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Simulation of exhaust gas reforming of natural gas in a microchannel reactor



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ABSTRACT

The present work is aimed to conduct modeling and simulation of exhaust gas reforming of natural gas in a catalytic microchannel reactor. Natural gas, an alternative fuel that can be used in internal combustion engines in compressed form, is modeled as methane only and methane/propane mixtures. The multichannel reactor is composed of a cordierite block with parallel channels, each of which is washcoated with Rh/Al₂O₃ catalyst. Due to the low thermal conductivity of cordierite, heat transfer between the channels is neglected, and a single, adiabatic microchannel is considered as the modeling domain representing the behavior of the multichannel unit. Two dimensional continuity and conservation equations for the fluid and porous washcoat phases are solved by the finite volume method using the ANSYS Fluent platform. Effects of feed temperature, fuel compositions (i.e. molar inlet steam-to-carbon (H₂O/C) and oxygen-to-carbon (O₂/C) ratios) and presence of propane in natural gas on temperature and product distribution are investigated in the context of a parametric study. It is observed that temperature is well distributed along the channel and no notable hot spot formation is observed. Increasing feed temperature favors methane conversion and hydrogen production, but results in less uniform temperature distribution. Feeding higher amounts of steam increases hydrogen formation, but slightly dampens methane conversion. Increasing O₂/C ratio at the inlet results in a proportional increase in methane conversion and temperature. Even though channel temperature is found to decrease, hydrogen production is favored upon using methane/propane mixture as the fuel.

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Introduction

Exhaust gas reforming (EGR) is a fuel conversion technique that offers the potential of increasing the efficiency and reducing the emissions of conventional internal combustion engines (ICEs) [1]. In EGR, a catalytic reactor, which is integrated into the exhaust gas recirculation loop, utilizes the heat, steam, CO₂, O₂, and traces of CO and some unburnt

hydrocarbons that are present in the exhaust gas together with externally injected fuel and steam to produce a H₂-rich reformate [1]. Temperature and composition of the exhaust gas stream depend strongly on the type of fuel processed as well as the relative amounts of fuel and air processed in the ICE. The resulting reformate is then sent back to the ICE for increasing the efficiency of the engine and reducing hazardous emissions. Another benefit of replacing a small fraction of hydrocarbon

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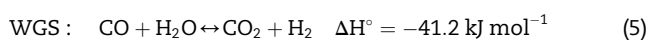
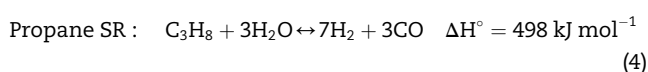
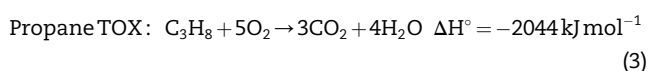
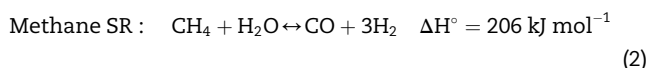
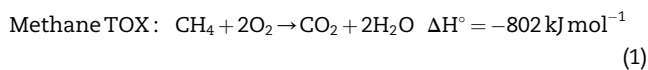
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fuel with H₂ is that it acts as a combustion promoter by supporting the autoignition of the fuel [2].

EGR is based on the oxidative steam reforming (OSR) process, which combines total oxidation (TOX) and steam reforming (SR) of the fuel in the same reactor volume [3]. This combination reduces the external energy demand, hence increases the energy efficiency of the H₂ production. When a combination of methane and propane is used as the fuel, the reactions involved in the system can be written as follows:



TOX is a highly exothermic and fast reaction, which is known to be capable of starting up even at low temperatures [4]. SR, on the other hand, is endothermic and requires high amounts of heat supply from the surroundings [4–7]. Heat of the exhaust gas alone is not sufficient for the energy requirement of SR, but enough heat can be raised when a fraction of the fuel is used by TOX [5]. Thus, heat dissipation along the catalyst bed, which is function of reactor geometry, is very important in OSR.

