Effect of Particle Shape on Catalyst Deactivation Using Particle Resolved CFD Simulations

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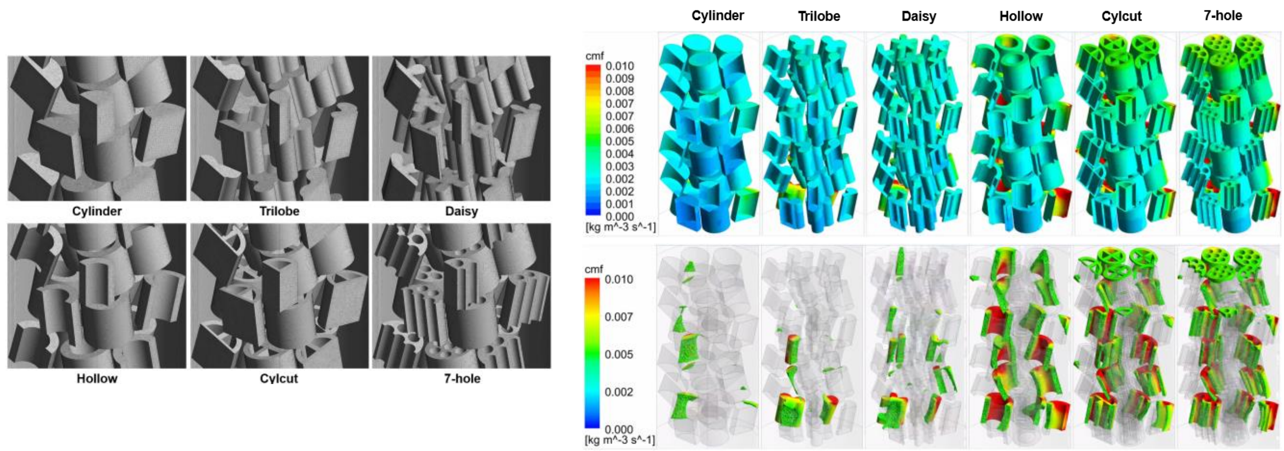
Highlights

- Development of methodology to model the catalyst deactivation inside the particle using propane dehydrogenation reactions.
- Particle resolved CFD simulations with six different particle shapes (cylinder, trilobes, daisy, hollow cylinder, cylcut, 7-hole cylinder).
- Analyze the impact of different particle shapes on the catalyst deactivation rate.

1. Introduction

Catalyst deactivation due to carbon formation is a major issue in many industrial catalytic reactions such as methane steam reforming (MSR) and propane dehydrogenation (PDH), as it decreases the reactor performance over a period of time. The particle shape has a major impact on the catalyst deactivation rate as it influences the local transport phenomena. Complex particle shapes (e.g. 7-hole cylinder for MSR) are being used for various industrial catalytic reactions due to their higher surface area to volume ratio. However, the impact of particle shape on the catalyst deactivation rate has not been explored.

Lower order models with over-simplified assumptions such as uniform heat and mass distribution on the particle surface are being used to predict the catalyst deactivation rate and its impact on the reactor performance. Unfortunately, these models fail to account for the effect of particle shape which influences the local fluid flow and hence the temperature and species distribution on the particle surface and within the particle. Particle resolved CFD simulations, mostly spheres or cylinders, are being used for accurate prediction of flow, temperature and species distribution around the particles and therefore reactor performance. These simulations can provide particle scale information and its impact on reactor performance which is otherwise difficult to obtain from lower order models and experiments. Therefore, the main objective of this work is to develop a methodology to model catalyst deactivation inside the particle and to perform particle resolved CFD simulations for analyzing the effect of particle shapes on catalyst deactivation rate for PDH reactions.



(a) *(b)*

Figure 1: *(a) CFD model and mesh used for different particle shapes and (b) distribution of carbon formation rate on particle surfaces (top) and iso-volume inside particles (bottom) for PDH reactions at Re=5000.*

2. Methods

A cylindrical packed bed (d_t=100 [mm], h_t=150 [mm]), containing 2 representative blocks of 15 particles each arranged in random manner, was used as the CFD model. Apart from cylinder (d_p=10 [mm], h_p/d_p=1), five different particle shapes namely trilobes, daisy, hollow cylinder, cylcut and 7-hole cylinder were considered (see Figure 1(a)). The particle volume (0.6*cylinder volume), outer diameter and height were kept constant, however the particle surface area varies owing to their shaping. Tetrahedral mesh was generated for all particle shapes leading to 14.3-30.6 million elements depending on the particle shape (Figure 1(a)). A user-defined scalar defined in particle and fluid domains with coupling at particle-fluid interface was used to model the reaction and diffusion in particle interior [1]. Propane dehydrogenation (PDH) with carbon formation reactions were considered for this work. The reaction kinetics, process conditions with particle and fluid properties were taken from the literature. The boundary conditions include mass flow inlet, pressure outlet, wall heat flux and reaction sources inside particles. The conservation equations were solved using ANSYS 18.0 with SST k-ω turbulence model for closure using 128 CPU-cores. Further details will be provided in the conference.

