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3D CFD simulations of steam reforming with resolved intraparticle reaction and gradients

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Abstract

Computational fluid dynamics (CFD) simulations are reported for flow, diffusion, reaction and heat transfer in a 120° segment of an $N = 4$ packed tube for the endothermic methane steam reforming reaction. The present work improves on previous approaches by explicit inclusion of intraparticle effects (conduction, species diffusion and reaction) coupled to realistic 3D external flow and temperature fields. It is shown that the usual assumption of symmetric species and temperature fields inside spherical catalyst particles holds for particles away from the tube wall, but particles placed in the strong temperature gradient near the tube wall show significant deviations.

Keywords: Computational fluid dynamics; Catalyst design; Heat transfer; Packed bed; Chemical reactors; Reaction engineering

1. Introduction

Catalyst performance in fixed bed reactors is generally modeled under the assumption that the intraparticle temperature field is symmetric, if not uniform. This approach is valid only if the reactor has a high tube-to-particle diameter ratio (N), with little temperature variation over the particle dimension. For methane steam reforming, the gas flow rate is very high, leading to the use of large catalyst particles to reduce pressure drop, and the reactions are highly endothermic, leading to the use of slim tubes so that heat may be supplied efficiently through the tube wall. Consequently, the fixed bed reactor tubes have low tube-to-particle diameter ratio (N), often in the range 4–8. As a result of the low N values employed, a large fraction of the catalyst pellets are placed in large temperature gradients, next to the tube wall.

The application of computational fluid dynamics (CFD) to packed bed reactor modeling, taking into account the actual packing structure, is on the increase and has recently been reviewed (Dixon et al., 2006). Studies have been made on fluid flow, heat transfer and dispersion in either low-N packed tubes or small periodic groups of spherical particles, without reaction. The inclusion of chemical reactions has so far been limited to lattice Boltzmann simulations of isothermal flows with surface reaction, and showed good agreement between the computations and the experimental results (Zeiser et al., 2001; Freund et al., 2003; Yuen et al., 2003). To date, no work from other groups has appeared in which detailed flow fields and heat transfer were computed by CFD for packed tubes with heterogeneous reaction taking place within the catalyst particles.

In our previous work, CFD simulations were reported for flow and heat transfer in a 120° segment of an $N = 4$ tube where temperature-dependent heat sinks inside spherical particles were used to mimic the thermal effects of the endothermic methane steam reforming reaction (Dixon et al., 2003). The analysis was later extended to cylindrical pellets (Nijemeisland et al., 2004). The objective of the present work is to improve on this approach by explicit inclusion of intraparticle effects (conduction, species diffusion and reaction) coupled to realistic 3D external flow and temperature fields in steam reformer performance simulation.

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2. Simulation model development

Our use of CFD codes to numerically solve flow, species and energy balances in complicated flow geometries found in packed tubes has been reviewed in detail (Dixon et al., 2006). The CFD approach for packed bed heat transfer has been validated by comparison to experiments (Nijemeisland and Dixon, 2001). For the present work, Fluent® version 6.2 was used, with the k–ε Renormalization Group (RNG) model for turbulence, and non-equilibrium wall functions. An unstructured tetrahedral mesh was implemented with finer mesh density near wall–particle and particle–particle contact points. To avoid contact points, the sphere diameters were actually reduced to 99.5% of their nominal value. In our previous work, we showed that a 120° wall segment (WS) model with two axial layers of particles and translational periodic boundaries at the inlet and outlet and symmetry side conditions gave good agreement with full bed simulations for $N = 4$. The segment structure is shown in Fig. 1(a).

The particles were represented in the present CFD model as porous regions, with species and energy reaction sinks/sources given by user-defined code. Species transport in the porous spheres was modeled by effective binary diffusivities calculated from straight-pore Knudsen and molecular diffusion coefficients, and corrected using pellet voidage and tortuosity. For pellet conduction an effective thermal conductivity was calculated from gas and alumina thermal conductivities using the pellet void fraction.

