Modeling of Porous Catalyst Pellets: Comparison of Diffusion Flux Models for Steam Methane Reforming

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Abstract

Introduction
Steam reforming of natural gas has been the most common method for producing synthesis gas (CO + H2) for the production of H2, MeOH and NH3 for over half a century [1]. Production of H2 in the USA using steam methane reforming (SMR) is ca. 9 MM tons/yr. Global production of NH3 using H2 derived from SMR was ca. 109 MM/yr in 2004. In the chemical industry, the SMR process is carried out in tubular furnaces where a Ni-supported catalyst is placed in tubes that are typically made from Ni-Cr alloys. The primary reactions that occur include methane reforming, water-gas-shift, and the methanation reactions.

$CH_4 + H_2O \rightleftharpoons CO + 3H_2$ $\Delta H_{298} = 206.2$ kJ/mol
$CO + H_2O \rightleftharpoons CO_2 + H_2$ $\Delta H_{298} = -41.5$ kJ/mol
$CH_4 + 2H_2O \rightleftharpoons CO_2 + 4H_2$ $\Delta H_{298} = 164.7$ kJ/mol

The catalyst pellets can vary in both size and geometry, and the compositions is usually Ni-on-Al_2O_3 [1, 4]. Some typical catalyst shapes are illustrated in Figure 1.
Comparison of diffusion flux models to describe species transport-kinetic interactions for SMR has been reported only for a spherical catalyst shape[2]. Dixon and coworkers [5] employed CFD to compare the effect of various catalyst geometries on heat transfer, pressure drop and methane conversion. However, a detailed performance comparison for various catalyst shapes using different flux models has not been examined.
The primary objective of this study is to describe a general modeling framework for various catalyst shapes using different diffusion flux models.
Use of Comsol Multiphysics
A 1D steady state model for a spherical catalyst shape is developed in COMSOL Multiphysics® software using Transport of Dilute Species and Heat conduction interfaces. The transport-kinetics interactions in the porous catalyst are analysed using various diffusion flux models such as Wilke, Wilke-Bosanquet, Stefan-Maxwell, and Dusty gas models. The kinetic equations from Xu and Froment are used to describe the reaction rates [3].
The steady state results using the various diffusion flux models are shown (Figure 2). The species mole fractions obtained using both the mass and mole formulations of the pellet model will be compared for each catalyst shape and for various diffusion flux models.

Reference

1. Kagyrmanova, A. P., Zolotarskii, I. A., Vernikovskaya, N. V., Smirnov, E. I., Kuz'min, V. A. and Chumakova, N. A., Modeling of Steam Reforming of Natural Gas Using Catalysts with Grains of Complex Shapes, Theo Found of ChE 40(2), 155 (2006).

2. Jannike, S., Stian, T., and Jakobsen. H. A., On the Consistent Modeling of Porous Catalyst Pellets: Mass and Molar Formulations, Ind. Eng. Chem. Res, 51, 8222 (2012).

3. Xu, J. and Froment, G.F., Methane Steam Reforming, Methanation and Water-Gas Shift: 1. Intrinsic Kinetics, AIChE J., 35(1), 88 (1989)

4. Bruno, S., Guillermo, F., and Gonzalez, M., Effect of the Geometric Characteristics of Commercial Catalysts for Steam Reforming, Chem. Eng. J., 39, 147 (1988)

5. Dixon, A. G., Boudreau, J., and Rocheleau, A., Comparison of Catalyst Geometries Using Computational Fluid Dynamics for Methane Steam Reforming, WPI B.Sc. Thesis (2010).

Figures used in the abstract

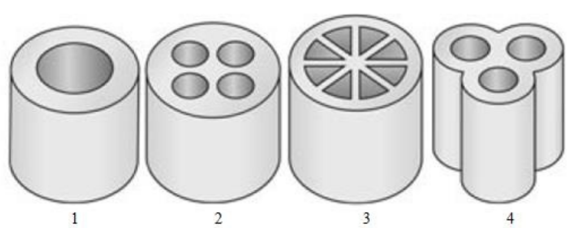

Figure 1: Catalyst shapes: (1) ring, (2) four-hole cylinder, (3) spoked wheel & (4) trilobe [1].

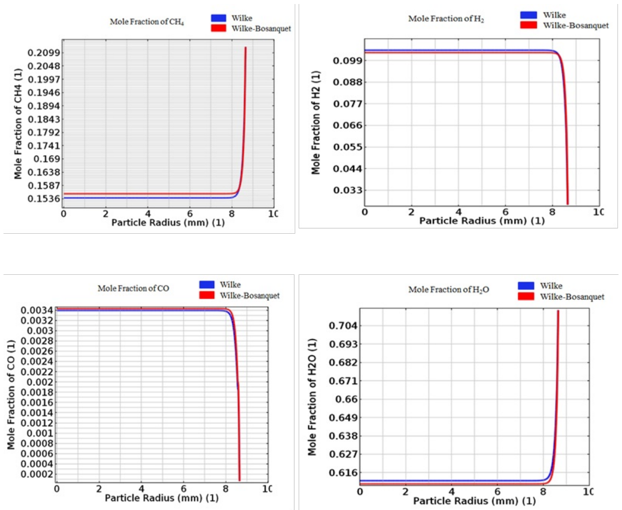

Figure 2: Mole Fraction profiles in a spherical catalyst pellet (Wilke and Wilke-Bosanquet Diffusion Flux Model)

Figure 3

Figure 4