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Effects of flow-field and mixture inhomogeneities on the ignition dynamics in continuous flow reactors

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ABSTRACT

The objective of this study is to characterize effects of turbulence and flow-field inhomogeneities on the mixing and ignition-dynamics in flow reactors. Specific focus is on investigating the ignition characteristics of hydrogen-containing fuels at gas-turbine-relevant operating conditions. Two different model formulations are developed to describe the mixing, induction, and subsequent ignition and combustion. Utilizing these models, parametric studies are performed in a generic flow reactor configuration that is representative of commonly employed facilities. Diagnostics is developed to quantify the ignition dynamics. Results show that in the case of an initially homogeneous mixture, the ignition process is fairly insensitive to the underlying flow-field. However, by considering inhomogeneities in temperature and mixture composition it is shown that the ignition process exhibits a more pronounced sensitivity to temperature perturbations, and the ignition delay is only weakly sensitive to initial equivalence ratio perturbations. Simulation results show that temperature fluctuations of less than 10% of the mean temperature are sufficient to significantly affect the ignition-onset. Results from this parametric study identify the need for quantitative measurements of temperature and composition to better characterize flow reactor facilities. A time-scale analysis is performed to characterize competing physical processes that are associated with turbulent mixing, autoignition, and flame propagation. Qualitative comparisons with experimental data suggest the possibility for deflagrative ignition modes that can occur at low temperature operating conditions.

1. Introduction

Prior to the large-scale production of shale gas, the utilization of coal-derived synthesis gas (syngas) and high-hydrogen content (HHC) fuels has been considered as an attractive opportunity for electric power generation [1]. The integrated gasification combined cycle (IGCC) process has been considered as viable technology for the syngas combustion in advanced power plants. In coal-based IGCC power plants, syngas – containing hydrogen (H_2) and carbon monoxide (CO) as primary fuel components – is generated through a coal gasification process [2]. After sulfur oxides and other pollutants are removed, the syngas is combusted in a gas turbine cycle, and excess heat is converted in a subsequent steam turbine cycle. Compared to conventional coal-fired power plants, IGCC-systems offer advantages for precombustion carbon capture and sequestration, significant reductions of emissions of pollutants and particulates, and the potential for higher thermal efficiencies.

However, despite the opportunities to achieve higher efficiencies and to reduce pollutant emissions, the stable operation of HHC-fuels in IGCC power plants introduces significant technological and scientific challenges. These challenges are primarily attributed to the lower density and higher diffusivity of hydrogen compared to conventional hydrocarbon fuels. These different thermo-diffusive properties lead to higher flame speeds, extended flammability limits, and lower ignition temperatures, which can adversely affect combustion stability and fuel conversion [3]. In addition, high flame temperatures associated with hydrogen combustion make NO_x emissions a concern, so that premixing and dilution strategies are required to reduce these emissions to acceptable levels [4].

Additional challenges arise from IGCC operating conditions that lie in the intermediate temperature range (600–1000 K) and high-pressure regime (up to 30 bar). Recent experimental and computational investigations identified considerable discrepancies between measured and predicted ignition delay times at these conditions [5–8]. Several reasons have been proposed to explain these discrepancies [5,7,9,10], including (i) sensitivity of rate coefficients at high-pressure/low-temperature conditions, (ii) effects of

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inhomogeneities in the mixture composition and flow-field structure on the ignition characteristics in experimental facilities, (iii) catalytic effects of HHC-fuel mixtures due to contaminations with metal carbonyls and nitric oxides, (iv) the significance of thermo-viscous boundary layers, wall-heat-transfer, and (v) turbulent mixing. While many of these non-idealities are often specific to a particular experimental facility, it remains a major challenge to experimentally isolate and systematically quantify individual contributions.

Ignition delay measurements are commonly conducted in shock tubes (STs), rapid compression machines (RCMs), and continuous flow reactors (FRs). Flow reactors are of particular interest for ignition studies at gas-turbine relevant operating conditions, since these facilities provide access to extended test-times beyond 500 ms. Currently employed FR-facilities share a similar setup [8,11–13]. In its simplest form, a flow reactor consists of a long pipe that is separated by a fuel injector module into a flow conditioner and a test section. Air is supplied from a pressurized tank through the flow conditioner in which the air is preheated. The fuel is subsequently injected via a mixing module, and different mixing strategies [14] are utilized to achieve an approximately homogeneous mixture prior to ignition. Following the mixing, the test-gas mixture passes down the test section in which autoignition takes place. To minimize heat losses and other secondary effects, the test section is commonly heated and thermally insulated. Thermocouples, photo-diodes, and pressure sensors are employed to detect the ignition location. The ignition location, x_{ig}, is then related to an ignition delay time, τ_{ig}, using the theoretical bulk flow velocity U_b [11,15]:

$$\tau_{\mathrm{ig}} = \frac{x_{\mathrm{ig}}}{U_b}. \tag{1}$$

To guarantee that the mixture ignites within the finite-length test section, mass flow rate and bulk velocity are adjusted, depending on the pressure and temperature conditions of the test gas mixture.

