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Li He

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Catalytic Methane Combustion in Microreactors

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and in accordance with
the decision by the College of Deans

and

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and in accordance with
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This thesis will be defended in public on

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Dedicated to my beloved family.

谨以此书献给我亲爱的家人

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Abstract

The current thesis deals with the catalytic methane combustion in microreactors with wall-coated Pt/γ-Al_2O_3 catalyst. The Pt/γ-Al_2O_3 washcoat preparation, the single- and multi-layer catalytic coating systems, and the different designs of microreactor geometries were particularly investigated. Various aspects were thus addressed, including the preparation procedures of the catalyst coating (e.g., the binder properties, pH value, initial Al_2O_3 particle size), the optimization of different reaction conditions with single- and multi-layer coating systems (e.g., temperature, flow rate, O_2/CH_4 molar ratio, Pt loading and coating thickness), the effect of internal channel configurations in the microreactor (i.e., involving straight parallel channels, cavity, double serpentine channels, obstacled parallel channels, meshed circuit and vascular network) on the reaction performance.

The thesis starts with a comprehensive literature review on the catalytic methane combustion at low temperatures, including catalyst, mechanisms, reaction conditions and reactor designs. Then, the preparation procedures of Pt/γ-Al_2O_3 washcoat catalyst have been studied in details, in order to improve its adhesion and uniformity on FeCrAlloy and stainless steel microreactors. A good adhesion could be obtained by using the slurry with 20 wt% γ-Al_2O_3 (particle size: 3 μm), pH = 3.5, and 3 to 5 wt% polyvinyl alcohol (molecular weight of 57,000 - 186,000). Based on the above-mentioned optimized preparation, Pt/γ-Al_2O_3 washcoat catalysts of various loadings were deposited inside the stainless-steel capillary microreactors and studied both in the single- and multi-layer catalytic coating systems. The influence of different operating conditions including the reaction temperature, total flow rate, molar ratio of O_2:CH_4, and the reproducibility of catalyst were tested. The results demonstrate that in general the methane conversion was improved with the temperature rise, and presented the highest at an oxygen to methane molar ratio of ca.1.5. An obvious decrease in the methane conversion could be found over the multi-layer systems compared to their respective single-layer counterparts (if the Pt mass in the catalyst was kept equal), due to the more significant internal diffusion limitation in thicker coatings. Among all the tested microreactor geometries washcoated with Pt/γ-Al_2O_3 catalyst, the highest methane conversion could be obtained in the double serpentine channel microreactor and the lowest presented in the mesh circuit microreactor, which can be explained based on the available coating surface area, flow distribution and residence time property. In order to achieve a desirable methane conversion in microreactors, a proper tuning of the catalytic coating properties (e.g., surface area, Pt loading and thickness), the residence time, the fluid distribution uniformity and other reaction parameters (e.g., temperature and oxygen to methane molar ratio) are required.

Keywords: Catalytic methane combustion; Pt/γ-Al_2O_3 catalyst; microreactor; coating; mass transfer