CHAPTER 4

MICROSCALE COMBUSTION MODELING

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Microreactor modeling encompasses intricate interactions between fluid flow, chemical kinetics, species transport, heat conduction in the solid, and external heat loss mechanisms. Numerical models are reviewed for stationary and transient microreactor simulation, focusing on both catalytic and non-catalytic systems. One-dimensional models and subsequently multidimensional simulations with detailed chemistry and transport are introduced and applied to investigate the combustion stability and start-up of microreactors, flame dynamics in catalytic and non-catalytic channels, and turbulent hetero-/homogeneous channel-flow combustion. Key comparisons of simulations with experiments are provided, demonstrating the capacity of the employed models. One-dimensional and two-dimensional simplified models based on perturbation theory (activation energy asymptotics) are also presented, providing closed-form solutions in micro-channel combustion (ignition criteria, flame propagation speeds). Limitations of the quasisteady-state assumption for the gas-phase in transient microreactor modeling and finite Knudsen number models based on the Lattice Boltzmann Method (LBM) are finally outlined.

4.1 INTRODUCTION

Microreactors used for portable power generation and fuel processing encompass a wide range of geometrical configurations and fuel conversion methodologies. The latter involve homogeneous (gas phase), heterogeneous (catalytic), and combined hetero-/homogeneous combustion processes. Microreactors for fuel processing exclusively employ catalytic approaches to achieve demanding product yields and selectivities. On the other side, all three aforementioned fuel conversion methods can be utilized for power generation applications, whereby total oxidation is required. The fundamental physicochemical processes in each microreactor type are outlined next so as to pave the way for the presentation of appropriate mathematical models.

Hetero-/homogeneous microreactors entail the largest complexity and are discussed first. Although various reactor configurations have been established, heterogeneous conversion in many small-scale power and chemical engineering processes is achieved in honeycomb structures comprising straight channels (Figure 4.1a), with each channel having a hydraulic diameter ranging from 0.5 to 2.0 mm. Such reactors are also used for large-scale power generation [1, 2], the main difference being the higher mass throughput per channel and the larger

number of channels per honeycomb structure as compared to microreactor applications [3]. Depending on the power requirements, single-channel geometries with large cross-flow aspect ratios have also been constructed (Figure 4.1b). Moreover, excess enthalpy (recirculation type) catalytic microreactors with complex three-dimensional geometries have been tested (configurations similar to the non-catalytic Swiss-roll microreactor shown in Figure 4.2a). The underlying physicochemical processes are shown in Figure 4.1c within the context of straight catalytic channel geometries.

Fuel and oxidizer are mixed and subsequently enter the reactor at inlet velocities that yield, for most power-generation and chemical processing applications, laminar channel flows [6, 7]. Both reagents diffuse transversely to the channel walls and react on the catalytic surfaces. Heterogeneous ignition (light-off) is achieved at a certain distance from the channel entry, which depends not only on the operating conditions (inlet temperature, velocity and pressure, fuel type, stoichiometry) and the particular catalyst formulation, but also on key in-channel heat
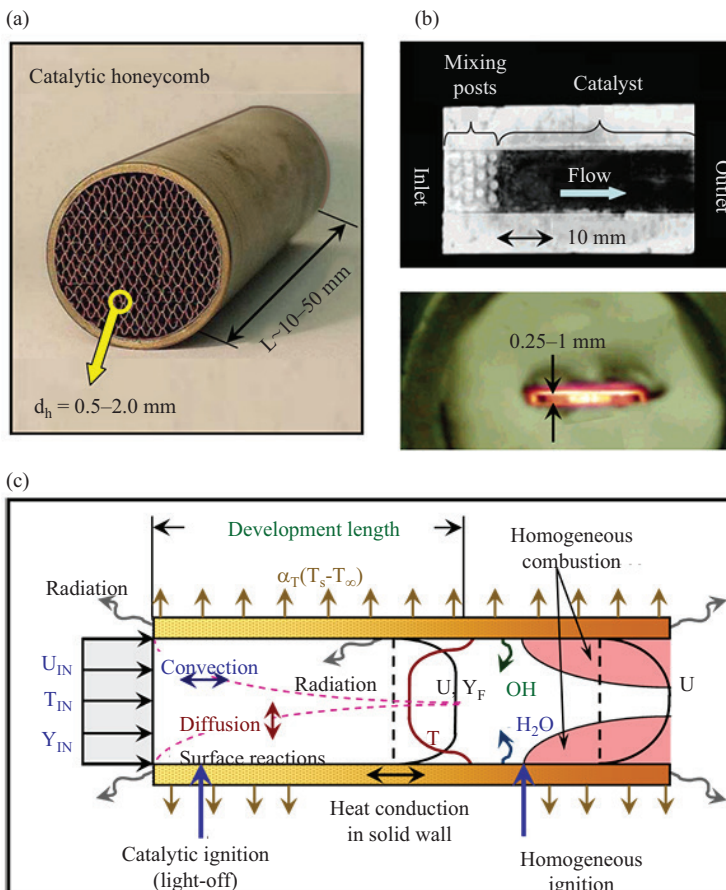

Figure 4.1. (a) Typical honeycomb catalytic structure made of FeCr alloy and coated with Pt, similar to the one used in Karagiannidis et al. [3] for mesoscale power generation, (b) rectangular-shaped, single-channel, Pt-coated catalytic microreactor (reproduced by permission from Norton et al. [4]), and (c) physicochemical processes within a catalytic channel.