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Understanding Local Dynamic Responses of Fixed Bed Reactors by CFD

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Coupling microkinetics with unsteady-state CFD has been studied to develop a method for tracking the dynamic behavior of temperature, species and surface sites in a fixed bed reactor. Transient simulations have been made for ethylene epoxidation in a small bed with inlet temperature reduction, and these show the start of thermal wave progression down the tube, and a maximum temperature above that given by steady-state models. Transient simulations have also been carried out for oxygen shut-down in the inlet feed, which illustrate the persistence of oxygen and reaction in catalyst pellets for a considerable time after the shut-down. Finally simulations of start-up after the initiation of oxygen flow have shown the evolution of oxygen mass fraction and temperature profiles in the pellets and their interactions.

Dynamic behavior of fixed bed reactors under transient conditions could lead to phenomena such as wrong way behavior and catalyst deactivation. These are highly unfavorable, that yet need to be addressed in the field. In order to study these phenomena a CFD integrated multiscale model has been developed to study fixed-bed reactors under dynamic reaction conditions. A detailed microkinetic reaction mechanism was coupled into the transport and flow in both the solid and fluid phases. The kinetics model was solved based on our prior ACS-PRF funded work using multivariate spline mapping with construction of generic computational libraries imported into the CFD code to evaluate the reaction rates and surface species coverages. The surface species reach steady state much faster than temperature and concentration in the reactor so they were assumed to be at steady state even for transient simulations.

Ethylene oxidation over silver catalyst was studied using the developed model. The computational domain comprised 400 spherical particles of size 6 mm in a tube of diameter 28.2 mm. Laminar flow was simulated, with inlet temperature and wall temperature of 503 K. The reactor tube was assumed to be adiabatic, in order to have a developed temperature profile in the relatively short tube length that could be considered in this study. Three different phenomena were studied: 1- wrong-way behavior, 2- reactor shut-off, and 3- reactor start-up. For wrong-way behavior and reactor shut-off, first steady state simulations were conducted then the simulations were switched to the transient conditions and dynamic behavior was studied.

Fig. 1 shows the temperature profile in the bed 10 seconds after the feed temperature was dropped by 70 K. The maximum temperature in the bed was raised about 6 K. The hot temperature profile also extended upstream in the bed. It was observed that the mid tube had a higher temperature than at the previous steady-state (about 25 K), while the inlet area was much colder (about 50 – 70 K). The dynamic conditions in the bed caused a significant change in the temperature profile and resulted in sharp gradients.

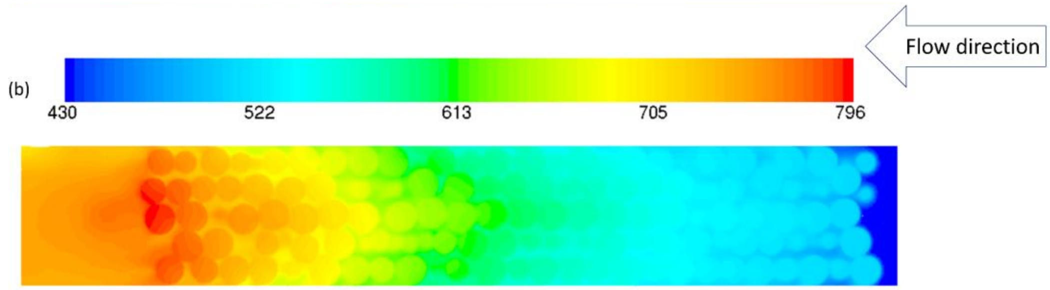

Figure 1. Contours of temperature (K) profile evolution along the tube. Inlet temperature has been decreased but temperatures in the center of the bed have increased from the steady-state values, demonstrating “wrong-way” behavior with variations at the local scale.

For start-up simulations, the velocity field was established with ethylene and methane at 503.15 K. Then at *t=0*, the inlet oxygen mass fraction was set to 0.08. Fig. 2 shows the oxygen profile time evolution along the tube. Oxygen moves fast in the void area of the reactor inlet in a uniform pattern. Once it reaches the packing it moves down the bed at a much slower rate. On the particle surfaces it starts to react with the ethylene in the bed and the concentration drops. After 20 seconds the fluid phase is mainly filled with oxygen. However, the particles are still mostly empty. After 90 seconds oxygen has almost completely diffused into the first row of particles. Notice that the oxygen by-passes along the tube wall due to the higher flow rate there caused by higher void fraction near the wall, creating a transient radial concentration profile in the fluid.

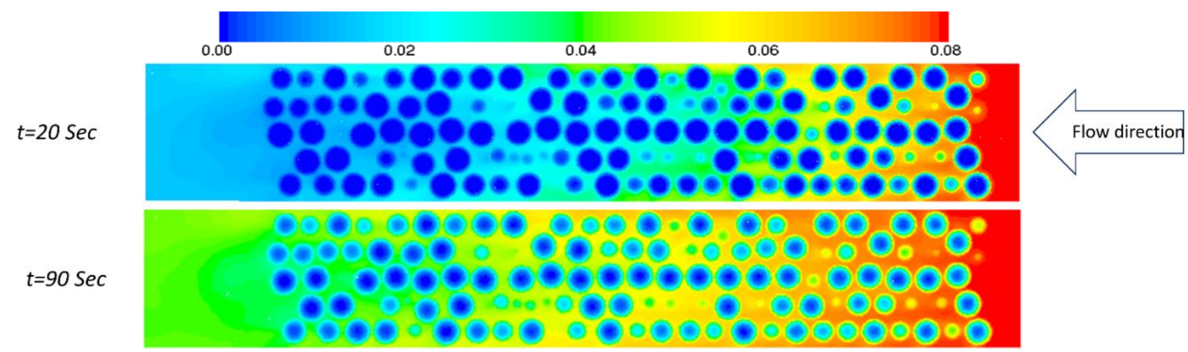

Figure 2. O_2 mass fraction profile along the bed on a mid-tube plane, for reactor start-up, at times 20 and 90 s after the start of oxygen flow.

The results show the importance of the temperature and oxygen profile evolution under dynamic reaction conditions and can show how they affect the reaction rate locally in the reactor.

The research has allowed the PI to broaden his CFD capabilities into the study of dynamic reactor phenomena, thus enabling study of a broader range of reactor performance and possible safety issues (e.g. moving hot-spots) than previously. The PhD student Behnam Partopour has been able to focus on research for the past two years of his graduate study, thanks to support from this grant. He graduated with Ph. D. in May, 2018 and is currently employed at Amgen in Cambridge, MA, while also teaching an evening graduate class at WPI. We continue to collaborate.