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Particle-resolved transient simulations of reactive transport in non-adiabatic packed bed reactors

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Introduction

Packed bed reactors are of high interest because of their wide use in the chemical and process industry. The selective oxidation of ethylene or the steam reforming of methane are just two of its applications. However, the widespread use is no guarantee for in depth knowledge about the multiscale processes happening inside. The processes taking place are the transport of momentum, heat, and species mass in between the pellets (interparticle) as well as the transport of heat and species, coupled via reactions, inside the pellets (intraparticle). The interparticle processes and the intraparticle processes are highly coupled, e.g. the temperature distribution in the bed depends on the heat conduction from the wall and the convective transport outside the particles, but the temperature of the particles is also influenced by the exothermic/endothermic reactions inside the particle. If the particles are not cooled sufficiently in the case of an exothermic reaction, a hot spot can be formed as the reaction creates heat which speeds up the reaction and hence creates more heat. Achieving the coupling is one of the challenges the review paper of Jurtz et al. [1] identifies in recent (2006-2019) studies of packed beds employing Computational Fluid Dynamics (CFD). Another challenge it identifies, is the simulation of transient operations, deeming them too computational expensive. We have tried to tackle these two challenges recently using particle resolved CFD [2]. In this talk, we will summarize that work, as well as highlight the challenges we currently try to solve.

Model description

The system that is studied consists of a fixed bed of 340 spheres in a slender column having a tube-to-particle diameter ratio of 5. This bed is generated via a Discrete Element Method as detailed in [3]. The sphere locations are mapped to the computational domain where we solve the transport equations on a uniform 3D Cartesian grid. To obtain the pressure and velocity we solve the incompressible Navier-Stokes equations (equation 1 and 2):

$$\nabla \cdot \boldsymbol{v} = 0 \tag{1}$$

$$\rho_f \frac{\partial \boldsymbol{v}}{\partial t} + \rho_f \boldsymbol{v} \cdot \nabla \boldsymbol{v} = -\nabla P - \nabla \cdot \boldsymbol{\tau} \tag{2}$$

Using a no slip boundary condition on the surface of the spheres and the tube wall, which is imposed via an Immersed Boundary Method (IBM) [4]. The solution is obtained via a two-step projection method which is detailed in [4].

The governing equations for mass and heat transport are given in equation 3 and 4.

$$\frac{\partial c_i}{\partial t} + \boldsymbol{v} \cdot \nabla c_i = \nabla \cdot (D_i \nabla c_i) - k c_i \tag{3}$$

$$\rho_i C_{pi} \frac{\partial T_i}{\partial t} + \rho_i C_{pi} \boldsymbol{v} \cdot \nabla T_i = \nabla \cdot (k_i \nabla T_i) - k c_i \Delta H_r \tag{4}$$

With the convection term being zero when i is solid (s), and the reaction term being zero when i is fluid (f). The internal transport in the solid is not resolved to the pore scale and hence an effective diffusion coefficient is used. The effective coefficient follows from $\frac{D_s}{D_f} = \frac{\varepsilon}{\tau}$ where ε and τ are respectively the

porosity and tortuosity which are generally experimentally determined. k is the reaction rate constant for the first-order reaction which is coupled to the temperature profile via an Arrhenius dependence.

The solid and fluid phase are coupled by imposing the continuity of mass and heat fluxes and the continuity of temperature and concentration at the interface:

$$-D_f \nabla c_f \cdot \boldsymbol{n}_{sf} = -D_s \nabla c_s \cdot \boldsymbol{n}_{sf}; \; c_f = c_s \text{ at solid-fluid interface} \tag{5}$$

$$-k_f \nabla T_f \cdot \boldsymbol{n}_{sf} = -k_s \nabla T_s \cdot \boldsymbol{n}_{sf}; \; T_f = T_s \text{ at solid-fluid interface} \tag{6}$$

To apply these boundary conditions a ghost-cell immersed boundary method is applied. To summarize this method, we consider equations 3-4 in their discretised form as given in equation 7.

$$a_p \phi_p + \sum_{nb} a_{nb} \phi_{nb} = b_p \tag{7}$$

with ϕ representing c_f, c_s, T_f or T_s. For cells close to the interface, one of the neighbours (subscript nb) could be in the other phase. For this neighbour (here labelled with subscript 0), we substitute a value following from a second order polynomial interpolation (see [4]):

$$\phi_0 = -\frac{2\xi_s}{1-\xi_s}\phi_1 + \frac{\xi_s}{2-\xi_s}\phi_2 + \frac{2}{(1-\xi_s)(2-\xi_s)}\phi_{sf} \tag{8}$$

where ϕ_1 is ϕ_p, ϕ_2 is the value at the opposing neighbour and ϕ_{sf} is the value at the interface. ξ_s is the dimensionless distance from the neighbour to the interface. As the value at the interface is not known, ϕ_{sf} is determined using a fit of two second order polynomials in the positive and negative direction normal to the surface, i.e. one into the solid phase and one into the fluid phase. The fit is obtained by probing the values of ϕ at distances Δx and $2\Delta x$ from the interface using trilinear interpolation from the values at the grid points. The probes together with the boundary conditions close the system of the two polynomials and lead to the following equation for the surface value:

$$\phi_{sf} = \frac{\Gamma_f\left(4\phi_{f1} - \phi_{f2}\right) + \Gamma_s\left(4\phi_{s1} - \phi_{s2}\right)}{3\left(\Gamma_f + \Gamma_s\right)} \tag{9}$$

with Γ either k or D. In case the second fluid probe lies in another solid, a first order fit is used.

