

Characterization of Mixing and Ignition Effects in Flow-Reactor Facilities Using a Particle Method

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Stationary power plants utilizing integrated gasification combined cycle (IGCC) technologies are considered to be a viable option for clean electricity generation. Considerable design challenges still exist that are attributed to the IGCC-operating conditions and requirements for the stable combustion of syngas and high hydrogen content (HHC) fuels. Recent investigations have indicated discrepancies between measurements and simulations of the ignition delay for syngas-mixtures at high-pressure and low-temperature conditions. The objective of this study is to characterize effects of turbulence and flow-field inhomogeneities on the mixing and ignition-dynamics in flow-reactors. To this end, an idealized flow-reactor is considered and a particle method is used to describe the mixing, induction, and subsequent ignition of the reactants. Utilizing this model, parametric studies are performed to quantify effects of flow-field and scalar inhomogeneities on the ignition-process for practically relevant temperature and pressure conditions. Results suggest that the unsteady velocity field affects the ignition location and residence time in the flow-reactor. Stochastic events, such as the formation of localized ignition spots upstream of the main ignition front could be observed in the simulation. Their impact on the global ignition dynamics needs to be evaluated in further research activities.

I. Introduction

The energy supply of a rapidly growing population under the constraint of limited resources and the goal of reducing greenhouse gas emissions is an increasingly important challenge for research. Predictions by the U.S. Energy Information Administration forecast an increasing demand for electric power over the next 25 years, particularly in the developing world.¹ The electricity is predicted to come from a mix of conventional and renewable sources. Because of its abundance, coal remains one of the most attractive energy carriers – provided that it can be used in an efficient and clean manner. Gasification techniques provide an opportunity to generate synthetic gas (syngas) from coal, and thereby removing particulates, sulfur oxides, and other pollutants. Syngas, containing hydrogen (H_2) and carbon monoxide (CO) as primary fuel components, can then be utilized in a gas-turbine cycle to generate electricity. In combination with heat recovery systems, these integrated gasification combined cycle (IGCC) power plants can exceed thermal efficiencies of conventional steam power plants.

Despite the potential of the IGCC technology, several engineering challenges remain to be solved in order to enable the stable and robust combustion of high hydrogen content fuel. In particular, the combustion of syngas under premixed conditions comes along with the challenge to guarantee

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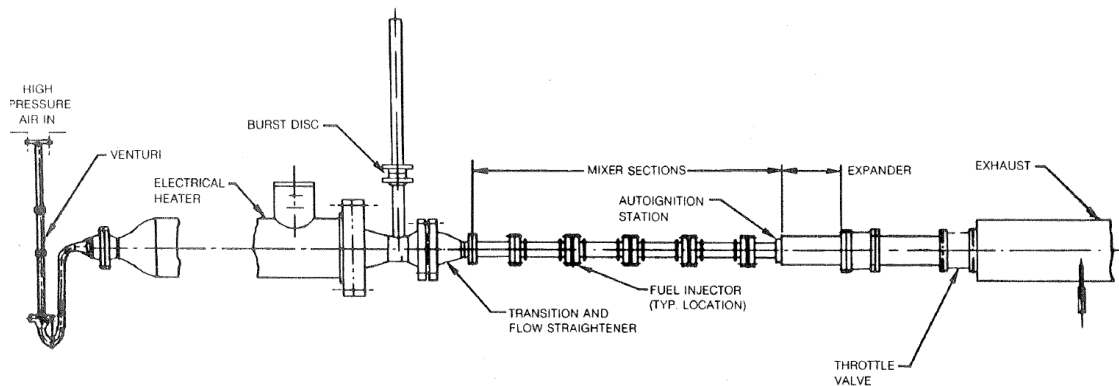


Figure 1. Schematic of flow-reactor (Figure taken from Ref. 2).

stable combustion. Under adverse conditions the flame can detach, lift-off, and eventually blow out. Other undesirable conditions include upstream flame propagation, spontaneous ignition of the mixture, and thermo-acoustic coupling. These unfavorable combustion regimes depend heavily on fuel properties and demand accurate prediction for advances in combustor design.

While there are several studies on ignition delay times in high temperature setups, such as shock tubes and rapid compression machines, there is only limited data of ignition delay times available at conditions that are relevant for gas-turbine systems.^{2,3} Flow-reactor experiments can fill this gap. These facilities allow to study autoignition of gas mixture under conditions that extend to low-temperature (<650 K) and high-pressure environments (> 20 bar). In its simplest form, a flow-reactor consists of a long pipe that is supplied by a continuous flow of preheated air. Through an injection system, gaseous fuel is rapidly mixed into the air-stream in order to achieve a homogeneous fuel/air mixture. The flow-reactor can be pressurized, and the walls are typically isolated or preheated in order to achieve nearly adiabatic operating conditions. A schematic of a flow-reactor facility is illustrated in Fig. 1. As the mixture propagates downstream, combustion occurs by autoignition. The ignition location, x_{ig} , is recorded by thermocouples or emission-probes and converted into an ignition delay-time using the Lagrangian relation

$$\tau_{ig} = x_{ig}/\langle U \rangle, \quad (1)$$

where $\langle U \rangle$ is the theoretical bulk-flow velocity.

Experimental studies of syngas/air mixtures⁴ and lean hydrogen/air mixtures⁵ indicate that measured ignition delay-times exhibit a broad distribution. Specifically, ignition could only be observed in some cases for nominally identical initial conditions.⁵ In cases of successful ignition, the ignition event occurred earlier than predicted by kinetic models for syngas combustion.³ These observations suggest that the ignition is not deterministic and most likely dependent on other effects that could include fluid-dynamics, scalar mixing, surface-catalytic effects, and mixture-inhomogeneities, among others.^{6,7} Therefore, a better understanding of effects of inhomogeneities in the temperature and mixture composition is crucial towards the characterization of ignition properties of syngas and HHC fuel.

The objective of this paper is to investigate effects of turbulent mixing and flow-field evolution on the observed ignition dynamics of syngas mixtures. To this end, a canonical flow-reactor configuration is considered that consists of a constant-area cross-section. The spatio-temporal evolution of the mixture composition inside the reactor is described using a particle method. Since flow-reactors are typically operated in the high Reynolds-number regime – with characteristic Reynolds numbers up to 2×10^5 – the flow-field inside the reactor is described from the solution of a turbulent pipe-flow.

Relevant aspects of the mathematical model are outlined in the next section. The computational setup and parametric variations of the operating conditions are described in Sec. III, and results are presented in Sec. IV. The paper finishes with conclusions.

II. Mathematical Model

II.A. Turbulent Flow-Field Description

For the description of the ignition inside the flow-reactor a particle method is employed. In this formulation, notional particles are advected through an irrotational velocity field. To investigate the effects of the unsteady turbulent flow-field evolution on the ignition dynamics, we describe the velocity field from the solution of a turbulent pipe-flow simulation. To this end, a highly resolved large-eddy simulation (LES) of a periodic pipe is conducted. A cylindrical grid with $128 \times 128 \times 128$ computational cells is employed. The grid uses an equidistant grid-spacing in axial and azimuthal direction, and the mesh in radial direction is refined in order to resolve the viscous boundary layer structure. In this study, four different conditions of increasing Reynolds number are considered. A comparison of the instantaneous field for the velocity component in axial direction is presented in figure 2. This qualitative comparison shows that the range of turbulent scales increases with increasing Reynolds number. A quantitative analysis of the statistical flow-field quantities in terms of outer and inner scaling variables is shown in figure 3.

From figures 3(b) and 3(c) it can be seen that velocity gradient at the wall becomes increasingly steeper with increasing Reynolds number, resulting in higher turbulent production. A closer view into the wall region shows that the location of highest turbulent fluctuation moves closer to the wall with increasing Reynolds number as the boundary layer narrows.

II.B. Description of Mixing and Ignition

The particle method describes the spatio-temporal evolution of the composition and location of the notional particles. Each particle has a location \mathbf{X} , velocity \mathbf{U} , and composition Φ . The stochastic differential equation (SDE) that describes the particle position is derived from a Langevin equation,^{8,9} and can be written as:

$$d\mathbf{X}(t) = \mathbf{U}(\mathbf{X}, t)dt + \sqrt{2\Gamma_T} dW(t), \quad (2)$$

where dW is a Wiener process, and Γ_T is the turbulent diffusivity.

The temporal evolution of the particle composition is obtained from the solution of the following deterministic differential equation:

$$\frac{d\Phi}{dt} = \frac{C_\Phi}{2} \Omega_m(\mathbf{X}, t) (\bar{\Phi} - \Phi) + \omega(\Phi), \quad (3)$$

The first term on the right-hand-side of this equation describes the mixing of the particle with its environment and is here modeled using an IEM-formulation. The mixing frequency Ω_m is evaluated as

$$\Omega_m = \frac{C_\Omega(\Gamma + \Gamma_T)}{\Delta^2}, \quad (4)$$

where Δ is the filter-size of the underlying LES-computation, and C_Ω is a model constant that is set to a value of 3. The second term on the right-hand-side of Eq. (3) is the chemical source term, which is directly evaluated from a detailed chemical mechanism.

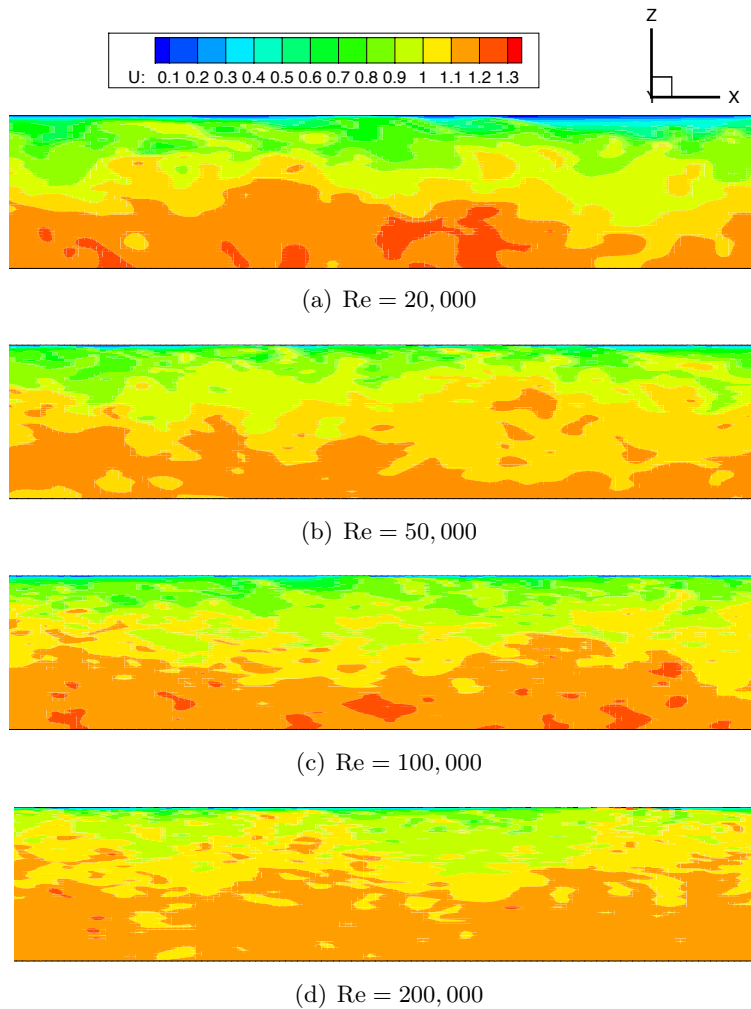


Figure 2. Comparison of the normalized instantaneous axial velocity field at four different Reynolds numbers; only the upper half-section of the computational domain is show.

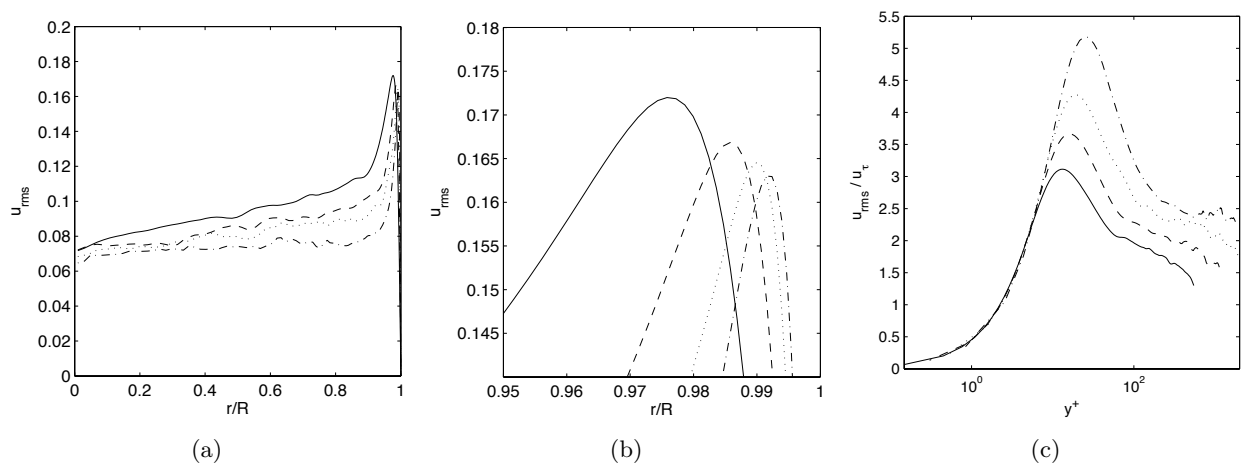


Figure 3. Statistical results of LES-pipe-flow computation: (a) Normalized root-mean-square velocity profiles as function of outer-scaling variables; (b) closed view on rms-velocity profile in wall-near region, and (c) velocity-profile in inner-scaling variables: solid line, $Re = 2 \cdot 10^4$; dashed line, $Re = 5 \cdot 10^4$; dotted line, $Re = 1 \cdot 10^5$; dash-dotted line, $Re = 2 \cdot 10^5$.

III. Computational Setup

In the experimental setup of flow-reactors, great care is taken to achieve a homogeneous mixture of fuel and oxidizer in the injector section. The mixing properties within a short distance downstream of the injector sector were studied by Samuelsen et al.¹⁰ and Santoro et al.⁴ using PLIF visualization of acetone. Close analysis of their results indicates that the mean mixture fraction is nearly homogeneous in the flow-reactor core and some stratification in the wall-near region is evident. To assess effects of this compositional variation on the ignition, the mixture composition is parametrically represented as inflow condition to the flow-reactor. This allows us to replace the geometric and facility-dependent complexity of the injector, and to describe the fuel/air mixture in terms of a distribution function with appropriate model-parameters that are obtained from experiments.^{4,10} Motivated by the analysis of PLIF-measurements by Santoro,⁴ we choose to represent the mixture-composition at the inlet of the flow-reactor by the following profile:

$$\bar{Z}(r) = Z_0 \tanh\left(\frac{R-r}{\delta_Z}\right) \quad (5)$$

where δ_Z is a parameter characterizing the mixture stratification. The coefficient Z_0 is evaluated so that

$$\frac{\phi Z_{st}}{Z_{st}(\phi-1)+1} = \frac{2}{R^2} \int \bar{Z} r dr, \quad (6)$$

where ϕ is the equivalence ratio and Z_{st} is the stoichiometric mixture fraction. Super-imposed to this mean-profile is a fluctuating component that is sampled from a beta-distribution with mean \bar{Z} and standard deviation σ_Z . Examples of the initial mixture composition for three different values of δ_Z and a fixed value of σ_Z are illustrated in figure 4.

