Time-Dependent Behavior of Reactions in Catalytic Monoliths

O. Deutschmann[1], S. Tischer[1], and J. Warnatz[1]

Abstract: Surface oxidation of hydrocarbons are very important processes. Examples are (1) catalytic combustion which leads to NO-free exhaust gas due to the absence of O atoms and CH radicals in the gas phase and (2) exhaust gas treatment like by the three-way catalyst for gasoline engines and lean DeNOx for direct-injected Diesel and Otto engines.

Catalytic multi-channel monoliths are used in these applications, and the modeling of the transient behavior of such monoliths is interesting in investigating, for e.g. the cold start of automotive catalytic converters or the start-up / shut-down of chemical processes. The aim of this project is to develop detailed models to describe the time-dependent physical and chemical processes in the monolith. The time scales of transport and chemical reactions in an individual catalytic channel are of the order of 1 ms, while those of the thermal variations in a monolith of the order of seconds and are thus decoupled. This difference in timescales is exploited: steady state solutions for the gas flow in the individual channels are used to provide heat source terms for a transient solution of the solid. The fluid flow in the channels is described using a boundary-layer approach with gaseous and surface reactions using elementary reaction mechanisms (DETCHEM [1]) and accounting for pore diffusion in washcoats. A transient heat conduction equation in 2d / 3d is solved accounting for heat losses at the outer walls and time-varying inlet conditions.

The result is a numerical simulation of the transient temperature field and the species concentrations for the entire catalytic monolith. As an example, catalytic combustion of methane [2] and catalytic exhaust gas removal after an Otto engine [3] are simulated in 2d and 3d where the outlet temperature and species concentrations as functions of time were predicted.

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[3] J. Braun, T. Hauber, H. Többen, J. Windmann, P. Zacke, D. Chatterjee, C. Correa, O. Deutschmann, L. Maier, S. Tischer, J. Warnatz: Three-Dimensional Simulation of the Transient Behavior of a Three-Way Catalytic Converter. SAE Technical Paper Series (2002), in press

[1] Interdisciplinary Center of Scientific Computing (IWR), Heidelberg University
Im Neuenheimer Feld 368, D-69120 Heidelberg, Germany
juergen@warnatz.de