

IR thermal imaging characterization of a hybrid catalytic radiant gas burner

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Abstract

The novel concept of hybrid catalytic combustion based on Catalytic Partial Oxidation + homogeneous flame combustion was tested for the first time with fuel mixtures of methane and hydrogen. A prototype radiant hybrid burner was characterized by IR thermal imaging while safely operating with up to 80% vol. of H₂ in the fuel and a primary equivalence ratio in the range 2.4 – 4.0. Outstanding NO_x emission levels were attained due to the effective reduction of both thermal and prompt NO_x formation.

Introduction

An increasing concern regarding environmental pollution during the last years has resulted in stricter emission regulations for NO_x, CO and greenhouse gases from combustion processes. The catalytic partial oxidation (CPO) of various fuels has been proposed as a preliminary conversion stage in hybrid burners for gas turbines with ultra low-NO_x emissions [1]. Recently the concept has been extended to develop a novel class of hybrid catalytic gas burners, with integrated interstage heat removal by IR radiation from the hot structured catalytic partial oxidation reactor/radiator [2,3], showing significant improvements with respect to the state-of-the-art fully premixed or blue-flame diffusive natural gas burners for domestic condensing boilers [2]. Since the air-fuel mixture fed to the burner is generally above its upper flammability limit, this technology is intrinsically safe and can be regarded as a candidate also for hydrogen-hydrocarbon blends. Structured CPO catalysts based on noble metals supported onto ceramic and metallic honeycombs, foams or gauzes have shown high syn-gas yields at very short contact times with hydrocarbon feeds from methane up to diesel and jet-fuels, alcohols and bioderived liquids [1-5]. In this work we set out to demonstrate the intrinsically safe multi-fuel operability of the novel hybrid catalytic burner and its low NO_x emissions when fuelled with H₂-CH₄ mixtures of variable compositions.

Experimental

Bimetallic Rh-Pt catalysts dispersed on commercial La- γ -Al₂O₃ (140m²/g) were prepared via sequential impregnations using aqueous solutions of Rh (NO₃)₃ and H₂PtCl₆ to achieve a total PM loading of 0.5+0.5 %wt [6]. The catalytic layer was

anchored by a dip-coating procedure [6] over a metallic gauze made of a FeCrAlloy knitted wire ($d \approx 120 \mu\text{m}$) in the form of flexible cylindrical sockets ($D=H=80\text{mm}$, thickness 2mm, apparent density 0.38 g/cm^2 , Figure 1). The hybrid catalytic burner under typical operation together with a rendering of its longitudinal section are presented in Fig. 1: the fresh gas mixture flowed radially through a perforated hollow cylindrical distributor, the CPO gauze reactor and then into the flame [2]. The hybrid burner was tested in upward position with secondary air for the purely diffusive flame combustion withdrawn by natural convection. The inlet ϕ was varied in the range 2.4 – 4.0 (i.e. above upper flammability limit). The H_2 content in the fuel mix was varied between 0 to 80% vol., at constant nominal power (6.6, 9.6, 19.2 kW). CO , NO_x and CH_4 emissions were corrected to 0% O_2 . A FLIR Phoenix infrared camera was set-up as a broadband sensor sensitivity in the range $1.5\text{-}5 \mu\text{m}$ to characterize the operation of the hybrid burner, obtaining simultaneously the temperature profile on the outer catalytic surface and an estimation of the flame temperature and structure. In particular, two optical spectral filters centred at $3.9 \mu\text{m}$ and at $4.25 \mu\text{m}$ were applied respectively in order to eliminate flame (gas) emissivity when measuring the surface temperature of the catalyst, and to estimate CO_2 (flame) temperature [7]. Moreover, the flame images filtered at $3.9 \mu\text{m}$ allowed us to follow the eventual presence of soot formation. Independent thermocouple measurements were preliminary performed to set appropriate emissivity factors [6]. All the IR pictures presented hereafter represent a 5s-average of the images collected at 340 frames/s. The temperature of the syn-gas emerging from the catalytic stage was also measured with a shielded (Inconel 600, $d=0.5\text{mm}$) K-type thermocouple placed 1 mm downstream from the catalyst.

