

Direct Numerical Simulation of Coupled Heat and Mass Transfer in Dense Gas-Solid Flows with Surface Reactions

Summary

Fluid-solid flows are frequently encountered in a wide range of processes. A typical example is fixed bed reactors, which are commonly used in chemical industry for catalytic reactions. In such processes, reactions take place at active sites deep inside porous catalyst particles. Therefore, it is important to know how reactants are transported to these particles and how products move out with the fluid flow. The performance of a catalytic process is determined not only by the efficiency of the catalytic reaction, but also by the efficiency of mass transport. In such highly heterogeneous systems, there are two important factors, specifically variable reaction rates and significant heat effects, which introduce additional complexities to the equipment design as well as the process performance.

In chemical engineering, most researchers focus on either reaction kinetics or transport processes, but in this thesis the flow of reactants and products around catalytic particles is modelled as well as the reactions that occur there. Using detailed models the performance of a catalytic process can be predicted by computer simulations. The simulations give insights in the interplay among mass transfer, chemical transformations and heat effects. Using the detailed information the models provide, real chemical processes can be improved and in that way technological development is accelerated.

In this thesis, an efficient ghost-cell based immersed boundary method (IBM) is developed to perform direct numerical simulation (DNS) of (coupled) mass and heat transfer problems in fluid-particle systems. The fluid-solid coupling is achieved by implicit incorporation of the boundary conditions into the discretized momentum, species and thermal energy conservation equations of the fluid phase. Taking advantage of a second order quadratic interpolation

scheme in the reconstruction procedures, the DNS model is capable to handle the Robin boundary condition at the exact position of the particle surface. This is the unique feature of this DNS model. A fixed Eulerian grid is used to solve the conservation equations for the entire computational domain. Several fluid-particle systems, with increasing complexity, are studied in the subsequent chapters with the aim to push forward the frontier of the application of DNS towards more realistic chemical reaction processes.

In Chapter 2, the governing equations, the numerical solution method and the IBM are introduced in detail. The newly developed quadratic interpolation scheme is verified by first comparing the numerical results for unsteady mass diffusion around a single sphere with the analytical solutions and, second, by comparing computed Sherwood numbers for convective mass transfer with the empirical values. In both cases, an infinitely fast surface reaction is assumed and excellent agreement is achieved.

In Chapter 3, a catalytic surface reaction is introduced into the DNS model. The Damköhler number is used to describe the interplay between chemical reaction and mass transfer processes in fluid-particle systems. First, a detailed verification study of unsteady mass diffusion is performed, where the concentration profile and the instantaneous Sherwood number are compared with the analytical solutions. Next, both single particle and multi-particle dense systems with different reaction rates are considered. It is noted that, the mass transfer performance of the dense particle array is improved to some extent with higher reaction rates.

In Chapter 4, mass transfer problems in particle clusters, specifically a nine-sphere cuboid cluster and a randomly-generated spherical cluster consisting of hundred spheres, are studied. The clusters are composed of active catalysts and inert particles, which are realized by the Dirichlet and Neumann boundary condition respectively. Mixed boundary conditions of individual spheres are handled consistently in the DNS model. In case of a cluster, the swarm effect decreases the mass transfer performance dramatically, however, dilution with inert particles can help to counteract this decrease quite a lot.

In Chapter 5, the heat transfer problems in fluid flowing through tubular fixed beds are studied. For the classical Graetz-Nusselt problem with both isothermal and isoflux boundary conditions, the DNS results agree well with the analytical results in a wide range of the Péclet

number. An advanced Graetz-Nusselt problem with hundreds particles randomly positioned inside a tube with adiabatic wall is then studied. The channeling effect at the wall region is noticed to significantly reduce the performance of the system, nevertheless large and passive particles will increase the heat transfer coefficient. The simulation results are compared with the results obtained by a team from the University of Twente using a different class of IBM, revealing good agreement.

In Chapter 6, independently coupled heat and mass transfer is studied in fluid-particle systems. The reaction rate is specified to be first order with a rate coefficient which is independent of temperature. The coupling of heat and mass transfer arises as a consequence of an exothermic reaction where the liberated heat heats up the particle that, subsequently, transfers the thermal energy to the fluid phase. For the cases of unsteady mass and heat diffusion around a single sphere, forced convection to a single sphere and the three-bead reactor, the temperatures obtained from DNS show excellent agreement with the well-established results. For the dense particle array, qualitative agreement is reached between DNS and a 1D model. However the quantitative discrepancy, which is further amplified at higher Reynolds numbers, is thought to originate from the heterogeneity inside the array.

In Chapter 7, building on the work of Chapter 6, the coupling of heat and mass transfer moves one step further to reality. The heat and mass transport processes are fully coupled, as the reaction rate is now influenced by the temperature. At the end of every time step, the temperature of individual particles is computed, and subsequently the reaction rate coefficient is determined by the Arrhenius equation. Realistic parameters from the process of partial oxidation of methane are used. Three fluid-particle systems with increasing complexity, specifically a single sphere, three spheres and hundreds spheres, are modelled. It is noticed that, the solid temperature obtained from DNS is extremely sensitive to the computed mass and heat transfer processes. Although the overall behavior correlates well with the average Sherwood (Nusselt) number and the superficial fluid velocity, no local correlation with simple isotropic quantities could be obtained.