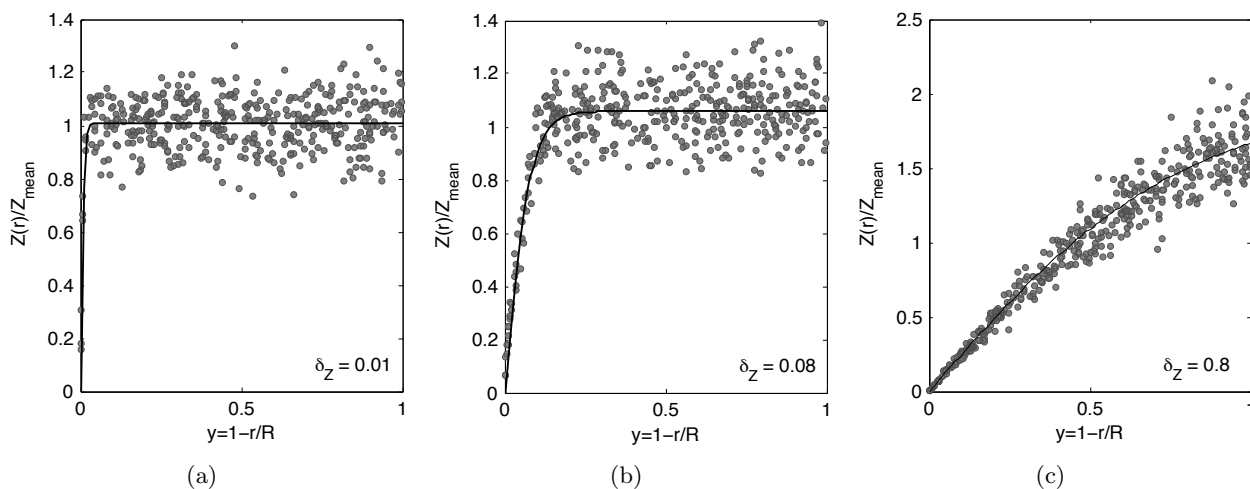


Figure 4. Initial mixture fraction distribution for different parameters of δ_Z and constant standard deviation $\sigma_Z = 0.10$. The solid line depicts the radial distribution of the mean mixture fraction using Eq. (5). The values of 400 randomly initialized notional particles are indicated by the markers.

IV. Results

In this section, results of a parametric sensitivity study are presented, analyzing the influence of the velocity profile and the mixture distribution on the ignition dynamics. A hydrogen/air mixture composition with an overall equivalence ratio of $\phi = 0.4$ and an initial temperature of $T_{init} = 950$ K is considered. The pressure is set to 20 bar. This condition translates to an ignition delay

time of approximately 30 ms (under idealized homogeneous reactor conditions). With a bulk-flow velocity of 10 m/s, ignition can be observed within a distance of less than 0.5 m downstream of the injection-section. The diameter of the flow-reactor is 45 mm and an azimuthal section of 22.5 degrees is considered. The flow-field is computed from a pipe-flow LES with $Re = 200,000$. The initial mixture composition is sampled from a distribution that is specified by $\delta_Z = 0.08$ and $\sigma_Z = 0.10$ (see Fig. 4(b)). This condition closely resembles the flow-reactor measurements of Santoro et al.⁴ The reaction chemistry is described using the high-pressure H_2/O_2 mechanism of Burke et al.¹¹