The surface value is calculated explicitly, but the ϕ_1 and ϕ_2 appearing in equation 8 are taken into account implicitly ensuring the stability of the numerical technique.

Results
We performed two simulations in the slender packed bed with different Thiele modulus (φ): 0.5 and 1.0. The Thiele modulus is based on the Arrhenius pre-factor as proposed by [5]. All other dimensionless numbers are given in Table 1.

For $\varphi = 0.5$, the rate of reaction is not fast enough for particles to self-ignite. This means the particles reside in the lower steady state and the enhancement compared to when the reaction would be carried out at inlet conditions is minimal ($\eta = \frac{\int_{V_s} kc\,dV}{k_0 c_0 V_s} = 1.25$).

For $\varphi = 1$, we do observe self-ignition which results in a creeping reaction zone [6]. Particles at the end of the reactor ignite first, leading to a high temperature wave that moves upstream. This is visible from the slices in Figure 1 and the corresponding concentration slices in Figure 2. In this case, multiple particles have self-ignited, meaning they are in the higher steady-state. This leads to significant enhancement for the overall reactor with a total effectiveness factor of 14.13.

Conclusion and outlook
In this work, a transient numerical model that accounts for the mass and heat coupling between the solid and fluid phase in packed beds is presented. The model is able to capture the moving reaction zone inherently avoiding uncertainties introduced in the determination of empirical coefficients used in classical 1D phenomenological models, which require an axial heat dispersion coefficient.

Next steps that will be discussed in the talk are to use more advanced diffusion models, i.e. Maxwell-Stefan and Dusty Gas Model, which take into account the interaction between species and possible pressure gradients introduced in the pellets due to the reaction. These more advanced diffusion models introduce a new challenge in coupling the inside and outside transport as the mass flux then depends on all species.

Table 1. Dimensionless numbers used for the packed bed simulations.

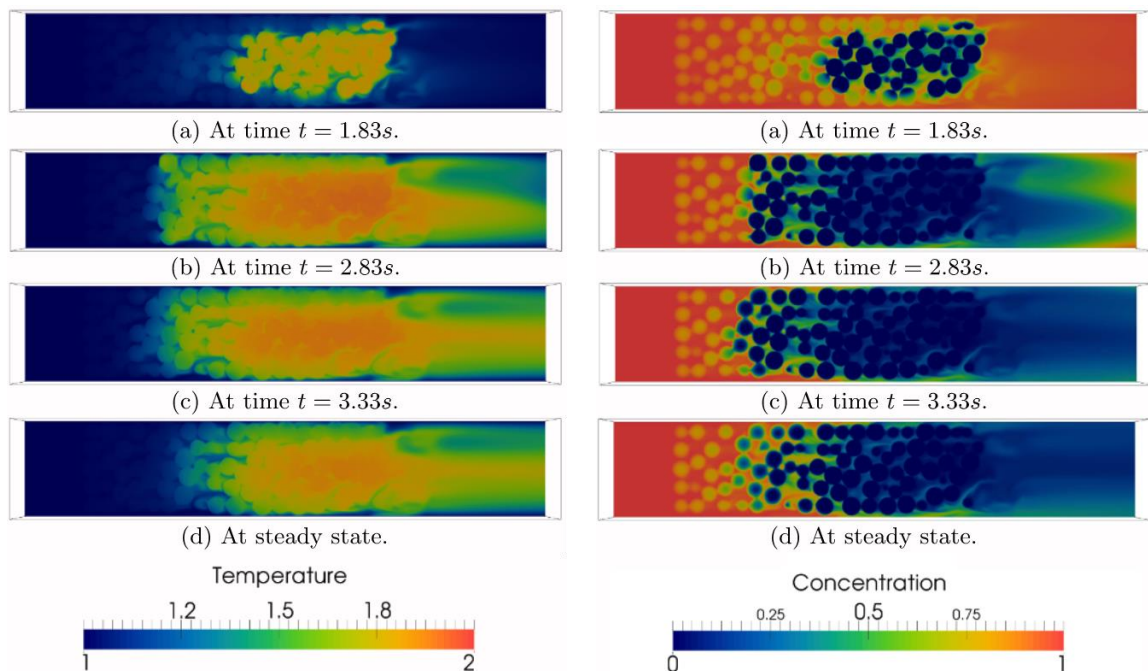| Dimensionless number	Definition	Value	Dimensionless number	Definition	Value
Reynolds number	$\frac{|\langle v \rangle| D \rho_f}{\mu_f}$	100	Fluid-to-solid molecular diffusivity	$\frac{D_f}{D_s}$	5
Prandtl number	$\frac{C_{pf}\mu_f}{k_f}$	1	Wall-to-inlet temperature ratio	$\frac{T_w}{T_0}$	1
Schmidt number	$\frac{\mu_f}{\rho_f D_f}$	1	Arrhenius number	$\frac{E}{RT_0}$	20
Fluid-to-solid thermal conductivity	$\frac{k_f}{k_s}$	0.1	Prater number	$\frac{c_0(-\Delta H_r)D_s}{k_s T_0}$	0.02

(a) At time $t = 1.83s$.

(b) At time $t = 2.83s$.

(c) At time $t = 3.33s$.

(d) At steady state.

Temperature

Figure 1. Temporal evolution of the dimensionless temperature profile for $\varphi = 1$

(a) At time $t = 1.83s$.

(b) At time $t = 2.83s$.

(c) At time $t = 3.33s$.

(d) At steady state.

Concentration

Figure 2. Temporal evolution of the dimensionless concentration profile for $\varphi = 1$

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