Results and Discussion

In the hybrid burner, the catalytic stage prepares a hot syn-gas which is the actual fuel for the following homogeneous (flame) combustion stage. During the CPO of pure methane over structured catalysts based on Rh and Pt, O_2 is rapidly depleted within a short front section of the catalyst bed and a significant portion of fuel is consumed to produce a mixture of H_2 , CO and H_2O [3,8]. Due to the limited oxygen availability in the feed, the catalytic performances of the CPO reactor are controlled by the inlet equivalence ratio ϕ . The partial substitution of CH_4 with H_2 at each fixed value of ϕ caused an increase of the temperature of the catalyst and of the product gas, in agreement with the trends of the adiabatic equilibrium temperature. CH_4 conversion also increased progressively, because of higher temperatures on the catalyst and higher partial pressures of water, that boosted steam reforming reaction [6,8]. It can be argued that H_2 is rapidly and oxidized instead of the hydrocarbon in the first part of the reactor due to its higher reactivity and diffusivity, causing an increase of catalyst temperature in this zone. Further downstream, H_2 is produced again by catalytic CH_4 steam reforming.

IR analysis of the hybrid burner (Fig.2) showed the presence of an axial-symmetric laminar diffusive flame structure surrounding the hot radiating catalytic surface.

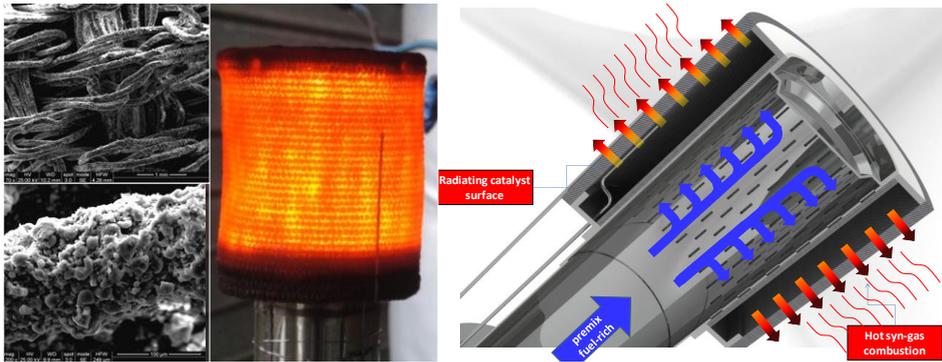


Figure 1. SEM images at increasing magnifications of the FeCrAlloy knitted gauze substrate with active washcoat layer (left); catalytic combustion head ($D \times L = 80 \times 80$ mm) during operation (centre), and rendering of its section (right).

The reactor presented a very uniform temperature distribution on the whole outer surface. The flame itself, which appeared blue in the visible range had a low-luminosity (typical of a premixed flame). No soot formation was found by Planar Laser Induced Incandescence, and this was confirmed by careful inspection of IR images collected at $F = 3.9 \mu\text{m}$ in the gas phase surrounding the burner (Fig.2 b).

Hot gases leaving the combustion head through the catalytic gauze were slightly colder than the catalyst itself. This was also confirmed by the thermocouple measurements [2] and implies that, inside the structured catalytic layer, heat generation occurs through surface oxidation reactions and the flowing gases are only heated by convection. Moving away from the catalytic surface along the radial coordinate we initially observed a flat temperature profile in the gas phase due to the absence of oxidation reactions close to the head: all of the oxygen from primary air was consumed by catalytic oxidation reactions (see next section), and more O_2 from the surrounding air had to diffuse to complete combustion. Thereafter, the gas temperature raised to a maximum in correspondence of a symmetric laminar diffusion flame front developed around the burner and above it, which was well captured by filtered profiles ($F = 4.25 \mu\text{m}$). Temperature profiles in the gas phase were obtained by measuring the main emission peak of CO_2 at $\lambda = 4.25 \mu\text{m}$. Those measurements are only qualitative due to the variation of the CO_2 concentration, the spatial integration of the emission, the flame thickness, the presence of an emitting/adsorbing/reflecting surface behind the flame [3,7]. However, this qualitative measurement is in a good agreement with temperature measurements operated by thermocouple.

