Catalytic Methane Combustion in Microreactors

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Chapter 1

Aim and scope of the thesis

Methane combustion has been a focus research topic for decades. The large volumetric flow rate of methane and a relatively high concentration in practice present a major challenge for the combustion technology development in order to abate the associated pollutant emission. Many researches on the catalytic methane combustion at low temperatures (typically < 600 ^oC) have been investigated towards achieving a lower light-off/working temperature, and with that less exhaust emission and more stable combustion. The ideal catalysts for the methane combustion endowed with high temperature stability, low activation energy and excellent resistance to poison still need to be further developed. Apart from the catalyst development, there is a quest for efficient reactor engineering concepts. Microreactors offer a great promise and can be broadly employed for intensifying (catalytic) reaction processes, especially for strongly exothermic reactions (such as the methane combustion). This is mainly due to the enhanced heat and mass transfer in microreactors, allowing the suppression of hot spot formation and a better catalyst usage for the improved reaction rate. The washcoat catalyst immobilized onto the walls of microreactors avoids high pressure drop and temperature gradient likely present in microreactors with packed catalyst bed. Thus, the catalytic combustion of methane over the washcoat catalyst (particularly of the promising noble metal based one) in microreactors represents an attractive research field, and is thus dealt with in this thesis. In this respect, a good adhesion and dispersion of the catalytic coating layer onto the microreactor wall is essential, and the influence of the coating properties, fluid flow, mass transfer and reaction parameters in various microreactor geometries should be thus well addressed towards obtaining an optimal performance of the catalytic methane combustion.

1.1. Research objective

The main aims of this thesis are to first develop the washcoating method of $Pt/\gamma\text{-}Al_2O_3$ catalyst in FeCrAlloy and stainless steel microreactors. The optimized coating method was then applied in single- and multi-channel microreactors to investigate the mass transfer behaviour and performance of the catalytic methane combustion, over both the single- and multi-layer coating systems. Typical tested operating conditions include the reaction temperature, total flow rate, O_2/CH_4 molar ratio, Pt loading and weight. Moreover, different internal channel configurations of microreactors have been experimentally studied for the catalytic methane combustion, where the preliminary results on the optimized reaction conditions and configurations were presented. The present thesis may serve as a useful reference for the catalytic methane combustion, by providing valuable insights into the microreactor design and operation, as well as the wall-coated catalyst for the potential upscaling in the industrial applications.

1.2. Thesis outline

The main structure and contents of this thesis are as follows:

Chapter 2

This chapter provides a comprehensive literature review on the catalytic methane combustion at low temperatures regarding catalysts, mechanisms, reaction conditions and reactor designs. A detailed description of the current understanding into the influence of various operational conditions and reactor types on the reaction performance have been well addressed. This chapter may offer an essential and comprehensive reference for developing the optimal strategies by selection of the active/stable catalyst equipped with appropriate reactors under different operating conditions.

Chapter 3

This chapter deals with the development of the methods for coating Pt/γ-Al_2O_3 catalyst onto microreactor walls in order to form a stable, homogeneous and well-adhesive coating layer. The influence of binder properties, pH value, γ-Al_2O_3 concentration, initial particle size and slurry viscosity has been particularly studied and discussed in details. The influence of substrate material and channel shape on the coating adhesion has also been evaluated. The different operational conditions and temperature distribution (along the microchannel) were briefly investigated in the microreactor for the catalytic methane combustion. The main aim of this chapter is to shed light on the preparation and characterization of the washcoat slurry preparation in the microreactor.

Chapter 4

This chapter presents the catalytic performance of single- and multi-layer Pt/γ-Al_2O_3 catalytic coatings inside the capillary microreactor, with regard to the effect of the reaction temperature, total flow rate, molar ratio of O_2 to CH_4, Pt loading or mass. The results provide a good understanding of mass transfer limitations that could be present during the catalytic methane combustion in microreactors. Additionally, the stability of catalyst have been examined. This chapter may serve as a good reference for the qualitative discussion about the effect of the (external and internal) mass transfer over the single-and multi-layer systems on the reaction performance in microreactors.

Chapter 5

Based on the findings of Chapter 3, this chapter firstly investigated mainly the influence of different operational conditions (e.g., working temperature, total flow rate), Pt/γ-Al_2O_3 coating properties (e.g., specific catalyst loading) on the catalytic combustion of methane in the straight parallel channel microreactor. In order to optimize the microreactor geometry, five other types of microreactors with different internal channel configurations have been coated with Pt/γ-Al_2O_3 catalyst, and tested regarding their performance in the catalytic methane combustion, which is experimentally rarely reported according to our best knowledge. The main aim of this chapter is to provide a useful reference on the optimization of microreactor designs that can ensure a good coating surface area, residence time and fluid uniformity towards achieving a favourable methane conversion.

Chapter 6

This chapter summarizes the main conclusions of this thesis, and provides an outlook with main focuses on the trends, challenges and future perspectives relevant to the catalytic methane combustion in microreactors. It is categorized into a short-term investigation that is required as a continuation of the current thesis work, and a long-term research for future investigation. The contents include the improvement of the noble metal catalyst activity, coating preparation, catalyst deactivation, reaction kinetics and mechanism, and different microreactor systems for coupling exothermic and endothermic reactions. These are deemed to provide a useful reference for the future research and application of microreactor systems in this upcoming field.