Hydrogen production by methane partial oxidation at moderate temperatures in Rh-impregnated microchannel reactors

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Introduction

Partial oxidation of methane is one of several methods suggested for hydrogen production [1]. However, a significant research into its possibilities and limitations was not initiated until during the early 1990's, when the renowned studies by Ashcroft et al. [2,3] and Hickman and Schmidt [4] were published. Significant contributions to the field and the most studied topics and challenges were recently reviewed [5,6].

Partial oxidation of methane is not thermodynamically limited, and at higher temperatures (above 1000 K) syngas yields are typically high with excellent selectivity to hydrogen and CO. However, avoiding the thermodynamically more favourable combustion reaction is still a major challenge at lower temperatures.

Rh is one of the intensively studied metals in the partial oxidation of methane [5,6]. It is also known to be among the elements with most promising properties for both steam reforming [7] and dry reforming [8,9]. An important reason for why it has been much studied in the partial oxidation of methane, besides its excellent resistance to re-oxidation once reduced, is the fact that results have indicated that the dominant reaction pathway over Rh is the formation of CO [10]. Most other metals appear to proceed through the formation of CO_2, followed by subsequent reforming to syngas. However, this is a controversial issue, and earlier results [11,12] including a temporal-analysis-of-product (TAP) [13] indicated a combustion-reforming mechanism over Rh. The operating conditions could be a key parameter in explaining the suggested differences in mechanisms.

Materials and Methods

The Fecralloy (72.6% Fe, 22% Cr, 4.8% Al) reactors used in this study has previously been investigated in the catalytic conversion of propane to hydrogen [14,15,16], while the Nicrofer (3220H) reactor was entirely new. The partial oxidation of methane has previously been studied in our group using a Rh sponge catalyst [17] and Pt/Rh gauze catalysts [18,19].

Results and Discussion

The results include effects on conversion, selectivity and bed temperature profiles by varying: the partial pressures of methane and oxygen, oven temperatures and total gas hourly space velocity.

Significance

The use of microchannel or microstructured reactors in the partial oxidation of methane offer several possibilities and advantages over conventional packed bed reactors. Because the reactors are made of high temperature resistant metal alloys they offer excellent heat transfer properties, especially when compared to alumina supported catalysts. This contribute to reduce the formation of undesired hot-spots from the exothermic reactions. The compact design of microchannel reactors may be especially interesting for mobile small-scale or distributed hydrogen production, and the small channel dimensions (20-1000 μm) ensure safe operation with respect to explosive gas mixtures [20].

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