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Homogeneous ignition of CH₄/air and H₂O and CO₂-diluted CH₄/O₂ mixtures over Pt; an experimental and numerical investigation at pressures up to 16 bar

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Abstract

The homogeneous ignition of CH₄/air, CH₄/O₂/H₂O/N₂, and CH₄/O₂/CO₂/N₂ mixtures over platinum was investigated experimentally and numerically at pressures $4 \text{ bar} \leq p \leq 16 \text{ bar}$, temperatures $1120 \text{ K} \leq T \leq 1420 \text{ K}$, and fuel-to-oxygen equivalence ratios $0.30 \leq \varphi \leq 0.40$. Experiments have been performed in an optically accessible catalytic channel-flow reactor and included planar laser induced fluorescence (LIF) of the OH radical for the determination of homogeneous (gas-phase) ignition and one-dimensional Raman measurements of major species concentrations across the reactor boundary layer for the assessment of the heterogeneous (catalytic) processes preceding homogeneous ignition. Numerical predictions were carried out with a 2D elliptic CFD code that included elementary heterogeneous and homogeneous chemical reaction schemes and detailed transport. The employed heterogeneous reaction scheme accurately captured the catalytic methane conversion upstream of the gaseous combustion zone. Two well-known gas-phase reaction mechanisms were tested for their capacity to reproduce measured homogeneous ignition characteristics. There were substantial differences in the performance of the two schemes, which were ascribed to their ability to correctly capture the p - T - φ parameter range of the self-inhibited ignition behavior of methane. Comparisons between measured and predicted homogeneous ignition distances have led to the validation of a gaseous reaction scheme at $6 \text{ bar} \leq p \leq 16 \text{ bar}$, a pressure range of particular interest to gas-turbine catalytically stabilized combustion (CST) applications. The presence of heterogeneously produced water chemically promoted the onset of homogeneous ignition. Experiments and predictions with CH₄/O₂/H₂O/N₂ mixtures containing 57% per volume H₂O have shown that the validated gaseous scheme was able to capture the chemical impact of water in the induction zone. Experiments with CO₂ addition (30% per volume) were in good agreement with the numerical simulations and have indicated that CO₂ had only a minor chemical impact on homogeneous ignition.

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Keywords: High-pressure homogeneous ignition of CH₄/air over Pt; Effect of H₂O and CO₂ on ignition; LIF and Raman

1. Introduction

The application of catalytically stabilized combustion (CST) to large-scale gas turbines has been actively pursued over the last years [1] as a means

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to mitigate NO_x emissions with direct combustion strategies rather than with indirect exhaust-gas aftertreatment techniques. The knowledge of the heterogeneous (catalytic) and of the low-temperature homogeneous (gas-phase) kinetics of methane is key to the development of advanced numerical CST models [2,3]. Validation of the heterogeneous reaction scheme of Deutschmann et al. [3] for the complete oxidation of CH_4 over Pt at pressures up to 16 bar was recently reported in Reinke et al. [4]; the catalytic reactivity was assessed with in situ Raman measurements of major species concentrations over a channel-flow boundary layer. The onset of homogeneous ignition is detrimental to the catalyst integrity and, therefore, the availability of validated gaseous reaction schemes is crucial in CST reactor design; such schemes can also be used to fine-tune analytical CST homogeneous ignition criteria [5,6], so as to provide a fast—albeit more restrictive—alternative to detailed computations. To this direction, gaseous reaction schemes in CST have been validated only at low-to-moderate pressures. Reinke et al. [7] have shown, encompassing earlier atmospheric-pressure studies [8], the applicability of the gaseous scheme of Warnatz and Maas [9] in CST of CH_4/air over Pt at pressures up to 6 bar, a range of interest to microreactors.

The present study undertakes a combined experimental and numerical investigation of CH_4/air CST over Pt, with the main objective of providing validated homogeneous reaction schemes at gas-turbine-relevant conditions. Particular objectives were to assess the CST applicability of gaseous schemes in the presence of large H_2O or CO_2 dilution (an issue of interest in gas-turbines with exhaust gas recycle [10]), and to study the hetero/homogeneous chemistry coupling at high pressures. Experiments were performed in an optically accessible catalytic laminar channel-flow reactor at pressures $4 \leq p \leq 16$ bar. The onset of homogeneous ignition was assessed with planar laser induced fluorescence (LIF) of the OH radical, and the catalytic processes preceding homogeneous ignition were investigated with one-dimensional Raman measurements of major species concentrations. The numerical predictions included an elliptic two-dimensional CFD code with elementary heterogeneous and homogeneous chemical reaction schemes and detailed transport.

2. Experimental

2.1. High-pressure test-rig

The test-rig (Fig. 1) consisted of a rectangular reactor, that formed a liner inside a high-pressure cylindrical vessel [7]. The reactor comprised of

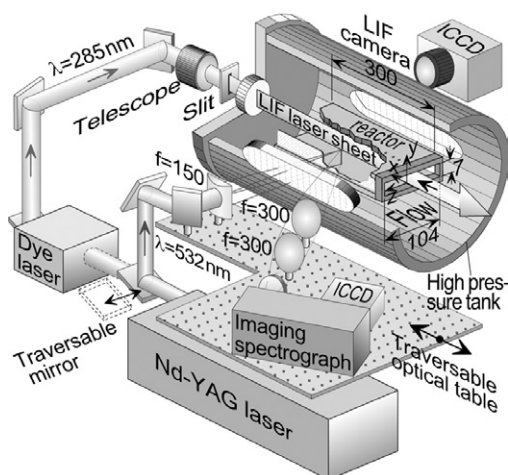


Fig. 1. Schematic of the test-rig and the LIF/Raman set-up.

two horizontal Si[SiC] ceramic plates (300-mm long (x), 110-mm wide (z), 9-mm thick, and positioned 7-mm (y) apart) and two 3-mm thick vertical quartz windows [11]. The inner Si[SiC] surfaces were coated via plasma vapor deposition with a 1.5 μm thick non-porous Al_2O_3 layer, followed by a 2.2 μm thick Pt layer. Measurements of the total and active catalyst areas with BET (Kr-physisorption) and CO-chemisorption, respectively, verified the absence of a porous surface structure. The surface temperature along the x - y symmetry plane was measured by S-type thermocouples (12 for each plate) embedded 0.9 mm beneath the catalyst, through holes eroded from the outer Si[SiC] surfaces. The plate temperatures were controlled by two resistive heaters positioned above the ceramic plates.

Air was preheated and mixed with CH_4 in two sequential static mixers. The CH_4/air premixture was driven into the reactor through a 50-mm long inert rectangular honeycomb section that provided uniform inlet velocity. A thermocouple positioned at the downstream end of the honeycomb measured the reactor inlet temperature. Optical accessibility from both reactor sides was maintained by two 350-mm-long and 35-mm-thick quartz windows on the high-pressure tank (see Fig. 1). Two additional quartz windows, one located at the rear flange of the high-pressure tank and the other (not shown in Fig. 1) at the exhaust section of the reactor, provided a counterflow streamwise optical access for the LIF experiments. In the tests with H_2O dilution, superheated steam was supplied by an AWTEC-DLR steam generator. In both H_2O - and CO_2 -dilution tests, the oxidizer was pure oxygen; nitrogen was separately added as a balance.

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