Efficient Implementation of Detailed Surface Chemistry into Reactor Models Using Mapped Rate Data

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Introduction

In recent years more and more mechanistic understanding of many catalytic processes becomes available. However, this understanding can frequently not be fully exploited in reactor simulations due to computational limitations. The approach of the current contribution is to substitute the full numerical simulation of the detailed reaction mechanism by the use of precomputed rate information. A spline-representation of the data [1] is used to compute efficient reaction rates during runtime of the reactor simulation.

While tabulated rate data has been used before for a number of gas phase systems[2,3], there are several reasons why this approach seems to be particularly advantageous for catalytic reaction systems :

- surface mechanisms generally show a wide bandwidth of timescales which makes the solution of the corresponding differential equations numerically demanding.
- Catalytic reactors frequently have a complex and deeply structured multi scale geometry. This makes reactor simulations a challenge.
- Identification of the steady state species is not a problem in heterogeneous catalysis since surface species are immobile and hence can be naturally treated as steady state species, at least for steady state reactor simulations. Identification of the steady state species is a major issue during the construction of rate tables for gas phase reaction systems.

Materials and Methods

The procedure mapping is demonstrated using a surface mechanism for CH_4 oxidation on platinum (11 surface species, 19 reactions) by Deutschmann et al. [4]. Reaction rates are computed by the program package Cantera. A 3D model of a monolith channel is implemented in the commercial simulation program Comsol.

Results and Discussion

It is shown that a mapping of precomputed rate data can be used to efficiently implement detailed surface mechanisms into reactor simulations. The spline representation reproduces the effective CH_4 conversion rates with an error of less than 0.5 % (see table 1). The mapping approach is implemented in a 1D model and the computed concentration profiles are compared to the results of a full numerical solution. In all cases the concentration and temperature profiles of the exact solution are perfectly reproduced by the mapping approach (Figure 1).

To demonstrate the potential of the approach for complex reactor simulations a 3D model of a monolith channel including the irregularly shaped washcoat is implemented in a commercial CFD program. A steady state solution of this model takes 20 s on a 1.5 GHz Pentium computer. This corresponds to a speed up of three orders of magnitude compared to the solution of the same model coupled to a full numerical solution of the surface chemistry.

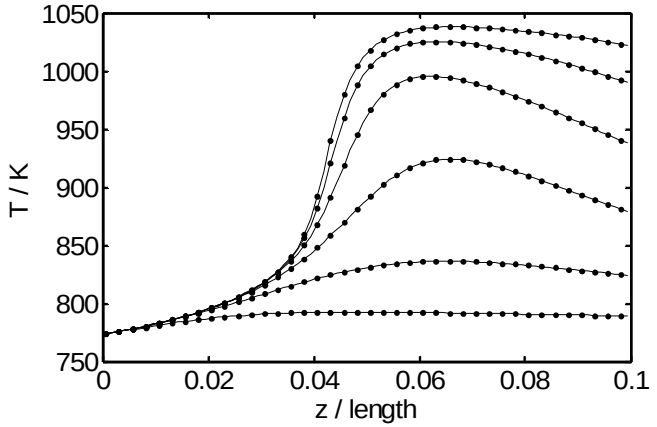
Significance

The promise of the approach presented in this paper is that it decouples the time needed for the numerical treatment of the surface processes from the computing time in a reactor simulation. This should allow the application of increasingly complex surface chemistry models that can not be implemented in realistic reactor models today.

Table 1. Performance of the rate approximation by the spline mapping

	t [s]	Speed up	Av. relative error
Linear spline	0.018	2350	2.8 %
Quadratic spline	0.032	1300	0.3 %
Cubic spline	0.079	540	0.16 %

Figure 1. Axial temperature profiles in a monolith reactor during ignition of the reactor. Lines: Full numerical simulation. Dots: Spline approximation



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