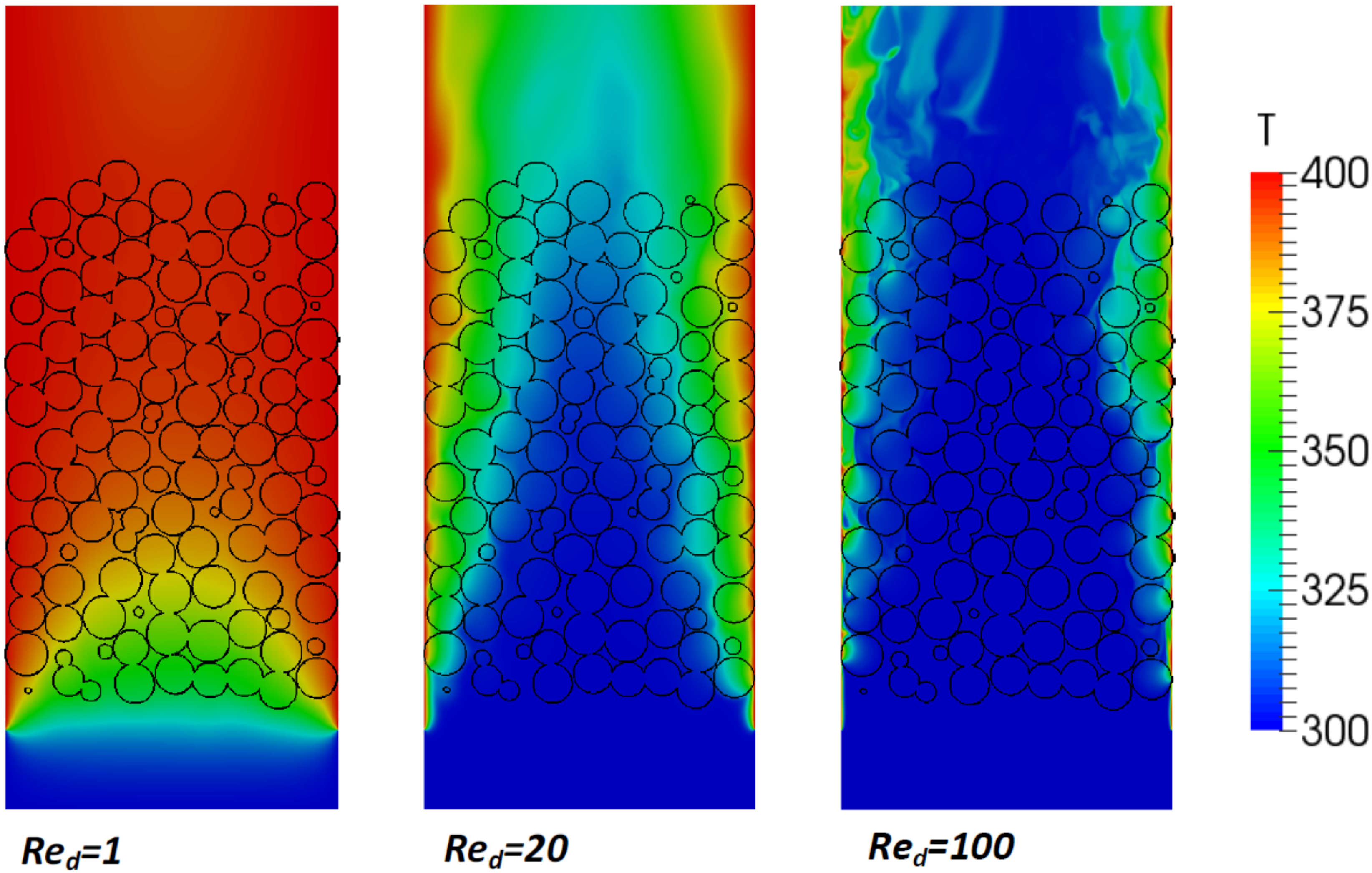
IBM BASED DNS MODEL

transport phenomena in packed bed reactors: temperature profiles



conjugate heat transport in fluid and solid phase

Das et al., 2017, CES

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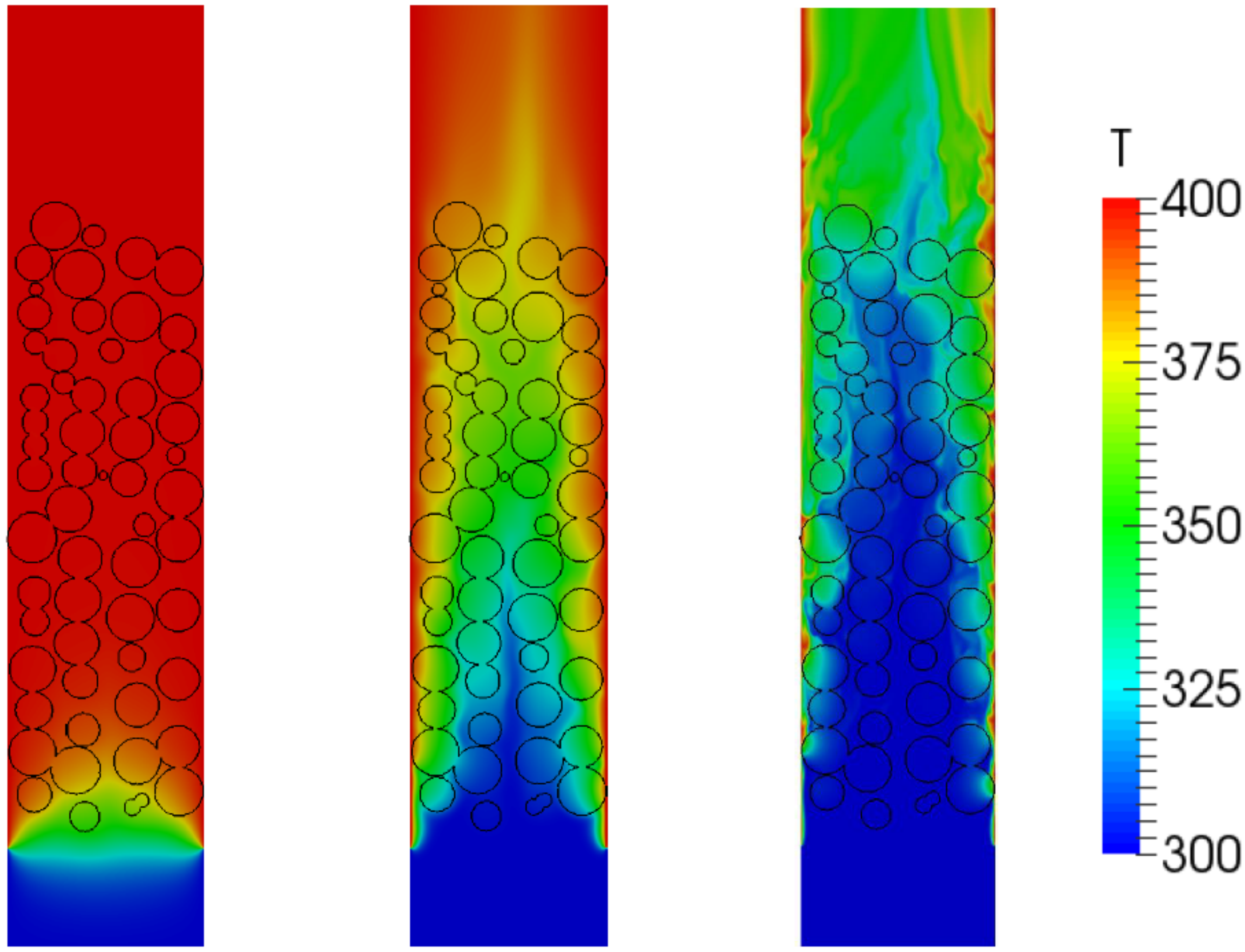
transport phenomena in packed bed reactors: temperature profiles



