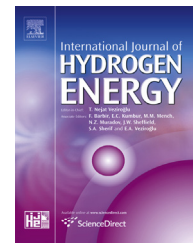




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Comparison between a micro reactor with multiple air inlets and a monolith reactor for oxidative steam reforming of diesel

Moa Z. Granlund ^{a,*}, Oliver Görke ^b, Peter Pfeifer ^b, Lars J. Pettersson ^a

^a KTH Royal Institute of Technology, Department of Chemical Engineering and Technology, SE-100 44 Stockholm, Sweden

^b Karlsruhe Institute of Technology, Institute for Micro Process Engineering (IMVT), D-76344 Eggenstein-Leopoldshafen, Germany

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ABSTRACT

In order to lower the emission from idling heavy-duty trucks auxiliary power units can be implemented. Due to limited space available on-board the truck the units needs to be both efficient and compact. One alternative for these units is a fuel cell supplied with hydrogen from a fuel reformer. Today, mostly monolithic reactors are used in the field of oxidative steam reforming of fuels, which has some challenges that need to be addressed before a possible breakthrough occurs on the market. One is the temperature gradient developed over the length of the monolith as a consequence of the sequential reactions. This could be improved by using a metallic micro reactor with better heat integration between the reaction zones and further improving the integration with multiple air inlets along the catalytic bed.

The aim with this study was to compare a conventional monolith reactor for oxidative steam reforming of fuel with a novel micro reactor design where air was dosed at four different positions along the reactor channels. The experiments were not necessarily conducted autothermal, i.e. a heating jacket was applied for operation.

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Introduction

It is estimated that heavy-duty vehicles are responsible for about 30% of the total emissions of nitrogen oxides (NO_x) and particulate matters (PM) [1]. In addition the engine is idling 30–40% of the time just to supply the vehicle with electricity [1,2]. The emission during idling is a considerable part of the total emission for the vehicle and there is an urgent need to

decrease those in order to meet future emission legislations [3]. One alternative to solve this could be to implement a fuel cell system where the hydrogen is produced via on-board reforming of diesel, a diesel fuel cell auxiliary power unit (FC APU) [4].

There are three different processes for reforming diesel to hydrogen-rich gas: partial oxidation, steam reforming and oxidative steam reforming (OSR); the latter could be operated self-sustaining as autothermal reforming (ATR). The three

* Corresponding author. Tel.: +46 8 790 91 50.

E-mail address: moazg@kth.se (M.Z. Granlund).
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processes have different advantages and disadvantages and for on-board fuel reforming OSR is the most suitable option [5]. The main reason is that the process is autothermal and therefore no external heating is needed. The autothermal characteristic is a consequence of the process being a combination of the exothermic partial oxidation and endothermic steam reforming. Other advantages are the short start-up time due to the fast thermal response and the relatively high hydrogen yield [6].

Due to the limitations of space onboard heavy-duty vehicles the design of the FC APU must be compact. An OSR reactor is normally based on a tubular reactor where the catalytic material is coated on a honeycomb cordierite monolith. This minimizes the pressure drop and also makes the reformer resistant to vibrations [7]. One of the most prominent disadvantages with this reactor design is the characteristic temperature gradient, developing along the monolith as a consequence of the combination of the two sequential reactions. The fast and exothermic partial oxidation reaction takes place at the entrance, causing a sharp increase in temperature and the slower, endothermic steam reforming reaction consumes the heat towards the outlet of reactor [8]. The heat transport from the initial partial oxidation reaction is transferred along the monolith only by convection and the interaction between the two reaction zones is poor. The initial raise in temperature can cause thermal deactivation of the catalytic material by the potential formation of hot spots. In order to limit the temperature increase in the beginning, better heat integration between the two reaction zones is necessary.

One option to decrease the temperature gradient over the reactor and to increase the heat and mass transfer could be to use a micro reactor with better heat integration [9,10]. With a micro reactor it is possible to make a design closer connecting the exothermic and the endothermic part of the reactor. Another advantage with micro reactors is that metal is used which increases the heat conductivity of the reactor structure compared to ceramic monoliths [11].

One completely different possibility, already successfully tested at the Institute for Micro Process Engineering at Karlsruhe Institute of Technology, is to indirectly couple endothermic steam reforming with the exothermic combustion of anode-off gas in two different foil arrangements inside a metallic micro reactor [12].

In the current publication, however, the attempt was to further improve the heat integration between the two reaction zones in a direct coupling. Therefore, a micro reactor with multiple air dosing points axially distributed along the reaction channels was designed. The idea is to further improve the heat management by dosing the air axially along the reactor channels by adapting the kinetics between the partial oxidation and steam reforming reactions. With a better heat integration between the partial oxidation and steam reforming parts of the catalyst the temperature stress that usually occurs in the beginning of a monolith would be decreased. The integration could potentially also lead to higher temperatures in the steam reforming part, increasing the rate of the steam reforming reaction with a higher H_2 yield as result.

The scope of our work is to evaluate if micro structured reactors are a viable alternative to monolith reactors for on-

board production of hydrogen. The deactivation of the catalytic material caused by thermal aging, i.e. sintering and collapsing of the pore structure, might therefore be decreased as previously shown in Ref. [11].

Experimental

Catalyst preparation

The catalytic material used in the comparative study between the two different reactor designs was 3 wt.% Rh supported on high surface area CeO_2-ZrO_2 (MEL Chemicals, 16.5% CeO_2/ZrO_2) prepared by the incipient wetness method. As metal precursor, Rh nitrate solution was used ($Rh(NO_3)_3$ solution, 9.53 w/w, Sigma–Aldrich). After the impregnation the catalytic material was calcined in air at 800 °C for 3 h. Subsequently, the catalytic material was suspended in ethanol and ball milled for 24 h.

For the monolith reactor the catalytic material was applied to a cordierite monolith (400 cps) by dip coating until the washcoat loading was approximately 20 wt.%. After the coating the monolith was calcined in air at 800 °C for 3 h. The inserted catalyst mass was 1.15 g. The diameter and length of the monolith was 20 mm and 30 mm, respectively, resulting in an inner reaction volume of approximately 5.35 cm³ [13].

Before coating the micro structured reactor, coating tests with single metallic micro structured foils were conducted. Prior to coating the Nicrofer[®] foils were calcined at 800 °C for 3 h in air to generate a thin oxide film on the surface. Subsequently the foils were coated dropwise with the catalyst slurry. The coated foils were then investigated with scanning electron microscope and microprobe analysis. The obtained coating showed an excellent adhesion on the micro channel foils and the catalytic material was homogeneously distributed on the foils, which can be seen in Fig. 1.

For the micro reactor with multiple air inlets the micro structured foil stack was welded by diffusion bonding and air inlets were attached to the flow chambers by tungsten inert-gas welding. The stack was calcined at 800 °C for 3 h and afterwards flow-coated with the same slurry as it was used for dipcoating of the single micro structured foils. After drying and calcination of the catalyst for 3 h at 800 °C in air, the flow chambers were welded onto the coated stack on the reaction side by tungsten inert gas welding. The inserted catalyst mass was also 1.15 g.

The fuel used in the study was synthetic diesel of type biomass-to-liquid (NExBTL, Neste Oil). The main advantage with using synthetic diesel is its high content of alkanes, implying low content of aromatic and polyaromatic hydrocarbons that are prone to coke and lead to catalyst deactivation. Another benefit with synthetic diesel is its low sulfur content [14]. The characteristics of the synthetic diesel are summarized in Table 1.

Reactor set-up

The OSR experiments were performed in a bench-scale setup where either the micro reactor or the monolith reactor was

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