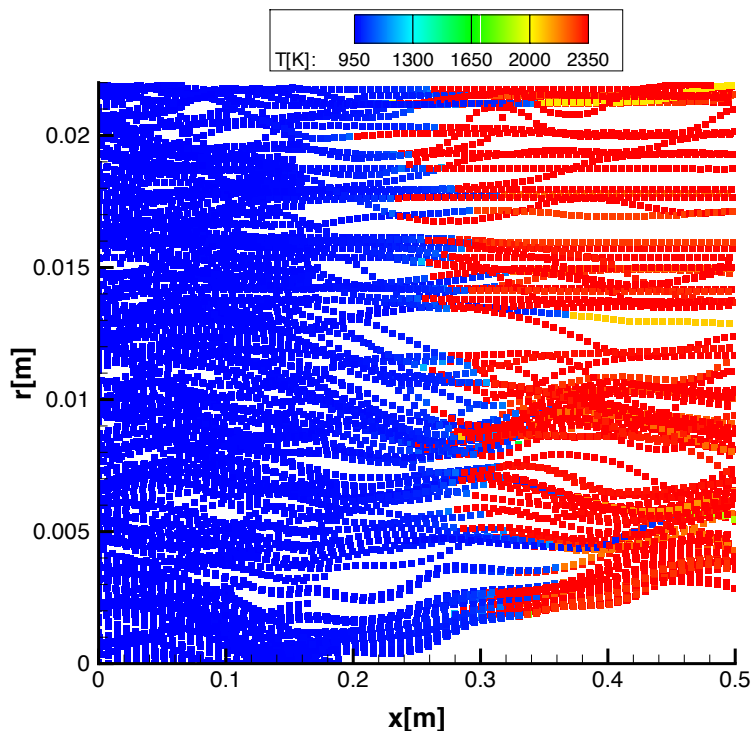


Figure 5. Trajectories of randomly selected particles in the flow-reactor. Particle are colored by the particle temperature. The initial particle temperature is $T_{init} = 950$ K.

Figure 5 shows the trace of a large number of randomly selected particles in a simulation with turbulent flow profile. The particles are colored by the particle temperature and the flow is from left to right. The particles follow individual – apparently random – trajectories and ignite downstream. By averaging the particles over the cross-section of the flow-reactor, a temporal evolution as shown in figure 6 is obtained. After initialization, the particles move downstream until all particles ignite and the final condition converges to the equilibrium composition.

A comparison of results for different flow-field profiles is shown in Fig. 7. For this, analytic velocity-profiles for a plug flow, Hagen-Poiseuille flow, and power law flow are investigated and compared against the turbulent LES flow profile. Figure 7 shows the cross-sectional-average temperature profiles as function of axial distance along the reactor. Two main effects are visible. The flow profile influences the propagation of the particles. Therefore the shape of the ignition front resembles the flow profile. This is particularly evident for the laminar cases. Furthermore, the progress of the chemical reactions and the final temperature is determined by the prescribed mean mixture fraction profile. Leaner mixture compositions delay the ignition at the wall in the absence of radial convection. However, turbulent fluctuations can entrain fuel-rich particles into the wall region. As a result, early ignition in the wall region can be observed in the scenario with LES flow-field. This effect appears also in the cross-sectional-average temperature plot, shown in figure 7.