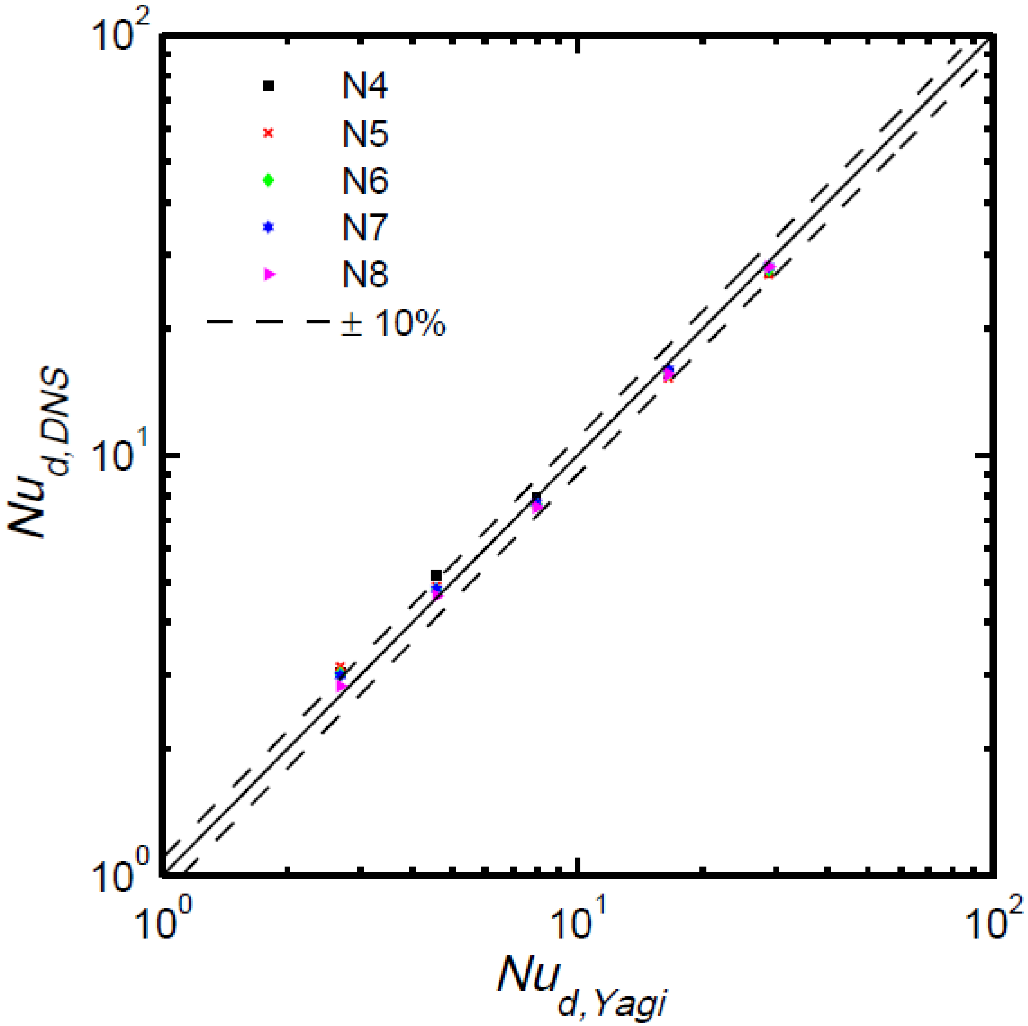
$Re_d=1$ $Re_d=20$ $Re_d=100$

conjugate heat transport in fluid and solid phase

Das et al., 2017, CES

IBM BASED DNS MODEL

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



Das et al., 2017, CES

IBM BASED DNS MODEL

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

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

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

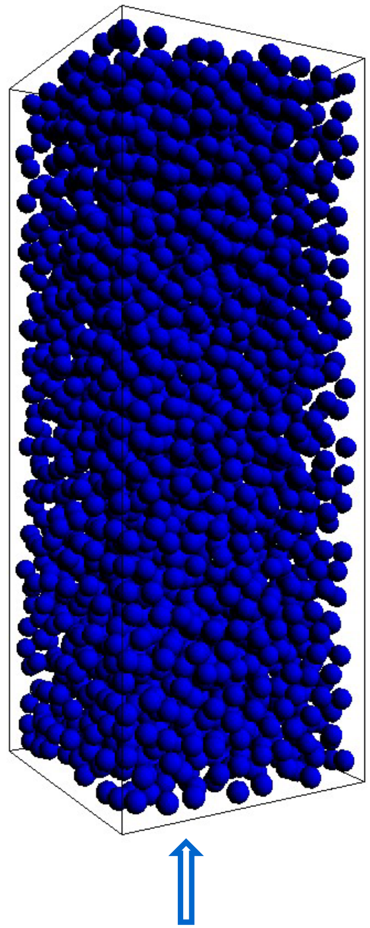
$$Sh_p = \left(7 - 10\varepsilon_b + 5\varepsilon_b^2\right)\left(1 + 0.7\,\mathrm{Re}_p^{0.2}\,Sc^{1/3}\right) + \left(1.33 - 2.4\varepsilon_b + 1.2\varepsilon_b^2\right)\mathrm{Re}_p^{0.7}\,Sc^{1/3}$$

