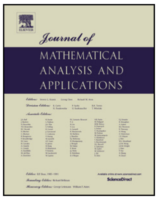
J. Math. Anal. Appl. 445 (2017) 677–709

Contents lists available at ScienceDirect

Journal of Mathematical Analysis and Applications

www.elsevier.com/locate/jmaa



# Global strong solutions for a class of heterogeneous catalysis models

Dieter Bothe [a], Matthias Köhne [b,*], Siegfried Maier [b], Jürgen Saal [b]

[a] *Center of Smart Interfaces and Department of Mathematics, Technische Universität Darmstadt, Alarich-Weiss-Str. 10, 64289 Darmstadt, Germany*
[b] *Mathematisches Institut, Heinrich-Heine-Universität Düsseldorf, Universitätsstr. 1, D-40225 Düsseldorf, Germany*

ARTICLE INFO

*Article history:*
Received 28 January 2016
Available online 11 August 2016
Submitted by Y. Yamada

*Keywords:*
Global existence
Strong solution
Chemisorption
Diffusion–sorption–reaction system

ABSTRACT

We consider a mathematical model for heterogeneous catalysis in a finite three-dimensional pore of cylinder-like geometry, with the lateral walls acting as a catalytic surface. The system under consideration consists of a diffusion–advection system inside the bulk phase and a reaction–diffusion–sorption system modeling the processes on the catalytic wall and the exchange between bulk and surface. We assume Fickian diffusion with constant coefficients, sorption kinetics with linear growth bound and a network of chemical reactions which possesses a certain triangular structure. Our main result gives sufficient conditions for the existence of a unique global strong $L^2$-solution to this model, thereby extending by now classical results on reaction–diffusion systems to the more complicated case of heterogeneous catalysis.

## 1. Introduction

Catalysis is a key technology in Chemical Engineering, employed not only to increase the speed of chemical reactions by up to several orders of magnitude, but also to change the selectivity in favor of a desired product against other possible output components of a chemical reaction network. In heterogeneous catalysis, the catalytic substance forms a separate phase which is advantageous concerning the separation of the products from the catalytic material. A prototypical setting, which also underlies the mathematical model below, consists of a solid phase catalyst brought into contact with a gas or liquid which carries the educts as well as the product species inside the chemical reactor. In this case, the overall chemical conversion consists of the following steps:

---

[*] Corresponding author.
*E-mail addresses:* bothe@csi.tu-darmstadt.de (D. Bothe), koehne@math.uni-duesseldorf.de (M. Köhne), maier@math.uni-duesseldorf.de (S. Maier), saal@math.uni-duesseldorf.de (J. Saal).

http://dx.doi.org/10.1016/j.jmaa.2016.08.016
0022-247X/

(1) the educt species are transported to the surface of the catalytic substance;
(2) molecules of at least one educt species adsorb at the catalyst surface;
(3) adsorbed molecules react, either with other adsorbed molecules or with molecules in the bulk phase directly adjacent to the surface;
(4) the product molecules are desorbed.

Of course, further processes will usually appear as well. For instance, adsorbed educt molecules may desorb back into the bulk before a chemical reaction occurs, or they can be transported along the surface by means of surface diffusion processes. For a recent view on the complexity of heterogeneous catalysis modeling see [12].

In the present paper, we only consider the case of pure surface chemistry, i.e. chemical reactions are only allowed between adsorbed species. This is actually no restriction, since one may otherwise introduce an artificial adsorbed form of the reaction partner which is in the bulk adjacent to the surface and assign to it an infinite adsorption rate such that all arriving bulk molecules immediately adsorb and, hence, are available for surface reaction.

In order for a heterogeneous catalytic process to be efficient, a large surface area is required. Therefore, in classical heterogeneous catalysis with solid phase catalyst, the latter is often provided as a porous structure, e.g. in so-called packed-bed reactors. In this case, the smallest unit is a single pore, into which the educts have to be transported in order to reach the pore wall, i.e. the catalytic surface. More information on this classical reactor concept can be found, e.g., in [4,16] or [30]. In recent years, with the advent of microreactor engineering technology, new reactor designs became feasible. Due to the large area-to-volume ratio at the micro scale, multichannel microreactors with catalytic wall coatings can replace classical porous structures and still provide fast and intense diffusive transport to the channel walls in order to facilitate the reaction speed or selectivity enhancement; see, e.g., [29,26]. Since the given and precise structure of microreactors together with modern control and measurement techniques allows for defined and reproducible operating conditions, this approach is much better accessible for detailed quantitative modeling and simulation; cf. [7,8]. Structured catalytic microreactors are also employed for efficient screening of potential catalysts for new reaction pathways; see, e.g., [17]. To tap the full potential of such microsystems approaches and to intensify also more classical heterogeneous catalysis processes, realistic and sound mathematical models are required as the basis for any numerical simulation. The most fundamental question then is whether a given model is well-posed, a necessary requirement to enable any reasonable numerical treatment.

In what follows, we consider a single pore as a prototypical element, where we allow for convection through the pore with a solenoidal velocity field which is assumed to be known and to satisfy the no-slip boundary condition at fixed walls. We focus on pores having smoothly bounded cross shapes. Let therefore $\Omega := A \times (-h, h) \subset \mathbb{R}^3$ denote a finite three-dimensional cylinder of height $2h > 0$ with cross section $A \subset \mathbb{R}^2$ being a bounded simply connected $C^2$-domain, such that $\partial A$ is a closed regular $C^2$-curve. The boundary of $\Omega$ decomposes into bottom $\Gamma_{\text{in}}$, top $\Gamma_{\text{out}}$ and lateral surface $\Sigma$, standing for inflow area, outflow area and active surface. The mathematical model consists of the partial mass balances for all involved chemical components, both within the bulk phase $\Omega$ (representing the interior of the pore) and on the active surface $\Sigma$ (representing the catalytic surface). Inside the bulk phase, the species mass fluxes are due to advection and diffusion, where we assume the latter to be governed by Fick's law. On the active surface, we only consider diffusive fluxes along the surface, again assuming Fick's law to be a reasonable constitutive relation. We allow for different diffusivities but the model ignores cross-diffusion effects. Let us note in passing that for high surface coverage, cross-effects between the transport of different constituents will appear which are not accounted for by our model. The mass exchange between bulk and active surface is due to ad- and desorption phenomena, which are usually modeled via kinetic relations in analogy to chemical reaction kinetics. Examples will be discussed below.