3. Results and discussion

Preliminary investigations were performed with cylindrical particle shape with different number of representative blocks and computational grid to optimize the model, thereby reducing the computational cost. Presently, we modelled only the initial carbon formation rate for fresh catalyst however the model will be extended to include the carbon accumulation and its impact on reaction rate, species diffusion and particle properties. A comparison of the carbon formation rate on particle surfaces and iso-surface inside particle is shown in Figure 1(b). For all the particle shapes, strong gradients exist on the particle surfaces particularly in the near-wall particles due to endothermic behavior with wall heat flux. However, it is relatively more for hollow, cylcut and 7-hole particles shapes due to better reactant access to the particle interior. This can also be seen from the iso-volume distribution. The non-uniform temperature and species distribution on the particle surface clearly points out the limitations of the lower-order models. For more quantitative comparison, propane conversion (see Figure 2(a)), volume-averaged <carbon formation rate> (see Figure 2(b)) and propane conversion-to-<carbon formation rate> (see Figure 2(c)) were analyzed as a function of particle surface area. The propane conversion was observed to increase with particle surface area due to reduction in diffusion limitations. Similarly, the <carbon formation rate> increases with particle surface area due to higher effectiveness factors of the catalyst. In summary, 7-hole offered the highest propane conversion however trilobe gave the highest propane conversion to <carbon formation rate>.

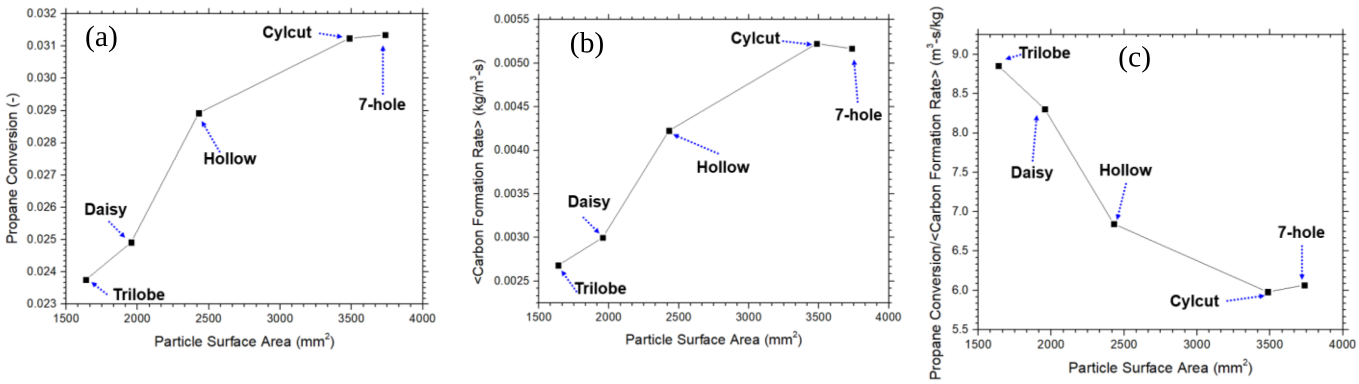


Figure 2: *Effect of particle surface area on (2) propane conversion, (b) <carbon formation rate> and (c) propane conversion-to-<carbon formation rate> for PDH reactions at Re 5000.*

4. Conclusions

Preliminary investigations showed that the <carbon formation rate> increased with particle surface area showing the importance of particle shape on the catalyst deactivation rate and thereby on reactor performance. The 7-hole offered the highest propane conversion, however, trilobe gave the highest propane conversion-to-<carbon formation rate>. The work will be extended to include the carbon accumulation and its impact on reaction rate, species diffusion and particle properties. The present work is important in understanding the effect of particle shape on catalyst deactivation rate for industrial catalytic reactions leading to development of optimal catalyst shape to maximize the reactant conversion and minimize the catalyst deactivation.

5. References

[1] Dixon AG, Taskin ME, Nijemeisland M, Stitt EH. Ind. Eng. Chem. Res. 2010; 49(19):9012-25.

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