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

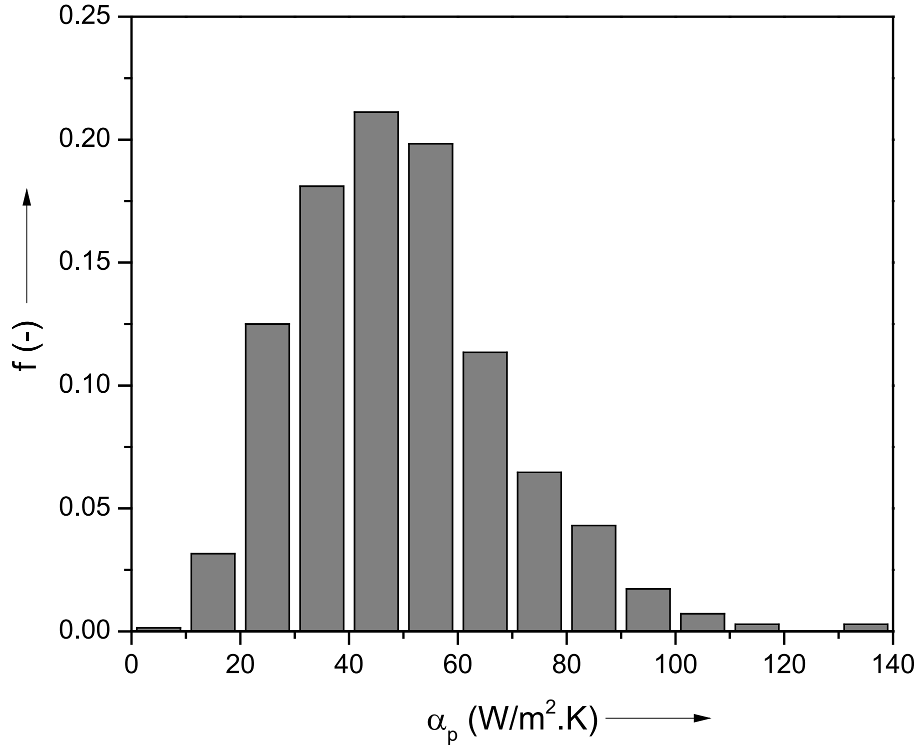
Deen & Kuipers, 2014, CES

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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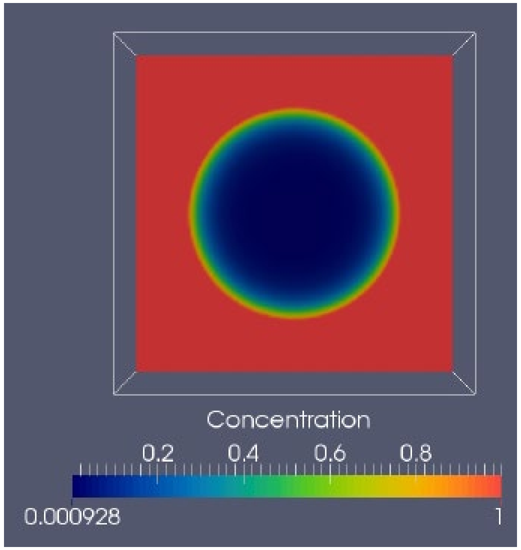
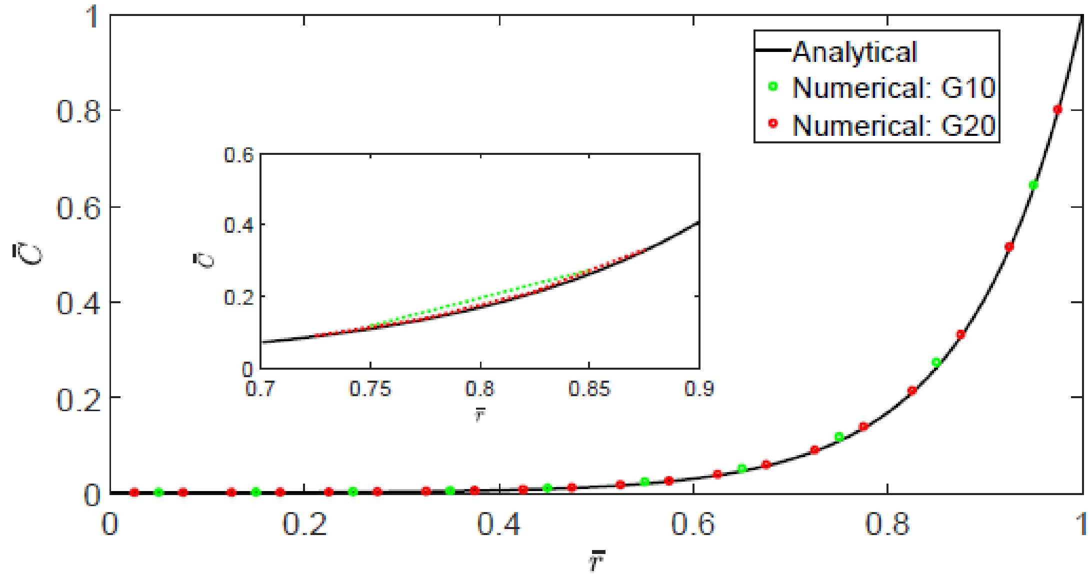
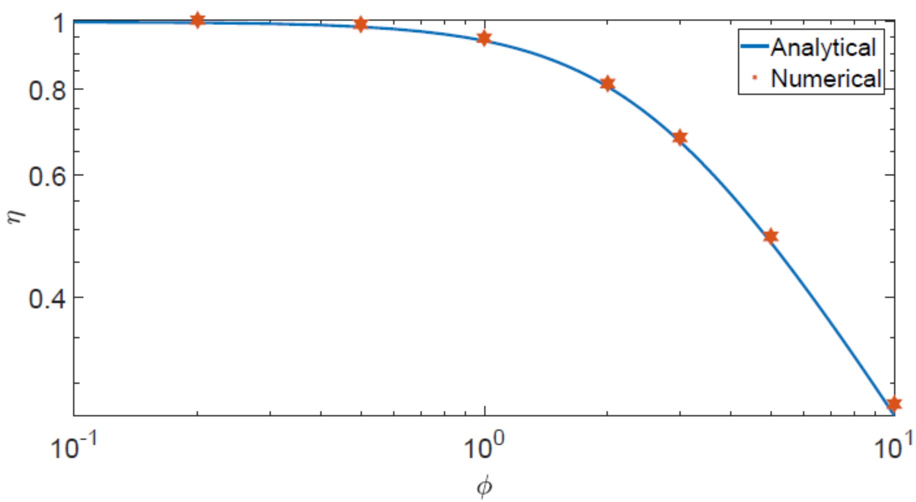
isothermal reaction with diffusion limitation in single particle







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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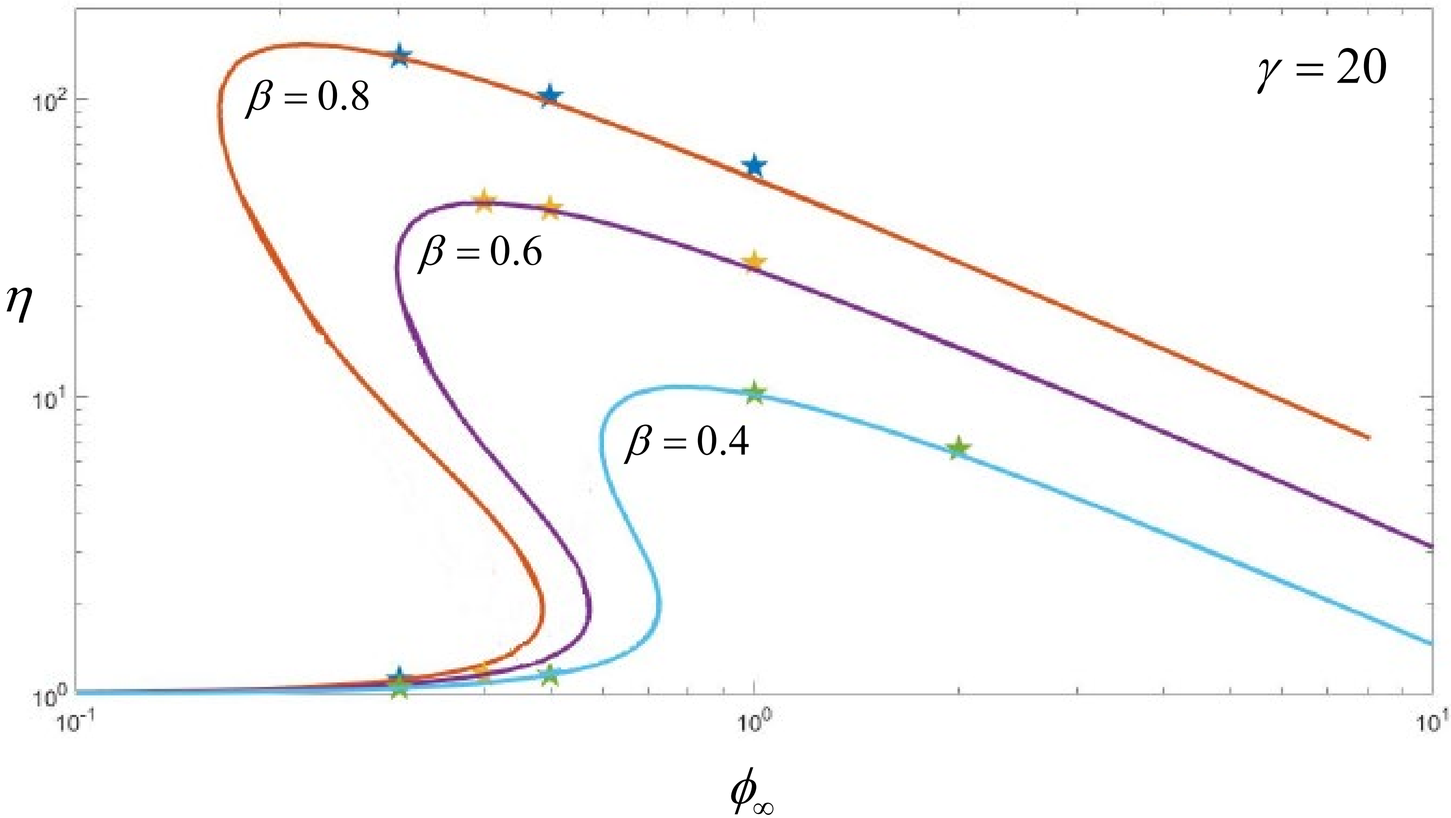
effectiveness factor versus Thiele modulus (single sphere)