Flow-reactor studies of HHC-fuels at gas-turbine-relevant operating conditions [8,11,13] have shown that the ignition delay is primarily controlled by the hydrogen-content of the syngas-mixture and is linked to the H_2/O_2 explosion limit. These measurements also revealed that the ignition exhibits a pronounced sensitivity to initial conditions. Often, ignition could only be observed in some cases for nominally identical initial conditions [8] or ignition events occurred at different locations in the test section [13]. In the case of a successful ignition, the ignition event occurred earlier than predicted from homogeneous isobaric reactor simulations. These observations suggest that the ignition is not deterministic and most-likely dependent on facility-specific non-idealities that are present in the flow reactor. In particular, Beerer and McDonell [8] pointed out that the sources for these sporadic ignition events could be linked to incomplete mixing between fuel and oxidizer, temperature variations in the preheater, turbulent mixing in the test-section, and the influence of the boundary layer in affecting the induction time and the ignition process. Therefore, a better understanding of the effects of inhomogeneities in temperature and mixture composition, and the role of the aerodynamics in affecting the ignition process is crucial towards the characterization of ignition properties of HHC-fuels.

The objective of this paper is to assess the sensitivity of turbulent mixing and inhomogeneities in temperature and mixture composition on the observed ignition dynamics of HHC-mixtures. To this end, an idealized flow reactor configuration is considered that has the same geometric dimensions as the UTRC flow reactor facility [11]. The flow-field inside the flow reactor is described by considering two different modeling approaches. In the first part, the spatio-temporal evolution of the mixture is represented by a particle method, and the high-pressure H_2/O_2 kinetic mechanism by Burke et al. [16] is used to describe the reaction chemistry. Effects of laminar and turbulent velocity profiles on the ignition are investigated. Motivated by previous experimental observations, the significance of inhomogeneities in temperature and mixture composition is then investigated, and results of this study are presented in Section 3. Because of the computational complexity of the particle method, a parabolized Eulerian model is developed in Section 4. This model assumes a stationary ignition-combustion process, which is described using a Reynolds-averaged approach. A tabulation method is used to describe the reaction chemistry. The computational efficiency of this model enables the consideration of an extended parameter space that is not accessible with the particle model. A time-scale analysis is performed, and three different ignition regimes are identified that can be associated with the competition between homogeneous ignition, flame-deflagration, and turbulent diffusion. Qualitative comparisons with experimental results, presented in Section 4.3, provide some evidence for the presence of premixed ignition regimes that are most prominent at low-temperature conditions. The manuscript finishes by offering conclusions.

2. Diagnostics and ignition criterion

The autoignition process in flow reactors is affected by the unsteady velocity field and the turbulent mixing. To analyze this stochastic ignition process, a statistical approach is considered. To this end, we define the probability density distribution (PDF), $\mathcal{P}(\psi; x, t)$, which is evaluated from a scalar field variable ψ and is a function of axial location x and time t. A cross-sectionally averaged quantity is then evaluated as:

$$\{\psi\}(x,t) = \int \psi \mathcal{P}(\psi; x, t)\mathrm{d}\psi. \tag{2}$$

By introducing the cumulative distribution function $\mathcal{C}$, the following ignition criterion can be defined:

$$\mathcal{C}(\psi_{\mathrm{ig}}; x, t) \equiv \int_{-\infty}^{\psi_{\mathrm{ig}}} \mathcal{P}(\psi; x, t)\mathrm{d}\psi \leqslant \mathcal{C}_{\mathrm{ig}}, \tag{3}$$

where

$$\psi_{\mathrm{ig}} = \alpha\psi, \tag{4}$$

(with α being a parameter) is an ignition threshold – for instance a certain temperature increase at the thermocouple or the detected light-absorption at a photo-multiplier. The ignition location, x_{ig}, is then defined as the most-upstream location at which Eq. (3) is fulfilled. In this study, we consider the temperature ($\psi \equiv T$) to characterize the ignition location. In a separate investigation, OH and OH* were also considered as ignition markers, giving very similar results.

3. Lagrangian flow reactor model

3.1. Mathematical model

In this section, a particle-field method is employed to describe the autoignition of the fuel/air mixture inside the flow reactor test section. This method describes the spatio-temporal evolution of an ensemble of statistically independent notional particles through a three-dimensional flow-field. The spatial location and composition of the nth notional particle in this ensemble is, respectively, denoted by $\boldsymbol{x}^{(n)}$ and $\boldsymbol{\Phi}^{(n)} = (\boldsymbol{Y}^{(n)}, T^{(n)})^T$, with $\boldsymbol{Y}$ being the vector of all species mass fractions and T is the temperature. The particle position $\boldsymbol{x} = (x, y, z)^T$ is obtained from the solution of the following stochastic differential equation [17,18]: