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Influence of a reaction mixture streamline on partial oxidation of methane in an asymmetric microchannel reactor

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ABSTRACT

Partial oxidation of methane (POM) has been tested in an asymmetric microchannel reactor with different inlet configurations. One inlet of the reactor provided successive splitting of an inlet flow into parallel channels, whereas the opposite inlet allowed the inlet flow to enter the parallel channels simultaneously. It was found that concentrations of carbon monoxide and carbon dioxide changed by 20–30% and the conversion of methane changed by 5–20%, depending on the rate and direction of the inlet flow. The hydrogen production rate practically did not depend on the inlet configuration and equaled 15 l/h at the inlet flow rates from 600 to 1400 cm³/min and at the methane conversion of 80%. The data obtained demonstrated that the use of different operating modes of the asymmetric microreactor allows changing the composition of produced syngas.

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1. Introduction

Hydrogen is widely anticipated to become an important energy source in the future [1–5]. However, large-scale applications of the commercially generated hydrogen-containing gas (syngas) are accompanied with a number of problems associated with the storage and transportation of large amounts of syngas. Recently started studies have shown that microchannel systems can be successfully used for the catalytic conversion of carbon-containing substrates into hydrogen-containing gas in the immediate vicinity of the syngas

consumption, which avoids many storage and transportation problems. Moreover, microchannel systems permit a considerable intensification of the catalytic hydrogen production compared with the traditional fixed bed catalytic reactors [6]. A promising technology for the hydrogen production is the catalytic partial oxidation of methane (POM) into syngas, which has a number of advantages over traditional processes [7]. Other examples of using microreactors in POM are described in Refs. [8,9].

M. Prettre et al. [10] were first to show that the temperature in the POM rises considerably at the inlet of the reactor and

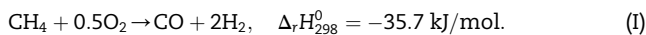
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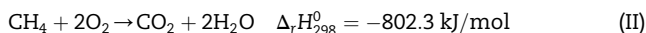
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then decreases to the temperature of a furnace in which the reactor is mounted. This observation may argue for the occurrence of exothermic reactions of methane oxidation with the oxygen indices from 0.5 to 2.0, while the reactions in the other regions are endothermic. In Refs. [11,12], authors showed that the POM can proceed via the direct mechanism at high temperatures (750–1000 °C), short contact times (0.02–0.2 ms), and preferably over a Rh catalyst:

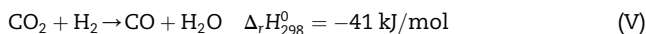
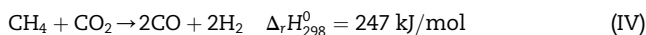
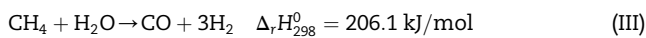


This fact is supported by the formation of carbon monoxide at the inlet of the reactor and by the formation of hydrogen at 1000 °C.

At longer contact times and over Pt catalysts, the POM proceeds via an indirect oxidation mechanism that involves total oxidation of methane followed by steam and carbon dioxide reforming reactions; this scheme is often referred to as the Combustion and Reforming Reactions (CRR) mechanism. At the inlet of the reactor, the total oxidation of methane (TOX) proceeds with the formation of carbon dioxide and water:



In the other regions of the reactor, the endothermic reactions of steam (III) and carbon dioxide (IV) reforming of methane, as well as the reverse water gas shift reaction (V), proceed:



The further works on numerical simulation of the process, as well as related experimental data, demonstrated that the CRR mechanism is valid for most cases, when the contact time is longer than 0.2 ms [13].

A significant problem associated with the mechanism of the POM and arising during long-term operation of POM microreactors is overheating of the front edge of the MC plates, which results in the corrosion and destruction of the MC plates and hence in appreciable reduction of their lifetime. In Ref. [14], we proposed to use specific Z-shaped MC plates with the countercurrent reagent flow to reduce overheating of their front edge.