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

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

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

Chandra et al., 2020, CEJ

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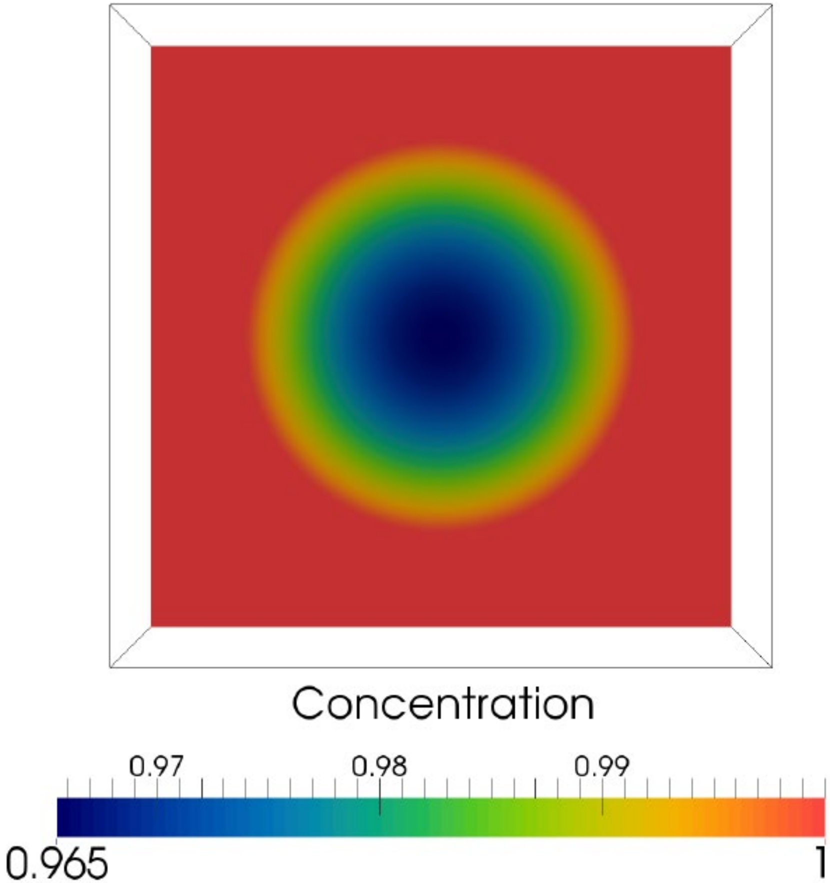
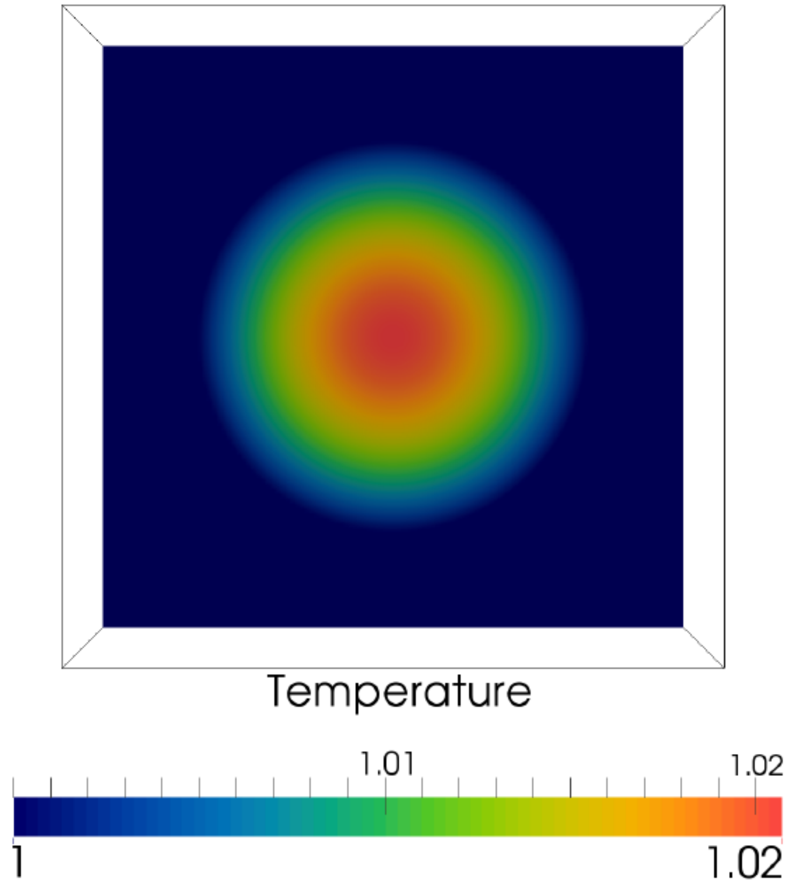
concentration and temperature distributions (single sphere)





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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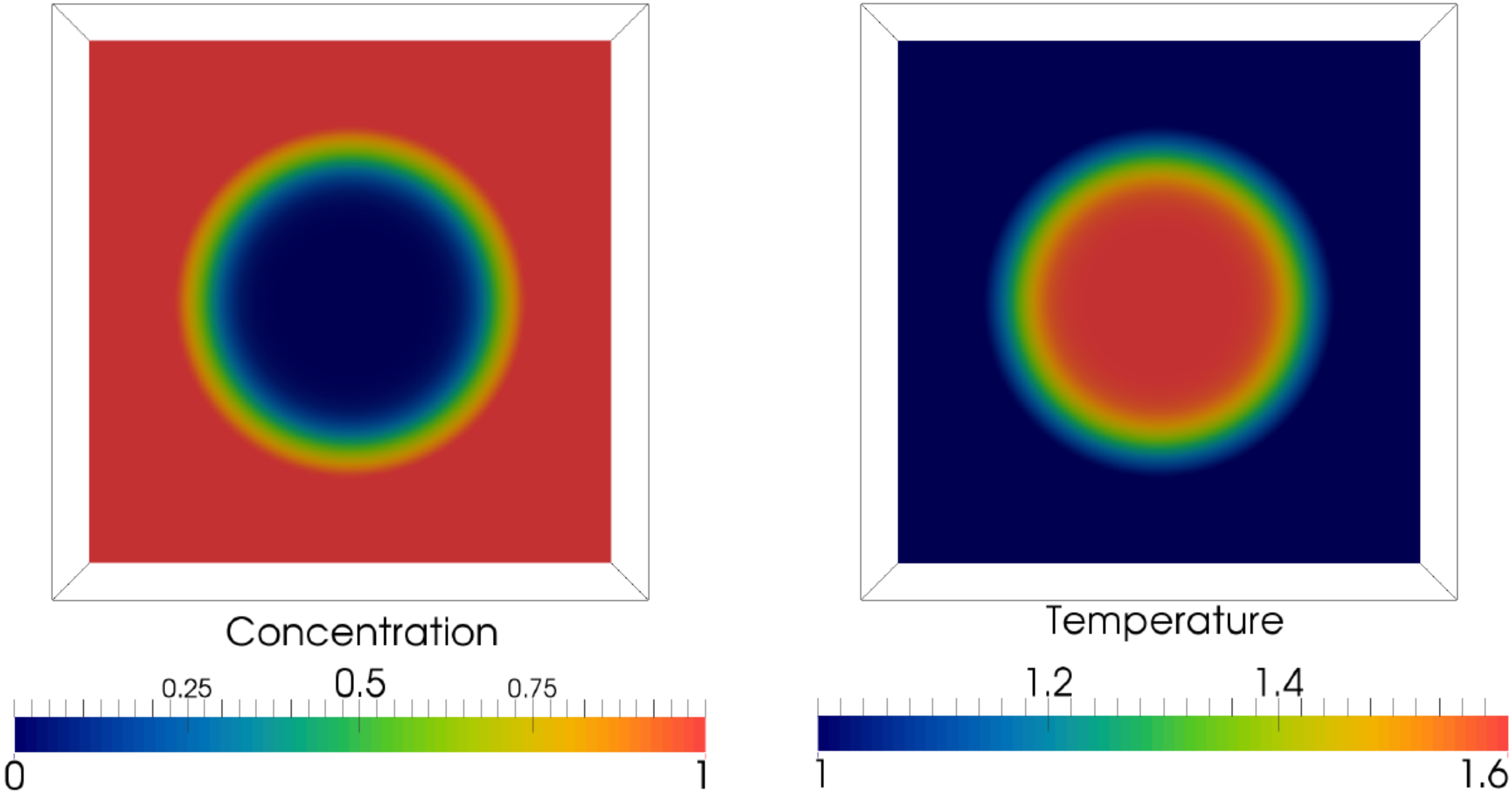
IBM BASED DNS MODEL

concentration and temperature distributions (single sphere)



