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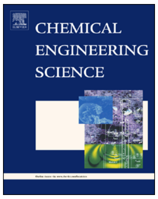

An experimental and numerical investigation of the combustion and heat transfer characteristics of hydrogen-fueled catalytic microreactors

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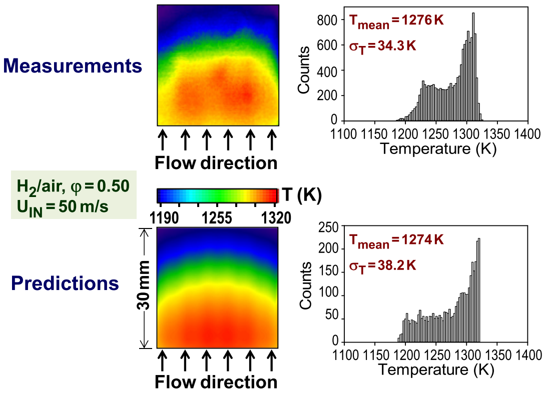
HIGHLIGHTS

- Optimization of a six-channel H_2-fueled catalytic microreactor with 3D simulations.
- Counterflow configuration superior to coflow in spatial uniformity of temperatures.
- Simulations reproduce measured higher moments of surface temperature distributions.
- Maximum surface temperatures up to 1311 K and standard deviations up to 18.6 K.
- Radiation efficiencies up to 76%, suitable for microreactor coupling with TPVs.

GRAPHICAL ABSTRACT



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ABSTRACT

The combustion and heat transfer characteristics of a hydrogen-fueled microreactor are investigated experimentally and numerically. The microreactor comprises a $30 \times 30 \times 4\ mm^3$ SiC-block equipped with six 1.5 mm diameter platinum channels. Combustion of fuel-lean H_2/air mixtures at equivalence ratios $\varphi = 0.25 - 0.50$ and inlet velocities 15–50 m/s is studied at coflow and counterflow configurations. Surface temperatures are measured with an infrared camera, while simulations are carried out with a 3D code that includes conjugate heat transfer, appropriate external heat losses, and detailed hetero-/homogeneous chemistry. Higher mass throughputs reduce the surface temperature spatial non-uniformities, while the onset of gaseous combustion lowers the catalyst surface temperatures and is thus detrimental for power generation applications. Four different channel configurations are tested for optimum temperature uniformity. Counterflow configurations are shown superior to the coflow configuration in attaining better surface temperature uniformities with standard deviations less than 19 K and maximum surface temperatures up to 1311 K. Comparisons of measurements and predictions are very favorable in terms of temperature probability density function (PDF) shapes and higher distribution moments. Counterflow configurations yield narrower PDFs slightly skewed to the low temperatures, while the coflow configuration yields mostly bimodal shapes. Radiation efficiencies increase with increasing inlet

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velocity and equivalence ratio. Application of the microreactor to power generation systems, in conjunction with thermoelectric devices, appears quite promising given the attained good spatial uniformity and the high values of surface temperatures.

1. Introduction

Hydrogen and hydrogen-containing fuels are under intense investigation for small- and large-scale power generation. Microreactors for small ($\sim$100 W_e) portable power generation devices fueled with hydrogen (Ghermay et al., 2010; Michelon et al., 2015; Norton et al., 2004; Seyed-Reihani and Jackson, 2004) or hydrogen-enriched hydrocarbons and syngas mixtures (Federici and Vlachos, 2011; Karagiannidis and Mantzaras, 2012; Seshadri and Kaisare, 2010) have been studied experimentally and numerically in the last years. In such systems hydrogen can be produced on-board from methane (Diehm and Deutschmann, 2014; Jiang et al., 2015; Kaisare et al., 2005; Stefanidis and Vlachos, 2009; Stefanidis et al., 2009) or high hydrocarbons (Casanovas et al., 2008; Donazzi et al., 2014; Eriksson et al., 2006; Hartmann et al., 2010; Holladay et al., 2004) using suitable microreformers. Most microreactors operate with heterogeneous (catalytic) combustion or with a variety of hybrid concepts, i.e. combined heterogeneous and homogeneous (gaseous) combustion (Schultze and Mantzaras, 2013), rather than with pure gaseous combustion. This is mainly dictated by the large surface-to-volume ratios of microreactors that result in much wider catalytic combustion stability envelopes compared to those of pure gaseous combustion (Ahn et al., 2005), the existence of a multitude of undesirable flame instabilities in tight geometrical confinements (Evans and Kyritsis, 2009; Fan et al., 2013; Kurdyumov et al., 2009; Pizza et al., 2010a) and the efficient suppression of such instabilities by coating the microreactor walls with a catalyst (Pizza et al., 2010b; Pizza et al., 2009).

