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Spatially Resolved Reactor Profiles: Understanding Catalytic Reactions at Industrial Conditions.

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Highlights

- Pilot scale fixed bed reactor for species and temperature profile measurements.
- Reactor profiles for n-butane oxidation to maleic anhydride on vanadyl pyrophosphate.
- Particle resolved CFD simulation of the reactor.
- Knowledge based optimization of temperature profile and maleic anhydride yield.

1. Introduction

Industrial catalytic fixed bed reactors are basically black boxes. It is known what flows in the reactor and what comes out of it. What happens inside, viz. how reactants are transformed into products, which reaction intermediates occur and how the temperature profile looks like, remains unknown. To get insight into what happens in industrial catalytic fixed bed reactors we developed a pilot scale reactor for spatially resolved measurements of species and temperature profiles at industrially relevant conditions and dimensions. The advantage of using industrial-like reactor tube diameters and catalyst shapes is, that heat- and mass transport phenomena are more realistic than in small-scale laboratory reactors. The selective oxidation of n-butane to maleic anhydride (MAN) was chosen as test reaction. Side products are carbon monoxide, carbon dioxide, acetic acid and acrylic acid.

$$C_4H_{10} + 3.5\ O_2 \rightarrow C_4H_2O_3 + 4\ H_2O \quad (1)$$

$$C_4H_{10} + x\ O_2 \rightarrow CO_2 + CO + \text{Acrylic Acid} + \text{Acetic Acid} \quad (2)$$

For knowledge based reactor optimization, the reactor was modeled using particle resolved CFD simulations. After fitting of key model parameters to a set of reactor profiles for training, the model was used to predict reactor performance for other operation conditions. Goal was to optimize the maleic anhydride yield and to eliminate hot spot formation being detrimental for catalyst lifetime.

2. Methods

The pilot scale profile reactor is based on the sliding capillary measurement principle developed by Horn et al. and described in detail in earlier publications, e.g. [1]. The reactor can accommodate a catalyst bed up to 500mm in length and 21mm diameter. Five independent heating zones control the reactor wall temperature profile. Reactor pressures up to 5bar are possible. The reactor is fully automatized and allows unattended measurement campaigns for extended periods of time. This enables studying the catalyst formation phase and the behavior of the catalyst under industrially relevant operation conditions, as well as its long-term stability. Three-dimensional computational fluid dynamics (CFD) modeling [2] with computer-generated packings by the discrete element method (DEM) was conducted to model transport processes inside the reactor. This combined DEM-CFD approach has been experimentally validated for the packing geometry (porosity profiles) and radial heat transfer [3]. In the present contribution the CFD model is extended by including reaction kinetics, i.e. a network for n-butane oxidation [4].

Fig. 1: Pilot scale fixed bed reactor for spatially resolved measurements.

3. Results and discussion

Figure 2 shows representative reactor profiles for n-butane oxidation with vanadyl pyrophosphate as catalyst [5]. In this experiment all five heating zones were set to 370°C.

The profiles reveal a nonlinear development of n-butane conversion. The temperature profile shows a 55 K

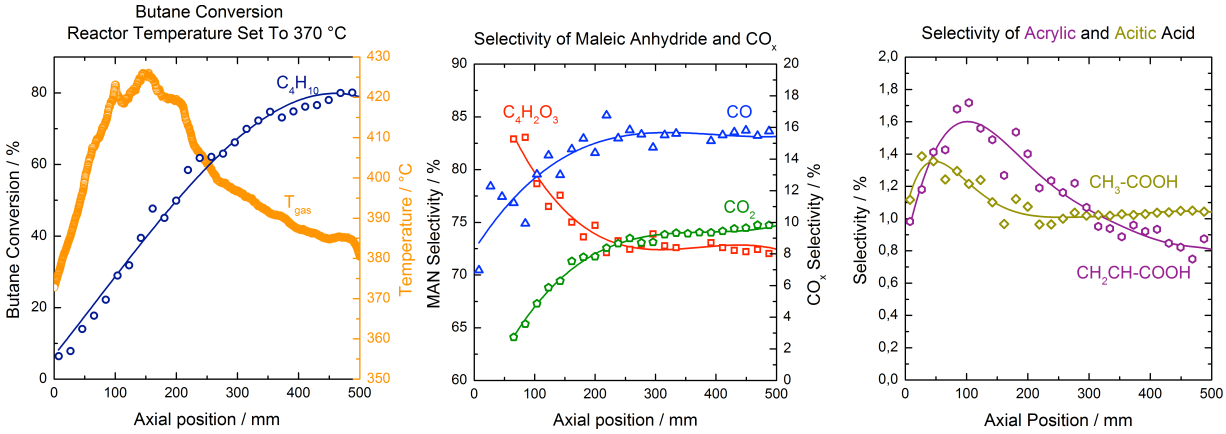

Figure 2. Reactor profiles for butane conversion, temperature and product selectivity at 370°C reactor wall temperature (lines to guide the eye).

hotspot, which correlates with the formation of acrylic acid as intermediate product. Acetic acid is another intermediate but peaks slightly earlier. The initial selectivity towards CO is larger than zero, indicating a second undesired butane oxidation pathway. Figure 3 shows a representative cut through the catalyst packing in the region of the hot-spot illustrating temperature inhomogeneity in radial direction caused by the finite rate of heat transport. To eliminate the hotspot a temperature profile was imprinted on the reactor wall using the five independent heating zones (360°C, 365°C, 370°C, 375°C, 380°C). By lowering temperature in the first half of the reactor where the n-butane concentration is highest and raising the temperature in the second half of the bed where the n-butane concentration is lowest a more uniform reaction rate could be realized throughout the reactor, the hotspot could be eliminated and the maleic anhydride yield could be improved.

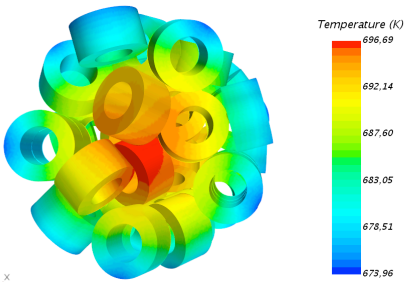

Figure 3. Inhomogeneous catalyst temperature in the hot spot of the reactor.

4. Conclusions

The profile reactor in combination with particle resolved reactor simulations is a powerful tool to prove or disprove kinetic models and understand the influence of local transport processes on the integral reactor performance.

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Keywords
profile reactor, butane oxidation, particle resolved CFD, knowledge based optimization