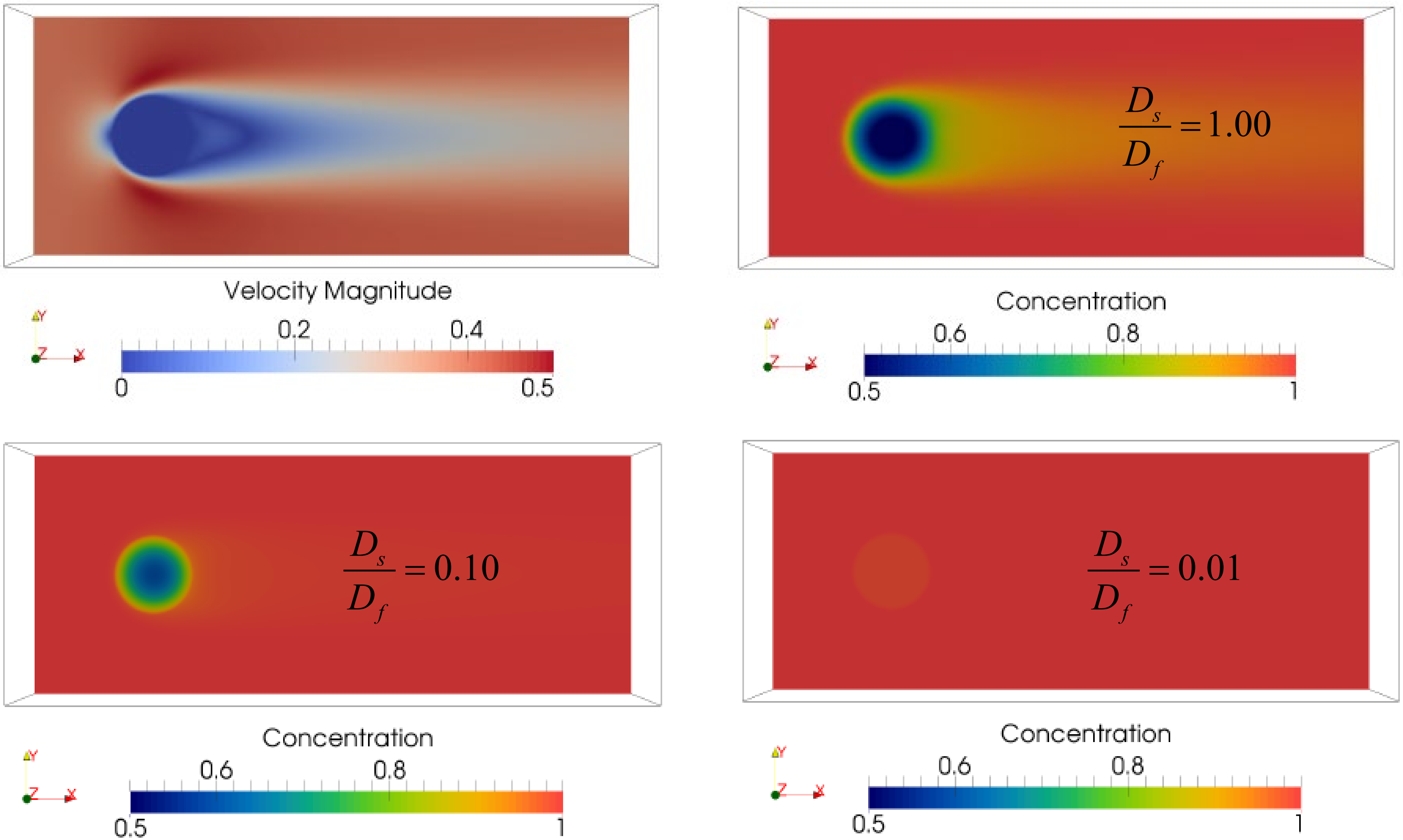
higher stable steady state:

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

Chandra et al., 2020, CEJ

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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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

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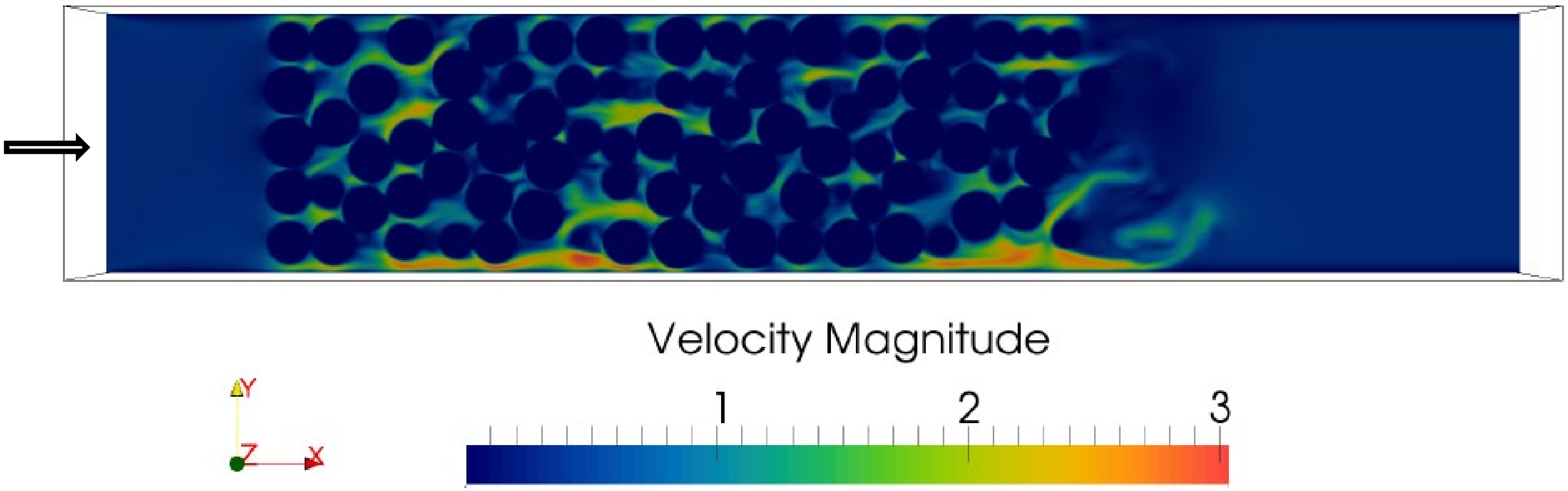
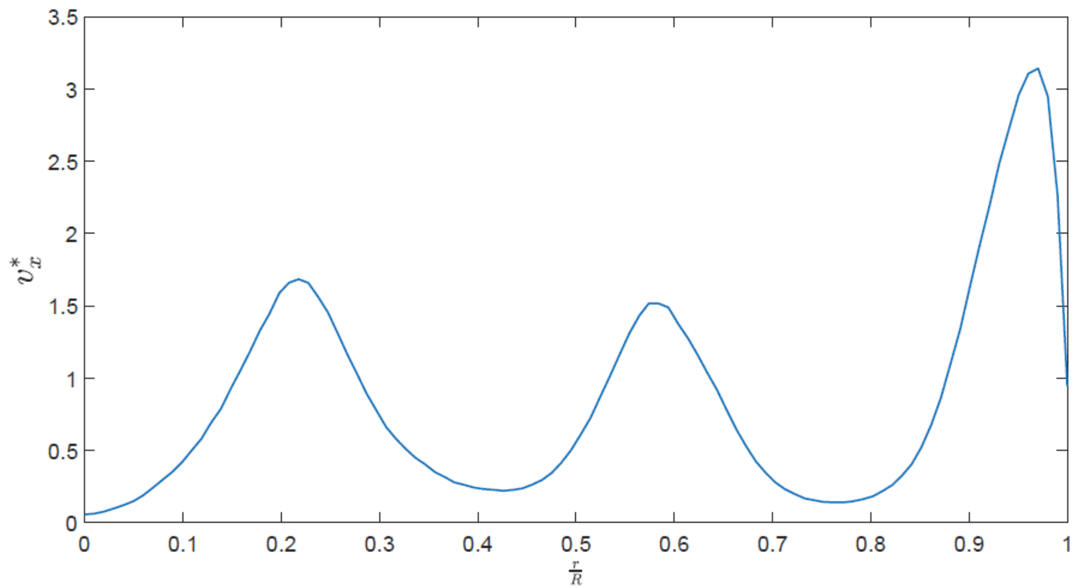
full bed simulations: flow field