It is clear that the flame front extended well above the catalytic head, due to the relatively slow diffusion of oxygen from the surroundings and to poor mixing effectiveness with secondary air achieved by natural convection.

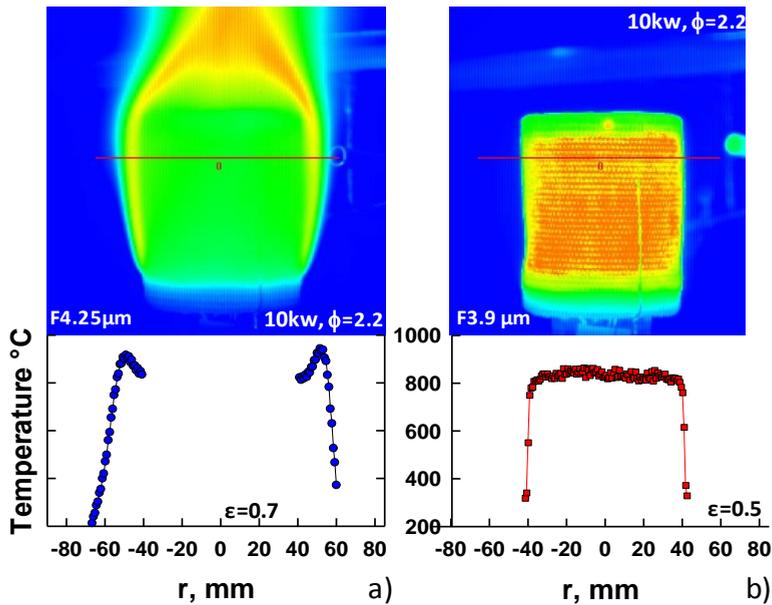


Figure 2. InfraRed images of flame structure (CO_2) and catalytic surface collected at $4.25\mu\text{m}$ (a) and $3.9\mu\text{m}$ (b) respectively, together with the temperature profiles at fixed height along the line shown. Feed $\phi=2.2$, $P=9.6\text{kW}$, pure CH_4 .

Figure 3 shows that the flame structure and appearance did not change significantly by increasing the H_2 content of the fuel, while operating the burner at constant input power and equivalence ratio. Nevertheless, the temperature of the outer surface of the catalytic gauze increased progressively for lower values of ϕ and for higher H_2 contents in the fuel fed to the hybrid burner (Fig. 3 and 4). The thermal resistance of the catalyst (i.e. its maximum allowable surface temperature) set the minimum permissible value ϕ , which in turn depended on how much heat was removed from the radiating catalytic head [2,3].

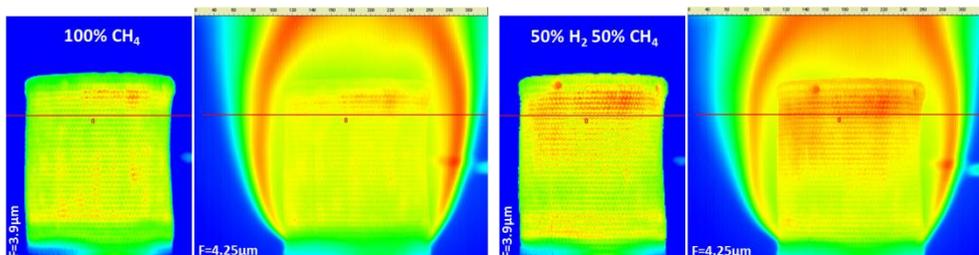


Figure 3. InfraRed images of catalytic surface and flame structure (CO_2) filtered at $3.9\mu\text{m}$ and $4.25\mu\text{m}$ respectively, during operation at $\phi=3.6$ and $P=19.2\text{kW}$ with pure CH_4 (left) and with a $50\% \text{H}_2 - 50\% \text{CH}_4$ mix (right). Surface temperature estimated with a solid emissivity $\epsilon=0.5$ at $3.9\mu\text{m}$.

