DESIGN AND MODELING OF A NOVEL MEMS-BASED MICROREACTOR FOR GAS-PHASE CATALYTIC REACTIONS

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Summary

Design methods and performance evaluations of a novel catalytic microreactor for catalytic gas-phase reactions based on MEMS fabrication techniques are described. The hardware platform is a commercial computer chassis whose slots accommodate special-purpose circuit boards for specific process functions, such as gas mixing, flow control, temperature control, chemical reaction, and process safety. The oxidation of ammonia over a 0.1 μm Pt film catalyst is used as a test reaction. Closed-loop PID temperature control was demonstrated at ca. 200Hz, which exceeds typical responses from a conventional laboratory fixed-bed reactor by at least one order-of-magnitude. Detailed 2D and 3D multiphysics models are developed that capture the transient and steady-state reactor performance.

Keywords

Micro-reactors, novel reactor technologies, process intensification, multi-scale analysis

Introduction

Miniaturization of electro-mechanical systems, analytical instrumentation, fluid transport hardware, process sensors, process analyzers, and other technological devices over the past decade has been a key driver in the development of Microreaction Technology (MRT) [1]. Advances in MRT for catalytic reacting systems have been presented annually since 1997 at the International Conference on Microreaction Technology (IMRET) with the latest one being IMRET-10 [2]. The advantages of MRT and miniaturization of reactor system versus conventional reactor systems include a smaller system footprint, lower process hazards, reduced reagent consumption, lower rates of waste generation, higher rates of heat and mass transport, and improved process dynamics and control [3]. Nevertheless, achieving these advantages also leads to new product engineering challenges, such as fabrication of precision miniature components and process sensors, creation of robust component packaging and device integration schemes, defining methods for incorporating catalysts, and creation of new instrument control schemes. Mathematical modeling of microreactor systems also introduces new challenges since typical geometries require a detailed accounting of scalar and vector field quantities (fluid velocities, fluid pressures, temperatures, species concentrations, etc.) in two or three spatial dimensions over time and length scales that often vary over many orders-of-magnitude. For these reasons, new opportunities exist for development and application of new modeling tools for microreactor design, fabrication, and analysis of system performance.

The primary objectives of this paper are to describe a new miniaturized reactor system for studying gas-phase heterogeneous reactions, and to interpret reactor performance data using the catalytic oxidation of ammonia as a test reaction. Analysis of reactor performance is based upon multidimensional steady-state and dynamic models that capture the system physics. The reactor hardware represents a notable departure from existing approaches for MRT systems since control of all key system functions, such as reactant flows, temperatures, gas-phase catalyzed reactions, and process analytical, is performed using an array of custom-designed circuit boards that are compatible with a Compaq PCI™ computer chassis. The reactor performance analysis takes advantage of recent developments in COMSOL Multiphysics Version 4.0 for solution of the microscopic temporal-spatial forms for the transport-kinetic equations.

Materials and Methods

Figure 1a shows a photograph of the Compaq PCI™ computer chassis that comprises the reactor system hardware platform. It contains two reactor circuit boards along with various supporting circuit boards that control various temperatures and reaction gas flow rates, and for monitoring of process safety interlocks. Each reactor board contains two parallel reactor channels on a single multi-laminate silicon wafer chip die. The microreactor socket is mounted on the reactor boards and is shown in Figure 1b. The reaction channel is a rectangular duct with dimensions of 50 µm x 50 µm x 500 µm and contains seven Pt metal heaters and temperature sensors. Fabrication is performed using methods employed in MEMS. The reactor die can be easily installed or removed using a modified industry-standard DieMate™ socket that is normally used for parallel testing of integrated circuits. Each reaction channel etched on the chip resembles a "Y" where each top leg is supplied by two independently-controlled feed gas streams with on-board mixing. The Pt catalyst is deposited as a thin 0.1 µm film on the underside of a SiN film that forms the top layer of the reactor.