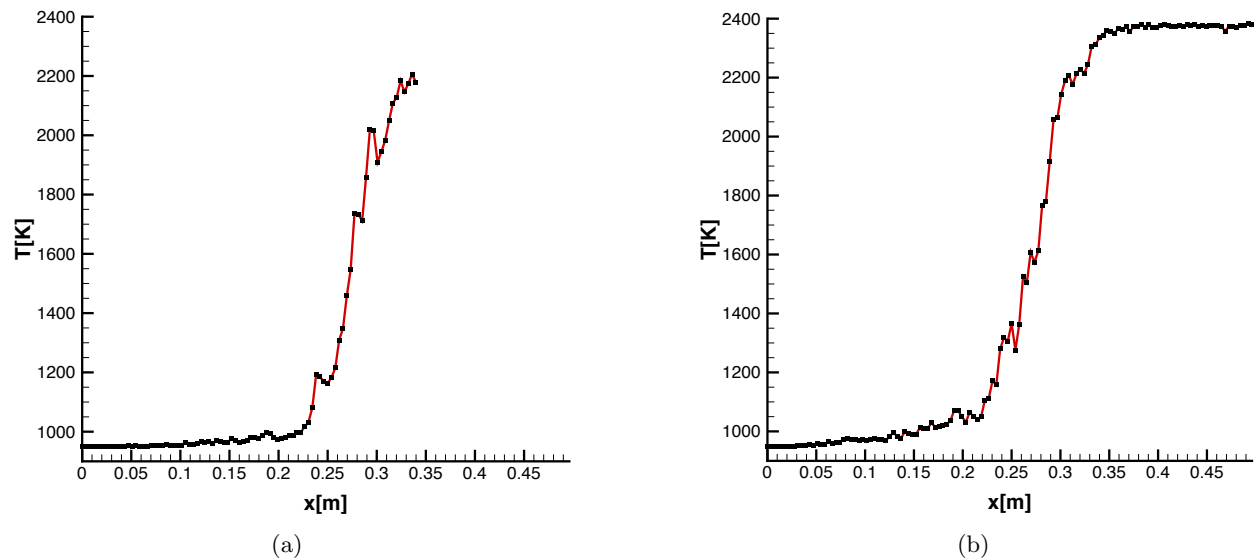


Figure 6. Evolution of the cross-sectional averaged temperature profile for the reference case at $T_{init} = 950\text{ K}$ at a time of (a) $t = 30\text{ ms}$ and (b) $t = 60\text{ ms}$.

While the location of 50% temperature increase is almost unaltered, the slope of the temperature evolution is sensitive to the flow profile.

A reduction of the mean flow velocity by an order of magnitude is analyzed in figure 8. From this comparison, we observe a stronger deviation from the homogeneous reactor curve for lower flow velocities.

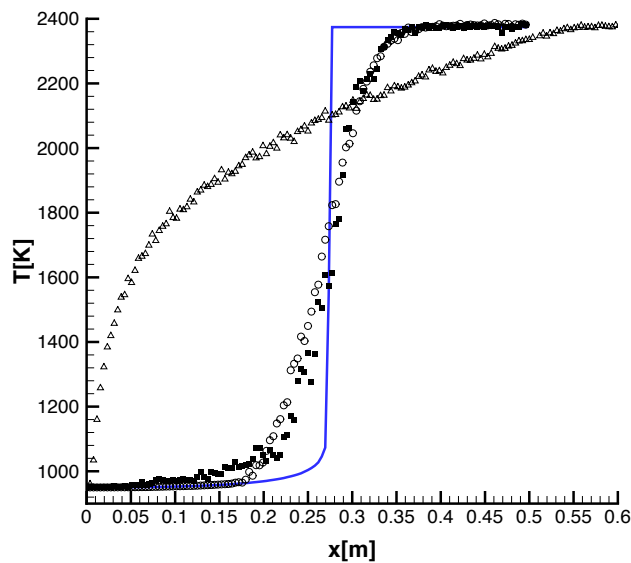


Figure 7. Temperature evolution for different flow profiles compared against a homogeneous plug flow-reactor represented by the solid blue line: LES flow-field (\blacksquare); parabolic profile (\blacktriangle); power law profile (\circ).

In addition, the sensitivity of the ignition characteristics to the fuel distribution was investigated. The corresponding profiles are plotted in figure 9. With an almost flat mean mixture profile ($\delta_Z = 0.01$), ignition events can be noticed already at the beginning of the flow-reactor. By contrast, using a profile with uneven distribution almost completely suppresses ignition events at the wall and postpones a complete mixture-ignition. However, the data scatter is fairly large for these conditions and indicates the pronounced stochastic dynamics during the ignition process.

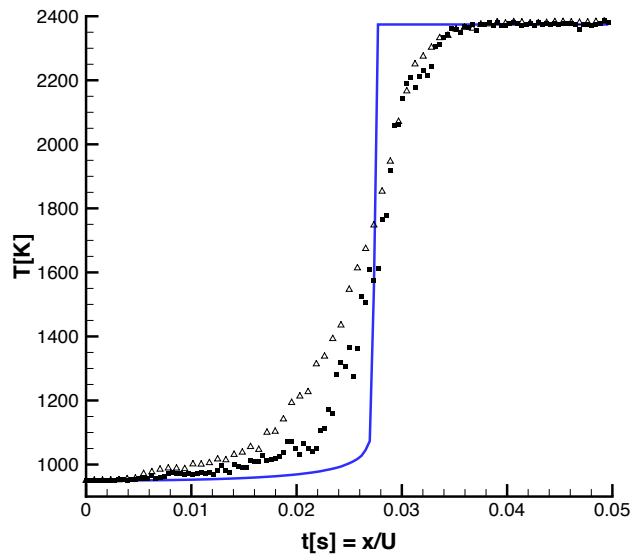


Figure 8. Temperature evolution for different flow profiles compared against a homogeneous plug flow-reactor represented by the solid blue line: LES pipe-flow with $Re = 200,000$ (■) and $Re = 20,000$ (Δ).