radial profile of the azimuthally averaged superficial axial velocity component

Chandra et al., 2020, CEJ

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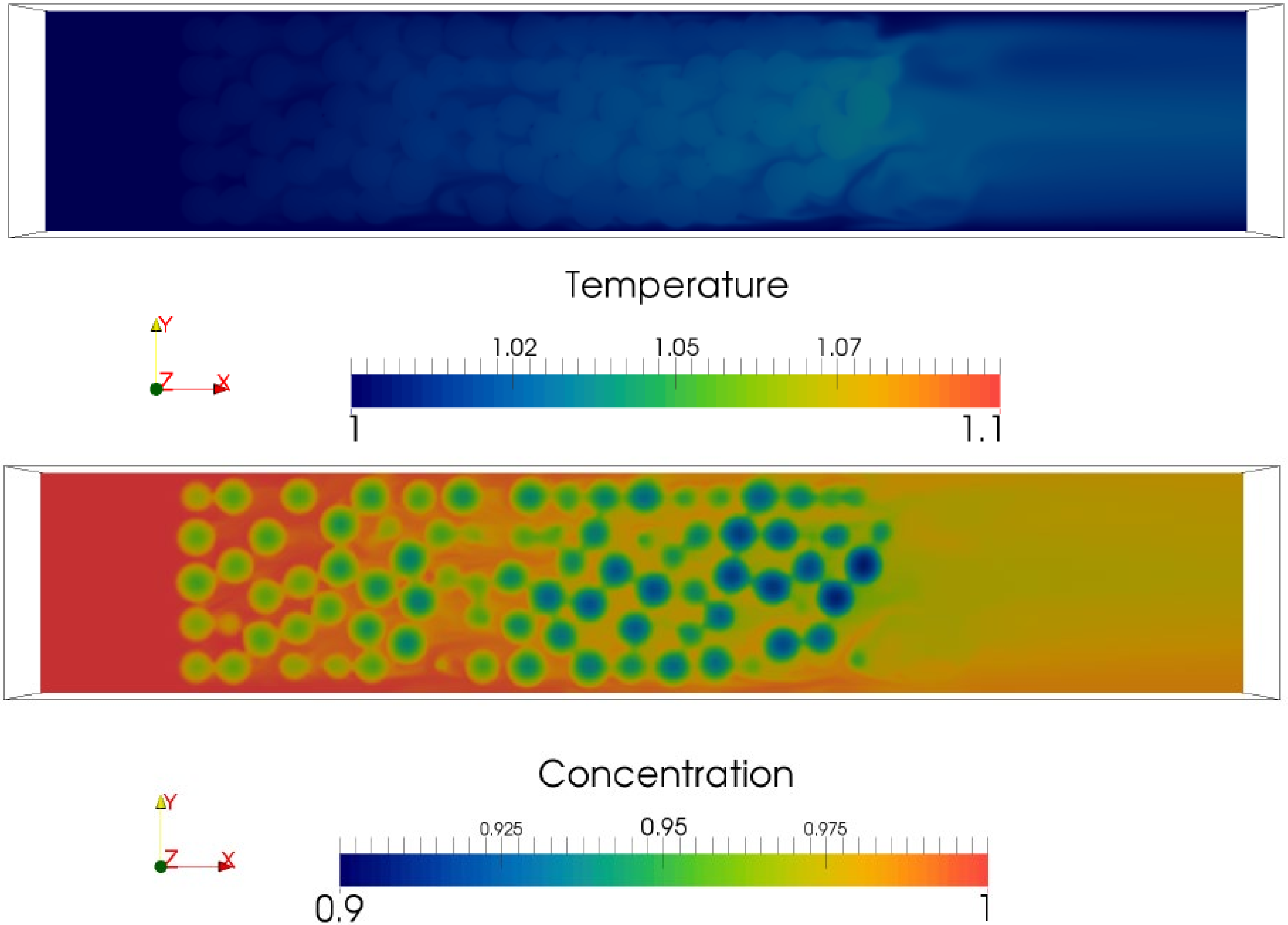
full bed simulations at ϕ_0=0.5: temperature and concentration distributions in central plane



Chandra et al., 2020, CEJ

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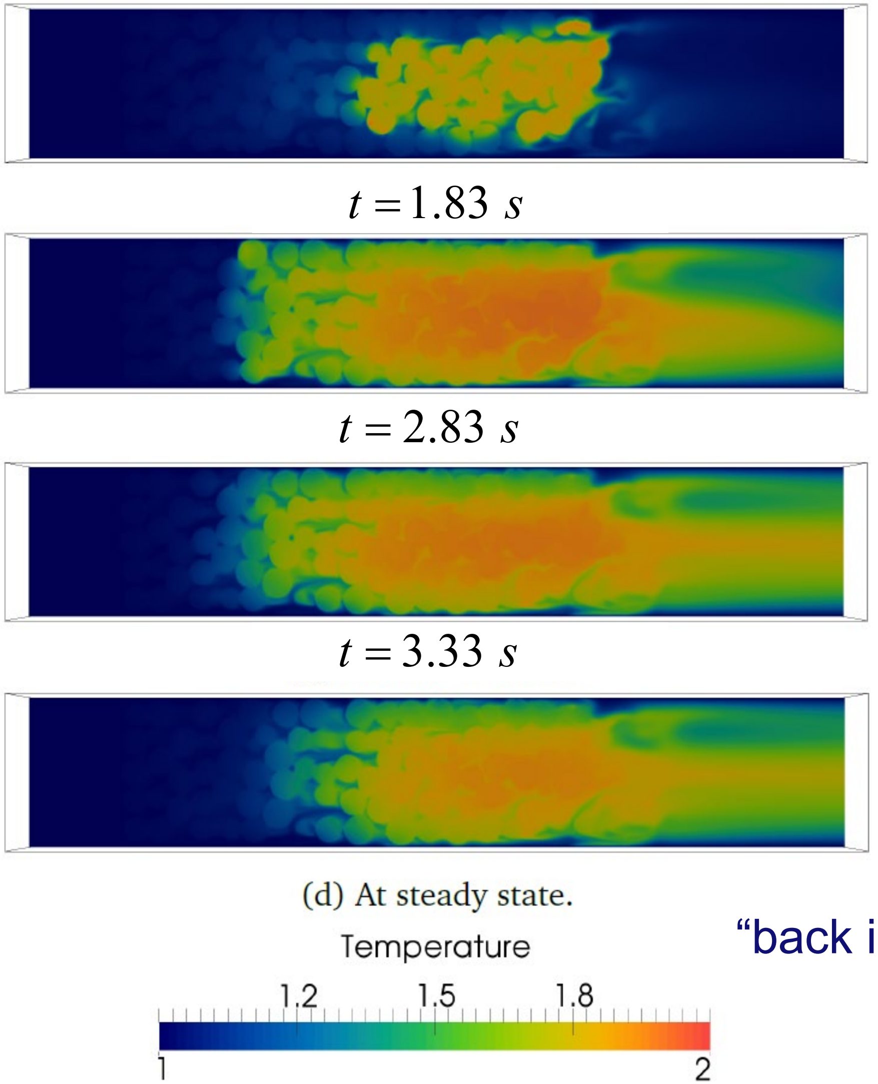
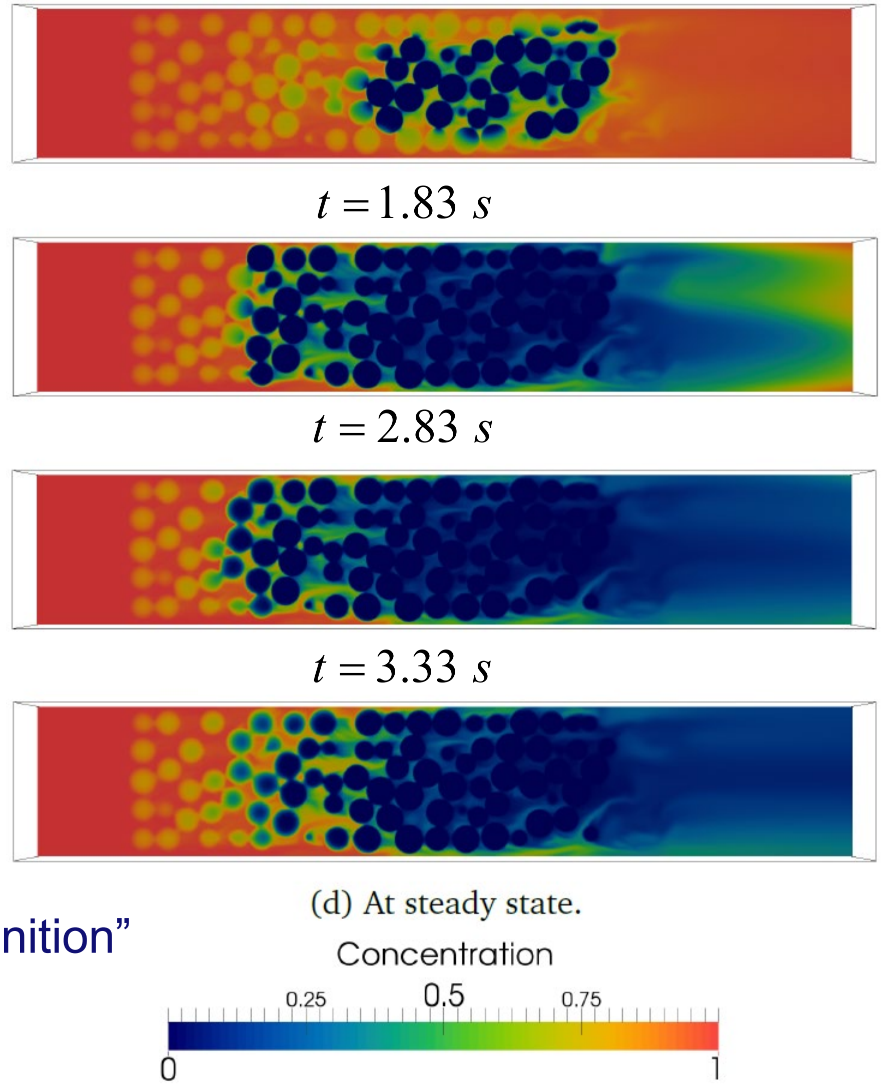
full bed simulations at ϕ_0=1.0: transient evolution of temperature and concentration distributions in central plane