Figure 4 demonstrates that fuel blends very rich in H_2 were safely handled with the hybrid catalytic burner by increasing the feed equivalence ratio, in order to compensate the increase in the temperature level of the catalyst. At the same time larger values ϕ effectively contrasted the tendency of the highly reactive fuel to give a flashback in the mixing zone upstream of the catalyst, and indeed this was never observed even at the highest hydrogen contents. Gas analysis at the exit of the catalytic partial oxidation stage invariably showed no trace of nitrogen oxides. Thus, NO_x formation occurred only in the homogeneous flame combustion stage: in the simple current configuration of the hybrid burner, syn-gases emerging from the catalytic gauze were burned under purely diffusive and laminar conditions.

In general it was found that the presence of up to 50% H_2 by volume in the fuel mixture did not alter the NO emission performance of the hybrid burner with respect to operation with pure CH_4 . A slight increase in the total NO formation was observed for higher values of the H_2 content in the fuel, probably related to the corresponding increase in the peak flame temperature achieved in the secondary flame. It should be remarked that the prompt NO_x formation mechanism in the syn-gas diffusion flame was progressively suppressed with increasing yH_2 in the fuel by two simultaneous factors: i) the decreasing concentration of (un-reacted) methane and CO in the syn-gas; ii) its larger dilution by H_2O [9,10]. Combustion tests run at different power levels for a fixed value of ϕ revealed that NO emissions increased slightly when the burner was operated at higher power, according to a proportionally lower fraction of the total heat of combustion which was transferred from the radiating element [2,3]. At the same time it was confirmed that up to 50% vol. of H_2 in the fuel had only a minor impact on the formation of nitrogen oxides. The high preheating of the syn-gas fuel, as well as its large H_2 content, facilitated the oxidation of other organic fuel fractions (CO and CH_4) even in a secondary flame with poor mixing judged by the standard of conventional design [10].

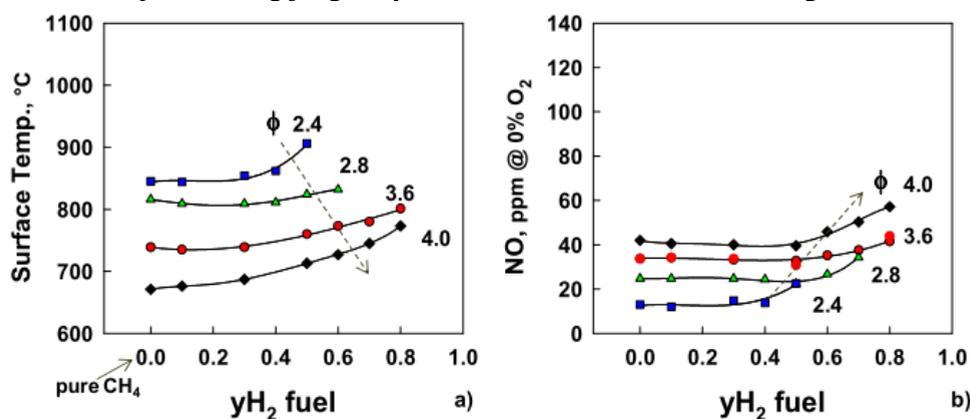


Figure 4. a) Maximum temperature of the outer catalytic surface and b) NO emissions in the exhaust from the hybrid burner as a function of H_2 content in the CH_4 - H_2 fuel mixture for several levels of ϕ at fixed input power (9.6 kW).

Conclusions

A prototype radiant hybrid burner was safely operated at atmospheric pressure with up to 80% by volume of H₂ in the fuel and a primary equivalence ratio ϕ in the range 2.4 – 4.0. Outstandingly low-NO_x emission levels were attained with a purely diffusive laminar flame combustion stage, due to an effective suppression of both thermal and prompt NO_x formation mechanisms. In particular it was found that the presence of up to 50% H₂ in the fuel mixture did not alter the NO_x emissions of the hybrid burner with respect to operation with pure CH₄, according to the small variations measured in the composition of the syn-gas burned in the secondary flame and to the slight increase in the temperature of the radiating catalytic surface. The rather large values of the feed equivalence ratio to the burner prevented flashback in the mixing zone upstream of the catalyst even at the highest hydrogen contents in the fuel.

References

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