Multiscale CFD model for the catalyzed ammonia oxidation: From a detailed surface kinetic model to an industrial reactor

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Highlights

- Multi scale model for ammonia oxidation
- Scale ranging from elementary surface mechanism to industrial reactor
- Model predicts meaningful conversions and selectivities

1. Introduction

With an annual world production capacity of 80 Mio. tons in the Ostwald Process [1], nitric acid is among the largest commodity chemicals. The first step of the process consists of the heterogeneously catalyzed oxidation of ammonia in air on Pt/Rh gauzes to form NO, with N_2 and N_2O being the main byproducts. Especially N_2O is highly unwanted, as it is a strong greenhouse gas and active in the depletion of the ozone layer [2]. Furthermore, the production of ammonia is an energetically expensive process, so that even small improvements in the NO selectivity allow significant energy savings.

Due to its industrial importance, a large number of surface science studies has been devoted to the investigation of the NH_3 oxidation on platinum, and today there seems to be reasonable agreement on the reaction mechanism. Based on this surface science work, a number of micro-kinetic models have been proposed. Despite all this fundamental research, there seems to be no published work that applies the developed reaction mechanisms towards the simulation of a catalytic gauze or even an industrial ammonia burner.

In this contribution, we present CFD simulation of an industrial ammonia burner based on a published surface kinetic model. Such a simulation is numerically challenging due to the combination of different length scales from ~100µm for the wires up to several m for the reactor dimensions. The different scales are bridged using a solution mapping approach. The idea is to solve the reaction diffusion equations at the gauze scale for a large number of local operating conditions [4]. These solutions are then represented by a spline map, so that in the reactor scale simulation the computation of the gauze scale can be replaced by a simple look-up operation.

2. Methods

Laminar CFD simulations at the level of a resolved gauze are performed in FLUENT using a surface kinetic model published by Kraehnert and Baerns [3]. A typical solution is shown in Fig. 1b. The gauze scale simulations were repeated for a defined parameter set (gas velocity, catalyst temperature and feed composition) and the resulting concentration changes over the whole catalyst due to chemical reactions were stored into look-up tables.

The reactor scale model was also implemented using the commercial package FLUENT. Here, the flow in the open reactor was described by a k-ω SST turbulence model [5] while the gauze was described as a porous zone, with the permeability and Forchheimer coefficient determined from the gauze scale simulations. During the simulation, species source terms describing the NH_3 combustion on the gauzes are interpolated by the spline maps using cubic Hermiete splines [6].

3. Results and discussion

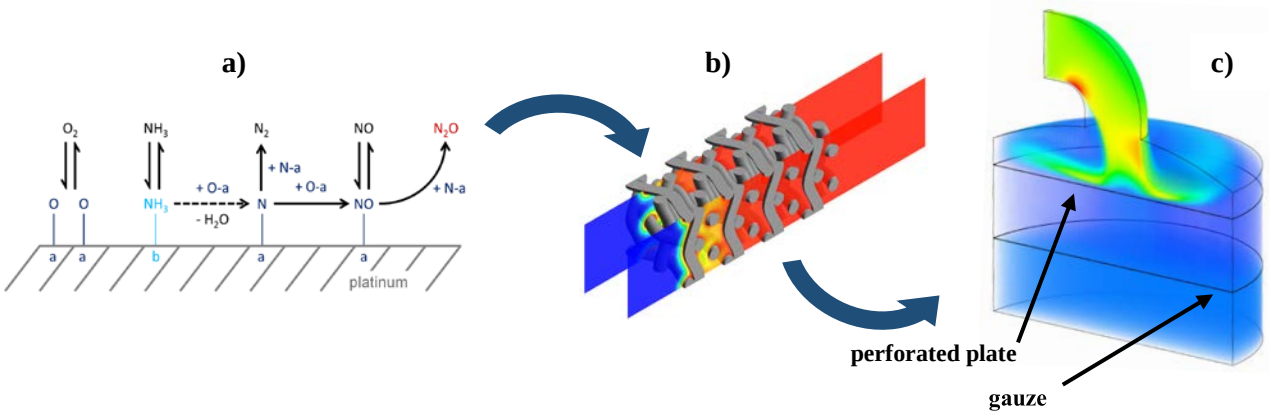

Figure 1. Hierarchical scheme of the multiscale reactor model.

A typical simulation result for a reactor containing a perforated plate for the flow homogenization is shown in Figure 1c. The inhomogeneous flow due to the curved inlet is homogenized by the perforated plate, resulting in an even distribution on the gauze. Using the tabulated gauze-scale solutions, a computation of the reactor-scale model with an 1.8 mio. element mesh is accomplished in about 6 hours on a parallelized machine using 12 cores. First results of the reactor simulation show meaningful conversions and selectivities.

4. Conclusions

This work presents the first attempt to develop a multi scale model for an ammonia oxidation reactor that uses solution maps obtained from CFD simulations of the resolved catalyst, coupled with a detailed surface kinetic model. Such a model can be used, to investigate how the reactor geometry influences reactor performance and therefor has the potential to aid the design of reactors exhibiting lower N_2O selectivities and higher NO yields.

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Keywords
Multi scale CFD model, Ammonia oxidation, CFD, Look-up tabulation, solution map, spline