

MODELLING NON-ISOTHERMAL NON-ADIABATIC PACKED BED REACTORS

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Tubular packed bed reactors have been widely used in the chemical process industry for decades in various chemical conversion processes such as the selective oxidation of ethylene, oxidative coupling of methane and the synthesis of phthalic anhydride. Multi-tubular packed-bed reactors are generally preferred for exothermic reactions where a cooling jacket enclosing the tubular wall is used for controlling the reactor temperature. Highly exothermic systems employ a multi-tubular design where each individual tube has a low tube-to-particle diameter ratio to ensure sufficient cooling across the entire length of the reactor. The dynamics of the non-isothermal reactor are described by non-linear partial differential equations having an Arrhenius type dependency for the rate of reaction and the heat production terms. The intrinsic two-way coupling between temperature and concentration along with the self-dependency of temperature on the heat produced during exothermic reactions causes the system to exhibit many unique features, such as a small change in the inlet feed concentration or temperature that can cause a dramatic change in the reactor effluent conditions.

Here momentum transfer is coupled with heat and mass transport and the numerical method used is Direct Numerical Simulation. The Navier-Stokes equations are solved for the fluid phase transport and the temperature and species balance equations are solved for both the fluid and solid phase transport. The no-slip boundary condition is imposed along the surface of the particles and the tubular wall to solve for the velocity field. The continuity of fluxes boundary condition is imposed along the fluid-solid interface of the particles. For temperature, a constant value is fixed along the wall of the tubular column to ensure cooling across the bed. For concentration, the wall is assumed to be inert with the zero-flux condition. The reactor is mapped on to a 3-dimensional Cartesian grid where the fluid phase events and the solid phase events are solved simultaneously evaluated using their respective properties. The fluid phase and solid phase solutions are then coupled by applying the required boundary conditions using the Immersed Boundary Method [1]. This technique employs a quadratic interpolation where the values at the grid nodes close to the interface are modified to account for the boundary condition effects. This is done at the level of the discretized set of equations governing the transport in the two phases.

Here, the work of Das et al. [2] is extended where flow and heat transfer was studied in a cylindrical column packed with spherical particles using the Discrete Element Method (DEM). The artificially generated fixed bed consists of 340 spheres randomly packed in a slender tubular column using the DEM approach with the tube-to-particle diameter ratio being 5. Intra-particle diffusion of heat and mass along with a simple first-order irreversible reaction governed by the Arrhenius equation is considered [3] in the catalyst phase.

The velocity field of the fluid across the mid-plane of the packed bed reactor is presented in Figure 1. The dimensionless axial superficial velocity distribution across the cross-sectional radius of the reactor is presented in Figure 2 whose value is averaged across the length of the reactor. The velocity distribution yields a unique phenomenon referred to as 'channeling', where a

large hump is observed close to the wall of the reactor. This arises from a combination of the packing of the spheres (where porosity reaches zero close to the wall) and the shear applied on to the fluid from the wall. The effect of flow-maldistribution is clearly visible in the figures presented below.