Larger fluctuations of the initial mixture composition broaden the zone of particle ignition. This leads to a significant temperature increase near the fuel-injector. From these results it can also be observed that the slope of the temperature evolution decreases.

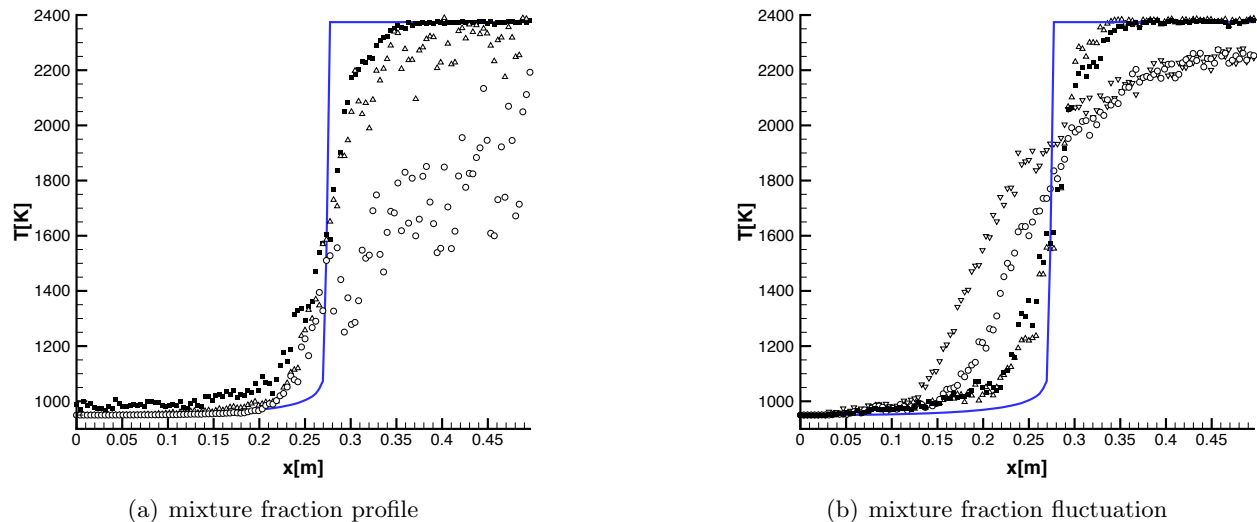


Figure 9. Temperature evolution for different mixture fraction distributions compared against a homogeneous plug flow-reactor represented by the solid blue line: (left) $\delta_Z = 0.01$ (■); $\delta_Z = 0.08$ (Δ); $\delta_Z = 0.8$ (\circ); (right) $\sigma_Z = 0.1$ (■); $\sigma_Z = 0.05$ (Δ); $\sigma_Z = 0.4$ (\circ); $\sigma_Z = 0.8$ (∇).

V. Conclusion

The influence of hydrodynamic effects on the autoignition of syngas mixtures in flow-reactors was investigated. The unsteady three-dimensional, and turbulent flow-field in the flow-reactor was modeled using a large-eddy simulation. A particle method was used to simulate the evolution of the composition and temperature of the mixture along the pipe. When compared against the result of a homogeneous flow-reactor, the results showed that the velocity field directly affects the ignition

location. Reasons for this are the velocity-induced change in the residence time of the particles and turbulent mixing. Regions of low velocity in the wall region lead to localized ignition upstream of the main ignition, and compositional fluctuations broadened the area of the ignition front. The simulation results reveal that the slope of the temperature evolution is sensitive to hydrodynamic effects and initial conditions. In turbulent flow, ignition kernels can be observed in proximity to the wall far upstream of the expected autoignition location. Hence, the definition of ignition delay time is not trivial for flow-reactors. Instead of a distinct position, the onset of ignition is a stochastic event. The implications of this finding should be examined closer.

Acknowledgments

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