Microchannel reactors are small, compact reactors made up of parallel, catalyst coated channels, the diameters of which vary between 10⁻⁶ and 10⁻³ m [8]. Due to the small channel diameters, microchannel reactors have very high surface area/volume ratios (up to ca. 5 × 10⁴ m² m⁻³), which result in high heat exchange surfaces and heat transfer coefficients. Therefore, compared to those of conventional reactors, microchannel units offer very high heat transfer rates which become crucial in the presence of highly exothermic and endothermic reactions [8]. In the case of OSR, it is important that heat produced by oxidation is distributed throughout the reactor without forming a hot-spot, since high temperatures can lead to the deactivation of the catalyst [9]. For an efficient EGR process, the heat of the exhaust gas and the heat raised by TOX should be transferred to the endothermic SR to increase the rate of reforming and to produce higher amounts of H₂.

EGR of various fuels such as propane (a model hydrocarbon for gasoline) [2], diesel and biodiesel [3,10–12] is investigated in the literature. These studies aimed to explore the product distribution in EGR of various exhaust gas streams obtained after combustion of the pertinent fuels in a relevant engine type at different fuel-air mixtures, and involved the use of supported precious metal based catalysts such as Rh in packed-bed type reactors for reforming the exhaust-gas into hydrogen-rich mixtures. In these studies, however, the effect

of reactor type on heat transfer and temperature distribution over the catalyst bed is not addressed. Comparison packed-bed and monolith reactors in the context of diesel fuel are reported in [13], and the latter, which has a geometry similar to that of a microchannel reactor, is found to lead to a more uniform evolution of reactor temperature. Sen and Avci [14] studied the use of microchannel reactor configuration in EGR. The work investigated the effects of different combinations of Pt and Rh based catalysts and of the operating conditions on methane reforming. It is concluded that methane conversions increased with feed temperature, H₂O/C and O₂/C ratios. The catalyst combination with Rh coated on both walls is found to exhibit the best performance in methane conversion [14]. Oxidation of methane in exhaust gas diluted reaction mixtures over Al₂O₃, ZrO₂ and CeO₂ supported Rh-based catalysts is studied by Eriksson and co-workers [15]. It is stated that while increasing Rh dispersion improved methane conversion with all supports, Rh/Al₂O₃ exhibited the best performance with higher synthesis gas yields and lower surface temperatures. Simsek and his co-workers [16] compared the performances of coated and packed microchannel reactors on methane OSR. The authors concluded that, in both reactor configurations, methane conversion increased with temperature, H₂O/C and O₂/C ratios, but CO selectivity is found to be significantly higher in the coated microchannel. In another study, propane EGR in a microchannel reactor involving combined use of Pt- and Rh-coated catalysts in the same volume is modeled with the aim of understanding the nature of heat exchange between TOX and SR reactions at the micro scale [17]. It is observed that addition of propane and H₂O to the feed increased syngas yield and increasing the feed flow rate improved heat distribution but decreased H₂ yield.

The objective of this study is to simulate EGR of natural gas in a Rh-coated microchannel reactor for providing insight into the effects of operating conditions and reactor geometry on temperature and product distributions. Natural gas is represented by methane only and by mixtures of methane and propane at certain ratios. Even though the main constituent of natural gas is methane, propane is one of the components of natural gas and is expected to affect heat utilization in EGR via Reactions 3 and 4, both of which have enthalpies much higher than those of methane TOX and SR (i.e. Reactions 1 and 2, respectively). To our knowledge, simulation of a binary fuel mixture in EGR is not investigated in the literature. The approach followed during modeling of the microchannel reactor, model equations and solution strategy is explained in ‘System description and mathematical modeling’ section. The effects of feed temperature, reactant composition (i.e. H₂O/C and O₂/C ratios) and presence of propane in natural gas on temperature and product distributions are reported and discussed in ‘Results and discussions’ section.

System description and mathematical modeling

Assumptions and working equations

The microchannel reactor to be simulated involves parallel, square-shaped channels, each of which having a height (H) and a length (L) of 7.5 × 10⁻⁴ m and 2 × 10⁻² m, respectively

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