The aim of the present work was to determine the influence of the channel configuration at the inlet of a microchannel reactor on the occurrence of POM and on the overheating of front edges of MC plates.

2. Experimental

A microchannel reactor of 52 × 18 × 14 mm (Length × Width × Height) in size and 10 MC plates were made of high-temperature stainless steel 310S AISI. The microreactor design is schematically shown in Fig. 1. A milling machine with the computerized control (SUDA SD-3025, China) was used to form branched channels and eight straight channels

with the trapezoidal cross-section (40 mm long, 0.5 mm deep, 0.7 mm wide at the top, and 0.3 mm wide at the bottom) on each MC plate 40 × 15 × 1 mm in size (Fig. 2). All MC plates were placed into a stainless steel housing. On one side of the microchannel reactor, the inlet flow entered one channel and then split into ten MC plates. In each MC plate, the flow split again to enter eight straight channels. The successive splitting made the flow to enter 80 straight channels. On the other side of the microreactor, the inlet flow entered first into an empty cavity (1.3 cm³) and then into 80 straight channels. The temperature was controlled by three chromel–alumel thermocouples welded at the inlet, outlet and in the middle of the microchannel reactor housing.

A Pt(2 wt.)/Ni_{0.5}/MgAlO_x composition deposited on the MC plates was used as a catalyst. The catalyst coatings were prepared and anchored on the MC plate surface according to the following procedure. To prepare a stable suspension of aluminum oxide, we used mechanochemical activation of a γ-Al₂O₃ powder (Fluka, specific surface area 230 m²/g) for 6 min. The powder was activated with a high-power planetary centrifugal mill (AGO-2 activator, Russia) filled with aluminum balls, which provided a 60 g acceleration of milled particles. The milled γ-Al₂O₃ powder was used to prepare a 5 wt.% suspension in a 0.3 M nitric acid solution upon continuous stirring for 72 h. An aqueous suspension (5 wt.%) of pseudo-boehmite AlOOH (Disperal P2, SASOL, Germany) was prepared using the same procedure. The prepared suspensions were mixed at the volume ratio γ-Al₂O₃/AlOOH = 2:1, then crystalline hydrates (Ni(NO₃)₂·6H₂O and Mg(NO₃)₂·6H₂O) were added to the suspension in the amounts required to obtain the overall mixture composition Ni_{0.5}Mg₁Al₁(O). To improve the adhesion between the particles of the prepared suspension and the metal surface of the MC plate, a thin oxide layer was formed on the MC plate surface by its preliminary annealing in air at 1000 °C for 4 h. The prepared suspension mixture was stirred for 4 h and then layer-by-layer brushed on the overall plate surface to fill all microchannels, including the region of channel splitting. Each deposited layer was annealed in air at 450 °C for 30 min. This procedure was repeated 6 times. To form a NiMgAlO_x layer, the as-prepared MC plate was annealed in a furnace at 860 °C for 6 h. Platinum was deposited on this layer by the incipient wetness impregnation from an

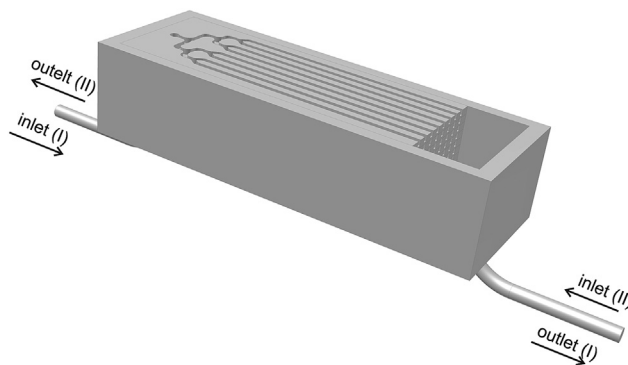


Fig. 1 – Configuration of the microchannel reactor 52 × 18 × 14 mm in size that contains 10 MC plates. Arrows show the direction of the flow for modes I and II.

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