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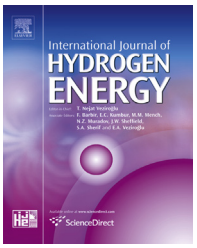

A systematic parametric study on the effect of a catalyst coating microstructure on its performance in methane steam reforming

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ABSTRACT

A systematic approach is presented to study the effect of a catalyst coating microstructure on its performance in a wall-coated steam methane reformer using response surface methodology. Three-dimensional simulation of diffusion and reaction are performed in several catalyst microstructures represented by packing of overlapping spheres. A surrogate model is developed based on Latin hypercube design of experiment and response surface methodology that relates the rate of hydrogen production in the catalyst coating microstructure to inter-particle porosity and average particle size. Two sets of simulations are done based on a kinetic model with two different sets of kinetic parameters and the results are compared. The comparison shows that the maximum rate of hydrogen production occurs at higher inter-particle porosity and smaller particle size, when the kinetics is faster and diffusion limitation is more severe.

Introduction

Methane steam reforming is a suitable option for efficient production of hydrogen in fuel cell systems. Wall-coated reactors have been used as compact fuel processors for hydrogen production and they have shown promising performance compared to conventional packed bed reformers [1,2]. Catalyst coating morphology has a crucial role in the optimal performance of a wall-coated reactor. The effect of the catalyst coating morphology on reformer performance can be demonstrated through multi-scale modeling in which, transport and reaction processes are simulated in computer-generated structures at difference length scales [3]. Random packing of spherical particles can be used as a practical representation of the catalyst morphology. It can capture the essential characteristics of the catalyst porous structure, like porosity, particle size and it can be used for parametric study and optimization. Reacting flow simulation in complex three-dimensional structures is computationally expensive to be used directly for structural optimization. In order to avoid performing many simulations, a statistical approach can be used, which combines design of experiments (DOE) and response surface methodology (RSM). RSM offers a surrogate

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model that establishes a functional relationship between the inputs and the outputs of the simulation based on the design points obtained from the DOE [4]. Then the surrogate model can be used for prediction and optimization purposes. Lian and Liou used an integrated optimization approach using RSM based on CFD simulations to redesign a turbo pump [5]. Ceylan et al. considered RSM for the optimization of an enzyme-catalyzed process, which involved polymerization of a phenolic compound [6]. Leon and Adomaitis applied RSM for a chemical vapor deposition (CVD) process to predict and optimize a thin film properties [7].

In our previous work, a multi-scale methodology was implemented in which the effect of the catalyst pore structure at different length scales was incorporated in a reacting flow simulation in the catalyst coating of a reformer [3]. It was demonstrated that for a given catalyst particle with certain pore structure, the inter-particle porosity and particle size have significant effect on the rate of hydrogen production in the coating. In this work, random packing of overlapping spheres is also assumed as the representation of the catalyst coating microstructure. This work focuses on implementing a systematic approach based on Latin hypercube design of experiment (LHD) and response surface methodology (RSM) to study the effect of inter-particle porosity and particle size on the volume-averaged rate of hydrogen production in a nickel-alumina spinel coating for methane steam reforming process. In this work, instead of assuming a certain intra-particle pore structure for the catalyst, X-ray nano-tomography (nano-CT) results are used to obtain a more realistic representation of the catalyst nanostructure. In the design of experiment, the range assumed for the particle size is based on the characterization results obtained from micro-computed tomography (micro-CT) of the coating. Details of the catalyst characterization by X-ray tomography as well as the discussion about the kinetic model used for the catalyst have been published in a separate paper [8].

Based on LHD, different microstructures with specific porosity and particle size were generated and a non-isothermal diffusion and reaction problem is solved based on boundary conditions obtained from a point near the reformer inlet, where transport limitations are significant and adjusting the pore structure can alleviate the limitations. The simulations are done using CFD software ANSYS FLUENT®.

Methodology

Latin hypercube design (LHD) is a widely used space-filling method for design of experiments [5,9]. It is specifically suitable when deterministic computer simulations such as this work are involved. LHD provides the flexibility to cover small and large design spaces with minimum unsampled regions, making it a suitable choice for computer experimenting [9,10].

In LHD, the range for each design variable is divided into '*n*' intervals with equal marginal probability. A random number is generated in each interval for each design variable. These '*n*' points for all the design variables are randomly matched to produce '*n*' design cases. For each variable, the '*n*' input values appear once in the design space [10]. Based on this method, there are numerous possibilities on how to scatter the design points throughout the experimental region.

The optimization of LHD has attracted considerable attention and has shown increasing popularity over the last few years [9]. Two criteria are considered to optimize the design of experiment; one is the orthogonality, which ensures minimum correlation between variables and the other, space-filling properties, which allow exploring all the portions of the experimental region [11]. In this work, the result of the method suggested by Cioppa and Lucas to generate nearly orthogonal LHD with good space filling properties is used [10]. The optimal LHD generation code is available online at http://harvest.nps.edu [12]. The LHD design generated for two design variables, namely, inter-particle porosity and mean particle diameter are shown in Fig. 1. Based on the design values obtained from LHD (Table 1), regression is done to fit the response surface model.

It should be noted here that for two factors, a typical factorial design with four levels and the center point (2^4 +1points) has good space filling properties [10] and may be good enough for this regression analysis, but this work is based on the assumption that this approach is used in the future for larger problems for which, LHD is a more efficient design for computer experimenting. The range for the inter-particle porosity is decided based on our previous work. It was demonstrated that the maximum hydrogen production occurs in the porosity range of 0.15–0.4 [3]. In this work, the mean particle diameter is assumed to be between 20 and 45 microns (Table 1).

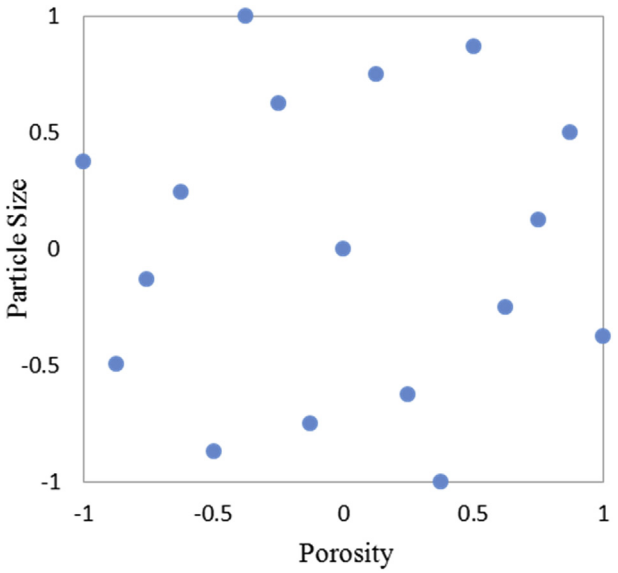

Fig. 1 – A Latin hypercube design with two design variables (inter-particle porosity and mean particle diameter) and seventeen design points, values for the variables are scaled to (−1, 1) range. Design values are shown in Table 1. The minimum and maximum values for porosity are 15% and 40% and for mean particle diameter are 20 and 45 μm. The center point (zero) in the LHD refers to 27.5% (porosity) and 32.5 μm (particle size).

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