“back ignition”

Chandra et al., 2020, CEJ

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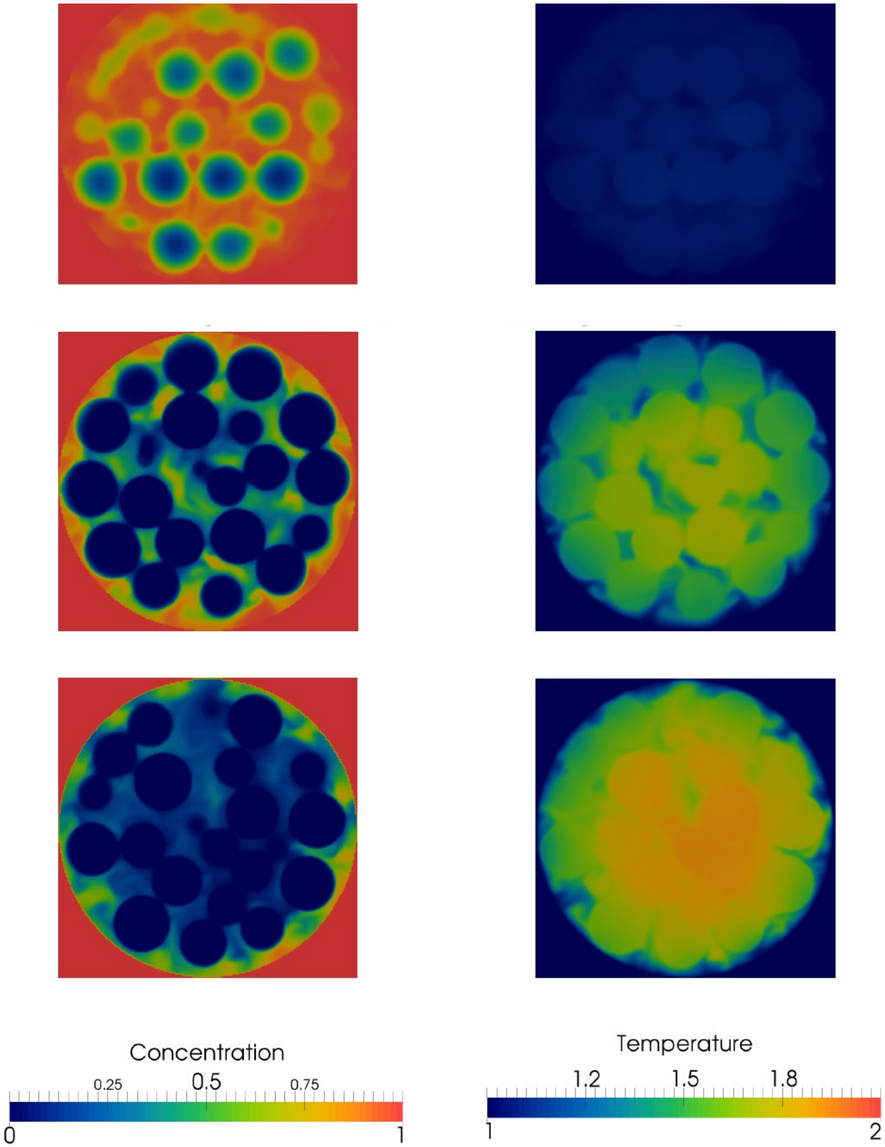
full bed simulations at ϕ_0=1.0: cross-sectional profiles of concentration and temperature

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

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

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



Chandra et al., 2020, CEJ

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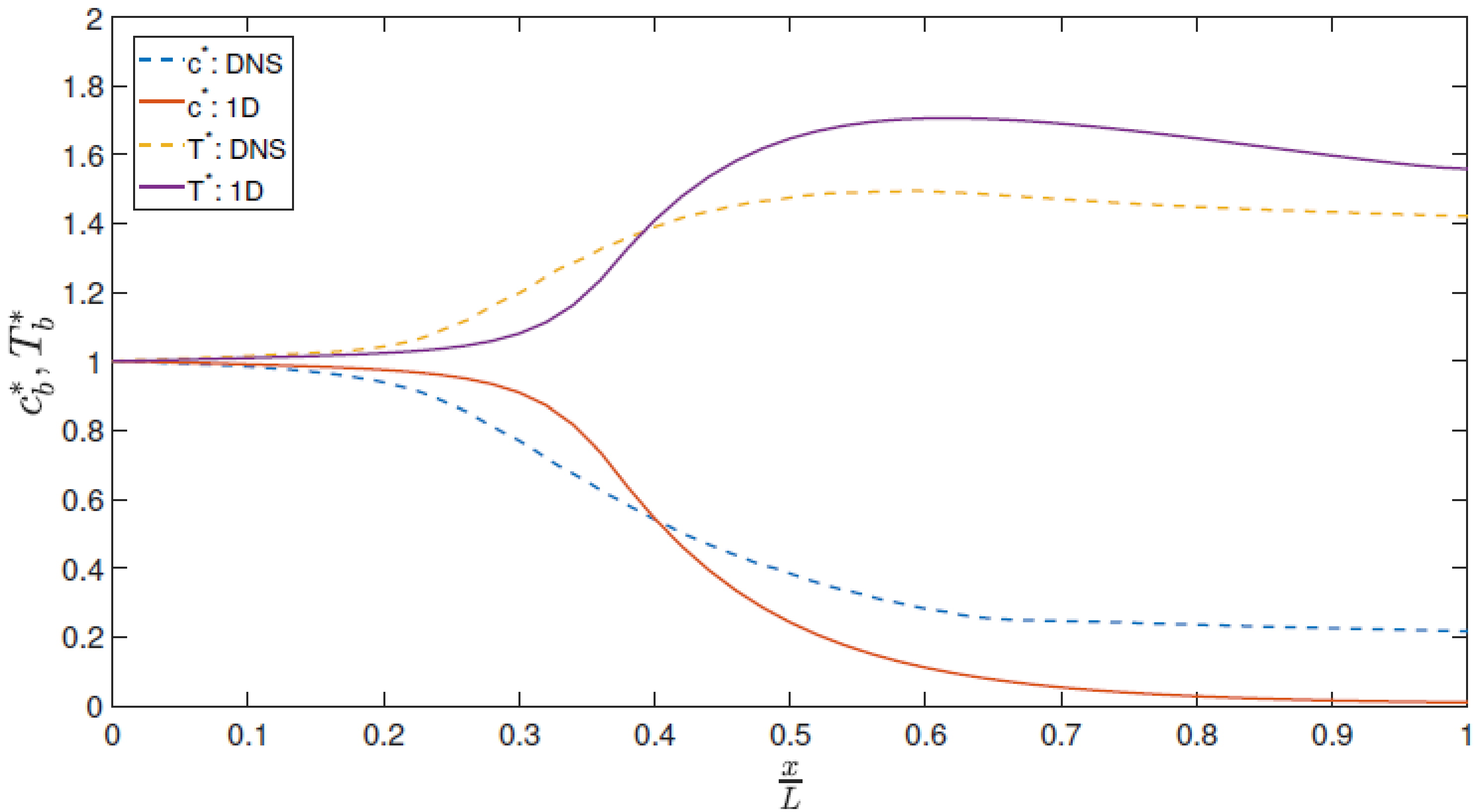
full bed simulations at ϕ_0=1.0 and comparison with 1D heterogeneous model