Although hybrid reactor designs have distinct heterogeneous and homogeneous combustion zones with the former preceding the latter, gas-phase combustion cannot always be neglected inside the designated catalytic combustion zone. Even at the large geometrical confinements of practical catalytic microreactors (e.g. catalytic channels with sub-millimeter hydraulic diameters) gas-phase combustion can be appreciable for hydrocarbon fuels, depending on the pressure, temperature and residence time (Karagiannidis et al., 2011; Reinke et al., 2002). A detailed parametric study of fuel-lean H_2/air combustion in platinum-coated channels (tubular or planar) has delineated (Ghermay et al., 2011) the regimes of wall temperatures, inlet temperatures, pressures, and channel hydraulic diameters for which gaseous combustion amounts to at least 5% of the combined catalytic and gas-phase hydrogen conversion. Catalytic combustion of fuel-lean H_2/air mixtures is particularly challenging due to the diffusional imbalance of the deficient hydrogen reactant (Lewis number of hydrogen $Le_{H2}\sim0.3$), which leads to superadiabatic surface temperatures (Bui et al., 1996; Mantzaras, 2014) that endanger the catalyst and reactor integrity. To mitigate such superadiabatic effects, an inverse hybrid concept for hydrogen has been recently proposed (Ghermay et al., 2010), whereby the gaseous combustion zone precedes the catalytic combustion zone.

Apart from microreactor applications, hybrid hetero-/homogeneous combustion of hydrogen is also of prime interest for large-scale power generation. One such approach is the catalytically stabilized thermal combustion (CST) (Carroni and Griffin, 2010), where part of the fuel is converted in a catalytic reactor and the remaining is combusted in a subsequent gas-phase burner. The CST hybrid methodology mitigates flashback by hindering upstream flame propagation inside the catalytic module due to the inhibiting effect of heterogeneous reactions on homogeneous combustion (Mantzaras and Appel, 2002; Mantzaras and Benz, 1999). On the other side, post-combustion CO_2 capture techniques currently apply large flue gas recycle (FGR) in order to increase the CO_2 content in the exhaust and thus facilitate its subsequent capture (Schneider et al., 2007; Tan et al., 2006). For the post-combustion CO_2 capture methods, inclusion of an upstream catalytic reactor enhances the combustion stability of the less-reactive FGR-diluted fuel mixtures.

There is nowadays increased interest in developing renewable energy sources for satisfying rising electricity demands. An ideal power source should provide reliable and continuous base-load power as well as peak-load power when needed to match supply demand. This requires a high Annual Capacity Factor (ACF), which is the ratio of the source's realized output in one year to its potential output when operated at full capacity over the same period. Within the European Union project Hybrid Renewable energy Converter for continuous and flexible power production (HRC-Power, 2015) a combined solar/combustor microreactor is pursued, capable of operating either in sole combustion mode, sole solar mode, or mixed combustion/solar mode, with a targeted 95% ACF. Fig. 1 illustrates the concept, which comprises a central microreactor block (2) made from a high thermal conductivity material. Under solar operation alone, the top microreactor surface (1) is coated with a special absorbing layer to efficiently collect concentrated solar radiation, while the bottom surface is coated with a selectively emitting infrared radiation layer (e.g. to be coupled with a thermophotovoltaic (TPV) module). In the absence of solar radiation or in the case of intermittent solar radiation, combustion inside the microreactor supplies all or part of the thermal energy necessary to heat the selectively IR-emitting bottom surface. A number of such microreactors can eventually be

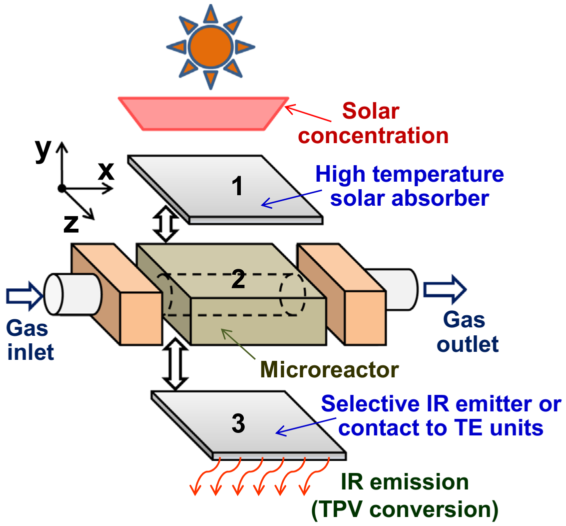

Fig. 1. Schematic of the solar/combustion reactor: (1) solar-energy-absorbing top surface, (2) microreactor block, and (3) IR-emitting bottom surface.