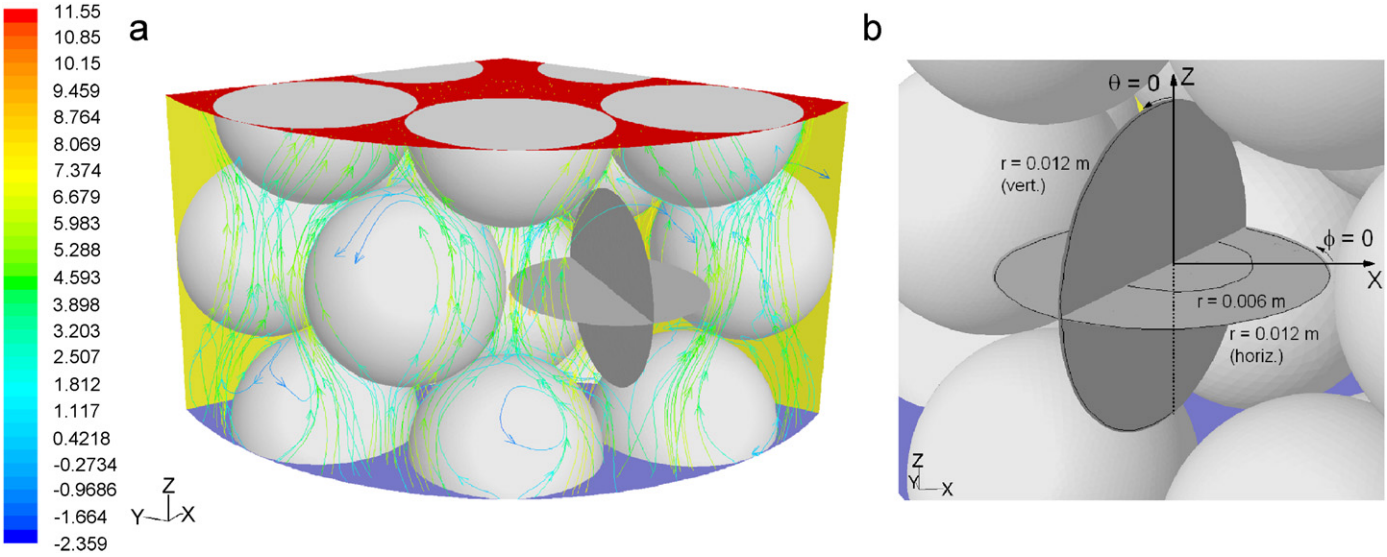

Fig. 1. Wall segment geometry: (a) three-dimensional view with flow pathlines scaled by z-velocity with particle 2–6 replaced by horizontal and vertical slices and (b) particle 2–6 with angular coordinates.

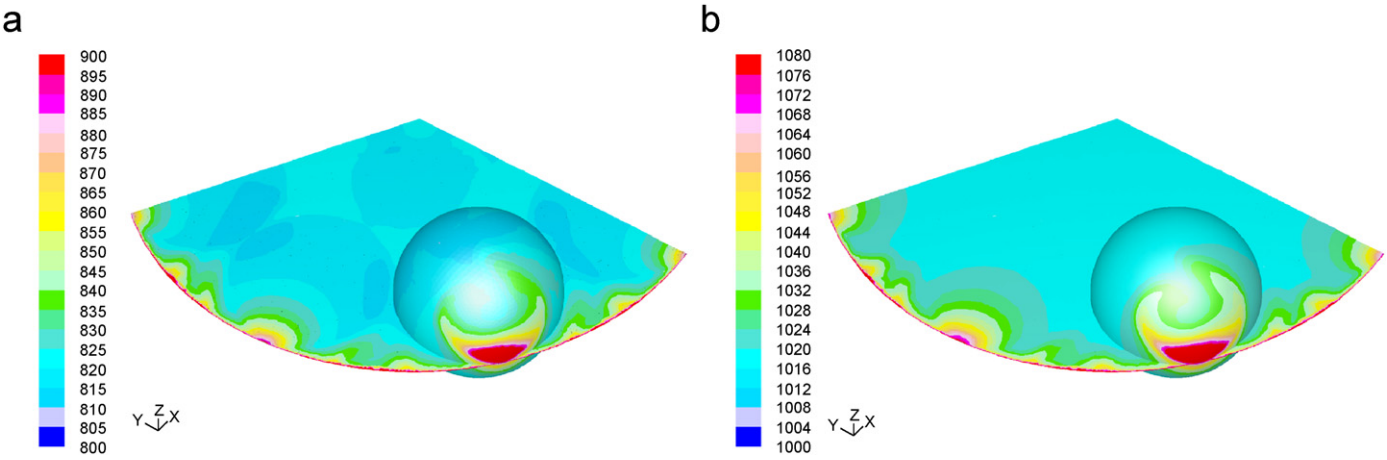

Fig. 2. Temperature contours on horizontal plane through middle of wall segment, and on surface of particle 2–6, for (a) inlet and (b) mid-tube conditions.