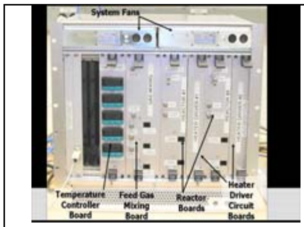

Fig 1a. Computer chassis showing various circuit boards for process functions.

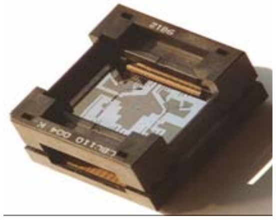

Fig 1b. Silicon chip-based microreactor with dual channels.

Experimental Results

Typical experimental results for the oxidation of NH_3 over a Pt film from one of the four microreactor channels are shown in Figure 2. The conversion of NH_3 at temperatures less than 200°C was not measurable, whereas the conversion approached 100% at temperatures in excess of 300°C. The selectivity data approach ca. 30% from 250 to 500°C and show that significant amounts of NO are produced. Small amounts of N_2O are also detected at an initial temperature of ca. 400°C.

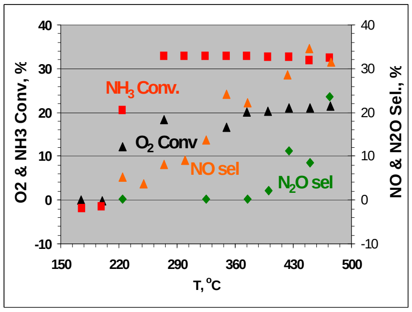

Figure 2. Microreactor data for NH_3 oxidation.

Reactor Modeling

Detailed 2-D and 3-D models of various microreactor designs that account for transport-kinetic interactions have been developed for interpretation of the measured performance data. The bases for these models are geometrical representations using Computer-Aided Design (CAD) tools. An example of a CAD drawing for a reactor design involving 7 Pt heaters that are evenly spaced along the underside of the SiN layer is shown in Figure 3. The feed streams containing the hydrocarbon and oxygen are introduced on opposite sides of the tee and then combined at the junction where fluid mixing and reaction occur. The reaction product is removed through a port located perpendicular to the plane of the die.

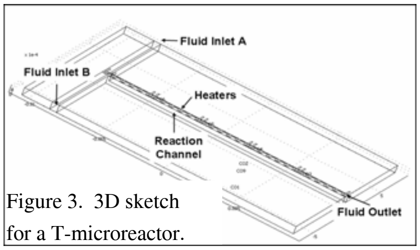

Figure 3. 3D sketch for a T-microreactor.

A 2-D representation of the actual 3-D reactor was used as the starting basis for modeling. The serpentine heaters were replaced by solid heaters that were distributed across the reactor width. The block length was equivalent to the one used for the serpentine design, except the heater thickness was adjusted to produce the same Pt metal volume. The model consists of the microscopic forms of the conservation equations which are solved using COMSOL Multiphysics Vers 4.0. Reaction kinetics for ammonia oxidation were taken from literature models and used as the starting basis for data fitting. Details are given in the full-length paper.

Preliminary Results

Figure 4 shows the model-predicted values for the NO concentration profiles at various feed gas flow rates. A full 3-D reactor model is needed to obtain an accurate representation of the averaged experimental specie concentrations measured at the reactor exit. Closed-loop PID temperature control was demonstrated at about 200Hz, which exceeds typical responses from a conventional laboratory fixed-bed reactor by at least one order-of-magnitude.

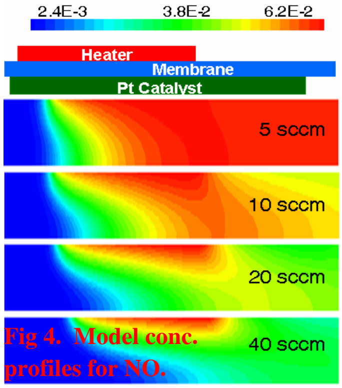

Fig 4. Model conc. profiles for NO.

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