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Three-dimensional modeling and simulation of a micrometer-sized particle hierarchical structure with macro- and meso-pores

Alessio Caravella [a,*], Shigeki Hara [a], Nobuo Hara [a], Akira Obuchi [b], Junko Uchisawa [b]

[a] National Institute of Advanced Industrial Science and Technology (AIST), Research Institute for Innovation in Sustainable Chemistry, Central 5, Higashi 1-1-1, Tsukuba, Ibaraki 305-8565, Japan

[b] National Institute of Advanced Industrial Science and Technology (AIST), Research Center for New Fuels and Vehicle Technology, Onogawa 16-1, Tsukuba, Ibaraki 305-8569, Japan

HIGHLIGHTS

- Decane oxidation is simulated in a hierarchical structure of macro- and meso-pores.
- The overall effective diffusivity includes porosity and tortuosity of ensemble.
- The isothermal effectiveness factor can be also used in non-isothermal cases.

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ABSTRACT

This study is focused on modeling and simulation of hierarchically structured catalytic media with macro- and meso-pores. The hierarchical structures are expected to enhance the contact between reactants and active catalytic sites. However, heterogeneous simulations of such structures are not computationally feasible for a practical scale, so that a multi-scale approach involving a pseudo-homogeneous approximation is required. This work aims at determining a suitable form for effectiveness factor in the reaction rate and effective diffusivity to be used in a pseudo-homogeneous approximation. To do that, computational fluid dynamics (CFD) simulations of hierarchical structure of particles are carried out in both heterogeneous and homogeneous mode within 240–300 °C and Thiele modulus around 2.4 or less. The analysis is applied to decane oxidation as representative of the oxidation of exhausted gas from Diesel engines. After providing a validation of the analysis on a single catalyst spherical particle, simulations in heterogeneous mode are carried out under isothermal conditions. The results suggest that the effective diffusivity for the pseudo-homogeneous approximation should include porosity and tortuosity of ensemble, which is evaluated considering the particles as dense. Additionally, detailed profile analysis suggests that the analytical expression of the effectiveness factor valid for spherical particles under isothermal conditions could be used in the pseudo-homogeneous approximation. Then, simulations in homogeneous mode are carried out under both isothermal and non-isothermal conditions to confirm the above-cited suggestions. The obtained results demonstrate the suitability of the chosen effectiveness factor and effective diffusivity.

1. Introduction

Monolith catalytic converters nowadays represent a standard technology for the treatment of exhausted gases from Diesel engines by oxidation process [1–3]. Traditionally, a uniform catalytic layer is deposited on the internal walls of the honeycomb channels. However, this solution can show performance problems in presence of mist, which can occlude the meso-pores (<50 nm in diameter) of the layer.

To avoid this type of problems and enhance the mass transfer toward the catalytic sites, hierarchical structures including meso- and macro-pores can be successfully used. Zeng et al. [4] prepared hierarchical macro–meso-porous structures of Pd/TiO_2 catalyst for styrene hydrogenation, showing that this dual-mode structure exhibits a higher catalytic activity than the meso-porous structure. Wang and Coppens [5] showed that the optimization of the macro-porosity and the macro-pore size in a Ni/Al_2O_3 catalyst hierarchical structure for the autothermal reforming of methane leads to a significant increase in conversion and selectivity to the main reaction product. Zhou et al. [6] prepared a Pd/Al_2O_3 hierarchical structure with macro- and meso-pores for the selective hydrogenation of the

* Corresponding author. Tel.: +81 29 861 4712.
E-mail address: a.caravella@aist.go.jp (A. Caravella).

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