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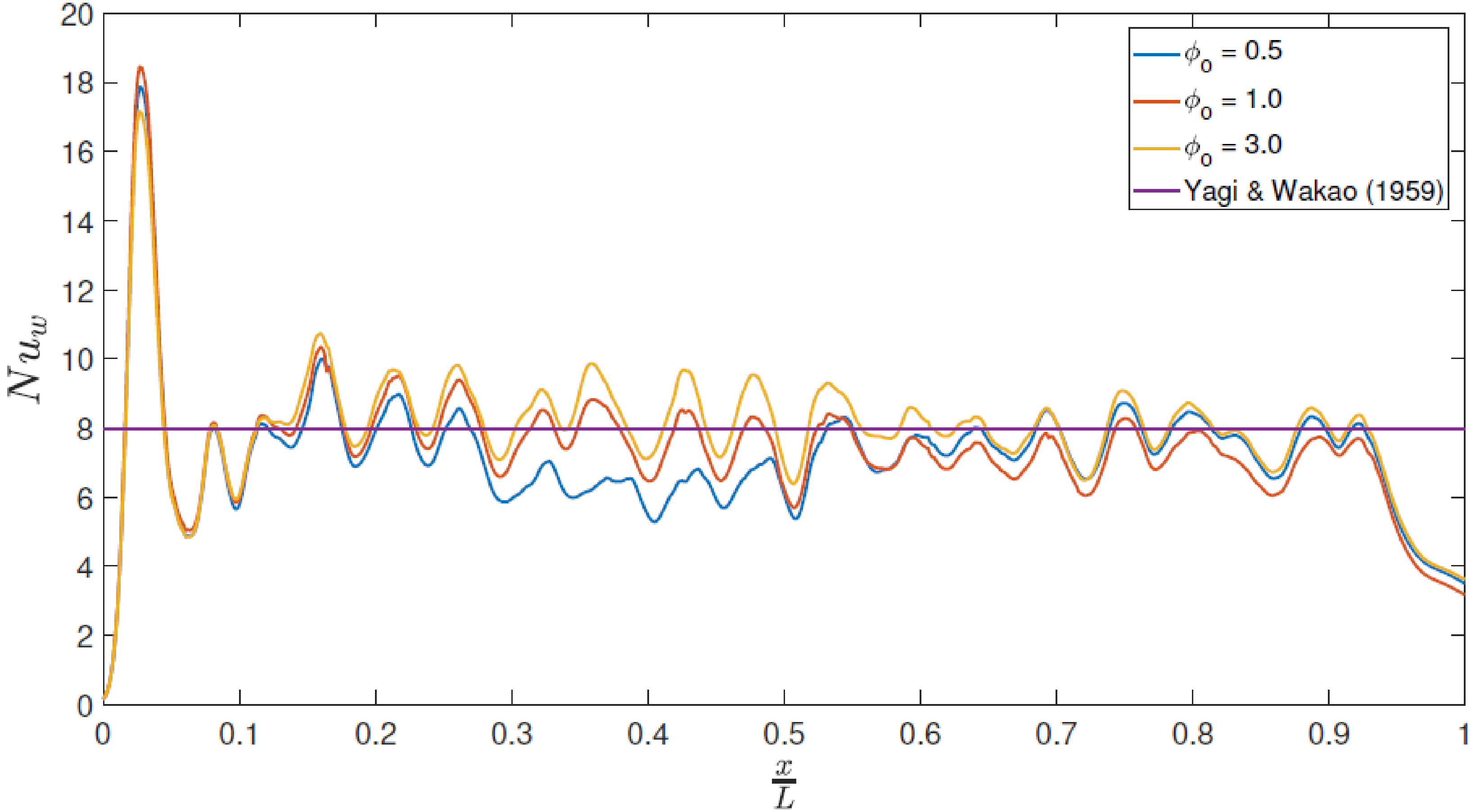
comparison of computed wall-to-bed heat transfer with Yagi & Wakao empirical correlation



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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ARC CBBC
ERC
MCEC
NWO
Shell

ADDITIONAL SLIDES

BACKUP

IBM BASED DNS MODEL
governing equations fluid phase

- CONTINUITY EQUATION

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

- MOMENTUM EQUATION

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

- 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

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

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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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closure equations

- FLUID-PARTICLE DRAG

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

separate evaluation of friction drag and pressure drag

- FLUID-PARTICLE TORQUE

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

- FLUID-PARTICLE HEAT AND MASS TRANSFER RATES

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

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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

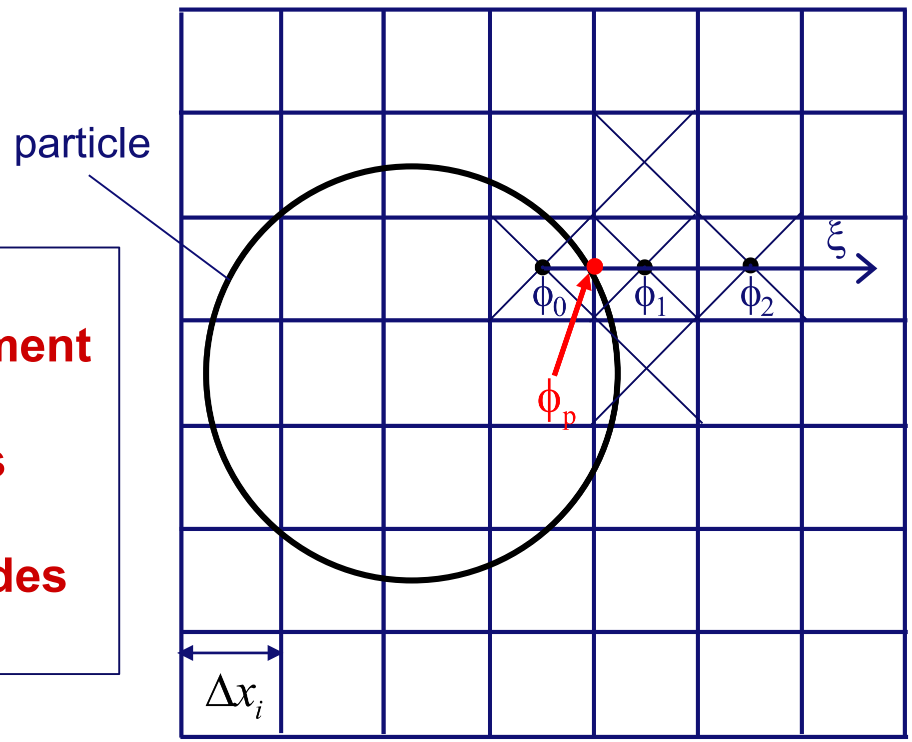
+ staggered computational mesh

+ 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

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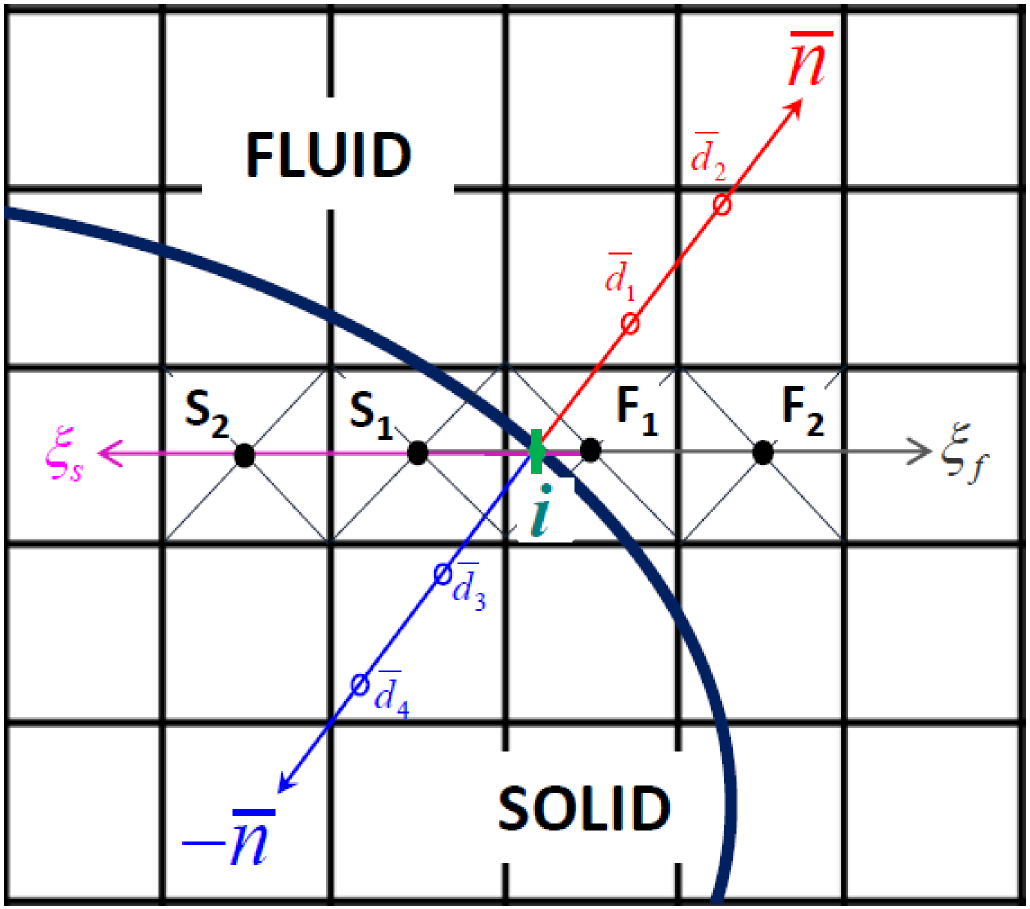
fluid-solid coupling with intra-particle scalar transport



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

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