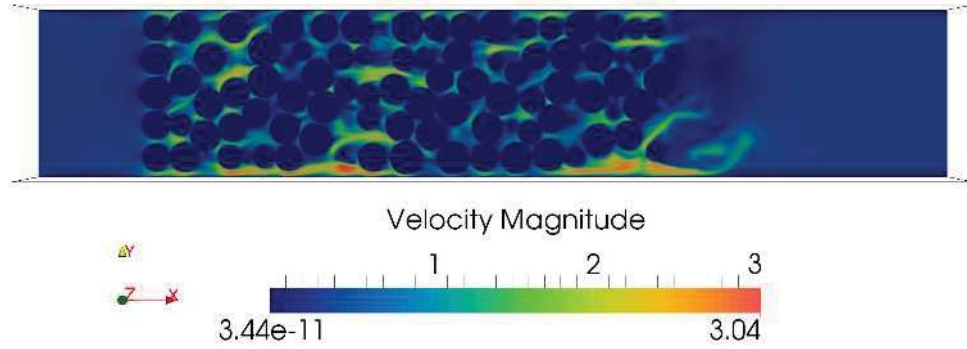


Figure 1

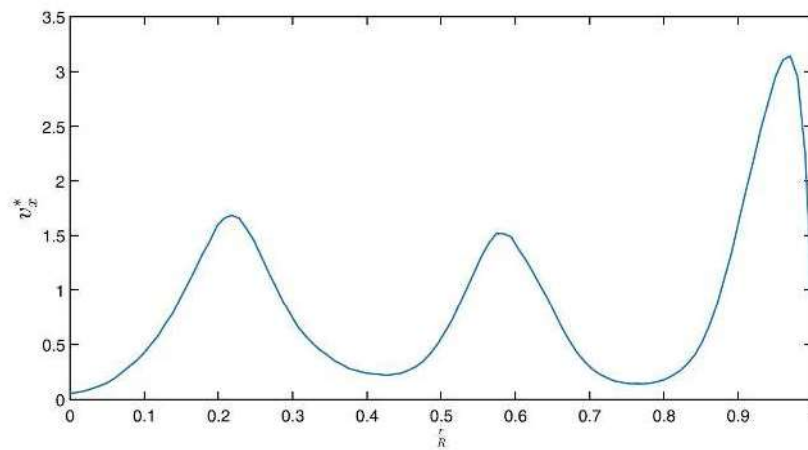


Figure 2

In Figure 3 and Figure 4, the concentration and temperature contour plots across the mid-plane of the reactor are presented where the rate of reaction is fixed such that the Thiele Modulus $\phi = R\sqrt{(k/D_s)} = 3$. Here the rate of reaction is three times the rate of diffusion within the catalyst particle. Due to the fast reaction, a large amount of heat is generated and this in return increases the temperature within the catalyst thereby enhancing the rate of reaction in return. Thus the system attains an auto-thermal operation condition wherein a feedback loop exists with the rate of reaction producing heat, in return enhances the rate.

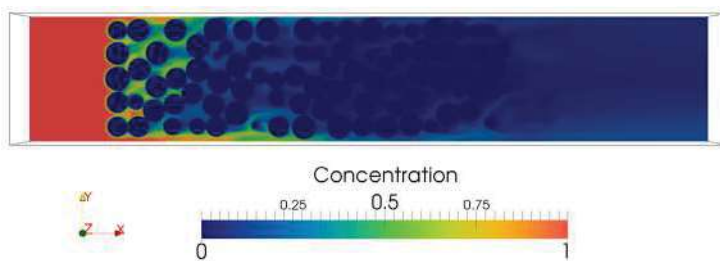


Figure 3

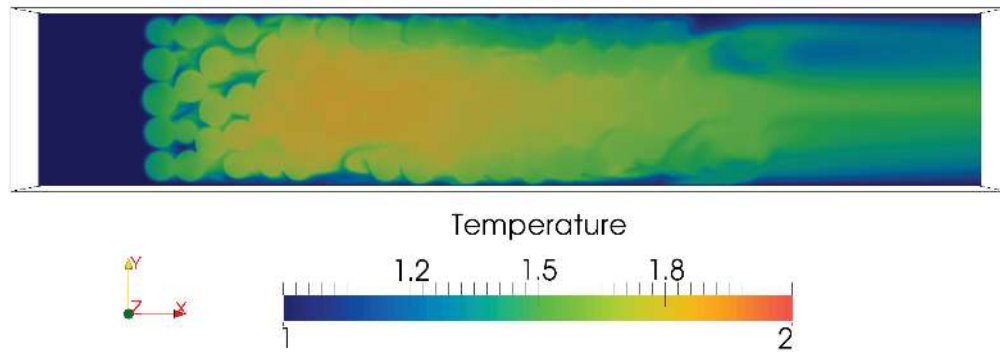


Figure 4

From the contour plots it can be seen that most of the reactant is converted away with the dimensionless concentration almost reaching zero at the outlet of the reactor. The dimensionless temperature yields that the reactor bed attains a value greater than the inlet feed temperature. It is assumed that the wall temperature equals the inlet temperature. Thus, the wall provides a cooling effect where the particles at the reactor exit are at a lower temperature than the particles in the middle of the bed. Plotting the temperature value within the fluid phase averaged using the velocity distribution, it can be seen that the temperature value reaches a maxima along the axial direction referred to as hot-spot. This hot-spot magnitude is dependent on the cooling effect provided by the wall. It is also influenced by the hydrodynamics where the flow-bypass provides a supplementary resistance to the heat removal mechanism across the radius of the bed. Figures 5 and 6 portray the temporal evolution of the bulk concentration and temperature values across the axial direction of the reactor bed, respectively.

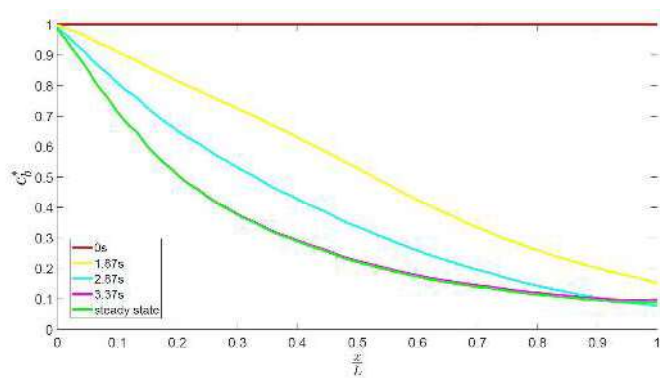
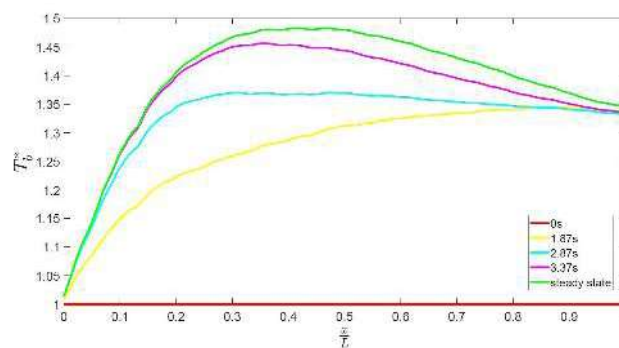


Figure 5

Figure 6



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